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Committee on Nuclear Science

MEASUREMENTS AND STANDARDS OF RADIOACTIVITY

Proceedings of an Informal Conference

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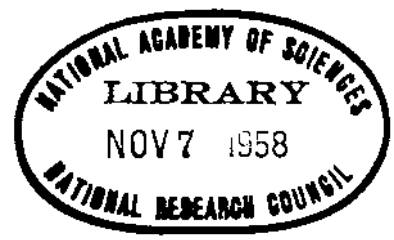

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FOREWORD

The proceedings of a three-day conference on Standards and Measurements of Radioactivity, held at the Tidewater Inn, Easton, Maryland, on October 9-11, 1957, are presented herewith. The conference was organized by the Subcommittee on Standards and Measurements of Radioactivity of the Committee on Nuclear Science of the National Academy of Sciences - National Research Council. The invited participants included experts in the field of radioactive standards both in this country and abroad.

In order to secure an informal productive atmosphere, the group was kept small in size and the notes were edited in such a fashion as to reflect the give and take of each session. These proceedings will provide a comprehensive record of the discussions for the benefit of the many individuals whom we would have liked to have had as participants but for the severe limitations of the conference size.

We are indebted to the Atomic Energy Commission for substantial encouragement and support and to Mr. J. S. Coleman, Executive Secretary of the Division of Physical Sciences, and Mrs. Catharine Parrish, of the Academy-Research Council staff, who were both efficient and indispensable in organizing the support for the conference and for their aid in reproducing this manuscript.

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SESSION I - ALPHA MEASUREMENTS AND STANDARDS

L. F. Curtiss, Chairman

Summary

The absolute measurement of the disintegration rates of radionuclides decaying by the emission of alpha particles has not in recent years received the attention that its importance should command. Dr. Robinson describes the application of modern techniques to the precise measurement of the rate of emission of alpha particles from a radioactive source. The result of his experiments is a considerable improvement in both the precision and accuracy. The over-all accuracy of his measurements appear to be of the order of 0.1 percent. One of the purposes of such accurate measurements on sources of alpha particles is the calibration of the rate of emission of alpha particles from specially prepared sources which may then be used in the calibration of other detectors of alpha particles.

Dr. Hayward describes the alpha particle standards now available from the National Bureau of Standards. The properties and limitations of these standards are presented with a discussion of further possible improvements.

The memory of the time when radium and its decay products were the principal sources of radioactive radiations dies out slowly in the face of more recent advances. Perhaps this tenacity is to be associated with the fact that our unit for the measurement of disintegration rates, the curie, is defined with the aim to bring it into agreement with the best estimate for the disintegration rate of radium, even though it is no longer tied directly to this figure. In the description by Dr. Kipfer of refined methods for measuring quantities of radium, the most significant contribution is the elegance of his method and the care obviously expended in the design of the equipment. The method has possible applications elsewhere.

Dr. Mann presents data to show that radium standards have recently been compared with a precision of the order of 0.1 percent to give agreement with the nominal values of the standards. He also recognizes that the usefulness of radium standards is rapidly decreasing and that cobalt-60 will undoubtedly take over the role in the future which in the past has been assigned to radium.

1. Alpha Standards

H. P. Robinson

A number of years ago we became interested in specific activity measurements on radioactive materials, and in particular alpha emitters. After a number of discouraging tries with small low geometry counters, we decided to build a large one with which we could get some very accurate measurements.

As a result of this we designed a low geometry chamber that was about 40 inches long. There have been other chambers made that have been even bigger, but this was our first large one. It was designed so we could measure all the important dimensions accurately and have a reasonable assurance that our measurements were reliable. We decided at that time, also, to use a phosphor and a phototube for detecting the alphas, rather than an ionization chamber which we used before.

Figure 1 is a photograph of the chamber mounted on the wall. It is about 40 inches high, not including the pre-amplifier, which is on top, and to the left we have the electronics mounted in a cabinet. We evacuate the chamber with an oil diffusion pump, but find that a mechanical pump is sufficient. We were a little concerned about scattering of the alphas by the residual gas in the chamber. It turned out to be completely unimportant. Evidently there are just as many scattered into the proper beam as there are scattered out. Even at 200 microns pressure we cannot detect any change in the counting rate.

This chamber has a factor of about 2600, which means that it is 1/2600th of 4π geometry.

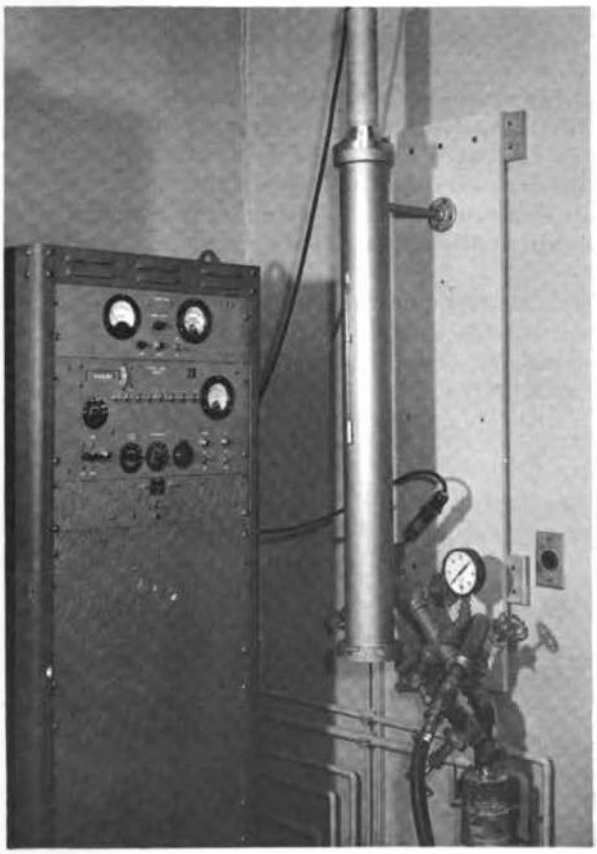


Figure 1. Precision low gometry chamber.

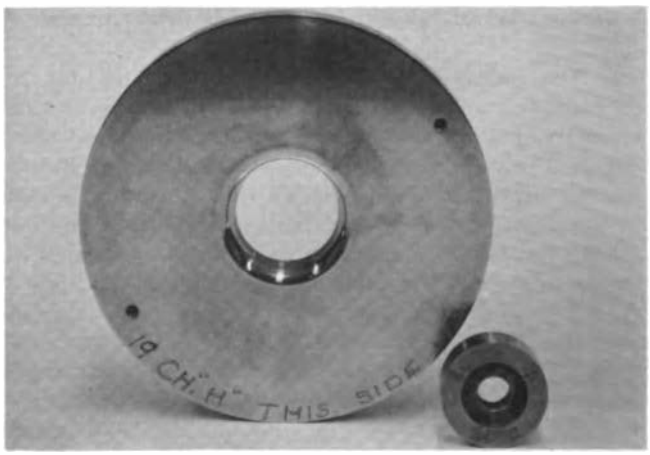


Figure 2. Schematic arrangement of chamber.

Figure 2 gives a schematic diagram of the arrangement. At the lower end is the source of activity. Then at the upper end is the collimator which is simply a hole in a disk. The hole has a 3-inch diameter and is measured very accurately. Above that is a film with a reflecting silver coating, and then phosphor deposited on glass, and above that oil connecting the phototube. The beam from the source is very accurately defined by the collimator, and knowing the distances we can accurately calculate the geometry factor.

I mentioned the silver film. That is a very

important part of it. If it were not there, light from the phosphor would escape downward and this would reduce the intensity of the pulse. As you will see later, it has a marked effect on the accuracy.

Also, there is another thing that is important; namely, the use of baffles. We have eight mounted along the wall, and these are spaced to eliminate scattering from the walls. If they are not there, the scattering accounts for almost one percent of the total count. That is intolerable, of course.

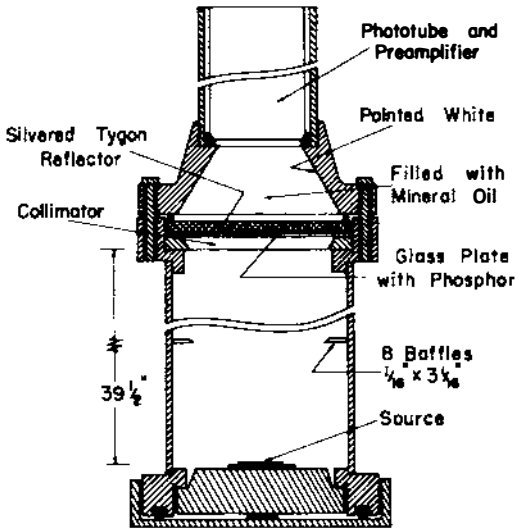


Figure 3. Collimator and gauge.

A 1-inch collimator is shown in Figure 3. In the lower right-hand corner is the gauge by which we measure the diameter of the collimator. It turned out to be a very simple system. The gauge is machined and ground to two or three thousandths of an inch smaller than the hole of the collimator, and the same thickness. We measure the diameter of the gauge by direct comparison with gauge blocks. Then we stick this gauge in the hole and measure the gap under a microscope, and this turns out to be a very convenient and accurate way of getting the diameter of the collimator. Of course, this diameter is very important, since one percent error in the collimator diameter represents two percent error in the geometry factor.

If all important dimensions are known to one part in 10,000, or so, then our counting accuracy is mainly limited by statistics. This is quite a limitation because it takes one million counts to get a standard deviation of a tenth of a percent. We take ten to one hundred million counts in order to improve the statistics, and run a number of experiments to detect any irregularities in the performance of the equipment.

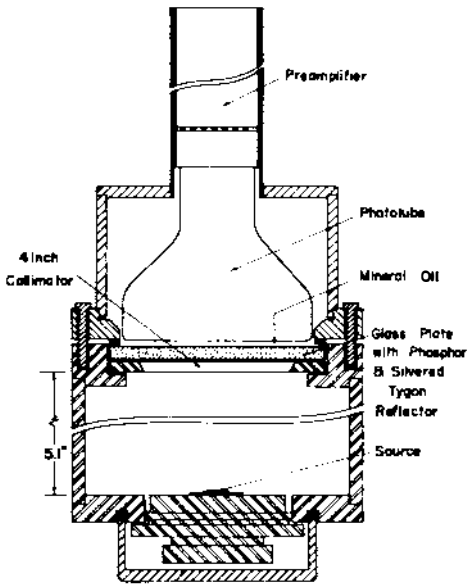


Figure 4. Precision medium geometry chamber.

Just recently we have designed a new chamber, as shown in Figure 4, having a factor of about 29. It will use a 5-inch phototube. With this chamber we can make precision measurements on sources a hundred times weaker than before. The important dimensions, which are the collimator diameter and the distance from the source to the collimator, can be accurately measured with gauge blocks and other techniques. So again we will know the dimensions quite accurately and, therefore, the geometry.

Figure 5 shows the effect of a silvered Mylar film. The middle curve was taken with the silver placed up so that it was the exposed silver surface that reflected the light up through the phosphor.

The top curve was produced with the silvered side down, which means that the surface of silver that was reflecting the light back up

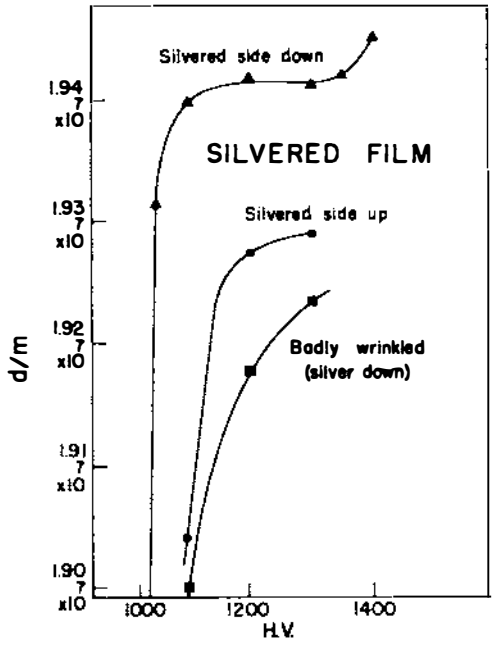


Figure 5. Plate counting plateaus obtained under various conditions.

was in contact with the Mylar, and, therefore, was presumably brighter or cleaner. I presume the exposed surface becomes tarnished a little and reduces the reflectivity of the light.

There is also shown the effect of a wrinkled surface. You can see that the total effect is of the order of a half to one percent between the best and the worst. This is too much for precision counting. The disintegrations per minute are plotted against the phototube voltage.

We now make the silver film in a different manner. First of all, we deposit the phosphor by letting the phosphor settle out of a water suspension on to the glass plate. After it has settled completely and a layer of about 15 to 25 milligrams per square centimeter is deposited, we drain off most of the water so that there is about an inch left over the phosphor, and then form a Tygon film on the surface of the water, and when this is "dry," we siphon off the rest of the water and that lowers the Tygon film onto the phosphor. This has to be done very slowly so that there is no disturbance. The phosphor is not anchored to the glass plate except by gravity, and it is very easy to disturb it.

When the film has settled completely and the water drained off, we leave it for a day or two until it is completely dry, and then it is quite durable. We silver the surface of the Tygon film in a vacuum chamber. In the one we made we overcoated with silicon monoxide, but I don't think this is necessary.

There is no difficulty from evacuation pulling the film away from the phosphor. We were a little concerned about this, but it has held up very well for a couple of years. We have not had very much experience with this technique because we made one film for a test, and a second one for use, and we have not had occasion to make any more.

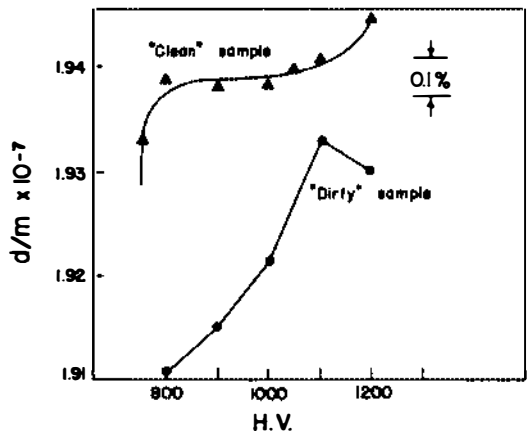


Figure 6. Effect of "dirt" on counting rate.

One of the things we encountered was a matter of cleanliness of the sources. After they sit around for a while, they acquire a small amount of dust or other contamination, and although it is almost invisible, it has a substantial effect on the counting efficiency. We found that we could very easily take care of this by washing the source off with alcohol before counting it. The lower curve in Figure 6 shows how a "dirty" sample performs. The upper curve is typical of what we get when we use a clean source. Every time we run a sample, we take a plateau on it. This shows up many of the defects that can occur in our measurements. We have to get a plateau or there is something wrong. Here is one example of it.

BORKOWSKI: What do you consider a

tolerable slope on the plateau, and what portion of the plateau do you operate at if it does have a slope?

ROBINSON: I really can't tell you what sort of a plateau we will tolerate. It is normally flat to ±0.05 percent or better. This one is satisfactory. We don't like to use too high a voltage. We think the lifetime of the phototube will be a little longer at the lower voltage. At the same time we take a point which is well above the knee of the curve.

WATTERS: From what particular source is the source material and how do you prepare these sources?

ROBINSON: This is an americium-241 source that was evaporated onto platinum.

WATTERS: Do you have any major particle size in the source?

ROBINSON: The sources in general are invisible. We could not assign any particle size. They go on presumably as atoms or molecules of americium.

SELIGER: This dirty sample intrigues me. How dirty is it?

ROBINSON: This is hard to say. If you look at it under the microscope, you see little specks of lint and other dust which look like boulders. An estimate of the fraction of the area covered suggests 0.1 percent in some cases. Possibly some of the dirt here was not dust, but an adsorbed film of some kind.

SELIGER: The curve indicates that half a percent in this particular case is completely adsorbed.

ROBINSON: Near the peak it is less than a half percent. It is more nearly a quarter of a percent. The alphas go through these dirt particles, and lose some energy. The resulting spread in the size of the light pulse causes the plateau to fall off sooner.

SELIGER: I am surprised you don't get any plateau at all with the dirty sample.

ROBINSON: Perhaps the plateau occurs above 1200 volts. We don't care to run our phototube any higher than that.

BAPTISTA: You mean there can be a self-adsorption of the quenching of the light in the case of the phosphor used?

ROBINSON: Why would there be any difference between the upper and lower curve? The quenching would be the same in either case.

BAPTISTA: In the clean source you don't consider that you are just counting all the light emission inside the phosphor. All the alpha particles that strike the phosphor are counted by the photomultiplier.

ROBINSON: I think we are counting all the alphas. We have made some interlaboratory comparisons both with Hanford and with Harwell, and in general the checks are of the order of 0.1 percent. Of course, this is quite encouraging. So we would say if we are losing counts, we are not losing more than of the order of 0.1 percent, or else other laboratories are also. The phosphor is silver-activated zinc sulfide.

PATE: What particle size did you get this down to and do you have any idea how thick the Tygon film was?

ROBINSON: The Tygon film is probably not more than 100 micrograms per square

centimeter in thickness. The silver deposited is of the order of one to two hundred micrograms. The Mylar film was about one milligram in thickness, but we no longer use this material.

PATE: How about particle size?

ROBINSON: I have measured this, but I just don't remember. It is the standard material supplied by RCA. It is not as fine as one can use, however.

CURTIS: How important is sample thickness, and did you take into account the sample area when you were computing your geometry factor?

ROBINSON: One of the reasons for choosing americium for standard sources is that it has a high specific activity. So for any activity that we want to use, the thickness will be essentially zero. These same samples are used by the alpha spectroscopy group in determining alpha energies to about 5 kilovolts, so there is no appreciable self-absorption. They are for all practical purposes weightless and usually are invisible.

The answer to the second question is yes. We made the chamber long enough to reduce the effect of the sample size. With a 40-inch low geometry chamber, and a 3-inch diameter source, the correction is only three parts in 2600, or about a tenth of a percent. It is quite easy to calculate. We don't even use the source size that large. It is more like three-eights of an inch diameter.

In the case of the intermediate geometry chamber, this will be a more important correction. It goes up as the square of the ratio of the source diameter to chamber height. The correction for a 1-inch source and 5-inch source-to-collimator distance is 0.5 percent.

GRUMMITT: If the thickness of the dirt in the dirty sample were very much greater than 100 micrograms per square centimeter, this would indicate you were losing, because of the Tygon film.

ROBINSON: Yes, although that Tygon film is much thinner than the Mylar film which we used to use. The Mylar is about 1000 micrograms per square centimeter. We have used the quarter mil Mylar satisfactorily with good results. That is another reason for believing that if we extended the voltage we would get a plateau. If the dust particles are not thicker than a milligram, we should count the alphas that get through the dust particles.

GRUMMITT: Would the Mylar shift the plateau to the right?

ROBINSON: I am sure it would. These results are taken with a given setting of the gain control.

BALAGNA: You have done no work where the phosphor is deposited directly on the phototube. Do you have any reason to believe that this is a better technique?

ROBINSON: It is easier, although we have made some alpha counters in which we deposit the phosphor right on the phototube. Having the phosphor away from the phototube has an advantage in that it tends to make the pulses more uniform.

LAZAR: I don't understand how you can raise the voltage and get a decrease in the counting rate.

ROBINSON: I think that is probably a statistical variation.

LAZAR: The point is that the increase in these so-called plateau curves indicates noise from the phototube entering into the counting rate, so increasing the high voltage with the dirty sample should increase the counting rate again. I don't see how you can get a plateau if one

doesn't show. Increasing the voltage should only bring in the noise according to the upper curve. It is not clear in my mind what is happening here, but it sounds to me that you are losing counts due to some sort of absorption somewhere.

BORKOWSKI: I think a really critical experiment in vacuum low geometry is to actually place absorbers on the sample source. We did this some time ago with a vacuum low geometry chamber. There one is amazed, obviously--one would expect that there should be very little effect of absorbers over the source until you get very close to the range of the particle. If you put a milligram per square centimeter of Mylar over the source, we found there was an undetectable change in counting rate. So your experience with Mylar films rather checks this type of thing.

I think it would be interesting to find out from an actual absorption measurement, not a self-absorption, but to run an absorption measurement using mica or plastic film directly over the source in order to find where the threshold is for your device.

ROBINSON: Yes. Probably in the future, as interest in this work grows, we will do more of this sort of thing. Perhaps this lower curve can be attributed also to changes in the operating characteristics of the electronics. I just can't say offhand. We know by experience that if we get a bad curve, we can often improve the results by washing the source with alcohol.

LAZAR: If in fact you do get dirty samples, it is not clear to me why you don't get dirty windows as well.

ROBINSON: These do not get dirty inside the chamber. It is when they are stored away.

LAZAR: When you pump on the system, pump oil could easily get on the photomultiplier as dirt on the sample.

ROBINSON: I don't think it was pump oil, although there is back diffusion of oil in a diffusion pump.

UNIDENTIFIED: Could this be adsorbed moisture by any chance?

ROBINSON: I really don't think so. You cannot imagine a milligram, or even a tenth milligram of moisture per square centimeter adsorbed on the source.

UNIDENTIFIED: You have monoenergetic alpha particles and what you suggest is that 100 micrograms per square centimeter reduces the ionization such that you need an increase of maybe 400 volts in your high voltage with a resultant gas multiplication increase of maybe a thousand.

ROBINSON: At the lower end of the plateau we are counting essentially 99.5 percent of the particles, so we are not losing on those. We are losing only a small fraction, but the effect is exaggerated on this expanded scale. Does that answer the question?

UNIDENTIFIED: No. I am not convinced, but that is not relevant.

BORKOWSKI: You mentioned 100 micrograms. Dust particles can be 10 milligrams per square centimeter. You run into the same problem with your soft betas.

GRUMMIT: If they are, they should be weighable.

BORKOWSKI: It depends on the disposition of the source itself.

ROBINSON: We have not tried to make a research project of this to see if they are dust particles. If you have some doubts about it, perhaps your doubts are justified. This set of data was obtained when we were experiencing trouble with dust. We have just given this tentative explanation, and it may be wrong.

GROSS: I wanted to ask you if you had done anything in the way of determining pulse height spectrum.

ROBINSON: At the very early stages of the work we did. It happened that at that time we had not put the baffles in the chamber, and we got very poor results. Afterwards the pulse analyzer was not handy so we didn't use it extensively. The pulse analysis looks very much better than the results would indicate. In other words, the pulses fall within a narrow energy range, and we could not estimate even a tenth of a percent in the tail. So it is something we can't pick up with the pulse analysis.

SELIGER: Approximately what resolution would you estimate you get?

ROBINSON: It is not good compared with a solium iodide crystal or a homogeneous phosphor of any kind. The spread in the energy is 20 or 30 percent.

SELIGER: This is a zinc sulfide phosphor, so I assume your signal to noise ratio is tremendous. What would you estimate is the cause for the plateau rising at 1100 volts?

ROBINSON: I don't know. Possibly it is noise. The other explanation is that our electronics are not the best in the world. The scaler and amplifier that go with it may be producing spurious results by overloading.

BORKOWSKI: What was the clipping time you used on this?

ROBINSON: The pulse is of the order of a couple of microseconds in width, so the amplifier can be overloaded and that may be the reason for the short plateau. I must confess we have not gone into that since we have found conditions where we could get good reproducible results.

SELIGER: I wonder if anyone has tried using a detector, a simple proportional counter, rather than a phosphor? It seems the resolution would be much better.

ROBINSON: Most people do use a proportional counter. We went to the phosphor because it eliminates some of the problems with the proportional counter. You don't have a gastight window; for example, this is a difficult problem with a 3-inch diameter collimator.

SELIGER: For alpha particles you are essentially counting a lower energy alpha particle because your incidence is just about normal in the low geometry case. Thus, the average absorption of energy from each alpha particle would be about the same.

ROBINSON: With the proportional counter.

SELIGER: With the window.

ROBINSON: Yes. It is here, too, or do you mean that the phosphor produces a greater spread?

SELIGER: The phosphor produces a much larger spread.

ROBINSON: I agree.

BAPTISTA: I might say that in using radium sources, we count simultaneously the alphas and betas on a 4π counter operating in the Geiger region. The results are quite good. What happens is that the plateaus are completely gone when the ratio between alpha and beta is increased. There is a deterioration of the plateaus in the Geiger region where it is completely flat for beta particles. If you count alpha particles in the 4π geometry, the plateau slope increases. Of course, you can get some accurate results by extrapolation of the plateau to the threshold. I still get some valuable results. I have no experience in the proportional region where I think you can get the best results.

YAFFE: We have actually done this in the proportional region, and we have no trouble with plateaus. You get very nice alpha and beta plateaus, using 4π counting.

ROBINSON: Hall and Glover at Harwell, on whose samples we made some measurements, used a proportional counter above the chamber. We checked to a tenth of a percent. We chose the phosphor and we have done our work with the phosphor, as an independent method. Whether it is best in the long run, I don't know. In the case of the proportional counter, there were some corrections that had to be taken into account, because of the support of the wire.

CURTIS: We also have used a proportional counter in several low geometry measurements. We count 2×10^2 disintegrations up to the curie level, and we estimate ±1 percent accuracy.

UNIDENTIFIED: I think it is important to point out that you have to be careful about overloading. People who have good counts either accidentally or otherwise have a good amplifier around.

ATEN: Are there any special precautions to be taken in construction of baffles?

ROBINSON: No. Our baffles were 1/16-inch brass. In the new chamber the size is large enough so that we don't need baffles.

ATEN: Is the number of baffles very important?

ROBINSON: No. We chose them very conservatively. We probably could have made use of half that number of baffles. We used eight.

CAMPION: Could you tell us how you measured the quantity of material evaporated onto your sources when you use this for specific activity measurements?

ROBINSON: The material was prepared not by vacuum evaporation but by depositing from solution, and evaporating to dryness. Jim Wallmann is the man who did this. He was interested in checking specific activity and, therefore, the half life of americium-241, which incidentally is less than the 470 years usually given. Wallmann has recently checked the figures of Hall and Markin, who got 458 years.

CAMPION: Are the characteristics similar with dry aliquots as opposed to evaporated sources?

ROBINSON: I can't personally say. I imagine they are, because Wallmann was satisfied with his results.

PATE: It would be quite easy to put this source preparation on a quantitative basis. We are doing some work at Brookhaven on distillation processes essentially with a closed system.

This is the same method we used at McGill for laying down evaporated sources on thin films. This is for the preparation of beta active sources for beta spectrum and 4π counting, and things of this nature. It is possible to arrange for a system such that you put a certain amount of material, a known quantity of your source material, into the furnace and transfer it quantitatively onto a suitable material, platinum in this case, the material setting itself down in a well-defined area defined by the geometry of the furnace system, and particle size something less than 200 Angstroms.

Another remark that might be interjected on this question of amplifiers; somebody made the remark that you get longer plateaus for alphas and betas if your radiation is entering the counting chamber essentially in a collimated beam. That is true. On the other hand, you can get yourself quite long alpha plateaus even with 4π geometry where your radiation is at low angles. We quite frequently observe, I think, 6 or 7 or 8 hundred volt plateaus, starting with 2 kv in the

type of counters used at McGill. These were good ±0.33 percent. So clearly, you have no trouble with plateau if you get yourself a good amplifier, whether by accident or otherwise.

ROBINSON: Yes, this is true. There is no pretense that this is anything more than the conventional amplifier. It has no extraordinary characteristics. We perhaps could extend the curve quite a bit if we had a good amplifier. This other point of transferring evaporated sources quantitatively is a very interesting one. We would like to know more about that. It is quite important in preparing a good sample.

PATE: We realize this is quite important. We have quite a program going. We are still in the early stages, however.

KOFOED-HANSEN: I would like to inquire about the spherical uniformity of the alpha particles coming off. If you are talking about one part in a thousand accuracy, wouldn't there be a possibility of some back scattering?

ROBINSON: As I understand it, back scattering is completely negligible if the angle is greater than 30 or 35 degrees.

SELIGER: If you prepared a source of this nature and counted it and gave it to someone else to count who counted it in a 2π geometry, there would be a different answer by about 1 to 1-1/2 percent.

ROBINSON: Yes. We don't think you can do precision work in a 2π counter. In a 4π, yes. We use 2π counters, and we use a source that has been calibrated in the low geometry counter to calibrate our 2π counters.

SELIGER: I think you might offend some people who do 2π counting here.

ROBINSON: I don't think so, if the people have looked into it, because you can't avoid back scattering in a 2π counter. We went to low and medium goemetry counting to avoid the problem of back scattering.

PATE: If you are giving a sample for people who measured disintegration rates, and come back with a half scatter, this can't be very good. They should be aware of this. The back scattering is primarily at small angles, smaller than you indicate.

ROBINSON: Yes. An angle of 30^{o} is conservative.

PATE: If you are detecting with a device like this, the back scattering increases with the energy; if you observe this phenomenon in an ionization chamber, the back scattering also increases with the energy. It becomes larger and larger as you observe it in angles closer and closer.

I agree with you if you wish to do alpha counting you should do it in 2π geometry or 2π geometry calibrated to the 4π geometry.

SELIGER: The point I was making was not the fact that you wanted to know exactly how many alpha particles were contained in your source, but in the use of alpha sources it is very interesting to know how many alpha particles are coming out of your source.

PATE: It is just a matter of semantics.

REYNOLDS: I have about three points. We have calibrated and have published an article on said calibration of an alpha 2π counter for alpha disintegration rate measurements and in 2π counting the back scatter effect is closer to 3 percent than the 1 percent that somebody mentioned. In other words, the counting yield for polished platinum is 51.3 to 51.5 percent, according to our measurements.

PATE: This depends on energy.

REYNOLDS: We have not looked into the energy dependence because we are not interested.

PATE: It is considerably more if you go into the 6 or 7 Mev region.

REYNOLDS: Yes.

Second, we have noticed a considerable effect on storage of standard plates for alpha spectrometry, if we store a set of standards in reasonably tight plastic boxes. The resolutions have gradually become poorer. We have been afraid to wash these sources with alcohol. We have flamed them and the resolution is then restored.

ROBINSON: Apparently a lot of people have observed this deterioration. I do not think it has been explained satisfactorily. Any crystals that would grow would be microscopically small. They would be much smaller than any effect you could detect.

REYNOLDS: We think this is probably pump oil or something of the sort, although we have not explained it. It may be probably a combination of effects, lint falling on the samples, pump oil, moisture, what have you.

Third, and one very minor point, we also used some low geometry counters with proportional detectors. The window could be made a milligram per square centimeter which I think was comparable with your total figures.

2. National Bureau of Standards Alpha Standards

R. W. Hayward

I intend to make this very brief report on the alpha standards that are available from the National Bureau of Standards, and try to get suggestions from you on what standards we should add to our stock. At the present time the Bureau puts out two types of standards. One standard is the uranium oxide source with an emission rate of about 10 to 12 alphas per second into the forward hemisphere. I stated it this way, because I have the ground taken right out from under me. We count all our sources in 2π geometry. This is how we specify our activity.

The other sources can range up to strengths of a thousand alphas per second into the forward hemisphere and these are polonium-210 standards. Formerly the Bureau issued radium D plus E plus F. These are lead-210 plus bismuth-210 plus polonium-210 standards, since this is a very convenient way of getting energetic alpha particles with a long half life. Unfortunately, these standards have to be prepared about three years in advance of their calibration because it takes about three years for the emission rate to reach about one-half of a percent of the ultimate equilibrium value.

The demand for these sources has increased so much in the past few years that we have been forced to go to polonium-210 sources, where the polonium itself is electroplated onto palladium. This is really no disadvantage, because the half life of polonium-210 is known to five significant figures. One can really get a massless source this way, while with radium E plus D plus F, one had to plate radium D down from a solution containing lead. One gets far less straggling in the spectrum of the pure polonium source than in the spectrum of radium E plus D plus F where the activities are about the same and distributed over approximately the same area. These are currently the sources that we are issuing from the Bureau. They can be made in practically any specific activity that one desires.

There is one drawback. I am feeling very uneasy at the present time about polonium sources. We have taken some audioradiographs of polonium sources that have been electroplated onto palladium, and looked at the same sources a year later by audioradiographs. These are shown in Figure 1. One can see that in the plating operation the polonium was laid out unevenly but it is still a massless source and this unevenness is of no consequence. But over a period of a year, the polonium has crept and evened itself out a little bit. However, I don't think it is due to free polonium salts, because these sources are quite abrasion resistant. I actually wrapped a source in an envelope and carried it in my pocket for a year. At the end of that time I was able to calibrate it and obtained a half life that corresponded to the accepted half life.

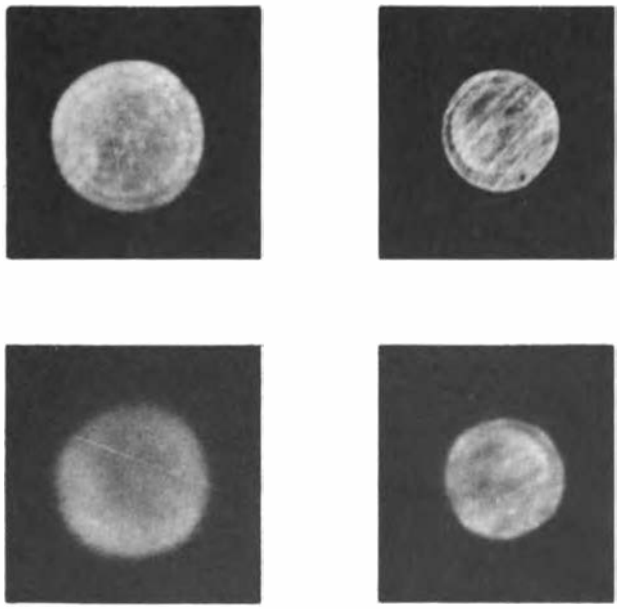


Figure 1. Audioradiographs of polonium sources.

We are currently experimenting with different backings for polonium sources. We are trying at the present time to plate polonium sources on monel, which is quite a corrosion resistant alloy of copper and nickel. The electromotive potential between polonium and copper and nickel is far greater than that of polonium and palladium, and perhaps we may get a more stable source this way.

Another thing we are planning at the present time is to investigate the preparation of americium-241 sources of fairly low activity, let us say up to a thousand DPS, which could be

supplemented by the very high activity sources that you have out in Berkeley. If there are any other sources that would be of value to people, we would like to know about it. Some people like a mono-energetic alpha which is convenient for the checking of counters and energy calibrations. Something like americium-241, which has quite a number of alpha groups on the other hand, is equally acceptable in checking the response of certain detectors to alpha energies.

This is really about all I have to say at the present time on the Bureau program.

ALLEN: May I point out something which is perhaps common knowledge? We used to use polonium sources electroplated onto both platinum and onto silver in a long tube low geometry counter, and after a few months, particularly if the source had been under vacuum, there were up to a few percent of polonium on the walls of the tube.

HAYWARD: Yes.

ALLEN: It might be better with polonium with monel backing. It would be rather interesting to know.

SELIGER: I would like to throw one problem that we have open which is analogous to that which Dr. Hayward brought up with regard to the alpha standards, and the audioradiographs. We have to go through a lot of difficulty to prepare radium D plus E sources, by electrodeposition, and at one time we thought it would be very nice to be able to deposit a small spot of radium D solution and alloy it with the palladium surface of the disk that we used. We, therefore, took a very small spot, in this case lead nitrate, and passed hydrogen through a tube in which the sample was placed at about 500°C, lead was reduced and the critical tables say that a very small amount of lead can go into solution in palladium. This was actually the case. We prepared a radium D plus E source which was extremely abrasion resistant which was essentially a point source, and we were quite happy. We put these away. The size of this point was about a millimeter. About six months later we re-examined these sources and we found that the activity had spread over a centimeter and irregularly, and there seemed to be about three or four times as much solids as we had seen originally. I doubt whether this is a diffusion process, but I assume that possibly this activity has progressed along surface defects in the palladium film which is on the surface. I just throw this open to people who are going to prepare sources this way, that it is rather difficult at the present time to prepare a source which will be stable over a long period of time.

KIPFER: The pictures you showed were pure polonium sources?

HAYWARD: Pure polonium metal.

KIPFER: And the backing of monel was for polonium sources or radium D sources?

HAYWARD: Polonium sources, but we have only made the monel sources two months ago and there is no evidence as yet of creep.

HAWKINGS: I would like to ask Dr. Selinger why he thinks that the spread in the polonium-210 on palladium is not due to diffusion.

SELIGER: I don't think that it is due to diffusion in the sense of atomic diffusion through the material itself. It is, rather, possibly a travel of the material along defects in the surface of the material. I don't know that at room temperature that metals have a diffusion coefficient so long as to give one centimeter traverse in six months.

PATE: I would like to ask if there is any information that exists concerning such phenomena being observed without polonium-210.

SELIGER: I would assume that if one proposes a process by which the polonium creeps, this should not be a particular property of polonium.

PATE: Unless you assume some mechanism which depends on the chemical nature of polonium--volatility of compounds has been brought up. However, if you have a polonium sample sitting under vacuum, it is not going to grow any lines. Yet we observe this phenomenon moving around, and this is the sort of thing we have observed in spectrometry.

SELIGER: I should think that wherever you have a sufficient number of atoms so that they can agglomerate, you will have this process occurring. If you lay a polonium surface down so that there is a fair distance between atoms, and if you do this under high vacuum where your accommodation coefficient is rather low, then you should not observe this. If you do it under a process where you distill your polonium, then you would tend to observe this creeping more.

ROBINSON: We have made autoradiographs of some of our americium-241 sources which are several years old, and there is no evidence of any migration of activity on the surface of the platinum. Has anybody done any work on any other material?

BALAGNA: We have not noticed the contamination problem with polonium. This is a point of information.

ZUMWALT: I remember at a previous subcommittee meeting, I think Dr. Patt and a number of others pointed out it was their experience that the americium-241 standards were very stable as they are prepared. So as far as experimental evidence goes, they would seem to be a good kind of standard from the stability point of view.

PEACOCK: I want to make a comment on the radium E plus D plus F sources at the risk it might be slightly off the subject at this point. I think one of the main purposes of this source was for the purpose of getting the beta rays as well as the alpha rays sources and for having the mixed radiation which is of some convenience in some laboratories. I certainly urge that although it may take time to restock on those sources, that they are available in quantities that are needed. I dare say they will be much more in demand as beta sources. I expect the demand will go up very substantially. They are very handy.

HAYWARD: You need to order a radium D plus E source from the Bureau and you get the alpha sources with them.

CURTISS: I think that Dr. Peacock was worried that you were going to discontinue the radium E plus D plus F sources.

MANN: We discontinued them two years ago.

SELIGER: Not the radium D plus E. You can wait a little while.

PEACOCK: It is plated out. I am satisfied.

HAYWARD: We can do that.

MANN: We are not standardizing the accuracy any more. They are just reference sources. We don't put them out as absolute standards. It is ±5 percent now. As a special favor we will prepare one that is ±2 percent. We normally put them out as ±5.

MANOV: Do you contemplate plutonium alpha standards?

HAYWARD: I see no reason why they could not be prepared very simply.

MANOV: I could foresee quite a demand for that.

HAYWARD: The demand for alpha particle standards has been going up exponentially to our surprise, and we don't really know why.

MANOV: You may need a market survey team. I may give you some answers later if you like.

REYNOLDS: On the matter of plutonium standards, is americium-241 accurate enough? In other words, does one need plutonium as such? The principal alpha energy is 5.5 Mev approximately. In this connection we are very much pleased that the Bureau is contemplating the americium-241 sources. This was suggested, I think, in an earlier meeting of the subcommittee as something very desirable. The americium has the advantage of moderately long half life, not so long as to require the deposition of substantial amounts of material. It also is one of the few alpha emitters which has a substantial gamma emission also, and, therefore, this could be alpha-gamma coincidence counted for accepted standardization independent of low geometry counting.

HAYWARD: We would like to know what alpha standards would be desired that are not presently issued by the Bureau of Standards. For example, plutonium-239 and americium-241, what strengths, and this sort of thing.

PATE: You want alpha standards, do you not, for two separate purposes? You want alpha standards as disintegration rate standards. One also wants alpha standards as standards for line energies. It is desirable to combine both functions in a single source for many experiments. But one usually runs into the trouble that many of the materials which are suited for one purpose are unsuitable for another purpose. Many alpha lines are too close for other purposes. That is where the consideration comes in. We at Brookhaven would certainly welcome the appearance of standardized materials which could combine the two functions if such could be prepared and would be available.

The americium lines are well enough separated and the material is available and it could be laid down; I think it would be very valuable to have it available. The same would apply to any other materials with different line energies that could be secured.

WATTERS: I would like to put in a vote of confidence for the americium samples because of the nice gamma and coincidence figures.

HAYWARD: Yes, it would be possible to calibrate them by independent ways and really arrive at what is the alpha back scattering.

WATTERS: The low energy gamma is a very sweet thing to have around.

ROBINSON: At one time we thought that plutonium-239 would be a nice standard to have around, but there are a couple of questions about it. One is that it is a little hard to get plutonium without some plutonium-241, and you have to worry about this. The second is, I suppose, there is now, and will continue to be, difficulty about exchanging plutonium samples abroad. Is that right?

MANOV: I think I can answer that. With those countries with which the United States has a bilateral agreement for cooperation, that agreement contains provision for getting up to, I believe, 10 grams of plutonium for research purposes. There is at present, however, no mechanism for exchanging small quantities of plutonium for counting purposes without having to go through a lot of red tape. I think it would be extremely useful if this could be removed. I suggest as we get into so-called exempt quantities in this country that we will be able to take this up.

MANN: The same applies to polonium-210. We find our standards tend to get a little mixed up.

MANOV: This is a silly situation but it is absolutely correct. The reason for this is the way in which the Atomic Energy Act was written; namely, that all naturally occurring radioisotopes are exempt from AEC rules, whereas the pile produced ones are not. You have to

decide between tweedle dum and tweedle dee when you separate out polonium-210 from one source or another. It is amazing how the amount of natural polonium-210 has risen to astronomical figures. We hope to get that machinery for interchange of research quantities of polonium as well as other associated purposes.

ROBINSON: One more point in that connection. When we received these samples from Harwell for comparison, these were plutonium samples and the State Department got involved. All this had to be arranged through the State Department because these same samples had to be sent out of the country. There was a lot of red tape involved in getting the plutonium samples in under conditions where we could get them out again. We would certainly like to avoid things like that.

MANOV: We hope we can straighten this out in the future.

ATEN: I would like to put in a word for plutonium standards. Even if you distribute americium samples, most people would use them for measuring plutonium anyway. One could get around the difficulties if the Bureau of Standards, which is best equipped to make standards, would send out instructions for making your own plutonium standards, and possibly having abandoned them for calibration purposes. It would not be essential that they be returned to the countries they came from.

PUTMAN: The longer lived materials such as americium can presumably be measured by more than one method. One can either use counting methods or one can use calorimetric methods and sometimes these materials are wonderful standards for a calorimeter or in the case of half lives of a few hundred years, one could weigh the sample and use accepted figures for half life. After all, a millicurie of americium is something more than four milligrams, I think. So this should be weighable and quite accurate. I wondered whether any intercomparisons of different methods of measurements could be made for these alpha standards if they are proposed. So far we have only been hearing as absolute methods about high and low geometry methods. It seems a pity to rely entirely on one method of measurement when unforeseen snags might arise.

REYNOLDS: We have done alpha-gamma coincidence counting on americium sources which have also been measured by low geometry counting. The results were within our statistical uncertainty in the coincidence counting. So we can say that depending on the source strength, bearing in mind our coincidence equipment demands, that the source be at least as strong as a certain amount and not stronger than a certain other level, the agreement is between a tenth and one percent by two independent techniques of three or four different sources.

MANOV: I would like to see some plutonium alpha standards, even if there are problems connected with them, to match so we have pairs. A plutonium alpha and a plutonium beryllium neutron source would have useful applications. This is worth thinking about, particularly for laboratories in foreign countries that are setting up cascade accelerators or setting up other alpha end measurements--a set of standards like that--relating to the same isotopes would be quite useful.

ZUMWALT: As far as neutron standards go, you can use americium very nicely. The americium pairs would be even better.

3. Radium Standards

W. B. Mann

As the only member of the program committee who was fortunate enough not to have to go to Europe for the whole of September, I have not had time to prepare a formal talk. I grabbed up a few slides last night and some results which have been obtained in our radon testing laboratory and I am, therefore, very pleased that I will have the opportunity, as you mentioned earlier, of being able to edit these remarks.

We recently carried out some intercomparisons of national primary radium standards--Hönigschmid standards.

TABLE 1

Intercomparison of Hönigschmid Standards							
Date	Standards	Electroscope	Radiation balance	Hönigschmid	Ratio of radiation balance ratio to electroscope ratio	Ratio of radiation balance ratio to Hönigschmid ratio	Ratio of electroscope ratio to Hönigschmid ratio
February 1954	A/B	2.441	2.450	2.450	1.003	1.000	0.997
Do	A/D	1.870	1.873	1.870	- - -	- - -	- - -
Do	D/B	1.305	1.308	1.310	1.002	0.998	.996
November 1955	A/G	2.608	2.612	2.617	1.002	.998	.996
Do	A/D	1.870	1.869	1.870	- - -	- - -	- - -
Do	D/G	1.395	1.398	1.400	1.002	.999	.996
December 1955	A/C	1.578	1.583	1.583	1.003	1.000	.997
Do	A/D	1.870	1.875	1.870	- - -	- - -	- - -
Do	C/D	1.185	1.184	1.181	1.000	1.003	1.003
February 1956	A/D	- - -	1.874	1.870	- - -	- - -	- - -
Average	A/D	1.870	1.873	1.870	1.002	1.002	1.000

Table 1 represents the American standard and B is the British standard. Just so you know what they are, we use A, B, C, D. A, American; B, British; C, Canadian; and D, the second United States standard which conforms to the same mnemonic: We refer to it affectionately as the Damyankee. G is the German standard.

We intercompared these standards by means of the gold-leaf electroscope which has been in use at the National Bureau of Standards for a great many years, and also by means of a microcalorimeter (the so-called radiation balance).

In the last three columns are the ratios of the ratios of the standards to each other obtained by the three different methods of measurement (including Hönigschmid's weighings). You notice all ratios in the radiation-balance-to-electroscope-ratio column tend to be high. They are on the borderline of our precision with deviations from unity of the order of 0.1 or 0.2 percent. There is a definite tendency for these ratios to be greater than unity. The ratio of the Canadian to the second United States standard is unity but these standards are approximately equal so that the self-absorption will be about the same.

On the other hand, the ratios of the radiation balance to Hönigschmid readings show a fairly

close proximity to 1.000, at least to within the precision of the measurements. If you take the ratios of electroscope to Hönigschmid weighings then the ratios are all less than unity. So it looks as though we are running into trouble with source self-absorption and that the micro-calorimeter is the only way of getting a really good comparison between these standards which does, in fact, agree with Hönigschmid's own figures.

Table 2 will show the best masses that we deduced.

TABLE 2

Values for Mass of Hönigschmid Standard					
Standard	A	B	C	D	G
Hönigschmid's mass	38.23	15.60	24.15	20.45	14.61
Mass derived from radiation balance and Hönigschmid	38.23_7 38.24_2 38.22_5	15.60_5 - - - - - -	- - - 24.15_4 - - -	20.43_1 20.42_1 20.45_0	- - - - - - 14.62_2
Mass derived from electroscope and Hönigschmid	38.21_6 38.21_6 38.22_3	15.62_8 - - - - - -	- - - 24.17_7 - - -	20.44_2 20.44_1 20.44_5	- - - - - - 14.63_3

As Mr. Perry pointed out, these are "Hönigschmid milligrams" because Hönigschmid never compared his weights with the standard kilogram in Paris. They were calibrated, however, in terms of each other. So, for what it is worth, we agree with Hönigschmid insofar as the mass ratios are concerned.

When you get down to the very great precision that Dr. Kipfer has in his very beautiful method, one in 10, 000, I have a feeling that we are going beyond the point of reason with these radium standards. In fact, I hope never to measure a radium standard again! The question, which I think Dr. Hayward is going to take up tomorrow in the gamma-ray session, is whether or not it would be more sensible to try to make a break from radium and go to cobalt-60. Radium is very much tied to the medical therapy, but radium clinically is being replaced by cobalt-60, and it might be best to make the measure ments in terms of disintegrations per second.

Dr. Hayward has already carried out a very nice comparison with Harwell and Chalk River in which I believe they used the same method of dissolving an approximately 30-millicurie source and measuring an aliquot by gamma-gamma coincidence counting.

At Geneva last year at the meeting of Committee I of the International Commission on Radiological Units, we decided to compare cobalt-60 standards internationally. They were to be sheathed in glass or metal-plated. This was left for National Physical Laboratory and National Bureau of Standards to work out. I am afraid with all our other diversions, especially for Dr. Hayward, who is going to try to look into this matter, we have not gotten around to it. We have a deadline of next August by which to do it.

The last thing I wanted to mention, is that we have recently prepared some radium solution standards. Even though solid radium standards may have their limitations and are awkward things to work with, we do have to have solution standards for the assay of ores and so forth by the radon method.

We recently used the calorimeter to prepare some standards which we call our 1957 series of radium solution standards. It may interest you to know the order of precision we have obtained. We had a special radium source prepared by the Radium Chemical Company in a

glass tube of approximately the same size of crystals in order that we could also make a gamma ray comparison using the National Bureau of Standards electroscope. By spreading the source along the length of the tube by tapping we could make a gamma ray comparison which would not be too subject to salt self-absorption corrections. With the microcalorimeter we obtained the following values for the rate of energy emission:

October 23, 1956	914.8_4 uw
November 1, 1956 . . .	914.8 uw
November 8, 1956 . . .	914.6_3 uw
November 28, 1956 . .	914.0_8 uw

This corresponded to 6.10_8 mg of radium element compared with 6.08 mg of radium element obtained by comparison with the primary standards using the N. B. S. electroscope.

We put these standards into solution and measured them by the method of radon analysis. We prepared some 10-microgram radium solution standards and we also prepared some 10^{-9} and 10^{-11} gram radium solution standards. We compared these with our 1947 10-microgram standards and also our 1940 10-microgram standards and got good agreement within the stated errors.

Measurements between the 1940, 1947, and 1957 10-, 20-, and 50-microgram standards were now carried using the National Bureau of Standards 4π beta-gamma ionization chamber and between the 1940 and 1957 10^{-9} gram standards. The initial results may be summarized by saying that, to within the precision or accuracy claimed for the various series of standards, there was agreement between the 1940, 1947, and 1957 microgram series of standards. These measurements were made by Mr. Garfinkel. In the case of the 10^{-9} gram standards, however, the ratio of 1957/1940 came to about 2.5 percent less than unity. This result was so surprising that we decided to check our procedures by taking six of the 1947 10-microgram standards, diluting these down, by two different routes, to 10^{-9} grams and comparing these with the 1940 10^{-9} gram standards. These dilutions were made by Mr. Schwebel and Mr. Stockmann and the comparison carried out by the latter and Miss Mullen. Once again we got approximately the 2.5 percent discrepancy. The details of our experiments will shortly be published but the final result for the 1957/1940 ratio of 10^{-9} gram standards is 0.9740. Our conclusion is that an error must have occurred in the dilution down to 10^{-9} gram standard in 1940 as we cannot detect any 2.5 percent disagreement between the microgram standards for these years.

As I have said there was good agreement between the electroscope and calorimeter. The calorimeter is shown in Figure 1. It can sit on a table covered with black paper but is rather sensitive to fluorescent light and to drafts. You can get of the order of half percent precision. If you put it in a temperature-attenuating enclosure so that it does not follow the temperature of the room so rapidly, you can get a precision of 0.13 percent. It is based on the Peltier cooling effect. You put a radium source, say, in the left cup and a dummy in the right. You compare the temperature by means of the six thermocouples, and you balance by passing a cooling current in such a way as to cool the left-hand cup and heat the right. You then interchange source and dummy and reverse the current, and quite simply the microwatts come out to be equal to twice the Peltier current times the Peltier coefficient. This is the calorimeter with which we have measured this new source in terms of the United States primary radium standards. We have measured against the two primary standards and we will be accepting the microcalorimetric value for this new series.

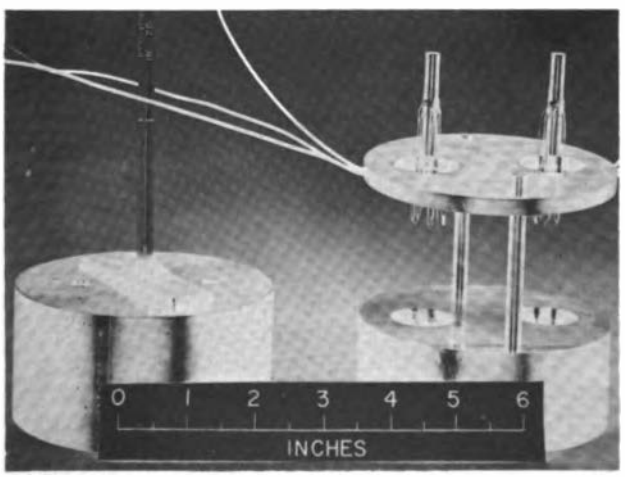


Figure 1. Standardization calorimeter.

ROBINSON: You mentioned these dilution errors. Do these errors result from adsorption?

MANN: It may be simply an error in calibration. We have no idea. It was carried out by someone who was at the Bureau and is no longer at the Bureau. We are not quite clear what the procedures were. Where you have to dilute so many times--there are about four dilutions in the chain--it is possible to introduce some error through calibration or lack of calibration.

PEACOCK: Do I understand that the error is in the 10^{-9} gram 1940 standard but as far as you know it is not in the 10 microgram standard?

MANN: Yes. We have some high activity standards from each year.

ATEN: Is there a special technique for avoiding absorption errors?

MANN: We use a barium carrier in acid solution. We believe we have no absorption errors.

ATEN: How much barium is put in and what acid solution, roughly?

MANN: Actually, Mr. Schwebel did this part, and I don't have the figures here. I can tell you later. It was hydrochloric acid and in 1947 nitric acid. Apparently the hydrogen ion acts as well as barium as a carrier. It prevents adsorption.

PERRY: I would like to give one or two figures of comparisons of NPL solution standards in the region of 10^{-9} curies with NBS 10^{-9} curie standards. We make two of our 10^{-9} standards different by one percent. We differ by one percent from the NBS value for one of their standards that we have; that is, we would make it 0.991 instead of 1. With the second 10^{-9} standard we only have two of these NBS standards. We would call it 0.979, instead of 1. We would say NBS was one percent lower than we are, or we are one percent higher than NBS, and two percent higher in the other standard.

MANN: This is worse, because we say our 1940 standards are not so active.

PERRY: We don't claim precision better than ± 1 percent on these measures. We have done this by the integration technique and alpha particle count technique. These differences, perhaps with the exception of the two percent I mentioned, are within the limits of our precision.

MANN: I should mention for the record that Mr. Stockmann and Dr. Youden have done a very beautiful job in arranging these experiments on radon. We have gotten a precision of 0.8 percent.

CURTISS: That is remarkable for radon measurements.

MANN: Yes. It was first done by the method of bubbling nitrogen through sintered glass filters and removing the radon. We reinstalled reflux condensors and got identical readings. It was a very thorough job.

CURTISS: I would like to say this about Hönigschmid standards, because every time I hear the name of Hönigschmid mentioned, I am uncomfortable. Dr. Hönigschmid actually determined the atomic weight of radium and had at his disposal five grams of radium. He was not interested in preparing radium standards, but he prepared very high purity samples of radium and redetermined the atomic weight and got essential agreement, and only slight improvement with previous values. He had this large quantity of very pure radium. He had to tube it up and return it to its owners, which he did. I don't think at any time he expected that it would be used as radium standards. I don't think he used any of the precautions that you ordinarily would use, but he tubed it up properly and returned it to the owners. The owners after some time had this five grams of very pure radium in terms of 100 and 200 mg and lesser amounts of radium. They wanted to get rid of it. They didn't want to put it all on the market as just ordinary medical

preparations. So they decided to distribute it as standards, subject to calibration by the International Radium Commission by the electroscope method. That is how these standards got on the market and you may find the discrepancy in weight in these standards which I think you will inevitably find.

MANN: I don't think there are any discrepancies in the weight, Mr. Chairman. I think the weight ratios are in extremely good agreement with Hönigschmid's results. You find that in very few cases are they outside the precision of the experiments that we are carrying out.

If I might add one further remark, there was some suggestion that we might take the residue of Hönigschmid's atomic-weight material and prepare a new set of radium standards. But I am "agin" it! I feel that radium has had its day!

CURTISS: All right. We will have your slide.

GEIGER: We just had the opportunity two years ago to acquire one of those Hönigschmid standards due to the kindness of the various laboratories. It could be prepared with the Hönigschmid standard and used with the former Paris and Vienna standards. The results are given in Table 3.

TABLE 3

Preliminary Values for New Canadian Primary Standard No. 5425 as of June 2, 1934

	Weight mgm Radium	No. 5425 Measured Value mgm	% Deviation from Weight	Remarks
#5425 Hönigschmid's weighting	24.153	------	-----	------
#5426 (German) at PTB	14.608	24.125	-0.11	Corrected for absorption
#5426 (German) at NRC	14.608	24.141	-0.05	Corrected for absorption
#5428 (Vienna) at Inst. f. Radium f.	23.407	24.123	-0.12	No correction necessary
#5330 (Paris) at Union Miniere	16.922	24.125	-0.11	Not corrected
#5432 (U. K.) at NPL	15.605	24.125	-0.11	Not corrected
#5437 (U. S.) at NBS	38.228	24.144	-0.04	Radiation balance
#5440 (U. S.) at NBS	20.446	24.216	+0.26	Radiation balance
#5440 (U. S.) at NRC	20.446	24.239	+0.36	Corrected
Former International at Union Miniere	16.573	24.178	+0.10	Not corrected
Former Vienna at Union Miniere	23.492	24.248	+0.40	No correction necessary
Former Canadian at NRC	24.256*	24.161	+0.03	Corrected

*This is a measured value as certified by the International Radium Standards Commission.

On the very top is the Hönigschmid weighing of our standard of 24.153 milligrams of radium. Then the measured weights by these various laboratories. In the third column, you see the percentage deviation from weight. Except for two cases this is better than 0.2 percent, which I think is very good. If you read Hönigschmid's paper, he really does not claim that he could weigh it better than 0.3 percent. In most cases absorption corrections have been applied as far as feasible when ionization chamber measurements were made.

KIPFER: I would add that I compared some of Hönigschmid's standards deviations of the same order of magnitude, about 0.2 and 0.3 percent.

MANN: There is the problem, Mr. Chairman, that these standards may be getting a little

dangerous now. We have to worry about them. We have now encapsulated our own at the Bureau in hermetically sealed containers. They can be radon tested. The air inside can be tested for radon. I am worrying about the permanence of these standards.

CURTISS: Is it not true that over a period of some years very high pressures can develop in these tubes? I think it is remarkable that so few accidents have occurred.

MANN: I believe that Hönigschmid took extreme precautions to make sure they were not damp. So they may be all right.

CURTISS: Small amounts of moisture can cause these things to blow up. But even without that, apparently over a period of some 30 or 40 years pressure does develop. It may not be due entirely to dissociation of water.

NOVEY: Has anybody ever opened any of these sealed standards?

CURTISS: Similar tubes were opened in Vienna, and the pressure was fairly high. It made quite a disturbance when it was broken.

KIPFER: I would ask if standards of 1911 are still living.

CURTISS: I don't know. There may be slight differences in preparation.

GEIGER: I may say that I think the radium in glass has the longest life time. We have a large number of encapsulated sources, and they don't seem to live more than 15 or 20 years. The glass sources definitely live much longer.

CURTISS: It is more difficult, I think, to completely seal a metal preparation. I think there is no doubt about that. But when it comes to an explosion, the metal is quite a protection.

SESSION II - BETA MEASUREMENTS AND STANDARDS

W. B. Mann, Chairman

Summary

The second session is devoted to a discussion of "Beta Measurements and Standards," the discussion being initiated by two papers by Dr. Peter J. Campion and Dr. Howard H. Seliger.

The paper by Dr. Campion is entitled "4π Beta Properties" but deals essentially with the theory of 4π beta-gamma coincidence counting. New equipment that has been built at the Atomic Energy of Canada Limited Chalk River Laboratories for 4π beta-gamma coincidence counting is also described. The discussion that follows is concerned mainly with the questions of source preparation, source self-absorption and with the possible use of liquid scintillators in place of sodium iodide crystals.

The paper by Dr. Seliger is on "Liquid Scintillation Counting." The author at first extends some of the comments on the previous paper dealing with the question of 4π beta counting. In the scintillation counting paper itself he first describes experiments with the method of 4π crystal scintillation counting and then proceeds to discuss liquid scintillation counting and the effects of wavelength shifters, oxygen quenching and thermal quenching. By attention to these factors the speaker and his colleagues have been able to produce pulses in a liquid scintillator that are at least as good as using anthracene in the 4π crystal scintillation counter. The initial discussion on this paper is concerned with scintillation counting but the session finishes with more general discussion on the problems of 4π beta source preparation.

1. 4π Beta-gamma Coincidence Counting

P. J. Campion

It is generally accepted that the method of 4π beta counting is the most universally applicable for the standardization of artificial radio-isotopes. The fundamental phenomenon which limits the accuracy of this method is that of self-absorption. The beta-gamma-coincidence method is in principle independent of the efficiency of either detector and hence free from any self-absorption correction. However, this merit is often outweighed by the uncertainties in several corrections which must be applied before the true disintegration rate can be determined. The method of 4π beta-gamma-coincidence counting is, of course, nothing new but I would like to review it here, and attempt to demonstrate that the corrections are well defined, by which I mean readily calculable, and that accuracies of the order of 0.1 percent can be achieved by this method.

I will start, therefore, by enumerating the various corrections which may apply to 4π beta-gamma-coincidence counting. Perhaps the most obvious advantage of this method is the fact that the correction due to any angular correlation betweeen the beta and gamma rays is greatly reduced. For 100 percent efficiency the correction is obviously zero. Further, it is well known that the preparation of sources by the evaporation of aliquots of active solutions generally yields deposits consisting of local aggregates as opposed to uniformly extended deposits. It is reasonable to assume, therefore, that to a first approximation the self-absorption will be isotropic and hence there is no correction required even for efficiencies of less than 100 percent of the 4π beta counter provided that this loss is due to self-absorption.

The next correction we will consider is that of possible internal conversion of the gamma ray following beta decay. Again it is clear that if the beta detector is 100 percent efficient and the internal conversion process follows the beta decay in a time short compared to the resolving time of the mixer circuit, the conversion electrons add nothing to the beta-channel counting rate. In the case that the efficiency is less than 100 percent a correction must be applied to take into account those conversion electrons which produce a signal in the beta detector when the associated beta particle was not observed. The relevant equations omitting dead-time corrections are the following:

$$N_\beta = N_O\left[\epsilon_\beta + \frac{\alpha}{1+\alpha}(1-\epsilon_\beta)\epsilon_{ce}\right]$$

$$N_\gamma = N_O \frac{\epsilon_\gamma}{1+\alpha}$$

$$N_c = N_O \frac{\epsilon_\beta \epsilon_\gamma}{1+\alpha}$$

where $\underline{N}_\beta$, $\underline{N}_\gamma$ and $\underline{N}_c$ represent observed counting rates in the three channels respectively, α is the total internal conversion coefficient and ϵ_β and ϵ_γ the over-all efficiencies of the beta and gamma channels respectively, and ϵ_{ce} is the efficiency for detecting conversion electrons. Since these are mono-energetic ϵ_{ce} is generally indistinguishable from unity. The small value of $(1 - \epsilon_\beta)$ coupled with the usually small value of α considerably reduces the magnitude of this correction. As an example we may take the case of mercury-203 for which the total internal conversion is about 20 percent and, because it has a low beta ray energy and therefore considerable self-absorption, the efficiency of the beta detector may be only 95 percent. Thus the correction amounts to one percent.

Another correction to be considered is the gamma sensitivity of the beta detector and possible gamma-gamma coincidences. For those isotopes which have a single gamma ray transition and provided we make the assumption that the gamma ray interaction giving rise to a signal in the beta counter cannot produce a coincidence, the equation may be written as

$$\frac{N_\beta N_\gamma}{N_c} = N_O \left[1 + \frac{(1 - \epsilon_\beta)}{\epsilon_\beta} (\epsilon_\beta)_\gamma \right]$$

where $(\epsilon_\beta)_\gamma$ is the gamma sensitivity of the beta detector.

If, on the other hand, such coincidences can occur or if a second gamma ray is present in the spectrum, we have the following set of coincidence equations:

$$N_\beta = N_O \left[\epsilon_\beta + (1 - \epsilon_\beta)(\epsilon_\beta)_\gamma \right]$$

$$N_\gamma = N_O \epsilon_\gamma$$

$$N_c = N_O \left[\epsilon_\beta \epsilon_\gamma + (1 - \epsilon_\beta)\epsilon_c \right]$$

whence

$$\frac{N_\beta N_\gamma}{N_c} \approx N_O \left[1 + \frac{(1 - \epsilon_\beta)}{\epsilon_\beta} \left\{ (\epsilon_\beta)_\gamma - \epsilon_c/\epsilon_\gamma \right\} \right]$$

where ϵ_c is the probability of detecting a coincidence when the beta particle is undetected.

We have measured these values as a function of gamma ray energy by covering sources of known activity with polythene sufficiently thick to absorb all the beta rays, and observing the response of the beta counter.

Figure 1 shows the results for $\epsilon_{\beta\gamma}$ as a function of energy. We have used cobalt-60 as one point since the two gamma rays have approximately the same energy. The other four isotopes all have single gamma rays in their decay.

The characteristics of this curve are very similar to those obtained by earlier workers who used Geiger counters and external sources. In our case the beta counter was constructed of aluminum.

The results for ϵ_c are shown in Figure 2 where the quantity $\epsilon_c/\epsilon_\gamma$ is plotted against ϵ_γ for various gamma ray energies. The efficiency of the gamma channel was varied by adjusting the width of the window of the single-channel pulse-height analyzer, the largest efficiency being obtained when the window was set to include almost all the pulse height spectrum. The value of $\epsilon_c/\epsilon_\gamma$ decreases very rapidly as the window is narrowed down to include only the photopeak, as is to be expected.

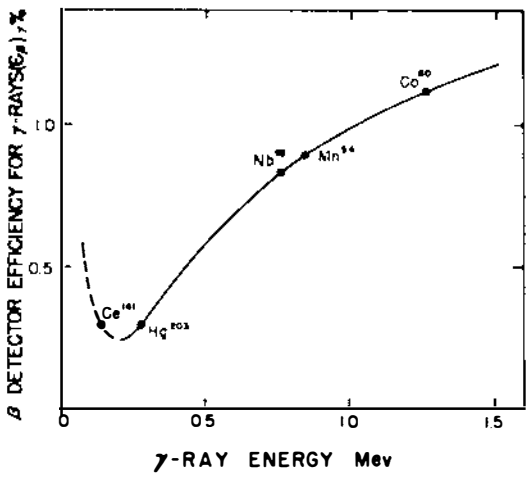


Figure 1. Beta detector efficiency for gamma rays.

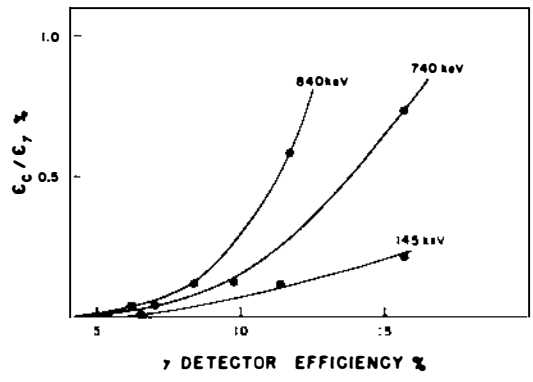


Figure 2. Coincidence counting efficiency.

From these two graphs it can be seen that the term $(\epsilon_{\beta\gamma} - \epsilon_c/\epsilon_\gamma)$ is small indeed and thus the correction becomes negligible for most practical purposes. It should be emphasized that the results shown in these figures are expected to depend on the experimental conditions and thus are strictly applicable only for the instrument with which they were measured. They do serve to show, however, the orders of magnitude involved.

SELIGER: For all practical purposes $\epsilon_c = 1 - \epsilon_\beta$.

CAMPION: No.

PATE: Times some coincidence efficiency.

SELIGER: All right.

CAMPION: Yes. The walls of the beta and gamma counter are sufficiently thick to stop the entry of beta particles into the gamma detector, but it is possible that the lower bias of the pulse height analyzer is set at sufficiently small value to allow the detection of bremsstrahlung quanta in the gamma detector. This condition will obviously occur when the maximum beta particle energy is greater than the succeeding gamma ray energy or if there is a branching to the ground state.

WATTERS: Would you say what you just said again?

CAMPION: Maybe I had better read it.

WATTERS: No, the last statement. In which case will you detect the bremsstrahlung in your gamma detector?

CAMPION: If the maximum beta ray energy is greater than the succeeding gamma ray energy, you could possibly pick up a bremsstrahlung quantum. Since the source in the 4π beta counter is usually mounted on a thin film, and is itself thin, the only beta particles which are not detected are those having very low energy, and which thus give rise to extremely weak bremsstrahlung quanta which are not recorded in the gamma counter. It follows that any quantum so recorded must be associated with the beta particle which produces a signal in the beta detector, and this type of event serves to increase the over-all efficiency of the gamma detector and requires no correction.

I will now turn to complex decay schemes. Mr. Putman has shown that the coincidence method is valid for isotopes with several beta branches provided that the efficiency of either the beta or gamma detector is the same for all branches. The use of 4π geometry is the obvious approach to this condition and it is instructive to take a simple hypothetical case in order to examine the magnitude of the correction involved. Suppose, therefore, we have an isotope which decays by two beta branches of fractional intensity a and b, such that $a + b = 1$. The coincidence equations and the counting rates in the three channels (again neglecting dead-time losses) for this situation are the following:

$$N_\beta = N_O(a\,\epsilon_{\beta_a} + b\,\epsilon_{\beta_b})$$

$$N_\gamma = N_O(a\,\epsilon_{\gamma_a} + b\,\epsilon_{\gamma_b})$$

$$N_c = N_O(a\,\epsilon_{\beta_a}\epsilon_{\gamma_a} + b\,\epsilon_{\beta_b}\epsilon_{\gamma_b})$$

whence

$$\frac{N_\beta N_\gamma}{N_c} = N_O\left[1 - \frac{ab(\epsilon_{\beta_a} - \epsilon_{\beta_b})(\epsilon_{\gamma_a} - \epsilon_{\gamma_b})}{(a\,\epsilon_{\beta_a}\epsilon_{\gamma_a} + b\,\epsilon_{\beta_b}\epsilon_{\gamma_b})}\right]$$

Here ϵ_{β_a} and ϵ_{β_b} are the efficiencies of the beta detector for branches a and b respectively while ϵ_{γ_a} and ϵ_{γ_b} are similar quantities for the gamma detector. These gamma efficiencies may include the contributions from several gamma rays in each branch. It can be clearly seen that the correction for N_O is zero if $\epsilon_{\beta_a} = \epsilon_{\beta_b}$ or $\epsilon_{\gamma_a} = \epsilon_{\gamma_b}$ as was pointed out by Mr. Putman.

An estimate of the maximum value of this correction can be made as follows:

The efficiency of the beta detector is determined primarily by the source mount and source self-absorption. The former can usually be made small compared with the latter by the use of thin plastic films rendered conducting by a vacuum-evaporated metal coating. Below about 0.3 Mev self-absorption effects become appreciable. Dr. Seliger has reported a 10 percent effect for cobalt-60 and at Chalk River we have observed efficiencies as low as 85 percent. Taking the most unfavorable case, namely, maximum self-absorption in one branch and a negligible amount in the other, we have $\epsilon_{\beta_a} - \epsilon_{\beta_b} = 0.15$.

Since the calculated efficiencies of sodium iodide crystals vary by little more than a factor of two over the range of gamma ray energies encountered in radioactive isotopes, it may be assumed for this purpose that the efficiency associated with the higher energy beta branch is 10 percent while that with the lower energy branch is 20 percent and hence, $\epsilon_{\gamma_a} - \epsilon_{\gamma_b} = 0.10$.

It should be noted that if cascading occurs with one or more gamma rays common to both branches, as is very often the case, the difference between these two efficiencies can be made much less. Finally the maximum value of ab is 0.25, for a = b = 0.5. Thus the correction in this rather extreme example amounts to about 2.6 percent. For all practical cases the correction is smaller by at least an order of magnitude and can generally be ignored for routine work. The most pessimistic values for the corresponding efficiencies, in the case of gold-198, lead to a correction of .01 percent. Thus the serious restriction of the beta-gamma coincidence method to simple decay schemes is largely removed by the use of 4π geometry in the beta detector.

We shall now turn our attention to those corrections which are functions of the counting rate, that is to say, the accidental coincidence rate and the dead-time correction. Owing to the finite resolving time of the mixer it is always possible to obtain a coincidence pulse from two unrelated events unless both detectors are 100 percent efficient. An expression for this accidental rate may be derived as follows:

It is assumed that the background rates are negligible, which is well justified for those instances in which the accidental rate is important. The probability of detecting an event in the beta counter, for example, when the associated gamma ray is not recorded in the gamma detector is $\epsilon_\beta(1 - \epsilon_\gamma)$. The probability that a second event will occur and be recorded in the gamma detector (the beta channel being dead due to the previous event) within τ_R, the resolving time of the mixer unit is $N_O \tau_R \epsilon_\gamma$. Hence the accidental rate due to this type of event is

$$N_O^2 \tau_R \epsilon_\beta \epsilon_\gamma (1 - \epsilon_\gamma)$$

A similar expression obtains for the type of accidental coincidence in which the first event is recorded in the gamma detector and the second in the beta detector. Thus, the total random rate N_{Acc} is given by the sum of these two rates

$$N_{Acc} = \tau_R N_O^2 \epsilon_\beta \epsilon_\gamma (2 - \epsilon_\beta - \epsilon_\gamma)$$

The sum of the efficiencies of ϵ_β and ϵ_γ lies close to unity. Hence, for a disintegration rate of 10^4 dps and a resolving time of 7×10^{-7} sec the accidental coincidence rate is about 0.7 percent of the true coincidence rate.

For the dead-time correction, we shall assume for simplicity that the dead-times of both channels are the same and equal to τ, and that there is no dead-time associated with the coincidence channel greater than τ. Further, the absolute disintegration rate of the source N_O is assumed to be such that the probability of two events occurring within a time τ can be neglected

compared with the probability of occurrence of one event, i.e., $(\underline{N}_O\tau)^2 \ll N_O\tau$.

The probability of no event occurring in an interval is thus $(1 - N_O\tau)$ and the probability of detection in the beta channel, for example, is therefore

$$\epsilon_\beta\left[(1 - \underline{N}_O\tau) + \underline{N}_O\tau(1 - \epsilon_\beta)\right] = \epsilon_\beta(1 - \underline{N}_O\,\epsilon_\beta\,\tau)$$

The first term in the square brackets expresses the chance that there was no event in the preceding interval τ while the second term is the chance that there was, but it went undetected owing to the inefficiency of the beta counter. Within the limits of the initial assumption this expression for the dead-time correction is the same as that generally adopted, which in this notation would be $(1 - \underline{N}_\beta\tau)$.

By extension of the argument to the coincidence channel and remembering that a previous count in either channel occurring within time τ will block the coincidence channel, we can write for the probability of detection

$$\epsilon_\beta\,\epsilon_\gamma\left[(1 - \underline{N}_O\tau) + \underline{N}_O\tau(1 - \epsilon_\beta)(1 - \epsilon_\gamma)\right]$$

where the second term is the probability that there was an event in time τ which was undetected by either counter. We have, therefore, the following equations:

$$N_\beta = N_O\,\epsilon_\beta(1 - N_O\epsilon_\beta\,\tau)$$

$$N_\gamma = N_O\,\epsilon_\gamma(1 - N_O\epsilon_\gamma\,\tau)$$

$$N_c = N_O\,\epsilon_\gamma\left[1 - (1 - \Delta)N_O\,\tau\right] \qquad \text{where } \Delta = (1 - \epsilon_\beta)(1 - \epsilon_\gamma)$$

whence, since $(N_O\tau)^2 \ll N_O\tau$

$$\text{i.e.,} \quad N_O \approx \frac{N_\beta N_\gamma}{N_c}\,\frac{1}{(1 - N_\gamma\,\tau)} \qquad \text{for } \epsilon_\beta \to 1.0$$

* * *

$$R = \frac{1 - N_O\,\tau\,\epsilon_\beta\,\epsilon_\gamma}{1 - N_\gamma\,\tau}$$

If now we make $\epsilon_\beta = 1$ the dead-time correction for the coincidence channel is just that for the beta channel, and the correction appearing in the usual expression for $\underline{N}_O$ is that for the gamma channel. Further, no appreciable error is introduced if we adopt the more usual expression for the dead-time correction. In order to obtain a quantitative estimate of the error thus introduced, I have calculated the ratio,

$$R = \frac{1 - N_O\,\tau\,\epsilon_\beta\,\epsilon_\gamma}{1 - N_\gamma\,\tau}$$

for a range of values of $\underline{N}_O\tau$, ϵ_β and also ϵ_γ. The results are tabulated in Table 1 which it is seen that, for the range of parameters chosen, the value of $\underline{R}$ differs from unity by about 0.1 percent for the most unfavorable case in which ϵ_β equals 0.8 and ϵ_γ equals 0.2.

TABLE 1

Values of the Ratio R for various values of $N_O\tau$, ϵ_β, and ϵ_γ						
$N_O\tau$	$\epsilon_\beta = 1.0$ $\epsilon_\gamma = 0.1$	$\epsilon_\beta = 0.9$ $\epsilon_\gamma = 0.1$	$\epsilon_\beta = 0.8$ $\epsilon_\gamma = 0.1$	$\epsilon_\beta = 1.0$ $\epsilon_\gamma = 0.2$	$\epsilon_\beta = 0.9$ $\epsilon_\gamma = 0.2$	$\epsilon_\beta = 0$ $\epsilon_\gamma = 0$
0.03	1.0000	1.0003	1.0006	1.0000	1.0006	1.001
0.02	1.0000	1.0002	1.0004	1.0000	1.0004	1.000
0.01	1.0000	1.0001	1.0002	1.0000	1.0002	1.000
0.005	1.0000	1.0001	1.0001	1.0000	1.0001	1.0002

One can go through a similar argument as I have outlined here and show that even for dead-times which are not equal--we have stipulated that the dead-time of both channels must be the same--if now you say that τ_β, where τ_β is now the dead-time of the beta channel, is not equal to τ_γ, one can show that the same equation holds provided you make τ_β greater than τ_γ. In other words, it is the dead-time of the beta channel which is determining the dead-time in the coincidence channel. This cancels out leaving you the dead-time of the gamma channel as the only correction involved.

As a check on the validity of these counting-rate-dependent corrections, we have carried out two experiments. But first I should briefly describe the 4π beta-gamma apparatus used. The beta detector is a flow type proportional counter while the gamma detector consists of two 3-inch by 3-inch sodium iodide crystals. A sketch of the instrument and shielding is shown in Figure 3. A more detailed drawing is given in Figure 4.

The beta counter is a conventional pillbox design but in order to avoid regions of weak electric field strength two shaped lucite pieces whose surfaces have been rendered conducting have been placed in each pillbox as shown. This approximates cylindrical geometry. The two pillboxes are separated by a turntable on which source mounts may be placed and rotated into the counter. A gas tight seal is effected by two "O" rings. The two sodium iodide crystals are mounted symmetrically above and below the beta detector and are optically coupled to Dumont 6363 phototubes.

A block diagram of the electronic circuitry is shown in Figure 5. The outputs of the photomultipliers are each fed via amplifying stages to single channel pulse height analyzers, and the outputs of these are then added before being fed to the coincidence mixer. This unit has continuously variable dead-time controls on each input channel. The resolving time currently used is 0.7 microseconds and the dead-time is 2.0 microseconds.

WATTERS: This 4π amplifier, since you qualify it as nonlinear, I hope is also qualified as nonoverloading.

CAMPION: Yes.

We will return now to the experiments to verify the counting-rate-dependent corrections. The first of these consisted of following the decay of a source over as great a range of activities

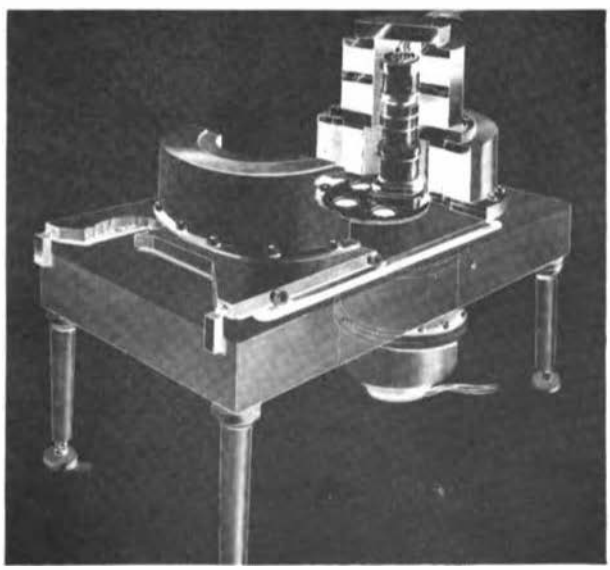


Figure 3. 4π beta-gamma coincidence counter.

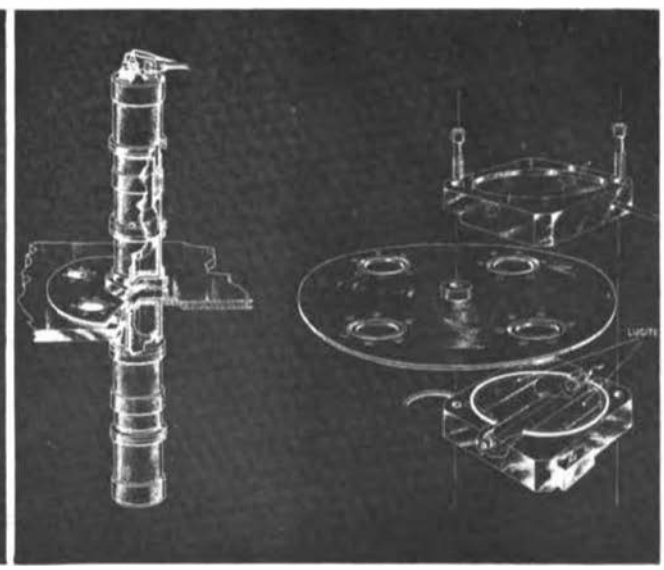


Figure 4. 4π beta-gamma coincidence counter.

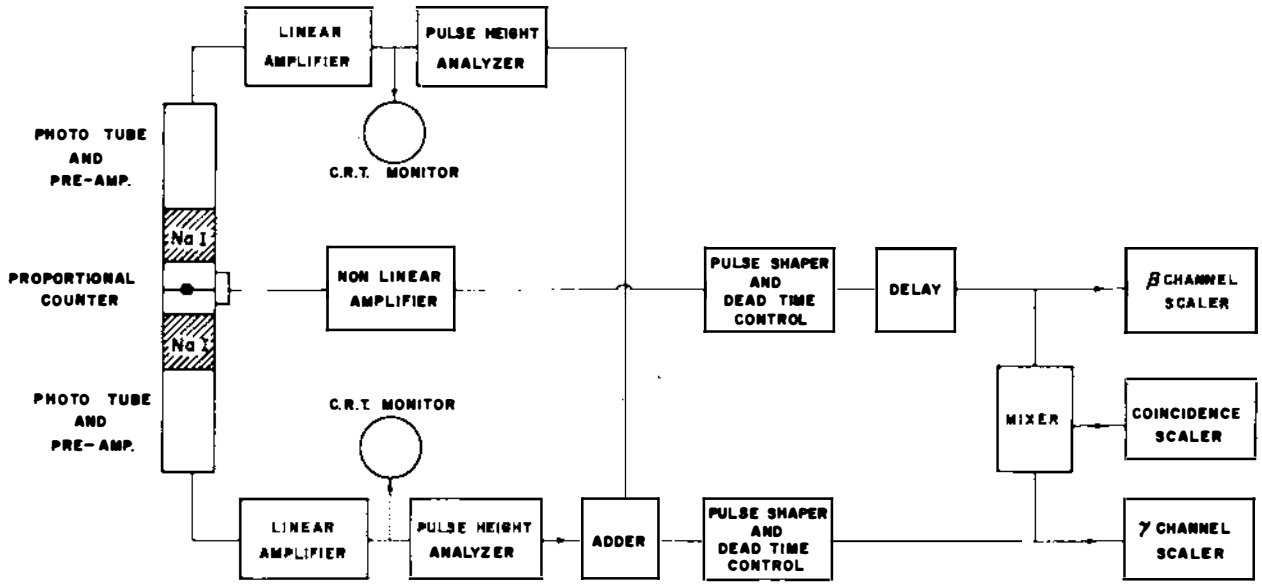


Figure 5. Electronic circuitry for 4π beta-gamma coincidence counter.

as practicable. For this experiment we chose sodium-24 since its large maximum beta ray energy enables sources to be made whose efficiencies are close to 100 percent, for which the expression that was shown on an earlier slide is exact. It has a sufficiently short half life so that the observations may be completed in a reasonable time. Further it may be readily purified by ion-exchange techniques. However, the half life must be well known and although several measurements of this quantity appear in the literature it was felt that a more accurate measurement could be made. The result of this determination was 14.96 plus or minus 0.01 hours.

A source of about 20,000 dps of sodium-24 was prepared and the absolute activity of this source, corrected to a zero reference time, was measured in the 4π beta-gamma instrument as a function of elapsed time from zero. The results are shown in Figure 6.

Curve (a) was obtained when both dead-time and accidental-rate corrections are included in the calculations. Curve (b) was obtained without including the dead-time correction, while curve (c) is the result when neither correction is taken into account. It is gratifying to note that the absolute activity is essentially independent of the time of observation and therefore the counting rate, provided that both the dead-time and accidental-rate corrections are included.

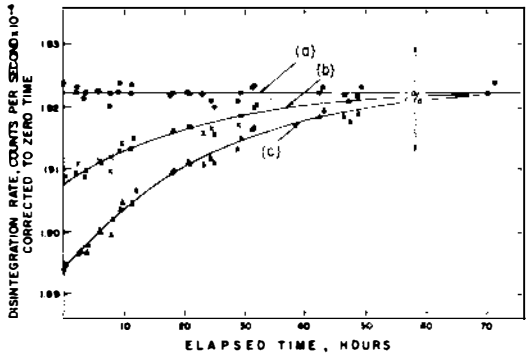


Figure 6. Effect of counting rate dependent corrections for sodium-24.

The second experiment does not rely so heavily on an accurate knowledge of a half life. The absolute activity of a bromine-82 source, free from any bromine-80 activity, was observed as a function of the efficiency of the gamma detector. This was achieved by varying the width of the pulse height analyzer windows and since there are several gamma rays emitted per disintegration of this isotope, the gamma detector efficiency could be varied from 2 to 40 percent.

The results including all corrections are shown by curve (a) of Figure 7. While curve (b) shows the result if the dead-time correction is not included. This is asymptotic to curve (a) at

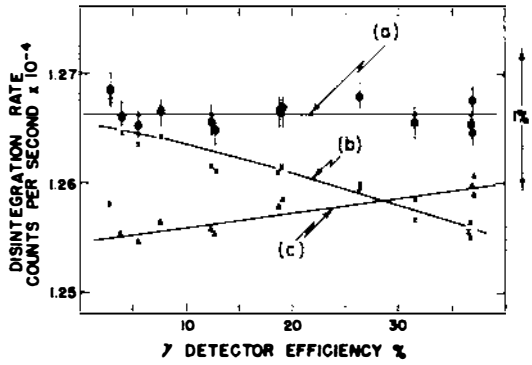


Figure 7. Effect of counting-rate-dependent corrections for bromine-82.

low values of the gamma detector efficiency but deviates by up to one percent at large values, whereas curve (a) is independent of the gamma detector efficiency. For the sake of completeness curve (c) shows the result obtained if the accidental-coincidence-rate correction is omitted from the calculation.

These two experiments serve to demonstrate that the counting-rate-dependent corrections proposed earlier are correct and should be included in the calculation of absolute activities by the 4π beta-gamma coincidence method. The dead-time correction may be insignificant in many cases where the gamma detector efficiency is low, but it is difficult to reduce the accidental rate except of course by reducing the activity.

Finally, I would like to compare the relative merits of 4π beta-gamma counting with those of straight 4π beta counting on the basis of the corrections involved. Considering first the counting-rate-dependent corrections we find that both methods are limited at the upper end of the useful counting rate range by the fact that $(\underline{N}_O\tau)^2 \ll \underline{N}_O\tau$ unless second-order corrections are to be applied. In 4π beta counting the correction in this region is simply that due to dead-time losses but in the coincidence method there are the two corrections as we have just discussed. The ratio of the fractional correction in the two methods may be written

$$\frac{4\pi\,\beta\text{-}\gamma}{4\pi\,\beta} = \frac{\tau_R \underline{N}_O (2 - \epsilon_\beta - \epsilon_\gamma) + \tau_D \underline{N}_O \epsilon_\gamma}{\tau_D \underline{N}_O \epsilon_\beta} =$$

$$\frac{\tau_R}{\tau_D}(1 - \epsilon_\gamma) + \epsilon_\gamma \quad \text{for} \quad \epsilon_\beta = 1 - 0$$

assuming that the dead-time, τ_D, is the same in the two methods. Since the resolving time can be made somewhat smaller than the dead-time the corrections are of comparable magnitude with the coincidence method slightly more favored.

The lower limit of the counting rate range is set by the background rates and for the 4π beta-gamma coincidence method the controlling rate is that in the gamma detector, the background in the beta and coincidence channels being negligible. Since the efficiency of the gamma detector is considerably less than that of the beta detector any arbitrary signal to background limit is reached first in the coincidence method. Thus, in the straight 4π beta-gamma method the remaining corrections are usually very small (of the order of 0.1 percent) and can be readily calculated to a fair degree of accuracy. On the other hand, in straight 4π beta counting, there are corrections due to film and self-absorptions. The former correction has been treated by several workers and estimates for its magnitude can be made. The self-absorption, however, still remains an unknown quantity.

MANN: I had the benefit of seeing this beautiful piece of equipment. Are there any comments?

BAPTISTA: It may be interesting that we had the same proposal as this one, but with sodium iodide crystals seen by a photomultiplier or a plastic scintillator crystal, with another photomultiplier.

CAMPION: Does this exist or is this what you propose?

BAPTISTA: As a matter of fact, I have only published the suggestion and not the results because I have trouble using the 4π plastic scintillator in extrapolation to zero discrimination. As a matter of fact, I consider the correction for the full absorption is quite easy because you can make very thin fluorescent plastic foils as a source mounting. Perhaps you can even put the source on the crystal without any foil. But even for the betas of cobalt-60, using these very thin fluorescent foils, you can have a negligible correction by the absorption of the betas in the foil.

There still remains the problem of self-absorption of the source.

PEACOCK: This may be the wrong time to ask the question, but do you have any intercomparisons with your calibration equipment and any of the others?

CAMPION: Not yet.

PEACOCK: I trust you are going to do that.

CAMPION: Yes. We have done sodium-24 with this equipment, but Dr. Mann tells me there are no results available as yet.

HAYWARD: A good one to compare would be gold-198 because here is the poorest agreement between 4π beta and coincidence counting. It has turned out that Steffen in the last few months has found that there is a 4 percent angular correlation between the beta and the gamma radiation. This may well explain the disagreement.

CAMPION: Yes.

PEACOCK: You will be able, I trust, to publish some efficiency curves for the 4π counter, too?

CAMPION: Yes. We have preliminary information on this.

BAPTISTA: I found that in the plastic phosphor we have, the extrapolation to zero was very short. I am just trying to study this thing to get some results. I will send you these.

YAFFE: I would like to congratulate Dr. Campion on a very nice piece of work. At the same time I would like to quarrel with the use of the word "estimates." He may mean this a little differently, regarding film absorption of 4π beta counting. I do not think these are really estimates now. They are experimental data.

CAMPION: I am sorry.

YAFFE: Perhaps I feel a little more strongly about this because I am involved in these things.

CAMPION: I intended it in that sense.

YAFFE: I agree that the remaining problem in 4π beta counting is self-absorption. We at McGill have extended the work which was begun by Dr. Pate when he was with us, and we have now covered a fairly extensive range. We started with tritium, and we have data for tritium as a solid source, nickel-63, carbon-14, sulfur-35, mercury-203, cobalt-60, arsenic-77, and phosphorus-32. This covers a range from 18 kilovolts all the way up to 1700 kilovolts. I only have preliminary data with me, but some of these are pretty fair. I think the important thing is to realize the amount of self-absorption that occurs in so-called thin sources. For example, carbon-14; it takes only five micrograms to cut the counting rate down by 10 percent, from 100 percent essentially to 90 percent. If anyone would like to see these data afterwards, I have them here. I want to emphasize that they are only preliminary data. I do not have these yet on an expanded scale. I have tried to organize everything on one sheet. This is a little difficult to do because of the large energy range which we have covered.

CAMPION: I would like to add that our experience backs up Dr. Yaffe's remarks, that the self-absorption seems to be much greater than one has been imagining in the past, perhaps. We have tried to get efficiencies greater than 99 percent. It is rather difficult to do this. One has to go to considerable extremes to get up there, even for sources like sodium-24. We are trying with some other isotopes. We have satisfied ourselves that it is not a geometry effect, and it does seem to be self-absorption.

REYNOLDS: Are you speaking of volatile solutions?

YAFFE: All the data which I have here are sources which have been distilled. You get very small particles and fairly uniform. We have taken electron-microscopic pictures of some of these. They are indeed very small.

PUTMAN: If I could raise one question about this self-absorption, I believe Dr. Campion said that the correction when using the coincidence method would be zero. If he meant zero to better than 0.1 percent, I am a little unhappy about this. In fact, it is only strictly true if the gamma counter is equally sensitive to all parts of the source, or if there is some particular relation between the errors of the gamma counter at one point and the differences in the beta counter at that particular point integrated all over the source. In fact, this is rarely realized.

I am wondering whether the sensitivity of the gamma system which he uses is uniform all over the source. I would like to suggest the use of the liquid scintillator which we have used completely surrounding the system for giving better uniformity of gamma efficiency.

May I raise one other small point in passing which I think will probably be taken up later on. I was not very happy about the plot of the beta detector efficiency for gamma rays being taken by successively screening the source with polythene. In fact, would not the gamma efficiency of the beta counter be affected thereby?

CAMPION: Yes. I did not quite tell the whole story there. We did vary the thickness of the polythene to check that no gamma rays were being converted in the polythene and thereby adding to the efficiency that we were trying to measure. This is true for those isotopes whose gamma ray energy is much greater than the beta particle energy because then the range of your photoelectrons is large compared with the thickness of the polythene that you are using, and therefore doubling it should produce an effect. This we checked and the effect was negligible. The polythene was also coated on the outside with a layer of gold similar to our normal source mounts in an endeavor to reproduce the same conditions.

PUTMAN: Did this layer of gold itself make a large difference?

CAMPION: No.

YAFFE: We have actually done the same experiment with only one nuclide, cesium-137, and we get an efficiency, I think, of 0.45 percent. This is 661 kilovolts. I did a visual extrapolation and this looks as though it fits rather well on your curve.

PATE: This is a very interesting paper and the technique is one which will clearly have a lot of applications in decay scheme studies, particularly where you are interested in determining absolute branching ratios.

With regard to the application to disintegration-rate determination on the type of standards distributed by the National Bureau of Standards, I am wondering, in my own mind, whether you indeed gain as much from this 4π beta-gamma coincidence technique as you would appear to. I had the impression that with the type of specific activities that one had met in the customary intercomparison standards, we were getting to the point where the self-absorption correction was under control. We could estimate it. We did do a few comparisons (from memory, gold-198 was one and sulfur-35 was the other, with sulfur the more interesting of the two), where we

seemed to be getting down to a few tenths of a percent. When you go away from the elegantly simple 4π beta counter, and introduce the coincidence method, certainly you begin to free yourself from self-absorption effects, but you introduce a lot of other things. We do a lot of coincidence work at Brookhaven, particularly in decay scheme studies and branching ratio studies, and there are lots and lots of difficulties with this which do not meet the eye. To start with, getting a coincidence efficiency which approaches 100 percent, even using the best available circuits and so on is difficult, and so is correcting your gamma rates with complex decay schemes for summing and the like. This is particularly true with a close geometry, with two large three by three inch crystals.

I am wondering whether one knows these sorts of corrections applying specifically to the coincidence method as well as one was beginning to know the simple self-absorption corrections in the simple 4π beta counting method.

MANN: I think we agree with you that the tool is completely adequate from the point of standardization, but it is nice to know where you are.

PATE: This will be extremely useful where one does not have the specific activity.

ALLEN: One might point out here that, when doing 4π beta-gamma work one should have the wall of the 4π beta counter reasonably thick or else bias fairly high in the gamma channel. For example, with a nuclide like mercury-203, if the gamma channel is biased fairly low, one can get internal conversion X-ray coincidences quite easily.

CAMPION: That is right.

ALLEN: This can very easily mess up the answer.

CAMPION: I have derived an expression for this and it reduces the correction due to internal conversion. It makes it even smaller. The expression is rather complicated. That is why I did not include it in my talk. It is easy enough to derive it.

MANN: Do you have any comments on this question, Dr. Seliger?

SELIGER: I guess I can say something about that now. The numbers that Dr. Campion quotes are quite reasonable. We have experienced the same sort of thing in cobalt-60 and also sulfur-35. In fact, for sulfur-35 it is very simple to take a source which has been prepared in a normal way by just evaporation of an aliquot of solution, count it, then manhandle it physically and chemically, count again and get a 15 percent higher counting rate.

YAFFE: This brings out the question which I would like to ask which may well be pertinent at this time, The question is propounded both of ignorance and indignation.

In my case it is certainly both ignorance and indignation. We have received from time to time, sent out by the National Bureau of Standards, samples of material for intercomparison. One of these was the sulfur-35 which Dr. Seliger has just mentioned.

PATE: This was a Chalk River sample.

MANN: The indignation is Canadian.

PATE: I think the National Bureau of Standards counted it, nevertheless.

YAFFE: The point is this. I think it applies to all of these. I would like to know what happens to these results and what use is made of them. In the case of sulfur-35 which in this case was sent out from Chalk River, I do not see much point in taking a series of 8 or 10 results, and because 8 of them happen to group around a certain figure to say that this is the "true"

accepted value, because there is always the possibility that the person who has a higher figure might have developed some new technique which may indeed be giving the proper answer. With all due modesty, I think this is what happened in the last sulfur-35 intercomparison. We could get values which agreed with what everybody else was getting by using normal techniques, but by using proper sample mounting techniques we got values which were 15 percent higher.

PATE: Not quite as much as that, but something of that order. It was several percent.

YAFFE: Yet this value stood out like a sore thumb, and in everybody's mind it was discarded as a bum value.

MANN: In the Bureau when we give results of an intercomparison we never quote a mean. We never attempt to average them. I think in this particular summary sent out by Chalk River, they did give an average value.

YAFFE: These things were averaged. I just do not see the point.

SELIGER: May I say something about this since I was "indignated" first?

YAFFE: The reason I accused the National Bureau of Standards of this is that Dr. Seliger sent me a letter saying how come you got this value.

SELIGER: I never quite received an answer to that letter.

I think that as far as the Bureau of Standards is concerned, if we distribute a standard of a particular radionuclide, we have to stand behind the value that we observe regardless of whether you may be correct or not. That may be shown later. I do not think this applies in the case of sulfur-35, because we did not issue the particular standard of sulfur-35 that was distributed. I think this is the only way in which we can operate and distribute what we call disintegration-rate standards. If we find at a later date that we have erred, then we can correct it. We cannot say because somebody gets a higher value that we had better change our standard.

MANN: I might add that our value has also stood up to the test of the $\underline{W}_{air}$ measurements. We made calometric measurements and disintegration-rate determinations of the sulfur-35. The calometric values were used at Columbia by Dr. Failla, and also used by Dr. Seliger, Harold Wyckoff and Z. Bay at the National Bureau of Standards. It was also tied in with the 4π beta counting. I think it was compared by scintillation counting with the Chalk River sulfur-35.

SELIGER: No. That was an ionization chamber using a fair number of sources prepared in exactly the same way.

MANN: I want to point out that the measurement does tend to support the fact that we are roughly in the right ball park and not 15 percent out.

SELIGER: This would tie in a good deal better with the mean energy of 50.4 kev which we calculated based on the calometric measurement of sulfur-35, and our 4π beta measurements of sulfur-35, which compares with 49.6 kev from the calculation from the Fermi distribution. This is within the 2 percent uncertainty that we assign to our sulfur-35.

MANN: We are always open to doubt but we do feel very confident about sulfur-35.

SELIGER: Quite.

HAWKINGS: I would like to answer this sulfur-35 problem, because it has been a very important point with me for about two years. When I was working on standardization at Chalk River, we distributed some sulfur-35 in an effort to compare the various methods used for counting sulfur-35. At Chalk River we did this by differential gas counting of sulfur dioxide.

Several other laboratories did it by the best possible 4π beta counting technique. Although at the time of distribution of the results of the intercomparison I did not have the National Bureau of Standards' results, I have been recently advised by Dr. Seliger that the result that he got agrees very, very closely with the result we got by differential gas counting.

Recently a paper by Jesse and Sadukas on the $\underline{W}$ value for air, as based on sources counted by Dr. Yaffe indicated that Dr. Yaffe's counting of sulfur-35 cannot be off by 11 percent. Yet the Bureau's results are also in agreement. We cannot find anything wrong with the gas counting of sulfur-35. All the 4π beta determinations are equal to or lower than this. I wonder whether the McGill determination of this was not possibly in error at this time. I can not believe by any conceivable means that the gas counting technique could be 11 percent low.

YAFFE: All I know is that we can duplicate the other measurements without any trouble at all by just putting our sources down normally. Then you can show an increase. In other words, you are cutting down the self-absorption. You can show an increase to what is essentially an asymptotic value.

HAWKINGS: Your results are not consistent because the counting you did for Jesse and Sadukas gives a reasonable value for $\underline{W}_{air}$. The Bureau of Standards agrees with us.

YAFFE: The samples were counted in the same instrument. You are bringing something else in here. All we are doing is counting in exactly the same instrument two samples of sulfur-35. Are you implying that in one case we counted this correctly and in the other case incorrectly?

HAWKINGS: This has been known to happen.

SELIGER: I would like to point out one thing about this $\underline{W}_{air}$ measurement. We used the techniques that we developed for the 4π beta counting for the preparation of sources for the measurement of $\underline{W}_{air}$. In this case we measured the total ionization current from the sources. The main reason for our entering into this was the fact that we thought we were able to prepare sources from which we could determine the amount of self-absorption. The counting measurement that we made in the 4π beta counter did not enter into the calculation of $\underline{W}_{air}$ because we used the calometric measurement. $\underline{W}_{air}$ depends on the total energy available, and the total ionization current measured. It does not depend on the 4π beta counting measurement.

MANN: When I said they tied in, I meant through the determination of the mean energy. We had good agreement there, within two percent.

PATE: With regard to reproducibility, I would like to emphasize exactly what Dr. Yaffe just said; namely, you can move the disintegration-rate value up and down, by aliquotting from a stock solution and using the two methods of source preparation. The particular method we used in this determination which gave the higher value--I speak from memory--was evaporation under an infra red lamp on a surface treated with insulin with a special rolling motion of the film to maintain the whole area wet until the final moment. This produces a deposit in which brown interference colors are seen. You can, however, reproduce a low or high value at will by using the two methods. The low result has some scatter in it, but you can reproduce these things entirely as you will.

MANOV: That ought to "indignate" somebody.

GROSS: I wanted to say you can leave the $\underline{W}_{air}$ measurement out of this completely, as Dr. Seliger pointed out. With the calorimeter measurement integrated energy what average energy was assumed to use the beta disintegration rate?

SELIGER: 50.4 kev.

GROSS: The last value I saw was the Jesse paper, and he used 48.7 kev.

MANN: What I am saying is that we calibrated the sulfur-35 in the calorimeter. We also used the same value for $\underline{W}_{air}$. It tended to show that the calometric value was right. The 4π beta counting also gave a mean energy close to the Fermi calculation.

GROSS: I think that last statement is sufficient rather than bringing the $\underline{W}_{air}$ in at all.

MANN: I am saying that it helped to give confidence in the calometric value.

SELIGER: May I add one point to that? There may be a reason for bringing the $\underline{W}_{air}$ value in at this time. We determined that the energy absorbed from a sulfur-35 source was approximately one quarter as important as the particle absorption. That means that for every four particles that I do not count in the 4π beta counter, I lose one quarter of that effect in terms of energy. Therefore, if the 4π beta counting results are 11 percent low, and sources were prepared in exactly the same manner for $\underline{W}_{air}$, $\underline{W}_{air}$ itself should be three percent high or low--I am not sure how the fraction works--and this three percent is a big number now in terms of other measurements.

PERRY: I would like to revert to the sulfur-35 that was distributed by Atomic Energy of Canada Limited. We reported a value based on 4π beta counting, and the sources had been prepared by precipitation of the sulfur with barium chloride. Subsequently, we did some further work on the particular sample in which we prepared sources by various techniques, and as Dr. Pate said, one can get any value of a range of plus or minus 10 percent.

We did eventually get what we would call an optimum value by using a wetting agent and evaporating in vacuo without freezing. That I think gave us a value which is something like 8 percent above the value we reported originally.

HAWKINGS: Your value reported originally was 1.32.

PERRY: Yes. I raise a question about the 90 percent as was stated.

MANN: He has been able to raise it by 15 percent as was stated.

PERRY: Yes. We can get an optimum by repeated precipitation, and it begins to fall off.

PATE: All of this tends to negate or sounds as though it tends to negate what I was saying about self-absorption in 4π beta counting being something which we know something about.

What I might emphasize is the following. This was essentially two years ago. The techniques one used at that time for many samples was the simplest one, namely, taking liquid aliquots, which we know how to do, to the required accuracy and making sure of using a method which gives 100 percent mounting efficiency. If you want to do a real job of self-absorption, it involves a research problem on each individual sample that you wish to mount.

Dr. Yaffe would doubtless be able to demonstrate using a suitable method--from memory we did some work distilling sulfur-35--he may now have a better method--laying down a source in this fashion with very much smaller particle size than you can ever possibly obtain with an evaporation process. You are going to do very much better than this discussion would appear to indicate.

PUTMAN: Is it easy to lay down such a source quantitatively?

PATE: No. Allow me to describe an experiment which is in the literature and which involved a half-life determination of nickel-63 which appears under Dr. Yaffe's and my own names. This involved, in our end of things, the determination by 4π beta counting of the disintegration rate of nickel-63. The method of preparing the source was as follows:

We had a stock nickel-63 solution. We pipetted aliquots of varying sizes into a platinum crucible whose aperture was more or less one square centimeter in area by accident. To this we added acid, followed by alcoholic dimethylglyoxime and ammonia. We evaporated everything in the crucible to dryness by infrared. We placed this in a radio-frequency heated furnace, with a 5 to 10 microgram/cm^2 source mount in close proximity to the opening of the crucible and raised the temperature to 200°C. In these conditions, the nickel dimethylglyoxime distills out of the mess in the crucible quantitatively. We obtained a self-absorption curve for nickel-63 in this compound by addition of inactive amounts of nickel from zero to 100 micrograms.

The scatter on the points was such that the reproducibility of the distillation must have been better than 0.2 percent or so.

YAFFE: I think a proper answer to Mr. Putman's question is simple. It is not easy to do this, but it certainly can be done.

PERRY: I would like to mention a method we are trying at the National Physical Laboratory which derives from a method suggested by Cardwell and Milstead in which they prepared very thin alpha ray sources by putting the solution in a small tube, and applying high voltage across the tube. We did try to make that quantitative by weighing before and after, but we found we were getting discrepancies of the order of 8 or so percent. We now think we will try using gold-198, which we can measure fairly easily in the 4π beta counter, as a tracer for sulfur-35. One would use a mixed solution of gold-198 and sulfur-35 and make two measurements at an interval to give one a fair difference in the two readings. One can derive equations whereby one can get the disintegration rate of the sulfur-35 in the mixed source. It does seem that one can use fairly high specific activity solutions which give you a fairly low solid content and use the gold-198 to determine the sulfur-35.

We have tried it with cobalt-60 and phosphorus-32 as a sort of exercise in the technique, and it does seem to be fairly promising. This I may say is being done by a colleague of mine. It is not my own work.

BORKOWSKI: I would like to know how well known is the low energy end of the sulfur-35 spectrum. Presumably the same problems exist in this area as in the thin source. The average energy per disintegration is presumably affected by knowledge of the low energy in the spectrum.

SELIGER: This has been investigated by Dr. Wu and also by some people at Iowa State. They have found it is an allowed transition down to about five or six kilovolts which is as far as they have been able to go. So we can assume that it is pretty well an allowed transition and we can use the Fermi distribution to determine the mean energy.

PATE: I think Dr. Wu has it down to three now.

SELIGER: I am not sure.

QUESTION: Is that a straight line down to three kilovolts?

PATE: Yes.

KOFOED-HANSEN: About the straight-line Fermi plot, I would like to say that at the Israel meeting, quite recently, some doubt was expressed as to the actual knowledge of the beta spectrum shape. Langer gave a report dealing with several cases which are normally being discussed as having straight-line Fermi curves where quite a little curvature was observed. Whether this applies to sulfur-35, I do not know.

Another thing about beta particles or rather electrons that come out of any source, I would like to mention here, is that any sort of disintegration tends to give a rather high ionization of the recoil nucleus. This is evidenced by the measurements of Snell and Pleasonton at Oak Ridge.

There will be quite a few electrons more than the ones that are beta particles. In order to demonstrate that quite clearly, I might mention they have observed in certain disintegration cases as high charge on recoil as 22. I think these things might go into this discussion.

PATE: In reply to the last point, if your beta geometry does approach 100 percent, then the effect of these things is reduced.

KOFOED-HANSEN: Certainly.

PATE: I would like to ask a question about the nonlinearity of Fermi distributions. Is this not a function of what you assume to be an allowed spectrum in calibrating the energy dependent effects in a spectrometer that you wish to use to measure whatever it is you are studying? In other words, is there not a lot of discussion regarding what is your initial standard of an allowed distribution?

KOFOED-HANSEN: No, these measurements show some curvature of the Fermi plot and one has tried to prepare as good sources as one possibly could and tried to calibrate the beta counter efficiency and tried to insert all sorts of diaphrams of different sizes so one could separate out effects from the edges of the diaphragms, etc.

In these measurements one does not assume that certain decays have straight-line Fermi plots. Here one tries to measure the basic idea of the spectral shape.

NOVEY: Aren't these effects of the order of a few percent of the Fermi? These are not really large effects.

KOFOED-HANSEN: Sure. Although there is no experimental basis for the following formula you may get an idea of the order of magnitude involved by stating that the spectrum might have the form $pEF(E, Z)(E^{max} - E)^2(1 + b/E)$ with $b \lesssim 0.12$. Carrying out an integration here you can see how much it would affect the calculations in question.

NOVEY: This does not mean a 12 percent error.

KOFOED-HANSEN: No, by no means. You have to sit down and calculate it out.

BORKOWSKI: I would like to ask whether any of the 4π beta counter users have measured the sulfur counting rate as a function of pressure in the counter. We made such measurements some time ago and we found that for low energy electrons the counting rate was affected by the pressure. One obtained a higher counting rate at lower pressures of gas, indicating that the lower energy electrons, at least on the surface, and the fields around the surface, were quite important. As soon as you got the electrons in the volume, the probability of collecting these events became higher. I wonder if any other people had experience for that.

BAPTISTA: Not for sulfur. I have some results for cobalt. I varied the total pressure from two centimeters to about 50 centimeters of mercury in a mixture of alcohol-argon.

BORKOWSKI: This was in the Geiger region.

BAPTISTA: Yes.

HAWKINGS: We have done some measurements like this. I cannot recall which nuclides they were on. I think cobalt-60 was one.

PATE: You did sulfur, too.

HAWKINGS: We varied the pressure of the methane from two centimeters of mercury up to two atmospheres and we could observe no detectable change in counting rate.

BORKOWSKI: I think this effect will only be apparent on films in which you do not deposit a conducting film.

HAWKINGS: That is true.

MANN: We found the same thing at the National Bureau of Standards by doing a mirror-image experiment. We forced the lines of electric force through the nonconducting film. Do you remember what the change was, Dr. Seliger?

SELIGER: About 3 percent in the case of iodine.

BORKOWSKI: The point I was getting at is whether one can then get essentially the same disintegration rate by not using a conducting film but operating at a lower pressure if such an effect exists. I do not know whether this would be reproducible.

MANN: After all, without a conducting film, the lines of force bend away from the source. You can get over it by lowering the pressure or putting a conducting film on the source. We found it easier at the Bureau to do the latter. You can show this effect by the mirror-image experiments.

HAWKINGS: Could I ask Dr. Borkowski what advantage he sees in eliminating the conducting film?

BORKOWSKI: None. I was curious to know whether there was any significant difference found in these low energy ranges, because we obviously ran into this. This was with nonconducting films.

HAWKINGS: We found you have to be extremely careful with these conducting films because many times you think you have a conducting film and some time later it will become nonconducting.

PATE: We too found this effect. The gold in films is a different element than in aluminum. The Montreal air being humid, one observed quite drastic effects. The color changed from nice purple to a horrible looking pink and the counter would misbehave. We got over this by storing all the films to the moment we used them over silica jell. That eliminated the trouble forthwith. I think we communicated this result to you.

HAWKINGS: We have been doing the same thing for a number of years. Also, if you subject these conducting films to any heat such as from an infrared lamp in an effort to hasten the drying of the source, you will end up with the same condition.

PATE: That is contrary to our experience.

CAMPION: Talking about conducting films, some time ago we performed an experiment to determine how much gold one should put on these films. The experiment was to aliquot given samples of thallium-204 over the film of known thickness and known thickness of gold coated onto them. The results are shown in Figure 8.

HAWKINGS: How thick were the films?

CAMPION: Five micrograms. With thin gold coatings the curve tails off to about 0.95. The interesting point was that in this region we were able to get, occasionally, good plateaus. Sometimes they were at the correct point on the voltage ordinate. If a typical plateau curve is as shown in Figure 9, then occasionally one can determine a plateau with thin gold coatings which is at the expected starting potential, but its absolute value may be as much as five percent down as shown by the dashed curve (a). More usually we find that the plateau has shifted away up in voltage and get a counting rate curve which looks like curve (b) in Figure 9 but still low in

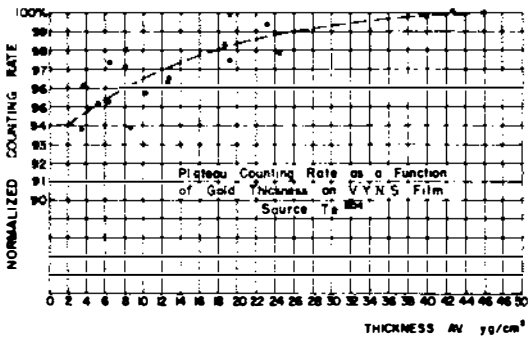


Figure 8. Plateau counting rate as a function of gold thickness on V. Y. N. S. film source thallium-204.

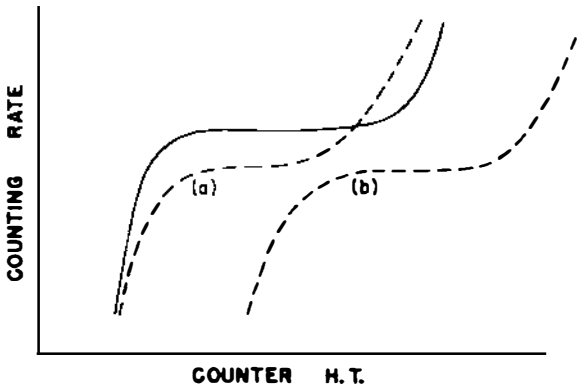


Figure 9. Full curve illustrates normal plateau obtained with sufficient gold coating on V. Y. N. S. film. Dashed curves illustrate plateaus obtained with insufficient gold coating.

counting rate. As Dr. Seliger said, in his talk, it is no criterion to say that you have a good looking plateau to comfort yourself that you have the correct conditions.

PEACOCK: How do you evaporate your gold?

PATE: Perhaps I can answer this point. This is old history representing an argument between McGill and Chalk River. We too have published these curves. With regard to this figure of 30 that Dr. Campion quotes, in our case it is approximately, as I recall, something of the order of 0.8 micrograms per square centimeter where these things turn over. This critically depends on the speed with which you do your distillations. We have a series of electromicrographs with a gold deposit. If you put the gold on with fast distillation rates the aggregate size you observe is considerably larger. Presumably the conduction effect from one aggregate to another is less and it takes more gold plastered on to give you a conducting film. We specify distillation rates which, if you stick to them, give you films which conduct in the sense defined here, giving you good plateaus in the correct position, at 0.8 micrograms per square centimeter. If you conduct an experiment in which you continue to add gold, the moment you go past 0.8 micrograms there is no further increase observed in the counting rate, right up to and including higher figures such as this. Consequently we adopted a safe figure of between two and five micrograms of gold for our working number.

I might say in connection with this question of comforting yourself with the fact that you get a flat plateau, it is true that you have to be aware of the observation of a plateau. If, however, you can get yourself a plateau of several hundreds of volts in terms of a percentage, say 30 to 40 percent longer than your starting voltage, if the counting rate remains constant over this plateau, if you change other conditions of the experiment, and the disintegration rate you compute from this does not vary more than a tenth of one percent or so, then indeed, I think you can start having confidence in the number you are getting here.

CAMPION: I would like to comment on that, starting at the tail end first. I think perhaps one of the first things one should vary as Dr. Pate suggests is the thickness of the gold you evaporate on. If you get a difference there, naturally one should go on until one does not get any difference.

Second, we also investigated this difference of the rate of evaporating the gold and could not get any significant difference between films as we varied the rate of evaporation. Further, I believe Holland,* in his book on thin films, suggests that aggregates are more readily formed when you evaporate slowly.

*Holland, L. "Vacuum Deposition of Thin Films" page 207 ff, Chapman and Hall, Ltd., London, 1956.

PATE: I might say we repeated the experiments on the speed of evaporation of gold and the thickness required for conductivity at Brookhaven after I arrived when we were setting up a production line for making these films on a routine basis by completely unskilled hands. The results we obtained were essentially in agreement with McGill. Here we have a one to one correlation between Upton, New York, and McGill.

YAFFE: I merely want to re-emphasize what Dr. Pate said, and I think Dr. Campion must have missed, and that is that the essential criterion in our case was to prepare a sample and to keep piling gold on until you got no increase in counting rate. This is essentially what the curves which we have shown are. That was the essential criterion. We took plateau, incidentally.

CAMPION: You deposited your source, your active material, and put the gold on top of it.

YAFFE: We did two things. We deposited gold, put a sample on there, and with an energetic beta emitter, deposited more and more gold on top of this.

MANN: You think it is the nonuniformity of the field you are eliminating?

YAFFE: Yes. As Dr. Pate was careful to point out, he said conducting in this sense. We have tried experiments and found them very difficult to do with our rudimentary equipment.

MANN: I think if this is the question, that it should be checked with plain plastic film. I do not know whether I made myself clear before. There is a powerful method of checking this question. All you have to do is to take two halves of 4π beta counter and replace one of the insulated loop anodes with a little sphere and give it negative polarity so that you simply force the lines of force through your nonconducting film. There can be no uncertainty whatsoever. So depositing extra thicknesses of gold should check with this fundamental experiment. You take the counting of both halves and the total, substitute the sphere in one half and determine the correction for that half. Then substitute the sphere in the other half of the counter and determine the correction for that side. There is absolutely no uncertainty then. This is an experiment that we did, and Dr. Seliger and I got a 3 percent correction.

PEACOCK: What thickness are you recommending?

MANN: None.

SELIGER: I would recommend a thickness such that the back scattering is negligible. What that means is that we have been using something very simple. We prepare a thin film and deposit our source on this thin film. If you evaporat aluminum on this film, regardless of which side you evaporate the aluminum on, the acid solution as we use it in most cases will soak through the plastic and interact with the aluminum. We perform all our chemistry on the thin plastic film, and lay on an aluminum leaf which is 240 micrograms per square centimeter.

For cobalt-60 and sulfur-35, then, we evaporate after we prepare our source, if we want to be really careful.

YAFFE: You found no trouble with this evaporated aluminum from oxidation?

SELIGER: We measured the sources within several hours after the evaporation. I do not know that we really can give you an answer to that.

MANN: These cross check with other experiments.

ROBINSON: I had a couple of little points in connection with the conductivity of these films. I understand that during the war considerable work was done with conductivity of thin films with gold in connection with the preparation of glass surfaces to make them conducting for use in airplane windows and such. It was found that the nature of the substrate had a tremendous effect

on the effective resistance of the gold. In other words, it had a large effect where the gold formed into aggregates. One of the things that helped to give you a good continuous film was bismuth oxide. In the case of these thin films of organic materials, it may be that one person is using one type of material and another, another type, and the amount of aggregation is different between them.

Another thing I have also heard, but have no experience with, is that carbon when evaporated does not do this. If you want to take the trouble to evaporate carbon, it is a good way of avoiding this aggregation.

PATE: I think what you say is perfectly correct. The difference between the observations at Chalk River and our own, so to speak, may be something other than the rate at which gold actually arrives on the surface per se. I think what it could well be is the temperature at which the substrate sits as the gold arrives. As I recall Holland's book, it is a question of surface mobility of material arriving on the surface from the vapor phase.

There have been some very interesting electron-microscope studies of things like inorganic crystals distilled onto surfaces, observed as a function of the temperature of the substrate. It is largely a question of the mobility. For instance, in this case, that which the gold atoms would have as they arrived on the surface of the film. In the McGill apparatus we had less bare filament showing, so that the films would heat less by infrared and if our surface temperature had been low, we would have achieved smaller crystal sizes than in another apparatus.

HAWKINGS: May I ask what film distance you used in your apparatus?

PATE: At McGill we had a hemispherical distribution of six films around the filament at about 10 centimeters from the filament.

MANN: I wonder if Dr. Vincent has any comments.

VINCENT: We generally evaporate about 20 micrograms of gold and we found these to be conductive. That is about all.

CAMPION: Did you find them conducting at less than 20 micrograms?

VINCENT: We actually have not tried that, because we wanted to be on the safe side, and we thought the 20 micrograms per square centimeter would not do any harm in low energy emitters.

CAMPION: I gathered that Harwell confirmed this figure of 20 to 30 to 40 micrograms. It is not exactly fixed at 30.

PUTMAN: Could we ask you to be a little more personal about Harwell, there being 6,000 or more people there.

HAWKINGS: With respect to the possible temperature of the substrate, I am not exactly certain of the film distance in the apparatus at Chalk River, but I seem to recall that it is somewhere of the order of 30 to 40 centimeters from the filament.

PATE: What about the question of bare filament area?

HAWKINGS: The bare filament area would be about a quarter of a square centimeter.

PATE: You have the gold sitting on the filament and the infrared radiation is considerably down on these areas compared to the bare filament itself.

HAWKINGS: The bare filament area is quite small because as you know, the gold spreads over the whole heated area of the filament.

There are two other points I would like to make. We have found that thin films of aluminum tend to appear to oxidize immediately on exposure to air. This is evidenced by the fact that a thin plastic film will remain quite taut in the evaporator, and have a mirror-like finish. The minute you let air into the system to remove it, it becomes quite slack.

YAFFE: On that I must say Chalk River and McGill do agree.

HAWKINGS: A third point on which we do not agree with McGill is that a plateau even of several hundred volts is not a sufficient criteria that you are getting maximum detection efficiency.

PATE: No. We agree. There is no disagreement about that.

YAFFE: No one has postulated this.

HAWKINGS: I thought you made a remark a short time ago to that effect.

PATE: Not just the plateau. Many other things.

WATTERS: Whether you could get a conducting film of less than 20 micrograms, I think, was Dr. Campion's question. We did some work at the Radioactivity Center at M. I. T. determining this with evaporated gold on metal slides, and then measuring the conductivity. Once you get up above 8 or 10 micrograms, then everything seemed to be very nice. Naturally you get a little more conductivity with a little more gold, but it is quite acceptable above 8 micrograms. This was at about 15 to 18 centimeters from the filament.

CAMPION: On metal slides.

WATTERS: No, on glass slides.

PUTMAN: I wanted to clarify whether in fact Dr. Campion was referring to our own group or some other group at Harwell.

MANN: I was going to suggest that you might have some comments to make anyhow, or Mr. Perry. I think we should reserve a little time for both of you gentlemen. We will resume the Chalk River-McGill function in a minute.

CAMPION: Cunningham.

PUTMAN: I do not know his results. We used 20 micrograms per square centimeter of aluminum. We have not found any difficulty there. We have not found any difficulty with the conductivity falling off with time. This is a fairly thick aluminum layer. We are working in the Geiger-Muller region. The criteria of conductivity are at least different in the Geiger-Muller region.

PERRY: I have a little comment on this thin film technique. We have not done any experiments on variation of response with the thickness of the film. What we tried to do is to get a uniform method of preparing the films. In order to do that, we include in the evaporating unit an insulator which itself gets covered with gold. The resistance of that is measured. That gives us a control of the amount of gold that is put on. As far as the preparation of the V. Y. N. S. film itself is concerned, we have adopted a centrifuge method. It briefly consists in mounting a glass plate on the shaft and a small quantity of the V. Y. N. S. solution of the right strength is put onto the glass plate, and the centrifuge in operation causes the liquid to spread very quickly and to evaporate very quickly. Then the plate itself is put in the evaporation unit and we deposit gold on that. Eventually we are able to cut the film with a razor blade and float them six or eight at a time. There is a little gradation in thickness from the center of the plate towards the edge, but that only amounts to 2 or 3 micrograms per square centimeter, and when we consider the ease with which we make these films, it is probably worth it.

HAWKINGS: Just to keep the record easy, V. Y. N. S. is a trade name for a mixture of copolymers of vinyl chloride and vinyl acetate, 85 percent chloride.

YAFFE: It is put out in this country by the Electrolyte Corporation in New York.

PATE: I have two remarks to make. I think this McGill-Chalk River controversy is becoming faintly vulgar. With regard to the question of having an insulator in the evaporator, we tried that. We ran into a lot of trouble with thermo-electric effects. We eventually went over, and I think it has been published, to a method of visual inspection, watching color changes, and we individually calibrate each film since the amount of gold each film receives is dependent on its position relative to the filament.

I have been puzzled for a while about a discussion of absorption corrections. People still think it is difficult. In light of the present discussion, I understand why. If you can prepare films with the appropriate conductivity of five to ten micrograms of V. Y. N. S., pluse two to five micrograms of gold per square centimeter, then your absorption correction for well nigh everything is essentially unity. For nickel-63, as I recall, it is less than one percent. If you have thicker films, you still need to apply a correction.

One last word on this question of conductivity. What the film has to do is to conduct the return current from the discharge to an extent such that the voltage drop between the area immediately adjacent to the anodes and ground shall be such so that there is no disturbing influence on the field. You can compute what the resistance for this would have to be for this to occur. The films we used at McGill are 2-1/2 centimeters in radius. There are 2 to 5 micrograms of gold per square centimeter which was more than enough by a considerable factor to satisfy this condition according to the way we prepared them. It is reproducible by witness of the experience of the National Physical Laboratory.

WATTERS: I was just going to bring up an old dog. I still prefer the old archaic way of dropping collodion in acetate on a water surface and packing the film out when you want it. You can make a couple of dozen an hour, with no trouble, of any size you want. I do not see the necessity for the centrifuge technique. I think it is a lot of trouble.

PUTMAN: Could I at this late stage say something that is probably going to "indignate" Dr. Seliger, which is that he made a remark, which was allowed to pass a little further back, that the thickness of his films is such that the back scattering is negligible. It is our experience that the back scattering of a supporting film is never negligible unless the absorption itself is also negligible. We would back this up with two series of tests, one in which the counting rate in the two halves of our 4π Geiger counter were compared, and compared with the total rate and it was found that the ratio of the two halves was ten times more removed from unity than the total loss in the added count, which indicates that at least at first the scattering of the radiation is the most important means by which it is removed from its original direction.

PATE: McGill confirms.

PUTMAN: Added to this, I now want to sling mud at the sandwich method of correction, if I may. If one starts to make a sandwich method of correction by placing on top of the source one film which is the same thickness as the one underneath it, one does not in effect double the reduction in counting rate. One does something quite different from this. If you increase the size of your sandwiches successively on both sides of the film together, you can get a continuous absorption curve. Yet a point corresponding to a foil on one side does not correspond to the absorption you get with half the thickness on both sides.

HAWKINGS: I would like to agree with what Mr. Putman says on sandwich technique. I do not endorse this any longer in the sense it was a technique developed originally to attempt to evaluate film absorption before very thin films were developed. I think it is a stage in the development of the 4π beta counting technique which has passed. I do not think anybody belabours it any longer.

PUTMAN: I thought Bryan Smith had shot this three years ago and I was a bit appalled to hear it was still supported, but maybe I was mistaken.

SELIGER: Maybe I should define what I mean by a film absorption correction so that we can determine what is negligible and what is not. Usually our film absorption correction ranges from a half percent to one percent. I think under those conditions the sandwich technique is valid.

HAWKINGS: As an approximate estimate.

SELIGER: When we make a one percent correction I would say that is good to ten percent.

HAWKINGS: Very true.

SELIGER: Yes.

PUTMAN: The smaller the correction the worse is the sandwich technique because the low energy electrons are beginning to play a part in the scattering.

SELIGER: When the correction is smaller you forget that the percentage of the low energy electrons is smaller and, therefore, of less consequence.

PUTMAN: The over-all correction is okay, but not proportionally.

SELIGER: The area of uncertainty there remains extremely small.

PATE: I cannot for the life of me understand what Bryan Smith will say, but we showed quite clearly that if you want a quick method of estimating your film absorption correction, put your second film on the other side from the source. It gives a straight-line extrapolation and it is quite accurate. Why do people insist on the sandwich method? It is not more difficult to put it on the other side. When the film is on the same side, the back scattering effect is not altered. You have a very, very similar system to the original film. A simple straight-line extrapolation gives you a very accurate estimate of the film absorption. You can use the films of Dr. Pate and Dr. Yaffe.

SELIGER: May I say one last thing on this? We have done this both ways. We have done this up to five layers on each side. It is linear. I agree with you. On your comment for sulfur-35, we agree in our final results. I cannot say that the small correction that is made, even if you want to consider the fairly large uncertainty in this small correction, is worth all this discussion.

PATE: It is worth it because if you put them both on the same side, the answers are always right. If you put them in a sandwich arrangement, the answer can quite definitely be wrong by quite a bit if the energy happens to be low. If you happen to be using extremely thick source mounts, you can run into tremendous errors. These errors are not liable to be small. We at McGill were shooting for one-tenth of one percent on favorable materials. When you are shooting for that close an answer, then these errors you are talking about are by no means negligible.

PUTMAN: I would say it starts to be worth it, at sulfur-35, and if you are starting to push the power by counting below those energies, it becomes worth more.

SELIGER: In our original discussion of the sandwich technique, we only talked about this for energies as low as iodine-131. When we went to cobalt-60, we used evaporated gold films of 20 micrograms per square centimeter and I still do not see where the discussion arises. You are applying a sandwich technique to nuclides and to thicknesses of films where it should not be used.

PATE: We are not applying it. We are implying you are.

2. Liquid Scintillation Counting

H. H. Selinger

I had one or two comments, but I thought there were other things that had to be "de-indignated" first. I thought I would save them until I got up to talk and had the floor.

I agrée with Dr. Pate that the main problem is source preparation. I think for the most part, although there are these isolated cases such as sulfur-35 where we seem to be at opposite ends, that this is where we should concentrate in order to do accurate standardization. Then I think the 4π beta-counting technique is really the most universal method for standardization. That includes its use for electron-capturing sources.

One suggestion that I have, and that to me seems to eliminate a lot of the difficulties, is one which interested me about a year ago. I have not yet had a chance to work on it, but plan to do so. We have always started with a solution of a certain radionuclide, have taken a definite aliquot of it and then tried to prepare a source in a way that it could be counted properly. The main problem is the delivery of a quantitative amount of this original solution to a source mount. It struck me, and because, I think, that we are all creatures of habit, it should have struck me a good deal darlier, that it would be much easier to concentrate on a rather simple method for evaporation of a high-specific-activity source onto a source mount within any definite limits of activity that you want. Let us say within 100 dps up to 10, 000 dps, not caring exactly what you have, which would depend on the specific activity, but in such a way that you get something even less than a mono-molecular or mono-atomic layer of activity on your film.

Then after counting this in the 4π beta counter, it should be a relatively simple matter to take the source and dissolve it completely, bring it up to known volume, and you have what you wanted to have in the first place, without the attendant difficulties, it seems to me, of trying to make your deposition method quantitative.

This method we intend to work on in the standardization of iron-55, for example. I think it is much more appropriate than working backwards the way we have been.

NOVEY: This then becomes only good as a reference. Someone cannot take this and put it in his counter and see if he is using the proper technique. Obviously then, they have to mount an aliquot of that quantitatively and you can't do that.

SELIGER: The question becomes one of purpose. As far as the Bureau of Standards is concerned, for example, I think this technique would be sufficient for us. If you wanted to find out not how your counter worked, but what activity you had, you could use any relative method you want if you have one of our solutions.

NOVEY: Provided you trust the solution.

SELIGER: There has been enough work done that has ended up in disagreement.

PATE: It sounds as though you are trying to put a stop to the disagreement.

SELIGER: A little less disagreement might not be out of line.

MANN: If we are not accurate scientifically, we are right legally.

NOVEY: You would eliminate competition, in other words.

SELIGER: Yes.

PUTMAN: Could one raise the point that while this method is probably highly desirable for

something for which you need a particular method for standardization which relies on producing a thin source, you may be able to intercompare sources by some other property, like gamma ray emission.

SELIGER: Yes.

PUTMAN: It does not apply so well for intercomparison of sources which rely on the formation of a thin source. You are still up against the same problems in intercomparing which you had to face in your original determination.

SELIGER: No, one then uses essentially infinite thicknesses. I shall bring that out in my discussion of liquid scintillation counting but this technique eliminates this problem.

CAMPION: One further difficulty which I foresee, but to which you may have an answer, is that you are rather limited in the amount of activity you can produce. You cannot count curies or even millicuries in any counter.

SELIGER: No, I don't intend to do that.

CAMPION: As I understand it, you take the source which you count in your 4π beta counter and dissolve it, and therefore you only have that amount of material to distribute.

SELIGER: No, we never distribute a primary solution. We shall then prepare secondary solutions for distribution.

CAMPION: I beg your pardon.

SELIGER: I have met several people who have been doing 4π beta counting, and they have said to me, "Bob Hawkings and you recommend the sandwich technique for the determination of film absorption, but this is not any good because Yaffe did not get any good results with it."

YAFFE: That is axiomatic, but true. It is also the fact that yours is theoretically a little difficult to swallow.

PATE: Also, it doesn't work.

YAFFE: And the fact that it doesn't give the right result. If you add all of those up, I believe this is true. Dr. Mann, Dr. Seliger, if you use up all the time in discussion there will be none left for your paper.

SELIGER: This to me is extremely interesting. I shall be glad to write this paper up for the symposium if I don't get a chance to talk about it.

I think the main thing that we stated, at least in our paper, when we talked about a sandwich correction was that this was valid provided the back scattering could be considered negligible. We did not propose its use for low energy beta emitters. We never proposed using the thickness of films that we spoke about for nickel-63. We should amend our paper; in fact, we should call it back and put on the bottom, "Don't use nickel-63!" But for those nuclides that we did use, the method worked admirably.

As one of the European contingent, I took a little bit of time to arrange my slides out of their random order. I would like to report on some scintillation counting that we have done, the main object of which has been to see whether other methods of detection of particles could be used for absolute counting, and in addition to determine, as we have for the 4π beta counter, those characteristics of the scintillation counting technique which would give you some idea--would give us some idea--of the intrinsic sensitivity of the method and its range of applicability. We never intended, and we never shall, I should think, replace the 4π beta counter with any of

either of the solid- or liquid-scintillation-counting techniques. However, we thought it was quite an interesting problem to see how far we could push the methods and whether we could determine their intrinsic sensitivity.

We became interested in liquid scintillation counting several years ago. We thought it would be quite interesting to put some activity into a liquid scintillator and measure the counting rate. Of course, this would give us what we thought would be the answer, because the problem of source self-absorption is eliminated, everything occurring on a molecular level and there being no aggregates or crystalites. As you know, this is not the ideal method. It is a substitution of a method with more attendant difficulties than the original 4π beta counting. We decided to go ahead anyway just to see how far we could push the method. We divided the problem into two parts, the first being the mastery of the electronics required to do the standardization.

I might mention that Dr. Mann and I spent many unhappy hours--some were happy, at least the final ones--working out coincidence and dead-time or coincidence-resolving-time and dead-time corrections similar to those which Dr. Campion has reported, although they are in a different form. I leave it to the reader to bring the two together. Sufficient to say that both methods seemed to work quite well.

We decided to start by using anthracene crystals rather than a liquid scintillator in order to develop the electronics required for the absolute beta count. The main problem here is a little different from that which you encounter normally in scintillation coincidence counting. That is, the coincidence equipment is used not to determine the efficiencies but only to limit the effect of the thermal noise due to the phototubes used. It is still absolutely necessary that the crystals detect every beta particle which is emitted because the "true" coincidence measured is not between a beta particle and a subsequent gamma ray, but is between the outputs of two phototubes looking at the same beta particle in the same crystal; thermal noise in the one phototube being non-coincident, in general, with the thermal noise in the second phototube is not recorded in the coincidence channel.

Figure 1 shows the experimental arrangement that we used for the 4π crystal coincidence counting. The anthracene crystals were approximately one inch in diameter and they ranged between one-eighth of an inch and one-quarter of an inch in thickness. The source was deposited directly on the face of the anthracene crystal in some cases and in other cases on extremely thin films so that the film could first be measured in a 4π beta counter, then transferred to the crystal counter and measured again. We thus had measurements on the identical source so that we could compare the 4π crystal counter directly with the 4π-beta-counting measurements. We made measurements "in addition" in which case the outputs of both phototubes were added together. This is a more efficient method since the criterion of each phototube seeing the light pulse is not present. But this runs into difficulties with phototube noise at some of the high gains required.

The data that we obtained is shown in Figure 2 where the counting rate--that is coincidence counting rate--is plotted as a function of the phototube voltage. The pluses and crosses indicate the observed values of coincidence rate and the solid line, the circles, indicates the corrected value based on both the approximate and rigorous calculation for coincidence-resolving-time and dead-time that Dr. Mann and I worked on.

There is one important point to bring out here. We were able to count thallium-204 with 99.5 percent efficiency compared to the 4π beta count. We were not able to count cobalt-60 with the crystal counter with the same efficiency as the source was counted in the 4π beta counter. This does not include anything regarding self-absorption, because it was the same source that was counted. Yet you will notice that there is an approximately flat plateau. This is one of the pitfalls that one has to be careful about in the coincidence-counting setup. It is possible to get a plateau and still not be counting at 100 percent efficiency. So you have to be quite careful in knowing what the intrinsic sensitivity of your system is, and in knowing the spectral distribution of the beta particles you are trying to measure in order to determine whether this is close to 100 percent efficiency.

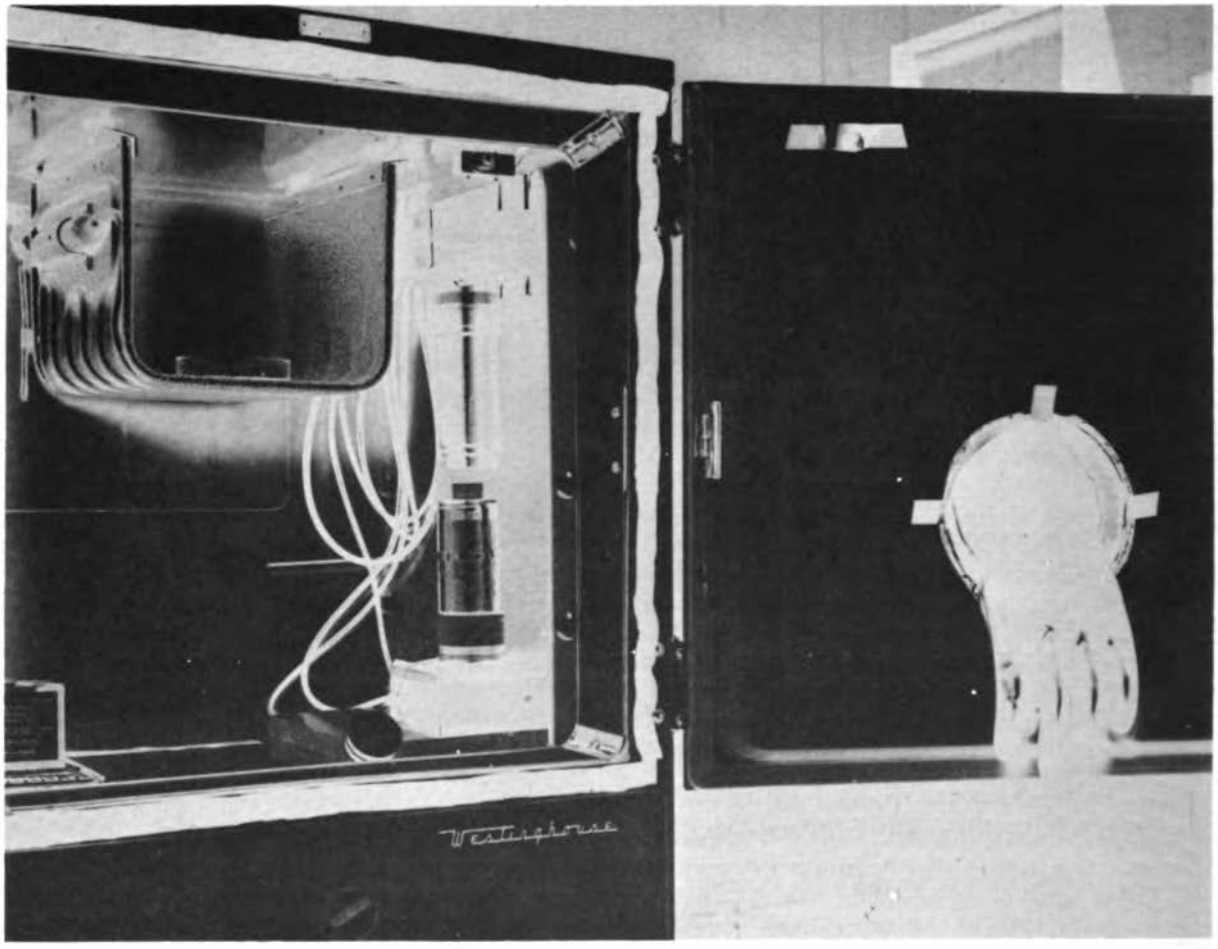


Figure 1. 4π crystal coincidence counting arrangement.

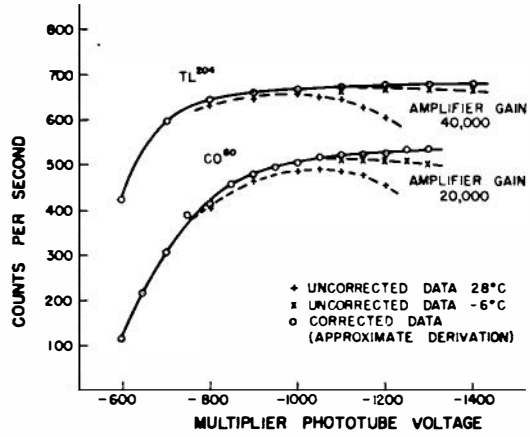


Figure 2. Coincidence counting characteristics.

In order to test these rigorous and approximate derivations that we used for correcting, we devised the experimental setup shown in Figure 3. The 4π crystal counter consists of a phototube on top and a phototube on the bottom of the two crystals with the source between. The true counts which are due to the source occur and are indistinguishable from the thermal noise of the phototubes. But in this experimental arrangement we arranged to fool our coincidence analyzer and make it think this was the case when we put into it three separate sources. The first two were just two phototubes. The thermal noise of these was measured and fed through the mixing circuit. The third was a phototube with a sodium iodide crystal and a constant source. This supplied "N_o", the true counting rate. We measured the coincidence counting rate as a function of the noise counting rates in either channel. This shows that even at

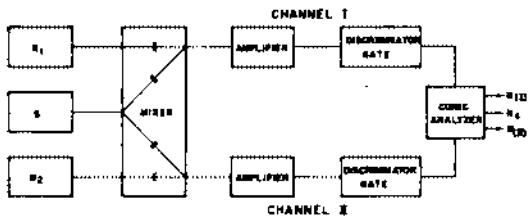


Figure 3. Experimental arrangement for testing counting rate corrections.

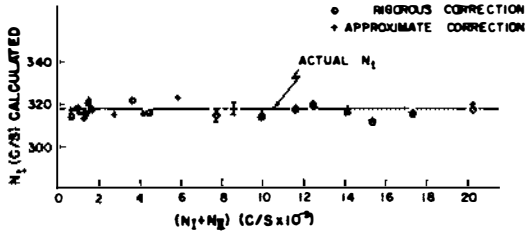


Figure 4. Comparison of counting rate corrections.

very high channel counting rates due to thermal noise, our corrections were valid.

The results that we obtained up in counting rates of the order of 20,000 counts per second are shown in Figure 4. That is 10,000 counts per second in each channel. You see the corrections that we did derive are quite valid. For example, the dead-times that we used in each channel--these are electronic dead-times--which we made slightly larger than the maximum resolving time of the electronic amplifiers, (which were non-overloading because again we had a terrific spread in pulse height distribution), were about 10 microseconds. The coincidence resolving time was about 1 microsecond. So we felt that we had a fairly good arrangement for the electronic side of the standardization in any case, except that we were a little disappointed to find that the anthracene crystal itself--perhaps not "disappointed" as we knew about this before--had such a high energy cut-off. We were concerned with the fact that the liquid scintillator was even less efficient than the anthracene.

YAFFE: Could you estimate where the cut-off occurred?

SELIGER: It requires about 120 to 240 electron volts. I can give you that more accurately later. For the liquid scintillator, it is twice that. It is 240 electron volts for anthracene. I am getting a little ahead of myself. It is 480 electron volts per photon for the liquid scintillator. This depends on the liquid scintillator. Now, we came upon something else. We started to put some liquid scintillators together and we were playing around trying to make some measurements, and we found the following effect in our phototubes. Let me explain how the curve shown in Figure 5 was obtained. Starting at point A, we would increase the voltage in discrete steps. This is purely noise. This is not a source. This is the thermal noise of the phototube. We would measure the thermal noise as a function of voltage. We would expect it would go up from A to B. From A to B took about 4 or 5 minutes. We let the phototube stand at voltage B and we waited for a while and lo and behold, it increased to point C. This C was a constant value, independent of time. That is, independent of any time longer than about an hour. When it reached point C, we took the voltage down the same way and we obtained curve C-D. If the phototube rested, and by rested I mean if voltage was removed from it, the counting rate, when one started again, would be at point A. This was reproducible.

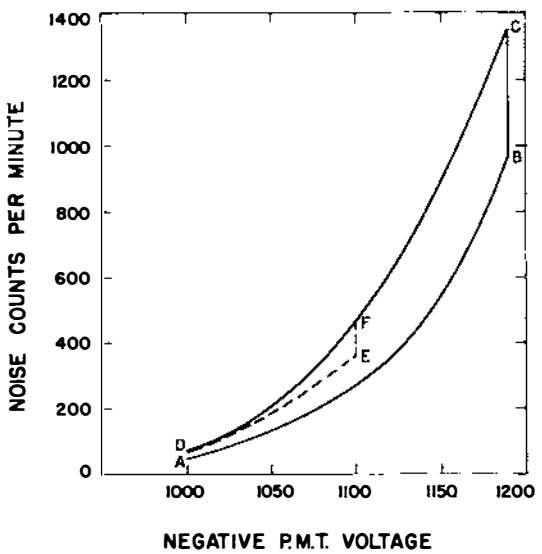


Figure 5. Variations of phototube noise.

GROSS: You were using Dumont phototubes?

SELIGER: Both Dumont and RCA.

GROSS: I have some information on this to tell you.

SELIGER: We found this with RCA also.

FROM THE FLOOR: Is this independent of the dynode structure?

SELIGER: Yes, because we got the same effect with the 5692 and the 5891. This effect is not visiable at room temperature. It is only visible at a temperature where the cesium vapor molecules do not cause any spurious counts, which is around 0°C. It is only visible when you are looking at the noise with a gain of about 20, 000.

This was fine. At least if they were going to vary on us, we would know how they varied so we could get reproducible counting rates. The suspicion that we have is that this is a surface effect due to the fact that there is negative voltage on the photocathode surface.

Now, I will get to the place where I can answer Mr. Putman's question. The arrangement shown in Figure 6 was used for the intercomparison of activities in the liquid scintillation counter. This is a blown up version of the slide arrangement in the spectrophotometer. You can see the little slots on the bottom of the slide. They fit over a screw so that the positioning is reproducible. We observe these with an ordinary phototube. We take care to reduce the reflection of the back surface of the slide and of any other surfaces so that we try to observe a volume in the cell itself. These are pyrex glass absorption cells with optical flats. We make our comparisons with the various samples in a Latin square arrangement to eliminate slight changes in efficiency or slight changes in even the reflectivity of the glass surfaces. We were able to compare tritiated water standards from Chalk River and Los Alamos with our own with a precision of about 0.2 of a percent. This is, I think, quite good for the liquid scintillation counter, as those who have worked with it, mostly people in biological work, will, I believe, agree. It is necessary to work in a refrigerator and to be careful about the normal things, such as long-term fluorescence in the glass and stability of all the electronic components.

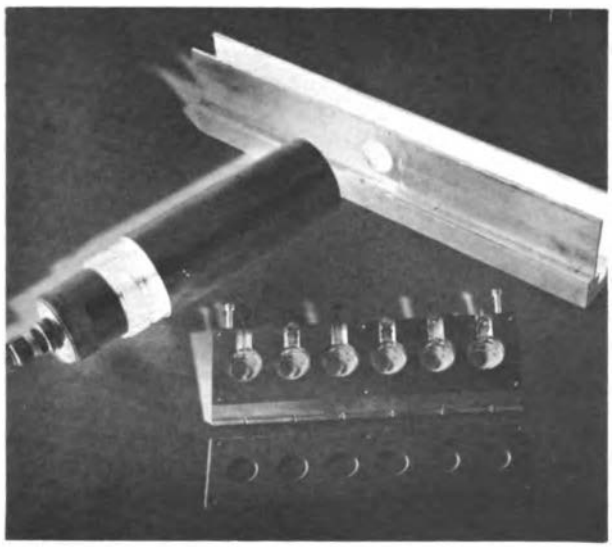


Figure 6. Spectrophotometer slide arrangements.

We were able to do this but with air-saturated solutions. The next problem was to see how stable our liquid scintillator was, and whether we could make it any more efficient than it was, because we knew this would be necessary.

Figure 7 shows an arrangement similar to the 4π crystal counting technique with a liquid scintillator in its place.

The first thing we investigated was based on some articles by Dr. Arnold that indicated that the removal of oxygen is quite important in a liquid scintillator. We later found out that while this was a rather new effect as applied to nuclear detection, the people who were interested in fluorescence had realized this about 27 years ago. We were a little embarrassed to point it up as a new effect, or even to be working on it as a new effect.

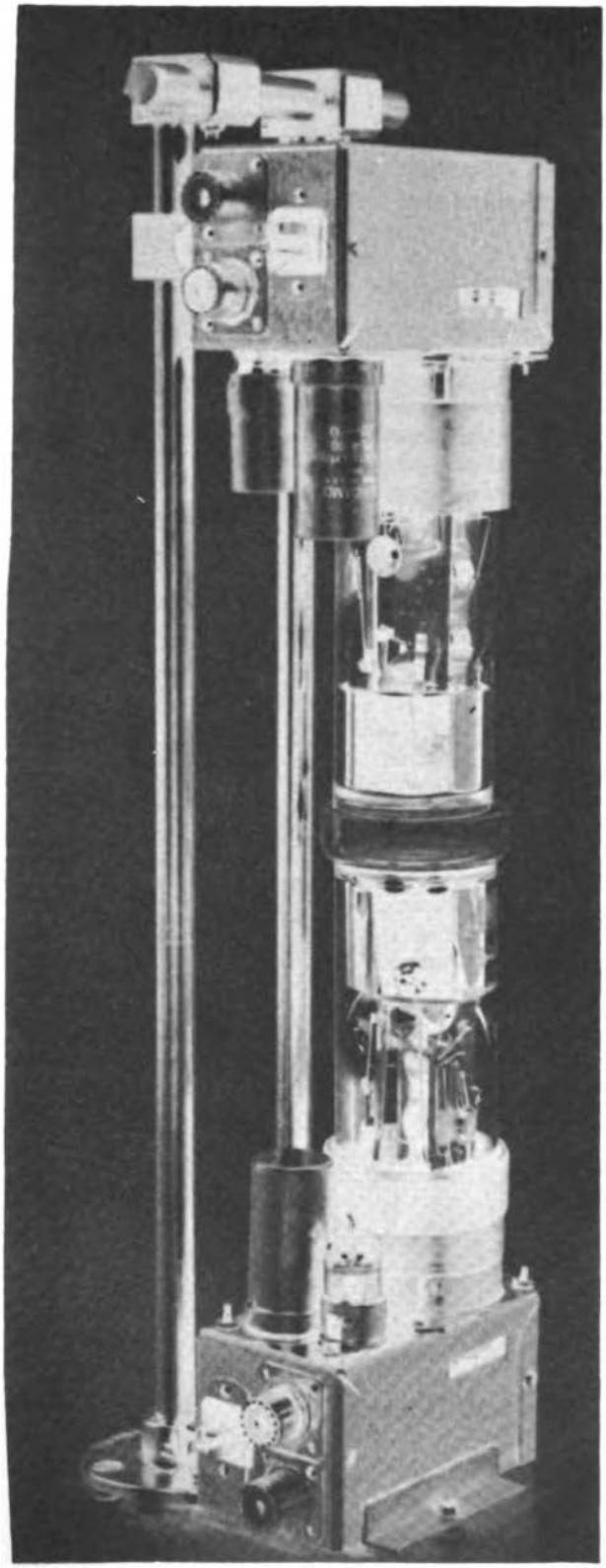


Figure 7. 4π type liquid scintillation counting.

We prepared our sources or our liquid scintillators in the numbered cells and xylene or toluene was distilled into these cells under vacuum using liquid nitrogen so that all gases were pumped out. These cells were then irradiated with a collimated gamma ray beam and the light output was measured in one of several ways; either by measuring the integrated current from a phototube or by using a rather high gain amplifier and low discriminator setting and measuring the integral number of counts observed. These quantities were measured as a function of concentration.

We obtained a set of curves such as shown in Figure 8. Just to show that what we were doing seemed to be in line with what other people were doing, particularly Kallmann, who is really the father of present day scintillation counting, we took Kallmann's data, the black dots, and compared it with our data, and the agreement was fairly good. We felt fairly confident that at least when we made a measurement we were noticing the proper trend. A, B, C, and D indicate the different things that were done to the liquid scintillator.

MANOV: What is α-NPO?

SELIGER: α-Naphthyl phenyloxazole. It is used as a wavelength shifter. We noticed something peculiar in analyzing the data from the previous set of curves. If we plot the percentage

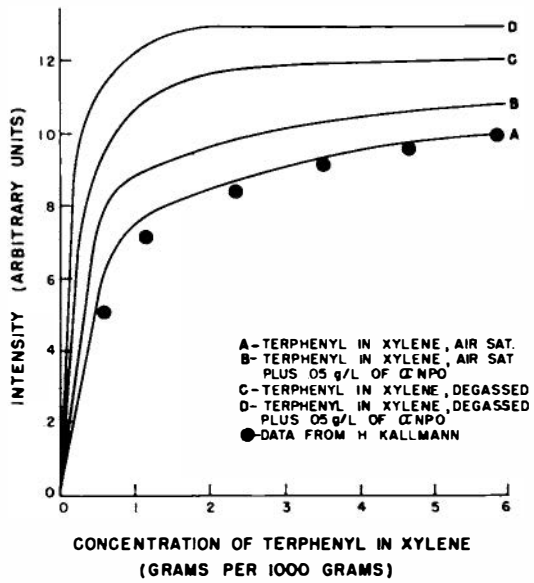


Figure 8. Effect of air and α-NPO on scintillator light output.

increase due to the addition of α-NPO for the degassed solution prepared under vacuum, and for the air-saturated solution, we noticed that there is a bigger increase on adding α-NPO to an air-saturated solution than to the degassed solution. If this were the case, it seemed to us, at first sight, that perhaps the α-NPO is competing with the oxygen, which is doing the quenching.

We approached the problem in reverse. We determined the difference between no α-NPO and α-NPO for degassed solutions and we found something again which indicates that the oxygen in an inverse sense is affecting the α-NPO. This is shown in Figure 9 and Figure 10.

It has usually been thought that the effect of oxygen--that is, the quenching effect of oxygen--was on the solvents. I say this is not incorrect, because Dr. Furst and I had a discussion about this at Northwestern University a month ago and we came to good agreement on both of our diverging results, which might be a good place to start. This line of research interested us. We have not done any standardization yet, but this was all very interesting. We performed the following experiment.

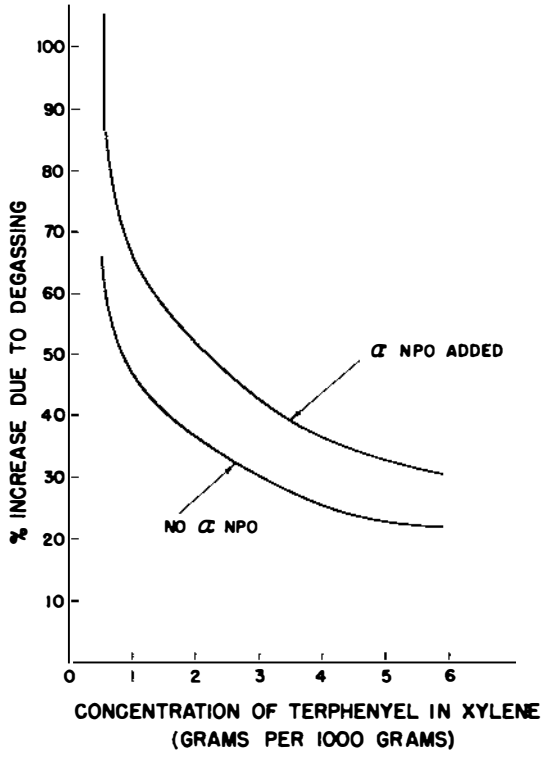


Figure 9. Effect of air and α-NPO on scintillator light output.

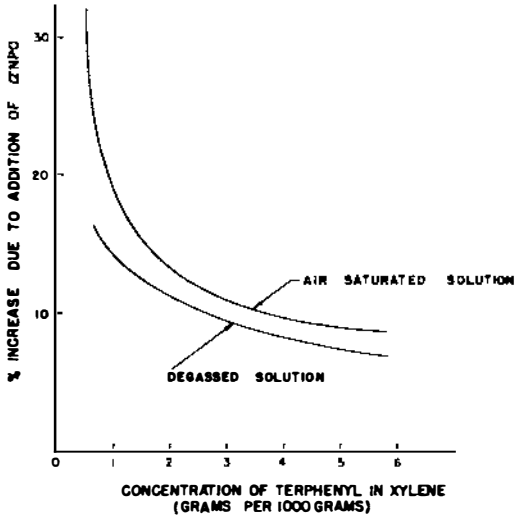


Figure 10. Effect of air and α-NPO on scintillator light output.

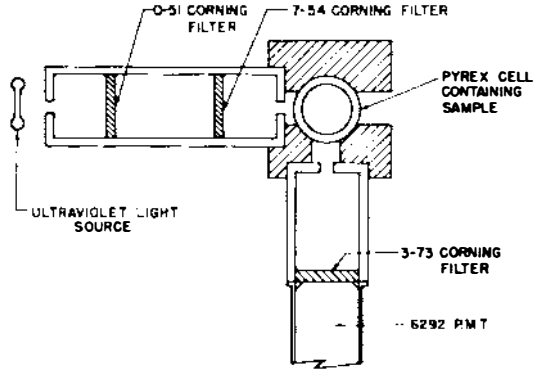


Figure 11. Experimental arrangement for studying fluorescent effects.

Figure 11 shows a black box arrangement. It was really a very simple type of experiment. We irradiated a pyrex cell containing a sample with ultra violet light and looked at it with a phototube at right angles. We used Corning sharp cut-off filters, such that only the long wavelength end of the solutes could be excited.

For those who have not been concerned with liquid scintillation counting, let me draw a very simple picture here. If the energy is transferred from the solvent to the solute, it usually means that there is some overlap between the emission spectrum of the solvent and the absorption spectrum of the solute. The emission spectrum of the solute will be at a higher wavelength side to its absorption spectrum.

In Figure 12 you see the absorption spectrum of diphenyloxazole, which is what we used. The sharp cut-off filters are the 7-54 and the 0-51. The absorption spectrum of the solvent, in this case xylene, is away over to the left at lower wavelength. So just the shaded area of spectral range was incident on the cell. The slight intersection of the long wavelength end of absorption of diphenyloxazole was the only region irradiated. So we think in this case that it was not possible for us to excite the solvent. In B of Figure 12 light is emitted at only the wavelength region above 4,000 Angstroms, which is the emission spectrum of DPO, and you notice that it is shifted to the right of the absorption spectrum. We think here that what we were doing was irradiating only the solute. If the solute would fluoresce, we would see it, and we would not see anything else.

We, therefore, decided to investigate this effect with oxygen saturated solutions and with solutions in which all the gases had been removed. If the oxygen affected only the xylene or solvent, then we should observe no increase in light output when oxygen is removed in this case.

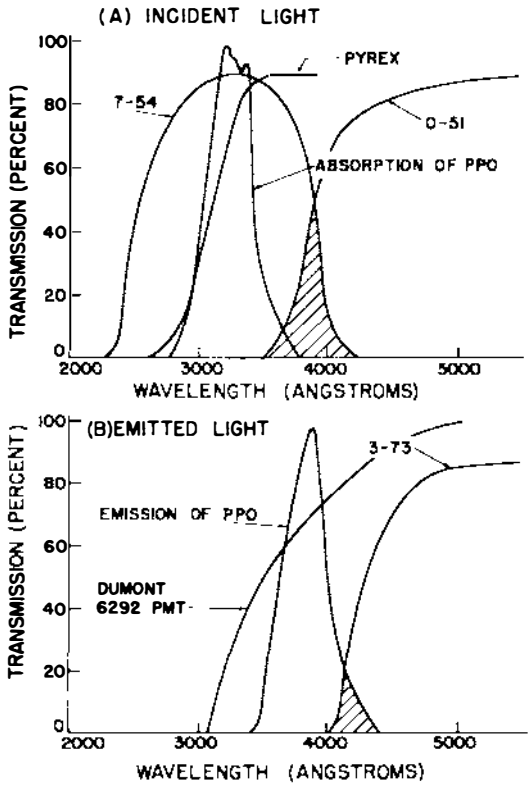


Figure 12. Observed incident and emission spectra.

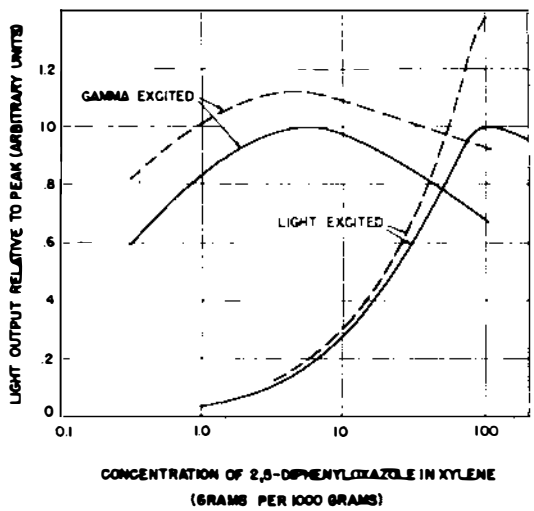


Figure 13. Comparison of gamma and ultraviolet excited light outputs.

Figure 13 shows what we observed. Let us look at the light-excited case first. The dashed curves represent the light output with all gases removed. The gamma-ray-excited curves now are the curves obtained in the ordinary way when gamma radiation is incident on a solvent and you measure the light output. The fractional increase in light output at the particular

concentration involved seems to be the same for the solute as it is in the case when the solvent is irradiated itself, which means that at these concentrations the oxygen quenching does occur in the solutes. Dr. Furst agreed that according to his determinations, the shapes of his light output versus concentration curves are determined at very low concentrations. It is particularly at these low concentrations of solute that the oxygen mainly affects the solvent. But at the concentrations normally used for scintillation counting, the quenching by oxygen occurs in the solute itself.

We had a method for increasing the light output of the liquid scintillator by removal of the oxygen. We found in Pringsheim that this oxygen effect had been noted for a long time so we decided Pringsheim would be a good book to read.

We found there that there is also a thermal-quenching effect. So quickly we investigated the thermal-quenching effect. We decided to investigate this thermal-quenching effect with gas free solutions. If you have a solution exposed to air and you reduce the temperature, you increase the solubility of oxygen in it and the additional quenching could very well negate the increase in light output due to the reduction in quenching by temperature.

We used the same sort of gas-distillation system to prepare our sources. We did one further thing. We took a gold trident and we dipped one tip of it into a polonium solution so we had essentially a point source of polonium on the gold trident. We were then able to measure a pulse height distribution of these alpha particles when the polonium was placed in the liquid scintillator. In this way we had our polonium in one place and we could look at it quite well. We did this for a gas-free solution, and therefore all that was necessary was to plot the relative position of the peak of the polonium pulse height distribution as a function of temperature to see whether this thermal quenching actually was present.

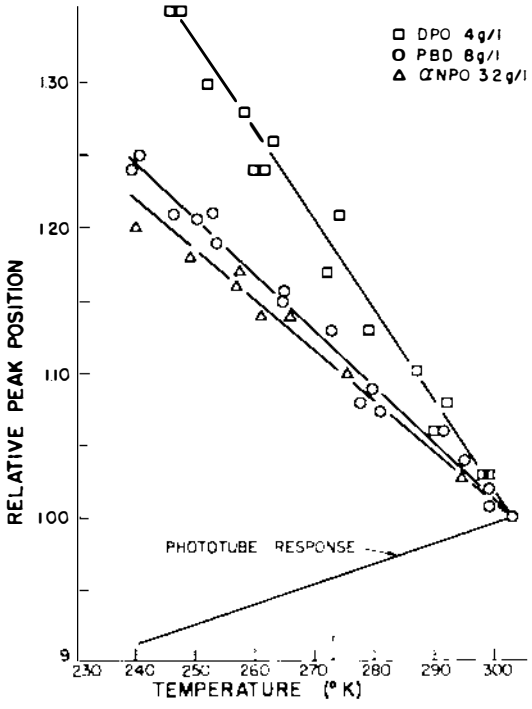


Figure 14. Thermal quenching effects.

Figure 14 shows the results we obtained for several of the liquid scintillators that we measured. We actually found that, in the case of gas-free solutions, as you reduce the temperature you do indeed reduce the thermal quenching and you can get a fairly large increase in pulse height. It was brought out at one time that it is possible that decreasing the temperature really does something else, because these are alpha particles that ionize very heavily.

Maybe this effect was only good for alpha particles and not for beta rays. It was really for beta rays that we wanted to use this. So we performed the same set of experiments except in this case we used a collimated beam of gamma rays and we observed the integrated current from a phototube--again from the sources which did not at this time contain any polonium. We got curves which were very similar except that they seemed to rise a little more steeply at the beginning and then to level off around 260 or 270°K.

In any case we did observe the fact that we were able to increase the pulse height appreciably by lowering the temperature. So therefore we had found a method of making our liquid scintillator at least as good and in some cases better than the crystal anthracene in our 4π crystal

counter. So while we were not happy because the anthracene itself cannot be used for low energy beta particles, and therefore neither can the liquid scintillator, at least we said we should be able to standardize roughly the same energy particles with it.

We have just started to do this. Before we did this I thought it would be very interesting to determine the intrinsic sensitivity of their liquid scintillator. You know that, similar to the internal gas counter, you would have a wall effect if you used beta rays. Let me start at the beginning. You would want to use a line spectrum. These are fairly energetic. You want to absorb all the energy in the liquid scintillator. In the same way as you have a wall effect in the internal counter with beta rays, you will have a wall effect in the liquid scintillator and this will spread out the resolution curve.

Therefore, to determine what the optimum resolution would be, we used alpha particles. The wall effect for alpha particles will be very much less. We put polonium alpha particles into our liquid scintillator without lowering the temperature or removing the air. We used just a simple pyrex glass cylinder sitting on the face of a phototube.

Figure 15 shows the type of pulse height distribution that we get for polonium alpha particles. Curve (b) is the integral pulse height distribution and curve (a) is the differential. It is possible by using a non-overloading amplifier to obtain quite f at integral counting rate curves. Therefore, it is quite possible to standardize alpha particles with not much difficulty at all in the liquid scintillator. We did this by preparing sources first for the liquid scintillator, and then for our 4π counter which gives an 800-volt plateau for alpha particles, but it does work quite nicely.

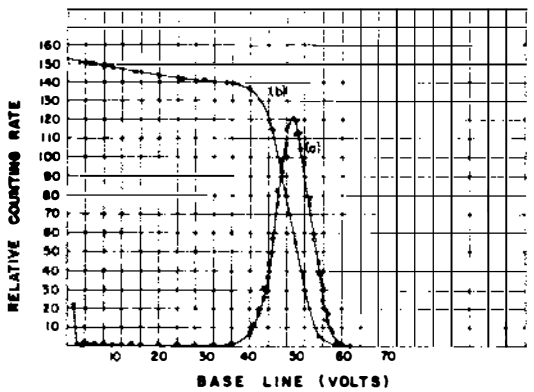


Figure 15. Pulse height distribution for polonium alpha particles.

ALLEN: The alpha source, is this a dissolved polonium source?

SELIGER: It is polonium nitrate in alcohol in toluene. Using phosphorous-32 and strontium-90, we were able to obtain better than 99.5 percent efficiency as compared with the standardization made in the 4π beta counter. At least with this technique it is possible to standardize high energy beta particles. One has to be quite careful, though, about the handling of the liquid scintillator because it is extremely sensitive to quenching, time, changes in temperature, light and just about anything else. For most purposes relative counting, if I were to expose a liquid scintillator to light, I think I could even make up the 11 percent for sulfur-35.

One other thing I might mention. These results seem to be in agreement with the results obtained from pulse height distributions for cesium-137 gamma rays in sodium iodide in this way. This is all qualitative. If one takes the resolution from the liquid scintillator as an indication of the original number of photoelectrons at the photocathode--that is, just a sample case where you say the resolution is equal to $\frac{1}{\sqrt{n}}$--and assume for all practical purposes 5 percent photocathode efficiency. In any case, it doesn't matter with the correlation between the cesium-137, sodium iodide results and these. If we assume this, then we can get the figure that I stated before, about 480 electron volts--I am sorry, let me not jump the gun. I find it requires 5.3 kilovolts per photon for alpha particles in a liquid scintillator. Then I took cesium-137 which has a 624 kilovolt line spectrum and I put it in. I was not as interested in resolution as I was in the position of the peak relative to where the alpha peak was with exactly the same experimental conditions of gain and I found that for beta particles it requires 480 electron volts per photon, a factor of 11 relative to alpha particles. I believe this figure has been obtained by other people as well.

If I can take 6 percent as the best resolution available experimentally for cesium-137 in the sodium iodide crystal, and work backwards in exactly the same way as for sodium iodide, I get 120 electron volts per photon in sodium iodide. We know from other measurements that anthracene puts out half of the light that sodium iodide does. We know also that the liquid scintillator puts out approximately half the light that anthracene does. Our number of 480 electron volts arrived at completely independent of this is just four times that. I think our results are quite consistent and what we have been able to do is reaffirm our faith in the 4π beta proportional counter. We have been able to show what the intrinsic efficiency of the scintillation counting technique is, and I think that it can be used as an alternative method for those cases where it might be impractical to use the 4π beta counter for a good many routine measurements or in those cases where we have high energy beta particles and it would be quite simple to put them in a liquid scintillator rather than to prepare 4π beta counting sources.

ARNOLD: I want to say that was a very interesting paper. I saw a lot of data I had never seen before, and I am very interested. There were a couple of questions. One was that nowadays almost everybody uses this thing with the awful name of popop as the spectrum shifter and if you combine all these virtues, you can get up to, according to Hayes, in the neighborhood of 80 percent of the pulse height of anthracene. Qualitatively it makes life better. It takes you from 800 to 700 vilovolts as the minimum energy you can get away with without loss. Did I understand you to say that early in the game you avoided the use of reflectors?

SELIGER: Yes.

ARNOLD: That seems to be a very high price to pay. You lose a tremendous amount of light that way.

SELIGER: Yes, but we found the reflection from the back surfaces only made more trips through the glass. Since the glass of the cells does vary in reflectivity it is better to avoid this.

ARNOLD: You mean the consistency is improved by removing the reflection.

SELIGER: Yes. We are not interested in measuring low activity sources. We have the range at our disposal and can choose the specific activity we want.

ARNOLD: That is right. I was not thinking of that, but of the rough energy cut-off. If you can push that down, you can measure more beta particles. If you can collect more light in principle you can push this energy cut-off down.

SELIGER: Yes.

ARNOLD: I wondered about this. I have not looked at it from the point of view you have. We used reflectors. We get a lot of consistency with carbon-14. We use relative standardization. We have a few thousand counts at the end in our counters. These are fairly consistent although not perfectly consistent.

I think your limits of energy have got to be very high. There was one other rough remark I wanted to make which I think is of no interest to people who are interested in as precise standardization as you are, but it has been known for years in the trade, I think everybody who worked in the field had independently done this--it is a crazy thing to do--if you take the counting rates in individual channels and square them, on the extremely naive assumption that the probabilities for observing a pulse in one of the two phototubes is uncorrelated with the probability of observing it in the other phototube, which is clearly wrong in any detailed analysis, still one gets quite excellent agreement in what one observes in coincidence. For tritium you might expect this to work fairly well because when the efficiency is very small, the assumptions become better. Even for carbon-14 where you are counting 60 or 70 or 80 percent of the betas, this is roughtly true.

We used to use it in the old days for approximate standardization. I once worked through the

theory of this. You can see it works a trifle better than you expect naively, but I think it is worth reporting that to within a few percent for a particular isotope, this is a rather reliable technique, if you have a high enough counting rate.

LAZAR: This figure of 5.3 kilovolts per photon of alpha on the resolution being proportional to $\frac{1}{n}$ for sodium iodide, apparently is not the case. We found that there seems to be an intrinsic resolution in sodium iodide which limits you to about six percent. In other words, if you take resolution from the total number of photons measured with a light pulse, and take the observed resolution and take them in quadrature such that the crystal resolution is equal to the observed resolution minus the phototube resolution, you get something left over for the so-called intrinsic resolution. This was reported a couple of years ago. I was wondering whether you have observed such an effect with anthracene or any of these solutions. You do not know what the number of photons are from a measure of the resolution of the peak.

SELIGER: I thought this effect was similar to the one that Morton introduced for the relation between the number of photoelectrons at the photocathode and the resolution, and when you are dealing with approximately 50 photoelectrons at a photocathode, it was not important enough. In this case $\underline{n}$ is large enough so it would not enter. I have not looked for this, and I cannot say anything about this being true for the liquid scintillator or for anthracene. I do not know.

LAZAR: I am not clear. $\underline{n}$ is small and so the resolution is dependent on the number of photons. When $\underline{n}$ is large, then the resolution should be dependent only on the intrinsic resolution.

PATE: That is right.

LAZAR: The better the resolution that you have, the more photons you have, the more you eliminate the phototube and the greater the resolution of the phosphor, if there is any.

ARNOLD: Can't one simply say that the resolution of liquids is always poorer than the resolution of good crystals so that intrinsic resolution is never important? If the best resolution you see for alphas is this 17 percent, then something that causes a 6 percent resolution cannot be very important.

LAZAR: What you say is true if the number of photons is small enought so that it determines the 17 percent. If the 17 percent is not determined by the number of photons, but by the phosphor, then you have an indication as to possibly why all the resolutions are poorer than you should expect.

ARNOLD: Maybe I should say--Dr. Seliger can defend himself--that I know of at least two independent studies which have arrived at essentially this same ratio for the amount of light emitted by alphas.

LAZAR: I do not question the ratio. I question the absolute number.

ARNOLD: Wouldn't these numbers fall down completely? Certainly 5 kilovolts or so is about right for alphas in liquid scintillators. At least that seems to be the consistent number that people get by a number of different methods. If that is true, that would tend to confirm in an independent way the kind of analysis that Dr. Seliger has done. If you can get this number, not from looking at the square root of $\underline{n}$, but a measure of the total light, which is what these other studies are, then the agreement of the two numbers is an argument that the analysis is roughly right.

SESSION III - GAMMA MEASUREMENTS AND STANDARDS

W. E. Perry, Chairman

Summary

During this session papers are presented dealing with the application of gamma ray measurements to the determination of gamma ray intensities and disintegration rates of sources. Dr. Lazar shows how the response of sodium iodide phosphors to gamma rays can be interpreted quantitatively within a few percent to provide data in the form of efficiency versus energy curves which can be used to estimate the absolute intensities of gamma rays. These curves vary with geometry much less than with size of crystal, and the method described should prove useful for many applications. Mr. Putman deals with a number of aspects of absolute and secondary standardization based on gamma ray measurements, including the extension of the range of calibration of sources to the multicurie level by a technique employing small ionization chambers of the type used for the radiation monitoring of personnel. He also refers to a gamma ray twin spectrometer incorporating two sodium iodide phosphors, and having a subtraction device, for the analysis of mixed sources and suggests that this technique may be useful for the detection of impurities in radioactivity standards.

Dr. Hayward's consideration of the cobalt-60 decay scheme and of the accuracy of the gamma-gamma coincidence method for disintegration rate measurements leads him to suggest that cobalt-60 is more suitable than radium as a primary gamma ray standard. Here one must compare the complications in the decay scheme of radium and the self-absorption problems associated with radium as a gamma ray standard with the uncertainty in the half life of cobalt-60, as revealed by measurements of specimens of this material. There must inevitably be a transition period during which the claims for cobalt-60 can be investigated, in particular from the viewpoint of the radiochemical purity of samples.

1. Measurement of Gamma Rays

N. H. Lazar

It is fortunate for me that the subject of this conference includes both measurements and standards, because the work I am going to discuss will certainly not cover standards in the same sense that we heard about in an earlier session. Measurements of gamma rays would certainly be a better description of the subject matter which I will cover.

The order of accuracy and precision which we have been able to attain with the scintillation counters, about which I will talk, has been from three to five percent up to as high as you wish. This certainly is an order of magnitude poorer than what you can measure with such beautiful techniques as Dr. Kipfer described yesterday. However, the scintillation counter approach to measurement of intensities does serve a purpose, even in this company, in that it is an independent method for determining the accuracy of the measurement of gamma rays, although the precision in itself is not particularly good. Since the accuracy is independently determinable, these results should give an indication as to whether these other very precise results are also accurate.

I would like to describe our methods for determining the efficiency of sodium iodide crystals for gamma rays of various energies. This will involve two different approaches to show the attempts we have made at checking these efficiency determinations.

I would then like to go on from there and describe some intensity measurements. I believe that will probably lead us well into the subject and probably any questions during the discussion will bring out specific applications.

The first method that we used is based on the following set of circumstances. You know that the pulse height distribution which you obtain from a sodium iodide crystal contains a peak (which people call all sorts of things, and which I will usually call the full energy peak) and a Compton distribution. In order to determine the intensity of particular gamma rays of complex spectra, one must interpret the number of gamma rays in terms of the peak area or peak height or something at least related to the peak, because the Compton distribution is very strongly a function of the surroundings and the particular experiment you are doing. Usually lead surrounds the crystal, and your spectrum will then show a peak at 75 kilovolts from lead fluorescent X-rays. If you have a thick backing for the source, you end up with a back scatter peak, and the Compton level is also increased due to gamma ray scattering from the surroundings. Just a measure of the total area in the spectrum (either from integration of the spectrum or electronically) obtained in some particular geometry is not a sufficiently accurate method of determining the intensity of any one particular gamma ray unless that particular setup has been calibrated in advance.

The method which we have been using for determining the intensity of unknown gamma rays is independent of the particular surroundings and geometry in which these measurements are made, but rather depends entirely on the crystal size itself. I think it has some universal application.

Consider a cylindrical crystal of sodium iodide of radius, r, and thickness, t. If you place a source on the axis a height, h, above the crystal, then one may calculate what we call the total efficiency for the detection of a gamma ray. By this I mean the probability that a gamma ray will be detected if it strikes the crystal. The resulting pulse may fall anywhere in the spectrum even if the original interaction were a Compton interaction since multiple Compton scattering in the crystal followed, possibly, by absorption of the gamma ray results in any pulse size up to the full energy peak. In fact, most of the full energy peak is due to this multiple scattering.

However, we ask only what the probability is, if a gamma ray strikes the crystal; i.e.,

that it will be detected, and do not concern ourselves, for the moment, with the pulse height distribution. For this purpose if we take an element of solid angle, $d\Omega$, at the angle, θ, then the path length through the crystal will be x, and the amount transmitted is $\exp(-\tau x)$, where τ is the absorption coefficient of sodium iodide in cm^{-1} and is known to about one percent. This, perhaps, is not very accurate in this company, but it is accurate enough to give us the precision we are interested in for these measurements. If $\exp(-\tau x)$ is the transmission, $(1-\exp(-\tau x))$ is the probability of absorption of the gamma ray, and the absorption times $d\Omega$ is the probability that a gamma ray entering $d\Omega$ from a point source at height, h, above the crystal will be detected in the crystal.

Now, x is a function of θ, but they are related by straightforward trigonometric expressions. What we have done is to integrate this function numerically over the entire face of the crystal.

This involves a numerical integration because as you get outside the cone defined by the bottom edges of the crystal and the source, x becomes a rather complicated trigonometric relation and the integral is not given in a closed form.

This integral represents the total efficiency, ϵ_T, times what I will call the solid angle, Ω. From the way in which the integral is taken, Ω is simply the solid angle subtended by the top face of the crystal. What we have done at Oak Ridge is to calculate this product for various values of τ and various heights, h, above the crystal. ϵ_T, itself, is a very strong function of the height above the crystal. When you get farther away there is less corner penetration since most of the gamma rays are going straight down, and so, since the path length is, on the average, long, the efficiency is large. These values have been tabulated for the three by three inch cylinders and the one and a half by one inch cylinders.

Since that time, Wolicki, at the Naval Research Laboratory, has done a more complete job and calculated these functions for all standard crystal sizes made by the Harshaw Company. The report number is NRL-4833 by Wolicki, Jastrow and Brooks.

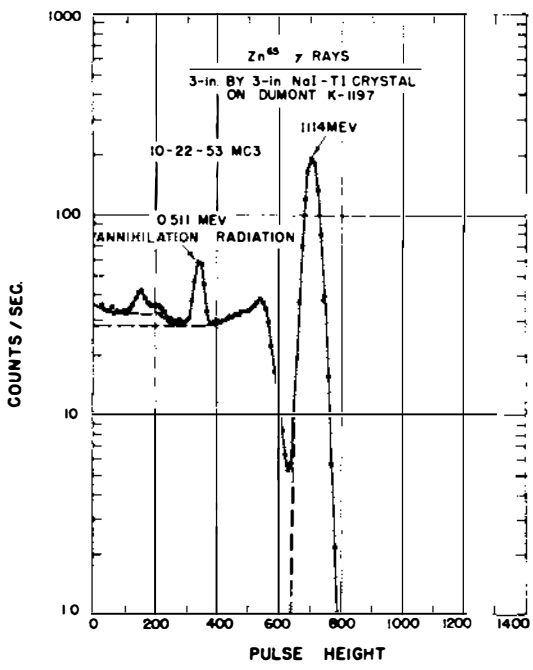


Figure 1. Zinc-65 gamma ray spectrum.

Unfortunately, this does not give the answer you are interested in because this is the total efficiency which tells you, again, what the probability is of getting a pulse but it does not tell you what the probability is that the pulse will fall in the peak.

To solve that problem a number of spectra were measured under conditions in which there was no observable scattering. From the spectrum, the ratio of peak area to total area was determined. Figure 1 shows what I mean by this. This was not one of the samples that were measured, however, the shape is typical of what you get from a sodium iodide three by three inch cylindrical crystal. The shape of the Compton distribution is drawn in as a straight line. We now know what the shape is from the other gamma rays near this energy. It probably tails down a little bit. Actually the difference is of the order of only one or two percent in the total area. In any case, this is typical of the type of spectrum that we would get, although the back scatter peak was not present for the spectra that we measured. We simply took the ratio of the area in the full energy peak to the total area and multiplied this by ϵ_T to determine ϵ_p, the peak

efficiency. In these measurements we were careful that the dial setting for zero pulse height of the multichannel analyzer that was used to take this data was well known, so you knew how far to extrapolate in order to include the total number of counts in the spectrum.

In addition, other precautions were taken to eliminate spurious counting in the region of the Compton distribution. For example, an attempt was made to use beta ray emitters in which the beta ray energy was very low, or better still K-capturing isotopes. For these cases, the bremsstrahlung contribution is considerably reduced below what you would normally get from high energy beta rays stopping in your absorbers. So we selected as best we could, and luckily we were able to find sources such that the amount of bremsstrahlung was quite small, or at least unobservable, in most of these measurements.

The room in which the measurements were made was completely cleaned out of everything except the crystal, the tube and the source. The measurements were made in a shielded counter room; the walls were two feet of concrete. The floor was something like six feet from the crystal. The ceiling was something like ten feet away, and the walls were also something like ten feet away. In this case the only appreciable scattering that you get with gamma rays which started off in the wrong direction is that from the air.

We did not evacuate the room, although this was the obvious next logical step. It is a step that you would have to take to improve the accuracy. At Arco, they have gone part of the way to the next step in that they put the source and crystal in a big balloon and filled it with helium. Their results agree pretty well with ours; thus, the scattering does appear to be quite small. What we measured, then, is the ratio, R, of photopeak area to total area in the spectrum and multiplied ϵ_T by R, thus obtaining E_p, the peak efficiency. I have simplified matters to a great extent. I have indicated that we use only isotopes in which there are single gamma rays. Unfortunately, you cannot find isotopes emitting only single gamma rays of high energies. So we did actually use some substances, such as sodium-24, which emit several gamma rays.

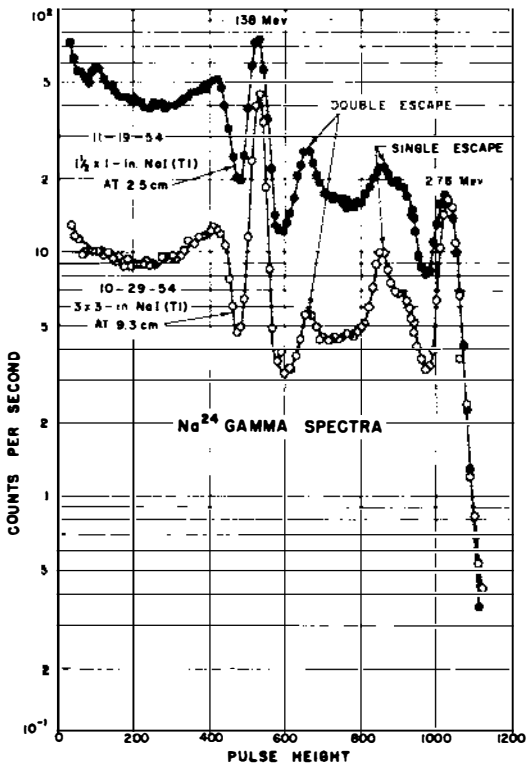


Figure 2. Sodium-24 gamma ray spectra.

Figure 2 shows the pulse height spectrum obtained from sodium-24. The upper curve is obtained with a one and a half by one inch crystal, and the other with the three by three inch crystal. The tremendous improvement that is obtained with a three by three inch crystal is clear. This is the reason we now use, almost exclusively, three by three inch crystals. These spectra were determined with h = 9.3 centimeters. In this case, we are interested in the efficiency for the 2.76 Mev gamma ray. It is clear that the peak to total measurement as I described above is not sufficient to obtain ϵ_p (2.76) because of the contamination of the spectra by the 1.38 Mev gamma ray. First, we simply added up all the counts in the spectrum. Then, in order to determine the contribution from the 1.38 Mev gamma ray, the extrapolation of the Comptons from the 2.76 Mev gamma ray under the 1.38 Mev peak was drawn in. Notice, the extrapolation is over a very small distance, indeed. We have also obtained spectra from gamma rays with energies near 2.76 Mev. For example, ThC'' emits a gamma ray of 2.62 Mev, and we also have observed gamma

rays near this energy produced at the Van de Graaff machine in (p, α) and (p, p') reactions which tell us approximately how this region should look. If one simply measures the area of the 1.38 Mev peak after subtraction of the extrapolation of the Compton distribution and then uses the previously determined value for the peak area to total area, R, one may determine the total number of counts due to the 1.38 Mev gamma ray. This was then subtracted from the total counts in the spectrum to get the total counts from the 2.76 Mev gamma ray. The determination of the peak area for the 2.76 Mev gamma ray could easily be performed and thus one has R (2.76).

This was done for sodium-24 and also for yttrium-88, which emits gamma rays of 1.85 Mev and 0.90 Mev. I believe those were the only two cases in which a measurement of this type was needed.

This, then, was the method originally used to determine the peak efficiency. I might just say in passing, you may notice that this does not require a knowledge of the decay scheme. The peak to total ratio and ϵ_T are just functions of the crystal and geometry. The peak area for an unknown is also just a function of the size of the crystal although the Compton distribution may now be dependent on the surroundings. The full energy peak is necessarily undistorted by, for example, absorbers. If there is any scattering of the gamma ray directed toward the crystal, it can only remove counts from the full energy peak. You now, simply, use the known absorption coefficients for the absorber and, divide the peak area, by the transmission to determine the number of gamma rays which left the source.

CAMPION: Is this true if you have a phototube with poor resolution?

LAZAR: It is true depending on the poorness of the resolution, in a sense. What you are saying is that with small angle scattering, gamma rays, scattered in the forward direction, still fall in the peak and you count them, essentially, twice--once in the peak and once by the correction for scattering. This is true. However, you fit a Gaussian to the peak and the small angle scattered gamma rays would distort the shape of the peak. To about a percent the fit is very good with no scattering. When you use very thick absorbers, you have to worry about this problem and to that extent the precision is limited, and you have to recognize this.

The question was raised about how to include the area of the pair peaks seen in spectra. We simply add up all the points in the spectrum to get the total number of counts and this automatically includes these peaks.

The question was asked whether R was independent of the distance from the crystal, and the answer is "No, it is not." It is very nearly independent of h, however. We have measured it at two distances, 9.3 centimeters (that happens to be the height of our stands that we normally use) and 3 centimeters. The reason we chose 3 centimeters is that it is very nearly the worst possible position. For a three inch diameter crystal, a source three centimeters from the face gives you, nearly, the maximum corner penetration. When you get very much closer than three centimeters, you have rather thick corners seen by the source. If you get very much further than 9.3 centimeters, the corners again thicken since the gamma rays are moving nearly in a vertical direction. R should be very much less sensitive to distance at greater distances. For these two distances, the difference in R is of the order of 10 percent or less. I think it will be clearer from Figure 3 which is a spectrum from a 7.5 Mev gamma ray obtained from the $Be^9(p, \gamma)B^{10}$ reaction with one Mev protons. The same ratio was determined as previously but, unfortunately, the scattering conditions were not nearly so clean. Also you can see artifacts of what appear to be other gamma rays. The 7.5 Mev gamma ray is a capture gamma ray, and there is probably some cascading to the ground state through other levels; thus, ϵ_p determined from this spectrum is not as accurate as the other values.

Figure 4 shows ϵ_p versus energy. For the three by three inch crystal there is a variation in efficiency between 3.0 and 9.3 centimeters. As a matter of fact, the variation is more a function of ϵ_T than R. If we attempt to determine the intensity of a gamma ray obtained at other than

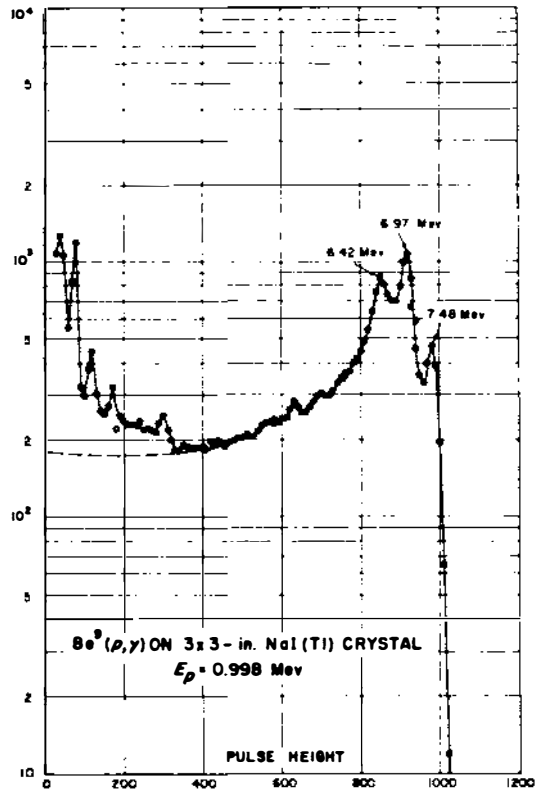


Figure 3. Spectrum from 7.5 Mev gamma ray obtained from $Be^9(p, \gamma)B^{10}$ reaction.

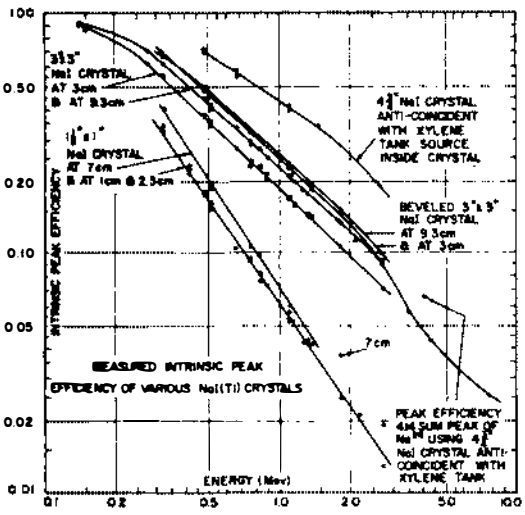


Figure 4. Peak efficiencies for various sodium iodide crystals.

these distances, we choose the photopeak to total ratio by interpolation; for distances comparable to 9.3 centimeters or larger we use R determined for the 9.3 centimeter data, and then correct ϵ_T for the different height above the crystal. Notice that the points do not scatter greatly from the curve which was drawn. The point farthest from the curve was three percent off. We think we know the efficiency up to 2.76 Mev, let us say, to the order of three percent. Certainly to one Mev, a three percent uncertainty would be a conservative estimate; at 2.76 Mev it may not be very conservative. However, I think the three percent value is valid over the entire region to 2.76 Mev. We used the value shown on the curve for the determination of the thorium half life. The 2.62 Mev gamma ray in ThC'' was measured in samples in equilibrium with the parent thorium, the samples were weighed, and the half life was found to be in agreement with that of Kovaric and Adams ($t_{1/2} = 1.42 \times 10^{10}$ y). We felt that these efficiencies were correct to three percent and this seems to confirm that.

We made another series of experiments which is a check on these values. It is an independent check and depends on the decay scheme only in the sense that we ask that all beta transitions proceed through the gamma ray. This was a coincidence experiment between beta and gamma rays in which we used a very small 4π counter (split anthracene crystal) to detect the beta rays. This was done by mounting two anthracene crystals, each of the order of a quarter inch square by about an inch thick, on the same phototube, and placing the source between them so you essentially have 4π geometry. The necessity for the small size crystals will be indicated shortly.

Coincidences were taken between beta rays detected in this geometry and gamma rays. If the decay scheme is such that every beta ray is followed by a gamma ray, the beta spectrum in the anthracene crystal will look something like Figure 5. The tail results from the summing of the detected gamma rays with some of the beta rays. Since essentially every beta ray is counted, whenever a gamma ray is detected in the anthracene, it removes a pulse from one portion of the beta spectrum and replaces it someplace else.

The area of the gamma ray peak observed with the NaI crystal in coincidence with beta rays, $P(\gamma)$, may be written as

$$P(\gamma) = C(\beta)\, \epsilon_p\, \Omega\, A$$

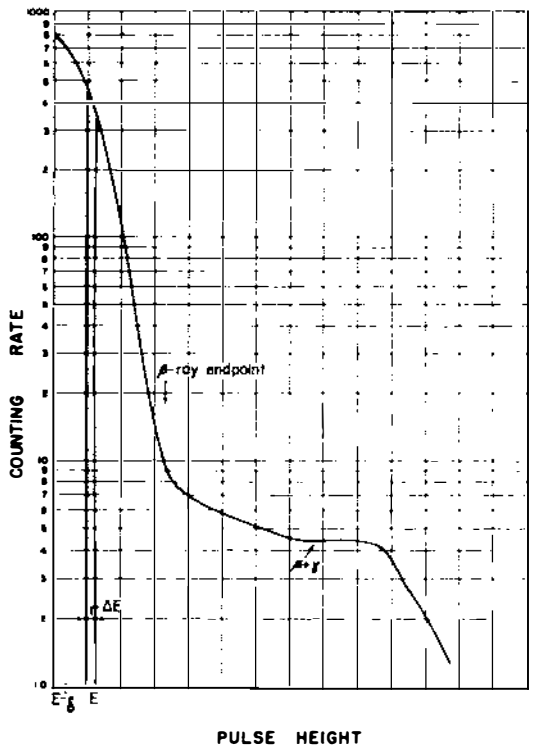


Figure 5. Beta spectrum in anthracene crystal for beta-gamma coincidence measurements.

where A is the absorption in anthracene and $C(\beta)$ is the counting rate in the beta channel. Now, you can see why we made the crystal small. We can measure the peak area accurately. We can measure the geometry accurately. We can even determine the absorption fairly accurately since the absorption coefficient for anthracene is very well known. However, in this experiment a portion of the beta spectrum was selected by means of a single channel analyzer and coincidences with these pulses were recorded. Thus a correction for pulses in the selected pulse height interval which arose from the summing of simultaneous beta ray and gamma ray detection is required. The thinner the anthracene crystal, obviously, the smaller the correction.

Consider electrons which are emitted at some energy, say $E-\delta$ (see Figure 5) and which sum with gamma rays so as to raise the pulse height to E. The number of counts in the window at E can not simply be determined by placing a beta ray absorber between the anthracene and the source, because doing so removes the summing effect. In addition, the scattering of gamma rays is increased. However, the probability for an electron in the window to be summed with a gamma ray to give a pulse in ΔE at E is just the probability of gamma ray detection $(1-e^{-\mu x})\Omega_\gamma$. where u is the absorption coefficient of anthracene, x, the thickness of the crystal and Ω_γ the solid angle for gamma ray detection (assumed = 1) times the probability the gamma ray pulse size size will be δ. If this is multiplied by $C(E-\delta)$, (the number of electrons at pulse height $E-\delta$) and then integrated over δ from $\delta = E$ to $\delta = 0$, the result gives the number of pulses in the pulse height interval in question which arose from electrons of lower energy.

To facilitate performing the integration, a flat rectangular distribution for the gamma ray spectrum was assumed. This assumption is not bad for anthracene where you have almost pure Compton scattering and in which the probability of multiple Compton collisions is negligibly small. Under these circumstances the Compton distribution is, in fact, very nearly rectangular. It turns out that with this assumption, all terms are independent of δ except $C(E-\delta)$ and the integration is simply the area under the beta spectrum from zero to E. Fortunately, the correction is only a few percent with the small crystal used and this procedure is then quite reasonable. To sum up, the number of counts in the window is mostly determined by the number of electrons of the energy in question and then there is a small correction for the gamma rays which sum with electrons to give pulses in the window.

This type of measurement was done for a series of isotopes. In fact, ϵ_p was found for mercury-203, scandium-46, niobium-95, sodium-24, gold-198, and sodium-22 for the two distances. The results are shown in Figure 5. One can see the order of magnitude of the scatter of points that was obtained. These results checked with the values obtained from the peak to total measurements and gave us a feeling of confidence in our values to the order of precision that we have quoted. I might just mention that the efficiency of a four and three quarters inch sodium iodide crystal is plotted on the same curve. This data was obtained relative to the three by three inch crystal efficiency for a source in the center of the larger crystal.

I would like to take a few minutes to describe the methods we have used, with these efficiencies, to determine the intensities of gamma rays. To determine the intensity of a gamma ray is a straightforward problem, but is not always recognized as being somewhat more complicated than one might naively expect.

If you have a single gamma ray of energy, say, γ, then $I(\gamma)\,\epsilon_p \Omega A = P(\gamma)$ where $I(\gamma)$ is the intensity of the gamma ray; ϵ_p, the peak efficiency, Ω, the solid angle subtended by the face of the crystal at the source; the factor A represents the absorption of the gamma ray by any absorbers that are placed between the source and the crystal and, $P(\gamma)$, the full energy peak observed for the gamma ray. This expression is correct only, however, if there is no summing of cascading gamma rays. Consider, however, the case exemplified by sodium-24 in which you have a beta transition feeding two successive gamma rays, γ_1 and γ_2 (Figure 6).

In this case, three peaks are obtained. The third peak is the coincident sum peak and arises when both γ_1 and γ_2 are completely absorbed in the crystal. The probability for this (neglecting factors like A) is

$$\epsilon_{p_1} \Omega\, \epsilon_{p_2} \Omega\, \overline{W(0^\circ)}$$

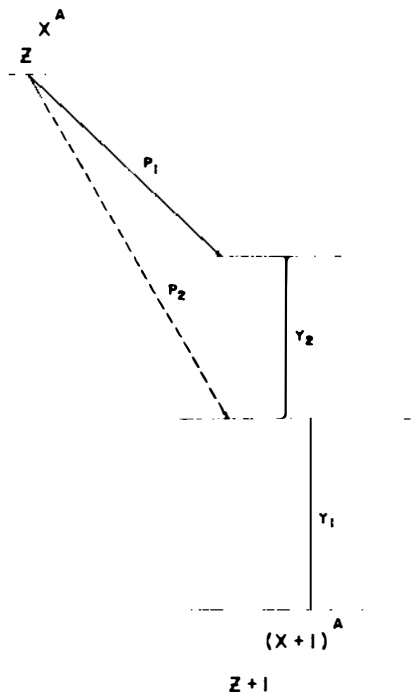
where $W(0^\circ)$ is the angular correlation of the two gamma rays evaluated with 0° between them (both enter the crystal) and corrected in the manner described by Rose for finite angular resolution. The intensity of the coincident sum peak is just the disintegration rate, N_o, times this probability.

If we are interested in the intensity of γ_1 only, from its particular full energy peak, then (again dropping factors like A)

$$N_o\, \epsilon_{p_1} \Omega = P(\gamma_1) + N_o\, \epsilon_{p_1} \Omega\, \epsilon_{T_2} \Omega\, \overline{W(0^\circ)}$$

or

$$P(\gamma_1) = N_o\, \epsilon_{p_1} \Omega \left[1 - \epsilon_{T_2} \Omega\, \overline{W(0^\circ)}\right]$$



DECAY SCHEME FOR SODIUM - 24

Figure 6. Decay scheme for sodium-24.

With the second term in the bracket zero, this is just what you have for only one gamma ray, thus the bracket represents a "correction factor" for coincident summing. This correction factor is in fact very small if Ω is small. But in any case it is finite and should be taken into account when precise gamma ray measurements are made. In fact, it may easily be as high as several percent. For example, in 2π geometry, $\Omega = 0.50$ and for $\epsilon_T \simeq 0.5$ this correction could be as large as 0.25. At 9 centimeters, Ω is of the order of 0.04, ϵ_T is still of the order of 50 percent, so the correction is just of the order of a couple of percent.

If the two gamma rays are in cascade, but if there is a branching ratio between them, i.e., if there are two beta groups such that the intensity of one gamma ray is greater than the other one--then an extra factor has to be multiplied in the expression, which takes into account the branching ratio between the two gamma rays. Again one must be careful what one means by this factor. If you ask "What is the intensity of γ_2?", you must remember that for every γ_2 there is a γ_1 and the "branching ratio" factor is actually unity. These considerations are rather

straightforward in the sense that it is fairly clear what has to be put in here. Very often these corrections are not applied, however, and the intensities, then, are not accurate to that extent, although they may be very precisely determined.

I believe that is about all I have to say about the determinations of intensities in this way.

SELIGER: We have available standards of zinc-65. I just wonder if you have had occasion to check this intensity measurement with the disintegration rate standards of zinc.

LAZAR: We have not checked it in that way. We have checked it in other ways, however. One approach is to simply check them against the results of other people in the laboratory, and this we have done and always find a good agreement.

Usually, these considerations are applied in decay scheme studies and in those cases consistent results are the only real measure of the precision or accuracy of these calculations.

As you imply, we have not tried to push the method to its ultimate precision. We probably should attempt to measure some National Bureau of Standards samples. We have never done this. Incidentally, most of our intensities are quoted to the order of five percent because of the uncertainties in solid angle and statistics on top of the three percent uncertainty in efficiency.

PATE: I found the last remarks in your paper quite interesting. We have been doing measurements of this sort, not particularly with standardization in view, but in decay scheme studies where one wishes to determine branching ratios and K-capture to positron ratios and things of this nature. What you say about the importance of summing effects is quite true. It is particularly true when you are doing gamma-gamma coincidence measurements.

I might mention here a technique that we use to make these corrections a bit more directly more in an experimental fashion. One cannot claim this is new. It is a fairly obvious sort of technique, but it might be worth mentioning here. It starts off in a simple fashion when you wish to extrapolate, as in your sodium-24 spectrum, the Compton distribution from your higher energy gamma ray underneath the photopeak of your low energy gamma ray in order to do an accurate subtraction. The method we use to make such corrections is to use coincidence technique. You gate a single channel analyzer on the lower energy gamma ray in the cascade pair, and display what the 100 channel analyzer sees in coincidence with the gate. If there is a cascade transition clearly you would display just the upper energy gamma ray. If you use the Compton distribution obtained under these circumstances, you can do an accurage subtraction, provided, of course, you worry about the random coincidences.

LAZAR: If I may interrupt for just a minute, that approach would be fine except for the efficiency determinations I have discussed, because placing another crystal nearby would cause scattering which would change the total area.

PATE This is true.

LAZAR: Each problem has to be looked at individually. There are practically an infinite number of problems each one of which would have to be done separately under these conditions.

2. Gamma Ray Measurements and Standards

J. L. Putman

We may divide the subject of gamma ray measurements and standards into two sections. In the first, let us consider briefly those methods in which gamma rays alone can be used for primary measurements of radioactivity, that is absolute measurements of disintegration rate. The second section is concerned with relative measurements of activity by the comparison of gamma ray measurements. It is upon measurements in this second category that I would like mainly to concentrate, but perhaps a few words on absolute measurements would not be out of place.

The use of Geiger type counter tubes for primary measurements, using gamma rays only, is obviously very limited because of their intrinsic low sensitivity and the difficulty in determing this sensitivity either theoretically or by direct methods involving only gamma rays.

Scintillation counters can be made to approach an efficiency of 100 percent for low energy gamma rays and so do offer means of direct measurement of gamma rays incident upon them, without the necessity for calibration by other methods.

The other ways in which gamma rays alone might be used for absolute determinations include the gamma-gamma coincidence method, and for this the scintillation counter is the obvious choice of detector; particularly since its energy discrimination can be used to overcome difficulties of spurious coincidences caused through scattering.

Our general feeling is that when another method for standardizing a nuclide is available, this is generally preferable to the direct measurement of gamma rays, at least for primary measurements. Most nuclides which emit gamma rays do so following the emission of some other type of radiation which is easier to measure; that is, alpha particles, beta particles, positrons or X-rays. So in making absolute measurements we try, whenever possible, to use these other radiations either directly or in coincidence with gamma rays. The only exception is in the internal conversion of isomers and very few of these seem to be of practical importance. One glaring exception to this generalization is metastable barium-137 following cesium-137 with a half life of 2.6 minutes. The importance of barium-137 is that cesium-137 is a pure beta emitter, and gamma rays are only emitted from the daughter produce barium-137. The use of 4π beta counting methods is valid for beta-gamma emitters because corresponding beta and gamma rays give rise to a single impulse unless the gamma ray is delayed by a time greater than the resolving time of the detector. If it were simply a delayed gamma ray, corrections could be made by screening the source and counting the residual gamma rays; but this gamma ray is also internally converted, and so far we have not been able to get any very definite figures as to the degree of internal conversion. Therefore, I submit that the direct measurement of cesium-137 by some other method such as the direct measurement of gamma rays would make an important contribution toward the standardizing, at least, of this isotope.

In using scintillation counters for absolute determinations, one drawback is the need to calculate the efficiency of the phosphor from absorption data. As an experimental approach, the efficiency might be determined by progressive thickening of the phosphor and extrapolation to 100 percent efficiency. This is rather an expensive business, especially when using sodium iodide phosphors. It is also complicated by the fact that thickening of the phosphor not only increases the intrinsic absorption of gamma rays, but also increases the internal absorption of photons. A correction for this effect would be necessary at least when using high energy gamma rays and consequently thick phosphors. Evidently, even if the phosphor thickness is increased until a maximum counting rate is achieved, there is no guarantee that this maximum represents 100 percent efficiency. We have not done this, incidentally, but the method may have possibilities, especially when using liquid phosphors.

Let us now turn for a moment to the absolute determination of radioactivity by indirect measurements, that is, those which do not involve direct counting of the radiations. These

include the use of ionization chambers and calorimeters. Ionization chamber measurements unfortunately depend on a number of factors which have to be determined independently. They depend on W, for example. Although I think temporary agreement has been reached on values of W, there seem to be periodic storms about this particular constant which tend to leave the experimenter with little confidence. Of course, the measurements also depend on the energy of the radiations, absorption coefficients, and on the material of the chamber. But it is of such importance to radiologists to know the radiation at a distance from a source that in some ways, for radiologically used isotopes, the source strength is more important than the disintegration rate itself.

Perhaps one of the most important reasons for measuring absolutely the disintegration rate of radiologically used isotopes is in order that the appropriate K factors can be evaluated; from these, the radiological doses can be calculated under various conditions, given the actual amount of isotope in use.

Let us now turn to the kind of measurement for which I think gamma rays are principally suited. This is for the comparison of sources which have been absolutely measured by other means.

Since the gamma rays are very penetrating they lend themselves naturally to a comparison of sources under conditions of negligible self-absorption. Also, since they are penetrating, they suffer very little absorption or scattering by air. Therefore, they lend themselves to measurements under conditions of large distances between source and detector, where the geometry of the arrangement is not critical. Gamma rays are also convenient for routine measurements approaching 4π geometry, in which a source is placed inside a reentrant detector and the radiations penetrate the walls without much absorption.

This has bearing, I think, on the important problem of extending absolute measurements to the higher levels of activity. It is well known that ordinary counting measurements and even some of the so-called absolute ionization chamber measurements concern themselves mostly with sources of fairly low activity, and direct counting determinations are generally confined to the microcurie level.

However, standard sources are now required with activities from one to several curies. If we can accurately compare the radiations from gamma ray sources, with those which have been standardized by other means, we can work our way in steps from microcurie or millicurie levels right up to the curie levels. This is the method which we, and probably many other people, have been using. For example, with cobalt-60, we started off with two sources, one about 160 millicuries, and these were compared with ionization chambers some years ago.

Then the 20 millicurie source was dissolved and measurements were made on dilutions of the solution. Having made the gamma ray comparison, the activity of the nominal 160 millicurie source could then be determined, and this was used to calibrate a further ionization chamber for high flux gamma ray measurements in a machine. This was done three or four years ago: the problem of calibrating large sources has since become more acute. My colleagues in the Isotope Division at Harwell have been working on a simple system for intercomparison of relatively large gamma ray radiographic sources.

For this the commercial type BD11 ionization chamber is used. It is normally intended for personnel monitoring and has a graphited plastic coating and a graphite center electrode insulated with polytetrafluoroethylene (Teflon). The chamber is open at the bottom so that it can be screwed into an electrometer for measurement. The measurements were made by my colleague, R. L. Otlet, who arranged thirty of these chambers at a distance of one meter from the sources he wished to compare. Radiographic sources in aluminum capsules were used. These were mounted at the top of a transparent polymerised methyl acrylate plastic (perspex) pillar about a quarter of an inch in diameter. The BD11 chambers were screwed on to separate perspex pillars, mounted on a perspex frame in an arc one meter from the source mounting, at which distance the inverse square law can be relied upon very accurately.

In fact, these comparisons do not rely on the inverse square law, but this provides another variable at our disposal. To minimize scattering, the whole arrangement was mounted on a bench in the middle of an otherwise empty large room. Twenty-eight chambers were actually used for the comparisons, and measurements were taken, not by comparing the loss of charge in the detectors over a fixed time interval, but by varying the time so that the loss of charge was almost exactly the same for two different gamma ray sources. Thus, the chambers were operated under identical conditions for the different sources, using the electrometer on the same part of the scale so that variations in its efficiency over the scale did not affect the comparison. Twenty millicuries of cobalt-60 was found, at the distance of one meter, to discharge a BD11 chamber from an initial 300 volts down to 25 volts in 15 hours.

The chambers were found to saturate at about 12 volts so were always operating under saturated conditions. The low final potential of 25 volts was chosen to reduce errors which might arise in estimating the sharing of charge between the electrode of the ion chamber and the electrometer. By leaving only a small residual charge in the ion chamber, the effect of the charge sharing errors on the total discharge measured was made small.

No correction was made for humidity. The original charging potential of 300 volts was only accurate to plus or minus a half percent.

Tests were made with the 28 chambers, using two cobalt-60 sources, each of activity about 20 millicuries, to check the reproducibility obtainable. Five measurements were made, three on one source and two on the other, and they were interspersed in time. Since 15 hours were required to obtain a measurement, variations of conditions over this period could be significant. They were reduced to some extent by taking measurements on either side of a mean time. The net result of this was that by taking, for each chamber, the average of the ratios found between the two sources in the five measurements a root mean square statistic fluctuation of ±0.35 percent was obtained.

On statistical grounds, a mean of the ratios obtained with the 28 chambers should now yield a root mean square error of about ±.07 percent in the comparison of the two sources. I think it is interesting that by using commercial chambers under such relatively crude conditions one can compare gamma sources with this sort of accuracy, and I can see no obvious source of systematic errors. If comparison is possible with this accuracy one is justified in taking a series of steps from a relatively low activity source, dissolved for standardization, up to curie or multicurie standards. Otlet has applied this technique for measurements of activity of separate sources up to 40 curies.

Let us consider how the accuracy might be further improved. First, the charging potential of the chambers was only measured to plus or minus one-half percent. It was only by averaging over five chambers that the reproducibility reported could have been achieved, on this account alone. Obviously, the accuracy of charging can be improved without great difficulty. Second, humidity changes could affect accuracy, and errors could be avoided by enclosing the chambers in a thin plastic envelope containing silica gel.

Third, no particular care was taken to arrange that the orientation of the chambers was always the same. Any departure from axial symmetry in the efficiency of the chambers could, therefore, introduce an error, and this could easily be avoided simply by marking one side of each chamber as a guide to orientation.

We hope that by taking such simple precautions as these, the deviation of 0.35 percent between individual chambers can be reduced by a factor of three. So we now have quite a simple and reliable method of comparing sources up to very high levels.

I should like now to consider another aspect of the use of gamma rays for comparing sources of like isotopes: that is the desirability of using gamma ray reference chambers for the maintenance and comparison of substandards. A good deal of work has been done in several

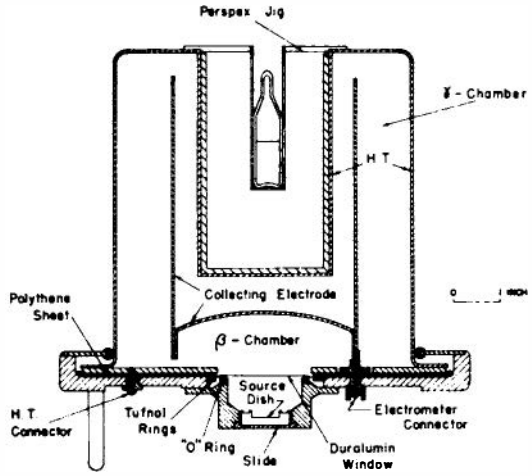


Figure 1. Ionization chamber for secondary standardization of beta and gamma ray sources.

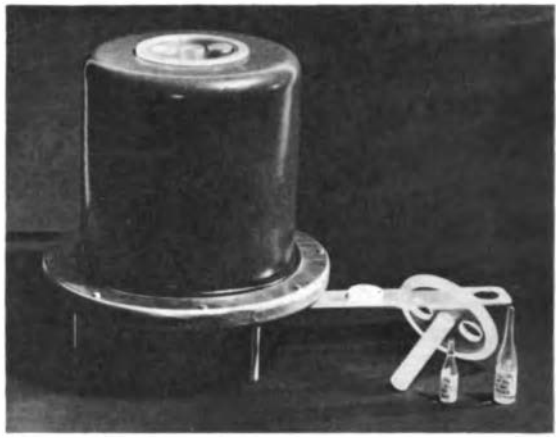


Figure 2. Ionization chamber for secondary standardization of beta and gamma ray sources.

countries on this problem, and a chamber has been designed in Great Britain, jointly by the National Physical Laboratory and Atomic Energy Research Establishment for routine measurements. This is a composite ionization chamber which can be used either as a gamma or beta chamber. It happens that our Chairman, Mr. Perry, is an authority on this subject. May I therefore suggest, Mr. Chairman, that you speak about this chamber.

PERRY: Figure 1 and Figure 2 illustrate the chamber. The first is a diagram which is self-explanatory. The gamma ray chamber consists of three coaxial cylinders; the beta ray chamber occupies the lower portion of the collecting electrode. The beta rays enter the chamber through a 0.68 mg/cm^2 duralumin window. The design is a compromise between that required for accurate secondary standardization and that for routine measurements in hospitals. The diameter of the central cavity which takes the gamma ray source is rather larger than one would choose for accurate work. Also it seemed desirable to provide for the measurement of beta ray sources in liquid form; hence, the thin window to separate the chamber from the polythene dish holding 1 ml of liquid. The outer cylinder is covered with an insulating material because it is at high voltage (i. e., 100 volts).

The original idea was to make a simple inexpensive chamber but it has not proved as cheap as we expected. It also took much longer to get into production than was anticipated. We made and tested the prototype at National Physical Laboratory in a few weeks, but it took about 2-1/2 years to get the production model. We received the first batch of ten chambers in March of this year. The tests of these chambers with a few gamma ray sources (sodium-24, cobalt-60, iodide-131, gold-198, and radium-226) showed that different chambers agree in their response to gamma ray sources to within ±1 percent. For beta ray sources (including sulfur-35) the limits are ±3 percent. We have now received 40 of these chambers, 30 of which are at the moment being tested; they appear to have much the same characteristics as the first ten.

Figure 2 shows the chamber with the polythene dish on the slide and the jigs which take the standard National Physical Laboratory ampoules containing 1 ml and 4 ml of solution, respectively. Another jig has been designed to take the 10 ml bottles in which solutions are normally supplied in the United Kingdom.

PUTMAN: In the beta gamma chamber just described the sensitivity of the gamma chamber is to some extent dependent on position of the source along the axis, but Mr. Perry has said why a chamber of this shape was chosen. As might be expected, an absorber placed in the bottom of the gamma source well can be used to produce a region of nearly constant response near the

middle of the well. We have found that with a brass absorber one centimeter thick, a two or three centimeters wide flat response curve can be obtained for gamma rays as different in energy as cobalt-60 and gold-198. An objection to such a system is that the actual efficiency of the chamber over this regon is quite a sensitive function of the absorber thickness, and is also a function of the length of the source, if this is over 3 or 4 centimeters. In the interests of simplicity and reproducibility, absorbers were omitted from the present design, in which the measurement of an accurately centered source is more independent of its length.

I should like to mention one other technique which may be of interest in determining the radiochemical purity of standards. We have found it convenient to use a simple gamma ray spectrometer for this purpose. Further advantage has been gained by using a twin spectrometer, incorporating two sodium iodide phosphors, as nearly identical as possible, connected through photomultipliers and exposed to different sources. These two detectors are connected to a subtraction device which merely switches over their responses every half second, and subtracts one from the other. The difference spectrum thus obtained with a single-channel kicksorter is plotted automatically. The electronics is identical with that used by D. H. Peirson for eliminating the Compton spectrum by subtracting the output of an anthracene phosphor from that of a sodium iodide one exposed to the same source. In the present arrangement, instead of using two counters measuring the same source, we use two counters exposed to different sources, and we can now subtract the gamma ray spectrum of source B from that of A.

We have found this valuable for analyzing mixed sources, in which any component, once recognized, can now be eliminated by subtraction. The method was developed in the first place for industrial applications involving small quantities of impurities determined by activation analysis. Its application to studies of radiochemical purity is illustrated by some measurements which were done by my colleague, W. H. Taylor. A composite source of selenium-75 and mercury-203 was simulated by placing together samples of the two isotopes on separate foils. In this way a composite spectrum was obtained, in which the mercury-203 spectrum was completely obliterated by that of the selenium. This composite source was placed in front of the first detector of the twin spectrometer, and a pure selenium-75 source was placed in front of the other detector. By adjusting the distance of the pure selenium source from its detector, the difference spectrum was arranged to give zero response at energies above those of mercury-203 gamma rays. The result was a beautiful reproduction of the gamma ray spectrum of mercury-203, which was correlated with the spectrum obtained on removing the selenium component of the composite source.

The agreement between the amplitude of the difference spectrum and the true spectrum of the mercury-203 was, in fact, quantitative within 5 percent. I think this is quite a reasonable method for finding relatively small impurities which may otherwise be swamped in a complex spectrum of gamma rays.

KOFOED-HANSEN: Since 15 hours of irradiation time was used in comparisons with the pocket chamber, what was the RC value of that chamber?

PUTMAN: An attempt was made to measure the leakage during the 15 hour period and it was found too small to measure.

KOFOED-HANSEN: The radiation may damage the insulator. We once had some small chambers of that kind and in 200 days under normal conditions, they showed no leakage. But after heavy radiation, they were discharged in 10 days. This will make quite a difference. This is visible after you remove the source.

PUTMAN: I believe that no specific tests for this affect have been made, but one would have expected this to be evident in the analysis of results. I am not sure whether polytetrafluoroethylene (Teflon) insulation is affected in this way.

PEACOCK: In polystyrene, you can get leakages during irradiation of the order of 2 percent

per week where the separation is of the order of a millimeter. Actually, over a considerable radiation range, it tends to improve rather than get worse. I do not know whether that holds if you get up to many millions of roentgens, but it probably holds up beyond 30 million roentgens.

GROSS: We have done a few experiments along these lines with ionization chambers, particularly in trying to make high dose chambers. We have tried practically every insulator we could find commercially available, and we find, especially at high dose rates that the leakage is just terrible. It does continue afterwards, but decays away so that they ultimately return to good insulating properties.

GEIGER: I have an additional remark about the source comparison measurements. We have made comparison measurements on a similar basis, essentially using cobalt sources, and we obtain different ratios when we use air equivalent chambers for the measurements and when we use our radium measurement machine which is an aluminum chamber lined with lead. This can give differences up to one or two percent. This seems to be due to a difference in the source encapsulation. The source encapsulation is very important when you are making measurements with air equivalent chambers. You get additional Compton scattering. This you would not get with the heavily shielded radium measurement chamber. For this reason when we measure cobalt sources routinely we report the strength in roentgens per hour at one meter output and not in millicuries. This is something that people who buy the sources are mainly interested in anyway.

PUTMAN: In the measurements which I have been describing, the primary interest is also in the effective activity of the source. In fact, in order to reduce it to absolute activity figures one has to be sure that the source size and encapsulation, besides the distribution of activity throughout the source, is the same in both cases. Of course, in making a cobalt-60 source by neutron irradiation, the internal screening of the neutrons produces a non-uniform distribution of activity through the source. All of these aspects enter into the picture. But provided one is comparing sources of the same type, of the same isotope, and provided they are physically identical, that is of the same size and shape, and identically encapsulated, I do not see how one can get variations in this way.

GEIGER: That is correct. I just wanted to point out that one has to be rather careful.

SELIGER: I would like to inquire of Dr. Hayward if he has any opinion why there should be so much difficulty in the determination of the conversion coefficient for the barium-137 gamma ray. There is a large discrepancy in this value and it does not seem to be too complicated a decay scheme.

HAYWARD: Offhand, I do not see why it would be difficult. There is a high energy beta group in cesium-137 that goes to the ground state which has to be subtracted off from the beta group that goes to the 660 kev level in barium-137 and this may lead to some trouble in determining the conversion coefficient. Other than that, I do not see why a good job could not be done.

NOVEY: I do not think it is as easy as it appears on the surface, because you have the problem of determining the total area under the spectrum as measured on a spectrometer because you cannot go to zero energy. You have to make some sort of a normalization. If you know that the boundaries of the conversion coefficient are 10% ± 1% or 2% this means 20 percent error in the conversion coefficient, but only a one or two percent error in an area measurement.

SELIGER: The conversion coefficient has been reported from six to nine percent. That is a rather large discrepancy.

NOVEY: I think figures have settled down somewhat better than that. It is true that nowadays one ought to be able to determine the absolute gamma ray emission. It is just not an easy problem. When you look into it, you find it is much more difficult than it appears. This is true in general of conversion coefficient measurements.

SINCLAIR: There was mention of the agreement that now exists on W. I wonder if you could say what the agreed value is.

PERRY: I do not know that there is an agreed value but the International Commission on Radiology now recommends 34 ev.

SINCLAIR: I am sure Dr. Gross will hardly agree with this. A number of other measurements that have been made during this year seem to be coming out at around 32.7 ev.

GROSS: No, I am sorry. My value is 33.6 ev.

SELIGER: I think possibly Mr. Perry might point out that the 34.0 is a rather arbitrary value arrived at around a conference table and not in a laboratory.

PERRY: I think that has been the case for a number of years.

GROSS: The three most recent values I have seen are the National Bureau of Standards', ours and Jesse's. I think Jesse gets 34.1, which is right on the 34.0 value. The Bureau got 33.7, and we got 33.6.

SINCLAIR: There is a value that is of fairly high precision. I think it was 33.1.

SELIGER: You are correct.

SINCLAIR: This was done again with slightly better precision still and I was told unofficially it was 32.7. It is not published yet. I just wanted to point out that there is still some doubt about this number.

VINCENT: I have a question with respect to the National Physical Laboratory chamber. Have you done any tests on this chamber with a gamma emitter having the same activity but a different amount of liquid to test the dependence of the activity on the amount of liquid?

PERRY: There were one or two limited tests on the National Physical Laboratory prototype, but since that differs in some respects from the commercial one, we are making tests on the commercial model now.

3. Gamma-gamma Coincidence Counting

R. W. Hayward

I intend to make this a rather brief report on some of the gamma-gamma coincidence counting measurements that we have made on cobalt-60 and then end up with a provoking remark and try to escape quickly.

Cobalt-60 at the present is rather unique in that its decay scheme is kinetically quite well understood compared to many other radioactive isotopes which, presumably, have settled decay schemes, and then something new turns up. I think cobalt-60 is in rather good shape. It decays by the emission of a beta group with maximum energy of 310 kilovolts or thereabouts, followed by two cascade gamma rays of 1.33 Mev, and 1.17 Mev.

The spin of cobalt-60 and nickel-60 are both known. The measured spin of cobalt-60 is 5 and that of nickel-60 is 0. The spins of the two intermediate states have been determined independently by several means, and of course they are based on the ground state spin of nickel-60, and will have spins of 2 and 4. That means that all the radiations involved here are pure radiations.

The lifetimes of these intermediate states are known to be less than 10^{-12} seconds, so that the angular correlation between the two gamma rays can be, to a very high degree of accuracy, the theoretically expected one.

There is a problem in counting cobalt-60 by gamma-gamma coincidences because there are some secondary effects that one has to take into account if one really wants to get precision in coincidence counting. The first of these is the fact that the two gamma rays are very close to one another in energy. You cannot in general distinguish one gamma ray from the other. If you were to be able to look at the pulse height spectrum of these two gamma rays separately--this, of course, is an impossibility, but by looking at such things as zinc-65 and sodium-22 one has a pretty good idea of what they look like--you would get a pulse height spectrum versus intensity for the 1.17 Mev gamma ray that would look like the figures that Dr. Lazar showed. You have a total absorption peak where the full energy of the photon is dissipated in the crystal, then a Compton distribution where some of the photon energy escapes, and then you usually have a back scattering peak from the photons interacting with the walls of the room or other objects and scattering photons back into your detector. You might say that the spectrum of the 1.33 Mev photon is quite similar.

Actually if your resolution is good you can resolve these two peaks fairly well. But the point is that one can make a rough efficiency calculation and determine that the total area under the 1.33 Mev spectrum is less than the area under the 1.17 Mev spectrum. So if one is going to do coincidence counting, one wants to know the relative efficiency of both counters for each of these two gamma rays.

One can allow for this by, say, looking at the pulse height spectrum integrally where you bias at a certain point and look at only the pulses larger than a certain height, and you arrange this bias so that the relative areas under these two curves are approximately equal. You also adjust your bias high enough so that you completely eliminate the possibility of a photon going into one of your coincidence counters and scattering back into the other and registering a coincidence. This will usually turn out to correspond to a pulse height corresponding to 300 or 400 kilovolts.

We would like to see what sort of error is introduced if you do not set this bias quite correctly. The expression which can be derived for the disintegration rate is

$$N_0 = \frac{N_1 N_2}{N_c} \frac{Z_1 Z_2}{(Z_1 + 1)(Z_2 + 1)} \epsilon(\nu)$$

where N_1 and N_2 are the counting rates in counters number 1 and 2, respectively. N_c is the coincidence counting rate. Z_1 and Z_2 are the ratio of efficiencies for the two gamma rays in counters 1 and 2, and $\epsilon(\nu)$ is a term taking into account the angular correlation between the two gamma rays. When $Z_1 = Z_2 = 1$, you have set your bias correctly, and you can see that this term reduces to one-half. But say you did not set your bias correctly and $Z_1 = 1.1$; it turns out that if it is 1.1 this term will be one over 2.004. In other words, you have made 0.2 percent error in your disintegration rate due to the missetting of the bias. If you are as far off as 1.2, then you make about a one percent error in this factor.

So really this is not too sensitive to the bias if you set your bias so that the relative efficiencies are approximately equal, say, within 10 percent. Then you make a 0.1 percent error in your determined disintegration rate.

At first sight, it looks as if this angular correlation term is a complication, but really it is very nice. It allows you to make several independent determinations of the disintegration rate under different geometries. Say you have a situation where your two counters are opposite one another and the source in between, you will have one value which gives you a low disintegration rate, and then you have another situation where you count coincidences between two counters oriented at 90° with respect to one another, and you get a different disintegration rate. One should be able to get a ratio of these apparent disintegration rates that would be just equal to the theoretical angular correlation between the two gamma rays. If you get this, it is an indication that you are not being bothered by scattered gamma rays and things of this nature, which is the chief reason why in the early days of angular correlations, people did get spurious angular correlations.

One has to evaluate the angular correlation term to take into account the finite resolution of the counters--the finite angular resolution. There will also be certain factors that attenuate the angular correlation from the theoretically expected one. These can be taken into account in a similar manner to the calculations that Dr. Lazar mentioned for determining the efficiency of a scintillation counter.

When this is done, it is possible to consistently get results that agree with one another within a tenth of a percent. Various members of the radioactivity section have done coincidence counting and can get a tenth of a percent accuracy for the absolute disintegration rate of cobalt-60 sources in the range of about 10^4 or 10^6 disintegrations per second. One can actually go up a strength of the order of 10 millicuries, but there you really have to use very fast electronics and you get into difficulties of missing true coincidences.

We have been quite successful in getting internal agreement between absolute disintegration rates as determined with 4π beta counting and gamma-gamma coincidence counting and beta-gamma coincidence counting, which is a third independent method that one can use. Of course, there is a fourth independent method for determining cobalt-60 just by observing the gamma rays, such as Dr. Lazar mentioned. So with cobalt-60 we have a readily understood disintegration scheme. We have approximately four independent ways of determining its disintegration rate. Because cobalt-60 now far outweighs radium in its uses for radiology, industrial applications and such, it seems to me that it is high time that we did establish standards of cobalt-60 gamma ray emitters in the region of millicuries up to a curie.

It seems to me that with the complications of the radium disintegration scheme, which even to this day people really do not understand, I would like to have something simple that can be understood. A problem such as self-absorption in a thick source can really be handled quite easily in the case of cobalt-60 because you know precisely what the radiations are that are present. This is the question or the provoking remark that I would like to make. Should we establish cobalt-60 as a primary gamma ray standard and abandon radium or maintain the two for a while until everybody gets convinced, or forget about the whole thing?

There is one question that I know somebody will ask--what about the half life for cobalt-60?

It is 5.25 years with an uncertainty in the second decimal place. If you established it as a standard, everybody would be looking at this for the next few years, and in time there would be a good value for the half life. The ease of intercomparison of cobalt-60 sources, as outlined by Mr. Putman, makes it a really valuable standard.

MANOV: I want to second Dr. Hayward's idea of a cobalt standard for the curie range. I think this is an excellent idea. Whether you get up to high strengths and you standardize the source or standardize an instrument as proposed, are two alternative approaches. But in industry and in the Atomic Energy Commission we are considering one source of 2 million curies of cobalt-60 in one place for irradiation in the preservation of foods. We have to get beyond the millicurie range. I want to put in a very strong second to your suggestion that we go ahead in this direction.

ATEN: How good is the half life of cobalt at present?

HAYWARD: It depends on who you believe. It is much better than it was a few years ago. I said I think it is 5.25 years with the last figure uncertain.

MANN: It does not matter anyhow. You can calibrate to 0.1 percent. As Dr. Hayward said, this can be done any time, every six months or every year. This is a minor exercise.

MANOV: At one of our meetings of the subcommittee last year, we looked over the half life of cobalt-60 in the literature and we found a range of values as you intimated. But we found something else that puzzled us, and I am not quite sure how the story came out; namely, for a given sample of cobalt-60 the half life with the time seemed to be getting shorter, not longer. I think the people who were at the meeting will remember that. I would like to ask what was in chapter 2 of this very interesting story. Does anyone recall what happened?

HAWKINGS: I think we were probably the instigators of this in the sense that in the year 1953 there was an intercomparison of cobalt-60 and the agreement, as I recall, was within 0.2 percent. At that time we remeasured one of the original National Bureau of Standards cobalt-60 standards that was issued in 1947. If we can believe the measurements which were made at that time, this source must have decayed with a half life shorter than 5.2 years. When this came up for discussion at the last subcommittee meeting, the arrangement was that we would try to get another intercomparison of cobalt-60, and if agreement was as good as it had been in the last, we would undertake to remeasure the previously issued standard of 1953 and thereby get another check on the half life.

SELIGER: We retained at the National Bureau of Standards several samples of primary cobalt solution which was standardized in 1953, and we have during the past several years been measuring the half life of these solutions. We have found that the half life has been increasing slightly as a function of time. At the present time it appears to be fairly stable and the half life comes to, I think, either 5.27 or 5.28 years. The half life over the first six months of 1953 or maybe the last six months of 1953--this was just at the time when we had our 4π gamma chamber in operation--was about 5.2 years. That is, 5.20. The precision of these measurements is much, much smaller than the difference between 5.20 and 5.28. It is about 0.01 or 0.02.

So there does seem to be something in the cobalt, and I have not any idea what it could be, that does make the half life increase slightly with time. I think this has been noticed by some of the people at Oak Ridge who have been measuring cobalt half life.

PERRY: If I may add to what Dr. Seliger said, we have also made measurements of the solution that was distributed in 1952 by the National Bureau of Standards. These measurements were made over a period of 4-1/2 years.

SELIGER: That is quite good. It is in fairly good agreement with our own.

PERRY: This compares very favorably with the result of measurements we have made on cobalt-60 from Harwell, which was 5.260 ± 0.034 years; the 0.034 is the standard deviation from the mean of the measurements of six sources over a period of 7-1/2 years. The method is the gamma ray ionization chamber comparison with radium sources under fixed geometrical conditions.

SELIGER: That was the same condition as our own.

REYNOLDS: The latest value that we have for Oak Ridge is also 5.30. This is not a very recent value. The measurement is still going on. I do not have the most recent value. I might remark that the earlier value was a little higher, if the members of the subcommittee remember. The value moved down to 5.30 from 5.38 earlier. But the 5.38 value was on a rather short time span.

GEIGER: We have measured the half life of cobalt over the last 7 years in comparison with radium and the latest value we found is 5.24 ± 0.034 and two years ago we found 5.21.

PEACOCK: Do you have a value based on the latest two or three years?

GEIGER: No.

SELIGER: Our value is based on the last two years, I believe.

GEIGER: This value is based on the last seven years.

HAYWARD: I should like to point out that all these values quoted, I think, are based on the gamma ray intensity. If there is something increasing over the years, it looks like there might be a very weak shorter lived impurity which would tend to give you a change in value. I think some effort should be made to observe this decay by various methods which might not depend on the gamma emitting impurity.

KULP: How much care has been given to the radiochemistry in each of these? Has everyone done the same thing?

HAYWARD: I haven't the slightest idea.

MANN: We had a very new and very old specimen of cobalt-60 to try to find out what this peak contribution might be.

PERRY: We can supply one of our sources for that purpose.

MANN: We might be able to pick up something that way.

HAWKINGS: We have examined the spectrum of a number of cobalt samples of various ages and as you might expect you cannot detect any difference. This is not too surprising in that an impurity of less than a few percent will hardly be detectable in the spectrum.

Another thing is that any future measurements of the half life of cobalt-60 should be made on cobalt that has been purified by radiochemical procedures because cobalt is now being made in higher and higher neutron fluxes all the time. If there is any side reaction which is interfering with it, the situation is going to change.

REYNOLDS: One possibility for an impurity is actually an isotopic impurity, cobalt-58, which would show up if one happened to get some cobalt which had been produced in a high fast flux and not been cooled very long. The half life of this isotope is 71 days.

HAWKINS: This is far too short to be of any consequence over a period of several years.

REYNOLDS: If one bases the value on all of the results over a period of several years starting when you had cobalt-58 present, then the over-all value would be different.

KIPFER: I would like to ask if Dr. Mann thinks that such a cobalt standard will replace the radium standards. A lot of people would be very glad to get rid of the radium standard because the problems involved seem quite insoluble.

MANN: I think that radium does still have one use. We had some discussion earlier about the requirements of the people who put out radium commercially--they are usually not more than two percent different from the calibrated values given by the Bureau of Standards, and I think this whole thing is a hangover from another age. I know that antibiotics, where you pay $16 for ten chloromyecetin pills, are far more expensive than radium, and I am sure it is not precisely two percent. It does not go to the Bureau of Standards to be measured. I think it is completely ridiculous with respect to radium. It does have one use and that is in calibration of calorimeters. We did have the recent interesting application in which Lord Rothschild in England was doing some work, and in wanting to calibrate, he asked our advice. We suggested that he take a radium source and get Mr. Perry to compare it with the British Hönigschmid standard which has been measured in microwatts. This has been known and has been tied in with more standards. Microwatts in my view is a much more fundamental unit than a Hönigschmid milligram, which is not even tied to the standard milligram.

PERRY: While we are discussing this half-life business we are losing sight of the main part of Dr. Hayward's paper. I do not know whether there are any questions on the gamma-gamma measurements.

PUTMAN: Might I raise the question as to whether the reproducibility which Dr. Hayward quoted of ±0.1 percent, and then in the same breath quoted the accuracy, are necessarily the same thing? Is the accuracy of ±0.1 percent of cobalt-60, to which Dr. Mann referred, a real accuracy or is this reproducibility?

HAYWARD: I would say it was the reproducibility.

PUTMAN: Could I make one other point about the use of cobalt-60 as a standard? This raises a little the philosophical question of what is a standard. A standard I would have thought was not something which has to be set up and remeasured every six months as Dr. Mann suggests. Surely there is a place for radium or some long lived high energy gamma emitter in maintaining reference chambers for measuring the cobalt-60 and such isotopes which can be checked by absolute measurements periodically but which should still have a reference which can be referred back to previous years not depending on half life.

MANN: I was making the same point in my discussion earlier that we still have use for radium for maintaining calibrations. I do feel this other use of standards is the proper use of a standard because the measurement in one country in microwatts is used in another country to calibrate the calorimeter. I think this is a proper use of the standard. I also agree we do want to continue our calibrations and we indeed are at the National Bureau of Standards and I am sure you are at National Physical Laboratory.

PERRY: Yes.

PUTMAN: Could we ask, to what extent in this case an ionization chamber can justifiably be calibrated or even held to its calibration by use of a gamma emitter emitting different gamma ray energies from the ones which we are trying to measure? In other words, are we justified in using radium as a long lived standard in the sense that it will maintain the behavior of an ionization chamber over a period when the ionization chamber is to be used for measuring cobalt-60 or cesium-137?

MANN: Yes, assuming constant geometry of the gamma ray chamber, I think this is

justifiable, and also provied that you do measure the current and keep a check on the current. That is, the value of the current and K constant do not change with time in radium, let us say. I believe this is your practice, Dr. Seliger.

SELIGER: Yes. We essentially use a double check of measuring the absolute current and also the ratio.

MANN: Here you are pretty safe. In a sense the chamber plus the radium is the standard. The radium is not the standard; neither is the chamber by itself. This is the only way we can maintain a primary standard.

PUTMAN: Supposing the chamber characteristics were to change, however. A proportional change in the radium response would not necessarily reflect in a proportional change in the iodine.

MANN: That is right. We check every so often. We do preliminary 4π beta counting and check our chambers. So far we have not run into this difficulty. It would be a catastrophe if we did. It would be a serious error.

SELIGER: I would like to point out that due to the nature of the ionization which is produced inside the sensitive volume of the ion chamber that aside from changes in geometry the only other changes with time, it seems to me, that could occur might be possibly a surface effect whereby the secondary emission from the surface might change with time, either due to oxidation or some other process. I think that the fact that it may not be in the same proportion, that it may be energy dependent, would give a second order effect in the ionization chamber measurements. At the present time I do not believe that it is our purpose to maintain these chambers to give a long term precision any better than 0.5 percent. We have not had any difficulty doing this for the past four years.

PERRY: Perhaps if I attempt to sum up, the idea seems to be that radium as a fundamental standard is rather unsatisfactory. Cobalt is probably the real answer to the fundamental radioactivity standard. But there may be a transition stage where we shall have a radium source as a reference source rather than a fundamental standard and in that transition period we may be able to establish cobalt to displace radium. I do not know whether that is a fair summing up.

MANN: With one addition, if I may remind you, at the International Commission of Radiological Units meeting 18 months ago National Physical Laboratory and the National Bureau of Standards were given the task of producing such standards for cobalt. As far as I know, nothing has been done.

PERRY: We have done a little but not for that specific purpose.

SESSION IV - ELECTRON-CAPTURE NUCLIDES AND EMITTERS OF LOW-ENERGY RADIATION

S. A. Reynolds, Chairman

Summary

One may attempt to measure electron-capture nuclides in three ways: (1) by counting auger electrons; (2) by counting X-rays; and (3) by measuring X-ray - internal bremsstrahlung coincidences. The counting of X-rays is the technique which has been studied most, and it appears to be best. The principal uncertainties are due to lack of knowledge of K fluorescence yields and K capture/L capture ratios. Groups at Harwell and Chalk River have used high-pressure 4π counters successfully. End-window counters have also been used, and a few intercomparisons between 4π and end-window counters have shown good agreement. X-gamma coincidence counting data confirm the measurements. Measurements with ionization chambers have also been made.

Some details of inner bremsstrahlung spectra were discussed. It appears improbable that electron-capture nuclides can be standardized by counting X-ray - bremsstrahlung coincidences in any simple way.

Improved techniques for measurement of beta activity in the gas phase have been developed and employed for measurement of krypton-85. They will also be used for carbon-14 and sulfur-35.

An intercomparison of measurements of promethium-147 has been made. The mean deviation of valid results was about four percent.

Our subject is measurement and standards of electron-capture nuclides and nuclides which emit low-energy radiation. We shall hear of techniques where the accuracy, at least in terms of intercomparisons, is of the order of a few percent, in contrast to the numbers around 0.1 percent reported in the first three sessions.

1. Electron Capture Measurements

R. A. Allen

I would like to remind you first of the problems involved in attempting to standardize electron capture nuclides. Electron capture is followed by the emission of either an X-ray from the K or L shell or an Auger electron. In certain cases the X-ray is accompanied in coincidence by an internal bremsstrahlung quantum. We therefore have three types of radiation which we might attempt to measure in order to standardize such nuclides.

The Auger electrons from low Z nuclides have very low energy. For example, the electrons emitted from chromium-51 have energies of the order of four to five kilovolts and less. Therefore, using normal source preparation techniques, it is very difficult to get all the Auger electrons from the source into the counting volume of a gas counter or into a crystal.

X-rays emerging from electron capture nuclides are mono-energetic. They are of fairly low energy. We feel there is more chance of determining the number of X-rays accurately than if one tries to measure the number of Auger electrons.

Finally, one may, as suggested by Dr. Rasmussen, attempt to measure the coincidence rate between K X-rays and internal bremsstrahlung. This is in principle a very elegant method but one must have exceptionally pure samples of material. There must be no gamma radiation emitted, it has been calculated that in iron-55, for example, the presence of 0.01 percent of iron-59 ruins the method.

We at Harwell decided to concentrate on measurements of X-rays from the lower atomic number nuclides (chromium-51, manganese-54, iron-55 and zinc-65) in the first instance. With the exception of iron-55 these nuclides have the advantage that some or all of the electron capture transitions are followed by gamma radiation, therefore one can make X-ray gamma coincidence measurements and so get two independent methods for determining the source strength. I will describe the piece of apparatus which we used to determine the number of X-rays which are emitted by these various materials. We used a 4π counter which consists of a cylinder subdivided by a plate onto which the source could be mounted. We, therefore, have in effect two D shaped cylinders with a counter wire passing lengthwise through each half.

The procedure was to lay down a source--an evaporated deposit from an aliquot of the stock solution--onto an aluminum foil about 0.8 μ (microns) thick and cover this source with another similar aluminum foil. In the case of chromium and manganese this thickness of foil is sufficient to absorb out all the Auger electrons emitted by the source. We have 4π geometry for counting X-rays and we were pleased to find that even in this rather awkward geometry the resolution was quite good. A graph, plotted auto - matically, relating number of counts and energy is shown in Figure 1.

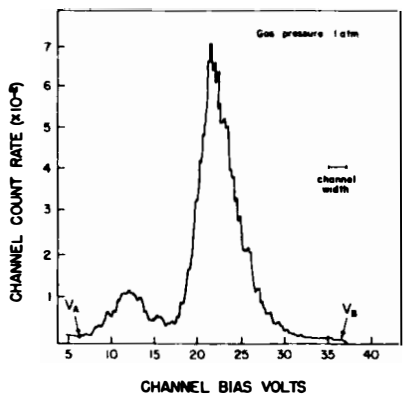


Figure 1. Pulse height spectrum of Mn-54 K X-rays in 4π proportional counter.

The filling gas was a mixture of 90 percent argon and 10 percent methane. The pressure in the counter was slowly increased and repeated runs of the spectrum were taken, adjusting the voltage to give the same peak pulse height at different pressures. The number of counts with bias set at V_A and also at V_B were measured; the difference was taken as the number of K X-rays detected. The logarithm of the number of X-rays counted was plotted as a function of the inverse of the filling pressure and we obtained a series of points lying on a straight line as shown in Figure 2.

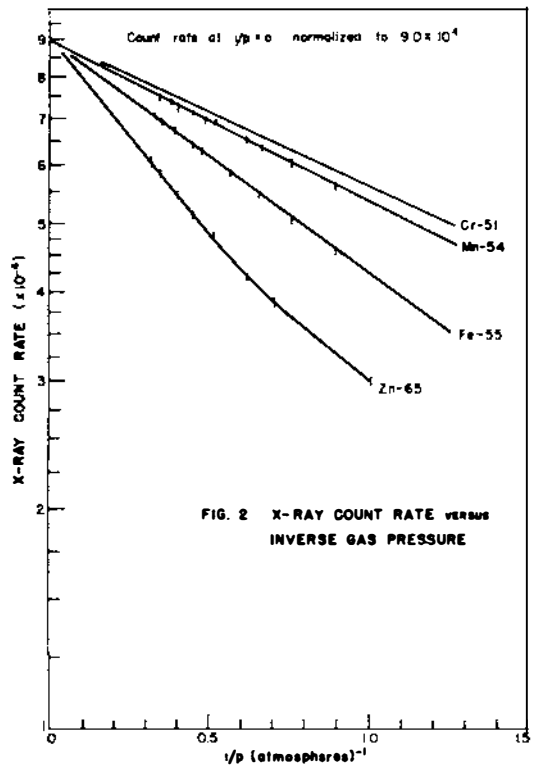


Figure 2. X-ray count rate versus inverse of gas pressure.

Extrapolation of this straight line back to zero is in effect an extrapolation to infinite pressure when all the X-rays will be stopped in the counter, and therefore will all be detected. This extrapolation, then, gives us the total number of X-rays N_K being emitted by the source.

ARNOLD: We did make up another large 4π counter. The one we used is approximately one and a half inches in diameter and about four inches long. We made one which was six inches in diameter and about 14 inches long, this should have had 100 percent efficiency at one atmosphere or so for chromium or manganese X-rays. In fact, the counter was so large, and the counting losses seemed to be rather large, that the apparatus was not really accurate at all and we abandoned the idea of using a large counter.

BORKOWSKI: You can in effect do the same thing by changing the gas.

ALLEN: That is true. We had to do that a little later in fact.

BORKOWSKI: We performed a similar experiment using both krypton and xenon, and in effect the same linearity and total absorption was observed in xenon gas.

ALLEN: Having determined the total number of X-rays, to get the total disintegration rate, one has to assume some figures for the K to L ratio, and for the K fluorescence yield in each of the nuclides concerned. One simply multiplies N_K by $1/\omega_K$ and $(K + L)/K$ to give the total number of disintegrations.

At present we have used only the theoretical figures for ω_K as are given in Burhop's book on the Auger effect, and K to L ratios as given in the article by B. L. Robinson and R. W. Fink in "Reviews of Modern Physics" (1955).

In order to check the results, we made K X-ray gamma coincidence measurements on the three nuclides. The 4π counter was used as the X-ray detector, and a one-inch sodium iodide crystal was used as an external gamma ray detector. This coincidence arrangement very nearly satisfies the criterion that one of the counters shall be equally sensitive to all parts of the source, provided the source is fairly small in diameter and the crystal is at a reasonable distance from the source.

The agreement of the results obtained by this method and those obtained by the pressure method using theoretical constants was between one and three percent. This can be considered two ways. One can either look at it as a confirmation that the pressure method is correct or one can look at it in the light that the product of theoretical value of $(K + L)/K$ times ω_K is not wrong by more than one or two percent.

There is one other correction. The source is sandwiched between two thin pieces of aluminum, and these, of course, absorb some of the X-rays. We believe the sandwich technique is, in general, suspect, but in this particular instance, where the radiation emerging from the source is mono-energetic and where all the energy loss is due to photo effect--in other words,

single scattering--we considered that the sandwich method was justified in determining the loss in one sandwich foil. The loss in 0.8 μ aluminum is approximately 10 percent for chromium going down to approximately one percent for zinc-65. The loss can also be calculated by evaluating Gold's integral.

In the case of manganese-54, we did both x-gamma and Auger electron-gamma coincidence measurements; these two and the pressure method agreed to within about three percent. We feel, therefore, that we can supply standards of iron-55, where we cannot check by the coincidence method, to better than ±5.0 percent.

The next thing was to try to use the method for higher Z numbers and we considered strontium-85; again the result can be checked by x-gamma coincidence measurements. There is a 0.9 microsecond delay between strontium-85 electron capture and the succeeding gamma ray. The coincidence resolving time must be increased until there is no further increase in true coincidence rate; we used an over-all resolving time of about 8 microseconds.

We tried strontium-85 with an argon filling in the counter and obtained a curve rather than a straight line on the pressure plot; this curve was extrapolated back (as a smooth curve) to zero. This extrapolation was of the order of 50 percent. The straight line extrapolation in the other nuclides varied from 20 percent for chromium-51 to 40 percent for zinc-65; it was decided to try using a heavier gas in the counter. The next obvious choice is krypton; krypton is the daughter element from the decay of strontium and therefore is on the absorption edge and is transparent to its own characteristic radiation. We used xenon and obtained another series of points which now lay on a straight line. This time the extrapolation was only about 10 percent and the end point was 15 percent higher than that obtained using argon. The K X-ray gamma coincidence results agreed to within 2 percent with the xenon extrapolation.

We are now trying to standardize cesium-131. We have not yet progressed very far with this problem. The K X-ray energy is about 30 kilovolts and the L X-ray has an energy of about 4 kilovolts. We found that with our small 4π counter no K-photopeak was observable. We can only pressurize our counter to about 4 atmospheres. Krypton does not have a much higher absorption coefficient at 30 kilovolts than argon, so for K X-ray measurements we may go to a long counter and defined solid angle arrangements in the way that Dr. Reynolds indicated.

The L X-ray peak, however, does show up very well in the small counter on top of the K Auger electron background. If we knew the true K to L ratio and further, if we knew how many K X-rays were followed by L X-rays then, it being a comparatively simple matter to determine the number of L X-rays it would be comparatively simple to estimate the source strength. What we are going to do is to measure the number of K X-ray - L X-ray coincidences and so attempt to find the number of true coincidences which occur. These can be added to the L/K ratio and so give a method of obtaining the source strength. We have obtained, so far, one curve of K-L coincidences which shows up the L peak quite well.

PUTMAN: Why do we not use 4π scintillation counters for these levels? At this sort of level, where it becomes difficult to count your K X-ray in the proportional counter, you could change over to the scintillation counter.

ALLEN: We hope to do something along these lines.

PEACOCK: I am sure everybody understands what an escape peak is, but I thought it might as well go into the record. You mentioned that you have an escape peak as well as the full energy peak.

BAPTISTA: I have still some doubt about the values at each value of the pressure. In view of the counting geometry is it not to be expected that as the voltage is increased you will not get a flat plateau but a slight increase of counting rate due to the end effects?

ALLEN: We have not found this in our experience though we have not done very extensive measurements on this effect. Our real criterion was that the pulse height curve should be at zero below and above the X-ray peak. When this was the case we assumed that the number of counts was correct.

SELIGER: I should like to make one comment on the method itself. I think the x-gamma or the Auger-gamma coincidence measurements are quite applicable for standardization, but I would hesitate to depend completely on a method that required so large an extrapolation as is the case for iron-55. Even though you have it bracketed I would be a little uncertain about it. At least more than maybe 5 percent.

ALLEN: It is unfortunate that we have not been able to pressurize this counter to a pressure greater than three atmospheres or so and so reduce the extrapolation. We must try using a heavier gas with the low Z nuclides.

PEACOCK: You mentioned the method proposed by Rasmussen on the internal bremsstrahlung X-ray coincidences. Have you tried that too?

ALLEN: No, we have not tried that method, simply because we have no iron-55 which is less than 0.01 percent pure in iron-59. Rasmussen, himself, arrived at this maximum permissible impurity figure.

SELIGER: For strontium-85, what resolving times did you use for your coincidence measurements?

ALLEN: An over-all resolving time of about 8 microseconds. We did several measurements of coincidence rate at increasing resolving times until the rate was statistically constant. We were quite sure that no delayed coincidences were being missed.

SINCLAIR: What values of ω_K and K/L were you using for iron-55?

ALLEN: The figure of 0.351 was used for ω_K for iron and 0.097 for the ratio of probability of L capture to K capture.

SINCLAIR: How do you arrive at this accuracy estimate of three to five percent then?

ALLEN: It is simply the fact that the pressure results agree with the coincidence measurements to three percent or so. Using the same set of theoretical values for the constants and remembering that the iron curve is bracketed by chromium, manganese and zinc, we tentatively assume that the accuracy of the iron result should be about the same as the other three.

SINCLAIR: Your total fluorescence yield would be out by about this percentage, your total ω is about 0.27 or so. We have standardized iron-55 with an accuracy that we thought was of the order of 5 percent by counting both Auger electrons and X-rays. A literature search has given an average value of 0.297 for the iron value. Experimentally, we obtained a value of 0.305 which is quite a bit higher.

LAŹAR: I might say that this is just the region where the data on fluorescence yield shows the most scatter. The latest K/L ratios (theoretical) that I know of are those of Rose and Brysk which were published in an Oak Ridge National Laboratory report. I do not know whether Robinson and Fink's data include it. This is certainly the most extensive calculation that I know of.

REYNOLDS: Dr. Campion has a short presentation on a similar subject.

2. Gamma Measurements and Standards

P. J. Campion

I feel like an intruder since I don't appear on the agenda at this point, but at Chalk River we have fallen into some pitfalls which Dr. Allen avoided, so if anybody is going to enter into this game, they might as well know these traps and take heed.

Figure 1 shows our first high pressure counter. It consists of an eight-inch sphere divided into two halves by a plate across the center. To begin with, we designed this to have a conventional loop type of electrode in both halves, but we found that the large voltages required at high pressures cause the loop to extend physically to the grounded plate containing the source. We, therefore, adopted the horseshoe shaped electrode with a thin wire stretched across the ends of the shoe as shown in Figure 1.

As you can see this results in a considerable dead space in some parts of the counter, which meant that higher pressures were required in order to compress the X-rays into the sensitive volume of the counter. Our philosophy at Chalk River is to cover up the source, in order to absorb out the Auger electrons and to count the X-rays. We also require that we get up onto a "pressure plateau." That is, that the counting rate does not increase as we further increase the pressure.

A typical counting rate plateau, that is counting rate against counter potential, for iron-55 is shown in Figure 2. The gas used was 90 percent argon, 10 percent methane. This plateau was taken at 40 pounds per square inch. By varying the pressure the value of the counting rate on the plateau can be increased until essentially all the X-rays are absorbed in the sensitive volume. Figure 3 shows a pressure plateau where the abscissa is absolute pressure in pounds per square inch, and the ordinate is the observed counting rate along the (conventional) plateau at each pressure. It can be seen that above about 55 pounds per square inch there is no increase in the counting rate, and this rate is assumed to be the X-ray counting rate from this particular source sandwich combination.

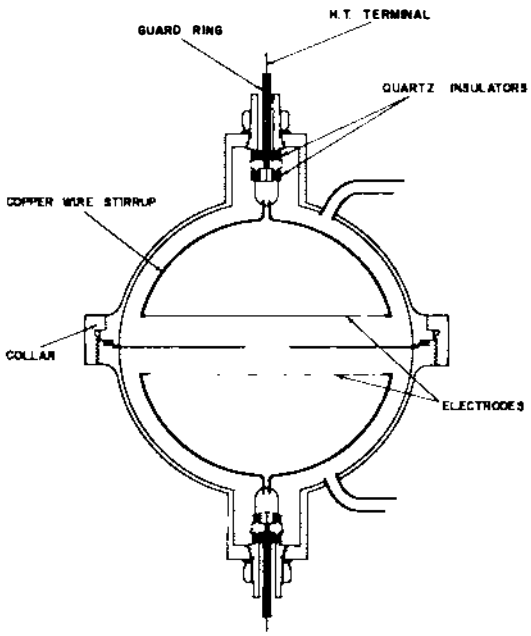


Figure 1. High pressure counter.

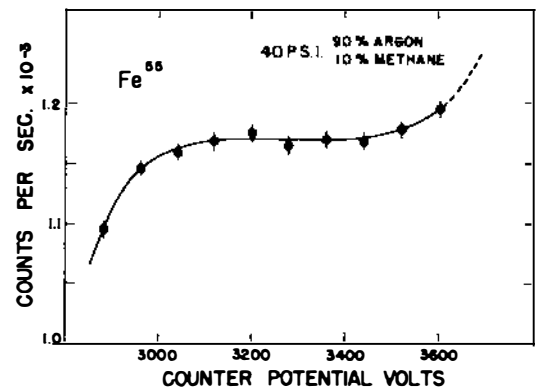


Figure 2. Curve of counting rate versus counter potential data taken with the counter shown in Figure 1.

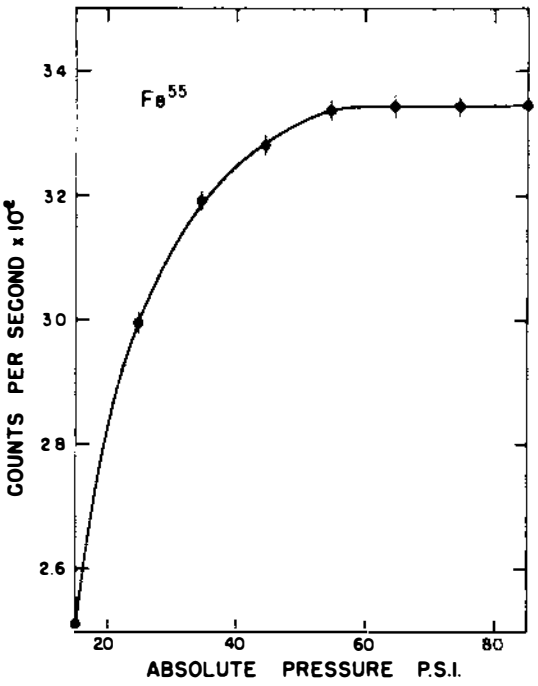


Figure 3. "Pressure plateau" for a source of Fe-55 data taken with the counter shown in Figure 1.

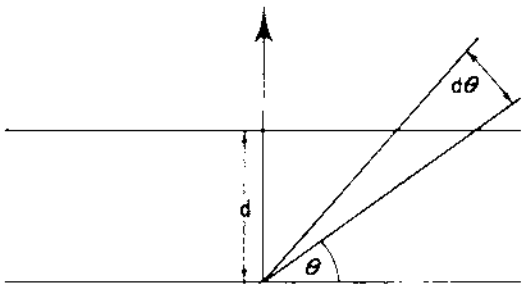


Figure 4. The calculation of the X-ray absorption in a film over 2π steradians.

Intensity of transmitted radiation at angle θ is
$N = N_o \int_o^{\pi/2} \cos\theta \exp(-\mu d \operatorname{cosec}\theta)\, d\theta$.

To solve, put $x = \sin\theta$, whence
$N = N_o \int_o^1 \exp(-\mu d/x)dx$

and making a second substitution $\mu d/x = y$ we obtain
$N = N_o \left| \exp(-\mu d) - \mu d \int_{\mu d}^{\infty} \frac{\exp(-y)}{y} dy \right|$.

This last integral is defined (see for example Jahnke and Emde) as $-Ei(-\mu d)$. Hence
$N/N_o = \exp(-\mu d) + \mu d\, Ei(-\mu d)$
$= f(\mu d)$.

There there is the problem of absorption in the aluminum foil, which is illustrated in Figure 4. This is quite an age-old problem. It was first discussed by Gold in 1908. We found this out only after we had gone through the calculations shown in Figure 4. The calculation is given here in a form more applicable to the present problem than the form given by Gold.

You have an X-ray source situated on one side of an absorber foil of thickness d and absorption coefficient, μ, and it is necessary to know what is the amount of radiation which is transmitted through this foil. The intensity is given by the first expression in Figure 4, which one has to integrate between zero and $\pi/2$. One goes through the calculation and winds up with an expression as indicated in Figure 4. This is defined in Jahnke and Emde as $-Ei(-\mu d)$. We have calculated this expression as a function of (μd) and the result is shown in Figure 5. Just for comparison the normal exponential absorption which one would get with good geometry, i.e., a source a long way from the absorber is also shown. There is a large difference as you see even for quite thin absorbers. We have tested the validity of the equation shown in Figures 4 and 5 by varying the absorber thickness and plotting the observed counting rate N against the function $f(\mu d)$. The results are shown in Figure 6. The points lie on a straight line within experimental errors. The two dotted lines

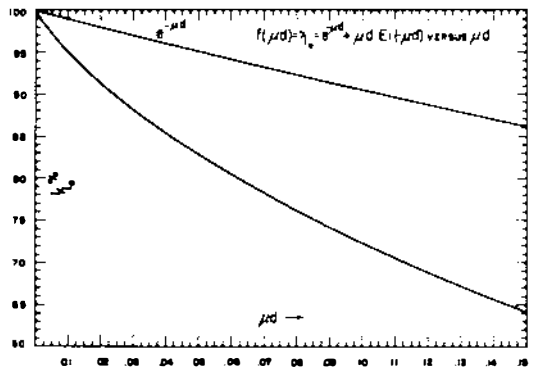


Figure 5. The bottom curve shows the function $f(\mu d)$ plotted against (μd). For comparison the usual exponential function is given by the top curve.

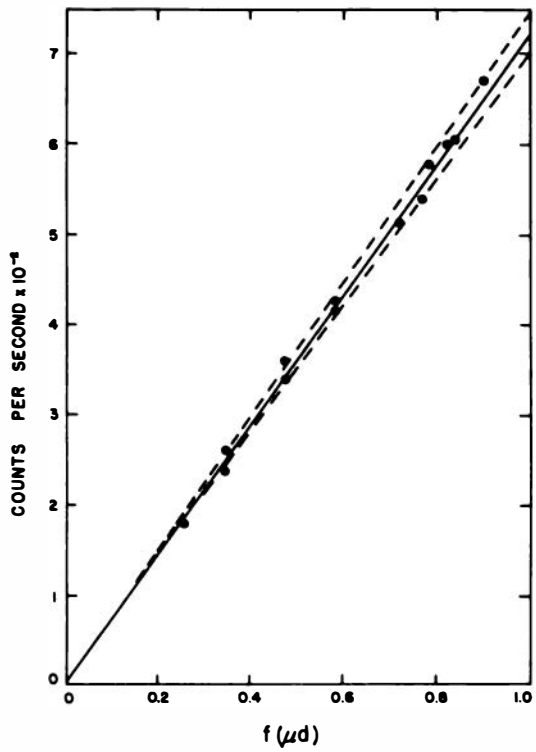


Figure 6. The observed counting rate, measured on the pressure plateau as a function of f(μd).

are considered reasonable extremes. Extrapolation to zero absorber thickness, i.e., f(μd) = 1.0, yields the absolute X-ray counting rate of the source.

We were not satisfied with the first counter that we built, which was designed to withstand 50 atmospheres pressure. We tried it with zinc-65 and did not get any plateau at all. This was attributed to the dead space mentioned earlier and the fact that zinc-65 emits radiations such as positrons and gamma rays which produce ionization across the whole volume of the counter. So another counter was designed to get rid of this, and a sketch is shown in Figure 7. This is the high pressure counter in use at present at Chalk River. It is cylindrical, being essentially two D shaped counters with a wire down the middle of each D. The internal dimensions are roughly six inches in diameter and nine inches long. The source is situated in the middle of the source tray. This counter is designed to go to 2,000 pounds per square inch, although we have not operated the counter above 500 psi as yet.

Gratifyingly enough, we can now count zinc-65 with this counter, and Figure 8 shows a (conventional) plateau obtained with zinc-65. The plateau has a slope of one-tenth of a percent per 100 volts. The high tension is now plotted in kilovolts. This data was obtained with 100

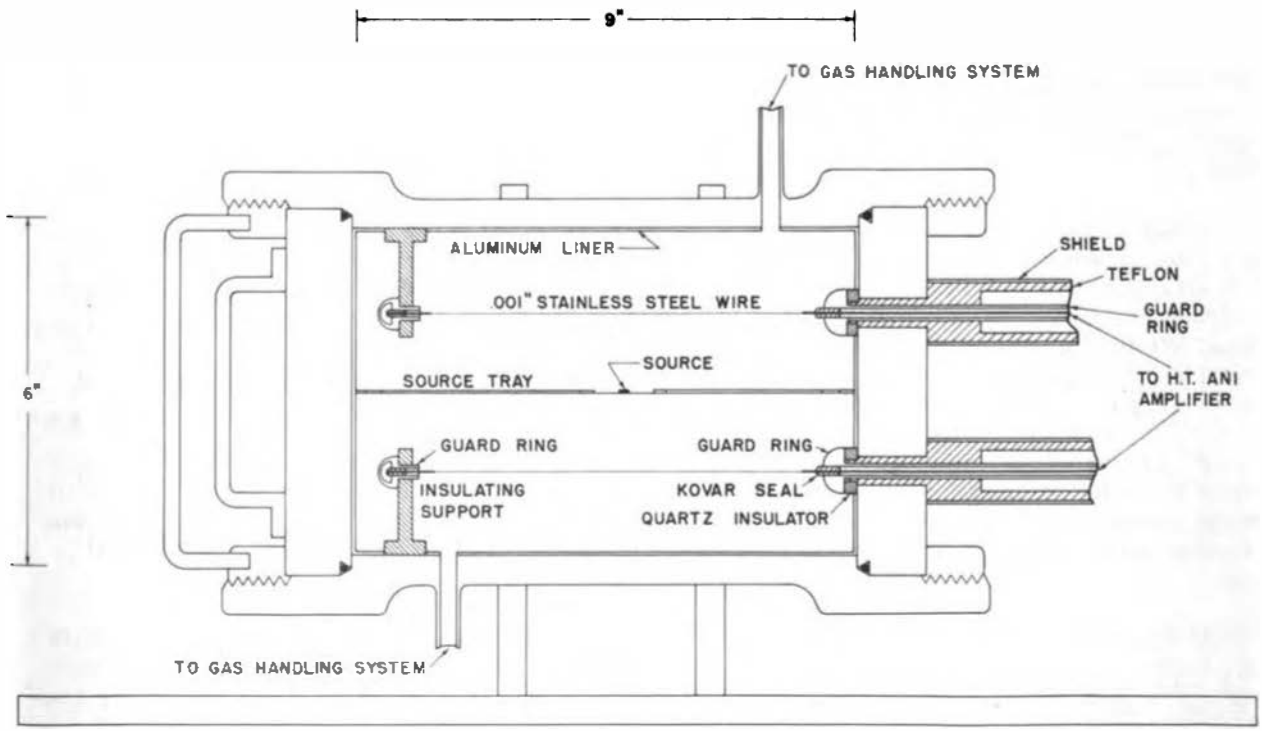


Figure 7. Chalk River high pressure counter.

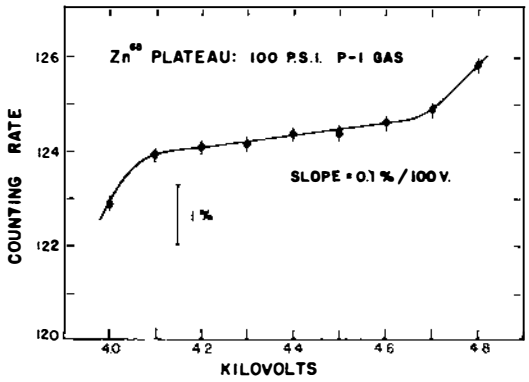


Figure 8. Curve of counting rate versus counter potential. Data taken in the counter shown in Figure 7.

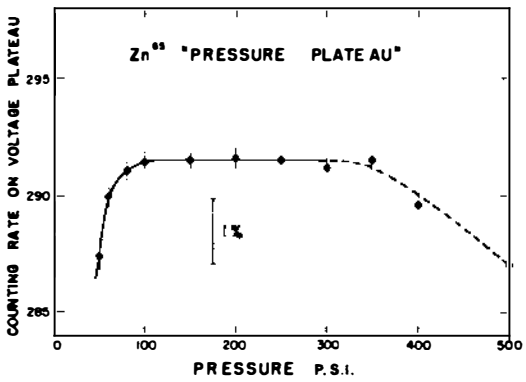


Figure 9. "Pressure plateau" for a source of zinc-65. Data taken with the counter shown in Figure 7.

pounds per square inch of P-1 gas. P-1 gas, that is argon with 10 percent methane, does not give a very good plateau, or reproducible results. If the amount of methane present is reduced quite satisfactory operation can be obtained. We expect as we go to higher pressures still, we will have to reduce the amount of methane still further.

Finally, Figure 9 shows a pressure plateau for zinc-65 which is flat to quite a high degree of accuracy over a range of 200 pounds per square inch. For some reason the plateau seems to tail off at high pressures. This is possibly due to the fact that there was a small cavity in our source sandwich. As the pressure is increased the gas in this cavity absorbs an increasing fraction the X-rays and hence the plateau tends to drop off at very high pressures. That is all I have to comment on at this moment.

LYON: Since we have heard about the internal counters somebody should mention that at Oak Ridge there are external counters for X-ray counting of the type developed by Dr. Borkowski. We have done this for a number of years. We have a beryllium window type argon or krypton-methane filled counter. Some of the results which we obtained were published in a paper which followed Dr. Allen's paper. We have used this counter to calibrate an iron-55 standard.

Recently Dr. Lazar and I, in connection with some other work, had occasion to make a very thin sodium iodide crystal with a beryllium window on it. Using this in conjunction with some very excellent electronics supplied by his group and a 20-channel analyzer we made a calibration of this crystal using coincidence counted X-ray sources of chromium, manganese, and zinc. We then measured this iron sample and found it checked within the experimental error which we would expect, indicating that our previous calibration using the external beryllium window argon-methane counter was quite satisfactory.

It is true, as Dr. Sinclair brought out a while ago, that perhaps the biggest uncertainty in all of these determinations is the fluorescent yield. However, as far as the reproducibility of these counters using any one fluorescent yield value is concerned, we are quite happy with the external type counters, although I will say we have not had any experience with the internal type.

SINCLAIR: I can add something to that. I don't know whether you are corresponding with Dr. Cole or not. We did this work. The agreement between his samples and ours was about 3 percent, as I recall. I don't remember the extent of the disagreement, but it was pretty considerable with this other X-ray-internal bremsstrahlung coincidence method.

REYNOLDS: I understand the latter method was 40 percent lower.

BORKOWSKI: There was one point on X-ray counting which might be of some interest. This is with regard to nickel-59. One of the methods we used to determine the absolute number of X-rays was with a 4π proportional counter, where one varies the pressure very much as described previously. However, the other method was an ionization chamber technique where one essentially measures the ionization current produced by the X-rays. Since they are all monoenergetic and the W factor for argon is in less dispute than W in air, one can determine the absolute number of quanta being absorbed by the gas, assuming one has a geometry where essentially all of the X-rays are absorbed. The geometry was the following:

An ordinary spherical glass flask was used for the ionization chamber. It had a standard tapered joint on it. The source was mounted in the center of the spherical volume. This was about a two liter flask, I believe. The source was mounted very much as one mounts a source for a 4π counter--using the sandwich technique in this instance to cut out the Auger electrons--the collecting electrode itself was a very thin piece of stainless steel. The X-rays essentially are radii from the center of the sphere. One measures the current, and from that computes the number of quanta being absorbed in the gas. In this particular instance it was argon at atmospheric pressure. Krypton was also used. The agreement between the ionization current measurement and the proportional counter measurement was within about three percent. This was rather interesting to us because the value of W for argon at that time was not, I think, held in quite the confidence that it has now.

The other point I would like to make is that possibly this same technique may be used to determine the Auger electrons. If one uses a spherical ionization chamber, and introduces the source as a gas--suppose one introduces iron carbonyl--it may be possible to measure the total disintegration rate of the source. There the wall effect would enter into it, but I believe with soft electrons, (four or five kilovolts), and by making the chamber large enough, one could minimize the wall effect to where the absorption by the wall would be very small.

One could also use other gases. For example, if one used helium, the X-ray absorption in the helium could be calculated, and it would be very small. One would be essentially then measuring only the Auger electrons. Then by counting another aliquot in a gas having a high absorption coefficient, one could measure the quanta separately.

SELIGER: I might mention one thing about using helium. You have to be extremely careful about purity.

BORKOWSKI: That is right. Hydrogen could be used.

SELIGER: That would be better.

GROSS: Yes, but you would be just as well off if you would add your impurity to the helium when you come to the low level, and then you would not have to worry about the purity of the gas.

BORKOWSKI: You run your own W measurement. Run the alpha and electron source in the two gases you are going to use.

SINCLAIR: Did you measure the fluorescent yield?

BORKOWSKI: No, we did not in this case.

3. X-ray Internal Bremsstrahlung Coincidence Measurement

R. W. Hayward

I am going to comment on some measurements that we have made on internal bremsstrahlung and perhaps try to illustrate why these measurements of X-ray internal bremsstrahlung coincidences may not be entirely reliable.

Internal bremsstrahlung is a second order process where an orbital electron makes a transition to a virtual intermediate state from which it is captured by a nucleus. In making this transition to the intermediate state it radiates part of its energy in the form of a photon while the remainder of the disintegration energy goes off with the neutrino after the electron has been captured by the nucleus.

Some calculations have been made recently by Glauber and Martin of Harvard University taking into account the capture from the K and the L shells. They in particular looked at the L-1 and L-2 shells, i.e., the 2S and the $2P_{1/2}$ electrons.

For the capture of K or 1S electrons, one gets an energy distribution of the photons that has an intensity distribution I = constant x $E(1-E/E_{max})$, where E_{max} is the total disintegration energy for K capture. For the 2S electrons, you have a similar situation except the spectrum is about a factor of 8 lower in intensity. This is just from the reduced overlap of atomic wave functions at the nucleus. When one takes into account the 2P electrons, the spectrum is completely different, for it blends right into the normal K X-ray spectrum and it can be considered as the wings of the K X-ray spectrum.

What happens here is that a $2P_{1/2}$ electron will make a transition to a virtual S state and be captured from the S state. It will emit electric dipole radiation in the process just as a normal K X-ray would. This spectrum should blend right into the normal K X-ray spectrum except that in the internal bremsstrahlung spectrum this unique K X-ray energy is forbidden because the 2P electron cannot make a transition to the precise 1S state, because that is occupied, and dual-occupation is forbidden by the Pauli principle. Glauber and Martin have worked out the energy and intensity dependences of these different spectra, and we have applied this theory to some measurements that we have made on the internal bremsstrahlung spectra from vanadium-49 and cesium-131.

I might also say that I can justify speaking about this because it involves a technique of gamma ray measurement that has not been mentioned yet. The technique is one of measuring a continuous spectrum, which is quite a bit more involved than the observation of discrete lines. I would like first to show the results and then to mention how we measured the spectrum.

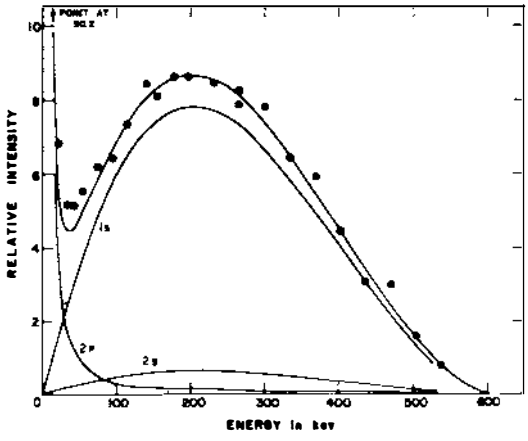


Figure 1. Spectrum from vanadium-49.

Figure 1 shows the 1S spectrum which would normally be used in the calibration of an electron capture isotope by the coincidence method. The main contribution over most of the energy range is due to the 1S or normal bremsstrahlung which would be in coincidence with K X-rays. The 2S and the $2P_{1/2}$ would not. Since the relative intensities of these spectra are very Z dependent and energy dependent, the $2P_{1/2}$ spectrum would be greatly enhanced if the disintegration energy was much smaller and the atomic number of the nucleus was higher.

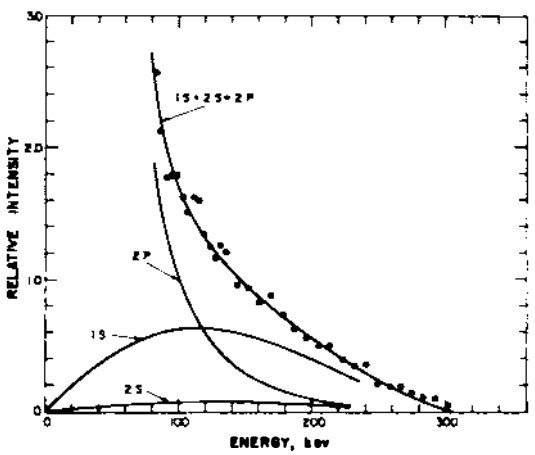


Figure 2. Spectrum of the inner bremsstrahlung from the cesium-131.

In Figure 2 the 1S and 2S spectra are very similar to the previous case, but the $2P_{1/2}$ spectrum is greatly enhanced. If one were to make coincidence measurements, one would really have to be careful to apportion that fraction of the inner bremsstrahlung that is due to the 1S state which is in coincidence with the K X-rays. This is where the difficulty will lie in standardizing the K capture emitters by X-ray inner bremsstrahlung coincidence.

It is also interesting to note that due to the recent developments of the non-conservation of parity that the 1S spectrum is completely circularly polarized, where the $2P_{1/2}$ spectrum is a non-polarized spectrum. So in principle one could separate out the 1S from the $2P_{1/2}$, but I doubt that in our lifetime we will see K capture emitters standardized in this way.

There is the problem of analyzing a continuous X-ray spectrum, taking a pulse height spectrum which one observes and converting it into a true energy spectrum. One needs to know what the differential response of your counting system is for photons of discrete energies. As we saw earlier, for any particular energy you have some sort of distribution. It means that if you have a continuous distribution, you have to start at the high energy end and take a small interval and say that the pulses at the high end are really due to photons of this maximum energy, and then subtract off what pulses you would say these photons are contributing at the lower pulse heights. By this process you can continue to subtract off groups of pulses due to photons of successively decreasing energy and arrive at a true spectrum. It is sometimes very difficult because the errors made in such a procedure are quite large. But in a spectrum that tails off at the high energy end, it is a fairly reliable process.

The measurements that we made on vanadium-49 were made in a well type sodium iodide crystal, three by three inches, with a quarter inch well extending down into the interior of the crystal. Such things as back scattering peaks and escape peaks are completely eliminated because your photons, if they back scatter, go right back into the crystal at the opposite side of the well. So one merely needs to correct for the absorption due to the walls of the well at the lower energy end.

You have to artificially introduce some absorber because the K X-ray is in time coincidence with the K inner bremsstrahlung, and you would get into some difficulties if you did not absorb out this X-ray.

In the case of vanadium-49 where your X-ray is 4.5 kilovolts and your spectrum extends all the way up to 620 kilovolts, there is no problem whatsoever. But in the case of cesium-131 where the X-ray energy is about 35 kilovolts and your upper limit of the spectrum is 320 kilovolts, there is not much difference between the absorption for these two energies. Here you have to go to a geometrical situation where the source is outside the crystal and introduce enough absorber so that you reduce to a high order of magnitude the chance for addition of the X-ray and the inner bremsstrahlung. This usually has to be done in a geometry where the source is located far away from the counter so that the chance for any time coincidence between the X-ray and the inner bremsstrahlung is greatly reduced.

These are essentially the only remarks I wish to make on inner bremsstrahlung. I might add a few remarks on the K to L capture processes.

For low Z elements, where the energy available for K capture is of the order of 50 kilovolts

or greater above the K binding energy, the K to L capture ratio is nearly constant. But as you go up in atomic number to, say, the region of 50, you already begin to deviate from this constant by as much as a factor of two. Even when your energy is as much as 500 kilovolts above the disintegration energy and in regions around uranium, the correction may be as great as a factor of 100.

There are calculations of Rose and Brisk who have done in detail the probability for K capture, L-1 capture, L-2 capture, and so forth.

I might also remark that these calculations are in some doubt because they are fraught with the same difficulties that the calculations of internal conversion coefficients are. There are such things as the finite size of the nucleus that still are not fully understood.

There is certainly disagreement in the theoretical internal conversion coefficients and the experimental values, and it is expected that the same order of disagreement should occur here.

BEAR: I would like to say that in discussing Dr. Rasmussen's measurements with him he mentioned that he was able, in his inner bremsstrahlung channel, to see this 2P effect, and to set his discriminator level so that he was at the point where the 2P was only 6 percent of the total bremsstrahlung.

HAYWARD: In an isotope like iron or other low Z elements where the disintegration energy is high, one could account for the $2P_{1/2}$ contributions very accurately. In high Z elements, the 2P spectrum is the dominant part of the spectrum and one would have to be very careful.

4. Counting-of Low Energy Radiation

W. B. Mann

What I have to say is mainly about instrumentation. I want to describe the internal gas counters we have recently assembled at the National Bureau of Standards with which we hope to standardize low-energy beta emitters and also electron capturers that can be put into a gaseous form for internal gas counting. Some ten years ago at Chalk River I suggested the use of compensating gas counters. Those were the good old comfortable days at Chalk River when we had only the bears to contend with and Leo Yaffe was on our staff, and presumably on our side, too. Bob Hawkings and I assembled a number of such counters. They were rather primitive ones, but they served to standardize carbon-14 with what we thought then was a fairly good accuracy, but which we have rather greater doubts about now, because of the discrepancies in the half life.

There was not anything particularly original about the suggestion of compensating gas counters because lots of people before had used internal gas counters of different diameter and different length, but nobody seemed to have used them together at the same time with the same diameter but different lengths. If you use two counters of the same diameter, as nearly as you can make them, but of quite different lengths, then you may hope to eliminate the end effects. If you try to close off the end as identically as possible and you assume you have the same spread of field--in other words, if you have a field distribution, as shown in the solid line of Figure 1, along the length of the long counter and the field distribution shown in the broken line along the short counter--then you can assume that the fall-off at the ends of the counters is the same, and although you will lose counts by the fact that you have a weaker field at the ends, you will lose the same in both counters. In fact, at certain points on the plateau the counts you lose by the weakening of the field may be just compensated by the number of extra counts you get by beta particles coming into the counting volume from the end volumes. Thus in some cases you can work out the end corrections from the readings obtained from two counters and you find you have zero correction.

You should never have a flat plateau because your field is always extending out further and further as you increase the anode voltage. The last thing in the world you want to get with gas counters, although some people seem to strive for this, is a flat plateau. If this counter is so short that you have no flat portion in the middle of your field distribution curve, then you don't get compensation. So when I went to the Bureau of Standards, I set myself the task of trying to make some precision gas counters. We decided that we would make three of three different lengths so we could see whether in fact the shortest counter was long enough to give compensation. This can be tested because if you take the long, the medium and the short counters, you can determine the difference in the volumes very accurately. You cannot determine these actual volumes very accurately but you can determine the differences in the volumes. We just used a simple method which will be illustrated in one of the figures. You can get the difference in volume of the long and the medium counters and also of the long and the short quite accurately.

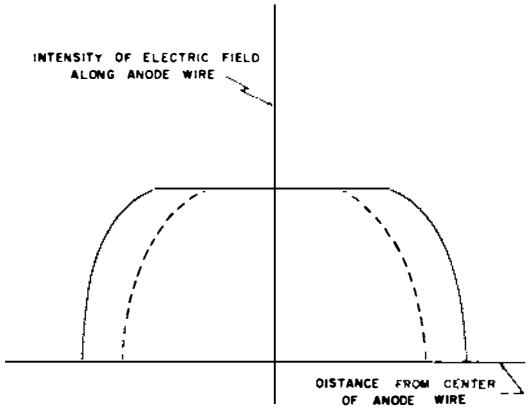


Figure 1. End effects in gas counters.

The criterion which I think is sufficient to test whether or not you have true compensation is that the ratio $\underline{V}_{L-M}/\underline{V}_{L-S}$ should be equal to the ratio of the counting rates $\underline{N}_{L-M}/\underline{N}_{L-S}$. If you have that condition satisfied, then you know that your shortest counter is really long enough to give a finite length of field which is the same intensity as in the middle of the long counter, and that you have compensation. You can then

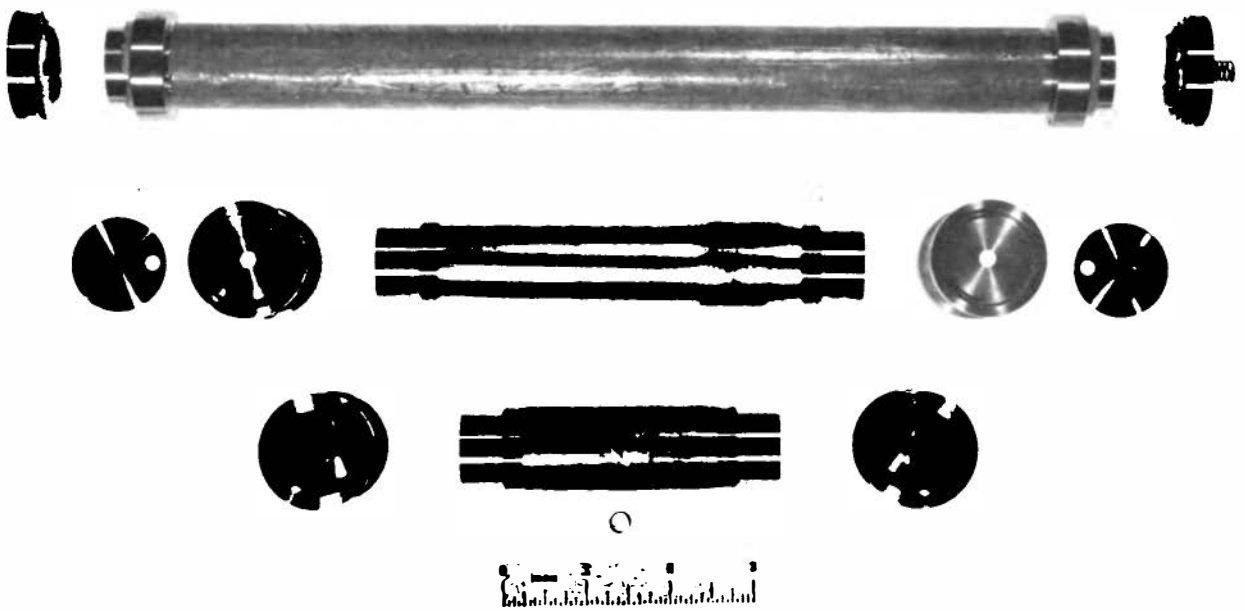


Figure 2. Stainless steel counters.

throw away the middle counter and do all your counting with the short and the long.

We made some stainless steel and copper counters. Figure 2 shows three stainless steel counters with the ends as nearly as possible the same. I picked out some pieces of glass and had them ground and matched them up in diameter in two different directions; these three were roughly the same in lengths, and those three were roughly the same. Here we have micrometer threads, 25 to the inch. These end pieces consist of essentially a recessed disk and another one which is screwed in. This one has a biggish hole so as to clear the wire and this one has a very small hole to locate the wire and there is another hole off-set here so you cannot see into the counter, but through which the gas can enter.

Dr. Seliger, Mr. Schwebel and I assembled those counters. It took a lot of work to assemble this particular set of counters. We tried to set the critical distances the same on a traveling microscope in each case to about two or three thousandths of an inch.

We thought it would be nice to construct another set of counters from copper. These are shown in Figure 3. I thought it might be possible to get away from the difficulty of the glass counter end-assembly and to throw the precision work onto the workshop by designing insulators which could be machined, with close tolerances, from teflon for the ends of the counters. There is a stainless steel shield around the teflon so that the counter sees the same outside world in each case. These counters are very easily assembled. They are simply screwed together and the wire mounted. It is easier to see the chamber in Figure 4. To measure the volumes we used a tungsten needle and a telescope. We then shone a light on the point of the needle and filled with water up to the point. Mr. Schwebel and I made these measurements and we obtained very nice reproducible results. We could not, however, get reproducible results until we put a wetting agent in. Then we observed when the surface of the water touched this very fine tungsten needle in the microscope, we got a reproducibility of one in 10, 000 in the difference in the volume. Using a tungsten needle mounted on a flat plate that sits on the end of the counters,

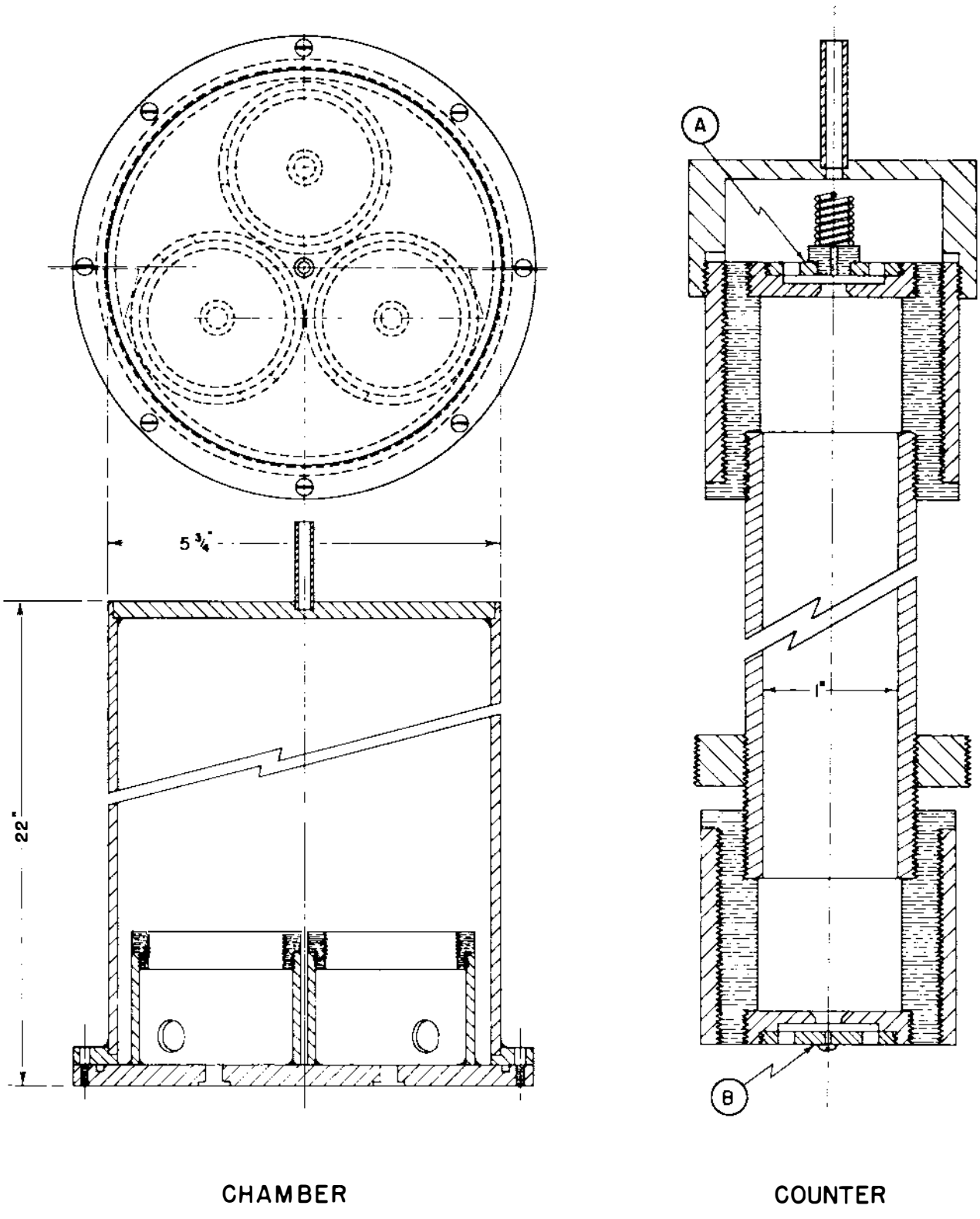


Figure 3. Copper counters.

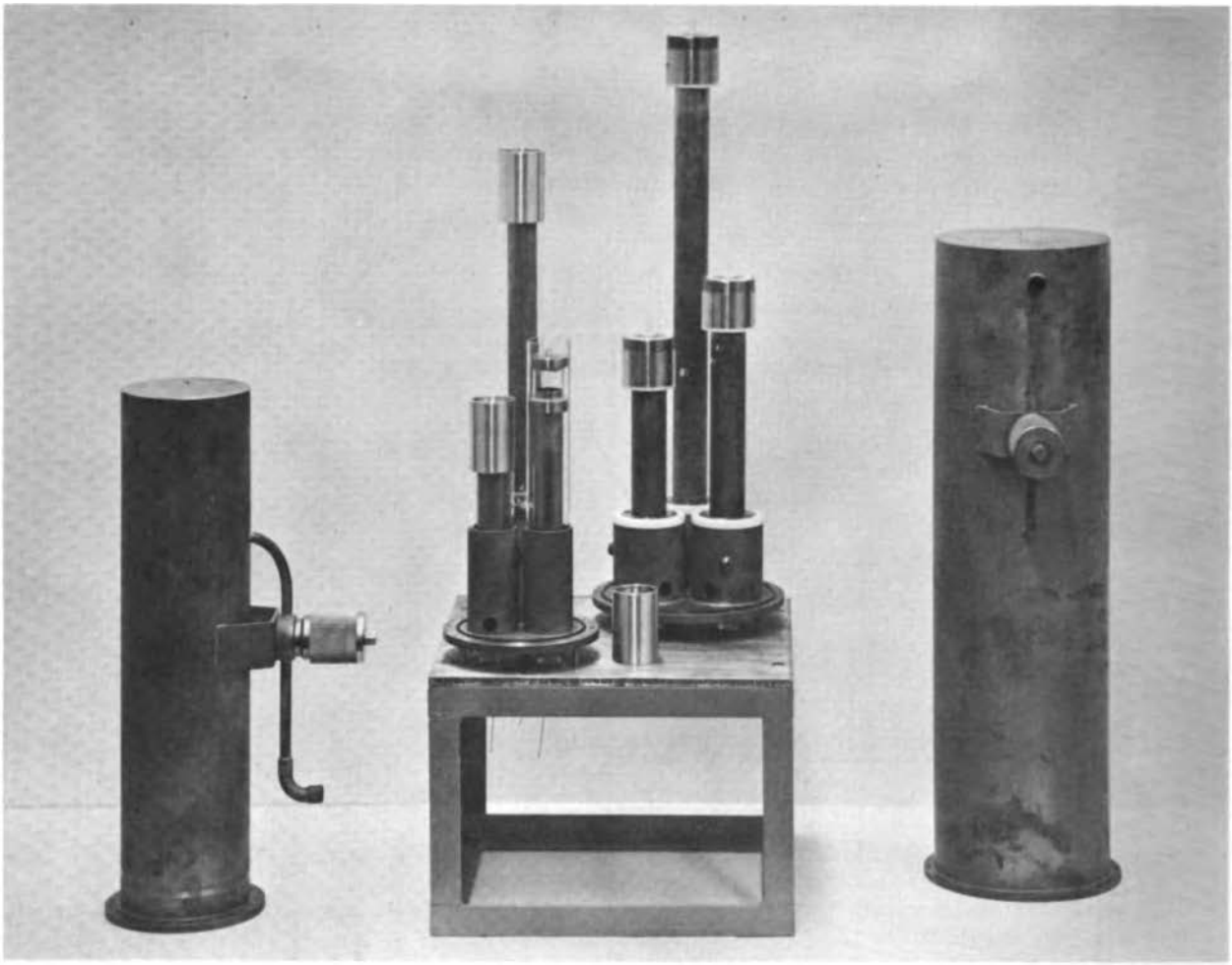


Figure 4. Mounted stainless steel and copper counters.

as long as you measure up to the point in each case, gives $\underline{V}_{L-M}$ and $\underline{V}_{L-S}$ which is all you want. You don't need absolute volumes.

These are the counters mounted--three stainless steel counters, three copper counters. The stainless steel counters also have shields over the end volumes so that they do not see the outside world. The field is the same. After we had gotten the system pressure tight we found it would retain a vacuum for the order of three months, so we can go on counting for a week without any worry about leaks.

Figure 5 shows the assembly which is a little complicated. Dr. Seliger kindly provided all this electronic equipment. I don't understand electronics, and am highly suspicious of it. This is a stable high voltage set whose meter, due to electrostatic charging of the glass, does not always read the same. It was once said that even instruments do not always tell the truth! It is much better to change the lead without cutting the high voltage! Dr. Seliger does not like my going in and changing things with the high voltage on, but I was doing this while he was away. It is much better to set to a given voltage and then to leave it there and then to count at the same voltage on each counter in turn.

This is a non-overloading amplifier and various auxiliary equipment. As I said, I only understand the dials and the face of these. Here are the gas counters and these are metal-Teflon valves.

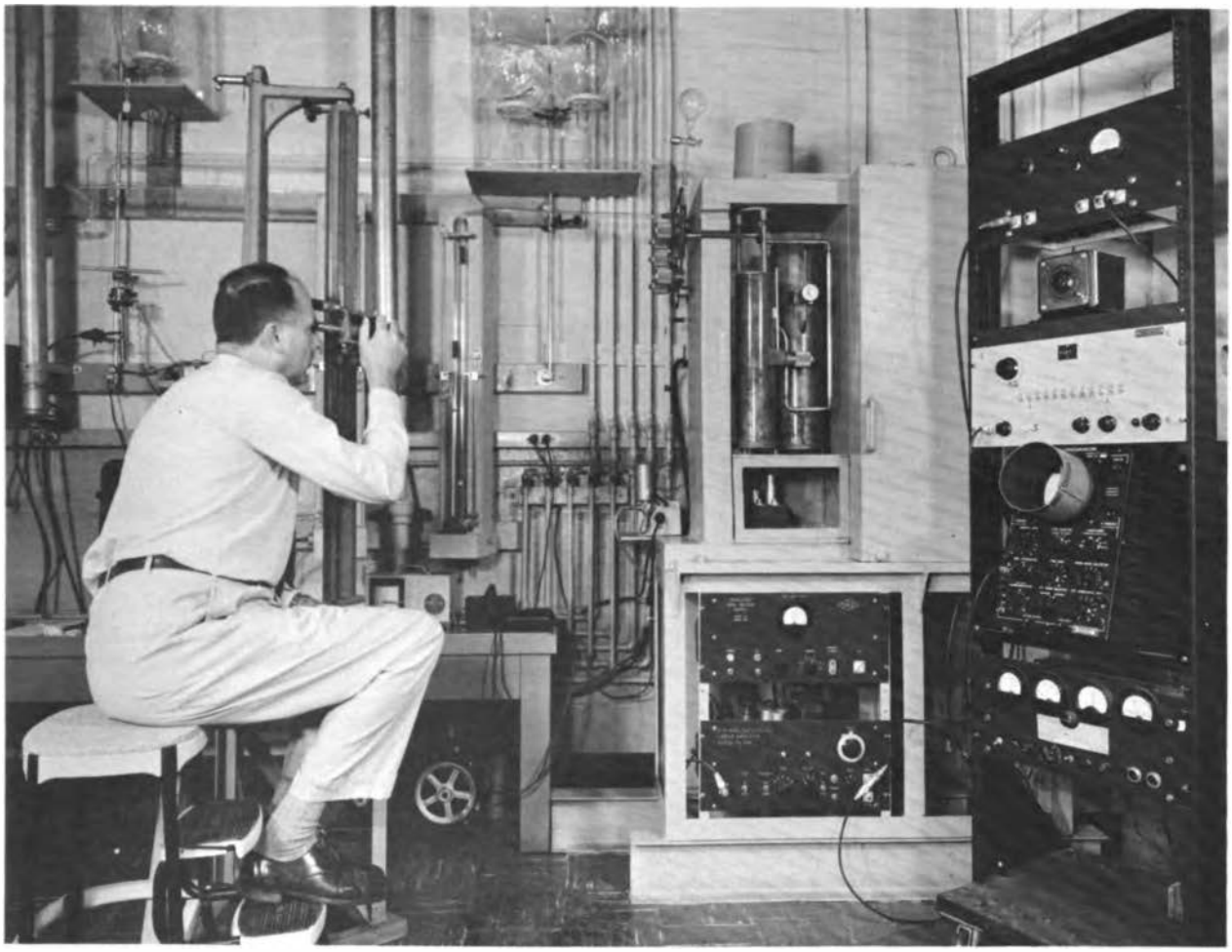


Figure 5. Experimental counting arrangement.

Here is a valve system of five valves and here an automatic Toeppler pump. Here is the gas handling equipment and a calibrated volume of five liters. We had to cover them with plastic to reduce temperature fluctuations.

Here are the barometers. We can get a reproducibility of about one or two parts in 70,000 with those. We try to keep everything in the gas handling and in the determination of volumes and gram molecules of gas to a precision of one in ten thousand so that we are then limited only by the statistics of counting.

We had to thermostat the room and keep it at constant temperature. Once you have the gases enclosed in the counters it is all right, but while you are trying to measure the number of gram molecules you have, or when you are mixing--I should have said we admit carbon-14 or krypton-85 to the right-hand side of the equipment, and its pressure is measured on the right-hand barometer. We admit our methane on the left-hand side, and measure its pressure on the other barometer. We know these volumes to a precision of three or four parts in 10,000, and we can thus calculate the total amount of methane and krypton. We mix them with the automatic Toeppler, back and forth. This is the point we noted at Chalk River, that you cannot easily get complete mixing. You can leave two gases for a week and they do not mix. You have to force them back and forth about a dozen times. Each time you expand you check the counting rate on one of the counters and then expand it back and forth until the counting rate stays constant. It

takes eight or nine such operations before you have it completely mixed and you have a constant counting rate. This is something almost unbelievable. You can wait a week and not get this constant.

SELIGER: I think that you might mention that in spite of the fact that this is the United States, we thermostated our room quite inexpensively.

MANN: Yes. We had an estimate of $15,000 to do it, and I think we did it ourselves for $150.00. We could do it only during the winter, because we had to wait until the temperature went down. We have a fan bringing cold air from the outside and electric heaters to heat it up, and a thermostat which switches on the heaters or the fan. The entire room keeps constant to about a quarter of degree Fahrenheit.

Figure 6 shows some results that Dr. Seliger and I got for carbon-14. As you see, they do not have a flat plateau. There is a definite slope. I was very pleased to find that I could go to a drawing board and force a line of the same slope through all three plateaus, which means, I think, that you have compensation. If you take $\underline{N}_{L-S}$ you can plot a voltage plateau. If you take $\underline{N}_{L-M}$ you get a series of counting rates which would be smaller, but you can multiply by $\overline{V}_{L-S}/\underline{V}_{L-M}$. You should get the same values of $\underline{N}_{L-S}$ and this I called "L-S" in Figure 6. These are the values, and you can see that the two sets of points overlap.

Figure 7 shows the results for the copper counters with the same pressure with a slightly bigger slope to the plateau but again quite nice compensation from 3100 to 3500 volts.

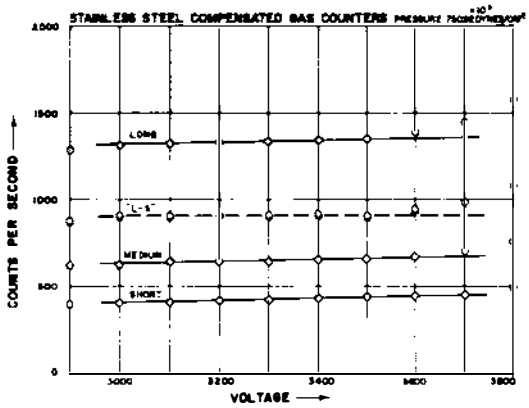


Figure 6. Stainless steel compensated gas counters. Pressure: 750.92 x 10^3 dynes/cm^2.

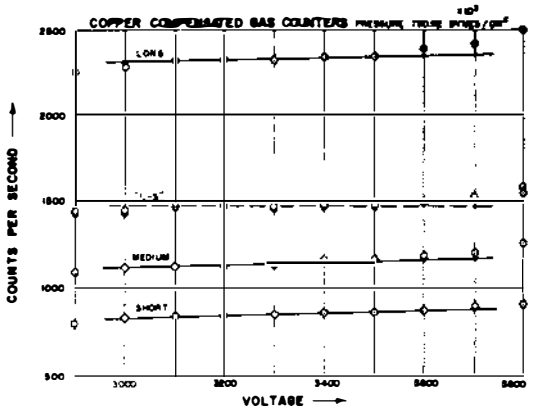


Figure 7. Copper compensated gas counters. Pressure: 750.92 x 10^3 dynes/cm^2.

Figure 8 shows the equipment for preparing krypton-85. We are trying to make up some krypton-85 standards. Krypton-85 and inactive krypton were carefully mixed and transferred to the bank of 100 ampoules which were then fused off.

Table 1 is a summary of the results for krypton-85 to date. These are about the worst of the three pressures. This is for the stainless steel counters, $\underline{N}_{L-M}$ counts per second, $\underline{N}_{L-S}$ counts per second, and $\underline{N}_{L-M}$ counts per second multiplied by the ratio of the volumes so these are the figures you compare giving the average counts per second per ml., which becomes 0.8209 for the stainless steel counters. The procedure for the copper counters is similar. The two figures you compare are the average counts per second per ml., namely 0.825 as against 0.821. This is at 659.97 dynes/cm^2.

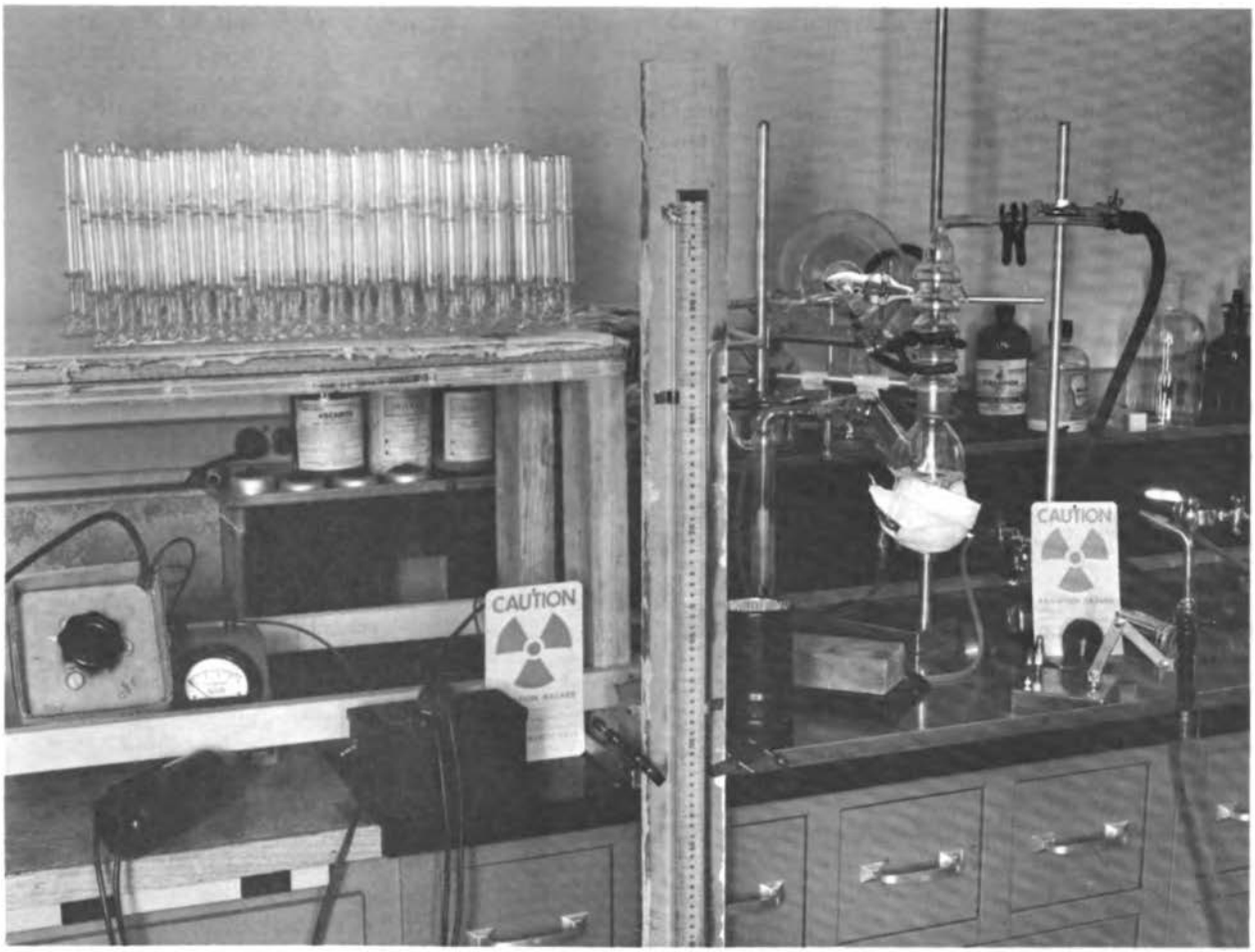


Figure 8. Equipment for preparing krypton-85.

TABLE 1

Summary of Results for Krypton-85 659.97 x 10^3 dynes/cm^2 (49.748 cm Hg at 24.18°C)					
Counters	Voltage	$\underline{N}_{L-M}$ c/s	$\underline{N}_{L-S}$ c/s	$\underline{N}_{(L-M)} \times \frac{V_{L-S}}{V_{L-M}}$ c/s	Average c/s per ml
Stainless Steel	3200	48.9	64.1	64.9	
Stainless Steel	3400	50.9	67.0	67.6	0.8209
Stainless Steel	3600	49.1	65.9	65.2	
Copper	3100	84.2	106.0	105.2	
Copper	3300	86.2	106.8	107.7	0.8254
Copper	3500	84.0	106.6	105.0	

TABLE 2

Summary of Results for Krypton-85					
Pressure in dynes/cm^2	Counter-sealing temperature (^oC)	Counts per second per ml		Counts per second per ml corrected to P and T of first reading	
		Copper	Stainless Steel	Copper	Stainless Steel
862.74×10^3	22.13	1.081	1.092	1.081	1.092
659.98×10^3	24.80	0.8254	0.8209	1.089	1.083
541.40×10^3	25.37	0.6759	0.6718	1.089	1.082

Table 2 shows the results at different pressures, namely at 65 centimeters, 50 and 45 centimeters of mercury. These are the results summarized from the previous table, together with those at lower pressures. If you take the ordinary gas law, you have to correct for the temperature of the counter at sealing and the pressure, and I have corrected back to the temperature and pressure of the first set of readings so that these remain the same.

This is not a Van der Waals correction which might be better but I did not have time to make this. I ordered 100 millicuries of krypton-85 which I thought should have given me about 10 microcuries in my counters, and I could have made the measurements in two days. However, there appears to be a discrepancy of a factor of 3 between the Oak Ridge and the National Bureau of Standards measurements which necessitated a much longer count. The factor of 3 makes a very big difference in the time you take. I would like to ask you, Dr. Reynolds, how krypton-85 is assayed at Oak Ridge.

REYNOLDS: The Oak Ridge National Laboratory Analytical Chemistry Division is not responsible for that measurement so we cannot answer the question.

MANN: At this point it looks as if we can abandon the stainless steel counters and just use the large copper one, and the smallest copper one which happened to be a bigger diameter with better counting rates and statistics, and do our standardization with these two, as it does look as though we have compensation. The only thing that bothers us is that we do not seem to be able to get a wall effect. If I remember rightly, we used counters of one inch and three inches in diameter at Chalk River for carbon-14 and could not find a wall effect. We calculated it was possibly 2 percent. We had hoped to plot counts per second corrected to standard temperature and pressure against 1/P and extrapolate to 1/P equal to zero.

HAWKINGS: I do not think we got any positive effect.

MANN: It is rather strange. I feel there should be a wall effect, but there does not seem to be.

HAWKINGS: What was the length to diameter ratio in your counters?

MANN: The copper counters are one inch in diameter and the stainless steel three-quarters of an inch. The long stainless steel is about 12 inches in length, and I think the long copper counter is 18 inches long.

HAWKINGS: You said that the slope on the plateau for each counter was the same. I can see how a counter can have a slope, but I should think on a percentage basis the longer counter should have a lesser slope because the contribution from the ends is considerably reduced.

MANN: No, the addition of each is the same. These are absolute counts. If this is flat

and you add the same amount here, using the end effects, you get the same slope, wherever it is. I should have said that I personally do not like the idea of trying to put guard tubes in. You try to put a tube along the wire to reduce the field to zero. In my opinion this is a very delicate operation, getting this tube centered at the right distance. But in this way we can make counters which are so simple, you just screw them together and you have no adjustments to make. I think Bob's last counters did have guard tubes, but I don't quite approve of this!

PEACOCK: Did you check even roughly to see whether or not the explanation for the Oak Ridge discrepancy versus yours related to the reported number of gamma rays per disintegration. If you have not done it, you probably are the ones that are nominated.

MANN: How would this come in? I am just counting electrons.

PEACOCK: I understand. They are calibrating and certainly they are likely to shift on the basis of the gamma ray abundance.

MANN: I do not know how they are calibrating. I wondered if they calibrated initially by beta counting and maintain by means of gamma rays. But then that would not affect it.

PEACOCK: They probably have a calibrated gamma chamber and on the basis of the reported number and energy of the gamma rays in a particular decay scheme estimate the disintegration rate. Since you probably have all the equipment necessary, it might be helpful if you would determine the gamma ray abundance. I do not mean after correcting it for internal conversion, either. I mean the gamma ray to disintegration ratio.

MANN: I did mean to add that Dr. Seliger and I are planning to use these counters for carbon-14 restandardization and wish to try to get a high-specific-activity source. We would like to get a new value of the half life. We are planning to calibrate sulfur-35, krypton-85, and we have quite a program ahead of us.

BORKOWSKI: One should not be surprised if there is no wall effect in gas counters, because the back scattering from these surfaces is very high. It can be as high as 80 percent, depending on the counter wall.

In addition, you have the secondary emission probability of soft electrons from walls. So it is exceedingly hard, and one has to work very hard indeed to try to have an electron impinge on a surface and not get at least one electron in return. So I think if one operates at a low enough bias (at several ion pairs), he will indeed observe no wall effect.

SELIGER: I might comment that this is extremely reasonable. Let me put it the other way. Several of the papers which have appeared where carbon-14 has been measured as CO_2 in the gas phase have shown this wall effect. Therefore, there is still this discrepancy to resolve.

BORKOWSKI: It is a small effect.

SELIGER: At about 30 centimeters in the one-inch counter with CO_2 and argon, it is about one to two percent. You can calculate it. There is only one set of data which has good enough precision to show this. I think these are Libby's data. It shows about a one percent correction which can be extrapolated at 1/P. Manov's data I think has too much of a spread to show the wall effect. At least I tried to look for it.

MANOV: My data are about nine years old. I am sorry I cannot remember that in detail. If I may add parenthetically, I am very happy to hear Dr. Mann's comments that work is going on to do the counting over again.

PEACOCK: This is a less than scientific comment but I want to comment on the wall effect, too, under the heading of shop notes, and not under the heading of science.

We have, among others, built for our own purposes a counter with a half-mil center wire and a diameter of about four and a half inches. We happen to use this in low background counting work. We have occasion to fill these counters over and over again. We have found that there seem to be, as far as we can tell, unaccountable variations when either air or perhaps pump oil--we are not quite sure which--contaminates the fill in such a way that we get what we refer to as an insensitive fill. In order to avoid the inaccurate result which would come about because we are actually assaying activity, we have constructed a little well in the side of the counter, about three-eights of an inch in diameter with a thin window over the end of it. Before actually counting a particular sample for the determination itself, we insert two separate sources into the counter well. The first is a collimated strontium source so that the beta rays include a lot of these which will tend to go clear across the chamber with relatively low specific ionization, and we know from a different counter what additional counting rate should come from the strontium source which is calibrated. Then we will put a thallium source in with an absorber over it which tends to produce more or less the softest beta rays which we can practically get into this counter, and also require that this have the proper counting rate. So we can get something very close to a wall effect and note that the wall in this case would be at the extreme because you have a large ratio of diameters here. If the fill is an acceptable one, there seems to be no appreciable difference from one fill to the next, even with CO_2, with its proper gas mixture, at several different pressures all the way from a fraction of an atmosphere to two atmospheres. This may account for some of the wall effects that were previously reported.

BORKOWSKI: You are particularly sensitive to electron attachment, because your fields are so low at the wall.

PEACOCK: The reason I brought this up is that it is most extreme. We have had these insensitive fills crop up over a period of years. By now we have gotten to a point where we are pretty well sure how to make a counter fill but it is not to a point where we can rely on it 100 times out of 100.

BAPTISTA: I wonder, and it is more in the nature of asking for information, if somebody considered the use of external cathode counters to study this problem? Even if I foresee some disadvantage, certainly it is a very simple method to change the sensitive volume of the counter by just changing the painted carbon cathode. I wonder if there are obvious disadvantages to this method.

YAFFE: Changing the which?

BAPTISTA: Using an external counter with a relatively soft glass you can change, and with reasonable care, limit externally the sensitive volume of the counter and just in one filling you can study the variation or the influence of these dead ends on the counting rate.

BORKOWSKI: You still have only one surface where the electrons are striking that one surface. That is, you are not changing the actual diameter.

BAPTISTA: Not for the wall effect.

BORKOWSKI: You are changing the electric field, are you not, within the counter?

BAPTISTA: I have a counter of just ordinary glass filled with gas that I want to count. I can limit the sensitive volume by painting carbon outside and defining the sensitive volume by painting the extremities with carbon and connect to the high tension unit. I can define here, if I take certain care, the sensitive volume. For this same filling I can clean the counter and change the sensitive volume for the same fill.

SELIGER: I do not think in a sense that this is true compensation because you have a different volume external to your sensitive cathode from which you can get counts.

BAPTISTA: Yes, but if the range of the beta particles is always maintained, that is, if I maintain the range infinite as compared to the beta count, the influence must be the same. Then I can study the influence of the electric field.

SELIGER: For carbon-14 that would necessitate a rather long counter.

BAPTISTA: You can work with very long counters.

MANN: The people using different counters and varying the cathode lengths and so forth are never measuring at the same time with the same filling. I forgot to come back to the point when I was talking but I even found it a good thing to take all the points on all three counters at 3200 volts and all at 3400 volts and not to run a plateau on each one separately. Take three counts with the same voltage. With different counters you are doing two different experiments at two different times. The thing that is nice about the compensated gas counting system is that you get your results with the same filling at the same time without changing anything. This proposal by Dr. Baptista is going back to the method which I feel is a little uncertain.

HAWKINGS: I think what is being proposed would not involve changing the filling.

MANN: I see. I misunderstood Dr. Baptista.

HAWKINGS: I should add that counters have been built with adjustable sleeves that slide in on a wire and thereby limit the sensitive volume of the counter.

BAPTISTA: I still feel that you can limit more accurately by this painting method than by the sleeve.

MANN: There is a tremendous amount of preliminary work that has gone into the volume determinations of these counters which I do not think would be so accurate with a system like that. Mr. Schwebel and I determined these volumes by one in 10, 000 reproducibility in precision and accuracy. I am not sure you would get that kind of precision by painting. I think there would be uncertainties. It is a good idea but I do not think I would like to give up our counters now!

MANOV: I am very pleased to hear about krypton-85 coming up. Could you tell us any more about availability or probable strength and so on?

MANN: On the availability, I made 100, as you saw. I tried to get about 10 microcuries per ampule. I carried out these measurements under rather pressing circumstances last week because I hoped I might have them in time for this meeting. As I mentioned before, I was working well into the night every night. From my initial calculations, I thought I would have a counting rate that would enable me to do the job in two days. From the fact that I took three times as long, I have roughly about a third of that activity (10 microcuries per ampule). I have not been able to make the complete assay or calculate the number of disintegrations per second per gram molecule of gas. We also have to remove the krypton ampule from the equipment and measure its volume up to the point below the break. I can say roughly that it is about three or four microcuries. I only showed the results for counts in the counters because I was interested in showing that they appear to give adequate compensation. I have not obtained the final value. There are about 8 ml of krypton at roughly atmospheric pressure in each ampule.

MANOV: You might be interested in knowing, we of the long hair group, how quickly some of the results of the standardization are put into practice. Krypton-85 is now beginning to replace strontium-90 for vehicle and deck markers and railway signal lights. You are familiar with the semaphore lights. An attempt is being made, successfully, to use krypton in a phosphor. Obviously we will need standards for measuring krypton. We have heard from two manufacturers that the total quantity of krypton which they can use in the next two years comes to between 200, 000 and 500, 000 curies of this material. You can see how quickly and how

important it will be to get some standards available and how big a dollar value basic research like this can have. I am very happy, Dr. Mann, that this work is going on.

MANN: Thank you, Dr. Manov. Actually, as to the speed, I am amazed myself, because we ordered this from Oak Ridge, and I also ordered some inert ordinary pure krypton from a local company, and I had not mentioned it to more than a few people that we are going to standardize krypton-85 and the next week we had an inquiry. I said who is asking for this one now? It was the company who sold us the inert krypton!

PUTMAN: Mr. Chairman, would it be in order to ask Dr. Manov if the Atomic Energy Commission is in any way embarrassed by these orders of large quantities of krypton?

MANOV: In these amounts, no.

KULP: I would like to make two remarks. First, regarding carbon-14, those of us in the radiocarbon dating business are extremely interested in the redetermination of the half life. However, if we have to bet on it, I think we will point out that the dated samples back to about 3,000 years ago measured by the more recent, more accurate gas counting technique, make it quite unlikely that the present half life can be wrong by more than 5 percent unless there are entirely compensating effects, such as cosmic flux changing.

Second, I would like to point out that one way to measure the decay constants of extremely soft emitters as we were discussing earlier is not to measure them at all, but measure some daughters and products. In this regard we have recently made a determination of the half life of lead-210 by a so-called geological method. This requires that we have secular equilibrium in the age of minerals. Essentially what we have done is to determine the efficiency of a counter by the indirect method of counting the polonium-210 alphas from a mineral of known age. Through the equation you see you can get the efficiency providing you know the age of the mineral and providing it was in equilibrium. Then you proceed to take a known amount of radon, let it decay to a known number of lead-210 atoms, and measure the known polonium-210 alphas. In this way one can get a half life which appears to be correct to a few percent. This seems significant because the last two reported values of this constant varied from 19.5 years up to 25 years. We get about 22.2 years.

REYNOLDS: I would like to present the results of an intercomparison on promethium-147 which was initiated by Oak Ridge National Laboratory. In November 1956 we distributed portions of a solution of promethium-147 containing about 1.4×10^7 disintegrations per minute per milliliter. We received measurements from several groups.

ARNOLD: I do not remember the radiations of promethium-147.

REYNOLDS: It is a pure beta emitter. The half life is 2.6 years, approximately. The beta energy is 0.222 Mev. This nuclide is a little difficult to measure. At least it seems to us to be difficult. The results received from several groups are shown in Table 3. I will not identify the groups except the Oak Ridge values. These disintegration rates were all corrected to November 30, 1956, using a half life of 2.6 years. By the way, I have taken liberties with some of these estimates of uncertainty. One person reporting said that his precision was a certain value, but his estimate of the true uncertainty in the value was quite a bit larger and I have used the latter estimate.

A word about the proportional counter result. This was a 2π proportional counter using copper disks as mounting support. This value is not entirely dependent on 4π. It is partially so, but in addition to the 4π result, the interpolation on 2π efficiency curves from two other reports was made, and the efficiency for promethium was estimated to be 70 percent.

The raw mean of these results is 1.36 ± 0.16, where the 0.16 is a mean deviation. It is not particularly significant. If we discard the lower results--I am beginning to be arbitrary here--

TABLE 3

Assays of Pm-147		
Organization	Instrument	Pm Activity[a] dpm/ml x 10^{-7}
A	G-M[b]	0.99
B	G-M	1.10
C	4π	1.39 ± 0.03
D	4π	1.39 ± 0.04
E	4π	1.41 ± 0.01
F	4π	1.52
ORNL	PC[c]	1.52
ORNL	4π	1.53 ± 0.04

[a] Uncertainties not defined
[b] End-window G-M counter
[c] 2π proportional counter

this becomes 1.46 ± 0.06. I am sure I will get some objections on this point. I regard this as the temporary or interim standard value.

HAWKINGS: Where did you cut off?

REYNOLDS: Between 1.10 and 1.39.

MANOV: Of course, you realize you are in a spot cutting off those two.

REYNOLDS: Yes, indeed.

MANOV: Do you have any explanation?

REYNOLDS: Yes, I think so. This is very tentative. These were both GM results and were presumably obtained by some sort of extrapolation technique to correct for air and window absorption. In these low energy beta spectra there are very many low energy electrons--a larger proportion of low energy electrons, if you will, than in a high energy beta spectrum. Therefore, the approximately straight line extrapolation which one makes for a high energy beta is no longer valid, but one must extrapolate along the curve. We have seen effects of this magnitude several years ago in the extrapolation of calcium-45 absorption curves. We made up at one time an essentially windowless counter with an absorber ring which could be rotated so that successively larger but very thin absorbers were passed between the source and the detecting volume in the counter, and we were able to show experimentally the break from linearity in the absorption curve in regions of two or three milligrams total absorption, and you realize, using a window counter, one does not even see this region. The total absorption is usually several milligrams. I think this is probably the explanation--that one has made linear extrapolations when actually there should be a rather sharp break upward at the lowest points.

PUTMAN: Can you say whether these measurements labeled 4π measurements are all the same sort of 4π measurements or are they partly Geiger-Muller and partly proportional or partly scintillation?

REYNOLDS: I should have specified. None of these is a scintillation counter result. Although I am not absolutely certain, I think they are all 4π proportional. We have some more correspondence on this, but I do not have it with me.

PUTMAN: By cutting out results A and B, you have based your standard on one method of measurement only.

REYNOLDS: Yes, that is a valid objection.

MANOV: Do you know what isotopes were used for comparison purposes in the GM counting?

REYNOLDS: One of these groups reported efficiencies based on a previous intercomparison of promethium. This, I believe, was a limited intercomparison and was based on essentially neutral water dilutions of promethium. This leads us into chemistry. We do not believe that a neutral aqueous solution of promethium is stable. Therefore, the fact that the group would get a low result would not, I think, be surprising. Also, I believe the same group A said that they had also used a commercially available promethium standard. This I know nothing about. Group B made a rather elaborate attempt at gaining an efficiency for 0.22 Mev betas from iron-59 which has, as you know, 0.26 and 0.46 Mev betas. This perhaps was a little questionable. It was an attempt, of course, to avoid the linear extrapolation. I have not talked to these people in detail. The technique appears at the moment not to have been successful.

HAWKINGS: I do not think it is really fair to consider these GM counter results in here at all. With all due respect to the people who attempted it, this is an almost hopeless extrapolation.

GRUMMITT: As a user of Geiger-Muller counters for low background counting--incidentally our total absorption is one milligram--I would hate to be given the job of standardizing this solution in those counters. I would certainly support Dr. Hawkings and go to a more realistic method. The extrapolation even over the one milligram would be a very difficult thing to do.

MANOV: This situation is not as bad as it was with iodine some years ago where we started 75 samples, got back 82 answers and nobody got the average.

YAFFE: Did the people who did the Geiger counting quote any error?

REYNOLDS: No.

YAFFE: In normal absolute counting with end windows, one is a little loath to put anything better than plus or minus 20 percent unless he really has a great deal of confidence in this.

REYNOLDS: On a low-energy emitter such as this I would certainly agree with you.

ALLEN: Can you tell me, in the 4π results, whether in certain cases calculated corrections have been added to any of the figures or whether there are supposedly thin sources on thin films with no correction? Is there any explanation of this trend from 1.39 to 1.52?

REYNOLDS: I do not recall that any of the people outside Oak Ridge reported such corrections. However, my memory may be faulty on that point.

LYON: Did you mention how many samples were sent out? That is, what percentage return you got?

REYNOLDS: The percentage return was rather good. We failed to get answers from only two out of a total of nine. This is probably par for an intercheck. We realize that this is a very limited number of results and any attempt to apply statistics or select the best value is questionable.

ALLEN: May I point out that in the intercomparisons which we occasionally do in England, when we send in our results, we invariably give the value as determined from the actual counting rate as well as any value which we might derive by any corrections. So at least you can see whether people counting by the same method got the same answer from their straightforward counting.

SESSION V - LOW LEVEL COUNTING

W. E. Grummitt, Chairman

Summary

When one considers the reasons for including a discussion of low level counting in a symposium such as this on methods of standardization, one expects to examine in detail the similarities in methods and measurements which the groups have in common. It is not too surprising then to discover that the requirements are in fact parallel and that there is a common interest in such things as the design, construction, and operation of suitable equipment. Both groups must apply much the same criteria in resolving problems such as the choice of detectors or of amplifiers of requisite stability.

It might be said that the distinguishing feature of a low level counting arrangement is the shield of anticoincidence counters used to cancel the effect of cosmic rays, though this is not strictly necessary with a low level scintillation counter. With this possible exception, it is a common meeting place of all laboratories in this field. Further problems of general interest to all are those of the selection of inactive counter and shielding materials and of contamination at a level where monitoring is difficult.

Various methods of meeting these problems and many of the more specific ones which occur in the measurement of nuclides such as carbon-14, beryllium-7, strontium-90, radon and tritium are discussed in detail in the papers which follow.

In the two papers of this session Dr. Arnold and Dr. Kulp will explain the techniques which are important in low level counting.

1. Low Level Counting

J. R. Arnold

I am not planning to cover all of the problems in this field for the excellent reason I do not know much about some of them. It has been a little difficult to decide just what this sort of group wishes to know about low level methods. The easiest thing to do is to not decide this at all, but to begin by making some remarks and having people interrupt me as you have other speakers, and thereby hold me down.

The first topic I shall treat is the general design approach that one uses for experiments in which very small quantities of radioactivity are involved. After you reach the conclusion that you have only about one disintegration per minute as the total activity of some source, you are forced as your first step to consider the choice of detectors. In the radioactivity field there is a rather embarrassing choice. There may still be some people who are not aware of the formulas that one applied to the choice of detectors, so far as sensitivity is concerned, but this is not the only criterion. With regard to sensitivity, there are some very simple parameters which we apply. These parameters are measures of the counting precision which is attainable with a given technique in a given time.

There are two possible cases of interest. The first case is where the sample counting rate S is small compared to the background. This is the unfortunate case. It is the case with which we frequently have to deal. In this case the parameter which one will maximize for sensitivity is $\frac{S^2}{B}$. Some people prefer to use $\sqrt{\frac{S^2}{B}}$. The rank order will be the same in either case. As between two detectors, the one which has the larger value of $\frac{S^2}{B}$, will be more sensitive. I call your attention to the fact that the sample count rate appears as the square and the background in the denominator only as the first power. This means that it is easily possible to exaggerate the value of background reduction in the improvement of the quality of the detector. I think all of us who have worked in this field for a while have been particularly subject to this particular neurosis. We have overemphasized the importance of background reduction. If one is talking about 20 or 30 percent reductions of background, these are desirable, if they are easy to do, but the improvement in performance will not be very startling. Increases in the sample count rate which are attained by better geometry or reduction of absorption or something of that kind are twice as valuable.

The other case of interest is the other extreme where we have succeeded in reducing the background to the point where the sample count rate, although small, is still considerably larger. In this case, the figure of merit is simply S. Here the count rate is even more important.

The other major criterion for choosing a low level detector is stability. People with your experience are very well off when it comes to understanding the problems here. The same criteria for stability have to be applied. Often one wants to control the stability with more accuracy than the accuracy of the final count just for comfort. Beyond this questions of the availability of equipment and of taste begin to enter the picture.

Those would be, in summary, all the criteria. Given those criteria we are led for each problem to certain detectors.

I would like to talk first about beta particles. I do not want to make the classification too rigid, but there are two different kinds of beta counting problems, depending upon whether the material to be counted is of low or of high specific activity. Let me illustrate this.

In carbon-14 dating, the specific activity of carbon-14 in contemporary carbon is about

15 disintegrations per minute per gram. Dr. Kulp might like 13 disintegrations per minute a little better. This is quite a low specific activity. We are interested in age determination in going back some 6, 7, 8 half lives. When one gets to specific activities which are very much lower, there is no point in using a counting method which does not have the possibility of introducing a large sample and counting with high efficiency to overcome the self-absorption problem.

On the other hand, in the work which we are now doing with beryllium-10 in nature, we have an isotope of an element which is very rare in nature. Although there is very little beryllium-10 around, the specific activity of beryllium-10 in a suitable deep sea core is in the neighborhood of thousands of counts per minute per gram. Since we are fortunate in having a sample of high specific activity, we are led to a very different choice of detector.

Then, too, there is the distinction which one can draw somewhere in the carbon region--in the region of 150 to 200 kilovolts of energy--between soft and hard beta particles, between energies where self-absorption and window absorption are extremely acute problems and energies where these things tend to become secondary. For all of these things there are different solutions.

Perhaps the hardest case to solve would be the one with low specific activity of soft beta emitters. This happens also to be the one which has been most studied because carbon dating is so popular, and also because tritium has become a very important isotope and its situation absorptionwise is even worse. The two techniques which are most widely used for low level counting of these soft betas are gas counting and liquid scintillation counting. Gas counting of carbon has been done in a great variety of ways. It has been done in the Geiger region. Dr. Crane at Michigan has a carbon-14 apparatus that runs in the Geiger region. I think the preponderance of numbers is overwhelmingly on the side of counting in the proportional region. People who actually take the plunge and do this seem to report uniformly the stability is better, the plateaus are flatter.

One other great advantage is that one has greater flexibility of counting gas. One can use pure hydrocarbon gases. One can use really pure carbon dioxide, which one cannot do in the Geiger region. There are many advantages to proportional counting, although the weight is not entirely on that side. For example, to count tritium, the technique of using an argon-ethylene filling with hydrogen in the Geiger region is quite competitive with other methods.

Here again it is a rather high specific activity application. Gas counters have been developed which take a very large sample--which take several grams or many grams of carbon--either in the form of carbon dioxide, acetylene, or methane. They have produced the great bulk of carbon-14 dates that have been measured.

All these detectors are used with a ring of counters in anticoincidence. This is probably a good place to stop and talk about the electronics and the kind of shielding that one uses for the entire device.

Seen end on, a typical arrangement will consist of a gas counter, quite frequently surrounded by a mercury shield about one inch thick which is in turn surrounded by a dozen or so Geiger tubes arranged in anticoincidence. There are many variants of this. One which should certainly be mentioned is the variant in which the outer anticoincidence ring is in the same gas-tight shell as the inner counter and in which one has only a very thin wall separating the two. This is something that has been worked on by Houtermans, Curran, and others. It is reported to give a considerable greater reduction in background because there is no chance for gammas to make secondaries between the outer counters and the central chamber. It has a few practical disadvantages.

The mercury shield is not absolutely essential. It is designed to absorb radioactivity from the counters in the ring--secondary electrons that might be produced in the wall of the outer

counter if the center one is quite thin--but chiefly it is designed as a further step for removing the effects of general gamma contamination. Outside of all this will be a further shield, which will be as thick as the laboratory can afford, and made of materials as pure as they are lucky enough to get. The outer shield is normally of steel and is perhaps normally eight inches thick.

The reduction of background is considerable in such instruments. A factor of 50 might be a rather typical figure for the reduction of background as between the central counter with the anticoincidence ring off and with it on. A hundred counts per minute might be reduced to two counts per minute.

The liquid scintillation counter gets its background reduction in an entirely different way in general. You have had an introduction to liquid techniques in Dr. Seliger's talk in Session II. Our low level counter looks similar to the one he described. One has a liquid cell between two phototubes, but it is often rather larger than Dr. Seliger's cell because of the low specific activity of our samples. The associated coincidence electronics to get rid of thermionic noise of the phototubes is more important to us than to him.

The thermionic noise of the phototubes is further reduced by cooling. All of the low level apparatuses that I know, with one exception, operate in a deep freeze at about -15°C. The shielding problem with liquid scintillation counters is in one sense simpler in that the detector is a great deal smaller than in gas counting. It begins to become economic to do all one's shielding with mercury, and this has been our own choice.

We have a simple two-inch mercury shield surrounding the central apparatus that we use. The source of background in liquid scintillation counter is quite different than in the gas counter. There the gamma sensitivity is low. The main things we count are mesons and a few other cosmic ray induced events. In any condensed phase one has a much higher gamma sensitivity. The gamma counts, even when one takes every possible pain to reduce them, still are more frequent than the meson counts.

There is a further reason why the meson counts are less important. The pulses which are produced by mesons traveling through a cell of any size are quite large. The pulses caused by the soft radiations we are interested in counting are small, and pulse height selection is used, although for beta particles we employ very wide windows. In this way we get reduction of background by eliminating Compton electrons from gammas when they happen to be hard enough and also by eliminating the effects of the mesons. There is little advantage in using Geiger tubes in anticoincidence in connection with a liquid scintillation counter. There may be some advantage in using a much larger liquid scintillator tank in anticoincidence. As far as I know, no one has tried this.

For carbon-14 one might give some indication as to what is currently possible with this method. We have been able to get a background in a 30 cc liquid detector, using a window such that 50 percent of the carbon-14 betas are detected, of five to six counts per minute. That is about a factor of four better than our published values. Most of the reduction is based on the following perhaps slightly unexpected phenomena.

There is bound to be a good deal of clear material around this system which is not fluorescent. This material is acted on by potassium beta rays in the glass of the phototubes and by the general flux of local gammas. These fast electrons may produce Cerenkov pulses which are quite small considering the size of the original energy pulse. Unfortunately, they are about the right size to give a pulse comparable to a small carbon-14 or a tritium pulse. One cannot eliminate these things by pulse height selection. One can do what one would do anyway; namely, reduce the local radioactivity to the lowest possible level. Second, one can reduce the amount of clear non-fluorescent material which can be seen by the detector. We do this by putting a reflecting coating inside the transparent cell in which the liquid is contained so that the cylindrical part of this cell cannot contribute pulses, by using thin end windows, and by putting black tape up and around the shoulder of the phototube so that any light which originates below the flat face is

absorbed on reflection. These things make a big difference, and they are essentially responsible for the reduction I am now describing.

With the liquid scintillation counter one has the advantage that the cell is now external to the actual detection system. This makes it possible to make certain background comparisons which are not easy to make with gas counters. It also results in a compensating disadvantage; namely, that the background over very long periods in a liquid scintillation counter varies a good deal more than in a gas counter which sits in place.

The other feature which is very useful for low level work is that one can vary the cell's size, and therefore the background to suit the size of the sample with which one works. Since this may well vary over one or two orders of magnitude the ability to use a cell from one cc to several hundred cc's in size without any modification of the system is an advantage.

I would like to say a word about chemistry. The liquid scintillation counter has a solution consisting essentially of toluene or another aromatic solvent in which various organic dyes are dissolved. At first sight it would appear that this would restrict its usefulness to carbon and hydrogen compounds which can easily be made into organic compounds. However, I have just attended a conference at Northwestern University on liquid scintillation counting, after which we concluded that nearly all substances of interest can be counted in this medium by one trick or another. Even proteins can be dissolved in useful liquid scintillation solutions. As for heavy metals, we have put 6 percent manganese into a liquid scintillation solution. Each case is a special one, but the proceedings of this conference contain enough information to solve most problems.

There are many cases in which it is more difficult to make the counting material into a gas for gas counting.

I would like to mention the technique for counting tritium in liquid scintillation counting at low levels without the problem of dissolving water directly in the liquid scintillators. One can dissolve a few percent of water in several systems fairly easily but there is this limitation. Dr. Nir of the Weizmann Institute in Israel has developed a clever technique which improves on this, based on some old work of Ingold, who finds that the hydrogen atoms in benzene will exchange with 100 percent D_2SO_4 and presumably, therefore, with other isotopes of hydrogen. Nir took both benzene and toluene and was able successfully to equilibrate the hydrogen atoms with water through which sulfur trioxide had been passed from a tank to make sulphuric acid. The exchange can be accomplished by shaking 50 cc or 100 cc in a shaking machine. When I first heard about this I thought it was the complete answer to low level tritium counting. It has a general utility, but one should be warned--this has not yet been published but is in press--that at the same time as the exchange reaction is going on sulfonation of these aromatice compounds is also going on. There is some difficulty in the adjustment of conditions, concentrations and so on, so as to get the exchange to proceed to virtual completion without losing an excessive amount of material. However, these workers in Israel have used this method for natural tritium with lower amounts of enrichment than is customary in the gas counting methods, because one can handle larger samples. They seem to find it rather useful.

PEACOCK: Can you give us any range of values for carbon-14 which will indicate the number of grams of sample that would be included in some of the solution used in scintillation counting?

ARNOLD: This depends on a lot of things. We are able to get about 15 grams of carbon into such a system by synthesizing before from living carbon dioxide. The counting rate of contemporary carbon-14 in this system is about 120 counts per minute. This is no longer a low level problem perhaps in the literal sense.

Let me say another word on sensitivity. To give an example of the difference between old and new methods, the first way that any of us learned to count carbon-14 was to count a barium

carbonate solid sample under an end window counter. If one has a low specific activity, one will choose to count an infinitely thick sample in order to get the largest possible number of counts. If one takes contemporary carbon-14 and puts it under such a detector, the background would be about 20 counts per minute inside the shield. The counting rate due to carbon-14 from living sources is 0.006. Thus, with modern counting techniques,

$$\frac{S^2}{B} \text{ has increased from } \frac{.006^2}{20} \text{ to } \frac{120^2}{5}.$$

Similar improvements have been made in gas counting. This is quite a few order of magnitude increase in sensitivity.

In the easier case of counting harder betas or some of these soft betas at high specific activity, there is an even larger variety of techniques available. Here one can deposit the sample externally to the counter or in a windowless counter without too much self-absorption.

A simple commercial end window proportional counter put inside an anticoincidence shield will have a background of about one count per minute. Considering that some of our other detectors have lower backgrounds for the same volume, I suspect that some of this is due to radioactivity in some of the materials of the counter. One might be able to do better. But a reduction from 20 to 10 counts a minute to one count a minute is certainly very useful for many purposes.

One advantage of having available any of these anticoincidence instruments is that you can take out the center counter and replace it with a whole variety of other devices. Any laboratory that plans to do many different low level problems should design its shield so as to have inside the umbrella a volume several times what is needed immediately, for this reason. Many different types of counters have been used inside an anticoincidence shield. The one with which I have had the most experience, and that I like best, probably for this reason, is another device invented by Libby, the Mylar thin wall counter. These things are simply a plastic frame with a shoulder around which a metalized Mylar film is wrapped. You can flow through P-10 gas ($A+10\%CH_4$) if you want to count in the proportional region, or Q gas for the Geiger region. The sample is mounted on a plastic split cylinder, sawed in half lengthwise, which is taped around the counter with Scotch electrical tape--not ordinary Scotch tape which is fairly radioactive. The sample can be changed easily. It is easy also to have around your laboratory a collection of these counters of varying size and to choose a size appropriate to the sample. To give some idea of the background in a device like this, the one which we use most frequently, whose diameter is about 1.5 centimeters, and length 6 centimeters, has a background of 0.15 to 0.4 counts per minute. This is largely because of the low Z and cleanliness of plastic materials.

The geometry in the counter is about 30 percent. One does not dare bring the sample cylinder closer in than a millimeter or so. Geometry can be calibrated with a standard source. It is quite reproducible by our 10 percent standards, as Dr. Grummitt pointed out. Even carbon-14 can be very usefully counted in a detector like this. The window thickness is a milligram per square centimeter or a little less. The absorption will be about 30 percent for carbon-14. A version of this counter which has also been used is to surround this with another jacket and make a jacketed gas flow counter. Someone I know has used this for studying very small quantities of helium-6 that he makes in a nuclear reaction. It is a very flexible sort of detector although I do not want to claim unique advantages for it. There are many others used similarly.

MANOV: Would you identify P-1 gas a little further?

ARNOLD: P-1 gas was spoken of earlier. It is 1 percent methane and 99 percent argon. P-10 gas is 10 percent methane and 90 percent argon. I do not know that Q gas is. It has been various things at various times.

Let me illustrate the problems you run into in low level work. We had some trouble getting the aluminized Myler when we started. We had an evaporator three laboratories down from us,

and we arranged with one of our friends to evaporate some aluminum on the film for us. The film came back with two or three counts a minute of alpha activity, and we were puzzled until we learned that this unit had been used for shadow casting in electron microscopy. Uranium is a favorite metal for this. Ensuring purity of materials and reagents is a major problem. It helps to buy them in large batches at infrequent intervals, checking each time. Lists of approved suppliers circulate privately, but this does not guarantee that the next batch will be free from activity. Everything should be exposed to circulating laboratory air only briefly, especially in bomb test periods.

Let me move on then to gammas. The first thing to say about gammas is that almost everybody owns a low level instrument for gamma rays. You can do very well indeed with an ordinary single channel scintillation spectrometer, not for the very highest energy gammas where the efficiencies are rather low, but for that particular ideal region up to four or five hundred kilovolts where the efficiency for a smallish crystal is quite high, where a large fraction appears in the photopeak.

We have used this instrument very extensively for beryllium-7, which has a gamma of 479 kev. We have also used it at lower energies and occasionally higher energies. What precautions should one take to make this as low a level instrument as possible?

I think the only precaution that has to be taken very seriously is that it is much better not to use the lead shields that have been customarily used. Mercury shields on the average are a good deal better than lead shields and do not, for such a small detector, require prohibitive quantities of mercury. Mercury ought to be surrounded by some further shielding material. The best thickness of heavy metal is probably at least four inches. A very common design is to surround the two inches of mercury with two or four inches of lead. The great advantage of mercury and the reason we have never been tempted to go to more exotic shielding materials, is that mercury is one thing that a chemist can purify for himself. It is quite easy to get free of base metals.

The resolution attainable in the instrument is relatively unimportant, because of the relative unimportance of the B parameter. If you improve the resolution from 10 to 8 percent in a detector you just get that fractional gain of reduction of background. The background in our experience does not show peaks at any energy. One sees a general fall off with energy indicating that many gamma rays are contributing. At the extreme upper end the background seems to be contributed by cosmic ray events. Our background does not seem to disappear even above the hardest gamma ray known to occur in nature. Some figures might be helpful here. With a two-inch well crystal, which we used for reasons of geometry, the background under the beryllium-7 photopeak with a crystal canned by Harshaw, is about 10 counts per minute in a mercury shield. This is with a 42 volt base setting and an 8 volt window. In the past we have run with quite wide windows to increase stability, but this is now less necessary. It is best to set the base line accurately where the count rate for the particular peak is at a maximum.

This background is considerably reduced if one gets a can of the proper kind, as Marinelli first pointed out. We have a crystal prepared for us by the Argonne National Laboratories which was canned in stainless steel and has a background of five counts per minute. It is traditional that aluminum is a rather radioactive material. However, someone recently told me about a very careful study in which a large amount of aluminum was used and found not to be radioactive. I can only say this is quite typical of the way the experience goes in the low level business. Copper is often a very much better material.

BORKOWSKI: It might be useful to know when trying to evaluate the relative backgrounds of various materials, that, if you happen to have access to a reactor, one of the fastest ways of determining radium containing materials is to determine the fission counting rate. A source which gives you one count per hour in a geometry like this may give you 10^4 fissions per minute in a flux of 10-inch neutrons/cm^2/sec. If one makes a screening test of many materials, one can very rapidly eliminate unsuitable materials, provided the neutron flux is available.

ARNOLD: This leads very naturally, I think, to the topic of evaluation of materials on which we have had a lot of progress in the last couple of years. Two other excellent tools can be used for this purpose. One is the Los Alamos human counter. This is a liquid scintillation device in which a hundred phototubes look into a chamber in which a human being is inserted. This device can count the amount of potassium in a human body to 1.5 percent in a hundred seconds. Nothing is ever counted in counts per minute but in counts per second. It is a very useful device for checking shielding materials.

We have had aluminum and various sorts of steels looked at in this device. It is a great comfort to have them look at something before you build up your unit and find it has been contaminated.

An even more sensitive tool for this purpose, is the technique that Marinelli and his group have developed at Argonne also for the purpose of studying radioactivity in human beings. This consists simply of a very large sodium iodide crystal in a room which has the requisite eight inches of iron surrounding it completely. The advantage of the Marinelli technique is that it is possible to do pulse height resolution and thereby to identify the undesired contaminants.

There is one further point that should be made on the matter of testing. Dr. Grummitt was one of the rediscoverers of what had become a lost art; namely, doing radiochemistry on the shielding materials before one uses them. One dissolves them up and extracts various of the obvious contaminants. This pushes your sensitivity to a very satisfactory level.

It is also very helpful, if you have one around, to use a multichannel analyzer for gamma work. You will accumulate your statistics a great deal more quickly and you will have a great deal more control over impurities.

LAZAR: We have looked at some sources of background and have found that the main source of background in scintillation counting was, as Dr. Arnold has said, the construction materials in the phototube. We have looked at the gamma ray backgrounds from crunched up phototubes.

Dumont supplied us with the glasses and the construction materials which they used, and then we crunched them up.

We found that the tube bases, mica filled bakelite, were notoriously hot. I do not have figures available with me, but the phototube bases were a major source of activity.

When we attempted to do low level work, and this is not at all comparable to the work that Dr. Arnold has described, the bases were the first thing that were determined to be hot. Second, the glass on some of the phototubes is hot. Here it depends on which section of the phototube you are talking about. They use several kinds of glass. The face plates they use are not particularly hot. When I say hot, what I am referring to is mainly potassium and radium and some thorium contamination.

ARNOLD: You have been looking at them with gamma techniques.

LAZAR: Yes, 2π geometry, essentially, with an anticoincidence mantle in a lead surrounding shield. The bases themselves contained mostly radium and potassium. The face plate glass was relatively cold and we could not determine the nature of the activity.

The tube sides of the photomultipliers were relatively warm. They were not nearly as hot as the bases. The hottest glass that is used is the lead glass inserts that are used to allow the wires to come through. They use lead glass because the expansion coefficient compares with that of the metal wire. This was found to be very hot and Dumont knows it is hot. They mentioned at the last scintillation conference a couple of years ago they were going to try to change this but I have heard nothing further.

I might mention that the aluminum that we use does not seem to be hot. One thing which we, surprisingly, found to be reasonably hot was a piece of wax which we used to seal the aluminum phototube. It turned out that this contained quite a bit of radium.

GRUMMITT: Does this apply to RCA tubes?

LAZAR: I do not know where RCA gets their glass. We have not crunched up any RCA tubes.

ARNOLD: The use of lead glass is a pretty universal practice in the industry and I think the RCA phototube bases are also the same material, at least by appearance.

LAZAR: If you can substitute the black bakelite which is not mica filled, this is not as hot. Therefore, we are now making our own bases.

CAMPION: What reduction in the background can you get by removing the bases?

LAZAR: This is a difficult question for me to answer for two reasons. One, I do not have the figures here, and two, it is a function of energy. When you go above 1.5 Mev, then the reduction is quite small. But the potassium content gives you a 1.46 Mev gamma ray, and you can observe a very distinct decrease in the potassium background. We have obtained reduction factors greater than two in potassium. This is just for the potassium-40 ray. At low energies the spectrum comes up rapidly and will be much less sensitive to this. I do not think that answers your questions but that is the best I can do.

ARNOLD: I am reminded of one item I did not cover and this is the question of counting time, although I think it was implicit in the earlier discussion.

The first thing that one has to get used to in low level work is counting for a long time. Forty-eight hours seems to be a common counting time. It has been distressing to me to discover that one of the great barriers to getting people to adopt low level techniques is that the idea of counting for longer than ten minutes is somehow repugnant to them. It is certainly necessary for any of these purposes to get over that particular repugnance. I have known people who counted for two weeks and even months on particular samples. Obviously, since the improvement in statistics goes only as the square root of the counting time, one must stop somewhere.

BAPTISTA: In this connection, can you be sure about the constancy of the background?

ARNOLD: This is a crucial problem. It can be tackled in different ways with different sorts of detectors. In the liquid scintillation counter where the cell is external and easily removed, one simply changes back and forth with sufficient frequency to have a running account of the background through the run and you are not then dependent on guesswork in that matter. Dr. Kulp will discuss this problem in gas counting. Certainly the constancy of your background and the constancy of the sensitivity of your detector must be checked, if possible, by external means. All questions of stability are crucial. This group surely has had some experience with stability, since for different reasons people concerned with standards also wish to have their units extremely stable in every respect.

BEAR: I just want to mention that, so far, most of the large scale shielding materials you have spoken of have been iron. At the radioactivity center we were thinking of building a low level walk-in room. We investigated a number of different materials and found some fire bricks made out of a material called "Dunite" which seemed to be good. We got a sample of these bricks and have built a test room with 18-inch walls and find that we get backgrounds, by lining this with a small amount of lead, that compare favorably with the results that Marinelli gets in his nine inches of steel. We think that this material will be less costly than steel.

KULP: If you have the volume, Libby's suggestion of sugar is still the best.

ARNOLD: I heard an Oak Ridge suggestion of water which I think is very interesting. Sea water is not so good because of its potassium content. We did some calculations as to what you could get by sinking a counter about 15 feet in Lake Michigan. The only thing you have to worry about is that your containing vessel for the equipment would have to be clean. But the figures are very impressive. Even five feet of water is really pretty good. Maybe I should mention some detectors that have not been really fully worked out.

One thing that we are anxious to try for very low level applications is a device which has been studied at Oak Ridge, as Dr. Lazar mentioned--the scintillation crystal surrounded by a large tank of liquid in anticoincidence. These people use it to sharpen up their spectra primarily but it has some potentialities as a low level gadget. I have spoken of these very large detectors in a slightly kidding way but in all seriousness they are potentially very valuable, not only as detectors for very low specific activities of gamma rays and material inserted in the center of a large detector, but also in solution.

Ernie Anderson and I had a little joke that went this way. For $100,000 we undertook--this was five years ago--to build a gadget which would go back 100,000 years for carbon-14, that is a dollar a year. This is a joke because the carbon-14 method will break down for other reasons before one can get to such sensitivities. A specific activity of a soft beta emitter of 10^{-5} disintegrations per minute per gram is surely within the present range of technology, if enough material is available.

PUTMAN: In defense of low background neurosis, this particular merit of $\frac{S^2}{B}$ depends on the supposition that the background is in fact random. Depending on the origin of the background this may well not be so. Doesn't this mean that it is quite a good thing to reduce the background to the lowest possible level?

ARNOLD: Yes. In fact, one good excuse for reducing the background is in order to make it easier to determine the origin of the remaining background. I would not like to leave everyone with the impression that the background does not matter. It matters for reasons, as you quite properly say, which go beyond the simple statistical limitation. Certainly non-statistical sources of background are one of the very largest and deepest "booby traps" in this business. The lower the background is reduced, the more easily sources of spurious counts, which tend to come in bunches, will tend to show up, and then they can be eliminated.

PEACOCK: One should emphasize that in low background counting you can go to great lengths to be sure that your pulses are random.

ARNOLD: That is certainly true. My students learn statistics before they learn anything about radiochemistry. We are always doing tests of significance.

An interesting type of instrument is the screen wall counter, although the device actually is not in very widespread current use. I think anyone who has read Libby's book has some idea what it is. It is simply a counter in a totally enclosed chamber, making provision for a direct sample-to-background comparison. A solid sample can be moved back and forth, in and out of the sensitive volume. It should be used more than it is because of this virtue which gas counting methods, with all their other advantages, lack.

GRUMMITT: I would like to describe two new approaches to the problem of counting external beta sources.

As both Dr. Arnold and Dr. Kulp mentioned, it is very easy to get an external counter down to one count per minute. Perhaps with effort down to one-half count per minute, but to go below that, and some of us seem to want to go below that, requires a great deal of effort.

One approach to this problem developed by W. Cross at Chalk River was to take a thin anthracene crystal with anticoincidence and iron shielding, and put the sample below it. The

geometry of his system is the following: A 0.015-inch thick anthracene crystal is mounted on a quarter inch of lucite, attached to a one and a half inch photomultiplier. This is surrounded with an anticoincidence shield which consists of a crystal and photomultiplier placed in anti-coincidence with the first one.

The source can then be introduced between the two crystals through a slide arrangement. In the best shield, and with a small amount of mass between the crystals, the best background to date is 7 counts per hour. He has made several attempts to reduce this, including putting it in the mercury-iron shield under 28 feet of concrete, but so far nothing brings it down below that value.

Because we had Geiger tubes available for anticoincidence and associated electronic circuitry, we preferred to set the thin crystal on a tray of 10- x 10-inch cosmic ray Geiger tubes. We also placed one around the side to provide a little better shielding. With lead surrounding the top, our background is 15 counts per hour, so it is fairly easy to get down into this region with this arrangement. I am pretty certain that this arrangement would give 7 counts per hour if we desired to change the shielding.

This system is fine for high energy betas. For yttrium-90 the efficiency at this level of background is 30 percent, so it is satisfactory for the strontium problem, but I would not advocate it for anything of energy less than 1 Mev.

The other approach was to apply the cosmic ray telescope technique to the standard end window counter. If we start with an end window Geiger tube surrounded with some sort of anti-coincidence shield, then one can place under this a thin double window counter. Coincidences are counted between the double window and the end window Geiger tubes with the source placed so that beta rays traverse both counters. The geometry of the system can be kept quite high. One can increase the diameter of the counters without affecting the background appreciably. Again one achieves a similar reduction of background.

One advantage of this system is that it is relatively insensitive to gamma rays so one can use lead for shielding. The background that we observe is 15 counts per hour at 30 percent efficiency.

ARNOLD: What would be the background of the upper counter in the anticoincidence ring but without the lower counter?

GRUMMITT: The background of the counter was high as I purposely used one that was slightly contaminated--3.3 counts per minute in the lead shield. Normally we would expect it to be 1.8. This difference is presumably contamination, or spurious counts from the wire, I am not certain which.

ARNOLD: That would really be eliminated by such an arrangement. My question was prompted by a little puzzlement that this was so effective in eliminating gammas. Compton electrons might be expected to behave very much like your electrons or even photoelectrons might penetrate from one unit into the other.

GRUMMITT: If they do penetrate, the efficiency of counting is the product of the two.

ARNOLD: No, because it is an electron. The electron goes through.

GRUMMITT: These electrons have a relatively short range.

ARNOLD: It depends on what you count.

PEACOCK: What kind of windows do you have in the region between the two counters?

GRUMMITT: They are Mylar window atmospheric pressure counters. The gain here, of course, is not startling. Neutrons will count in this system as in the other one because they will trigger both counters. Any uncancelled cosmic rays also will count. So one has to have a perfect cosmic ray shield, which we did not have in this case. There was a small escape area above.

SELIGER: You are counting strontium here.

GRUMMITT: This will count anything with reasonable efficiency. One can count down to moderately low energies with this arrangement, because one is only limited by the choice of windows and the mass of gas in the counter which is perhaps a half to one milligram per square centimeter.

2. Low Level Counting

J. L. Kulp

The pressure to develop low level counting techniques has come from disciplines other than pure physics and chemistry. The most important immediate applications of these techniques have been in the fields of geology and archaeology although the potential contribution to medical science is equally great. In all cases the power of the technique lies in the extreme sensitivity that is available. For example, at the Geochemistry Laboratory at the Lamont Observatory such exotic measurements as one or two disintegrations per minute of tritium per 30 gallons of sea water or a disintegration per minute of carbon-14 in some ancient piece of wood resurrected out of a lake bottom are made.

Our experience in low level counting began with the construction of a system for the black carbon method for radiocarbon dating in 1950 with the help and advice of Drs. W. F. Libby and J. R. Arnold. The need for greater sensitivity and freedom from fission product contamination led to the conversion to proportional counting techniques in 1952 and 1953. The development of these rather neat proportional counting methods, particularly for carbon-14, is largely a result of major break-throughs made by Dr. G. J. Fergusson in New Zealand and Dr. H. DeVries in Holland. These two men quite independently showed that proportional counting of carbon dioxide was possible. About the same time Dr. Hans Suess developed a method for the proportional counting of carbon-14 using acetylene.

During the next two to three years a number of academic and commercial laboratories developed different subspecies of these low level techniques based on proportional gas counting. Dr. Arnold has very well covered the background and general principles involved in low level counting. Therefore, it might be useful to describe in some detail the Lamont apparatus and technique which represents a typical modern carbon dioxide proportional counting system and discuss some of the parameters so that you can see the kinds of problems involved, and the levels of precision and accuracy that are possible.

The key to most of low level beta counting is an anticoincidence ring. Figure 1 gives the view inside the Lamont shield which consists of 8 inches of iron, successively inside of which are anticoincidence ring, the half-inch of mercury encased in a steel container, and two five-liter proportional counters made of copper.

The proportional counter has the advantage of near 100 percent detection efficiency for soft beta emitters that can be put into gaseous form. By using a fairly large volume the sample count rate can be maximized. Using carbon dioxide at two atmospheres about five grams of carbon are contained in the counter which yields something on the order of 60 counts per minute for a contemporary carbon sample.

In low level beta assay the selection of the counting gas is important. If carbon-14 is the isotope to be measured there are quite a variety of choices. Methane behaves well in the proportional counter but is fairly difficult to prepare. Acetylene has twice as many carbon atoms per liter and is rather straight-forward to make, but considerable skill is required to prepare this routinely without accidents. With both methane and acetylene the chemical preparation produces minor isotopic fractionation, hence carbon-12/carbon-13 measurements must also be made to monitor this effect if high precision (one-half of one percent) is required.

Carbon dioxide is attractive from the point of simplicity of preparation but apparently was avoided before 1950, as reports in the literature suggested that it was a poor gas for proportional counting. Fergusson and DeVries found that this problem was one of traces of electronegative impurities and if these were removed, carbon dioxide was a good counting gas.

Dr. Manov asked about the purification procedure. Actually in principle it is very simple. The most common offending gases are oxygen, water, hydrogen chloride, nitric oxide, and

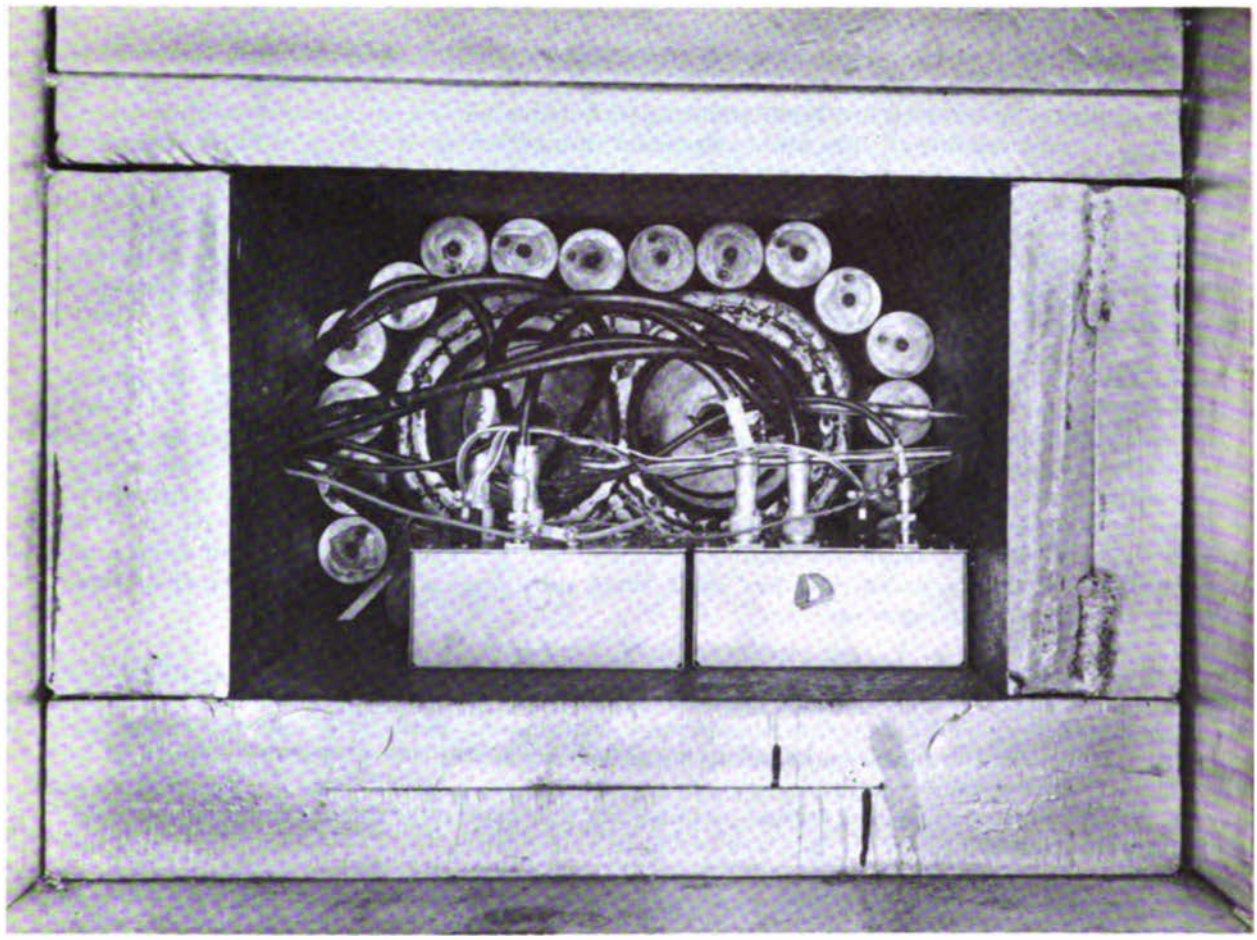


Figure 1. Lamont low level gas counting equipment.

sulfur dioxide. Chlorine, if it were present, would interfere, but we have never been able to identify it.

ARNOLD: We once thought we got some.

KULP: The chemical procedure for preparing the pure carbon dioxide is about as follows: If the material is combustible, it is burned. If it is in the form of carbonate, phosphoric acid is used to liberate the carbon dioxide. Once the carbon dioxide is obtained the gaseous impurities must be removed. The removal of water is relatively simple, because the gas goes through a dry ice trap and a phosphorus pentoxide drying tube before it goes to the counter. Oxygen contamination at certain levels is not a problem. It has been determined that oxygen at the level of about five parts per million or less does not give a significant decrease in counter sensitivity. In fact, if a limited amount of air is mixed with pure carbon dioxide and if the carbon dioxide is simply frozen out with liquid air, the system evacuated, and then the process repeated a few times, the carbon dioxide is perfectly suitable for counting again. If too much air is used, apparently sufficient sulfur dioxide is added to contaminate the carbon dioxide to a concentration such that purification by this procedure is not possible.

The real difficulties appear with hydrogen chloride and sulfur dioxide, particularly in samples which are prepared from burning where these gases commonly reach the parts per million level. It seems that they are of the order of at least one hundred times worse in their effect on the sensitivity of the counter than pure oxygen.

The purification consists of passing the carbon dioxide through two bubblers, silver nitrate solution to remove hydrogen chloride and a cleaning solution to pick up sulfur dioxide. The partially purified carbon dioxide is absorbed in ammonia, precipitated as calcium carbonate which is then thoroughly washed, and carbon dioxide is released again with phosphoric acid. After this, still another step is required because although this carbon dioxide is quite pure, it still commonly contains tens to maybe hundreds of parts per million of things like sulfur dioxide.

The last step of purification is to react the carbon dioxide with pure calcium oxide at about 700 degrees, lower the temperature to 500, evacuate, and then raise the temperature to about 900°C, at which temperature the carbon dioxide is again released, this time without the electronegative impurities. This last step cannot be used alone since excess sulfur dioxide or hydrogen chloride will sufficiently contaminate the calcium oxide that subsequent purification will not be effective. The total purification process reduces these impurities below the parts per million level, yielding carbon dioxide with excellent proportional counting characteristics.

One other point about this chemical procedure concerns radon contamination. The effect of radon can be eliminated electronically, which Fergusson has done by a rather elegant system. It is simpler to get rid of any radon by carefully selecting calcium carbonate for the calcium oxide. Merck's reagent grade calcium carbonate that is very low in barium has proved adequate. The particular lot of calcium carbonate that is selected is analyzed in our laboratory for its radium content by low level methods to insure that the amount of radium in the calcium oxide is sufficiently small so that, for the time in which the carbon dioxide is in the calcium oxide furnace, the amount of radon generated would contribute negligibly to the radioactivity of the sample gas.

Tritium is generally counted as molecular hydrogen mixed with other gases. Argon-ethylene fillings have been used as a Geiger gas, but subsequently it was found that consistently better performance could be obtained by proportional counting of hydrogen gas in methane. Tritium can be analyzed in natural waters at the level of 0.1×10^{-18} T:H ratio, provided that a 2,000 fold pre-enrichment by electrolysis is effected. The problem here as first worked out by Libby was to start out with gallons of water and electrolyze down to a few cubic centimeters, monitor the tritium enrichment by the deuterium enrichment, and then count hydrogen gas at the level of 10^{-15} T:H.

MANOV: How can you be sure that your tritium is retained quantitatively on such an electrolysis?

KULP: The enrichment of the deuterium is determined quantitatively, and from that the amount of tritium which is retained can be calculated.

MANOV: But you have to make certain assumptions regarding your electrolysis of tritium versus deuterium versus hydrogen which may not hold as your electrolysis proceeds.

KULP: Yes, but certain samples have been run with tritium spikes that indicate that the theoretical fractionation is about right. The error from this source does not exceed five percent which is quite adequate for the precision of absolute tritium determinations in natural samples.

Turning to the counter characteristics, a counter such as is shown in Figure 1 has a background of the order of 2,000 counts per minute on the laboratory bench. By putting that inside our iron shield, this will be reduced to about 500 counts per minute. Then by placing it inside the anticoincidence ring, the background is reduced to about 20 counts per minute. Finally, by putting an inch and a half of mercury around it, this is reduced to about 15 counts per minute. The routine counters shown in Figure 1 give a modern carbon sample count of about 60 counts per minute over a background of 15. For standard laboratory procedure a 20 hour count is used for most of the low level work at Lamont. This is convenient because the system can count all night and the sample is changed during the middle of the day.

In order to determine the stability of the electronics, two sorts of things are done. First, an external cobalt-60 source is used at the beginning and end of a run. The stability of the electronics now in use is such that the variation due to this source is less than 0.2 percent of the count.

A less precise (1-2 percent) but continuous monitor of the sensitivity is obtained by counting the total counts in the detector, i.e., cosmic ray mesons plus background plus sample, and comparing that with the cosmic ray meson flux for the run as determined from the total mesons from the anticoincidence ring. By subtraction the meson count in the sample counter and thus the average sensitivity can be determined. Such a measurement provides for the detection of a series of spurious counts particularly during the night. In practice it turns out that the electronic stability is not a significant problem, and day after day the same sensitivity is maintained within a few tenths of a percent providing the gas is pure.

The midpoint of the steep portion of the count rate versus voltage curve is extremely sensitive to gas purity, hence measurements at this point are made at the beginning of each run. Ten parts per million of oxygen will only affect the plateau value on the order of about a half percent, whereas it will change the count rate at the purity check point by 40 percent. If significant impurity is present the sample is reprocessed prior to counting. At the present level of operation, this occurs about one out of ten times.

In trying to give some comparison of the different systems that have been used, keep in mind the count rate of 60 over a background of 15 in the present Lamont system. At Berne, Professor Houtermanns and co-workers have developed a system which employs the sample gas in the anticoincidence volume. The center wire is surrounded first by a cylinder of aluminized plastic which forms the cathode of the sample counter. In between this aluminized plastic and metal casing a series of additional wires form the anticoincidence ring.

This system has certain advantages because electrons that are kicked out of the metal wall will not enter the sample counting space, resulting in a reduction of background of about 20 percent. Both the system employed at Berne and the system that DeVries is using in Holland will give something like 15 counts per minute for modern carbon with a background of 1-2 counts per minute.

It can be shown that as far as $\frac{S^2}{B}$ (the usual "figure of merit") is concerned, there is not a great deal to choose between these two systems. In the case of carbon-14, using either system, it is possible to go back about 45,000 years, a time of the order of 6 or 7 half lives. In the case of modern samples, these systems are a bit superior for a 20-hour count, i.e., a statistical error of ±.3-.4% as compared with ±.5-.6% might be obtained.

These considerations point up the problem which several asked about during Dr. Arnold's lecture concerning the statistical distribution of the background. The important thing is to define the constancy of the background. If it is a random distribution in time then an accurate background can be defined by continued counting, and the error will at all times be determined by the uncertainty in the sample count. In reality there are step fluctuations in the background due to cosmic rays. With the large Lamont counters, it is somewhat easier to observe this phenomenon than with the smaller counters that have more limited background. It was first observed that there was a consistent fluctuation in the background as a function of barometric pressure. It was not a one to one correlation for two reasons: First, the barometric pressure at sea level is not directly related to the total mass of air above, and second, the nature and origin of the particles that cause these variations would be expected to be independent of barometric pressure which was indeed the case. The next step was to examine the relation of background variation with fluctuations in total meson count. It was found that the correlation with this parameter was much closer than with barometric pressure. Except for the night of the solar flare in early 1957, where the background went up by a factor of two or three, these variations amounted to ±0.6 counts per minute on the background of 15.0 counts per minute.

It was found, however, that by applying a correction of 2.2 percent change in background for every one percent change in meson flux, that the variation of background was reduced to ±0.2 counts per minute, whereas the statistical variation just due to the counting of the background is ±0.1 counts per minute. So at the present time, by taking the anticoincidence ring count and applying this linear correction, the background uncertainty has been reduced by a factor of three.

DeVries observed this phenomenon independently and decided that it looked like a neutron phenomenon. He proceeded to put boric acid filled paraffin inside the iron shield. In doing this, the background was reduced in several of his counters, depending on size, shape, and wall thickness, by 20 to 30 percent and, possibly even more important, almost completely eliminated the background variation. He found that by putting the paraffin on top of the shield, he did not get as good improvement indicating that a good proportion of these neutrons were being formed by events in the shield. DeVries has also put a neutron detector inside the shield to monitor the total neutron flux, and has used this as a direct correction. This is satisfactory, although it would appear preferable to eliminate the problem rather than employ additional expensive electronics.

One other observation was that whenever methane was used in these counters, the background was consistently higher than when we used carbon dioxide. This pointed to a neutron phenomenon, but it was not pursued further until DeVries came out with a rather complete description of the situation. Most workers in low level counting have at one time or another put additional layers of cosmic anticoincidence tubes in the ring and found that this does not improve the background. Therefore, this is not a matter of losing the mesons through the anticoincidence ring.

GRUMMITT: Does cadmium reduce the background at all?

KULP: I do not think it has been tried.

HAYWARD: Do atomic bomb tests materially affect the background temporarily at all?

KULP: Since the black carbon method is no longer used (which can absorb particles readily from air) there is no significant effect from airborne fission products. I think the reason is that the shields are moderately tight. Even if the gamma background goes up slightly in the room the iron shield takes care of this.

YAFFEE: May I ask about the neutron absorption? You first slow these down. What happens after that? What absorbs the neutrons?

KULP: Boron mixed with paraffin. DeVries seems to feel there is a fast neutron component which does not cause an appreciable background in the carbon dioxide, but does cause an appreciable background in the propane or methane counting. He did not observe the same improvement with methane counting using the boric acid-paraffin shield as he did with the carbon dioxide. He feels that there is still a fast neutron component that he really has not controlled in the methane counting.

LAZAR: One absorber which we used is boron carbide, which you can get rather thin and which does not produce gamma rays to any large extent. The intensity of gamma rays is not what you get with cadmium. It has a somewhat flatter response. We find that it does make an appreciable difference to the neutron component, not at very low energies, but we do see an observable effect. It may be of some help in this program.

KULP: That is true.

GEIGER: I think I might point out that the neutron production by cosmic rays goes up exponentially. This makes it very worthwhile to use shields.

KULP: Yes.

GEIGER: Those neutrons have about the same spectrum as the radium beryllium source has.

KULP: Thus the iron would be better than lead shielding.

GEIGER: Iron would be much better than lead and mercury.

PEACOCK: To be controversial, it would mean that iron was better than mercury if you got it pure enough.

KULP: You want to restrict the volume inside the shield so you are pretty much restricted to a high atomic weight material such as mercury. I suspect, though, that most of the neutrons are produced in the massive iron shield and not the relatively small quantity of mercury.

In summary, this type of counting for carbon-14 will allow plus or minus 0.5 percent precision on modern carbon, where modern carbon gives about 13 to 14 disintegrations per minute per gram of carbon. Further, it is possible to go back about seven half lives until you reach the two sigma level which is where most of us put a "less than so many years."

There is one other thing. To get either greater precision or greater sensitivity is a very difficult problem, because everything begins to work against you exponentially. If you try to expand the size of your counter, then the neutron effect goes up and more material of which the counter is made is present to contribute activity.

At Lamont carefully selected electrolytic copper is used for counter construction but even this is not entirely free of radioactive contamination. The copper is welded in a helium arc so there is no solder except at the Kovar-copper joint on the outside. Even the Kovar for the metal glass joints and the low potassium glass is specially chosen.

ARNOLD: One should say that solder is quite normally a very radioactive material.

KULP: This is probably one of the best reasons for the mercury shield because the anti-coincidence ring has a bit of solder. This problem is avoided in the Houtermanns' system.

BORKOWSKI: What about increasing pressure?

KULP: This is quite feasible. The Humble Oil Company laboratory started with this procedure. They started with a small counter of a half liter to a liter, and ran it up to 5 or 10 atmospheres of carbon dioxide, thereby getting generally the same $\frac{S^2}{B}$ of the other systems described earlier. This requires up to 10,000 or 15,000 volts, and produces certain stability problems. This technique is possible but I believe that, after some considerable experience, they are running routinely at somewhat lower pressures.

I might say also that the voltage required for the 5 liter counters is about 7,000 for two atmospheres, but 5,000 of it is put on the case and the rest on the center wire in order to avoid too high a voltage across the condenser. This seems to help in maintaining the stability. There are serious problems with increased pressure but certainly some increase appears to have merit. This is another advantage of carbon dioxide over acetylene which supposedly has explosive characteristics above atmospheric pressure.

This concludes my remarks on soft beta counting. I would like to touch on several other types of low level counting.

Consider the low level counting of solid sources of reasonably hard beta emitters such as the kind of problem involved in the world-wide strontium-90 assay. Here again, particularly in

human bone samples, one to a few disintegrations per minute per sample are all that are available. There are two methods that are being used for this at the present time, both using massive shielding and anticoincidence. I won't bother you with the chemistry except to say that you wind up with a pure strontium-calcium solution from which you milk the yttrium-90 daughter, which is what is counted. This can be put on as small and well-defined an area as you wish. Once this yttrium precipitate is obtained, it is counted in one of two ways. Some of the laboratories which are in this business have been counting it by the cylindrical flow counter, described by Dr. Arnold and developed by Dr. Libby, which employs a thin metal-coated plastic film as the container. In this particular case the solid sample is slurried on to hemicylinders and placed around the counter. This has the advantage of a somewhat higher efficiency and better geometry factor than an end window counter, but obviously suffers from lack of convenience. Further, in this type of work the half life of the yttrium-90 must always be checked since you are dealing with a few disintegrations per minute in the presence of other possible emitters. Therefore, the samples must be handled repeatedly.

A more convenient method utilizes end window counters. A thin tabular end window counter of the Anton type that has a low background ($\sim$2 cpm) is placed inside an anticoincidence ring and lead shielding. Such a counter has a 25 percent efficiency for yttrium-90 betas mounted as yttrium oxylate on a planchet covered by a thin plastic film. This permits handling essentially as is done in ordinary tracer level techniques.

The background of these end window counters can be reduced further, although for the strontium-90 problem it really is not necessary. I think Tracerlab's new flow counter of similar geometry but made largely of plastic, has a background of something under 1 cpm.

PEACOCK: Five- or six-tenths of a count per minute.

KULP: There is a British company that is now making a sealed Geiger tube where the housing is out of plastic, sputtered for condution. They claim a background of 1 cpm which should do the same job as the Anton tubes of 2 cpm, but we have had three successive defective tubes from them. It turns out that in going from 2 cpm down to a 0.5 cpm there is no large gain.

Now, just a few comments on low level alpha counting. Thick source alpha counting is required for assaying rocks for total alpha activity. It is also useful in thorium free rocks for determining radium or estimating radium content. The best way to do this where adequate sample is available is to use a 5-inch photomultiplier tube coated with zinc sulfide phosphor as proximate to the sample as possible. By taking special care with the grease that is used and selecting the phosphors, a background as low as 30 counts per hour background is possible. This permits assay of 10^{-14} to 10^{-15} grams of radium per gram of solid.

The system used at Lamont over the past five years for low level radium measurement consists of a four liter ionization chamber which is arranged as a slow pulse device. A vibrating-reed electrometer is used to amplify the individual ionizations due to alpha particles from radon and its decay products. There are two ways in which this has been done. One is the so-called fast pulse counter which uses high voltages and can handle a large range of activity levels, but has inherent problems of microphonics and other instability. We have used the slow counter which employs a 300 volt potential between the center probe and the walls. This is an attractive technique for low level work since the gas can be quite impure without affecting the efficiency; the system is not microphonic, and the chemical and electronic systems are simpler.

In order to achieve the desirable background of 10-15 counts per hour, the four liter chambers must be carefully selected. Stainless steel was fine up to 1952 or so when metallurgists decided to flux stainless steel by introducing some rare earth oxide. Now stainless steel is hot as far as low level counting is concerned. Electrolytic copper appears to be the best material at present.

With this system it is possible to measure 1×10^{-13} grams of radium, equilibrated with radon and bubbled out of the solution into these cans, with a reproducibility of 5 percent. The absolute accuracy is known better than this because the counters are calibrated with the Bureau of Standards' radium solutions which are known to within a percent or so.

It is quite simple in geologic studies or human bone analysis to run a hundred gram sample. Therefore, it is quite easy to go down to 1×10^{-15} grams of radium per gram of sample at the plus or minus 5 percent level for most materials. It is more difficult, but not impossible, to use a kilogram of material and, if a standard deviation of 30 percent is acceptable, then 1×10^{-17} g Ra/g may be determined with this technique.

BAPTISTA: Do you think that for these radium measurements you can use the system that consists of a vessel coated with zinc sulfide and put on the photomultiplier? Using discrimination you can get a very, very low background, and I think an efficiency of about 60 percent.

KULP: The system I described gives about 80 percent efficiency for those alphas that enter the gas phase. Further, the zinc sulfide contains some radium. As you know, it is hard to get activated zinc sulfide that has no radium. We have done this experiment just trying to count phosphors to try to get the lowest phosphor. Most of these tend to be in the range of 10^{-14} g Ra/g. Since the background in the ionization chamber-counter method is almost entirely from the walls it is hard to see that the zinc sulfide scintillation technique would yield a lower background per volume of gas.

BAPTISTA: I think for some results that I saw you can get a comparable background by these systems.

KULP: I think in principle one might do as well with it.

PEACOCK: Dr. Kulp, do you want to mention a little cookbook experience on cleanliness around the laboratory or keeping things like the natural decay products from getting into the equipment, or have you run into that problem.

KULP: This can be a serious problem, particularly on things like this strontium-90 type of analysis. In that case you control matters adequately by checking the half life providing no one in the laboratory has any hot strontium-90 solution. In the case of the gaseous emitters such as carbon-14 and tritium, it is no problem at all, because it is so easy to clean them up. In the case of these alpha problems, I think normal cleanliness takes care of it, because you are dealing in this case only with radium. It is certainly specific. Maybe you were referring to something else.

PEACOCK: I thought it might be interesting to know whether or not it was a problem. I know that in the case of some of your low level Frisch grid chambers, if you are not particularly careful to keep the parts away from the atmosphere after you have gotten them well cleaned up--if you leave them lying around and dust them off--a certain amount of polonium contamination may get into them, something that people might not think of. It may be worse or it may not be as bad as some of the chemical procedures.

KULP: Polonium is the bad actor as far as natural air contamination is concerned.

ARNOLD: On this general question, the low level game is surprisingly free of horror stories of this kind, although there are a goodly number. There have been cases of laboratories where people were unable to operate for carbon-14 even, because of radon in the air. As Dr. Kulp said, the old carbon dating with carbon black was really pretty awful this way. Most of the rest of us, I think, are not famous for running our laboratories in a sterile fashion. We try to use good technique and we know in individual cases things to avoid. I would not think that anyone who has had five or ten years of lab experience would need to be told. The one thing you do is to try to physically separate projects if you have a lot of other radioactive work going on. You obviously don't try to run the two side by side.

PEACOCK: May I suggest again for the record, would you recommend that these be installed in air conditioned laboratories or filtered air laboratories or is it perfectly all right to use a shed?

ARNOLD: This is an excellent excuse to have an air conditioned laboratory, but my lab, for example, has never been air conditioned.

REYNOLDS: Do you think it is wise from the point of view of thoron uptake and, therefore, thorium series activities getting into your equipment, to take air in from some distance, say 50 feet above the ground? In other words, with a sort of inverse stack. This has been proposed for our low level facility at Oak Ridge.

KULP: The contribution to the carbon-14 counting system due to the gamma radiation from the air in the shield is negligible compared to the rest of the background, so this would not be worth the effort. I imagine in gamma analysis you might have the worst case. Even there I think the walls will contribute much more than the air.

ARNOLD: Many of these labs are located in the basement because of the weight of the shield. In our basement lab before we moved in, we did a rather extensive air filter study, and there was no activity to speak of. The air does come in from the roof, but also from open windows.

KOFOED-HANSEN: It is our experience that the thoron contamination comes from other places in the neighborhood so the stack would not help. There is a correlation with the wind direction in Scandinavia.

BORKOWSKI: During winter, of course, in the coal burning areas, this would be the worst place to remove or get an air supply. The thoron and radon background around Knoxville during the winter is tremendous.

GRUMMITT: The only place we find natural contamination is in the counting of alpha sources deposited on a platinum tray. During the first few hours you can sometimes observe a few counts per hour. But as it takes perhaps a week to count a sample at that level of activity, it is very easy to put the sample in the counter and leave it for a few hours before starting the count. This is the method we use to overcome this difficulty. We are also very careful to cover the slide and any part of the counter on which deposit might fall.

ARNOLD: It is always a good practice to enclose samples which are going to be recounted.

SESSION VI - NATIONAL PROGRAMS

G. G. Manov, Chairman

Summary

It has been said that technical meetings are the very lifeblood of science. It is particularly true in a field such as the standardization of radioisotopes where the fine points of techniques simply cannot be communicated by the written word. The opportunities for face-to-face, unhurried discussions generally result in an autocatalysis of ideas as we have seen in the past three days.

The program of the various countries reported in this session show a degree of sophistication both as to comprehensiveness of coverage and precision of measurements as to be unthough of only two or three years ago. Measurements of alpha standards to a precision of 0. 1 percent have been reported; intercomparisons of beta standards by several methods have been performed by several countries during the past several years, and each year sees a greater concordance between different methods as well as between different laboratories.

Two approaches to the problems of standardization are being pushed. One is the production of standard samples of radioactivity. The United States in particular has pursued this course most assiduously and, in order not to overwork its research personnel at the National Bureau of Standards, has arranged for a commercial company to prepare and issue the requisite quantities of short-lived standards on a continuing basis. Adequate cross-checking is an integral part of this program. The British, who also issue their own standards, are investigating the approach of preparing standard instruments, such as ion chambers. This approach, while it can be used for measuring the activity of long-lived isotopes, is particularly useful for short half-lived materials.

Both approaches are useful; undoubtedly both will be used widely in the future and particularly by laboratories other than those who themselves perform primary standardizations. It is hoped that the technical-industrial societies, for example, the American Society for Testing Materials, will assist in making such standard instruments a recognized part of testing procedures, in the same manner as has been done for thermometers, distillation flasks, etc.

It is a keen source of pleasure to me to see that standardization programs are under way in so many countries for the training obtained in making such absolute measurements is of the very highest calibre and useful in a variety of ways in other programs.

This session will deal with the description of the national programs of various countries.

1. National Program - United Kingdom

W. E. Perry

I would like first to give you some idea of the strength of the radioactivity measurements group at the National Physical Laboratory. This group is part of the Radiology Section of the Physics Division. Two other groups in that section deal with X-ray dosimetry. I think our group represents 10 percent or less of the total strength of the Division. There are three scientific officers, four experimental officers, and one assistant.

The main methods we use for disintegration-rate measurements are the 4π beta proportional counter method (Mr. R. Sara), the beta-gamma coincidence method (Mr. W. J. Callow and Mr. B. Owen), the 4π beta-gamma coincidence method (Mr. R. A. Lloyd) and the gamma-ionization chamber method, using graphite-walled ionization chambers (Mr. J. W. G. Dale).

Figure 1 shows the beta-gamma coincidence equipment. The beta detector is an anthracene disc 2 cm diameter and 1 mm thick, and the gamma detector, in the lower housing, is a sodium iodide crystal 4 cm diameter and 2.5 cm long. Clamps on the vertical support provide for the adjustment of the positions of the counters and the source. The figure shows the arrangement without any shielding against extraneous radiation; we have used lead shielding, particularly round the gamma counter, to reduce the background. Figure 2 is a diagram of the 4π beta-gamma arrangement. The 4π counter is of the pill-box pattern. There are two anode wires, and the source is mounted on a thin film. We generally operate the counter with pre-mixed argon (90%) - methane (10%). The gamma counter is a sodium iodide crystal with a photomultiplier. Figure 3 shows the 4π counter open, and illustrates the method of mounting the counter so that the source is as near as possible to the sodium iodide crystal. Here again we have used various shielding arrangements to reduce background. Figure 4 is a photograph of the equipment for gamma-ray ionization measurements using graphite-walled ionization chambers. The method is basically that described some years ago by L. H. Gray; the gamma ray activity of the source is compared with that of a radium standard, and, using the figure for the gamma ray emission from radium, together with the radium content of the standard, we determined what is in effect the gamma-ray dose rate in roentgens at the center of the chamber due to the source. Then from the so-called K factor, or what is now referred to as specific gamma ray emission, in r/mch at 1 cm, we derive a reasonably accurate value for the disintegration rate of the source. The figure shows the chamber mounted at the end of an evacuated brass tube 1 meter long, through which passes the wire connecting the central electrode to the Lindemann electrometer. The source holder and optical system for the electrometer are also shown. We find this apparatus very convenient for checking the strength of samples that we receive from Harwell and the Radiochemical Center, before we dilute down to the levels required for our standards.

We have been doing ionization measurements and beta-gamma coincidence measurements since about 1950 in connection with comparisons with other laboratories. Table 1 summarizes the results of measurements of two samples of cobalt-60 distributed by the Atomic Energy Research Establishment, Harwell to British laboratories in 1950 and 1951, of a solution supplied by the National Bureau of Standards in 1952, and of a solution we prepared ourselves from a solid source in 1955; measurements of our 1957 solution are not yet finished. Comparisons of the gamma ray activities of the solutions show that the consistency of the ionization measurements is about ±0.5 percent. The limits given for the beta-gamma coincidence values are standard deviations except for the 1955 values, for which the limits represent the total estimated error, which includes the variation with geometry in the case of the beta-gamma coincidence measurements (f). We have found that the 4π beta-gamma measurements using a proportional counter show less variation over the years than do the beta-gamma coincidence measurements. By that I do not mean that we suspect the beta-gamma coincidence method itself but it is just that we are not satisfied that we have the best experimental arrangements.

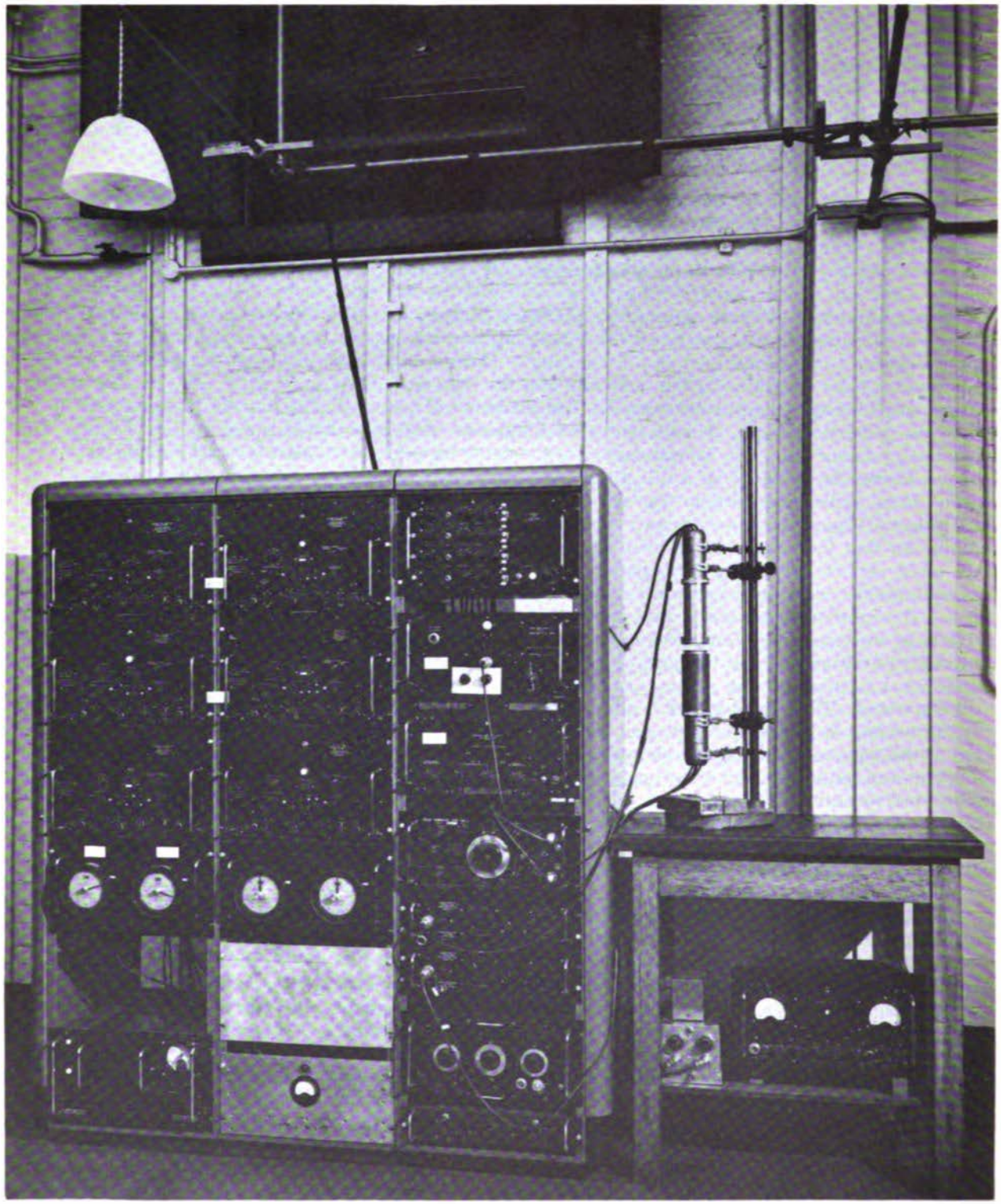


Figure 1. Equipment used at N. P. L. for measuring disintegration rates by beta-gamma coincidence counting.

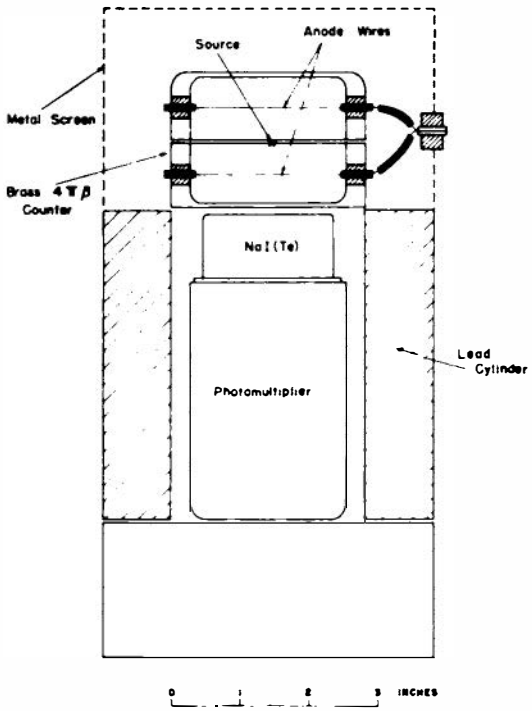


Figure 2. N. P. L. 4π beta-gamma coincidence apparatus.

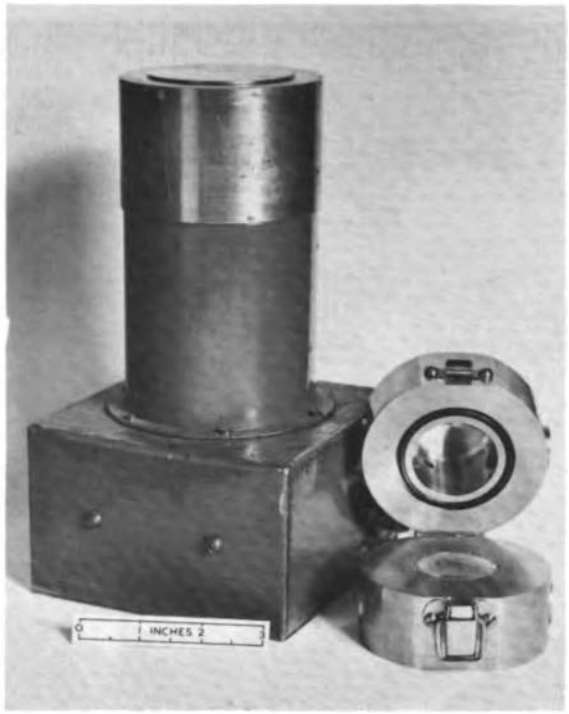


Figure 3. The 4π beta counter (open) and scintillation counter NaI (Tl) used at N. P. L. for 4π beta-gamma coincidence measurements of disintegration rate.

We use thin film for mounting the sources and keep the size of the sources as small as possible, but with the anthracene and sodium iodide crystal arrangement, we have not yet been able to get disintegration rate values which are independent of the geometry of our system. For example, if we separate the counters--the maximum separation is about 6.5 cm--and move the position of the source, we do not get the same apparent disintegration rate. In fact, values in closest agreement with the 4π beta-gamma values are obtained when the two corrections--that is, the correction for gamma ray sensitivity of the beta counter and the corresponding gamma-gamma coincidence correction--are about equal.

The minimum correction we have been able to apply so far, using the usual absorber method, is 6 percent. We are now using a much thinner anthracene crystal and are trying to improve the conditions to see if we can in fact eliminate the variation with geometry in order to compare the 4π beta-gamma coincidence measurements and the beta-gamma coincidence measurements a little more closely. If our measurements had been entirely consistent, all the ratios in the final column (Table 1) would be the same. It does not really matter what they are since the actual values in column 2 are somewhat arbitrary, depending as they do on the values adopted for W and the gamma output from radium.

We infer from the results that the beta-gamma apparent disintegration rates are tending to get a little bit low with time, but this question has not been finally settled. As I mentioned earlier, we are measuring another solution now and the results have not yet been fully examined.

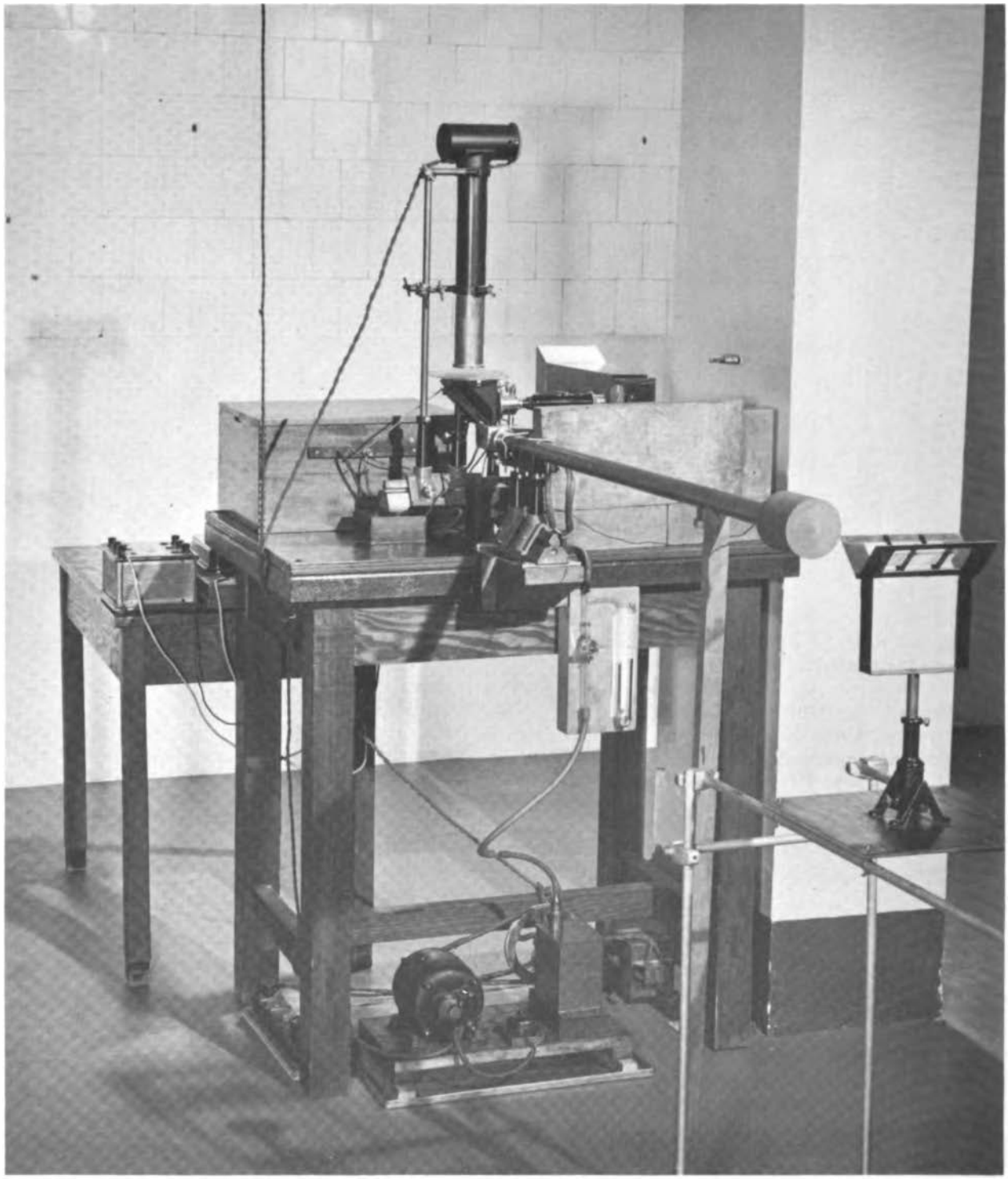


Figure 4. N. P. L. apparatus for gamma-ray ionization measurements, showing graphite chamber and electrometer system.

TABLE 1

Results of Measurements of Co-60 Solutions by N. P. L. 1950-1955			
Solution and year of measurement	Activity values mc/g at the time of measurement		Ratio of coincidence value to ionization value
	Ionization chamber[+]	Beta-gamma and 4π beta-gamma coincidence	
Co-60 solution (1) distributed by the A. E. R. E. in 1950 1950	0.534	(a) 0.534±0.007	1.000±0.014
Co-60 solution (2) distributed by the A. E. R. E. in 1951 1951	0.492	(a) 0.497±0.003 (a) 0.487±0.015	1.010±0.008 0.990±0.031
Co-60 solution distributed by the N. B. S. in 1952 1952	0.785	(a) 0.799±0.011 (b) 0.787±0.031 (c) 0.792±0.008 (d) 0.771±0.011	1.018±0.015 1.003±0.040 1.009±0.011 0.982±0.015
Co-60 solution prepared by N. P. L. in 1955 1955	0.173	(e) 0.1694±0.011 (f) 0.1701±0.025	0.979±0.011 0.984±0.016

[+]The ionization chamber values are based on 34 eV for W (energy per ion-pair) and 8.3 r/mg h at 1 cm from radium screened by 0.5 mm Platinum.
(a) GM detectors for beta particles and gamma rays.
(b) GM detector for beta particles; NaI (Tl) detector for gamma rays.
(c) 4π GM detector for beta particles; NaI (Tl) detector for gamma rays.
(d) 4π proportional counter for beta particles; NaI (Tl) for gamma rays.
(e) As for (d) but with improved counters and electronic equipment.
(f) Anthracene detector for beta particles; NaI (Tl) for gamma rays.

TABLE 2

N. P. L. Measurements of the Half-life of Co-60 Samples					
Cobalt Sources		Pile Factor	Gamma ray activity (1950) (mg Ra Equivalent)	No. of Measurements 1950 - 1956	Half-life y
A. E. R. E. Batch No.	Wire No.				
Co-70 I	I	8	19.57	10	5.282±0.024
"	II	"	19.45	10	5.320±0.021
Co-71 II	I	2.5	2.74	10	5.218±0.023
"	II	"	2.77	10	5.210±0.022
Co-72 III	I	0.5	0.346	9	5.228±0.031
"	II	"	0.343	9	5.254±0.023
			Over-all Mean Value		5.252±0.042

Table 2 is shown as a matter of interest to amplify what I said previously about the half life of cobalt. This shows our 1956 value, (5.252 ± 0.042) y. The value I gave the other day (5.260 ± 0.034) y includes measurements which we made in January 1957. Column 4 gives the gamma ray equivalent when the measured gamma rays are filtered by the 5 mm thick lead front of the chamber, column 3 the number of measurements. The half-life values and the standard deviations for the individual samples have been derived by the least squares method.

Another part of our work is the measurement of low-activity radium solutions by the radon method (Mr. W. J. Callow and Mr. J. W. G. Dale). Figure 5 shows the 4-liter electrolytic copper ionization chamber connected to the filling system. The flask, fitted with a reflux condenser, contains the radium solution. The standards range from 10^{-9} g to 10^{-11} g total radium content. The radon expelled from the solution passes through KOH in the drying tubes on its way to the chamber. The method of measurement is by alpha particle counting, i.e., slow counting in the air. With this equipment we can measure down to 2×10^{-13} g of radium total in the 4-liter ionization chamber. We have also used exactly the same pattern of chamber with a Lindemann electrometer to measure the integrated ionization in order to compare the alpha-particle counting method with the integrating ionization chamber method. We have obtained good agreement between activity ratios.

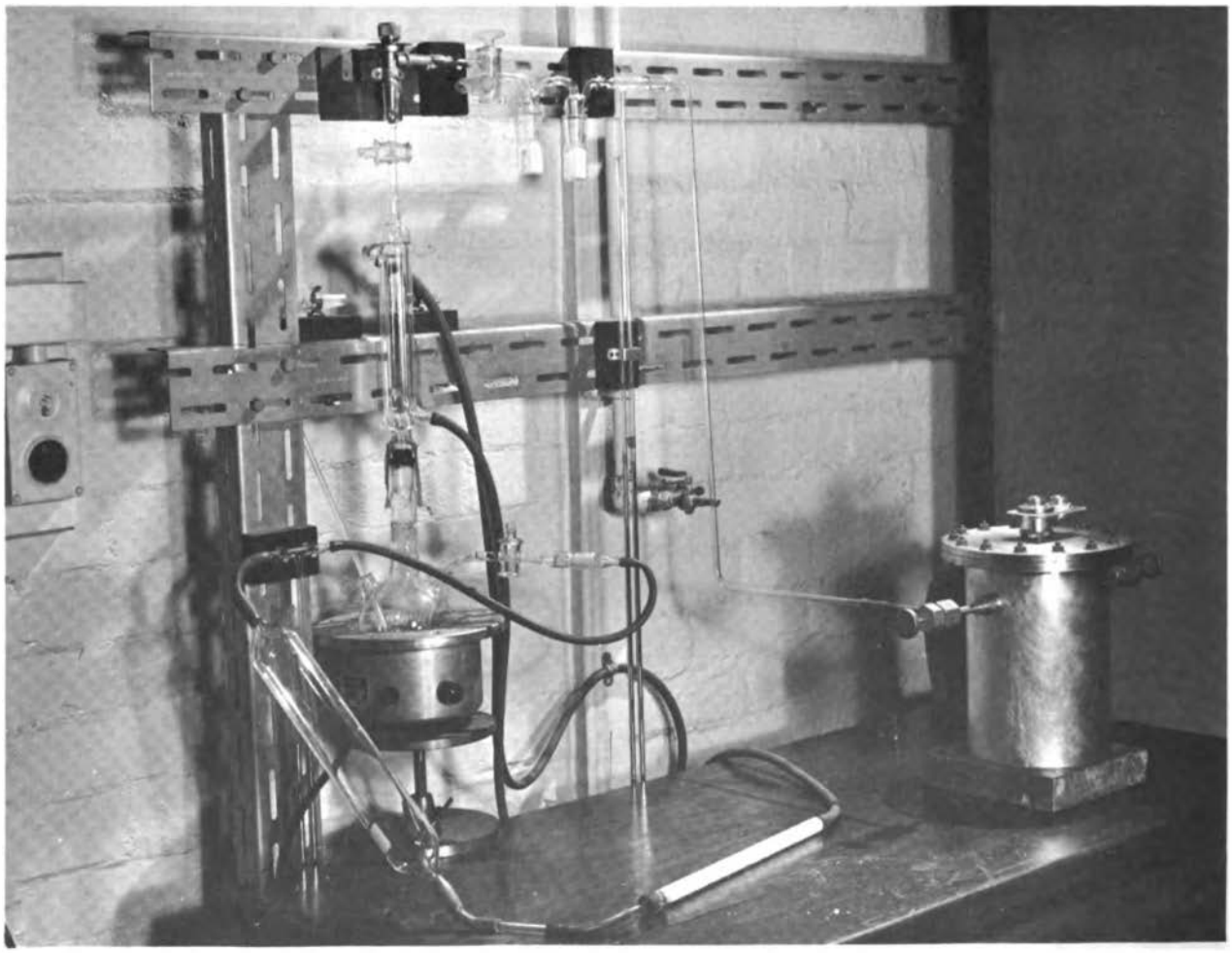


Figure 5. Ionization chamber and radon transfer apparatus used at N. P. L. for measuring low activity radium solution.

I would like now to digress and make a few remarks about the beta-gamma ionization chamber which I discussed earlier. First, I would like to say that although it is referred to as the N. P. L. chamber, it is really a product of the National Physical Laboratory Advisory Committee on Radioactive Standards, and derives essentially from a small subcommittee consisting of Dr. E. W. Emery, Dr. W. K. Sinclair, and myself. The design was modified by Mr. E. W. Pulsford and Mr. F. Wade of the Atomic Energy Research Establishment, Harwell, in collaboration with the manufacturers in order to make it suitable for production. Mr. Dale and Mr. Pulfer of the N. P. L. have done the measurements and have made a number of suggestions leading to improvements in the design.

One aim in producing the chamber that I did not mention earlier was that it might provide users of radioactive isotopes with an instrument having calibration factors which are known to an accuracy of perhaps two or three percent in the case of gamma rays and plus or minus five percent in the case of beta rays. In short, if a user was not able to get standards or was in an isolated situation, he would be able to make use of the calibration figures determined at the N. P. L. or at any other organization, for any particular isotope, for approximate standardization purposes. It was also hoped that in this way the demand for successive issues of short-lived radioactive standards might be reduced. We have yet to see whether this will in fact be one result of the issue of this chamber.

We have been taking part in a series of intercomparisons, not only the regular intercomparisons with the National Bureau of Standards and the Atomic Energy of Canada, Ltd., which have been in progress for a number of years, but with the Max Planck Institute at Göttingen, and the Commissariat a l'Energie Atomique. Some results of these measurements have already been published. [1]

Table 3 shows the results for Na-24 and P-32. The values agree to within ±2 percent. Table 4 shows the results of measurements of N. P. L. I-131 and Au-198 standards. Here again there is agreement to within ±2 percent. The N. P. L. standard of I-131 is maintained by means of the graphite ionization chamber on the basis of a specific gamma ray emission of 2.25 r/mch at 1 cm. We have always done counting measurements with either the 4π Geiger counter or the 4π proportional counter as a check. So far as I remember, the ratio of the ionization chamber value to the counting value does not vary by much more than one percent.

YAFFE: Would you elaborate on the defined solid angle absorption correction of 58 percent?

PERRY: I cannot because I do not know the method of derivation.

VINCENT: There is a paper, I think, by Dr. Grinberg bearing on the defined solid angle.

PERRY: These results were kindly supplied by Dr. Grinberg of the Commissariat a l'Energie Atomique (France) and I do not recollect that in his notes to me he said how he derived the 58 percent. He stated that it was determined for the experimental arrangement he used, and takes into account the absorption in air (5 cm path), in the counter window (1.9 mg/cm^2) and in the gas of the counter.

There has been another program of intercomparisons this year, in which the National Research Laboratory, Pretoria, and the Physikalisch Technische Bundesanstalt, Germany, have joined, but it is not yet completed. We still have the Au-198 intercomparison to do. The results are coming out very similar to those shown already. The order of agreement does not seem to improve from plus or minus two or three percent.

[1] Strahlentherapie 102:370 (1957).

TABLE 3

Measurements of N. P. L. Na-24 and P-32 Standards				
Measured by	Method	Source mounting	Absorption correction	μc/g
	Na-24 (March 1956)			
N. P. L.	4π Ppl	Al foil 200 μg/cm^2	none	2.50
Max Planck Inst. Göttingen (Germany)	4π Ppl	Celluloid foil 15-20 μg/cm^2	none	2.52
	Beta-gamma coin	Celluloid graphite foil 50-80 μc/cm^2	-	2.52
Commissariat a l'Energie Atomique (France)	4π GM	Polystyrene 1.35 mg/cm^2	+0.6%	2.57
	P-32 (June 1955)			
N. P. L.	4π Ppl	Al foil 200 μg/cm^2	none	25.8
Max Planck Inst. Göttingen (Germany)	4π GM	Cellulose acetate 15-20 μg/cm^2	none	25.3
Commissariat a l'Energie Atomique (France)	4π GM	Metallized polystyrene 1.5 mg/cm^2	+0.45%	25.6
	Def. solid angle	Polystyrene 1 mg/cm^2	+8%	25.2

YAFFE: I am a little curious about the source mounting. I was wondering if you were concerned with this one measurement, and I would like to know how you make celluloid at 15 μg/cm^2.

VINCENT: At that time we did it by dipping a glass plate in a solution of cellulose acetate and drying it and cutting the edges with a razor blade and floating it off in water. That is the way we made these foils. Of course, if you have a glass plate which is large enough you can easily weigh the foil and determine how thick it is.

PERRY: As to our future program at N. P. L., we are constructing a high pressure 4π proportional counter for measurement or rather investigation of the measurements of low energy beta particle emitters and possibly electron capture nuclides. We are building a multichannel pulse-height analyzer for use in this connection, and for other applications to standardization.

There is also a project for setting up for the United Kingdom a carbon-14 dating service, which, so far, has not gotten further than the preliminary administrative stage. We are hoping, in this connection, that we may also be able to enter into the field of absolute carbon-14 measurements and so perhaps provide some information on disintegration rate and half-life values. We shall, of course, continue to look into such matters as self-absorption in 4π beta measurements and all the other factors that we meet in this standardization work.

TABLE 4

Measurements of N. P. L. I-131 and Au-198 Standards

Measured by	Method	Source mounting	Absorption correction	μc/g
	I-131 (October 1955)			
N. P. L.	Ion. Chamb.			25.1
	4π Ppl	Al foil 200 μg/cm^2	+2.2%	25.24
Max Planck Inst. Göttingen (Germany)	4π GM	Cellulose nitrate 15 μg/cm^2	none	25.16
Commissariat a l'Energie Atomique (France)	4π GM	Metallized polystyrene 1.55 mg/cm^2	+1.8%	26.06
	beta-gamma coin			
	Def. Solid angle	Polystyrene 1 mm	58%	25.26
	I-131 (April 1956)			
N. P. L.	Ion. chamb.			25.0
	4π Ppl	Al foil 200 μg/cm^2	+2.2%	25.28
Max Planck Inst. Göttingen (Germany)	4π Ppl	Celluloid foil 15 μg/cm^2	none	24.98
	Au-198 (November 1955)			
N. P. L.	4π Ppl	Al foil 200 μg/cm^2	+1.5%	25.6
	Beta-gamma coin	Plastic + graphite		25.5*
Max Planck Inst. Göttingen (Germany)	4π GM	Celluloid 15 μg/cm^2	none	25.89
	Beta-gamma coin	Celluloid + graphite foil		26.0+
Commissariat a l'Energie Atomique (France)	4π GM	Metallized polystyrene 1.5 mg/cm^2	+0.8%	26.5*
"	Beta-gamma coin	Polystyrene 1.5 mg/cm^2		25.42**

* A "Decay scheme" correction of -0.6% was applied to the measured value.

** A correction of -4% was applied to the measured value to allow for internal conversion electrons.

\+ A correction of -2% was applied to the beta counting rate to allow for internal conversion electrons.

I would like in conclusion to refer again to the Advisory Committee that we have, which began as a very informal committee. Through this committee, and in other ways, we maintain very close relationship with the Atomic Energy Research Establishment at Harwell, the Royal Cancer Hospital, the Medical Research Council, the Radiochemical Centre, and various other establishments, so that we do present on the whole quite a good combined effort in this field. Although there is a sense of rivalry and competition, I think we do get along fairly well together, and have made fairly good progress in the past. I hope we shall continue to do so in the future.

MANOV: I am very pleased to see the progress that is being made in the field of standardization, because as recently as three years ago we could have looked at a similar slide and seen agreement to within 3 to 5 percent instead of 1 to 2 percent as we see now. This is one of the indications of the real progress that has been made in this field.

2. National Program - Canada

P. J. Campion

Just as we have heard in the United Kingdom and as we have seen in this conference just recently, there is indeed a certain amount of internal rivalry among the Canadians. However, likewise, there is good interchange among ourselves.

I really do not know quite where to begin so perhaps I should start by describing what we do at Chalk River. This strictly is not the laboratory which is responsible for maintaining standards, but nevertheless we do it. The National Research Council in Ottawa, to which Dr. Geiger belongs is the government laboratory, and he does radium standardization as you learned in the last few days.

At Chalk River we concentrate mainly on beta ray standardization. We have 4π proportional counters which are fairly well known. We have 4π beta-gamma counters. We also have gas counters. The original gas counters were deveoped by Dr. Mann and Dr. Hawkings. We also have a 4π gamma ionization chamber, and recently, as you have heard, we are trying to get into the field of standardizing electron capture nuclides.

The program as it stands at the moment is mainly directed to improving the technique of laying down sources so that we can get better and better efficiencies in our beta counters and try to reduce the self-absorption correction. This technique is being tackled from a different viewpoint by Dr. Yaffe at McGill.

Recently we have also looked into the question of the associated electronics. We have been trying to design a suitable amplifier for a 4π counter and while we seem to have had a little bit of success, it is rather too early in the game to make any further comments than that.

We are rather divorced from commerce up in the backwoods of Ontario, and we do not do any routine standardization for our commercial products division, although from time to time they ask us to calibrate specific samples. We are also asked, more or less on a private basis, to do standardizations for people from universities throughout Canada, and we, of course, take part in any international intercomparison that anybody likes to invite us to.

There is a certain amount of internal standardization in the project, some of which is routine and other which is not routine, and which requires more or less of a minor research program to carry out.

This, I think, covers the general field of the program we have up there.

3. National Program - United States

W. B. Mann

As six of the fourteen papers in the last three days have been from the National Bureau of Standards, I feel there remains little for me to say for the record. I might say that in addition to the papers given here, we have written a circular which is coming out soon. We have reached the situation where we have too many standards chasing too few people. We were very pleased, therefore, when a commercial firm came along and took over our short-lived standards, and relieved us for more research and more standards.

Actually we have come to an arrangement with Atomic Energy of Canada, Ltd., and the National Physical Laboratory whereby we continue to interchange these short-lived standards. The International Commission on Radiological Units and Measurements asked us to compare these standards internationally and so we have arranged to send three of our short-lived standards during the current year to A. E. C. L. and N. P. L., and gold-198 was sent last November to the National Physical Research Laboratory in Pretoria. There was very good agreement among the laboratories to within about one percent. We have now received two N. P. L. standards and we have one more of ours to go out.

We have had interchecks on all five short-lived standards this year, that is, the primary standards. So even though Nuclear-Chicago has started the distribution of short-lived standards and we have retired, we still continue to compare the primary standards so that there may be no falling off of accuracy in this respect.

This should give us more time for research. It is the policy of the National Bureau of Standards to encourage research because we feel that only as we have good research can we have decent standards and can we attract the right kind of personnel to produce the standards. This latter argument does not seem to work always, I find. The extra time we have had as a result of shedding these short-lived distributions seems to have disappeared.

We are very proud and happy to have had two members of the radioactivity section participating in the parity experiments and another one talking about liquid scintillation counting at the meeting of UNESCO in Paris.

We try to devote at least 30 percent of our budget to research as distinct from standardization.

We were recently asked to contribute to the Brussels World's Fair. This may be of interest to you. Figure 1 is a photograph of a so-called nuclear clock which we have provided for this exhibition. We were approached first of all by the United States Brussels Fair authorities to put some carbon-14 standard into the corner stone of the United States Pavillion, but the architect later shaped the pavillion like a doughnut without any corner for a stone. We then suggested we furnish a plastic block with two of our standards in it, so Mr. Vernon, Miss Harmon, Dr. Seliger and I got together and designed this plastic block.

On the left is a tantalum-182 standard decaying with a half life of 115 days, and on the other side there is a radium solution standard. There are stainless steel blocks in the center with holes for Geiger counters. We then hope to plot decay curves and show the passage of time. We will explain in an accompanying panel that in the same way that carbon-14 can be used to measure the passage of the centuries, so may people know how long the Fair has been open by means of the tantalum-182. We hope to have a reasonable exhibit.

I think the only other point I might mention is that in connection with the International Commission of Radiological Units (ICRU) Committee I on Standards and Measurements of Radioactivity for Radiological Use, of which Mr. Perry is the chairman, there has been set up in this country a corresponding subcommittee of the National Committee on Radiation Protection (NCRP)

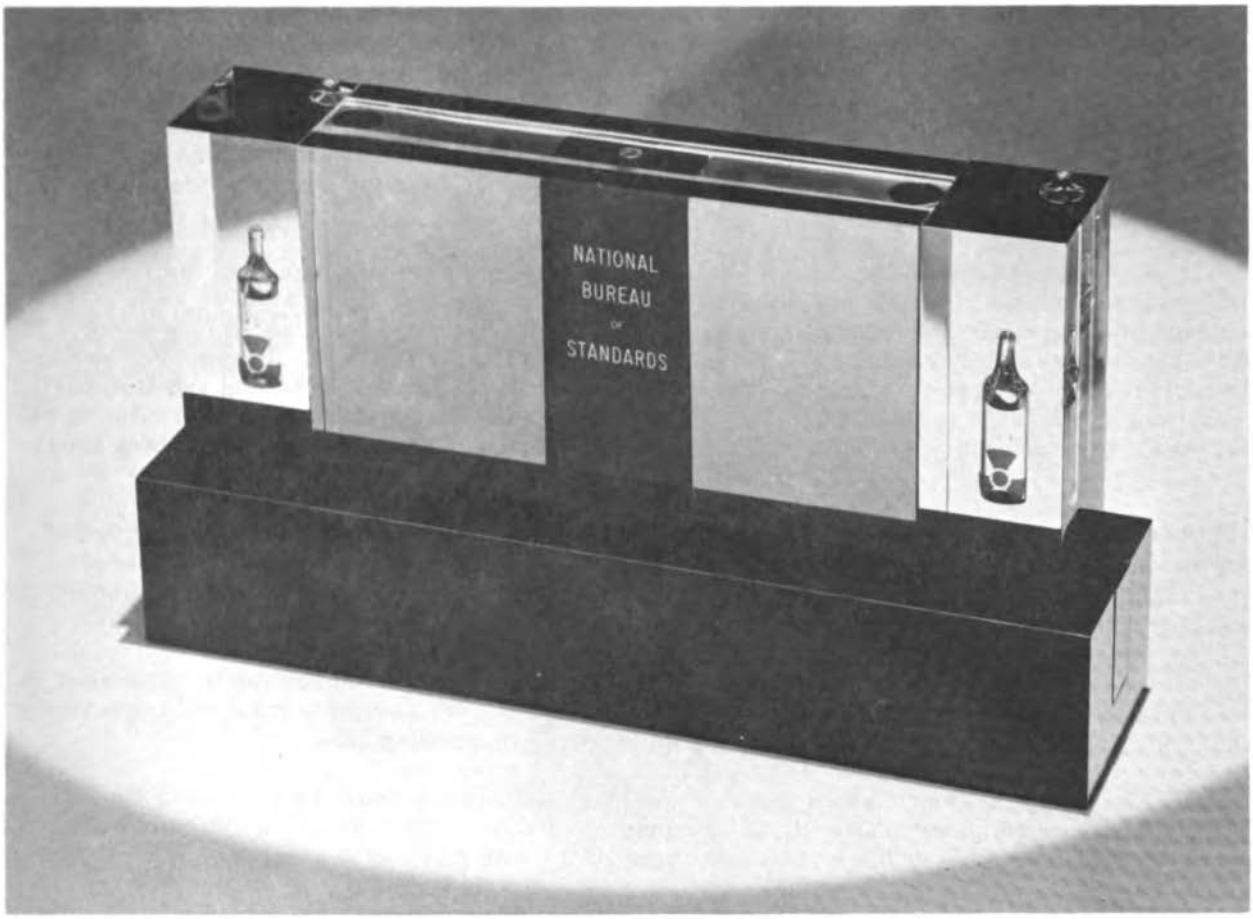


Figure 1. Brussels World's Fair nuclear clock.

with the same name, Subcommittee M-1 on Standards and Measurements of Radioactivity for Radiological Use, with the idea that this subcommittee should feed the views of the North American continent into its two representatives on the ICRU committee. This subcommittee and its task groups are trying to prepare a report right now for the use of the North American representatives of Committee I of ICRU, and include representatives of the standardizing laboratories, Dr. Geiger, Dr. Campion, Dr. Seliger, Dr. Reynolds, Mr. Garfinkel, and representatives from the hospitals, Dr. Quimby, Dr. Sinclair, Dr. Rugh, Dr. Gross, and Dr. Eberhardt. There are medical men and physicists to try to produce some sort of report on the needs of the hospitals in the use of radioactive isotopes.

We are also arranging to check the N. P. L. combined beta-gamma counter and various other simple devices which can be put in the hands of a hospital physicist and so that with little expense and effort they can standardize their preparations to ±5 percent or ±10 percent. This is dangerous ground between ± 5 and 10 percent, because if you say the latter to a medical man he does not often agree with you, so I usually compromise by saying ±8 percent, and really they frequently do not need any greater accuracy.

You also might be interested in a breakdown of our last year's distribution of standards. During the fiscal year 1957 radioactivity standards distributed by the National Bureau of Standards were as follows: alpha ray standards 88; short-lived standards 346; long-lived standards 457; radium ore standards 16.

BAPTISTA: May I ask a specific question on a very particular problem. It is about the strontium correction. Some time ago, I did an experiment with a 4π beta counter. We tried to separate completely the yttrium from the strontium and measure the strontium activity in a certain number of samples. After waiting about 15 days to attain equilibrium between the strontium and the ytterium, and assuming that because of the high energy of the betas of yttrium, they will not be absorbed in the source and in the foil, we tried to evaluate the correction for the strontium betas, resulting from self-absorption in the source, and the absorption in aluminum foil of 100 $\mu g/cm^2$.

In about 16 samples the strontium was almost pure strontium, and then after waiting for equilibrium--you get a surprisingly high value for this correction. If I remember well, it was an average of about 8 percent for the strontium. This means a correction of about four percent for the strontium-yttrium. I could not find a simple explanation for this fact. I remember seeing a report by Dr. Mann published in the International Journal of Applied Radiation and Isotopes containing a reference to the strontium-yttrium source, and you report quite a high value is reported for this correction for the strontium-yttrium source. Can you tell me something about this?

SELIGER: May I say something about that? Dr. Mann may have said in his paper that if one is not careful you can get a fair amount of self-absorption in strontium-yttrium sources, but I do not believe that his paper gave any numbers, because we do not assume a number for any correction in strontium-yttrium.

In a previous paper we gave some data on what can happen to strontium-yttrium sources in a 4π beta counter if the sources are not properly prepared. We showed that as you improve the chemical preparation of the source, you get an increase in counting rate.

MANN: I did say that it was a surprisingly large self-absorption. I was quoting Dr. Seliger's private communication. It was surprising in view of the high-energy yttrium beta particles that there was such a large correction, of the order of 5 or 6 percent.

BAPTISTA: Yes. I have not prepared a source with special precautions. I made a simple preparation in aluminum foils and I get this value. I was rather surprised because for 0.6 Mev betas it is a rather high correction.

YAFFE: We have measured isotopes with a beta energy of 690 kilovolts. You have to remember that in a 4π counter the self-absorption is tremendous. Comparing this data, I do not think the figures you quote are out of line at all.

BAPTISTA: We used aluminum foil which was quite thick. I was surprised because in the case of iodine-131, one can consider that the average beta energy is lower than in the case of the strontium. Certainly I get corrections of about two percent or something like that.

REYNOLDS: I have something that may tend to corroborate this finding. We were looking at cesium-137 a few months back which has a beta energy about the same as strontium-90, I believe, 0.5 Mev. The decay scheme is not well defined. By assuming various values of branching and conversion of the gamma, we were able to estimate the efficiency with which we were beta counting cesium-137. Our results curiously enough indicated that with no particular precautions in the source preparation, by simple evaporation on the film, we were getting between 92 and 94 percent efficiency. This I think correlates very well.

BAPTISTA: Yes.

MANOV: I would like to ask Dr. Mann a little about the circular he mentioned in passing, that is in preparation. Could you tell us a bit more about it and possibly your target date of publication?

MANN: I think it will probably be out in about three or four months' time, and it consists really of a combination, with an extension and bringing up to date, of Howard Seliger's and my articles in the first volume of the Journal of Applied Radiation and Isotopes with an introduction. I had only three weeks to write my article for the International Journal, and so there was not time to prepare graphs or pictures for it. We have brought it up to date by adding descriptions of techniques used at the National Bureau of Standards. The two articles really dealt with standardization on an international basis. We have added at least 40 pictures, diagrams, and so forth, with fairly descriptive captions dealing with Bureau techniques.

MANOV: I am sure we all look forward to seeing this circular, and I hope that it will become available to all the people here either by your sending it to them directly or alternatively sending it to them through the subcommittee. This paper should, I think, find large acceptance in many countries that do not have standardization programs of their own, or particularly primary standardization programs. I want to stress how much I think this will be of help to them.

4. Other Programs

A. H. W. Aten, Jr., A. M. Baptista, P. Kipfer,
O. Kofoed-Hansen, and D. H. Vincent

MANOV: I think we might profitably spend a few minutes hearing from representatives of other countries as to what sort of programs their countries have. I would like to suggest that we ask each man in turn who has not spoken if he would be willing to spend five minutes to tell us what is going on in his country. I would like to call on Dr. Aten.

ATEN: This is a rather easy duty because not very much goes on in the Netherlands. At one time we thought it would be nice to do some primary standardization and my colleague and his students did a number of calibrations mainly by coincidence measurements which turned out quite well in comparison with other countries. But then we figured other people could do it better than we could do it ourselves, and we discontinued it for a moment, although we shall probably come back to it.

In our country it is not so much the lack of people. At present it is the lack of space for people to do this kind of work because one needs a little room not to get messed up with other people's experiments.

We have been doing quite a bit of work on secondary standardization, chiefly on the reproduction of measurements. This type of secondary standardization is parallel to the British work on the standard type ionization chamber. In our case, however, we have been trying to do the same thing with Geiger counters, due mainly to the fact that in Holland an excellent type of Geiger counter is being produced that remains practically unchanged for quite a number of years. It is a counter which works at a very low voltage, a few hundred volts, and usually has a very long plateau. So we thought this might be useful for standard values. We mount them in racks in order to make certain that they are not influenced by the surroundings. This type of counter we use for beta and gamma counting.

We have a well type counter which is quite similar to the English type of ionization chamber. We check its stability by using different kinds of long-lived isotopes to safeguard against the influence of chemical impurities. Still, one of the main difficulties in our case is probably the radiochemical purity of these nuclides. We realize that it might be more satisfactory to use radium standards than the strontium-90, cesium-137, and the cobalt-60 standards we use at present, although I personally do not know how pure radium is in the condition one buys it, and whether one can obtain radium which is guaranteed free from short-lived components.

We have a few things that especially interest us. Thulium-170 is used as a secondary standard for gold because the beta energy is practically the same. You can see that very nicely from absorption curves.

We are presently looking at the possibility of using terbium-160 as a secondary standard for iodine-131, but I cannot give any data at present because the experiments are still being run.

A third thing we would like to do is the standardization of sodium-24 by comparison to natural potassium where one uses thick samples and has to compensate for scattering and absorption. We would do this by comparing a sample of sodium chlorate to a sample of potassium nitrate, and if you get the exact values and the exact square values, you come to very nearly the same figures for these two comparisons. So the comparison ought to be quite reasonable and also the beta absorptions quite similar. One has to correct, in this case, for the counting of the gamma rays in the sodium-24, but if one does not make the samples too thick, one still gets quite a reasonable comparison rate with normal potassium, and the correction for the gamma influence from the sodium-24 is not so large that it becomes an appreciable factor in the correction for the beta activity. Still it is not reasonable to hope for an accuracy in such an experiment better than maybe something like four percent. This is what interests us at present. This

is mainly a thing we do as an exercise of our own, although it might have some practical use.

MANOV: Thank you, Dr. Aten. Let me now call on Dr. Baptista.

BAPTISTA: Our program in Portugal is still a rather small program. In the beginning, you start studying these problems in the medical and biological applications for radioisotopes, because we have some special problems with phosphorous, iodine, and gold for medical and biological applications. Our work has been mostly in the field of 4π beta counting. We have built quite a number of these counters. They are only variations of the same theme. You can even build an external cathode 4π counter just for the fun of the thing, and it works.

At first we built glass 4π counters because we thought at the moment they would be cheaper, but after one week the technicians dropped the counter. You then realize that it is really cheaper to go back to the metallic counters. We have done absolute measurements on phosphorous, iodine, gold, strontium-yttrium, and other ones only through curiosity.

Related to this work we have made quite a number of secondary counting systems. We have quite a nice well counter that measures the betas of phosphorous-32 by measuring the bremsstrahlung in aluminum. We have ionization chambers of the type described here by Dr. Perry to measure gamma emitters with relatively high activity in the millicurie region. We have some special arrangements of Geiger-Muller counters to standardize the beta emitters. As a matter of fact, to compare cobalt sources, we have been using a method similar to the one described here by Dr. Putman, using the small graphite chambers that really are quite convenient and accurate.

We are also doing some work on radium. We have considered as our standard a very old radium preparation that the Cancer Institute in Lisbon got from, I think, America. Quite recently we received half a gram of radium from the Belgian company. We checked by a simple electroscope measurement the quantity received with this standard and the agreement was surprisingly good--less than one percent.

In addition we have been doing some work in 4π scintillation counting. Using liquid scintillation methods we found that for high energy beta emitters the agreement with the measurements by the Geiger-Muller method is quite good. We are struggling now with the problem of the low energy beta emitters. I am afraid I cannot give you many results because we have just started working with this method by using sodium iodide crystals on a photomultiplier. Inside the well you have a 4π beta arrangement formed with plastic scintillators. Of course, the other scintillator is viewed by another photomultiplier. We feel that the system has certain advantages if you look into the theoretical aspects of the problem that you heard Dr. Campion speak about. You have increased efficiency for the gamma radiation. There is no problem in ideal circumstances about the gamma sensitivity of the beta counter. No problems also for the internal conversion of the gamma radiation. So we are hopeful.

We have not yet published any results because by studying this simple plastic 4π arrangement, we have found out that for the very low beta energies, looking with only one photomultiplier, the extrapolation to zero discrimination was quite difficult. We hope to solve this problem in the future.

I think that this covers almost all the work we have been doing. There is really no official organization responsible for the standardization of radionuclides. We provide samples and everything, and we guarantee so far as we can the activity we give to the people that are working in some research problems, and who want to know the activity they are playing with.

MANOV: Thank you, Dr. Baptista. I would now like to call on Dr. Kipfer.

KIPFER: I could say the same thing as Dr. Baptista, "There is no official organization in Belgium for standardization." The Center of Research for Nuclear Energy, known as CERN, is

doing some standardization work but only for their own purposes, and for perhaps medical people who wish to have their radioisotopes measured. It is quite natural that Belgium is still interested in standardization of radium standards because a lot of countries are still using such standards, and there are still some demands for such standards.

The third field in which we are working is that of very low activity in connection with the work of Dr. Pejustos. He is interested in determining the age of deep sea and other geological samples. This work has been done until now by nuclear emulsion techniques combined with mass spectrographic analysis. Because of the interest the half life of radium has for this work, we think perhaps we will begin to make new a determination of this value because we do not believe it is the last value of 19.5 which has been advanced recently. Personally I am interested in a determination of the half life of radon and I think with the device I described, it would be very easy to obtain very good precision.

I think that is all that I can say on Belgium.

MANOV: Thank you, Dr. Kipfer. I would now like to call on Dr. Kofoed-Hansen of Denmark.

KOFOED-HANSEN: I am very sorry I cannot describe a large program because we, just as you have heard from the previous two speakers, have no official committee for that sort of thing. Standardization work has only been done where it was felt necessary for some other experiment in Denmark. This has been done especially with short-lived radioactivities like sodium-24 such as when Dr. Hassen wanted to measure a cross section for the disintegration of deuterium by sodium-24 gamma rays. Nowadays there are several laboratories in Denmark that are interested in low level counting and standardization from different points of view, mainly, for medical uses and secondary standardization.

There is a laboratory in which Dr. Hilda Levi works with low level activity, especially for the Danish archeologists who like to know how old the Vikings were. In addition, there is a laboratory run by Ambrosen. They are very much interested in fall-out measurements of all sorts. In our laboratory at the Atomic Energy we plan quite an extended program for standardization since we want to produce radioisotopes and are concerned with the problem of neutron flux measurements. We are beginning to gather instruments for that purpose, but I would rather wait until we have something more satisfying before I discuss it.

MANOV: Thank you, Dr. Kofoed-Hansen. I would like to call on Dr. Vincent to tell us a little bit about the situation in Germany.

VINCENT: I am not really entitled to talk about a national program because the Krupa authority in Germany is an organization which is not represented here. In addition to the radium standardization which they have been doing for quite a number of years, they have recently taken up isotope calibration. That was about two years ago. They are making good progress but I should like to concentrate on what our laboratory is doing.

We started 4π counting with the 4π Geiger counter in about 1951-52. You saw some of the results of the intercomparisons in Mr. Perry's tables in session III. We have now changed from the Geiger counter to a proportional flow counter which we are now using for 4π beta counting.

We are also doing beta-gamma coincidence measurements, and we have developed our own type of counter for that. We found it very difficult to find the corrections for the gamma contribution in the beta counter when beta-gamma coincidence counting, so we developed a counter which has a negligible gamma efficiency. It consists essentially of a brass tube which is about 10 cm in diameter and 20 cm in length. Down below is a source mount and around this there is a very small counter which consists of a wall of wire. The diameter of the wire is 0.2 mm. The center wire of the counter is 0.05 mm in diameter. This is with a number of wires of which the wall is made up, ten in number, and the center wire is again shielded so that the

actual counting volume for the betas here is about two to five cubic centimeters. Gamma rays which are emitted from the source, and which will produce secondary electrons here will be counted with a very low efficiency because of the very small counting volume of the beta counter.

On the other hand, since the beta source is mounted in such a manner as to form one of the walls of the beta counter--the efficiency for the betas is very high, approximately 30 to 40 percent. So we think that we will have very, very few gamma counts in the beta counter. We have a sodium iodide crystal on the other side for the gamma count.

We are also doing internal gas counting of C-14. We have a number of counters with different lengths and diameters, but so far we have not actually calibrated them. We have worried about end effect and wall effects. I think a paper will be published on the results of this research in the near future.

Finally, we are also trying to calibrate electron capture isotopes. We have a spherical counter for that. We did not hope to obtain the linear extrapolation which Dr. Allen got in his counter, so we thought it might be best to have a spherical counter, where the half lengths for the X-rays emitted from the source would be equal all over the solid angle. The construction consists of the two half spheres with a brass plate in the middle where the source is mounted, and two wire loops. If you want to do K X-ray gamma coincidence counting, you just dismount one of the half spheres and replace it by a brass plate, and then you can put your sodium iodide crystal very near the source. I found out recently that this gives about 10 percent efficiency for the manganese-54 gamma rays if you only consider the total energy peak. This gives a fairly high counting rate. The counting time for these K X-ray gamma coincidence measurements is fairly low.

We have our own program of distributing calibrated sources of irradiated material, mainly phosphorus-32 and iodine-131 and gold-198, to those of our customers who wish to have calibrated sources. We have started the program this year and in March we distributed 11 phosphorous-32 sources, and in April about 15 iodine-131 sources, and then in May, 4 gold-198 sources. These go mainly to medical people, to hospitals and the like.

MANOV: Thank you, Dr. Vincent. Are there any general questions with respect to the national programs?

YAFFE: I do not happen to represent Russia, but I am wondering whether anyone has seen the Russian 4π counter, and whether you would be interested in it? I have seen one and I can describe it in about 30 seconds.

This counter which is a 4π Geiger counter has a wire in the vertical direction going through a block of Teflon, which is the biggest piece of Teflon on any small instrument. Incidentally, Teflon seems to be an international commodity. The source is mounted on a thin film and the wire passes through the film at right angles to the plane of the film. They merely burn a hole in the center of the film with a microburner and spread the source as close to a hole as they possibly can. The wire then goes down through this. This has obvious advantages. You can get very good field distribution. With all due respect to those present, it is one of the neatest 4π counters I have ever seen. They use this, incidentally, on a routine basis for people who are interested in agricultural research. They standardize isotopes, and then send them out to those people. It is a beautifully engineered piece of equipment. I bring this out merely because I have never seen any of their absolute counting work described in the literature, although some people may have.

MANN: As I remember the counter, it was for carbon-14. I felt it was only a 2π counter since the source holder was such that the other side would not see the betas from carbon-14. I forget the details but they took it apart and showed it to me. The instrument is shown in Figure 1. I asked him how they used it with carbon-14 and I got no answer to that.

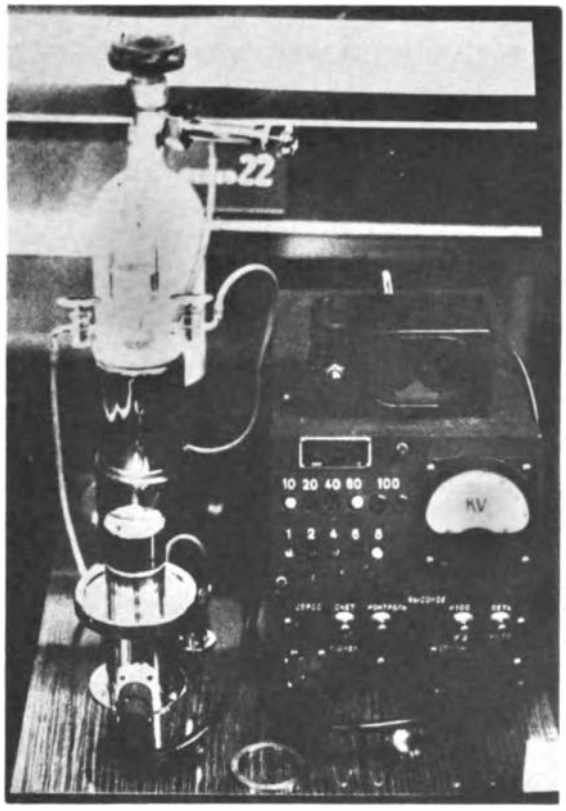


Figure 1. USSR 4π Geiger Counter.

YAFFE: No, I think this must be a different one. I asked them what isotopes they had standardized and he wrote down carbon-14, phosphorous-32, iodine-131, a fairly thin film. The film is about half-way up in the counter.

MANN: This is the same.

BAPTISTA: I saw a recent Russian paper on the 4π counter which is the same as the French.

SELIGER: There is an English translation of that at Brookhaven which is three months old. They do not claim any better than plus or minus two or three percent.

INDEX OF ISOTOPES

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