An Application of Solid Particles in Fuel Cell Technology

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Abstract

Solid fuel particles will become increasingly important in the future. Present energy conversion systems for solid fuels are too inefficient. New energy conversion systems for solid fuels with higher energy conversion efficiencies are possible. Fuel cell technology is a key-technology in these new conversion systems. The direct carbon fuel cell (DCFC) operates on carbon particles obtained from a variety of solid fuel feedstocks. The DCFC is the only fuel cell designed to directly oxidize carbon particles in a special anode chamber. The particles are generally graphite structure with high purity. The electrolyte used is the high temperature solid oxide, molten carbonate or hydroxide electrolyte. Since a pure stream of CO₂ is produced the stream can easily be sequestered and disposed. Pure carbon dioxide produced as a by-product would also have a market in many industries. A well defined technology roadmap identifying key research and development (R&D) issues is necessary to provide a framework for the development of these systems and to prevent entrenchment in inherently inefficient technologies. This review paper describes the direct carbon fuel cell and its system, how it works, the developmental status, the characteristics of the carbon particles needed, and the research and development issues for the technology.

Keywords: Carbon particles, Direct carbon fuel cell, High efficiency

1. Introduction

The DCFC has been under development for many years. It was researched as early as 1896, but an intense research and development is necessary to realize its potential. William W Jacques, a US electrical engineer and chemist, described a DCFC in 1896 in US Patent 555511 for a “Method of converting potential energy of carbon into electrical energy”. Jacques used coke electrodes in a molten NaOH electrolyte.

Molten carbonate fuel cell (MCFC), solid oxide (SOFC) or hydroxide electrolyte (AFC) technologies can be used as the basis for the DCFC technology. A schematic of the direct carbon fuel cell in the MCFC mode is shown in Figure 1. The molten carbonate fuel cell operates at approximately 650°C (1200°F). The high operating temperature is needed to achieve sufficient conductivity of the carbonate electrolyte, yet allow the use of low-cost metal cell components. A benefit associated with this high temperature is that noble metal catalysts are not required for the cell electrochemical oxidation and reduction processes. For the hydrogen reaction the half cell electrochemical reactions are
\[ H_2 + CO_3^- \rightarrow H_2O + CO_2 + 2e^- \]

at the anode, and

\[ 1/2O_2 + CO_2 + 2e^- = CO_3^- \]

at the cathode. The overall cell reaction is

\[ H_2 + 1/2O_2 + CO_2 \text{(cathode).} H_2O + CO_2 \text{(anode)} \]

In the case of carbon and the DCFC the reaction at the anode is generally replaced by

\[ 1/2C (s) + CO_3^- = 3/2CO_2 + 2e^- \]

For the case of the electrolyte being a molten carbonate salt, the carbonate ions move from the oxygen cathode to the special anode chamber where they react with the carbon fuel particulates dispersed in the molten salt and form CO\textsubscript{2} in high concentration. The particulate carbon dispersed in the molten salt is converted to CO\textsubscript{2} which emerges from the anode compartment of the cell at 100% concentration. MCFC current materials, thicknesses and processing techniques are shown in Table 1\textsuperscript{2).} The materials for the anode listed in Table 1 are for use when the fuel is hydrogen or methane. A special anode is required for the DCFC.

The SOFCs (600-1000°C) being developed, particularly the planar types, have unique designs, are constructed of various materials, and are fabricated by different techniques. The electrochemical reactions associated with hydrogen fuel are:

\[ H_2 + O^- = H_2O + 2e^- \]

at the anode, and

\[ 1/2 O_2 + 2e^- = O^- \]

at the cathode. The overall cell reaction is

\[ H_2 + 1/2 O_2 = H_2O \]

In the case of carbon and the DCFC the reaction at the anode is generally replaced by

\[ 1/2C(s) + O^- = 1/2CO_2 + 2e^- \]

SOFC materials widely vary. The basic SOFC materials, thickness and processing techniques are shown in Table 2\textsuperscript{2).}

<table>
<thead>
<tr>
<th>Table 1 MCFC Materials</th>
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<tr>
<td>Anode: Ni-Cr/Ni-Al/Ni-Al-Cr; 3-6 ( \mu )m pore size; 45 to 70 percent initial porosity; 0.20 to .5 mm thickness; 0.1 to1 m\textsuperscript{2}/g</td>
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<td>Cathode: Lithiated NiO-MgO; 7 to15 ( \mu )m pore size; 70 to 80 percent initial porosity; 60 to 65 percent after lithiation and oxidation; 0.5 to 1 mm thickness; 0.5 m\textsuperscript{2}/g</td>
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<td>Electrolyte: ( \gamma )-LiAlO\textsubscript{2}, ( a )-LiAlO\textsubscript{2}; 0.1 to12 m\textsuperscript{2}/g; 0.1 to 0.1 mm thickness</td>
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<td>Support: 62 Li-38 K; 60 Li-40; tape cast 0.5 to1 mm thickness</td>
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<th>Table 2 SOFC Materials</th>
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<td>Anode: Ni/ZrO\textsubscript{2}-cermet; Deposit slurry, EVD fixed; 12.5 ( \times ) 10\textsuperscript{-6}cm/cm\textdegree CTE; ( \sim )150 ( \mu )m thickness; 20 to 40 percent porosity</td>
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<td>Cathode: Doped lanthanum strontium manganite; (LSM); Extrusion, sintering; ( \sim )2 mm thickness; 11 ( \times ) 10-6cm/cm\textdegree CTE from room temperature to 1000\textdegree C; 30 to 40 percent porosity</td>
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<td>Electrolyte: Yttria stabilized ZrO\textsubscript{2} (YSZ) (8 mol percent Y\textsubscript{2}O\textsubscript{3}); EVD; 10.5 ( \times ) 10\textsuperscript{-6}cm/cm\textdegree CTE from room temperature to 1000\textdegree C; 30 to 40 ( \mu )m thickness</td>
</tr>
<tr>
<td>Cell Interconnect: Doped lanthanum chromite; Plasma spray; 10 X 10-6cm/cm\textdegree CTE; ( \sim )100 ( \mu )m thickness</td>
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The materials for the anode listed in Tables 1 and 2 are for use when the fuel is hydrogen. Special anode materials selective for carbon oxidation are available and will be discussed later. In addition to hydrogen, carbon monoxide (CO) and other hydrocarbons such as methane (CH₄) can be used as fuels. It is feasible that the water gas shift reaction involving CO (CO + H₂O = H₂ + CO₂) and the steam reforming of CH₄ (CH₄ + H₂O = 3H₂ + CO) in the high temperature environment of SOFCs produce H₂ that is easily oxidized at the anode. The direct oxidation of CO in fuel cells is also well established. In the SOFC case oxidation of carbon could occurring at the surface is quite different than oxidation after vaporization of the carbon.

In the AFC mode the hydroxide ion approach which occurs at 100-500°C is believed to have several advantages - lower cost lower-temperature materials of construction, more electrochemically active carbon and greater production of CO₂, although hydroxide reacts with carbon dioxide.

For the hydrogen reaction the half cell electrochemical reactions are

\[ \text{H}_2 + 2\text{OH}^- = 2\text{H}_2\text{O} + 2\text{e}^- \]

at the anode, and

\[ \frac{1}{2}\text{O}_2 + \text{H}_2\text{O} + 2\text{e}^- = 2\text{OH}^- \]

at the cathode. The overall cell reaction is

\[ \text{H}_2 + \frac{1}{2}\text{O}_2 = \text{H}_2\text{O} \]

In the case of carbon and the DCFC the reaction at the anode is generally replaced by

\[ \frac{1}{2}\text{C}(s) + 2\text{OH}^- = \frac{1}{2}\text{CO}_2 + \text{H}_2\text{O} + 2\text{e}^- \]

Electrode development in circulating electrolyte AFCs (hydroxide electrolyte) has concentrated on 1) multi-layered structures with porosity characteristics optimized for flow of liquid electrolytes and gases (H₂ and air), and 2) catalyst development. Most developers use noble metal catalysts; some use nonnoble catalysts. Spinels and perovskites are being developed in an attempt to lower the cost of the electrodes. Development of low-cost manufacturing processes includes powder mixing and presssing of carbon-based electrodes, sedimentation and spraying, and high-temperature sintering. AFC electrolyte development has been restricted to KOH water solutions with concentrations ranging from 6 to 12 normal. Still, use of less expensive NaOH has been considered. Minimal cost advantages appear to be far outweighed by performance reductions due to wetting angle and lower conductivity. However, NaOH as an electrolyte increases the lifetime of electrodes when CO₂ is present, because sodium carbonate, although less soluble than potassium carbonate, forms much smaller crystals, which do not harm the carbon pores.

2. Performance Potential of DCFC

For direct oxidation of carbon to CO₂ the maximum intrinsic thermal efficiency of conversion of the enthalpy (heating value) of the carbon to electricity is 100% at any temperature or pressure. This is because the entropy of oxidation of carbon is zero (ΔS = 0). Thus, the enthalpy of oxidation equals the free energy (ΔH = ΔG). This is not the case for the hydrogen fuel cell. The maximum intrinsic thermal efficiency for hydrogen fuel cells is plotted in Fig. 2 as a function of temperature. In the case of hydrogen as a fuel the maximum intrinsic thermal efficiency decreases with increasing temperature. Pressurization of the fuel cell to 30 atmospheres further improves hydrogen fuel cell performance, but it does not equal the 100% theoretical achievable by the DCFC. It is no mystery then why the interest in the DCFC.

Fig. 3 shows the expected fuel cell efficiency performance of carbon graphite solid particles in a simple SOFC hybrid system as a function of temperature with only 75% carbon utilization³. The reaction

![Fig. 2 Fuel Cell maximum intrinsic thermal efficiency. DCFC thermal efficiency is 1.0 at all temperatures.](image-url)
occurs almost to completion as soon as the conductivity of the SOFC electrolyte, LSGM in this case, is sufficient to conduct oxygen ions to complete the direct carbon oxidation reaction.

3. DCFC Developmental Status

There are several DCFC developers - Lawrence Livermore National Laboratory (LLNL) and Contained Energy (MCFC), SRI (MCFC + SOFC), SARA (hydroxide) and CellTech (SOFC). There is some work around the world at other organizations such as Stanford University, Akron University, Virginia Polytechnic Institute and State University, University of Colorado, Technical University Delft, Brown University, Duke University, AIST, and NEDO that has occurred over the years.

The LLNL and Contained Energy design utilizes an MCFC cathode, a nickel or high-fired graphite anode and porous ceramic separator. The anode reaction is carbon and carbonate ions forming CO₂ and electrons. De-ashing processes, such as those developed by UCC Energy (Sydney, Australia), the New Energy and Industrial Technology Development Organization and South African Synthetic Oil Limited could yield fuel at competitive costs (about $2–3 per million BTU). LLNL has demonstrated the technology in small, experimental cells with areas between 3 and 60cm². LLNL has tested a five-cell, self-feeding bipolar stack using pure fine carbon. LLNL reports that in repeated tests the cells deliver up to 0.1 watt/cm² continuously and are 80% efficient at 80 milliwatt/cm².

The reported performance of the SARA design is average output power in Mark III-A of 12-20 watts (over 540 hours) with peak output power (sustained for 5 to 10 seconds) 35-50 watts. The concepts have achieved 300 millivolt at 40 amperes in Mark III-A and 100 milliamperes/cm² in Mark III-A 250 milliamperes/cm² in Mark II-D. The efficiency was 60% measured in the non-optimized Mark III. They project the maximum efficiency of the DCFC at ~ 85-90% and the practical DCFC plant efficiency at ~ 70-75%.

The SRI design combines advances in SOFC and MCFC technology with the concept of using a circulating liquid-molten salt anode that incorporates the carbon-containing fuel. SRI is examining various molten salts, including molten carbonate, and using a YSZ electrolyte and a lanthanum strontium manganite cathode. A variety of fuels have been tested, including biomass, coal, tar, coke containing 6% sulphur, acetylene black, plastic and mixed waste. Using conventional coal without pretreatment, SRI has achieved power densities >100mW/cm².

CellTech technology builds upon conventional solid oxide fuel cell materials and components. Oxygen ions are extracted at the cathode and pass through the electrolyte to the anode (fuel) side where they combine with hydrogen or carbon monoxide from the fuel to form water or carbon dioxide. Meanwhile, electrons are released at the anode and travel through the load producing useful electrical energy on their way to the cathode. The cathode is made of conventional cathode materials such as lanthanum manganite doped strontium - LSM. The electrolyte is YSZ. The design is currently a cathode-supported with a tubular configuration. The anode is a p-orbital-electron metal such as tin which is molten at operating temperatures. The tin is held in place by a ceramic matrix. Tin is converted to tin oxide at the anode. In a separate reaction, the tin is “recharged” by the carbon. CellTech has integrated the reforming reaction (the reduction of tin oxide to tin) into the fuel cell, but uncoupled that reaction from the electrical production reaction (the oxidation of tin to tin oxide).
It is hardly expected that the DCFC development will precede the state-of-development of its base AFC, MCFC or SOFC technologies. There state-of-readiness appears eminent, however.

4. Solid Carbon Particle Fuel Production and Quality Requirements

The problem of the application of fossil fuels for powering fuel cells is to process the hydrocarbons in fossil fuels to produce elemental hydrogen and/or solid carbon. This can be accomplished by thermal cracking (decomposition) and pyrolysis processes. According to recent reports\textsuperscript{10}, the carbon formed in the plasma reactor is of a quality suitable for the molten carbonate cell type DCFC.

Graphite particles are commercially made from the sooting of methane; however, graphite is also available from coal, coal coke, petroleum coke, etc.. Sulfur, chlorine, sodium and trace metals are components of coal known to present problems to catalytic surfaces such as that of the fuel cell anode. Fouling, erosion, blockage of active catalyst sites, and chemical reaction are known problems occurring with coal ash. In addition, with coal the production of an aromatic or cyclic carbon compound rather than an active graphite solid particle is a problem inherent with the use of coal feedstock. What happens to the contaminants in coal with or without coal cleaning is an issue. Coal contains volatile components, so the DCFC system must be designed so that the coal can be introduced to the hot fuel cell without the explosive release of volatile components.

The cost of the chemical clean-up of coal particles to a low-ash, clean carbon has been estimated at $60/ton\textsuperscript{11}. Lower cost carbon particle clean-up systems obviously need to be developed. The estimated cost of oil agglomeration and advanced flotation are 17 and 12 $/ton of clean coal, respectively. Depending on the carbon content of the beneficiated coal, this could be in the economic range and competitive with other carbon formation processes providing the carbon content was high and the carbon reactive. For the MCFC-type DCFC, the anode chamber can be a carbonate melt similar to the MCFC electrolyte. This permits actually cleaning of impurities in the melt. For a low ash content carbon or coal this could be a batch process.

Nanoparticles of carbon if produced could be highly reactive. These hold great promise for subsequent development. LLNL\textsuperscript{6,7} has examined the relationship between the nanostructure of carbons and their electrochemical reactivity in molten salts by correlating significant differences in the 3D atomic structure of carbon fuels with their electrochemical reactivities. The more disordered the carbon atoms, the more easily they yield electrons. DCFC performance depends on disorder more than on carbon purity or even aggregate surface area. Extremely fine (10–100 \(\mu\)m) carbon particles with a high degree of structural disorder on the 30–100nm scale work well. Fine carbon particles, when mixed with molten carbonate to form a dense paste or slurry, operate like rigid electrodes when the melt is brought into contact with an inert metallic screen or graphite current collector.

5. DCFC Systems

Thermal cracking (decomposition) and pyrolysis processes can occur in a variety of reactors. There could be a good match between the hydrogen plasma black reactor (HPBR) and the DCFC for producing electric power and/or hydrogen and maximizing the power cycle efficiency\textsuperscript{12}. The HPBR combined with the DCFC and SOFC in a combined power cycle is an example of an integrated power plant for use with the DCFC which a high power cycle efficiency. The HPBR produces hydrogen and CO for the SOFC and carbon for the DCFC in an efficient manner. In the SOFC oxygen ion is transmitted through the SOFC ceramic membrane and oxidizes the CO and H\(_2\) to CO\(_2\) and H\(_2\)O with the production of DC power to complement the direct oxidation of carbon in the DCFC.

LLNL has designed a DCFC system that incorporates a self-feeding cell that can be refueled pneumatic with cleaned coal. The DCFC incorporates a

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Fig. 5 Thermal efficiencies of power plants Source: DOE
pyrolysis step that produces hydrogen (which can be used as fuel for a conventional fuel cell or as a chemical reactant), low-molecular-weight hydrocarbons and a nonagglomerating char that could potentially be reactive and conductive and usable in the DCFC.

One of the most efficient systems incorporate fuel cells and turbines - the fuel cell turbine hybrid. Fig. 5 shows the efficiencies of stationary and transportation systems. Hybrids are the highest efficiency power plants yet devised. Hybrids are combinations of energy conversion devices. Many combinations of energy conversion devices are possible, but the combination of the high-temperature fuel cell with the gas turbine is a very high efficiency one\textsuperscript{13,14}. In the direct fuel cell turbine hybrid concept the fuel cell replaces the turbine combustor. The turbine expands the high-temperature fuel cell exhaust.

Although the DCFC is capable of 100% energy conversion and 100% fuel utilization at any temperature, this efficiency and utilization will probably not be realized. Therefore, one would want to operate at in a hybrid system to take advantage of fuel cell underutilizations, irreversibilities and inefficiencies. The high exergy exhaust from the higher temperature DCFC is quite suitable to perform useful expansion work in a gas turbine.

The plant costs of various power plants have been projected by EPRI in Fig. 6\textsuperscript{15}. In this comparison DCFC long-term compares very favorably having the lowest plant cost projection. Specific applications for DCFC systems may exist in low-cost power, combined heat and power, and peak shaving/load-management applications in which DCFCs help utilities to balance supply and demand during periods of high electricity demand. For DCFC systems to be competitive, however, they must present an attractive alternative not only to today’s technologies, but to technologies that will be available when DCFCs are eventually commercialized.

6. Research and Development Needs

The major development required is for the anode and anode chamber, for all the modes of the DCFC. The reactor strategy - batch or flow - depends on the purity of the solid graphite particles.

In the SOFC-mode case special anode materials - tungsten carbide, vanadium carbide (VC), zirconia carbide, titanium carbide, nickel metal, and carbon - have been researched\textsuperscript{16}. Several transition metal carbides were investigated as anodes. Observed values of open circuit voltage of carbide anode SOFCs were found to be in the same order as the oxygen potentials calculated for the respective metal carbide/metal oxide equilibrium. All the carbide anodes showed better performance than graphite anode. No CO\textsubscript{2} was detected even at high oxygen potentials due to its reduction. The VC was able to best activate the carbon solids. Redox reactions of anodes play an important role in activating carbon. Oxidation of the anodes themselves hinders the electrochemical reactions. The CO evolution was not proportional to the current density due to consumption of oxygen for the oxidation of TiC (See Fig. 7).

US DOE-funded research has been conducted on V2O\textsubscript{5}-Pt catalysts for carbon oxidation anodes in DCFCs at the University of Akron and on carbon ionic conductors for use as electrolytes in novel DCFCs at Duke University\textsuperscript{14}.

For the molten carbonate DCFC type\textsuperscript{17} Selman has identified the following research needs, primarily dealing with the anode chamber:

1. Wetting characteristics of various types of carbon, in melts of variable basicity (cation composition, and OH-added ) and temperature

![Fig. 6 EPRI Capital Cost Comparison](image-url)

![Fig. 7 Outlet CO evolution rates from VC and TiC anodes as function of current density](image-url)
2. Spectroscopic characterization of molten carbonate in reducing medium, in contact with carbon, to establish identity of stable and semi-stable species
3. Molecular Dynamics calculations to establish the relative stability of carbon-carbonate-CO-CO₂ configurations in the melt
4. Monte-Carlo (stochastic) modeling of assemblies consisting of cells representing carbon particles, gas bubbles (CO₂, CO), and carbonate melt. Buoyant forces, interfacial forces, and electrochemical reactions to be accounted for, cell-to-cell.

Cell and stack area and height scale up, heat removal, performance degradation as a result of electrolyte contamination, and electrolyte purification and re-injection processes are also critical issues which need to be addressed for all DCFC technologies.

Other special R&D requirements vary with the developers technology and system development schemes identified in this paper.

7. Conclusions

In theory, the DCFC holds the highest promise of any fuel cell. An intense period of research would be required to develop the anodes and anode chamber for various designs. Carbon particle reactivity, impurity level and cost will play major roles in any eventual DCFC commercialization. It can hardly be expected that the DCFC development will precede the state-of-development of its base AFC, MCFC or SOFC technologies.

References

Mark C. Williams

Mark C. Williams received his Ph.D. in Materials Science and Engineering in 1985 from the University of California at Berkeley. Dr. Williams is the author of over two hundred papers and articles in various peer-reviewed technical journals, society proceedings, book chapters, encyclopedia chapters, and other publications. With Dr. Alan Lloyd, the California EPA Secretary, he received the inaugural U.S. Fuel Cell Council’s Pathfinder Award in 2003 for eighteen years of dedicated leadership to the fuel cell industry. In 2005 he became a Fellow of The Electrochemical Society “for sustained, internationally recognized contributions to and promotion of electrochemical energy conversion technologies, especially fuel cells”. In 2006 he was an ASM Visiting Lecturer to India. In 2007 he was a Japan Society for the Promotion of Science Fellow working with AIST on exergetic studies of intermediate temperature electrolytes. He has been active in helping formulate the fuel cell programs of the U.S. Department of Energy, Taiwan and Austria. Dr. Williams serves as on the Board of the Fuel Cell Seminar and on the Boards of Advisers of fuel cell companies. In addition, he serves as the Chief Technology Officer of an incipient fuel cell company. He is now Adjunct Professor, University of Utah. Mark can be reached at markcwilliams1@verizon.net.

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