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NUCLEAR SCIENCE SERIES
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RADIOCHEMISTRY OF CARBON, NITROGEN AND OXYGEN

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FOREWORD

The Committee on Nuclear and Radiochemistry is one of a number of committees working under the Board on Chemical Sciences and Technology of the Commission on Physical Sciences, Mathematics, and Resources of the National Academy of Sciences--National Research Council. Its members are drawn from academic, industrial, and government laboratories and represent the areas of nuclear chemistry, radiochemistry, and nuclear medicine.

The Committee has concerned itself with those areas of nuclear science which involve the chemist, such as the collection and distribution of radiochemical procedures, specialized techniques and instrumentation, the place of nuclear and radiochemistry in college and university programs, the training of nuclear and radiochemists, radiochemistry in environmental science, and radionuclides in nuclear medicine. A major interest of the Committee is the publication of the Nuclear Science Series of monographs on Radiochemistry and on Radiochemical Techniques. In 1982 a third series on Nuclear Medicine was initiated.

The Committee has endeavored to present monographs that will be of maximum use to the working scientist. Each monograph presents pertinent information required for radiochemical work with an individual element or with a specialized technique or with the use of radionuclides in nuclear medicine.

Experts on the various subjects have been recruited to write the monographs. The U.S. Department of Energy sponsors the printing of the series.

The present monograph consists of two reviews. The first section deals with radiopharmaceutical and biomedical applications. The second section deals with analysis of carbon, nitrogen and oxygen in different materials by use of nuclear techniques. This monograph is published as part of our continuing effort to update, revise, and expand the previously published monographs to keep them current and relevant.

Edward S. Macias, *Chairman*
Committee on Nuclear and Radiochemistry

PREFACE

This report on the radiochemistry of carbon, nitrogen and oxygen has been prepared as one of a series of monographs on the radiochemistry of the elements under the sponsorship of the Committee on Nuclear and Radiochemistry within the National Research Council.

This report consists of two reviews. The first section deals with radiopharmaceutical and biomedical applications. This section covers the production and the preparation of the important precursors of carbon-11, nitrogen-13, oxygen-14 and oxygen-15. The production yields for the important nuclear reactions are tabulated. The second section deals with analysis of carbon, nitrogen, and oxygen in different materials by use of nuclear techniques. A survey of the radiochemical methods (Table IX) for analysis of carbon, nitrogen, and oxygen in various matrices will be of interest to the scientist working in the field. A collection of radiochemical procedures for carbon, nitrogen, and oxygen as found in the literature is also included.

It is hoped that the bibliography is sufficiently extensive to serve the needs of the radiochemist. However, it is to be expected that important references may have been unintentionally omitted. The authors would appreciate being made aware of such omissions so that the references may be included in a future update of this monograph.

The authors appreciate the assistance of Miss Mabeth P. Martinez for typing the monograph.

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I. GENERAL REFERENCES ON THE RADIOCHEMISTRY OF CARBON,
NITROGEN AND OXYGEN

A. Radiopharmaceutical Chemistry and Biomedical Applications

The Chemistry of Positron Emitting Nucleogenic (Hot) Atoms with Regards to Preparation of Labeled Compounds of Practical Utility." R.A. Ferrieri and A.P. Wolf. *Radiochimica Acta.* 34, 69 (1983).

"The Synthesis of Carbon-11, Fluorine-18, and Nitrogen-13 labeled radiotracers for biomedical applications." J.S. Fowler and A.P. Wolf. *Nuclear Science Series. NAS-NS-3201* (1982) U.S. Department of Energy. 124 pp.

"Nuclear Data Relevant to Cyclotron Produced Short-Lived Medical Radioisotopes." S.M. Qaim. *Radiochimica. Acta.* 30, 147 (1982).

"Cyclotrons for Biomedical Radioisotope Production." A.P. Wolf and W.B. Jones. *Radiochimica. Acta.* 34, 1 (1983).

"In-Beam Production of Labeled Compounds." G.A. Brinkman. *Int. J. App. Radiat. Isot.* 33, 525 (1982).

"Special Characteristics and Potential for Radiopharmaceuticals for Positron Emission Tomography." A.P. Wolf. *Seminars in Nuclear Medicine Vol. XI*, 2 (1981).

"Some Aspects of Radioanalytical Quality Control of Cyclotron-Produced Short-Lived Radiopharmaceuticals." G.J. Meyer. *Radiochimica Acta.* 30, 175 (1982).

Synthesis with Isotopically Labeled Carbon. Methyl Iodide, Formaldehyde and Cyanide." R.D. Finn, T.E. Boothe, M.M. Vora, J.C. Hildner, A.M. Emran and P.J. Kothari. *Int. J. App. Radiat. Isot.* 35, 323 (1984).

"Accelerator production of medically useful radionuclides," D.J. Silvester in Radiopharmaceuticals and Labeled Compounds. IAEA, Vienna, Vol. 1, 197 (1973).

"The chemical form of N-13 produced in various nuclear reactions and chemical environment." R.S. Tilbury in Short-Lived Radionuclides in Chemistry and Biology. (J. Root and K. Krohn, Eds). *A.C.S. Advances in Chemistry Series Monograph*, American Chemical Society, Washington, D.C. 1981.

"Preparation of short-lived radioactive gases for medical studies," M.J. Welch and M.M. Ter-Pogossian. *Radiat. Res.* 36, 580 (1968).

"Production and use of N-13 gases and aerosols," N.J. Parks in Applications of Nuclear and Radiochemistry. (R.M. Lambrecht and N.A. Morcos, Eds). Pergamon Press, New York. p. 15 (1982).

Radioactivity and its measurements. W.B. Mann, R.L. Ayres and S.B. Garfinkel, 2nd Ed. Pergamon Press, Oxford. (1980). 282 pp.

"Brain oxygen utilization measured with oxygen-15 radiotracers and positron emission tomography." M.A. Mintun, M.E. Raichle, W.R.W. Martin and P. Herscovitch. J. Nucl. Med. 25, 177 (1984).

B. Radiochemistry in Materials Science Applications

There is no general review published. We list here reviews published for different Activation Analysis Methods.

"Neutron-Capture Prompt γ -ray Activation Analysis for Multi-element Determination in Complex Samples." M.P. Failey, D.L. Anderson, W.H. Zoller, G.E. Gordon and R.M. Lindstrom. Anal. Chem. 51, 2209 (1979).

"In vivo neutron activation analysis : state of the art and future prospects." S.H. Cohn. Med. Phys. 8, 145 (1981).

"The present state of in vivo neutron activation analysis in clinical diagnosis and therapy." S.H. Cohn. Atomic Energy Review, Vol. 18. IAEA, Vienna, p. 183 (1980).

"Determination of H, C, N, O content of bulk materials from neutron-attenuation measurements." J.C. Overley. Int. J. App. Radiat. Isot. 36, 185 (1985).

II. Table of Isotopes of Carbon, Nitrogen and Oxygen

<u>Isotope</u>	<u>Abundance or Half Life</u>	<u>Decay Modes</u>
^9C	0.1265 s	β^+ , p2 α
^{10}C	19.2 s	β^+
^{11}C	20.38 m	β^+ 99.76%, EC 0.24%, no γ
^{12}C	98.89%	
^{13}C	1.11%	
^{14}C	5730 y	β^- , no γ
^{15}C	2.449 s	β^-
^{16}C	0.75 s	β^- , n > 98.8%
^{12}N	11.0 ms	β^+
^{13}N	9.96 m	β^+ , no γ
^{14}N	99.63%	
^{15}N	0.366%	
^{16}N	7.13 s	β^-
^{17}N	4.17 s	β^-
^{18}N	0.63 s	β^-
^{13}O	8.9 ms	β^+
^{14}O	70.60 s	β^+
^{15}O	122 s	β^+ 99.89%, EC 0.11% no γ
^{16}O	99.76%	
^{17}O	0.038%	
^{18}O	0.204%	
^{19}O	26.9 s	β^-
^{20}O	13.5 s	β^-

III. INTEREST AND USE

Hudis (1a) prepared the first review of the radiochemistry of carbon, nitrogen, and oxygen nearly 28 years ago. The interest in the radiochemistry of the three elements lay relatively dormant until the past 10 years. The justification for an updated review is evident now due to the intense demand for radiochemists: (1) for the production and application of the short-lived positron emitters (Carbon-11, Nitrogen-13 and Oxygen-15) in radiopharmaceutical chemistry and biomedical research; and (2) for accurate analysis of trace levels of carbon, nitrogen and oxygen impurities in semi-conductors utilized for microchips, solar energy conversion devices, and various refractory methods (eg: nickel) utilized in the nuclear power reactor industry. The radiochemical methodology utilizing carbon-14 as a label of organic and biochemical substances, as well as in archeology for radioisotopic dating of objects of historical interest are established techniques and are not included in the present review.

Stable and isotopic tracers are utilized very widely in different areas of research. New applications are not uncommon, such as use of carbon-14 to estimate the microbial biomass in lake sediment (27) and the fate of crude oil spills (28). Mention is made, however, to the new field of Accelerator Mass Spectrometry (AMS) which has overlapped into the radiochemistry of carbon, particularly for determining carbon isotopic ratios ($^{14}\text{C}/^{12}\text{C}$ and $^{14}\text{C}/^{13}\text{C}$) and its extension

to a routine radiocarbon dating tool (29, 30). Litherland (31) has cited 28 facilities which routinely perform AMS measurements. The AMS method consists of counting of single radioactive isotopes that have been ionized, accelerated to high energy, and mass analyzed to select and identify individual atoms. This type of measurement permits smaller samples for determination of older ages than is possible with standard radiochemical methods. Radioisotopic dating with AMS was a topic of an international symposium (32) and of reviews (33-35) to which the reader is referred for details of sample preparation, ion services and problems of accelerator physics that are outside the expertise of the radiochemical specialist.

A complete set of radiochemical separation procedures for carbon, nitrogen and oxygen is not practical as the variations between the different investigator's techniques are not usually drastic. However, we have chosen eight (8) representative examples in a variety of matrices. The table IX and the bibliography contain specialized additional citations. As a general practice, citations in less common language are excluded, as are many references to laboratory reports and conference proceedings that are difficult to obtain. If a citation in the the bibliography was published in a language other than English, this is indicated at the end of the citation.

The use of SI (Systeme International) units as the standard for scientific communication has been agreed by the

international scientific community. However, the traditional units continue to be the more commonly used in the day-to-day practice. Therefore, the data has been reported in the units used by the authors. The unit of activity in SI units is bequerel (Bq). One bequerel is equal to one disintegration per second. The conversion factor i.e. $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$ can be used to convert the traditional units of curie to bequerel.

IV. CHEMICAL AND BIOMEDICAL SCIENCE APPLICATIONS

In this section, we will only focus on the aspect of chemistry, i.e. the production of nuclides. The synthesis of labeled compounds with these isotopes have been previously reviewed in detail (1b). Table 1 lists the different methods of production of the commonly used medical radioisotopes [Carbon-11, Nitrogen-13, Oxygen-14, and Oxygen-15].

The selection of the production method is dependent on the type of cyclotron available. Nuclear reaction cross section data are needed for the production of radioisotopes. The data are important for:

- (i) determining the optimum energy range for the production of a specific radioisotope,
- (ii) calculating the expected thick target yield of the isotope under production conditions.
- (iii) calculating the yields of radionuclidic impurities for a given thickness and enrichment of the target material.

The selection of the projectile energy range that will maximize the yield of the desired product and that will minimize the yields of radioactive impurities is of great importance in optimizing a production method.

Table I
Methods of Production for Medically Useful Isotopes

<u>Reaction</u>	<u>% Natural Abundance</u>	<u>Threshold (MeV)</u>	<u>Reference</u>
$^{10}\text{B}(\text{d}, \text{n})^{11}\text{C}$	19.90	0	19
$^{11}\text{B}(\text{p}, \text{n})^{11}\text{C}$	80.10	3.02	19, 20
$^{14}\text{N}(\text{p}, \alpha)^{11}\text{C}$	99.634	3.13	3, 4, 5
$^{12}\text{C}(^3\text{He}, ^4\text{He})^{11}\text{C}$	98.90	0	19
$^{12}\text{C}(\text{p}, \text{pn})^{11}\text{C}$	98.90	21.8	10
$^{13}\text{C}(\text{p}, \text{n})^{13}\text{N}$	1.10	3.23	20, 21
$^{12}\text{C}(\text{d}, \text{n})^{13}\text{N}$	98.9	0.35	21, 19
$^{16}\text{O}(\text{p}, \alpha)^{13}\text{N}$	99.762	5.52	10, 11
$^{14}\text{N}(\text{n}, 2\text{n})^{13}\text{N}$	99.634	-	23
$^{14}\text{N}(\text{d}, \text{n})^{15}\text{O}$	99.634	0	7, 9
$^{16}\text{O}(\text{p}, \text{pn})^{15}\text{O}$	99.762	16.68	9
$^{15}\text{N}(\text{p}, \text{n})^{15}\text{O}$	0.366	3.78	8
$^{16}\text{O}(\text{n}, 2\text{n})^{15}\text{O}$	99.762	-	24
$^{16}\text{O}(\gamma, \text{n})^{15}\text{O}$	99.76	-	25
$^{14}\text{N}(\text{p}, \text{n})^{14}\text{O}$	99.634	6.32	26, 6
$^{12}\text{C}(^3\text{He}, \text{n})^{14}\text{O}$	99.90	1.375	6
$^{12}\text{C}(\alpha, 2\text{n})^{14}\text{O}$	99.90	28.93	6

The thick target yield for a particular reaction can be calculated by an integration of the measured excitation function over the energy range covered by the target, using the relation,

$$Y = \frac{N \cdot H}{M} I (1 - e^{-\lambda t}) \int_{E_1}^{E_2} \left(\frac{dE}{d(\rho x)} \right)^{-1} \sigma(E) dE \quad (\text{eq. 1})$$

where N is the Avogadro Number, H is the isotopic enrichment of the target nuclide, M is the mass number of the target element, I the projectile current, $(dE)/(d\rho x)$, stopping power, $\sigma(E)$ is the cross section at energy E, λ is the decay constant of the product and, t is the duration of irradiation. The subject of cross section and stopping power has been described in detail elsewhere (2).

The calculated yield value represents the maximum yield which can be expected for a given target system. In practice, however, the yields are much lower than the theoretical values, mainly due to inhomogeneity in the incident beam, loss of the product due to the beam heating effect (or density reduction), etc.

Excitation functions for Carbon-11 (3-5) via $^{14}\text{N}(p, \alpha)^{11}\text{C}$; Oxygen-14 (6) via $^{14}\text{N}(p, n)^{14}\text{O}$; Oxygen-15 (7,9) via $^{14}\text{N}(d, n)^{15}\text{O}$, $^{15}\text{N}(p, n)^{15}\text{O}$ and $^{16}\text{O}(p, pn)^{15}\text{O}$; and Nitrogen-13 via the $^{16}\text{O}(p, \alpha)^{13}\text{N}$ nuclear reactions have been published. These references give calculated saturation yields as a

function of a particle energy on the target. Data from these papers are listed in Tables II - VIII. Yields are reported for 20 μA and 50 μA beam current. A beam current of 20 μA is usually used in medical radioisotope production runs, but the new generation of cyclotrons can deliver external beam currents up to 50 μA . Therefore, the calculated yields are reported for 20 μA and 50 μA currents. The yields listed in Tables II - VIII are the theoretical yields, and are not routinely achieved at high beam currents with the available technology.

The reduction in gas density during bombardment in gas target is mainly responsible for reduced yields. This problem has been addressed in the literature (12-15). The cross sections are measured using gas targets at 1 μA beam current or less. However, yields cannot be estimated based on thin or thick target yield at 1 μA by multiplying by beam current. These theoretical yields can only be used as a guideline. All gas and liquid targets require thin metal foil windows to separate the target from the cyclotron vacuum. Therefore, the maximum energy of any machine exceeds that which is available on the target. The thick target yield is calculated using the cross section and stopping power data. Nitrogen-13 and fluorine-18 can be produced by the proton bombardment of oxygen, gas and water. While calculating the thick target yield, one must use appropriate stopping power values such as tabulated by Williamson et.al. (16) and by Northcliffe et.al. (17).

Table II
 Thick Target Carbon-11 Yields

$^{14}\text{N}_2$ Target	$^{14}\text{N}(p,\alpha)^{11}\text{C}$ $t_{1/2} = 20.4 \text{ min}$	Threshold	3.13 MeV	
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 20 μA 20 min (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
17	184	3680	1840	9200
16	172	3440	1720	8600
15	154	3080	1540	7700
14	135	2700	1350	6750
13	115	2300	1150	5750
12	94	1880	940	4700
11	77	1540	720	3850
10	62	1240	620	3100
9	49	980	490	2450
8	38	760	380	1900
7	22	440	220	1100
6	5	100	50	250

Table III
 Thick Target Nitrogen - 13 Yields

$^{16}\text{O}_2$ - Target	$^{16}\text{O}(p,\alpha)^{13}\text{N}$ $t_{1/2} = 9.96 \text{ min}$	Threshold	5.52 MeV
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
15	37	736	1850
14	31	616	1550
13	26	520	1300
12	21	420	1050
11	15.0	300	750
10	8.0	160	400
9	6.5	130	325
8	2.6	52	130

Table IV
 Thick Target Nitrogen - 13 Yields

$^{14}\text{N}_2$ Target	$^{14}\text{N}(p,pn)^{13}\text{N}$ $t_{1/2} = 9.96 \text{ min}$	Threshold	11.4 MeV
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
26	116	2320	5800
25	103	2060	5150
24	91	1820	4550
23	79	1580	3950
22	67	1340	3350
21	56	1120	2800
20	45	900	2250
19	35	700	1750
18	26	520	1300
17	18	360	900
16	11	220	550

Table V
 Thick Target Oxygen-14 Yields

$^{14}\text{N}_2$ - Target	$^{14}\text{N}(p,n)^{14}\text{O}$ $t_{1/2} = 70.6 \text{ s}$	Threshold	6.3 MeV
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
14	8.4	168	420
13	6.9	138	345
12	5.5	110	275
11	4.4	88	220
10	3.3	66	165
9	2.3	46	115
8	1.3	26	65
7	0.3	6	15

Table VI
 Thick Target Oxygen-15 Yields

$^{15}\text{N}_2$ - Target	$^{15}\text{N}(p,n) ^{15}\text{O}$ $t_{1/2} = 2.03\text{m}$	Threshold	3.78 MeV
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
17	172	3440	8600
16	156	3120	7800
15	140	2800	7000
14	123	2460	6150
13	105	2100	5250
12	86	1720	4300
11	70	1400	3500
10	60	1200	3000
9	52	1040	2600
8	43	860	2150
7	29	580	1450
6	13	260	650

Table VII
 Thick Target Oxygen - 15 Yield

$^{14}\text{N}_2$ - Target	$^{14}\text{N}(d,n)$ 150 $t_{1/2} = 2.03\text{m}$	Threshold	0
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
12	111	2220	5550
11	100	2001	5000
10	89	1780	4450
9	77	1540	3850
8	65	1300	3250
7	53	1060	2650
6	40	800	2000

Table VIII
 Thick Target Oxygen - 15 Yields

$^{16}\text{O}_2$ - Target	$^{16}\text{O}(p,pn)^{15}\text{O}$ $t_{1/2} = 2.03\text{m}$	Threshold	16.7 MeV
<u>Energy on Target MeV</u>	<u>Saturation Yield mCi/μA</u>	<u>Yield at 20 μA (mCi)</u>	<u>Yield at 50 μA (mCi)</u>
32	271	5420	13550
30	208	4160	10400
28	150	3000	7500
26	100	2000	5000
24	56	1120	2800
22	31	620	1550
20	15	300	750
18	5	100	250

V. PREPARATION OF PRECURSORS

This subject has been reviewed in detail (18,36). These are important intermediates which are required for the introduction of radioactive species into complex organic molecules.

V-1. CARBON-11

Carbon-11 labelled precursors such as carbon monoxide, carbon dioxide, cyanide (as HCN and NaCN), acetylene and methyl iodide have been prepared.

The most useful reactions for ^{11}C production are the proton and deuteron reactions on natural and enriched boron oxide targets (19,20) and proton reactions on nitrogen (3,4,5).

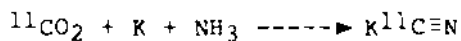
a) ^{11}CO and $^{11}\text{CO}_2$ preparation: [^{11}C]-Carbon monoxide and [^{11}C]-Carbon dioxide can be prepared by bombarding Boron-11 or enriched Boron-10 with protons and deuterons respectively. The production, storage and the targetry involved in this method has been reported in the literature (37-41). The chemical form of the product nuclei recovered from the target is largely determined by the target sweep gas composition. [^{11}C]-Carbon monoxide is prepared in high activity by reducing $^{11}\text{CO}_2$, by passing the $^{11}\text{CO}_2$ over Zn held at 400°C (39,42). [^{11}C]-Carbon dioxide can also be produced by irradiating nitrogen gas with protons (43-45,48). The nitrogen gas target has several advantages over the boron oxide targets for the

$^{11}\text{CO}_2$ production. The amount of activity produced per $\mu\text{A}\cdot\text{hr}$ is over five times greater, and there is essentially no requirement for maintenance of the target. The trace amount of oxygen in the tank nitrogen ($\sim 1\text{ppm}$) is sufficient for the oxidation reaction to occur. Higher oxygen concentration results in the formation of nitrogen oxides from the $^{16}\text{O}(p, \alpha)^{13}\text{N}$ reaction and from radiolysis processes. [^{11}C]-Carbon monoxide contaminants can be eliminated by passing the target gas over CuO at 500°C for catalytic oxidation.

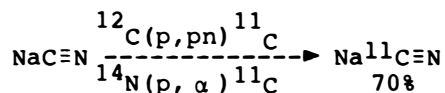
b) ^{11}CN Preparation: A number of methods for the preparation of H^{11}CN has been reported (46-48).

Lamb, et.al. (46) have reported recoil synthesis of ^{11}C -cyanide via direct bombardment of solid LiNH_2 target or $\text{N}_2 + \text{H}_2$ gas. The direct bombardment of $\text{N}_2 + \text{H}_2$ gas mixtures yields H^{11}CN , however, the radiolysis of the product in the target gas reduces the yield. The analysis of ^{11}C -activity in the gas stream from the target during bombardment showed 61% H^{11}CN , $\sim 28\%$ ^{11}C -hydrocarbons, $\sim 9\%$ ^{11}CO and $\sim 2\%$ $^{11}\text{CO}_2$.

Finn, et.al. (47) have reported a chemical synthesis method. [^{11}C]-Carbon dioxide generated from the boron oxide target was reacted with potassium and ammonia yielding K^{11}CN .

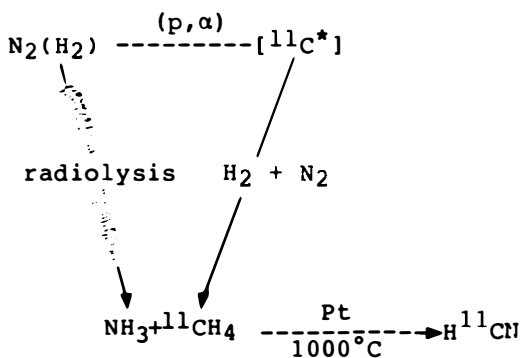


In another method, NaCN target bombarded with protons resulted in the production of Na¹¹C≡N due to recoil labeling.



The ¹¹C-cyanide yield from this target is high but gives poor specific activity. The cyanide specific activity was 11.5 and 62.7 mCi/mmol/100 μA-min for NaCN and B₂O₃ target, respectively.

The method developed by Christman, et.al. (48) is most commonly used for the production of ¹¹C-cyanide. The procedure is described below.



*indicates nucleogenic (energetic) carbon-11

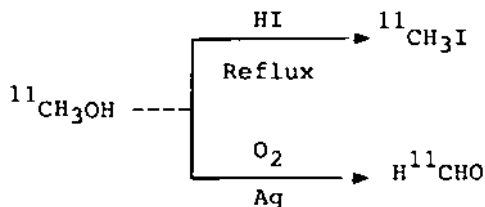
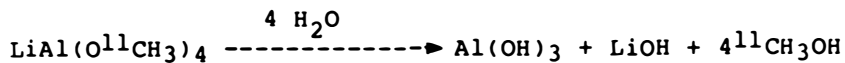
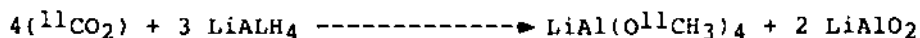
This procedure gives ¹¹C-cyanide in very high specific activity. A routine production method of H¹¹CN by remote

control using the above method has been reported (49). The target gas stream is bubbled through a 3M ammonia solution and sent over 15 g of platinum wire heated at 870°C. [^{11}C]-Cyanide is trapped in water.

Iwata et.al. (149) have recently investigated the catalytic production of H^{11}CN from $^{11}\text{CO}_2$. [^{11}C]-Carbon dioxide was reduced to $^{11}\text{CH}_4$ with H_2 on Ni and then converted to H^{11}CN by reaction with NH_3 on Pt in a radiochemical yield of more than 95%. This method has the advantage that in addition to H^{11}CN other ^{11}C -labeling precursors can be available with a single target system for the production of $^{11}\text{CO}_2$.

c) $^{11}\text{CH}_3\text{I}$ and H^{11}CHO Preparation: The synthetic route commonly used for the preparation of labeled methyl iodide and formaldehyde is the lithium aluminum hydride reduction of labeled carbon dioxide to yield methanol (50-52).

Appropriate subsequent reactions lead to the desired product as shown in the scheme below:



[^{11}C]-methyl iodide has also been produced by direct recoil synthesis by proton irradiation of N_2/HI flow target system (53). The radiochemical yield of $^{11}\text{CH}_3\text{I}$ under optimized condition was 25%.

Recently, (54) diphosphorus tetraiodide solid reagent has been used to convert $^{11}\text{CH}_3\text{OH}$ into $^{11}\text{CH}_3\text{I}$. The reagent was found to be reusable for about fifteen preparations without loss of efficiency and without any contamination of the apparatus resulting from volatile decomposition products. The radiochemical purity of the product varied between 98.5% and 99.5%. The mean specific activity of all the preparations was $3.4 \mu\text{Ci}/\mu\text{mol}$. A fully automated, microprocessor controlled system for the production of $^{11}\text{CH}_3\text{I}$ has been reported (55). Automated systems are now commercially available (56,57).

d) [^{11}C]- C_2H_2 Preparation: [^{11}C]-Acetylene was first prepared by bombarding solid barium carbonate with protons which generated $\text{Ba}^{11}\text{CO}_3$ (58). Upon reduction to [^{11}C]- BAC_2 and subsequent hydrolysis, [^{11}C]- C_2H_2 is released in good yield. [^{11}C]- C_2H_2 has also been produced by ^3He bombardment of calcium carbide (59) to obtain [^{11}C]-labeled carbide followed by hydrolysis with water to obtain acetylene, but this method gives acetylene of very low specific activity (few $\mu\text{Ci}/\mu\text{mol}$). The same type of target when irradiated with protons (60) results in a better carbon-11 production yield, and therefore ^{11}C -acetylene of higher specific activity.

[^{11}C]- C_2H_2 has also been obtained by proton bombardment of simple hydrocarbons (methane, cyclopropane) gas targets (61-63). The highest yield of [^{11}C]- C_2H_2 was ~50% when cyclopropane was used as the substrate.

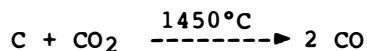
A different approach was developed which generated [^{11}C]- C_2H_2 from the carrier added pyrolysis of $^{11}\text{C}\text{H}_4$ in an inductive argon plasma (64). Sixty percent of the methane is converted instantly into acetylene, 30% remains as $^{11}\text{C}\text{H}_4$. The acetylene obtained in this manner has a specific activity of 150mCi/ μmol and a purity of 98%.

V-2. NITROGEN-13

Radiolabeled N_2 , NH_3 , N_2O , NO_2^- and NO_3^- are of growing interest for use in nuclear medicine as well as in biological, agricultural and environmental studies.

The production methods of important synthetic precursors are described.

a) ^{13}N -gas Preparation: Buckingham and Clark (65) have described a method for preparing saline solution of nitrogen-13, which utilizes graphite as the target material and carbon dioxide as the sweep gas. Nitrogen-13 is produced by the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reaction. In the presence of CO_2 , the graphite which is raised to a temperature in excess of 1450°C by the beam power, is eroded continuously as the following reaction occurs.



The continuous erosion leads to the release of volatile ^{13}N -labelled molecules previously trapped in the graphite lattice. Presumably, reaction of nucleogenic ^{13}N with dinitrogenous contaminates leads to ^{13}NN .

Crouzal and Comar (67) irradiated high purity CO_2 with deuterons, condensed the CO_2 and ^{13}NN in liquid nitrogen. CO_2 was removed by pumping with a Toepler pump and obtained 30 mCi of ^{13}NN from an irradiation of 40 minutes by a 20 μA beam of 9 MeV deuterons. The dissolved ^{13}NN in physiological saline had a final activity of 1 mCi/ml.

Clark and Buckingham (68) have compared the various ways of producing nitrogen-13 solutions. Targetry, production parameters, advantages and disadvantages of each technique are discussed.

Austin, et.al. (21) reported another method using proton bombardment of enriched ^{13}C -targets which is very useful for obtaining high specific activity of ^{13}NN by the $^{13}\text{C}(\text{p},\text{n})^{13}\text{N}$ reaction. They have reported a specific activity of ~20mCi/ml. The bombarding energy of the protons was 11 MeV. The irradiated material is converted to $^{13}\text{N}^{14}\text{N}$ gas by automated Dumas combustion and compressed into 1 ml vials. The entire procedure takes less than 15 min.

Parks, et.al. (69) produced ^{13}N -labeled atmospheric gases via proton irradiation of high pressure oxygen target by the

$^{16}\text{O}(p, \alpha)^{13}\text{N}$ nuclear reaction. The nitrogen atoms are incorporated into the primary products, $^{13}\text{N}_2$, $^{13}\text{N}_2\text{O}$ and $^{13}\text{NO}_2$ in a ratio of 8:1:3.5 respectively. The recoverable fraction of all ^{13}N -labeled compounds was 10%.

Jones, et.al. (70) have described a system for the production of ^{13}N labeled molecular nitrogen (^{13}NN) dissolved in physiological saline. The $^{12}\text{C}(d,n)^{13}\text{N}$ reaction with 50 μA of 6 MeV deuterons from a small medical cyclotron was used to produce ^{13}NN as the sole product in the target chamber. Carbon dioxide is both the target and the sweep gas in a flow system that terminates with continuous absorption of CO_2 in a NaOH bubbler. ^{13}NN is shaken with physiological saline to produce 20 ml of sterile, pyrogen-free injectate with a specific activity of 270 $\mu\text{Ci/ml}$.

Suzuki and Iwata (71) has described a simple method for the ^{13}NN production by irradiating an aqueous solution of ammonia with 15 MeV protons. Irradiation of 0.1M ammonia solution at 10 μA for twenty minutes produced 113 mCi of greater than 99.9% ^{13}NN .

Ferrieri, et.al. (22) have developed a solid ^{13}C -enriched target for the production of ^{13}N as nitrogen gas using the $^{13}\text{C}(p,n)^{13}\text{N}$ nuclear reaction. The target is of the flow-through type which allows for on-line recovery and separation of the radionuclide from the enriched target matrix and permit reuse of the target without detriment to its production

capability. At low beam current the untreated target yielded mostly oxides of ^{13}N . However, reduction in the absorbed oxygen concentration through a heating pretreatment of the target served to enhance the labeled dinitrogen products 8-fold. At 10 μA beam intensity the pretreated target yielded in greater than 99% radiochemically pure ^{13}N -nitrogen gas.

b) ^{13}N -Ammonia Preparation: [^{13}N]-Ammonia is the most widely used ^{13}N -labeled precursor for synthesis. It has also been the most exploited form of Nitrogen-13 for medical use. Ammonia has been prepared by irradiating solid inorganic carbides with deuterons i.e. ($^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$). For example, aluminum carbide target upon dissolution in acid solution gave $^{13}\text{NH}_3$ and $\text{CH}_3^{13}\text{NH}_2$ as the major product (66). However, ^{28}Al is also produced by the irradiation of this carbide target. This constitutes a considerable radiation hazard in handling and processing the target.

Tilbury, et.al. (72) have suggested an alternate procedure, which involves the deuteron bombardment of methane. The methane was flowed through a glass target chamber into a bubbler containing isotonic saline solution. Analysis of the solution in the bubbler by radioactive gas chromatography indicated $^{13}\text{NH}_3$ (95%), $\text{CH}_3^{13}\text{NH}_2$ (2%), HC^{13}N ($> 3\%$) and $\text{C}_2\text{H}_5^{13}\text{NH}_2$ (trace).

Presently, the most commonly used method for $^{13}\text{NH}_3$ is the proton irradiation of an H_2O target (73-76). Nitrogen-13

labeled $^{13}\text{NO}_3^-$, $^{13}\text{NO}_2^-$ and $^{13}\text{NH}_4^+$ are produced during the bombardment. The nitrate and nitrite is chemically reduced to ammonia by using De Varda's alloy.

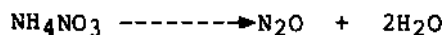
The irradiated water also contains ^{15}O and ^{18}F along with the ^{13}N . Fluorine-18 gives no problem as this radioisotope does not distill, but ^{15}O is present in the final ^{13}N -ammonia solution. Usually, the overall time necessary for the preparation of the ammonia solution and the transport of the radiopharmaceutical is long enough that most of the ^{15}O has already decayed prior to delivery of the labeled compound.

In a recent study, $^{13}\text{NH}_3$ was synthesized on-line by exposing ^{13}N -labeled nitrogen gas plus hydrogen to a microwave discharge (22). The reaction of the atomic ^{13}N with hydrogen was enhanced four-fold by placing pre-reduced palladium metal fillings (heated to 200°C) after the discharge cavity. A ten-minute discharge resulted in 65% conversion of the ^{13}N -labeled nitrogen gas introduced into the recirculation loop, to ^{13}N -ammonia. However, the efficiency of conversion to ammonia was found to be very sensitive to the trace presence of trace chemical contaminants.

c) $^{13}\text{N}_2\text{O}$ Preparation : Nickles, et.al. (77) have reported two methods for the synthesis of $^{13}\text{N}_2\text{O}$ by pyrolysis of NH_4NO_3 in sulfuric acid.

Method 1

Carrier NO_3^- and $^{13}\text{NO}_3^-$ were reduced in the DeVarda's alloy to $^{13}\text{NH}_3$ which was distilled from basic solution and collected by bubbling through 25 ml of cold H_2SO_4 which contained an additional 3mmol NH_4NO_3 . The apparatus was swept by a Nitrogen gas stream.



The H_2SO_4 was heated to 260°C and N_2O produced by the reaction was quantitatively evolved and collected over water.

Method 2

The irradiated water was combined with carrier NH_4NO_3 and concentrated to 1 ml by rotary evaporation. Twenty-five ml of H_2SO_4 containing 3 mmol of additional NH_4NO_3 was added and N_2O evolved at 260°C as in Method 1.

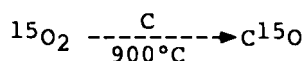
V-3. OXYGEN-15

Oxygen-15 can be produced by the $^{14}\text{N}(\text{d},\text{n})^{15}\text{O}$, $^{15}\text{N}(\text{p},\text{n})^{15}\text{O}$ and $^{16}\text{O}(\text{p},\text{pn})^{15}\text{O}$ nuclear reactions. By the first two reactions, oxygen-15 can be produced in high specific activity.

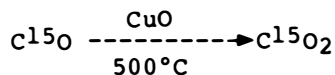
a) $^{15}\text{O}_2$ Preparation: ^{15}O -labeled oxygen can be produced by using a flowing gas target of 0.5 - 4.0% $\text{O}_2 + \text{N}_2$ with the $^{14}\text{N}(\text{d},\text{n})^{15}\text{O}$ reaction. Whereas, high purity oxygen gas was

used as a target gas for the $^{16}\text{O}(p,pn)^{15}\text{O}$ reaction. If the O_2 carrier gas was greater than 2%, the $^{15}\text{O}_2$ yield increases to about 97%. The radiochromatographic (9) analysis of the target gas for $^{14}\text{N}(d,n)^{15}\text{O}$ reaction showed $^{15}\text{O} = 97.5\%$, $\text{N}_2^{15}\text{O} = 2.3\%$, $\text{C}^{15}\text{O}_2 = 0.54\%$ and $^{13}\text{N}_2 = 0.15\%$. Whereas, the analysis of the target gas for the $^{16}\text{O}(p,pn)^{15}\text{O}$ reaction showed $^{15}\text{O} = 99.5\%$, $^{11}\text{CO}_2 = 0.2\%$, $^{13}\text{N}_2\text{O} = 0.1\%$, $^{13}\text{N}_2 = 0.1\%$ and $^{11}\text{CO}_2 = 0.04\%$. The CO_2 , N_2O and CO can be removed by passing the target gas through a soda lime and an activated charcoal trap at ambient temperature. Further details about ^{15}O - O_2 production are given in the literature (68). The production yields of all the reactions for ^{15}O -production were recently compared (9).

b) C^{15}O and C^{15}O_2 Preparation: Carbon monoxide labelled with oxygen-15 can be prepared by passing the target gas through activated charcoal maintained at 900°C .



Reaction at 500°C generates a mixture of C^{15}O and C^{15}O_2 . Conversion of C^{15}O to C^{15}O_2 can be obtained on-line with CuO combustion furnace maintained at 500°C .



C^{15}O_2 can also be produced on-line by using a flowing N_2/CO_2 gas target.

c) H₂¹⁵O preparation: The preparation of oxygen-15 labeled water (H₂¹⁵O) was first prepared in small amount by passing labeled molecular oxygen together with hydrogen gas over a heated palladium catalyst (78).

Welch and co-workers (79) have prepared H₂¹⁵O by an exchange method. This technique utilized the fast exchange reaction between labeled carbon dioxide (C¹⁵O₂) and water. Using this method, water with a specific activity of 80 mCi/ml was prepared.

Oxygen-15 labeled water can also be prepared by recoil labeling technique (80). Oxygen-15 is produced in a flowing N₂+H₂ (5%) target via the ¹⁴N(d,n)¹⁵O reaction. The ¹⁵O reacts directly with molecular hydrogen in the gas phase to produce labeled no-carrier-added water.

Nickles, et.al. (81) have described a catalytic generator for the production of H₂¹⁴O and H₂¹⁵O. The initial activities of ¹⁵O₂ and ¹⁴O₂ were produced from the ¹⁴N(d,n)¹⁵O and ¹⁴N(p,n)¹⁴O nuclear reactions, respectively. The target gas was mixed with hydrogen and carrier oxygen at 500 and 100 ml (STP)/min., respectively. The ternary gas mixture flowed over a platinum-on-alumina catalyst contained in a water-cooled quartz tube. At an incident energy of 8 MeV, the H₂¹⁵O activity saturated to 15mCi/μA.

d) N₂¹⁵O Preparation: Recently, an on-line synthesis method of ¹⁵O-labeled nitrous oxide has been reported (147). The

oxygen-15 was produced by the $^{14}\text{N}(d,n)^{15}\text{O}$ reaction. ^{15}O -Nitrous oxide was produced by catalytic oxidation of anhydrous ammonia in a gas mixture containing ^{15}O -labeled molecular oxygen. Anhydrous ammonia was mixed with the gas containing $^{15}\text{O}_2$, and after preheating to about 200°C was carried through an oven containing a Pt catalyst kept at about 310°C . Labeled gas was purified in H_3PO_4 and KOH . Radiochemical purity of the ^{15}O -labeled nitrous oxide was greater than 98%, and the specific activity at the end of synthesis was about 50 mCi/mmol.

VI. RADIOCHEMISTRY IN MATERIALS SCIENCE APPLICATIONS

The knowledge of trace levels of carbon, nitrogen and oxygen in different solid materials is of scientific, technological and economic importance. For example, the presence of trace amounts of oxygen in pure metals and alloys often has a detrimental effect on parameters such as ductility and electrical conductivity. Semiconductor behavior can be adversely affected by low level C, N, O impurities. Conventional vacuum fusion and inert gas fusion techniques are adequate for the assay of the relatively large amounts of oxygen (97) e.g. $> 5 \mu\text{g}$. However, the level of oxygen currently of interest in the field of pure materials and semiconductor research are very low ($< 10 \text{ ppm}$); additionally, the amounts of sample material available are often small. Under these conditions, the conventional fusion techniques are of limited use due to the problem associated with apparatus and reagent blanks and surface contamination of the samples. The micro-Kjeldahl and the fusion extraction are the most useful method for the determination of trace nitrogen in solids. These methods are capable of determining nitrogen down to 1 ppm. The blank problem is usually the limiting factor for these techniques.

Several nuclear techniques based on different principles have been developed for the determination of trace contents of carbon, nitrogen and oxygen. The following are the important activation techniques.

- a) Charged Particle Activation
- b) Photon Activation Analysis (PAA)
- c) Neutron Activation Analysis (NAA)

1) CHARGED PARTICLE ACTIVATION

The charged particle activation technique has been described elsewhere (97,98). Different types of interaction occur as a result of ion-beam interaction with matter. They can be at the atomic or nuclear levels, and some of these interactions result in the emission of signals which are element or isotope specific. The probability of the different interactions varies with the type and the energy of the bombarding ions. Three types of interactions can be exploited for chemical analysis.

- 1) The emission of characteristic X-rays induced by charged-particle beams has been used for elemental analysis. Inner shell vacancies created due to ion-atom collision are filled from outer shells. This results in the emission of K, L, or M X-rays. This is the basic principle behind proton induced X-ray emission technique (or PIXE). Details about PIXE are described elsewhere (82-84).
- 2) The energy of the elastic scattered projectiles are used to mass analyze the element in the target surface. This technique is known as Rutherford back scattering spectrometry (85).

3) Nuclear reactions yielding new radioisotopes. The characteristic γ -ray and the amount of activity is used to identify and quantify the parent isotopes. The bombarding particles are protons, deuterons, tritons, helium-3 ions and helium-4 ions or alphas.

The elements may be determined singularly or in combination due to natural occurrence and similarity of possessing a short half-life and detection by annihilation radiation.

Pierce, et.al. (86) have determined the concentration of carbon in steel by measurement of the prompt γ -radiation emitted during proton bombardment. Targets were in the form of steel disks. During irradiation, a 2.3 MeV γ -ray was emitted, and this was measured in these determination. By this method, they were able to measure the concentration of carbon to as low as 700 ppm.

Goethals, et.al. (87) have determined the concentration of carbon in aluminum by deuteron activation analysis via the $^{12}\text{C}(d,n)^{13}\text{N}$ reaction. The samples and standards were placed in a water-cooled target holder, and irradiated with a 7-MeV deuteron beam. The ^{13}N was separated from other activities by steam distillation. A carbon concentration of $0.25 \mu\text{g.g}^{-1}$ with a standard deviation of $0.05 \mu\text{g.g}^{-1}$ was obtained for aluminum of 99.5% purity.

Clemenson, et.al. (88) determined total carbon in atmospheric aerosol. Deuterons of 7.6 MeV were used to bombard the target

to produce ^{13}N by the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ nuclear reaction. This method was non-destructive, which permitted the sample to be studied by additional methods. Comparison of carbon found by deuteron activation analysis with that found by independent, but destructive combustion methods showed a standard deviation of 10% for 15 samples analyzed over a wide range of carbon contents. The detection limit was estimated to be $0.5 \mu\text{g}\cdot\text{cm}^{-2}$, corresponding to a carbon concentration of 0.2% in a sample of total thickness $250 \mu\text{g}\cdot\text{cm}^{-2}$.

Bottger, et.al. (89) have developed a sensitive method for the determination of carbon in silicon by deuteron and ^3He particle activation followed by chemical separation of the measured nuclide (^{13}N and ^{11}C). ^{13}N was produced by the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reaction; whereas ^{11}C was produced by the $^{12}\text{C}(^3\text{He}, \alpha)^{11}\text{C}$ reaction. The detection limit, was improved by a separation of ^{30}P and ^{30}Si formed by deuteron reaction in silicon by inert gas fusion extraction. The detection limit was reported as ~3 ppb for deuteron activation, and ~1 ppb for ^3He activation.

Vandecasteele, et.al. (90) have determined carbon in gold layers electroplated on brass by deuteron activation analysis using the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ reaction. The results range from 2 to $1300 \mu\text{g g}^{-1}$ and the relative standard deviation from 0.9 to 13%.

Barit, et.al. (91) have determined the carbon concentration in thin layers using a 2 MeV Van de Graaf accelerator. Carbon

was determined by nondestructive method in layers of 3-10 μm thickness. The method was based on using the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ nuclear reaction with simultaneous account of surface contamination by means of the $^{12}\text{C}(\text{d},\text{p})^{13}\text{C}$ competitive reaction. Variation of the amount of ^{12}C nuclei on the sample surface per cm^2 and the temperature of annealing for 100 μm molybdenum foil were studied. Minimal surface carbon content was achieved in the case of annealing samples previously oxidized to surface oxygen contents of 2-5 $\mu\text{g}/\text{cm}^2$. The irradiation of the samples by proton or deuteron beams of high intensity can cause the diffusion of ^{13}N as well as carbon from the surface to the bulk of the sample. Diffusion of ^{13}N is not influenced, since the total yield of ^{13}N was independent of the location in the sample. The temperature and time of annealing was chosen to minimize the diffusion of carbon. The accuracy of the method was verified by comparison with other analytical methods.

Macias, et.al. (92) have reported the development of nondestructive method for the determination of carbon, nitrogen and sulfur in atmospheric aerosols. The method was based on γ -ray emission following the inelastic scattering of 7.0 MeV protons. The inelastic scattering of protons excites the target nucleus to a low-lying excited states which deexcite by the emission of γ -ray. The γ -ray spectrum from each sample, recorded during the bombardment, contains at least one γ -ray from each element of interest. A calibration for

nitrogen, sulfur and carbon deposited on filters was carried out using standard methionine aerosol ($C_5H_{11}O_2SN$) samples. The results obtained by γ -ray method were compared with other methods. Total carbon was obtained by using the Dohrmann DC-50 Carbon Analyzer. An average deviation of 7.3% was found between the results of two methods. Carbon, nitrogen, and hydrogen were also measured using a Model 240 Elemental Analyzer. The average difference between this and γ -ray methods for nitrogen was 11% and 16% for carbon. The likely explanation of the poorer agreement for carbon lies in the method of blank subtraction. Detection limits for atmospheric samples were in the $\mu g/cm^2$ range with a precision of 5%.

Strijckmans, et.al. (93) have determined carbon, nitrogen and oxygen in nickel by charged particle activation using the reactions $^{12}C(d,n)^{13}N$, $^{14}N(p,n)^{14}C$ and $^{16}O(^3He,p)^{18}F$. The ^{14}C , ^{13}N and ^{18}F were separated by oxygen combustion followed by trapping of $^{14}CO_2$ in NaOH and by steam distillation of $^{13}NH_3$ and of H_2SiF_6 - ^{18}F , respectively.

A method for the analysis of the rare stable isotopes of carbon, nitrogen and oxygen using charged-particle induced prompt γ -rays have been reported (94). The sensitivity of the method depends on various experimental conditions such as background, incident particle energy, beam distribution, sample homogeneity, target overheating, and the performance of the counting equipment. Interfering nuclear reactions can affect the sensitivity of the method. To obviate

interferences, the choice of a suitable projectile and incident energy was important. Protons gave the best result for ^{13}C . Deuterons were ideal for assaying ^{15}N , because the analysis was free of interference. All three particles can be used for the analysis of the stable tracer of oxygen.

Clayton & Wooller (95) have used helium backscattering to determine hydrogen, carbon, nitrogen, oxygen and sulfur in hair samples. Spectra were measured with a beam of 1.4 MeV $^4\text{He}^+$ ions incident normally to the sample surface, at a target current of 50 μA . The backscattered particles were detected with a silicon surface barrier detector set at 135° to the beam.

Clemson, et.al. (99) have determined total nitrogen in atmospheric aerosols by proton activation analysis. Radioactive ^{11}C was detected via its 0.511-MeV annihilation radiation following the $^{14}\text{N}(p,\alpha)^{11}\text{C}$ reaction. A comparison of nitrogen found by proton activation method to that found by destructive combustion method gave an average percent difference of 14% for 17 samples analyzed over a concentration range of two orders of magnitude. A detection limit of approximately 0.1 $\mu\text{g}/\text{cm}^2$, (corresponding to 200 ppm) was calculated for an aerosol sample of 500 $\mu\text{g}/\text{cm}^2$ thickness.

Sastri & Krivan (101) have proposed a new activation method for the determination of nitrogen with beryllium-7 as the indicator radionuclide. Beryllium-7 was produced by the $^{14}\text{N}(p,2\alpha)^7\text{Be}$ and $^{14}\text{N}(d,2\alpha n)^7\text{Be}$ nuclear reactions. Both

activation methods were applied to the determination of nitrogen in niobium. The result obtained were compared with that found by the Kjeldahl method. The detection limit for deuteron activation was 25 ppb (at 25 MeV). The detection limit for proton activation, was as low as 3 ppb (at 22 MeV).

Gonzi, et.al. (101) have used the $^{14}\text{N}(d,p)^{15}\text{N}$ reaction for measurements of nitrogen depth distribution in single seeds of wheat. The measured nitrogen distributions show striking correlations with parameters describing the nitrogen level of fertilizer, the time of harvesting, the grain position in a head and the variety of wheat being analyzed. Gonzi also discussed the possibility to use the technique for selection purposes in plant breeding.

Morales, et.al. (102) have reported nitrogen determination with $^{14}\text{N}(p, \alpha)^{11}\text{C}$ reaction at 6.81 MeV performed in organic compounds. Positron annihilation from ^{11}C allowed determination of nitrogen contents to as little as 0.35% dried weight. The results were identical with the Kjeldahl method. The protein content of an agriculture product is the most important indicator of its nutritional value. A conversion factor of 6.25 related nitrogen to protein content in cereals.

Constantinescu, et.al. (103) have performed the analysis of protein content in cereal by total-nitrogen proton activation. The total nitrogen was determined using the $^{14}\text{N}(p,n)^{14}\text{O}$ reaction. The ^{14}O activity is detected by means of its

characteristic 2.312 MeV gamma rays. A mechanized system was constructed, which was capable of analyzing samples at a rate of one per minute. The radiation damage was concentrated in a layer of 1.7 - 2 mm under the irradiated surface of the seeds. The embryo region was protected by selecting a suitable geometry of the grains. The future germination of the analyzed seeds was quite normal. This is an important advantage of the method as it permits seedlings of richest protein grains.

Shikano, et.al. (104) have determined carbon in gallium arsenide by deuteron activation analysis using the $^{12}\text{C}(d,n)^{13}\text{N}$ reaction. Interferences of activities induced from matrix and impurities were removed. Produced ^{13}N which might be present in various chemical species such as $^{13}\text{NH}_4^+$, $^{13}\text{NO}_3$ and $^{13}\text{NO}_2$ was converted to $^{13}\text{NH}_3$ by steam distillation with Dewarda alloy and consequently precipitated substoichiometrically as ammonium tetraphenylborate. It was found that the decontamination factors of coexistent elements such as ^{72}Ga , ^{76}As and ^{11}CO to ^{13}N were large enough to determine carbon as low as 1 ppb in gallium arsenide.

Shikano, et.al. (105) have also reported the determination of oxygen in gallium arsenide by ^3He activation analysis using substoichiometric precipitation. ^{18}F is produced by $^{16}\text{O}(^3\text{He},p)^{18}\text{F}$ reaction. The radiochemical separation of ^{18}F was required for the accurate determination of oxygen at the ppb level. The nuclear reaction of $^{75}\text{As}(^3\text{He},2n)^{76}\text{Br}$ produced

^{76}Br (β^+ , $t_{1/2} = 15.9$ h) which interfered with the measurement of the ^{18}F activity. Fluoride was separated by distillation of fluoride as fluorosilic acid and finally precipitated as lanthanum fluoride. Two distillations followed by substoichiometric precipitation were applied to determine the oxygen concentration in gallium arsenide.

Nozaki (106) had reported that ^{76}Br was decontaminated to approximately 10^{-7} by the radiochemical separation of ^{18}F as KBF_4 . The chemical yield of the separation was corrected weighing the precipitate of KBF_4 . Oxygen was determined in gallium arsenide.

A method for the analysis of the differential and integral distributions of light elements admixed to the surface layer of materials have been proposed (107). The method was based on anomalous scattering of α - particles at projectile energies above the coulomb barrier on light nuclei. It was found that the minimum numbers of admixed nuclei of carbon, nitrogen and oxygen, which could be determined, were 2×10^{13} , 1.5×10^{14} and 1×10^{13} nuclei.cm⁻², respectively.

Lee, et.al. (108) have developed a radioactivation method to determine trace concentrations of oxygen in copper by using ^3He as the bombarding particle. The ^{18}F was produced by the nuclear reaction of ^{16}O with low energy ^3He ions. At ^3He energies below 6.3 MeV, no interfering radioactivities from reactions with the Cu matrixes were present, therefore, good

results can be achieved by nondestructive analysis. A radiochemical separation method of ^{18}F as Pb^{18}F_2 was reported. Fluorine, if present, would produce ^{18}F ; via the $^{19}\text{F}(^3\text{He},\text{p})^{18}\text{F}$ reaction. The cross section for the $^{19}\text{F}(^3\text{He},\text{p})^{18}\text{F}$ reaction is 15 mb at 6.5 MeV; whereas it is 350 mb for the $^{16}\text{O}(^3\text{He},\text{p})^{18}\text{F}$ reaction. It will require 24 times as much fluorine as oxygen to produce an equal amount of ^{18}F at 6.5 MeV. If fluorine is present in considerable amounts, a chemical separation would be necessary. The oxygen content of 1×10^{-4} inch thick Cu foil was measured to be 550 ± 30 ppm. The relative standard deviation was 5%.

Wai & Dysart (109) have used proton activation analysis with the $^{18}\text{O}(\text{p},\text{n})^{18}\text{F}$ reaction for the determination of oxygen in molten alkali halides. The ^{18}F produced was separated radiochemically as LaF_3 . The solubility product of LaF_3 is extremely small ($K_{\text{sp}} = 3 \times 10^{-19}$). The radioactive ^{18}F produced in the chloride salt samples was precipitated in the form of LaF_3 by the addition of LaCl_3 and $\text{NH}_4\text{F} \cdot \text{HF}$ to the acid solution. Co-produced ^{64}Cu , ^{72}Ga , $^{69\text{m}}\text{Zn}$ were separated from the ^{18}F . In the case of alkali fluoride salts, the $^{19}\text{F}(\text{p},\text{d})^{18}\text{F}$ reaction can cause interference for oxygen determination. The Q-value of this interfering reaction is -8.2 MeV. This can be easily eliminated by keeping the bombarding energy of the projectile below 8.2 MeV.

Ricci and Hahn (110) have developed an approximate method for the rapid computation of the average cross section to

facilitate the calculations of sensitivities and interferences in terms of bombarding energies in ^3He activation analysis. The method is based on the straight-line fitting of excitation functions, which leads to the final simple formula $\bar{\sigma} = 2/3 mE - a + b/E^2$. The constants m , a , b and other important parameters were given for most significant reactions. The average error of the method was not found to be larger (and in most cases much less) than 13.5%.

Proton activation can be used to determine the oxide film thickness on metals (111). The method was based on the activation of naturally occurring oxygen-18 with protons according to the reaction $^{18}\text{O}(p,n)^{18}\text{F}$. With a proton energy of 4 MeV and beam current of 10 μA , the lower limit of detection of the method corresponds to a thickness of the order of 1Å. The upper limit is $10^5 \mu\text{A}$. Elements interfering with the measurement were copper, nickel, zinc and titanium. The carbon distributions in metals can be examined by the nuclear microprobe method (112). Pummery and McMillan (113) improved the sensitivity of the nuclear microprobe method from 20 ppm to 1 ppm in pure metals. The improvement was obtained by optimizing counting and irradiation conditions. Excellent agreement was obtained between nuclear microprobe and gamma-photon activation analysis of carbon in nickel at concentrations between 10 and 1000 ppm. Techniques for, and the limits of detection of, CPAA have been discussed by Engelmann (150-158).

2) PHOTON ACTIVATION ANALYSIS (PAA)

Nuclear reactions may be induced by the interaction of photons with target nuclei. The production of the high energy photon beam is usually accomplished by accelerating electrons in a Linac, and then allowing them to impinge upon a tungsten target. The slowing-down of the electrons causes the emission of photons which are known as "bremsstrahlung." The bremsstrahlung may be used to induce nuclear reactions in a second target. Such reactions are called photonuclear reactions. Photon activation analysis is useful for a number of elements such as carbon, nitrogen, oxygen, iron and lead which are not highly activated with thermal neutrons. In addition, severe difficulties due to sample self-shielding are encountered when a matrix to be analyzed contains substantial amounts of elements with large thermal neutron absorption cross sections. High energy photon activation is useful in many of these situations.

Lutz (114) has calculated the sensitivities for photon activation analysis. The expected specific activity has been estimated from bremsstrahlung produced by an electron accelerator. The photon flux distribution was calculated for electrons striking a 0.6 cm tungsten target at different energies. The yield (i.e. flux times cross section) for photonuclear reactions of analytical utility were integrated. The disintegration rates of the reaction products for most of the elements under conditions of different electron energies

and irradiation times were calculated. Nordmann, et.al. (115) have determined oxygen in cesium. The technique developed enabled metal analysis in the bulk of the sample by mechanically scraping the surface of cesium after its irradiation in the solid state. The oxygen-15 was separated. By this method, $0.1 \mu\text{g g}^{-1}$ of oxygen could be determined.

Sharma, et.al. (116) have investigated the accuracy in the determination of C, N, O, F, D and K by the photon activation method. The method can be applied for the estimation of the elements with a precision of 0.6% in a compound provided that the amount of one of the elements is known. However, the limitation can be overcome by intimately mixing known amounts of a compound as an internal standard in the sample provided the elements in the compound are not present in the sample. It was shown that the elemental estimation in both inorganic and organic compounds can be carried out with good precision.

Rodionov, et.al. (117) have reported the determination of carbon in high purity iron and molybdenum. The irradiations were performed at 30 MeV. After the irradiation, the samples were etched with three successive acid baths in order to remove a layer approximately of 50 μm . For iron the bath was HNO_3 90% + HF 10%. For molybdenum the bath was H_2O 45% + H_2SO_4 45% + HNO_3 10%. The sample was then introduced in an alumina crucible containing the oxidizing bath of 87.5% Pb_3O_4 and 12.5% B_2O_3 and a piece of iron in order to start the fusion and to provide carbon carrier. The crucible was heated

by a high frequency furnace of 2.5 KW. The CO₂ formed during fusion was carried by an argon flow. The gas was passed through Schutze reagent in order to oxidize the eventually formed CO, and finally the CO₂ was adsorbed on KOH. The detection limit of 0.01 μg.g⁻¹ was obtained.

A method of carbon determination in chromium by photon activation analysis has been described (118). The samples were irradiated by photons emitted from a platinum target submitted to an electron beam of 35 MeV. After the irradiation, the sample was etched in hot 1 N HCl, in order to remove surface contamination. The sample was then oxidized and dissolved in a fused bath containing 1.5 g of NaOH, 5g of NaNO₃ and 5 mg of Na₂CO₃ in a nickel crucible heated by gas burner. The oxidation was performed in less than two minutes. The bath was then cooled, and the obtained solid residue was transferred to flask A of the apparatus. (Fig. 1). This apparatus was swept by air at a flow rate of about 0.2 l.min⁻¹ which was bubbling through two absorbers containing 12 M KOH. The residue was dissolved in 9 M HNO₃. The solution was heated to boiling and the air flow maintained for 10 minutes. The control was performed by using radiotracer in conditions quite similar to the analytical procedure. The radioactivity of the KOH absorbers was measured during the whole procedure. Evaporation of CO₂ took place just after acidification and was accelerated by heating the solution. The detection limit was 0.03 μg.

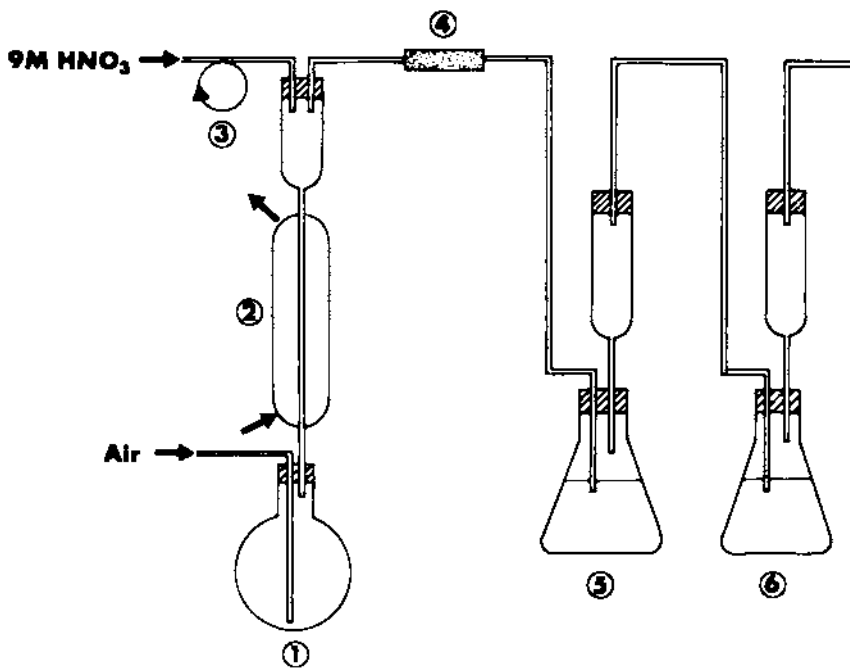


Figure 1 Apparatus for the separation of carbon. 1 - flask for the dissolution of the solid residue of the alkaline bath, 2 - refrigerant, 3 - peristaltic pump, 4 - quartz-wool filter, 5,6 - 12 M KOH absorbers.

Oxygen can be analyzed by the $^{16}\text{O}(p,p2n)^{14}\text{O}$ and $^{16}\text{O}(\gamma,2n)^{14}\text{O}$ reactions. These reactions have been used to determine the oxygen contents in the organic acid. (119). The main interfering element, if protons were used, is nitrogen because it constitutes a direct source of ^{14}O due to the $^{14}\text{N}(p,n)^{14}\text{O}$ reaction. In the presence of nitrogen, photon activation according to $^{16}\text{O}(\gamma,2n)^{14}\text{O}$ should be the method of choice.

Williams, et.al. (120) have determined trace levels of oxygen using the reaction $^{16}\text{O}(\gamma,n)^{15}\text{O}$. The ^{15}O was separated from the matrix by inert gas fusion technique. The post irradiation etched sample, together with a suitable flux, was placed in graphite crucible. The purpose of the flux was to ensure intimate contact between the fused sample and the crucible material and to provide inactive oxygen carrier. The flux can also inhibit the release of certain undesirable elements from the melt. The choice of the flux depends upon the matrix to be analyzed. The limit of detection for oxygen lies in the range of 0.2 to 0.02 μg . Oxygen was analyzed in the following materials: steel, iron, copper, nickel, molybdenum, germanium, arsenic and indium phosphide.

Fedoroff, et.al. (121) have reported a systematic study of radiochemical separations of carbon in molten salts. The following three basic mixtures of molten salts were studied: $\text{Pb}_3\text{O}_4\text{-B}_2\text{O}_3$, $\text{NaOH} - \text{NaNO}_3$ and $\text{H}_2\text{SO}_4 - \text{KIO}_4$ with some addition of complexing agents. Selected procedures for the determination of carbon by photon activation were reported for

the following samples: Ag, Al, Cr, Fe, Mg, Mo, Ni, Si, Ti, W, Zn, Zr, AlMg, AgZn and ZnMgTe. The detection limit for carbon was 10^{-8} g and the accuracy from 5% to 10%.

Schmitt, et.al. (122) have measured the concentration of nitrogen and fluorine in Air-Dust samples by PAA. The air-dust was collected on a "mikrosorbanfilter" (polystyrene) 250 mm in diameter by filtering 7160 m³ of air. Photon energy of 15 MeV was used to avoid interference of carbon and oxygen. Copper and nitrogen have approximately the same threshold for activation, the same half-life and the same β^+ radiation producing the annihilation radiation of two γ -rays of 511 KeV. Neither nuclide emits characteristic radiation which can be used for γ -spectroscopy. Radiochemical separation of nitrogen was not possible because of the chances of nitrogen loss during ashing of the filter material. Copper was analyzed by atomic absorption spectrometry (AAS) after the activation analysis. The sample of about one milligram of dust contains less than one microgram of copper. The interference due to copper was negligible due to the low copper concentration. The activation was followed by measurement of the decay curves of nitrogen and fluorine. The detection limit for the instrumental method was 4 μ g for both nitrogen and fluorine.

Davidson and Landsberger (123) have measured the concentrations of carbon in coal by instrumental photon activation analysis by means of the $^{12}\text{C}(\gamma, n\alpha)^7\text{Be}$ reaction. A rotating target holder was used to ensure a uniform exposure

of samples. Accuracy was determined by analyzing coal standards from the National Bureau of Standards. The overall error was about 4.7%, and carbon detection limits ranged from $19 \mu\text{g}\cdot\text{g}^{-1}$ to $44 \mu\text{g}\cdot\text{g}^{-1}$.

Fedoroff, et.al. (124) have determined simultaneously nitrogen and carbon in silicon by PAA. The analytical procedure included irradiation by 30 MeV photons followed by a chemical separation by evolution from molten KOH. Two radiochemical states of nitrogen were observed, i.e. NH_3 and N_2 . Finally, all ammonia were converted to N_2 which was absorbed on a titanium sponge. Carbon was converted to CO_2 , which was absorbed in a KOH solution. Detection limits of $0.01 \mu\text{g}$ carbon and $0.1 \mu\text{g}$ nitrogen were achieved.

3) IN VIVO PHOTON ACTIVATION ANALYSIS

Recently, measurement of total-body oxygen, nitrogen, and carbon in vivo by photon activation analysis has been reported (125). The γ -ray beam of a 45-MeV betatron and a whole body counter was used for the measurement ^{15}O , ^{11}C and ^{13}N . The decay curves were corrected for interfering activity from ^{30}P , ^{38}K , and $^{34\text{m}}\text{Cl}$, and in the case of live animals, also corrected for a substantial fraction of ^{11}C lost through exhalation. With a radiation dose of 40 cGy, total body O, N and C were measured in dead rats with estimated accuracies of 1.4% , 4.5%, and 1.5%, respectively.

4) NEUTRON ACTIVATION ANALYSIS

Neutron activation analysis has been reported in the literature for the analysis of carbon, nitrogen, and oxygen.

The samples can be analyzed using the conventional neutron activation analyses (NAA) or neutron-capture prompt γ -ray activation analysis (PGAA). In conventional NAA the bombarding particle is absorbed and a new radioactive isotope is produced. This isotope has a characteristic half-life and gamma-ray. Whereas in PGAA, one observes γ -rays emitted while the sample is being irradiated with neutrons. Nuclei formed in capture have excitation energies equal to the binding energy of the added neutron, from 5 to 11 MeV. The excitation energy is released by emission of one of several "prompt" γ -rays over times $< 10^{-14}$ s. In PGAA, the neutron capture does not necessarily form a radioactive species. The resulting product may be stable, have a very short or long half-life or emit no intense γ -radiation. The details of the PGAA technique have been described by Failey, et.al. (126). Three basic analytical configurations are utilized. Fast neutrons from the spontaneous fission of ^{252}Cf are thermalized by moderators before impingement on the sample. This neutron source is rugged and portable, but its detection limit for carbon and nitrogen analyses suffer from low neutron fluxes. Two different reactor based methods are available. A neutron beam may be extracted from the reactor, and the sample placed into this beam and observed with a nearby Ge(Li) detector.

Alternately, the sample may be placed directly in the thermal column of the reactor, and observed at some distance by a Ge(Li) detector (127,128). The former has a much lower neutron flux, but a more efficient counting geometry.

Bobrova & Navalikhin (129) have reported the determination of the total nitrogen content and its incorporation into titanium carbide with fast neutron activation analysis. The nitrogen-13 was separated by distillation as ammonia. It was established that the bulk of the nitrogen was completely bonded to the matrix in the form of titanium carbonitride and other chemical forms, as well as in the form of nitride. NAA has also been used to measure the nitrogen, oxygen, fluorine, phosphorus, sulfur and chlorine in aluminum alloys (130). Measurements of nitrogen in various plants (131) have also been reported. Semei, et.al. (134) have used the $^{14}\text{N}(n,2n)^{13}\text{N}$ reaction to measure nitrogen content in some foodstuff. The nitrogen concentration values were used to calculate the protein contents.

Lavrukhina, et.al. (132) have measured the nitrogen content in meteorites and rocks. The method is based on the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction. The samples were irradiated for 129h in a high flux reactor. Carbon-14 was isolated as CO_2 . Gas-filled proportional counters were used. The detection limit of 0.001 μg was obtained.

Rouchard and Fedoroff (136) have also applied this reaction for determining nitrogen in steel and semiconductors. The

sample was irradiated for 70h in thermal-neutron flux. A selective chemical separation was performed. The samples were counted in a liquid scintillation counter. A limit of detection of less than $1 \mu\text{g}\cdot\text{g}^{-1}$ of nitrogen was achieved.

Anderson (133) developed a procedure for assaying for low levels of oxygen in uranium. The nuclear reaction used for oxygen assay was $^{16}\text{O}(n,p)^{16}\text{N}$. The neutron energy was 14 MeV. The 6.13 MeV gamma-ray of $^{16}\text{N}(t_{1/2}=7.13 \text{ s})$ was used for oxygen assay. The procedure described has been successful for assaying oxygen in uranium in the 0.1 - 1.0 wt% (U/O) range. Because of the poor counting statistics, multiple irradiation-counting cycles were required to obtain results with reasonable uncertainty.

Recently, Chong, et.al. (135) have reported that the values of nitrogen concentration obtained by the $^{14}\text{N}(n,2n)^{13}\text{N}$ reaction need to be corrected. ^{13}N is also produced by the $^{12}\text{C}(p,\gamma)^{13}\text{N}$ and $^{16}\text{O}(p,\alpha)^{13}\text{N}$ reactions. The necessary protons to produce the reactions are recoil hydrogen nuclei resulting from (n,p) or neutron-scatter process either in the sample matrix or nearby packaging material. These correction amounts to 0.1 to 1% "apparent nitrogen content." Carbon and nitrogen are also determined accurately by neutron-capture prompt gamma-ray activation analysis (PGAA). PGAA has been used for measuring carbon and nitrogen in different National Bureau of Standards coal reference materials (137).

Gladney, et.al. (138) have PGAA to measure nitrogen and carbon in different environmental materials. Useful, simultaneous and non-destructive analyses with 1-g samples can be achieved at concentration greater than 500 ppm and 10% respectively.

Schofield, et.al. (139) have reported a comparative study for the analysis of carbon, nitrogen and hydrogen in environmental standard reference materials by instrumental combustion analysis and thermal-neutron capture gamma-ray spectrometry. It was suggested that PGAA should be used for quality assurance materials development.

5) IN VIVO NEUTRON ACTIVATION ANALYSIS

Nitrogen is central to the structure of living matter. It is not only present in all amino acids, which form the fundamental proteins for the body, but is also present in such important biological molecules as the DNA of the cell nucleus. Knowledge of nitrogen balance, or of changes in body nitrogen finds useful applications in medicine and biochemistry.

In vivo neutron activation has provided investigators with a new tool for research on body composition in relation to nutritional status.

Whole body nitrogen, an index of body protein, is presently measured in vivo by two methods.

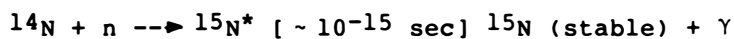
- a) The $^{14}\text{N}(n,2n)^{13}\text{N}$ method.
- b) The $^{14}\text{N}(n,2n)^{13}\text{N}$ prompt capture technique.

Leach, et.al. (140) have described the $^{14}\text{N}(n,2n)^{13}\text{N}$ method in

detail. The body was irradiated with 14 MeV neutrons. This reaction has a threshold of 11.3 MeV and the product, ^{13}N , decays by positron emission to ^{13}C . In the decay of ^{13}N there is no gamma ray characteristic of nitrogen, the only radiation being the 511 KeV annihilation quanta. This non-specificity poses a particular problem to the method. The fast neutron flux used produces (n,2n) reactions in other elements where the reaction threshold energy is sufficiently low, resulting in neutron deficient nuclei that generally decays by β^+ emission. A significant amount of nitrogen-13 was also produced from $^{16}\text{O}(p,\alpha)^{13}\text{N}$ reaction due to the knock-on proton flux. There was some error due to oxygen present in the body. This error was found to contribute 19% to the nitrogen count. This has also been reported elsewhere (141,142).

The Birmingham University Group (143) was the first to use prompt gamma photons from neutron capture for the in vivo measurement of total body nitrogen (TBN).

The following nuclear reaction provides the prompt-gamma radiation:



This reaction is most probable at thermal neutron energies. The lifetime of the compound nucleus $^{15}\text{N}^*$ is of the order of 10^{-15} sec; it de-excites to the ground state by emission of a cascade of gamma photons. In slow neutron capture by ^{14}N , the

total energy available is 10.83 MeV. Approximately, 15% of the de-excitations take place directly to the ground state of ^{15}N . Since no other major body element has a neutron-capture gamma of this energy, it is possible to determine body nitrogen by measuring 10.83 MeV photons.

Varstsky, et.al. (144) have described a facility for the measurement of TBN based on the $^{14}\text{N}(n,\gamma)^{15}\text{N}$ reaction. This system uses an 85 Ci ^{238}Pu - Be neutron source and uses the measurement of neutron capture prompt gamma-rays of body hydrogen as an internal standard. The precision of the method was $\pm 3\%$. The in vivo neutron activation analysis has been reviewed in detail (145,146).

VII. SELECTED RADIOCHEMICAL PROCEDURES

The selected radiochemical procedures for carbon, nitrogen and oxygen are reported as examples. The radiochemical methods for analysis of carbon, nitrogen and oxygen in various matrices are listed in Table IX.

Table IX : Survey of Radiochemical Methods for Analysis of Carbon, Nitrogen and Oxygen in Various Matrices.

Analysis for	Material	Method	Reference
Carbon	Silicon	CPAA	89
Nitrogen	Meteorites	NAA	132
C,N,O	Nickel	CPAA	93
C	Aluminum	CPAA	87
O	Copper	CPAA	108
O	Gallium	CPAA	105
N,C	Silicon	PAA	124
C	Metals	PAA	121
C,N,O	Si, SiC, W, Mo	CPAA	147
N	TiC	NAA	129
C	Cr	PAA	118
C	Fe, Mo	PAA	117
N	Gallium Arsenide	CPAA	104
N	Metals and Semi-Conductors	NAA	136
N	Organic Compounds	CPAA	102
N	Cereals	CPAA	103
N	Atmospheric Aerosols	CPAA	99
C	Coal	PAA	123
N	Air-Dust Samples	PAA	122
N,C,O	Environmental Materials	PGAA	138
O	Uranium	NAA	133
O	Aluminum	PAA	150
N	Aluminum	PAA	151

PROCEDURE 1

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Analysis for : Carbon
Element Separated : Carbon
Reaction : $^{12}\text{C}(\gamma, n)^{11}\text{C}$
Target Material : Ag, Al, Cr, Fe, Mg, Mo, Ni, Si, Ti, W,
Zn, W, Zn, Zr, AlMg, AgZn and ZnMgTe.

Time of Separation : 30 min.
Sensitivity : 10^{-8}g

- 1) One gram of the sample is irradiated.
- 2) After irradiation, remove the surface contamination by etching. Select the etching solution depending on the nature of the sample. The etching solution are given in Table X. After the etching, the sample should be rinsed with water and then with acetone.
- 3) Weigh the sample again.
- 4) The separation is performed by oxidizing carbon to carbon dioxide. The oxidation is performed by fusion in an oxidizing bath. Depending on the sample, select the appropriate reagent for sample dissolution. $\text{Pb}_3\text{O}_4\text{-B}_2\text{O}_3$, $\text{H}_2\text{SO}_4\text{-KIO}_4$ and Alkaline bath are used.

Table X
Etching conditions

Sample	Reagents	Temperature	Duration
Al and AlMg	H ₃ PO ₄ (d=1.7) 75%	100°C	1 min
	H ₂ SO ₄ (d=1.83) 20%		
	HNO ₃ (d=1.38) 5%		
Ag and AgZn	2M HNO ₃	Boiling	1 min
Cr	1M HCl	Boiling	2 min
Fe	30% H ₂ O ₂ + 2% HF	20°C	2 min
Mg	1M HNO ₃ in methanol	20°C	1 min
Mo	H ₂ SO ₄ (d=1.83) 45%	20°C	1 min
	HNO ₃ (d=1.38) 5%		
	H ₂ O 50%		
Ni	3M HNO ₃	Boiling	1 min
Si	HF (d=1.13) 15%	20°C	1 min
	HNO ₃ (d=1.38) 30%		
	H ₂ O 55%		
Ti	3M HF - 3M HNO ₃	20°C	2 min
W	3M HF - 3M HNO ₃	20°C	1 min
Zn	1M HNO ₃	Boiling	1 min
Zn _{1-x} Mg _x Te	3M HNO ₃	20°C	1 min
Zr	HF (d=1.13) 10%	20°C	1 min
	HNO ₃ (d=1.38) 10%		
	H ₂ O 80%		

5) Pb₃O₄-B₂O₃ bath

The sample is introduced in an alumina crucible containing the oxidizing bath of 87.5% Pb₃O₄ and 12.5% B₂O₃ and a piece of iron in order to start the fusion and to provide the carbon carrier. The crucible is heated by a high frequency furnace of 2.5 KW. The CO₂ formed during the fusion was carried by an argon flow of 50 cm³ per minimum. The gas is passed through a cotton filter, then Schutze reagent/I₂O₅ on Silica gel/in order to oxidize the eventually formed CO, and finally through powdered KOH, which absorbs CO₂. The activity is measured using NaI(Tl) detector.

6) H₂SO₄-KIO₄ bath

The composition used for this bath is shown in Table XI. The apparatus used is shown in Figure 2.

The bath and the sample were introduced in the flask A of the apparatus. The flask was then heated to gentle boiling. During the whole operation, the apparatus was swept with air at a flow rate of 0.1 l.min⁻¹. The air stream is passed through an HF-HNO₃-H₂O (2-2-1 in volume) washing bottle (c) for zirconium samples and through a 9M HNO₃ washing bottle (D) in all cases. The gases were then treated on a copper oxide catalyst in a silica tube (length = 20 cm., diameter = 15 mm) at 800°C. This catalyst was prepared by absorbing 40

Table XI
Composition of sulfuric baths

Sample	H ₂ SO ₄ (d=1.83) cm ³	KIO ₄ g	H ₃ PO ₄ * (d=1.7) cm ³	NaF, g
Ag and ZnAg	20	2	-	-
Mg	10	2	20	-
Zr	20	2	-	2

* The H₂SO₄ - H₃PO₄ mixture was preevaporated to approximately 25 cm³ before adding KIO₄, in order to decrease the water concentration of the bath.

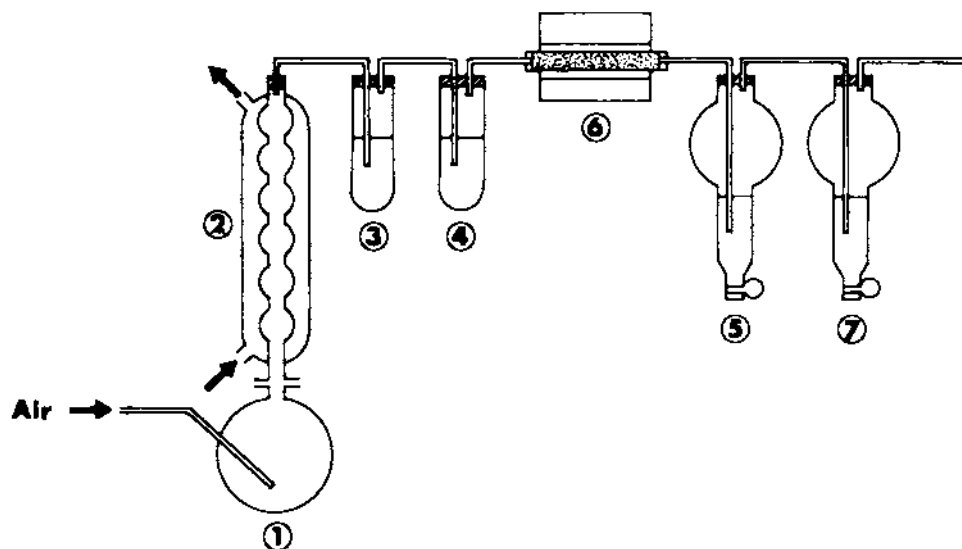


Figure 2 Apparatus for carbon separation in sulfuric baths. 1 - dissolution flask, 2 - refrigerant, 3 - HF - HNO₃ - H₂O (2 - 2 - 1 in volume) absorber (40 cm³), 4 - 9M HNO₃ absorber (40 cm³), 5 -copper oxide on porous alumina in a furnace at 800°C, 6, 7 - 12M KOH absorbers (10 cm³ each).

cm³ on an aqueous solution of 7.6g Cu(NO₃)₂ in 50g of porous alumina of 350 m²/g specific area in balls of 2 mm in diameter. It was treated 2h at 800°C in air before using. The gases passed then through two absorbers containing 10 cm³ 12M KOH. After dissolution of the sample, which lasts between 2 and 5 min., and complete sweeping of the apparatus, the KOH solution is poured into special flasks for radioactivity measurements with a NaI(Tl) detector.

7) Alkaline bath

The composition of baths for each material are shown in Table XII. The fusion was performed in a nickel crucible heated by a gas burner. The sample was introduced in the already molten bath. The heating was adjusted so that to obtain a regular dissolution without spattering. This dissolution lasts between 3 to 5 min. In the case of a bath containing fluorides, it is better to mix it during the fusion with a nickel spatula.

After the end of the dissolution, the crucible was quickly cooled and its content transferred to the flask A of Fig. 3. The crucible was carefully washed with hot water which was also added to flask A. The whole apparatus was then swept with air at 0.1 l/min., and 50 cm³ of 9M HNO₃ was added to a flask A by a peristaltic pump at 20 cm³/min. The solution of flask A was heated to boiling. The air stream passed through

Table XII
Composition of alkaline baths

Sample	NaOH g	NaNO₃, g	NaF, g
Al and Amg	3	15	6
Cr	1.5	5	-
Mo	1	5	-
Si	2	2	-
Ti	6	2	-
W	3	3	-
Zn	5	3	2
Zn_{1-x}Mg_xTe	5	3	2

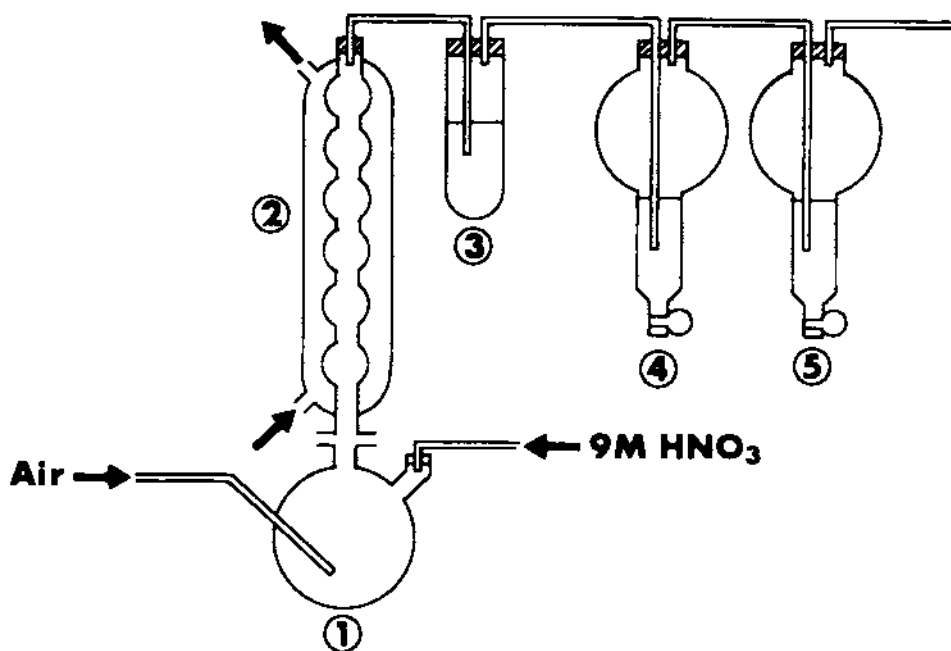


Figure 3 Apparatus for carbon separation after fusion in alkaline baths. 1 - dissolution flask, 2 - refrigerant, 3 - 9M HNO₃, absorber (40 cm³), 4,5 - 12M KOH absorbers (10 cm³ each).

a 9M HNO₃ washing bottle and then through two 12M KOH absorbers. The sample were counted on a NaI(Tl) detector.

PROCEDURE 2

Federoff et.al. J. Radioanal. Nucl. Chem. Articles 88, 45 (1985)

Analysis for : Nitrogen and Carbon
Element Separated : Nitrogen and Carbon
Target Material : Silicon
Reaction : $^{14}\text{N}(\gamma, n)^{13}\text{N}$ and $^{12}\text{C}(\gamma, n)^{11}\text{C}$
Time for Separation : 20 min

- 1) The irradiated sample of Silicon was etched twice for 15 seconds in an HF-HNO₃ (1/1 concentrated acid in volume) in order to remove the superficial contamination.
- 2) Introduce the sample in 30g of fused dehydrated KOH contained in a nickel crucible disposed in a quartz tube at 500°C (Fig. 4).
- 3) Sweep the apparatus by argon at a flow rate of 0.3 l/min.
- 4) Pass the gas through granulated copper oxide and then through copper oxide on porous alumina at 900°C.
- 5) Carbon dioxide was absorbed in 8 cm³ 12 M KOH.
- 6) Nitrogen is absorbed on Titanium sponge at 900°C.
- 7) The gas is passed for 10 min.

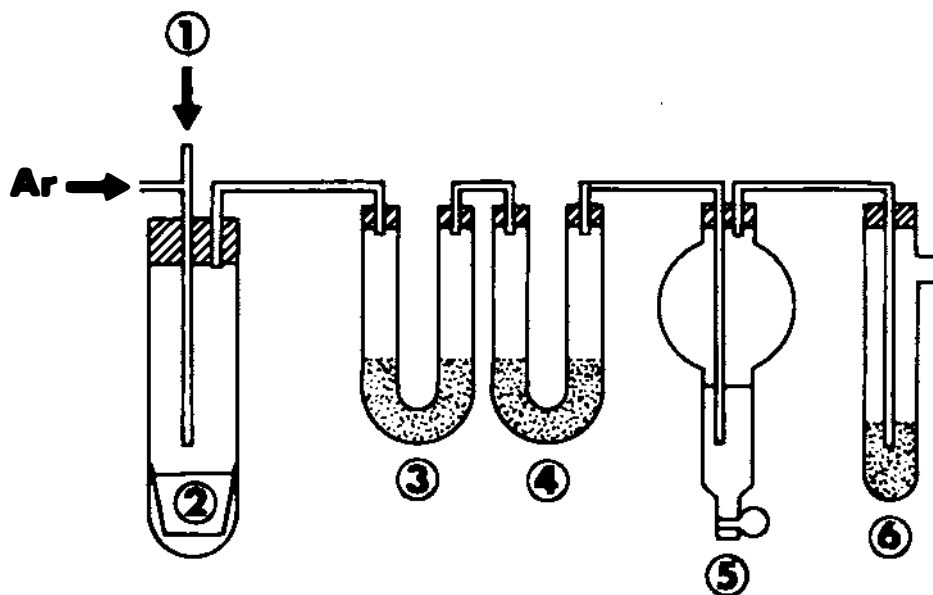


Figure 4 Apparatus for the radiochemical separation of nitrogen and carbon from silicon. 1 - sample introduction, 2 - KOH in nickel crucible at 500°C, 3 - granulated CuO at 900°C, 4 - CuO on porous alumina at 900°C, 5 - 12M KOH, 6 - titanium sponge at 900°C.

- 8) The KOH solution and the titanium were measured for 511 KeV annihilation radiation by two NaI(Tl) detectors couple to a coincidence unit and a multichannel analyzer.

PROCEDURE 3

Shikano et.al. J. Radioanal. and Nucl. Chem. Articles 91, 81 (1985)

Analysis for : Oxygen
Element Separated : Fluoride
Target Material : Gallium Arsenide
Reaction : $^{16}\text{O}(^3\text{He},p)^{18}\text{F}$
Time for Separation : 2 hrs.
Decontamination factor of ^{76}Br : 10^6

- 1) Gallium arsenide pieces are etched in a mixture of sulfuric acid, hydrogen peroxide and bidistilled water in the ratio of 3:1:1 at 70°C for one minute.
- 2) The etched sample is dissolved in a solution of nitric acid and hydrochloric acids containing 12 mmol sodium fluoride.
- 3) Fluoride solution is transferred to the distillation flask connected to a steam generator. Add 150 ml of perchloric acid and 5g silicon dioxide. After the temperature in the flask reached 150°C, steam was introduced and the temperature was kept at 150-155°C. The distillate of 150 ml volume was collected within 60 minutes.
- 4) To the distillate, add 0.04 mmol of carriers of gallium arsenide, ammonium bromide and hydrozine. Adjust the pH to 4 and distill again.

- 5) To the second distillate, add 0.04 mmol of carriers of gallium arsenide, ammonium bromide, and hydrazine.
- 6) Add 1 mmol of lanthanum solution.
- 7) Adjust the pH to 5-6.
- 8) Digest on a water bath at 80-85°C for 5 min.
- 9) Cool with ice water.
- 10) Centrifuge after decanting.
- 11) Count the LaF₃ precipitate.
- 12) Measure the activity using a single-channel analyzer with a well type NaI(Tl) detector, or a 4096 channel pulse height analyzer with a Ge(Li) detector.

PROCEDURE 4

Lee, et.al. Analytical Chemistry, 42, 994 (1970)

Analysis for	:	Oxygen
Target Material	:	Copper foil
Element Separated	:	Fluoride
Element Concentration Determined	:	Oxygen
Reaction	:	$^{16}\text{O}(^3\text{He,p})^{18}\text{F}$

- 1) Copper foils after the irradiation are transferred to a 15 ml centrifuge tube.
- 2) Add 2 ml of 4 M HNO_3 .
- 3) Add 10.0 mg of F^- carrier and 2 mg of scavenger carriers, i.e. Zn^{2+} and Ga^{3+} as nitrates.
- 4) After the copper is dissolved, NaOH is added to adjust the pH to 9.0.
- 5) Zn^{2+} , Cu^{2+} and Ga^{3+} are precipitated as hydroxide.
- 6) The hydroxide precipitation is repeated once more with a few mgs of scavenger carrier.
- 7) The solution F^- is then adjusted to about pH of 5 with an acetate buffer solution and an excess of $\text{Pb}(\text{NO}_3)_2$ is added to precipitate PbF_2 .

- 8) The PbF_2 precipitate is washed with distilled water.
- 9) Mount the precipitate on filter paper with alcohol.
- 10) The sample yield of F^- is determined gravimetrically by re-precipitation of the F^- as PbClF .

PROCEDURE 5

Goethals, et.al. Analytica Chimica

Acta 108, 367 (1979)

Analysis for : Carbon
Element Separated : Nitrogen
Time of Separation : 30 min.
Reaction : $^{12}\text{C}(d,n)^{13}\text{N}$
Target Material : Aluminum

- 1) Remove surface contamination of the sample by etching with a 7:2:1 (V/V) mixture of concentrated phosphoric, sulphuric and nitric acids for five minutes at 80°C.
- 2) Place sample in a conventional steam distillation apparatus and dissolves with slight heating (50-60°C) in 25 ml of 10M NaOH.
- 3) After the dissolution, raise the temperature to 110-120°C and introduce the steam.
- 4) Collect 70 ml of distillate for quantitative recovery.

PROCEDURE 6

K. Strijckmans, et.al. Fresenius

Z. Anal. Chem. 303, 106 (1980)

Analysis for : Boron
Target Material : Nickel
Element Separated : Carbon as CO₂
Reaction : $^{10}\text{B}(\text{d},\text{n})^{11}\text{C}$
Type of Bombardment : Deuteron

- 1) The nickel sample is wrapped in a copper foil and placed in an alumina combustion tray.
- 2) The combustion tray is introduced in the SiC furnace with a maximum working temperature of 1500°C under a 0.2/min argon flow.
- 3) The metals are melted for five minutes at a 0.1 bar overpressure to avoid reaction with atmospheric oxygen.
- 4) For 15 minutes an oxygen flow of 0.2 l/min is maintained at an under pressure of 0.1 bar, to avoid leakage.
- 5) The gas flow passes through a condenser, Schutze reagent (I₂O₅) to oxidize carbon monoxide to carbon dioxide and through two absorption vessel containing 0.5 M NaOH to trap carbon dioxide.

PROCEDURE 7

Strijckmans et.al. Fresenius

Z. Anal. Chem. 303, 106 (1980)

Analysis for : Carbon
Element Separated : Nitrogen
Target Material : Nickel
Reaction : $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$
Type of Bombardment : Deuteron

- 1) The nickel sample is placed in 25 ml solution of 140 mg l^{-1} $(\text{NH}_4)_2\text{PtCl}_6$ in 6 M HCl. The $(\text{NH}_4)_2\text{PtCl}_6$ is added to speed up the dissolution.
- 2) The sample is dissolved by heating.
- 3) The total volume is adjusted to 50 ml.
- 4) The solution is transferred to steam distillation apparatus and neutralized by adding 40 ml of 7.5 M NaOH.
- 5) 75 ml of distillate is collected in 25 ml of 6M HCl.

PROCEDURE 8

Lavrukhina et.al. J. Radioanal. Nucl. Chem. Articles 88, 45 (1985)

Analysis for : Nitrogen
Element Separated : Carbon
Target Material : Meteorites
Reaction : $^{14}\text{N}(n,p)^{14}\text{C}$
Detection limit : 0.001 μg
Yield : 99 \pm 1%

- 1) The meteorites and terrestrial samples are crushed into 100-200 mg pieces. The sample is crushed in a spherical sampling mill under argon.
- 2) The process of isolation is performed in a vacuum system. The sample is mixed with oxidizing flux and carrier and placed in a quartz reactor. The oxidizing flux is $\text{PbCrO}_4 + \text{K}_2\text{CrO}_4$ (1:10) or V_2O_5 . Marble is used as carrier.
- 3) The reactor is evacuated and the mixture is heated under vacuum at 200°C for three hours for the isolation of occluded gases.
- 4) The reactor is filled with oxygen, containing no CO_2 . The fusion of the samples with $\text{PbCrO}_4 + \text{K}_2\text{CrO}_4$ is effected at 1100°C. Whereas the sample is fused at 800°C with V_2O_5 .

- 5) The isolated gases are passed through CuO ($T=500^{\circ}\text{C}$) for the oxidation of CO to CO₂.

- 6) The CO₂ is trapped at liquid nitrogen temperature.

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