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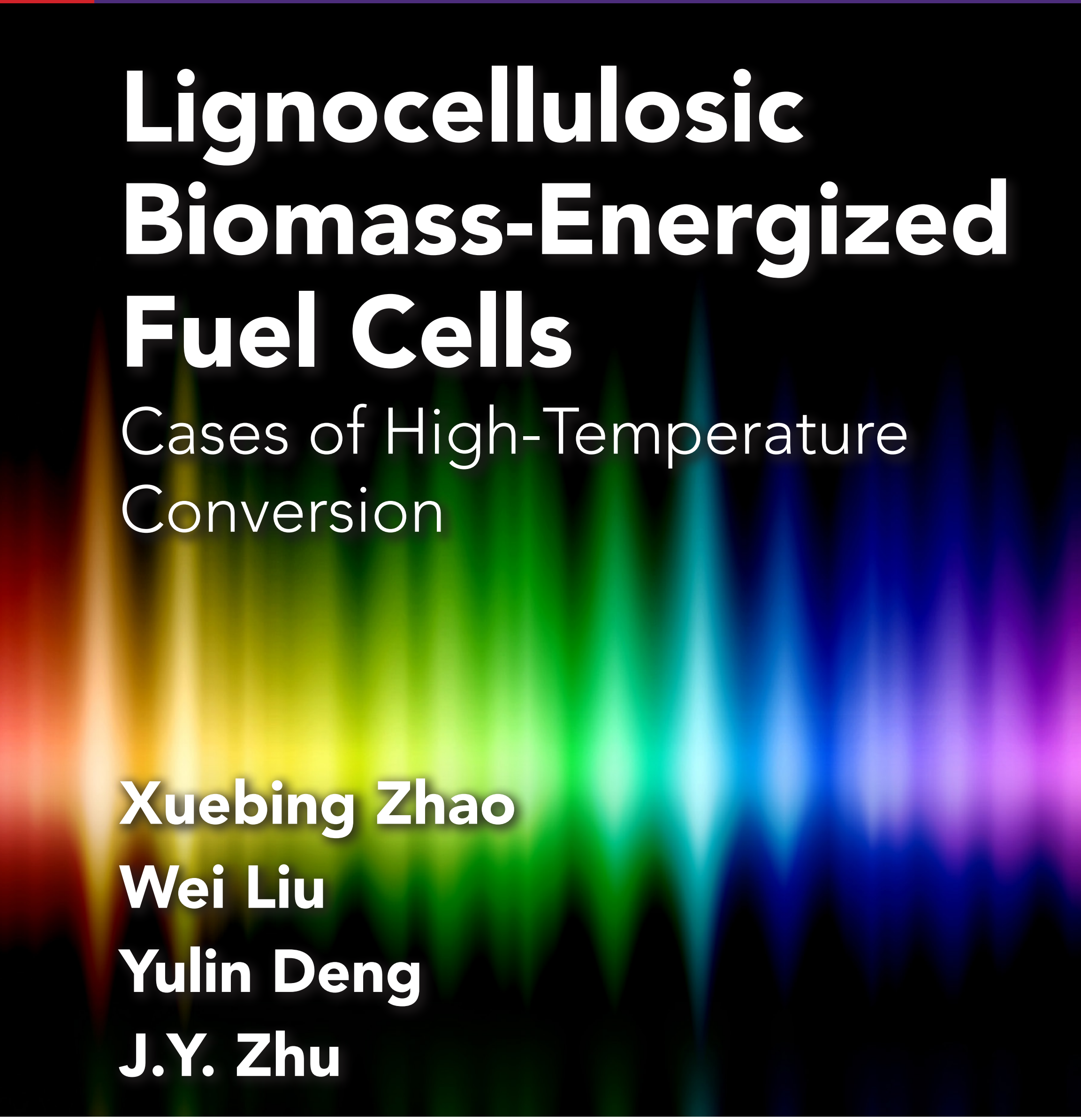

Lignocellulosic Biomass-Energized Fuel Cells

Cases of High-Temperature Conversion

Xuebing Zhao
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ABSTRACT

Fuel cell technology has attracted great interest in recent decades. However, progress in lignocellulosic biomass-energized fuel cells has been slow. This is because that lignocellulosic biomass generally cannot be directly used for electricity generation in a fuel cell with high efficiency. As a renewable resource available in large quantities in many regions of the world, lignocellulosic biomass can be a promising feedstock for sustainable electricity production using fuel cell technologies. In this monograph, we focus on the electricity generation in fuel cells that are operated at high temperatures with high efficiency using lignocellulosic biomass-derived fuels. More specifically, we discussed biomass conversion coupled solid oxide fuel cell and direct carbon fuel cell, the state of the art in technology development, the challenges and the perspectives on future development.

KEYWORDS

direct carbon fuel cells, fuel cells, gasification, lignocelluloses, solid oxide fuel cells

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CHAPTER 1

INTRODUCTION

Demand for electricity has been growing continuously as a result of increasing global populations and the development of third-world nations [1]. The importance of electric energy to modern society can be seen from many electricity-driven daily technologies or gadgets that contribute to our life quality. Electricity has been mainly produced through turbines [2] driven by hydropower at remote dams or hot gases or steam from combustion of fossil fuels. For example, the amount of electricity produced from the combustion of coal and natural gas in the United States in 2014 was 39 percent and 27 percent, respectively (U.S. Energy Information Administration, http://www.eia.gov/tools/faqs/faq.cfm?id=427&t=3, last accessed September 2015). Owing to the concerns over the air emissions of greenhouse gases and particulates and pollutants from combustion of fossil fuels, conventional fossil-fuel-driven power plants are facing increased challenges. Thus, sustainable and environmentally friendly pathways for alternative electricity production using renewable resources need to be developed.

Lignocellulosic biomass as one of the major renewable resources can be sustainably produced in large quantities and has been used for energy production through combustion in many regions around the world. A recent study suggest that approximately 1.3 billion dry tonnes of lignocellulosic biomass can be potentially produced annually in the United States alone [3]. This is equivalent to 2 trillion kWh of electricity (assuming 30 percent conversion efficiency from thermal energy to electricity), or approximately 50 percent of the total U.S. electricity production in 2014, which illustrates the potential impact of using lignocellulosic biomass for electricity production. Lignocellulosic biomass has been used to produce electricity commercially using steam or gas turbines through combustion or gasification (Figure 1.1). However, distributed operation at relatively small scale is preferred because of the low energy density of lignocelluloses.

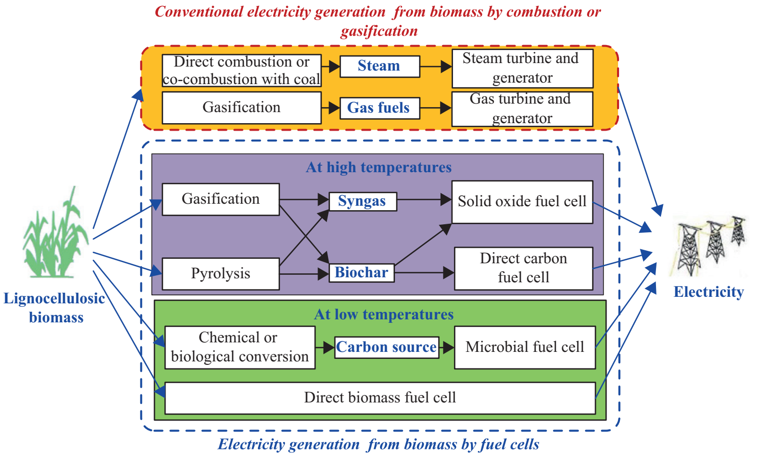

Figure 1.1. Different pathways for electricity production from lignocellulosic biomass.

Fuel cells are clean, are portable, and can be an alternative electricity production technology for a variety of applications. Fuel cells using fuels such as syngas, biogas, or biochar derived from lignocellulosic biomass by thermal–chemical or biological conversions or indirect biomass fuel cells (IDBFC) have been intensively studied in recent years. On the other hand, direct biomass fuel cells is a new technology that uses biomass directly to generate electricity. It has not been well developed, with only limited research being carried out in the laboratory. Therefore, in this monograph, we examine a variety of IDBFC with the focus on solid oxide fuel cells and direct carbon fuel cells that are operated at high temperatures but with high efficiency. Our aim is to present some basic principles, technological challenges, and future developments of these technologies.

CHAPTER 2

OVERVIEW OF THE CHARACTERISTICS OF LIGNOCELLULOSIC BIOMASS

Lignocellulosic biomass including biomass from forest and agricultural land such as harvesting forest residues, short rotation woods, agricultural residues (e.g., corn stover and straw), and energy crops (e.g., switchgrass) is renewable, is carbon neutral in terms of reduction of CO_2 emission, and can be sustainably produced in large quantities [4]. The elemental compositions of lignocelluloses are C, H, O, N, P, and S. The major elements C, H, and O mainly come from cellulose, hemicellulose, and lignin; N, P, and S mainly come from the minor components such as protein. Dry lignocelluloses commonly have a C content of approximately 50 percent, lower than that of coal (75 to 90 percent); an O content of about 45 percent, higher than that of coal (<20 percent); and, therefore, has a lower heating value of approximately 20 MJ kg^{-1} [5, 6], lower than that of bituminous coal of 26 MJ kg^{-1} [5, 6]. As shown by the *van Krevelen diagram* in Figure 2.1, lignocelluloses have higher H:C and O:C ratios than those of fossil fuels. Moisture content of freshly cut wood is approximately 50 percent [7].

The major components of lignocelluloses, that is, cellulose, hemicellulose, and lignin, account for more than 80 percent of the total dry weight [8, 9]. However, the proportions of the three major components differ among different species of plants and different parts of the same plant [10]. Lignin is an aromatic polymer composed of three basic monomeric units: *p*-hydroxyphenyls (H), guaicyls (G), and syringyls (S), which vary between species and cell tissue type [11]. Lignin has higher carbon content than cellulose and hemicelluloses, thus having a higher heating value. The heating value of woody biomass is higher than that of herbaceous biomass

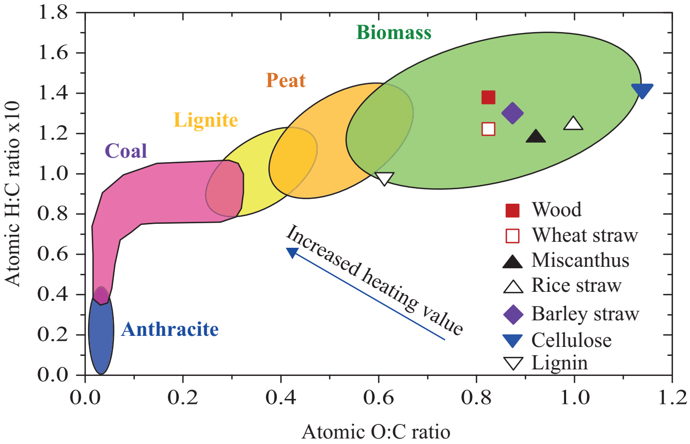

Figure 2.1. van Krevelen diagram of several solid fuels.

Source: Adapted from McKendry [5].

because of the higher lignin content of woody materials. Cellulose is a polysaccharide consisting of a linear chain of several hundreds to more than 10,000 β (1 → 4) linked D-glucose units. Hemicelluloses are heteropolymers of several monosaccharide groups and uronic acid groups [12]. Details of the structures of cellulose, hemicelluloses, and lignin can be found in literature [12–14].

CHAPTER 3

INDIRECT BIOMASS FUEL CELLS AT HIGH TEMPERATURES

Indirect lignocellulosic biomass fuel cells refer to fuel cell technologies that use biomass-derived fuels for electricity production. A first-step conversion of biomass is required. There are generally two types of indirect biomass fuel cells operated at high temperatures with high efficiency: solid oxide fuel cells (SOFC) and direct carbon fuel cells (DCFC) (Figure 3.1). In a biomass-fueled SOFC, lignocelluloses are first converted to syngas or biogas containing H_2, CO, and CH_4 using thermal–chemical or biological conversions, and then the fuel gasses are subsequently converted to electricity by electrochemical oxidation using air (oxygen). In a biomass-fueled DCFC, lignocelluloses are first carbonized, usually by pyrolysis, and the obtained biochar is used to produce electricity by direct electrochemical oxidation of the carbon. Several parameters were used to characterize the performance of a fuel cell. Open-circuit voltage (OCV) is the maximum voltage available from a fuel cell at zero current, V or mV. Current density is the current per unit area of electrochemical-active electrode (anode), mA cm^{-2}. Power density is power output per unit area or volume, mW cm^{-2} (mW m^{-2} or W cm^{-2}) or mW cm^{-3}. Coulombic efficiency, also called Faradic efficiency, is the fraction of total produced coulombs to the theoretical amount of coulombs available from the fuel, percent. These parameters will be exclusively used throughout this monograph.

3.1 SOLID OXIDE FUEL CELLS

3.1.1 PRINCIPLES OF SOFC

SOFC are a promising technology for efficient electric energy generation from hydrogen, natural gas, syngas, methane, or other similar light hydrocarbons [15, 16]. Intensive research progress has been made on SOFC since the 1950s, including the fundamentals [17], technology advances [18–20], electrode materials and modifications [21–28], cell design [29, 30], and applications [15]. However, work on SOFC using biomass has been limited.

The working principles of SOFC are graphically shown in Figure 3.1. A SOFC essentially consists of two porous electrodes separated by a dense, oxygen-ion-conducting electrolyte [31]. The fuels, such as H_2, CO, CH_4, or their mixtures, enter the anode chamber where they are dispersed by the anode over its interface with an electrolyte. The anode further catalyzes the electrochemical reactions to release electrons from fuel molecules and conducts the electrons to an external circuit. In the cathode chamber, oxygen molecules are distributed at the cathode interface with the solid electrolyte and reduced by electrons from the external circuit, thus producing oxide ions. Oxide ions diffuse through the solid electrolyte to the anode to react with fuel molecules forming H_2O or CO_2, depending on the fuel type [32]. The Gibbs free energy (or chemical potential energy) of the global reaction of fuel and oxidizer is converted to electricity and heat. The anode half-cell oxidation reactions are

$$\mathrm{H_2\,(g) + O^{2-}\,(el) \leftrightarrow H_2O\,(g) + 2\,e^-\,(a)} \tag{3.1}$$

$$\mathrm{CO\,(g) + O^{2-}\,(el) \leftrightarrow CO_2\,(g) + 2\,e^-\,(a)} \tag{3.2}$$

$$\mathrm{CH_4\,(g) + 4O^{2-}\,(el) \leftrightarrow 2H_2O\,(g) + CO_2\,(g) + 8\,e^-\,(a)} \tag{3.3}$$

and the cathode half-cell reduction reaction is

$$\mathrm{O_2\,(g) + 4\,e^-\,(a) \leftrightarrow 2\,O^{2-}\,(el)} \tag{3.4}$$

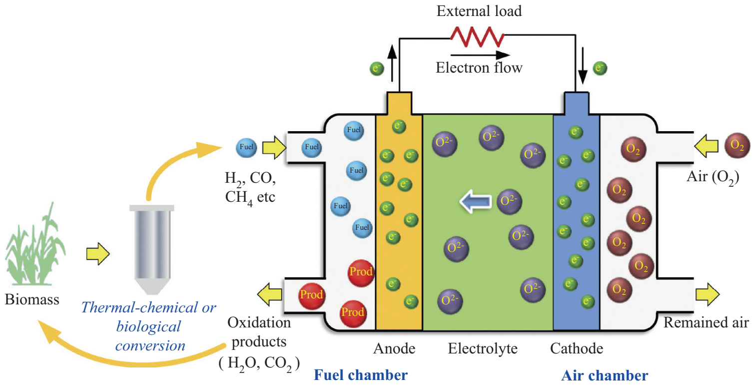

Figure 3.1. Working principle of biomass-fueled SOFC.

Corresponding global reactions are

$$H_2\,(g) + 1/2O_2\,(g) \leftrightarrow H_2O\,(g) \tag{3.5}$$

$$CO\,(g) + 1/2O_2\,(g) \leftrightarrow CO_2\,(g) \tag{3.6}$$

$$CH_4\,(g) + 2\,O_2\,(g) \leftrightarrow 2H_2O\,(g) + CO_2\,(g) \tag{3.7}$$

Therefore, for a given fuel composition, the theoretical maximum potential difference that the cell could achieve, namely, the reversible cell potential (the Nernst potential) E_{rev} between the fuel and oxidizer, can be calculated by the Nernst equation [17]:

$$E_{\mathrm{rev}} = -\frac{\Delta_r G}{n_e F} = -\frac{\Delta_r G^{\circ}}{n_e F} = -\frac{RT}{n_e F}\,\mathrm{In}\left(\prod_i p_i^{v_i}\right) \tag{3.8}$$

where $\Delta_r G$ and ΔG^0 are the temperature-dependent Gibbs free energy of reaction and standard-state free energy change, respectively, associated with the global oxidation reaction; n_e is the number of transferred electrons in mol; F is the Faraday constant (96,485.34 C mol^{-1}); R is the universal gas constant (8.314 J K^{-1} mol^{-1}); T is the thermodynamic temperature; p_i is the partial pressure of the species i in atmospheres; and v_i is the stoichiometric coefficient in the global reaction. However, the actual power output of SOFC is influenced by the actual fuel compositions. Pure hydrogen usually gives the highest power density, while the presence of inert gas such as H_2O, N_2, and CO_2 can dramatically decrease the power output (Figure 3.2a). The actual power output is also affected by many other factors, such as material of the electrode, type of electrolyte, operation parameter, impurity concentration, and cell design. The actual cell voltage is affected by losses from various electrochemical processes, mainly activation polarization, ohmic polarization, and concentration polarization [27], as expressed by the equation

$$V_{\mathrm{act}} = E_{\mathrm{rev}} - IR_{\mathrm{o}} - \eta_{\mathrm{cathode}} - \eta_{\mathrm{anode}} \tag{3.9}$$

where E_{rev} is the Nernst potential of the reactants, I is the current through the cell, R_{o} is the ohmic resistance of the cell, and η_{cathode} and η_{anode} are the cathodic and anodic polarization losses, respectively. Activation polarization depends on chemical reactions in the anode and cathode, and it is the combination of the reaction rate and the electron or ion transfer [27]. High operating temperatures of SOFC cause extremely fast reaction kinetics;

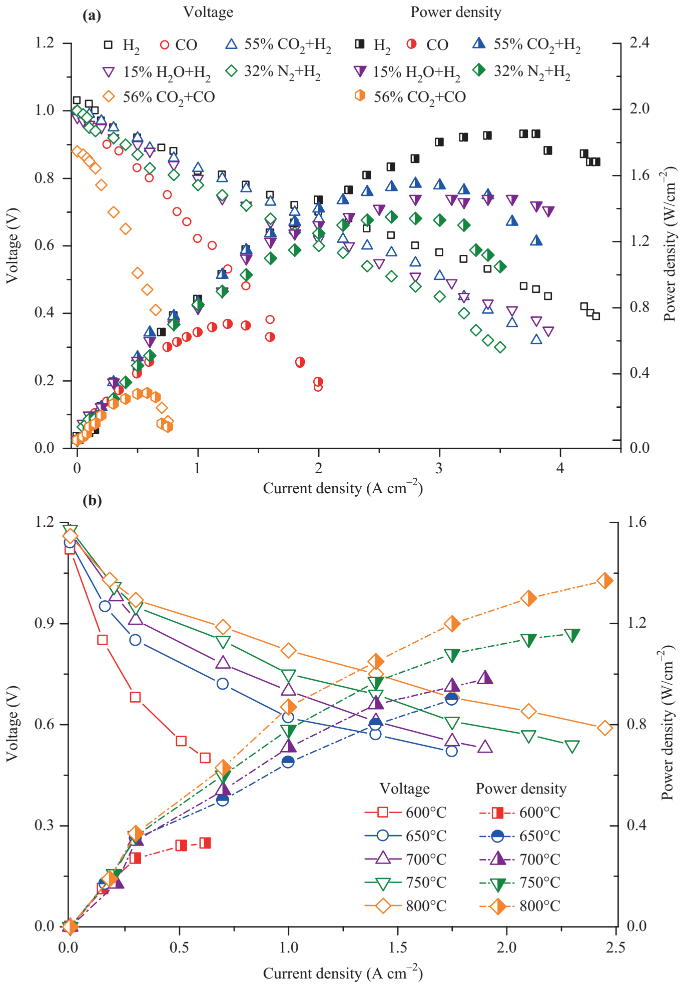

Figure 3.2. Effects of fuel and operating temperature on power density of SOFC. (a) effects of fuel (data from Jiang and Virkar [36]); (b) effects of operating temperature with hydrogen as the fuel and air as the oxidant (data from Minh [19]).

hence, the voltage drop because of the activation polarization is small [33]. The concentration polarization arises because of the transportation of the reactants from their respective streams to the fuel cell, while ohmic polarization is a combination of resistances to electron and ion transport throughout the fuel cell [27, 34]. Increasing operating temperature can reduce the ohmic polarization, thus increasing the power density of SOFC (Figure 3.2b).

3.1.2 ELECTRODE AND ELECTROLYTE MATERIALS FOR SOFC

The electrode materials for SOFC must have catalytic activity, electronic conductivity, chemical stability and compatibility, high morphological stability without sintering, mechanical compatibility with the electrolyte and interconnect, and low cost of fabrication [35]. The anode catalyzes electrochemical oxidation of fuel(s) and conducts electrons released during oxidation to reach the current collector. In a typical anode-supported SOFC, the anode support is a Ni–yttria-stabilized zirconia (YSZ) cermet with thickness between 0.5 and 2 mm [36]. This composite is electron conductive (because of Ni) and also ion conductive (because of YSZ) [37]. Ni anode materials show good electrochemical activity for hydrogen oxidation with high electrical conductivity at SOFC operating temperatures [38]. However, for hydrocarbon fuels such as CH_4, Ni can also actively catalyze carbon formation by reforming reactions (e.g., $CH_4 \rightarrow C + 2H_2$) [39, 40]. The deposit carbon can block the reaction sites and gas diffusion pores and degrade cell performance [40, 41]. Modifications of the Ni–YSZ anode by adding a small amount of metals such as Au, Rh, or Ru [18] or developing anodes without Ni can reduce the propensity for carbon deposits. For example, when using a mixture of CH_4 and CO_2 as fuel, porous Ni–Gd-doped ceria (GDC) cathode/thin porous GDC electrolyte (50 μm)/porous $SrRuO_3$–GDC anode system [42], Ru–GDC cathode/porous GDC electrolyte/Ru or $SrRuO_3$–GDC anode system [40], and Ni−GDC cathode/porous GDC electrolyte/Cu−GDC anode system [43] had no significant carbon deposition. Another issue associated with Ni anodes is low tolerance to impurities such as sulfur that can deactivate catalytic activity of Ni. Thus there would be a potentially significant impediment to the direct use of untreated syngas [34] or biogas produced from lignocellulosic biomass because of impurities. Some other materials, such as rare-earth-doped CeO_2, perovskite, pyrochlore, and tungsten bronze, have been developed as SOFC anodes [25]. However, the catalytic and electronic conductivities of these anodes are low and need improvement.

One of the basic functions of cathode is to electrically catalyze oxygen reduction on its surface, and the electrocatalytic activity of a cathode is highly dependent on the materials used and its microstructure features, such as porosity, grain size, connectivity between grains, and its adhesion with other SOFC components [44]. The most common type of cathode material for SOFC is perovskite [45]. A perovskite-type oxide has the general formula of ABO_3, in which A and B are cations with a total charge of

+6. For most of the perovskite-type cathodes in SOFC, the A-site cation is usually a mixture of rare and alkaline earth metals (such as La and Sr, Ca, or Ba), while the B-site cation is a reducible transition metal such as Mn, Fe, Co, or Ni (or a mixture thereof) [46]. The frequently used materials for cathode are a porous mixture of lanthanum–strontium-doped manganite, $La_{1-x}Sr_xMnO_{3-\delta}$ (LSM), and YSZ or a porous mixture of lanthanum–strontium-doped cobaltite, $La_{1-x}Sr_xCoO_{3-\delta}$ (LSC), and Sm-doped CeO_2 (SDC), as listed in Table 3.1 [36]. Lanthanum strontium cobalt iron oxide or its mixture with SDC is also used. However, high operating temperatures (800°C to 1,000°C) are necessary for these cathodes to effectively catalyze the electrochemical reduction of oxygen. Thus, great efforts have been devoted to the development of intermediate-temperature (500°C to 700°C) SOFC. Alternative cathode materials, therefore, need to be developed to reduce the overpotentials, particularly at temperatures below 700°C. $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O3_{-\delta}$ (BSCF) was developed as a new cathode material for reduced-temperature SOFC operation. High power densities (1,010 and 402 mW cm^{-2} at 600°C and 500°C, respectively) were obtained using humidified hydrogen as fuel when BSCF was incorporated into a thin-film-doped ceria [47].

Electrolyte is an important component of SOFC. It conducts oxygen ions from cathode to anode. Therefore, materials used for electrolyte must have a high oxygen ion conductivity (i.e., >0.1 S cm^{-1}) over a wide range of oxygen partial pressure [18]. Apart from high ionic conductivity, the electrolyte must be electrically insulating to avoid leakage of electrons and impervious to gas flow to prevent gas leakage. The electrolytes must also have good chemical and thermal stability because SOFC are always conducted with oxidant at high temperatures. The widely used electrolyte is YSZ, in which yttria is added to stabilize the conductive cubic fluorite phase and to increase the concentration of oxygen vacancies, thus increasing the ionic conductivity [48]. The optimum addition of yttria is 8 mol percent to obtain the highest conductivity. Further increasing yttria concentration leads to a reduction in defect mobility and thus conductivity. Another promising dopant for zirconia is scandia, which has been reported to have the highest conductivity in the zirconia-based oxide systems [49]. Scandia-stabilized zirconia (ScSZ) also has excellent stability in oxidizing and reducing environments with better long-term stability than YSZ. However, ScSZ has not been widely used because of the lack of conductive stability when it is aged at a high temperature and high cost of scandia [50]. Some other electrolytes have also been developed, such as rare-earth-doped ceria and lanthanum strontium gallium magnesium oxide [51]. GDC and cerium gadolinium oxide (CGO) show

Table 3.1. Some reported works on SOFC using fuels that can be potentially produced from lignocellulosic biomass

Fuel(s)	Fuel can be potentially produced from biomass by	Anode	Electrolyte	Cathode	Temperature (°C)	Power density (mW cm^{-2})	Reference
Pure H_2	Gasification or bioconversion	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~1,800	[36]
H_2 + 15% N_2	Gasification or bioconversion	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~1,500	[36]
H_2 + 15% H_2O	Gasification or bioconversion	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~1,500	[36]
H_2 + 15% CO_2	Gasification or bioconversion	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~1,400	[36]
H_2 + 14% CO	Gasification	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~1,700	[36]
H_2 + 3% H_2O	Gasification or bioconversion	Ni – YSZ	YSZ	LSCF–GDC	600–800	~200–1,440	[219]
H_2 + 3% H_2O	Gasification or bioconversion	NiO–SDC with Ru–CeO_2 as the catalyst	SDC	LSCF–SDC	400–600	~100–1,050	[220]
Pure H_2	Gasification or bioconversion	Cu–CeO_2–YSZ	YSZ	LSM–YSZ	700, 800	220, 310	[37]

(*Continued*)

Table 3.1. Some reported works on SOFC with fuels that can be potentially produced from lignocellulosic biomass (Continued)

Fuel(s)	Fuel can be potentially produced from biomass by	Anode	Electrolyte	Cathode	Temperature (°C)	Power density (mW cm^{-2})	Reference
Pure H_2	Gasification or bioconversion	Ni–YSZ	Co–YSZ	LSM–YSZ	800	~1,200	[221]
Pure H_2	Gasification or bioconversion	Ni–YSZ	Mn–YSZ	LSM–YSZ	800	~1,000	[221]
H_2 + 3% H_2O	Gasification or bioconversion	Pd-SDC/ SYT	YSZ	Pt–YSZ	800–900	~160–280	[222]
Pure CO	Gasification	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~700	[36]
CO + 56% CO_2	Gasification	Ni + YSZ	YSZ–SDC	LSC + SDC	800	~300	[36]
CO + 3% H_2O	Gasification	BaO/Ni–YSZ	YSZ	SDC–LSCF	750	700	[55]
96% CO + 1% H_2 + 3% impurity gas	Gasification	BaO/Ni–YSZ	YSZ	SDC–LSCF	850	1,080	[55]
CH_4	Gasification or bioconversion	Ni–YSZ	YSZ	LSCF–GDC	600–800	~200–1,270	[219]
CH_4	Gasification or bioconversion	Ru–Ni–GDC	GDC	SSC	600	750	[54]

CH_4 + 3% H_2O	Gasification or bioconversion	Pd-SDC/SYT	YSZ	Pt–YSZ	800–900	~130–220	[222]
CH_4 + 3% H_2O	Gasification or bioconversion	Ni–YSZ	YSZ	LSM–YSZ	800	960	[223]
CH_4 + 70% CO_2	Gasification or bioconversion	Ni–GDC	GDC	LSCF	800	93	[39]
CH_4 + 60% CO_2	Gasification or bioconversion	Ni–GDC	GDC	LSCF	800	181	[39]
CH_4 + 50% CO_2	Gasification or bioconversion	Ni–GDC	GDC	LSCF	800	280	[39]
CH_4 + 40% CO_2	Gasification or bioconversion	Ni–GDC	GDC	LSCF	800	334	[39]

GDC, gadolinia-doped ceria; LSC, lanthanum strontium cobaltite; LSCF, lanthanum strontium cobalt ferrite; LSM, lanthanum strontium manganite; SDC, samaria-doped ceria; SSC, samarium strontium cobaltite; SYT, yttrium-doped $SrTiO_{3-\delta}$; YSZ, yttria-stabilized zirconia.

higher conductivity than YSZ and ScSZ, especially at low temperatures. However, low electronic conduction at low partial pressure of oxygen and weak mechanical stability, as well as the high price of Gd, have limited the application of GDC [52]. Progress in developing new materials and modifications of electrolyte has been made in recent years. The stability of electrolyte, however, still needs improvement, and the cost of the materials is still prohibitive.

3.1.3 BIOMASS-DERIVED FUELS FOR SOFC

Hydrogen and carbon monoxide are typical fuels for SOFC and can be consumed directly in electrochemical reactions. Typically, these gases can be produced via coal gasification or methane steam reforming. Hydrogen can also be generated by electrolysis of water at different scales [53]. Other gas fuels such as CH_4 and light hydrocarbons (e.g., C_2H_6 [54], C_3H_8 [54, 55], and *n*-C_4H_{10} [37]) have also been studied. Some liquid fuels (at room temperature), such as iso-C_8H_{18} [56, 57], toluene, *n*-decane, and synthetic diesel, have been directly used in power generation without reforming [58]. Studies on electricity generation from fuels produced from lignocellulosic biomass are summarized in Table 3.1. The presence of inert gases such as H_2O, CO_2, and N_2 have negative effects on power output (Figure 3.2a). The dilution of fuel gas by these inert gases increased the anodic concentration polarization. Diluent with higher molecular weight have more serious negative impact on power output [36]. Three types of gas fuels produced from lignocellulosic biomass, namely biohydrogen, biogas (including hythane), and syngas, are discussed in the following sections.

3.1.3.1 Biohydrogen

The term biohydrogen here refers to hydrogen produced from biological conversion of biomass to differentiate from hydrogen produced from thermochemical conversion. Biohydrogen production using microorganisms can be classified into five different processes: (a) direct biophotolysis, (b) indirect biophotolysis, (c) biological water-gas shift reaction, (d) photofermentation, and (e) dark fermentation [59]. Currently, dark fermentation is the most feasible process for biohydrogen production from renewable biomass because of its higher rate of hydrogen evolution in the absence of any light sources as well as the versatility of the substrates used [60].

The widely used substrate is glucose. The theoretical hydrogen yield from glucose is dependent on the fermentation end products. Maximum hydrogen yields are 4 and 2 mol mol^{-1} glucose when end products are acetic acid and butyric acid, respectively, as shown in Equations 3.10 and 3.11, while coproduction of acetic acid with butyric acid or with ethanol produced maximum 2.5 or 2 mol (hydrogen) mol^{-1} (glucose). However, actual hydrogen yield is still much lower than theoretical yield. One reason is that actual yields are reduced by hydrogen recycling because of the presence of one or more uptake hydrogenases, which consume a portion of the produced hydrogen [61]. Another factor influencing hydrogen yield is the operation conditions in dark fermentation, which is closely associated with bacterial metabolic activities. Temperature, pH, hydraulic retention time, gas partial pressure, substrate concentration, and soluble metabolic profile had significant influence on hydrogen production [60–63]. Other substrates, including pure monosaccharides (xylose, arabinose) [64–66], biomass hydrolysate [67–70], and pretreated lignocellulosic biomass [71–74], have also been used for hydrogen production. Using biomass or its hydrolysate as carbon sources is of great interest; releasing sugars from biomass, however, is one of the most important limitations for low-cost production of biohydrogen.

$$C_6H_{12}O_6 + 2H_2O \rightarrow 2CH_3COOH + 4H_2 + 2CO_2 \qquad (3.10)$$

$$C_6H_{12}O_6 \rightarrow CH_3CH_2CH_2COOH + 2H_2 + 2CO_2 \qquad (3.11)$$

$$4C_6H_{12}O_6 + 2H_2O \rightarrow 3CH_3CH_2CH_2COOH + 2CH_3COOH + 10H_2 + 8CO_2 \qquad (3.12)$$

$$C_6H_{12}O_6 + 2H_2O \rightarrow CH_3CH_2OH + CH_3COOH + 2H_2 + 2CO_2 \qquad (3.13)$$

In recent years, hydrogen production using synthetic biology and cell-free system has attracted much interest. Zhang et al. [75] demonstrated a synthetic enzymatic pathway consisting of 13 enzymes for producing hydrogen from starch and water. The theoretical yields of biohydrogen from this pathway from hexose and xylose were 12 H_2 per glucose molecule [75, 76] and 10 H_2 per xylose molecule [77], respectively, and were much higher than the theoretical limit (4 H_2 per glucose) of anaerobic fermentations. This pathway seems to be promising to produce low-cost hydrogen from biomass for fuel cells, but demonstration in a larger scale is needed.

3.1.3.2 Biogas and Hythane

Biogas can be produced from organic materials by anaerobic digestion. It contains 50 to 75 percent (v/v) CH_4, 25 to 45 percent (v/v) CO_2, 0 to 2 percent (v/v) N_2, 0 to 2 percent (v/v) CO, 0 to 1 percent (v/v) H_2S, 0 to 1 percent (v/v) H_2, 0 to 1 percent (v/v) NH_3, 0 to 2 percent (v/v) O_2, and 2 to 7 percent (v/v) water vapor [78]. During anaerobic digestion, the conversion of organic matter into biogas is carried out by a consortium of microorganisms through a series of metabolic stages. Traditional anaerobic methane fermentation normally consists of four steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis [79]. Taking lignocellulosic biomass as an example, biomass is first hydrolyzed to monosaccharides such as glucose or xylose by extracellular cellulase enzymes secreted by hydrolytic or fermentative bacteria. The monosaccharides are then fermented by acidogenic bacteria in a process known as acidogenesis into a mixture of CO_2, H_2, alcohol, and low-molecular-weight volatile fatty acids (e.g., acetic, propionic, and butyric acids). In the final stage, acetotrophic and hydrogenotrophic methanogens transform acetate, H_2, and CO_2 into a mixture of CH_4 and CO_2 (the biogas) [78, 80]. Various lignocelluloses, such as wheat straw [81, 82], rice straw [83, 84], corn stalk [85, 86], wastepaper [87, 88], and forestry residues [89], have been used for biogas production with yields between 0.1 and 0.4 $m^3\ kg^{-1}$ VS (volatile solid) and an average yield of 0.24 $m^3\ kg^{-1}$ of dry organic matter [90]. Note that H_2 is also produced in the anaerobic digestion process; thus coproduction of H_2 and CH_4 is possible via a multistage fermentation. Actually, the coproduction of a mixture of H_2 and CH_4 (also known as hythane) can obtain higher energy recovery efficiency from biomass than either single-stage hydrogen or single-stage methane production [79]. After parameter optimization for a two-stage fermentation using 10 $g\ L^{-1}$ glucose as substrate, H_2 yield was increased to 2.75 $mol\ mol^{-1}$ glucose and the CH_4 yield increased to 2.13 $mol\ mol^{-1}$ glucose, which corresponded to a total energy recovery of 82 percent [91]. Biomass hydrolysate and pretreated lignocelluloses, such as agricultural residues, weeds, algae biomass, and livestock manure, have been successfully converted to hythane in laboratory scale by multistage anaerobic fermentation, with H_2 yield of 10 to 200 $L\ kg^{-1}$ VS and CH_4 yield of 100 to 700 $L\ kg^{-1}$ VS [92–96]. However, biomass recalcitrance limits the biodegradation efficiency of cell wall polysaccharide, even when a consortium of bacteria is used. Therefore, pretreatments such as physical comminution, thermal–chemical, chemical, and biological pretreatments must be used to increase biogas or hythane yield [97].

Using biogas or simulated mixed gases as a fuel for electricity generation in SOFC has been studied [39, 98–103]. Generally, the power density of SOFC increased with the increase in CH_4 concentration of fuel gas [39]. However, directly using biogas as a fuel for SOFC has faced several issues. First, a strong temperature gradient generated in the cell by an endothermic reforming reaction can cause cell fracture [100]. Second, impurity contained in biogas, especially H_2S, can poison the anodic catalyst by blocking the anode-active area, thus lowering the power output [104]. Poisoning by H_2S at 1 ppm caused approximately 9 percent voltage drop and 40 percent decrease in reaction rate of internal reforming using a simulated biogas (mixture of CH_4 and CO_2) [102]. Third, carbon deposition on anode caused by Ni-catalyzed reforming of CH_4 may decrease the cell performance or even cause cell deactivation [40, 41, 105]. Various processes have been developed to remove H_2S, including precipitation by Fe^{2+} ions or Fe^{3+} ions, adsorption on activated carbon, chemical absorption using NaOH or iron-oxide-coated ($Fe(OH)_3$ or Fe_2O_3) supported material, and biological treatment [106]. Another solution to H_2S poisoning is to modify the Ni-based anode or developing novel anode material that is not sensitive to sulfur, but such materials can be costly. A promising solution to carbon deposition is to combine an external reforming with SOFC, such as steam reforming or catalytic partial oxidation using either air or pure oxygen as oxidant [107]. Modification of the anode with the addition of a second-phase catalyst, such as K_2O or Ru–Ni, was also effective in suppressing carbon deposition [108].

3.1.3.3 Syngas

3.1.3.3.1 Syngas Characteristics and Formation Chemistry

Syngas (or synthesis gas) is mainly produced by gasification of biomass. It is the most promising biomass-derived fuel for SOFC at large scales. Syngas mainly consists of CO, H_2, CH_4, and a small quantity of other light hydrocarbons (C_nH_m), CO_2, water vapor, and N_2 from the air supplied for gasification [109]. The composition of syngas is affected by gasification conditions, such as temperature, equivalent ratio, and pressure, and the biomass characteristics such as biomass type, chemical compositions, and moisture content [109]. Generally, biomass syngas from a bubbling fluidized bed contains 5 to 26 percent H_2, 13 to 27 percent CO, 12 to 40 percent CO_2, less than 18 percent H_2O, 2 to 11 percent CH_4, less than 3 percent C_{2+}, less than 0.11 percent tars, ppm magnitude of H_2S, less than 0.2 percent O_2, and 13 to 56 percent N_2 [110]. During gasification of lignocellulosic biomass, drying, pyrolysis and devolatilization, reduction, and combustion take place [111]. The drying step removes moisture in the biomass; during pyrolysis, feedstock undergoes heat-induced decomposition, which liberates volatile constituents to the gas phase in the form of CO, CO_2, and light hydrocarbons and liquid long-chain hydrocarbons [111]. Gasification usually takes place at high temperatures (600°C to 1,000°C) under the deficiency of oxidants. Typical oxidants are air, steam, CO_2, oxygen, or the combinations of these [112].

Biomass gasification can be described by a global reaction (Equation 3.14), where $CH_\alpha O_\beta$ denotes biomass, neglecting sulfur and nitrogen. The chemical reactions involved in gasification of biomass include solid–gas phase and gas–gas phase reactions (Equations 3.15 through 3.23) [112, 113]. Energy from the combustion reactions is used to drive the gasification reactions to produce syngas [113].

$$CH_\alpha O_\beta \text{ (biomass)} + O_2 \text{ (21\% of air)} + H_2O \text{ (steam)} \rightarrow CH_4 + CO + CO_2 + H_2 + H_2O \text{ (unreacted steam)} + C \text{ (char)} + \text{tar} \quad (3.14)$$

Partial oxidation reaction:

$$C(s) + 1/2O_2(g) = CO(g)\ (\Delta H^0 = -110.5 \text{ kJ mol}^{-1}) \quad (3.15)$$

Complete oxidation reaction:

$$C(s) + O_2(g) = CO_2(g)\ (\Delta H^0 = -393.5 \text{ kJ mol}^{-1}) \quad (3.16)$$

Hydrogen oxidation reaction:

$$H_2(g) + O_2(g) = H_2O(g)\ (\Delta H^0 = -241.8 \text{ kJ mol}^{-1}) \quad (3.17)$$

Hydrogasification reaction:

$$C(s) + 2H_2(g) = CH_4(g)\ (\Delta H^0 = -74.8 \text{ kJ mol}^{-1}) \quad (3.18)$$

Water-gas shift reaction:

$$CO(g) + H_2O(g) = CO_2(g) + H_2(g)\ (\Delta H^0 = -41.2 \text{ kJ mol}^{-1}) \quad (3.19)$$

Steam reforming reaction:

$$CH_4 + H_2O(g) = CO(g) + 3H_2(g)\ (\Delta H^0 = 206.0 \text{ kJ mol}^{-1}) \quad (3.20)$$

Water-gas reaction:

$$C(s) + H_2O(g) = CO(g) + H_2(g)\ (\Delta H^0 = 131.3\ \text{kJ mol}^{-1}) \quad (3.21)$$

$$C(s) + 2H_2O(g) = CO(g) + 2H_2(g)\ (\Delta H^0 = 90.1\ \text{kJ mol}^{-1}) \quad (3.22)$$

The Boudouard reaction:

$$C(s) + CO_2(g) = 2CO(g)\ (\Delta H^0 = 172.5\ \text{kJ mol}^{-1}) \quad (3.23)$$

3.1.3.3.2 Impurities of Syngas and Their Impacts on SOFC Performance

Biomass syngas contains a variety of impurities that have negative effects on the performance of SOFC. The effects of impurities in syngas on SOFC performance have been intensively reviewed by Aravind and de Jong [32]. These impurities mainly include particulates, tars, H_2S, hydrogen chloride (HCl), alkali metal compounds, and nitrogen-containing contaminants. Particulates can deposit on anode of an SOFC, causing clogging of the anode porous structure, thus hindering gas diffusion, blocking the active catalytic area, or causing anode layer delamination because of mechanically induced tensions [114]. Filtration is the most commonly used process to remove particulates in syngas.

Tars present in syngas are generally assumed to be largely aromatics [115]. The variations in the composition of tars range from primary oxygenated pyrolysis products produced at lower gasification temperatures to high-molecular-weight deoxygenated products at higher temperatures and severe reaction conditions [32]. The effects of tars or model aromatic compounds, such as toluene, benzene, or naphthalene, on SOFC performance have been studied [114, 116–119]. No significant impact on an Ni–GDC anode was observed up to 110 ppm tar (naphthalene was used as representative). An Ni/GDC (gadolinium-doped ceria) anode operating at temperatures greater than 750°C was not susceptible to carbon deposition from a typical biomass gasification syngas containing 15 g m^{-3} benzene [116]. These studies suggest that SOFC anodes have a high tolerance to tars. Because the amount and composition of the tars present in biomass syngas depend on the type of feedstock and the operating conditions and oxidant [120], more intensive investigation on the subject is still needed.

Sulfur compounds had serious negative effect on SOFC performance, especially on those with an Ni–YSZ anode. Sulfur is predominantly present in syngas as H_2S, with concentrations ranging from 20 to 200 ppm. Syngas from nonwoody biomass usually has a higher H_2S concentration than that from woody biomass [32]. Various studies have proved that H_2S in the syngas poisons the catalyst even at low-ppm levels [121–126]. The polarization resistance and overvoltage of the electrode increased when H_2S concentration exceeded 0.05, 0.5, and 2 ppm at 1,023, 1,173, and 1,273 K, respectively, [124]. Cell voltage dropped with periodical addition of 2 to 100 ppm H_2S to an H_2-containing fuel in 24-h intervals. Fortunately, cell performance can be recovered after removing H_2S from the simulated fuel gas [122, 125]. This suggests that the poisoning effect of H_2S is perhaps primarily caused by the adsorption of H_2S at active sites to block Ni particles, which does not cause significant changes in the microstructure of the anode or the formation of an insulating layer [32]. H_2S can be removed by chemical adsorption using metal oxides such as zinc, cesium, copper, and other transition metal oxides [127–129] or molten carbonate (MC) [130].

HCl has been detected as the predominant halide gas in biomass syngas, with concentrations up to 200 ppm [131] depending on the biomass type. It is formed by the vaporization of alkali metal salts contained in the biomass and reacts with water vapor at high temperatures [131, 132]. Cell performance decreased when HCl was added into a simulated gas fuel for SOFC [133–136]. An Ni–YSZ cermet anode can tolerate up to 10 ppm HCl without significant performance degradation [133]. However, a higher concentration of HCl, such as 100 ppm, caused notable decrease in the power output. HCl mainly caused the corrosion of cell components and reacted with the nickel anode, resulting in permanent changes of the surface microstructure of the nickel particles [135]. Moreover, HCl can react with other contaminant species in the syngas to form salts, such as ammonium chloride (NH_4Cl) and sodium chloride (NaCl), which cause fouling of cell component or blocking cell pipe [137]. HCl generally can be removed at high temperatures by chemical adsorption using sorbents such as carbonates [32], metal oxides [138], and alkali earth metal compounds [132] to less than 1 ppm. The removal efficiency is significantly influenced by gas flow rate and temperature.

Alkali metal compounds, especially sodium and potassium, in biomass syngas are formed from vaporization of ash materials of biomass at gasification temperatures. However, only a minor fraction of the biomass alkali remains in the gas phase after gasification. The reported alkali compound concentrations range from sub-ppm to a few ppm, depending on biomass types and gasification temperature [139, 140]. Grass biomass has higher ash content than woody biomass, resulting in a higher concentration of alkali compounds in syngas. Alkali compounds can reduce cell performance by corroding cell components, deleteriously affecting fuel-reforming catalysts, and probably negatively influencing fuel cell electrodes [141]. Alkali sorbents such as bauxite, kaolinite, and activated alumina are usually used to remove alkali compounds to a few ppm or sub-ppm levels [142].

The primary nitrogen-containing compound in biomass syngas is ammonia (NH_3), with concentrations ranging from a few hundreds to 1,000 ppm depending on biomass and gasification method [143]. No detailed information on the negative impact of NH_3 on SOFCs was found in the literature. The addition of ammonia can suppress coke formation in direct methane SOFC using a conventional nickel cermet anode. Furthermore, hydrogen produced by decomposition of ammonia also acted as an additional fuel source for the SOFC [144–147], resulting in high cell performance. The operational stability of fuel cells was also improved when NH_3 was added [148]. These studies suggest that NH_3 has positive effects on SOFC performance.

3.1.3.3.3 Integration of Biomass Gasification and SOFC for Electricity Generation

Integrating biomass gasification and SOFC has attracted much attention, and various studies have been reported, particularly on process thermodynamics relating to energy efficiency [149–158]. Models predicted that integrating SOFC with biomass gasification produces higher electrical and exergy (the maximum useful work that can be obtained from a system at a given state in a given environment) efficiencies with lower greenhouse gas emissions than the conventional biomass-fueled power production system using a steam turbine [159]. In a typical integration (Figure 3.3), syngas from biomass gasification is first cleaned to remove various contaminants and then reformed to obtain H_2-rich fuel. The off gas from SOFC containing unconverted fuels is further burned in a combustor to produce heat for steam. The steam can be used for heating a reforming unit and driving the steam turbine for electricity. Gasification is the limiting step for electricity generation in such a combination; however, higher power density was obtained when catalysts such as Fe_2O_3 and K_2CO_3 were added into coconut shell carbon to accelerate fuel gas production [160]. Energy and exergy analyses of an integrated SOFC with gasification had an energy efficiency of 37 percent and an exergy efficiency of 23 to 27 percent for steam turbine process, depending on feedstock [161]. Fuel cell, gasifier, and combustion chamber were the main units with high exergy destruction. A similar conclusion was drawn from a pilot experimental study using a bagasse gasification unit integrated with an SOFC [151]. The major exergy destruction is the gasifier, ranging from 75 to

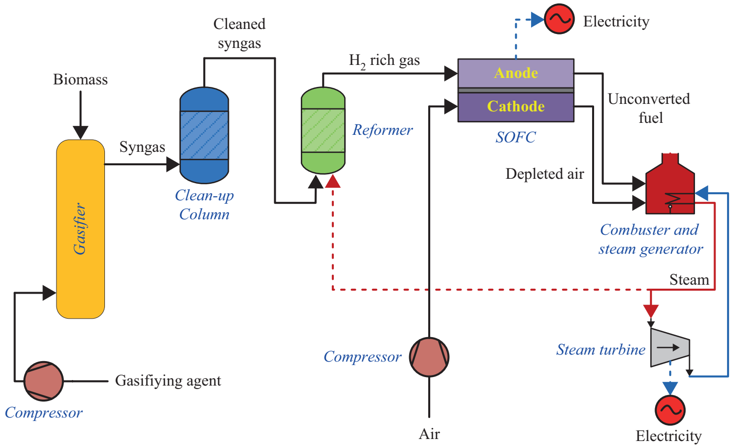

Figure 3.3. A typical scheme of integrating biomass gasification with SOFC for electricity generation.

80 percent of the total loss. Gasification agents were also found to have notable influences on the biomass-to-electricity conversion efficiency. Using steam as the gasification agent yielded higher electrical efficiency, power-to-heat ratio, and exergetic efficiency, but lower fuel utilization efficiency (50.8 percent), compared with values obtained using air and enriched oxygen [150]. The total efficiency of integrated SOFC with gasification was greatly dependent on cell and gasifier operating conditions and biomass moisture content [162]. Several economic evaluations on biomass–gasifier–SOFC systems indicated that a biomass–gasifier–SOFC system can obtain a high biomass-to-electricity conversion efficiency than any other biomass-fired electricity generation system; however, the operating cost of this system is very high. Gasifier, SOFC, and steam generator are major units with high investment costs [163–165].

3.2 DIRECT CARBON FUEL CELL

3.2.1 TYPES AND PRINCIPLES OF DCFC

DCFC directly use a carbon-rich material (e.g., coal and biochar) as a fuel [166]. The overall reaction of DCFC is

$$C + O_2 \rightarrow CO_2 \ (\Delta G = 395.4 \text{ kJ mol}^{-1}, E^0 = 1.02 \text{ V}) \qquad (3.24)$$

DCFC have several advantages over other fuel cells [166, 167]. First, a DCFC has a theoretical electrochemical conversion efficiency slightly

Table 3.2. Anodic and cathodic reactions of major DCFC developed currently

Cell type		*Typical carbon fuels*	*Anode and main anodic reaction*	*Electrolyte*	*Oxidant and cathodic reaction*	*T (°C)*
MH-DCFC		Coal, biochar, graphite rod, carbon black etc.	Graphite or metal (e.g. nickel alloys) $C+4OH^- = 2H_2O\ (g)+CO_2 + 4e^-$	Molten hydroxides ← OH^-	Air or pure oxygen as oxidant $O_2 + 2H_2O + 4e^- = 4OH^-$	500–650
MC-DCFC		Coal, biochar, activated carbon, graphite particle etc.	Carbon particles in molten carbonates $C+2CO_3^{2-} = 3CO_2 + 4e^-$	Molten carbonates ← CO_3^{2-}	Air or pure oxygen as oxidant $O_2 + 2CO_2 + 4e^- = 2CO_3^{2-}$	600–900
SO-DCFC	DCSO-DCFC	Coal, wood charcoal, biochar etc.	Carbon particles in a fluidized bed $C+2O^{2-} = CO_2 + 4e^-$	Oxygen ion conducting solid oxide such as YSZ, GDC, SDC ← O^{2-}	Air or pure oxygen as oxidant $O_2 + 4e^- = 2O^{2-}$	700–1000
	MMSO-DCFC	Coal, carbon, plastic, biomass etc.	Carbon particles in liquid metal (e.g. tin) $Sn\ (1) + 2O^{2-} = SnO_2 + 4e^-$ $C + SnO_2 = CO_2 + Sn\ (1)$			~1000
	MCSO-DCFC	Coal, biochar, activated carbon etc.	Carbon particles in molten carbonates $C + 2O^{2-} = CO_2 + 4e^-$ $C + O_2^- = CO + 2e^-$			700–1000

Source: Adapted based on Giddey et al. [166].

exceeding 100 percent because of the positive entropy change for the cell reaction (Equation 3.24, $\Delta S = 1.6$ J mol K^{-1}). Second, the fuel fed and product gases are distinct phases; therefore, their chemical potentials (activities) are fixed and independent of the extent of conversion of the fuel or location within the cell. Third, the DCFC system is mechanically simple because no reformers or heat engines are required. DCFC are classified into three main types according to the electrolyte used, namely, molten hydroxide (MH), MC, and solid oxide (SO), as compared in Table 3.2. With the development of materials and fuel cell technologies, some hybrid DCFC have demonstrated improved performance and are promising for long-time operations [168].

3.2.1.1 Molten Hydroxide Direct Carbon Fuel Cell

The molten hydroxide direct carbon fuel cell (MH-DCFC) was first patented in 1896 [169] and further developed in the mid-1990s by Scientific Applications and Research Associates (SARA) Inc. [170]. This type of fuel cell uses MH (LiOH, NaOH, KOH, or their mixture) as the electrolyte in a metallic container that also acts as a cathode. Typical operating temperatures of MH-DCFC are 500°C to 650°C [171]. The advantages of hydroxide electrolyte are its high electrical (ionic) conductivity, high electrochemical activity of carbon, lower operation temperatures, and ability to use less-expensive materials for cell fabrication [172]. However, the drawback is the chemical instability of the electrolyte because of the reaction with anode product CO_2 ($2OH^- + CO_2 = CO_3^{2-} + H_2O$) or electrochemical reaction of carbon with hydroxide ($C + 6OH^- = CO_3^{2-}$

+ $3H_2O + 4e^-$), resulting in hydroxide to carbonate conversion, which reduced cell performance and life time [170, 173]. Using high water content in the electrolyte or oxide additives such as SiO_2, As_2O_3, Sb_2O_3, ZnO, and MgO have been found to prevent or reduce carbonate formation to some extent [173, 174].

Several types of MH-DCFC configuration have been developed by SARA. A novel design (Figure 3.4a) consists of a cylindrical pure graphite rod immersed into molten sodium hydroxide serving as both the anode and the fuel, with a cylindrical or prismatic shell serving as the container for the electrolyte and the cathode. The electrolyte around the anode (the anolyte) is separated from the electrolyte around the cathode (the catholyte) by a porous separator. The separator allows for the transport of hydroxyl ions but prevents air (oxygen) from coming in contact with the anode [175]. This design can overcome a number of drawbacks of a conventional MH-DCFC. Graphite rods have been used as a fuel, and the average power output of 40 mW cm^{-2} at 140 mA cm^{-2} over 540 h with a peak power output of 180 mW cm^{-2} was achieved. The maximal current density achieved was greater than 250 mA cm^{-2} [173]. However, no studies were reported using a biomass-fueled MH-DCFC.

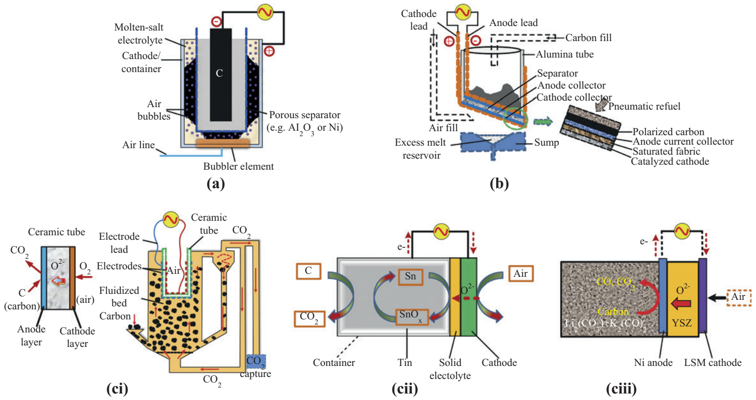

Figure 3.4. Schematics of different types of DCFC. (a) An MH-DCFC by SARA Inc. Adapted from Rastler [175]; (b) a DCFCs with a tilted orientation design by Lawrence Livermore National Laboratory (LLNL, Livermore, CA). Adapted from Giddey et al. [166] and Cooper [240]; (ci) an SOFC combined with carbon fluidized bed by CCE. Adapted from Cao, Sun, and Wang [167]; (cii) an SOFC fueled with solid carbon in molten metal (tin) developed by CellTech Power LLC. Adapted from Heydorn and Crouch-Baker [241]; (ciii) an SOFC fueled with solid carbon in MC.

Source: Adapted from Jain et al. [196].

3.2.1.2 Molten Carbonate Direct Carbon Fuel Cell

Molten carbonate direct carbon fuel cell (MC-DCFC) is a type of DCFC using MC as the electrolyte. This type of fuel has been considered as one of the most promising fuel cells for commercialization. Actually, fuel cells with MC as electrolyte have been developed to convert gas fuels such as hydrogen [176, 177], natural gas [178], biogas [179–181], and syngas [182–185] to electricity. Using MC (commonly Li_2CO_3, K_2CO_3, and Na_2CO_3 or their mixture) as electrolyte has a number of advantages, such as high ionic conductivity [166, 171] and good stability in the presence of CO_2 with a long-term operation [186, 187]. The carbonate can also catalyze carbon oxidation [188]. However, MC-DCFC has to be operated at higher temperature than MH-DCFC, typically at 600°C to 900°C, because of the high melting point of carbonate. The actual OCV of the cell is affected by CO_2 partial pressure at the anode and the cathode and O_2 partial pressure at the cathode. In particular, lowering CO_2 partial pressure in the anode by inert gas purge will result in an increased OCV [189]. The desirable carbon fuel for MC-DCFC should have high mesoporous surface area and rich oxygen-containing surface groups [190]. The anodic performance of a cell may also be improved by using carbon of small particle size and high stirring rates.

A tilted orientation design was developed by Lawrence Livermore National Laboratory (LLNL, Livermore, California) (Figure 3.4b). A 32 percent Li_2CO_3–68 percent K_2CO_3 melt was used as electrolyte. Various carbon fuels have been tested including coconut-activated carbon and peach pit char. The current density achieved ranged from 58 to 124 mA cm^{-2} at a cell voltage of 0.8 V [191]. Although MC-DCFC showed various merits, technical issues still exist, including high cathode polarization, loss of cathode performance with time, corrosion of metal bipolar plates, difficulties associated with fuel delivery, low power densities, short cell lifetimes, and the need for keeping the cell under constant polarization to avoid the reverse Boudouard reaction [166]. Fuel processing and fuel delivery systems are also key limits to a long-term operation of MC-DCFC.

3.2.1.3 Solid Oxide Direct Carbon Fuel Cell

In solid oxide direct carbon fuel cells (SO-DCFC), SOs similar to those used in SOFC are used as electrolyte for conducting oxygen ion (O^{2-}). The most common electrolyte being investigated is stabilized zirconia (8 to 10 mol percent Y_2O_3-stabilized ZrO_2, YSZ) at operating temperature of 800°C to 1,000°C [171]. There are three main classes of SO-DCFC depending on the contact type between the fuel and the anode, namely, solid carbon or fluidized bed (DCSO-DCFC or FBSO-DCFC) (Figure 3.4ci), solid carbon in molten metal (MMSO-DCFC) (Figure 3.4cii), and solid carbon in MC (MCSO-DCFC) (Figure 3.4ciii) [166]. In FBSO-DCFC, the anode directly contacts the carbon particles. For continuous fuel feeding to the anode–electrolyte interface, a fluidized bed arrangement with fluidizing gas, typically CO_2, can be arranged, as proposed by Clean Coal Energy (CCE, Stanford, California) (Figure 3.4ci) [167]. However, poor contact between the carbon anode and the electrolyte is still the main issue of FBSO-DCFC.

In MMSO-DCFC, molten metal, such as tin [192–194], is used as the anode and solid carbon fuel carrier, as developed by CellTech Power LLC (Westborough, Massachusetts) (Figure 3.4cii). The oxygen anions transported through the SO electrolyte react with tin. The tin oxide formed can be converted back to tin by chemical reaction between tin oxide and carbon (Table 3.2). Carbon can be directly converted to electricity without reforming when liquid tin was used. The operating temperature of an MMSO-DCFC is around 1,000°C. Fuel efficiency is up to 50 percent when coal is used and 34 percent when biomass is fed [194]. Sulfur is not a poison element anymore in this type of fuel cell. The high solubility of sulfur in molten tin (8 percent at 1,000°C) allows dissolved sulfur to be oxidized to SO_2 and carried out with anode exhaust [194]. The major technical hurdle is the excessive anodic polarization losses because of the use of porous ceramic separator. However, few studies on electricity generation using biomass or derivate charcoal as fuel were carried out.

MCSO-DCFC is a combination of MC fuel cell and SOFC, in which the anode is filled with a slurry of MC and carbon fuel (Figure 3.4ciii). This hybrid fuel cell can achieve direct conversion of solid carbon to electric power with high efficiency. MCSO-DCFC has some advantages, such as no need for CO_2 circulation, ability to protect the cathode from MC, and use of advanced cathode compositions already developed for SOFC. It can also avoid cathode flooding and corrosion in MC-DCFC [166]. A typical MCSO-DCFC consists of a NiO–YSZ anode, an LSM–YSZ cathode, YSZ as the oxygen ion conductor, and a mixture of Li_2CO_3 and K_2CO_3 as the solid carbon carrier [195, 196]. The actual anodic reactions are more complex than simple direct oxidation of carbon to CO_2. Partial oxidation of carbon to CO also takes place ($C + O_2^- \rightarrow CO + 2e^-$), evidenced by the CO detected in the gas products [196]. Power densities of 13.0 and 6.9 mW cm^{-2} at 900°C and 700°C, respectively, and OCV of 1.5 V at 550°C to 700°C (much higher than the theoretical value of 1.02 V) were achieved from this hybrid fuel cell using carbon black as fuel [195]. The major issues of MCSO-DCFC are corrosion of the nickel anode and other cell components and relatively poor stability of YSZ electrolyte in MC environments (e.g., formation of lithium zirconate in the presence of Li–K carbonate eutectic mixture at 1,000°C). Technical issues such as low power densities, very short lifetime, and slow electrode kinetics must be solved for large-scale applications [166].

3.2.2 BIOMASS AS A FUEL FOR DCFC

3.2.2.1 Biomass-Derived Charcoal (Biochar) as a Fuel

Currently, the main fuels used for DCFC are coal and graphite. However, carbon produced from lignocellulosic biomass (biochar) is promising for DCFC because biochar from biomass is inexpensive, easy to store, and readily available worldwide. Biochar also contains very low sulfur and nitrogen and no mercury. It has high electrical conductivity, large surface areas, and many reactive bonds at relatively modest temperatures [197]. Using biochar as a fuel for electricity generation in DCFC has been reported in recent years (Table 3.3). The reported power densities are lower than 100 mW cm^{-2}. Both biochar and fuel cell types show significant influence on power output. When comparing pine charcoal (PCC) with bituminous coal, demineralized bituminous coal, and anthracite coal as fuel in a direct carbon SOFC and an MCSO-DCFC, PCC showed the greatest power output [198]. A similar conclusion was reached by the same authors when comparing apple tree biochar with graphite rod in an MH-DCFC. Biochar was much more reactive than graphite rod, probably because of the structure of the carbon matrix and the degree of crystallinity of the biochar carbon [199]. Biochar [200] can eliminate or reduce anode degradation due to low sulfur content [189]. The chemical and physical properties of fuel such as composition, structure, surface area, surface functional groups, and mineral impurities all affect the electrochemical oxidation of carbon fuel in DCFC [201]. Biochar is preferred for obtaining higher power density due to its high carbon content [202]. The electrochemical oxidation of biochar can be enhanced by the oxygen-containing groups at high concentrations. These oxygen-containing groups are bound within the interior of the graphite crystal or between the separated layers in the graphite crystals [203]. The oxygen-containing groups have low thermal stability and can reduce polarization losses and increase electrochemical oxidation rate at fixed potential when they emerge at the reacting edges

Table 3.3. Some reported power densities by biomass-derived carbon-fueled DCFC

Fuel	**Fuel cell type**	**Peak power density ($mW\ cm^{-2}$)**	**Reference**
Apple tree biochar	MH-DCFC	10–41.7[a]	[199]
Apple tree biochar	MH-DCFC	22.4	[200]
Energetic willow biochar	MH-DCFC	~22.3	[200]
Pine wood charcoal	MH-DCFC	~20	[200]
Commercial biochar	MH-DCFC	~34	[200]
Coconut activated carbon	MC-DCFC	102	[191]
Peach pit char	MC-DCFC	124	[191]
Willow carbon	MC-DCFC	18.48	[224]
Carbonized wood biomass	MC-DCFC	25.15	[202]
Wood charcoal A	DCSO-DCFC	54	[197]
Wood charcoal B	DCSO-DCFC	70	[197]
Coconut char	DCSO-DCFC	25 (with N_2 as the purge gas)	[204]
Coconut char	DCSO-DCFC	60 (with N_2 as the purge gas)	[204]
Biochar	DCSO-DCFC	10	[204]
Pine charcoal	MCSO-DCFC	12	[198]
Miscanthus carbon	MCSO-DCFC	77.41	[224]
Almond shell biochar	MCSO-DCFC	127	[203]
Fiberboard biochar	MCSO-DCFC	70–878[b]	[225]

[a] The peak power density varied depending on the electrolyte composition (ratio of NaOH, LiOH, and KOH) and temperature.

[b] The peak power density varied depending on the purge gas and cathode materials used. The maximum power density was obtained by using LSC cathode with flowing air.

and participate in the electrochemical reaction [203]. The main issue with biochar is its high ash content, particularly in grass-derived biochars, which can reduce the amount of surface area for reaction and block charge transfer through the cell to result in reduced cell performance [204]. Biochar properties are influenced by biomass type, carbonization techniques, and pretreatment and posttreatment procedures. Therefore, preparation of biochar is important and should be optimized.

3.2.2.2 Preparation of Biochar from Biomass

Biochar is a stable carbon-rich by-product synthesized through carbonization of plant-based biomass [205]. It has been defined by Shackley et al. [206] as "the porous carbonaceous solid produced by the thermochemical conversion of organic materials in an oxygen depleted atmosphere that has physicochemical properties suitable for safe and long-term storage of carbon in the environment." Biochar has received considerable interest in recent years for remediation of contaminated soil [207] and carbon sequestration [208]. Biochar can be produced from various carbonaceous biomass by different carbonization technologies such as pyrolysis, gasification, hydrothermal carbonization, and flash carbonization [209], of which pyrolysis is the most commonly used. The yield and quality of biochar are greatly dependent on pyrolysis conditions such as peak temperature, heating rate, retention time, reactor, and biomass characteristics such as biomass type and chemical composition, especially lignin content, particle size, and moisture and mineral salt contents. The wide range of process parameters leads to the formation of biochar products that vary considerably in their elemental and ash composition, density, porosity, pore size distribution, surface area, surface chemical properties, water and ion adsorption and release, pH, and uniformity of physical structure [210]. Fast pyrolysis, with a heating rate of above 200°C min^{-1} and short residence time of approximately 2 s, usually favors the formation of bio-oil of approximately 75 percent yield, with charcoal yield typically lower than 12 percent [211]. Slow pyrolysis, with heating rate of generally less than 10°C min^{-1} to over 400°C, can obtain charcoal yield of approximately 30 percent, or as high as 51 percent [212].

Major components from lignocelluloses have been used for biochar production under different pyrolysis conditions (Table 3.4). Pure cellulose generally results in the lowest biochar yield, while lignin results in the highest biochar yield. Increasing pyrolysis pressure to 1 MPa can substantially increase biochar yield [213]. Woody biomass typically results in greater char yields than those derived from herbaceous feedstock because of their higher lignin content. Furthermore, biochar from woody biomass has higher carbon content than those from herbaceous biomass [214]. High carbon content is important for DCFC, as discussed previously;

Table 3.4. Some reported slow pyrolysis of lignocellulosic biomass for biochar production

Biomass	Heat rate and soaking condition	Pressure (MPa)	Biochar yield (%)	Reference
Woody biomass				
Alder wood	2°C min^{-1}, 450°C soaking for 60 min	0.1	30.5	[226]
Birch wood	2°C min^{-1}, 450°C soaking for 60 min	0.1	28.8	[226]
Oak wood	2°C min^{-1}, 450°C soaking for 60 min	0.1	31.2	[226]
Pine wood	2°C min^{-1}, 450°C soaking for 60 min	0.1	32.1	[226]
Spruce wood	2°C min^{-1}, 450°C soaking for 60 min	0.1	32.1	[226]
Beech wood	10°C min^{-1} up to 377°C with no soaking	0.1	29.7	[227]
Eucalyptus wood	10°C min^{-1}, 450°C soaking for 15 min	1.0	41.8	[212]
Nonwoody biomass				
Corncob	10°C min^{-1} up to 377°C, 477°C and 577°C with no soaking	0.1	26.0, 23.2, 21.5	[227]
Cocopeat	10°C min^{-1} up to 500°C and maintained for 1 h	NA	38.7	[228]
Paddy straw	10°C min^{-1} up to 500°C and maintained for 1 h	NA	41.0	[228]
Miscanthus	350°C–450°C for residence time of 14–15.1 min	NA	70 to 31	[229]
	10°C min^{-1} up to 500°C and maintained for 1 h	NA	27.2	[230]
Rice husk	5°C min^{-1} up to 350°C to 650°C with soaking for 20 min	0.1	~56 to ~45	[231]

Straw-stalk of rapeseed plant	5°C min^{-1} up to 500°C, 600°C, 700°C, 800°C	0.1	39.4, 35.6, 32.6, 29.6	[232]
Sugarcane bagasse	10°C min^{-1} up to 500°C and maintained for 1 h	NA	24.5	[228]
Switchgrass	300°C, 350°C, 400°C for 1–3 h	0.1	~84 to ~50 at 300°C; ~72 to ~40 at 350°C; ~60 to ~36 at 400°C	[233]
Wheat straw	5°C min^{-1} up to 350°C to 650°C with soaking for 20 min	0.1	~40 to ~27	[231]
Biomass components				
Cellulose				
Commercial cellulose fibers	5°C min^{-1} up to 600°C with no soaking	3–15	17.5	[234]
Commercial fibrous cellulose	5°C min^{-1} up to 429°C to 700°C with no soaking	2	~20 to ~12	[235]
Avicel PH105	20°C min^{-1} up to 800°C with no soaking	NA	~5	[236]
Sigma cellulose product	10°C min^{-1} up to 900°C with no soaking	NA	~7	[237]
Hemicellulose				
Beechwood xylan	5°C min^{-1} up to 600°C with no soaking	3–15	22.4	[234]

(*Continued*)

Table 3.4. Some reported slow pyrolysis of lignocellulosic biomass for biochar production (Continued)

Biomass	Heat rate and soaking condition	Pressure (MPa)	Biochar yield (%)	Reference
Beechwood xylan	20°C min^{-1} up to 800°C with no soaking	NA	~23	[236]
Birchwood xylan	10°C min^{-1} up to 900°C with no soaking	NA	~20	[237]
Lignin				
Wheat straw lignin[a]	Continuous pyrolysis in a bubbling fluidized bed at 500°C	0.1	35.6	[238]
Wheat straw or grass lignin[b]	Continuous pyrolysis in a bubbling fluidized bed at 500°C	0.1	39.0	[238]
Hardwood organosolv lignin	Continuous pyrolysis in a bubbling fluidized bed at 500°C	0.1	43.0	[238]
Crofton weed lignin[c]	10°C min^{-1} up to 900°C with no soaking	0.1	37.8	[239]
Crofton weed lignin[d]	10°C min^{-1} up to 900°C with no soaking	0.1	33.7	[239]
Sigma alkali lignin	5°C min^{-1} up to 600°C with no soaking	3–15	48.5	[234]
Sigma alkali lignin		NA	~40	[237]
Sigma lignin	20°C min^{-1} up to 800°C with no soaking	NA	37	[236]

[a] Extracted by organosov process.

[b] Extracted by soda process.

[c] Extracted by the Milox process.

[d] Extracted by acetosolv process.

carbon content of biochar increased with temperature as consistently demonstrated using various pyrolysis processes [215]. A carbon content of greater than 80 percent can be achieved at 500°C or higher, 60 to 80 percent at 400°C to 500°C, and 15 to 60 percent below 350°C [216]. Carbon content of biochars increased at an average of 41 g C kg^{-1} for each 100°C rise in pyrolysis temperature [210]. Pyrolysis temperature also can significantly affect biochar properties, such as pore structure, surface area, and adsorption properties [215].

CHAPTER 4

PERSPECTIVE OF BIOMASS-FUELED SOLID OXIDE FUEL CELLS AND DIRECT CARBON FUEL CELLS

Different biomass-energized fuel cells are compared in Table 4.1. Compared with direct carbon fuel cells (DCFC), solid oxide fuel cells (SOFC) show higher power density and stability (Table 4.1). Large-scale, utility-based SOFC power generation systems are in pilot-scale demonstration stages in the United States, Europe, and Japan. Small-scale SOFC systems are being developed for military, residential, industrial, and transportation applications [16]. It is also the most commercially promising fuel cell technology for electricity production from lignocellulosic biomass. In this system, biomass is first thermochemically or biologically transformed to fuel gases. The conversion efficiency of biomass to fuel gases is a limiting step. Biological conversion of lignocellulosic biomass to biohydrogen or biogas can be conducted at mild conditions with various microorganism species. However, fermentation is slow. New enzymatic pathways for producing biohydrogen, while promising, needs to be demonstrated at large scales. Thermochemical conversion seems to be more promising to produce fuel gases (syngas) for SOFC because of much higher reaction rates. Gasification of biomass is also a relatively mature technology for producing combustible gas. Thus, it can be integrated with SOFC at a large scale. However, the efficiency of common high-temperature SOFC with integrated gasification of solid feedstock is often lower than the efficiency of SOFC operated with pure hydrogen or methane because additional system components such as gasifier reduce efficiency. Hence, common fuel cell systems with integrated gasification of biomass will hardly reach

Table 4.1. Comparison of SOFC and DCFC fueled with different lignocellulosic biomass-derived fuel sources

Fuel cell type	Fuels	Operation temperature (°C)	Power density (mW cm^{-2})	Developing stage	Advantage	Technical challenge
SOFC	Biohydrogen	600–900	100–2,000	Laboratory	High power density; high reaction rate to convert fuel to electricity; long-term operation stability; high fuel flexibility; low emissions; relatively low cost; can be constructed for wide range of applications	Very high operation temperature with resulting materials and cost limitations and operating complexities; easy deactivation of catalyst by impurities; relatively low efficiency for bioconversion of biomass to hydrogen or biogas
	Biogas		100–1,500	Laboratory-commercial scale		
	Syngas		100–1,500	Laboratory-commercial scale		
DCFC	Biochar	500–1,000	10–900	Laboratory	Direct conversion of solid carbon to electricity; high theoretical electrochemical conversion efficiency; mechanically simple cell system; expensive noble metal electrode is not required	Relatively high operation temperature; relatively poor operation stability; poor contact between solid carbon and catalyst when no molten salt is used; corrosion of cell components when molten salts are used

electrical efficiencies above 30 percent [149]. Biomass gasification also has several challenges including tar formation to cause catalyst deactivation, operation interruption, and production of carcinogenic elements [109]. The presence of excessive moisture in biomass reduces the molar fractions of combustible components [217], therefore, the quality of the product gas, as well as the general performance of the system because of the excess energy consumed for water evaporation [218]. The impurities of syngas can cause degradation of SOFC. Gas clean-up reduces electricity production efficiency.

The following aspects should be considered to increase the efficiency of syngas–SOFC systems: (1) optimizing gasification in terms of process operation, catalyst, and reactor design and necessary pretreatment to remove moisture or desulfuration; (2) developing novel electrode and catalyst for SOFC with strong tolerance to impurities in syngas; (3) heat integration to decrease energy consumption and increase energy efficiency for power generation; and (4) comprehensive economic estimation of the system to identify the most expensive units for further optimization.

DCFC has the advantage of directly converting carbon to electricity. Biochar produced from lignocellulosic biomass is a promising alternative to coal for fueling DCFC. Using biochar as a fuel can reduce net CO_2 emission. Most biochar-fueled DCFC can only produce a power density less than 100 mW cm^{-2} that needs to be at least doubled for commercial applications. Biochar significantly lowers power density than coal in most cases. This is related to not only carbon content of biochar but also the properties of the fuel. DCFC is also still at an early stage of development compared with the conventional SOFC systems. A significant number of technical challenges still need to be overcome, such as poor power densities, high degradation rates caused by corrosion of cell components, poor fuel feed system, difficulty in scaling up to kilowatts and larger size systems, and need of establishing fuel processing requirements [166]. Poor contact of solid carbon with anode catalyst is a major challenge to DCFC, because fuel cannot diffuse to the active sites within a porous electrode where chemical oxidation can occur. The use of molten materials can not only improve power output but also increase the degradation of the system, either by chemical attack on other system components or through rapid reactions with fuel impurities because molten salts can alloy with even small quantities of metal within the fuel [166].

To improve the performance of biochar-fueled DCFC, the following areas of research are needed: (1) comprehensively investigating the effects of biochar properties (including elementary composition, impurity contents, moisture content, physical features such as particle size and its distribution, porosity, surface area, crystallinity, electrical conductivity, and wettability) on power output and establishing the "quality criteria" for biochar as a fuel in a DCFC for maximizing power output; (2) optimizing the carbonization or pyrolysis process, coupled with pretreatment or post-treatment processes, to maximize the production of biochar that meets the "fuel criteria"; (3) developing anode materials that can extend the triple-phase boundary of the reactant (carbon), oxygen ion conductor (electrolyte), and an electronically conducting material (current collector) to increase solid fuel reactive sites; (4) developing electrode-supported cell designs to minimize resistive losses across the electrolyte and enhance fuel transport or diffusion through the anode; and (5) integrating and globally optimizing biomass pyrolysis and DCFC systems to minimize running cost.

CHAPTER 5

CONCLUDING REMARKS

Lignocellulosic biomass is a promising feedstock for fuel cells because it is renewable, carbon neutral, and sustainable. For highly efficient conversion of biomass to electricity by fuel cell technology, biomass is usually first converted to simple fuels in external reactors, which refers to indirect biomass fuel cells, mainly including solid oxide fuel cells (SOFC) and direct carbon fuel cells (DCFC). Lignocellulosic biomass has to be chemically or biologically converted to syngas, biogas, or biohydrogen for fueling SOFC. This type of fuel cell shows high power density and seems to have promising chances to evolve into full-scale commercial applications, based on currently achieved power output and conversion efficiency. However, increasing the conversion efficiency of biomass to fuel gases is one of the most important considerations in reducing the cost of biomass-fueled SOFC. Lignocellulosic biomass can also be converted to biochar and used as a fuel for DCFC. This type of fuel cell can directly convert solid carbon to electricity after carbonizing lignocellulosic biomass without complex processing. However, poor contact of solid carbon with the anode catalyst is the major challenge of DCFC, and the cell performance must be improved for commercialization.

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Lignocellulosic Biomass-Energized Fuel Cells

Cases of High-Temperature Conversion

Xuebing Zhao • Wei Liu • Yulin Deng • J.Y. Zhu

Fuel cell technology has attracted great interest in recent decades. However, progress in lignocellulosic biomass-energized fuel cells has been slow. This is because that lignocellulosic biomass generally cannot be directly used for electricity generation in a fuel cell with high efficiency. As a renewable resource available in large quantities in many regions of the world, lignocellulosic biomass can be a promising feedstock for sustainable electricity production using fuel cell technologies.

In this monograph, we focus on the electricity generation in fuel cells that are operated at high temperatures with high efficiency using lignocellulosic biomass-derived fuels. More specifically, we discussed biomass conversion coupled solid oxide fuel cell and direct carbon fuel cell, the state of the art in technology development, the challenges and the perspectives on future development.

Xuebing Zhao is an assistant professor at Tsinghua University, Beijing, China. Dr. Zhao published extensively in the area of biorefining of lignocellulosic biomass to produce biofuels and chemicals via chemical and biological approaches.

Wei Liu is a PhD student at Georgia Institute of Technology whose thesis focuses on direct biomass fuel cell (DBFC). The first author of a recent paper on DBFC published in *Nature-Communications*. Mr. Liu has extensive research experience in polyoxometalate chemistry.

Yulin Deng is a professor at Georgia Institute of Technology in chemical engineering. Prof. Deng is the recipient of the 2013 American Institute of Chemical Engineers (AIChE) Andrew Chase award for outstanding and life time contribution to the advanced utilization of forest renewable biomass.

J.Y. Zhu is a scientific leader at the USDA Forest Service, Forest Products Laboratory. Dr. Zhu's research covers a board area of wood and fiber utilization for the production of sugar/biofuel and cellulose nanomaterials. He is the Fulbright-Aalto University (Helsinki, Finland) Distinguished Chair in Energy and Sustainable Use of Natural Resources for the 2015–2016 academic year.

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