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Molten Carbonate Fuel Cell Networks: Principles, Analysis, and Performance

Technical Note

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Preface

The project "Molten Carbonate Fuel Cell Networks: Principles, Analysis and Performance" was largely performed under the direction of Drs. J.F. Osterle and D.H. Archer, faculty members of Carnegie Mellon University's Department of Mechanical Engineering. An earlier report on this project was submitted in partial fulfillment of the requirements for the degree of Master of Science in Mechanical Engineering.

This project has also provided professional training within the Project Manager Development Program at the United States Department of Energy's Morgantown Energy Technology Center (DOE/METC). The concept of networking fuel cells is of interest to METC's fuel cell product team and process and project engineering branch.

Acknowledgments

Gratefully acknowledged are the contributions of the following individuals:

- **David H. Archer:** The initiator of this project, Dr. Archer first suggested the concept of placing fuel cell stacks in series. His expertise, guidance, and continual assistance were the cornerstone of this effort.
- **J. Fletcher Osterle:** The guidance which Dr. Osterle provided from his expertise in thermodynamics and electrochemistry was essential to the careful proof and clear explanation of this concept.

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1 Introduction

The U.S. Department of Energy's Morgantown Energy Technology Center (METC) sponsors the research and development of engineered systems that utilize domestic fuel supplies while achieving high standards of efficiency, economy, and environmental performance. One of the most promising electric power generation systems currently being sponsored by METC is the molten carbonate fuel cell (MCFC).

Presently, METC is striving to improve upon conventional MCFC system designs, in which MCFC stacks are typically arranged such that reactant streams flow among them in parallel. In an improved design, called an MCFC network, reactant streams are ducted such that they are fed and recycled among multiple MCFC stacks in series.

Before beginning an analysis of fuel cell networks, however, some introductory principles are presented.

2 Molten Carbonate Fuel Cells

2.1 Structure and Operation

Figure 1 illustrates an MCFC. The main components of an individual cell are the anode, the cathode, and the molten carbonate electrolyte. Electrode materials are usually porous nickel alloys for reducing atmospheres (anode) and nickel oxide for oxidizing atmospheres (cathode). The electrolyte, typically a combination of molten, alkali (Li, K, Na) carbonates, is contained within a porous ceramic matrix, commonly made of lithium aluminate (LiAlO_2). An individual cell is approximately 6 mm thick. The electrolyte is about 1 mm thick.

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Figure 1. Operation of an MCFC

The molten carbonate electrolyte, sandwiched between the anode and cathode, partially fills these porous electrodes. Electrochemical reactions take place at a three-phase interface formed by the electrolyte, the electrodes, and the gas streams. Carbonate ions are formed at an oxidant/electrolyte interface in the cathode and are transported through the electrolyte to a fuel/electrolyte interface in the anode. There, the carbonate ions react with the fuel, releasing electrons into the anode. The electrons then travel through an external circuit and through the

load, suffering a voltage drop. Finally, the circuit is completed as the electrons return to the cathode.

Although developers have produced various MCFC designs, the above describes the general operation of any MCFC. For analysis in this study, an MCFC designed to internally reform methane fuel was chosen. The following section describes the reactions that occur within an internally reforming molten carbonate fuel cell (IRMCFC).

2.2 IRMCFC Reactions

Methane Reforming: For an MCFC, methane must be reformed into hydrogen, which is more reactive. Within the anode gas chamber of an IRMCFC, methane is reformed into hydrogen and carbon monoxide.

This process eliminates the need for an external fuel processor and heat source, and is well-suited to take place inside the IRMCFC. Internal reforming directly utilizes the waste heat generated by the overall fuel cell reaction. In addition, excess steam produced by hydrogen oxidation is consumed by methane reforming. With the required catalyst, the conversion of methane happens quickly, approaching completion by the time 50 percent of the hydrogen has been utilized. According to Le Chatelier's principle, low pressure drives reactions (1-2) forward. Therefore, an IRMCFC is well-suited for operation near atmospheric pressure (Kinoshita, McLarnon and Cairns 1988, 82).

Anode Reactions: When the reformed fuels diffuse through the thin electrolyte film that wets the porous anode, a three-phase interface is created at which oxidation reactions (3-5) can occur.

For hydrogen, diffusion to the electrode/electrolyte interface can also take place through the anode, since hydrogen is sufficiently soluble in many metals (Selman 1963). The gaseous fuels react with molten carbonate ions in the electrolyte, releasing electrons directly into the anode. The products, carbon dioxide and steam, diffuse back to the electrolyte/gas interface and into the anode gas chamber. The freed electrons are conducted through the external circuit to the cathode. The diffusion of gaseous reactants through the electrolyte film to an electrode surface is usually the transport process which imposes an upper limit on the rate of electrochemical reaction (Kinoshita, McLarnon and Cairns 1988, 7).

Water-Gas Shift: Hydrogen is oxidized in the anode much faster than carbon monoxide. The steam produced by hydrogen oxidation is consumed by methane reforming, reactions (1-2), and reaction (6), the water-gas shift.

The water-gas shift reaction occurs very rapidly at IRMCFC operating temperatures (Kinoshita, McLarnon and Cairns 1988, 72). As hydrogen is depleted, the reaction is accelerated, since steam and carbon dioxide are produced in equal amounts at the anode. Essentially all the carbon monoxide is shifted to hydrogen before it can react at the anode. Consequently, only hydrogen is considered to be electrochemically active, and reaction (5) is omitted from the anode reactions (Appleby and Foulkes 1989, 541).

Carbon Deposition: Carbon deposition in an IRMCFC, which can plug gas passages in the anode chamber, must be prevented. There are four major reactions by which carbon deposition can occur (Shah 1988, 15):

Maintaining a high steam content in the anode ensures that carbon monoxide will be oxidized in the water-gas shift reaction. Consequently, when the ratio of carbon dioxide to carbon monoxide is kept high enough, carbon deposition by the Boudouard reaction (7) is prevented (Appleby and Foulkes 1989, 541). Operating at high temperatures also discourages the Boudouard reaction, since its equilibrium constant decreases rapidly with increased temperature (Kinoshita, McLarnon and Cairns 1988, 148).

Cathode Reaction: Air, which is commonly used as the oxidant for an IRMCFC, supplies oxygen and, unfortunately, the diluent nitrogen to the cathode. To supply the flow of

carbonate ions, the air oxidant must be enriched with carbon dioxide. Reaction (11) occurs when oxygen and carbon dioxide diffuse to the cathode and combine with electrons to form carbonate ions. The carbonate ions complete the electrical circuit by diffusing through the electrolyte to the anode.

Overall IRMCFC Reaction: The overall fuel cell reaction can be obtained by considering the above reactions, which occur simultaneously. Concentrations of the seven anode reactants can be fully described by a set of independent equations that contains each of them. Anode reactions (1,3,6,7) are independent, contain all the anode reactants, and can be combined to form the remaining anode reactions. (Independent reactions are starred [★].) Assuming that these reactions are kept in equilibrium, one can address the electrochemical reactions separately. When the single, independent cathode reaction (11) is combined with the anode reaction (3), the electrons and carbonate ions cancel and the overall IRMCFC reaction results:

where the subscripts a and c denote the anode and cathode gas chambers, respectively.

In aqueous electrolytes, ionic concentration gradients can exist without violating the principle of electroneutrality. However, the subject electrolyte is not aqueous, but a combination of molten, alkali (Li, K, Na) carbonates contained within a porous ceramic matrix. Within the matrix, there is only pure alkali carbonates (nothing is present for dilution), and electroneutrality demands that the concentration of carbonate ions is the same throughout.

Carbon dioxide was not canceled in the overall reaction because its concentration at the anode is not necessarily equivalent to its cathode concentration. Gas partial pressures, and therefore gas concentrations, become important when calculating the Nernst potential.

2.3 Thermodynamics

For a fuel cell, the first law of thermodynamics can be used to relate the decrease in enthalpy, ΔH , to the electrical work generated, W , and the heat absorbed, Q :

$$\Delta H = Q - W. \quad (13)$$

The decrease in enthalpy can be expressed in terms of the fuel cell's operating temperature, T , and the changes in entropy, ΔS , and free energy, ΔG :

$$\Delta G + T\Delta S = Q - W. \quad (14)$$

Free Energy: A fuel cell converts a portion of the free energy of reaction directly into electrical work. Free energy that is not converted to work is released as heat. However, when a fuel cell operates reversibly, all the free energy of reaction is converted into electrical work. The only heat absorbed by a reversible cell is the product of cell temperature and entropy change.

$$\begin{aligned} W_{REV} &= -\Delta G. \\ Q_{REV} &= T\Delta S. \end{aligned} \quad (15, 16)$$

A reversible fuel cell must operate completely in equilibrium. Every reaction throughout the entire cell must be reversible, and no concentration gradients can exist in the gas streams. Since current cannot exist without concentration gradients, the number of electrons transferred in the overall fuel cell reaction (12) must be considered virtually infinitesimal.

When calculating the change in free energy for the overall reaction, the standard change in free energy for each gaseous reactant must be corrected for nonstandard pressure (standard pressure is one atmosphere, or 101.325 kPa). The partial pressure of each gaseous reactant is the product of its mole fraction and the total chamber pressure.

$$\begin{aligned} \Delta G = & G_{(H_2O),a}^{\circ} + RT \ln (X_{(H_2O),a} P_a) \\ & + G_{(CO_2),a}^{\circ} + RT \ln (X_{(CO_2),a} P_a) \\ & - G_{(H_2),a}^{\circ} - RT \ln (X_{(H_2),a} P_a) \\ & - \frac{1}{2} G_{(O_2),a}^{\circ} - \frac{1}{2} RT \ln (X_{(O_2),a} P_a) \\ & - G_{(CO_2),c}^{\circ} - RT \ln (X_{(CO_2),c} P_c) , \end{aligned} \quad (17)$$

where: G° = standard free energy, $\left(\frac{J}{\text{gmol CO}_2^{\circ}}\right)$,

R = Universal Gas Constant, $\left(8.314 \frac{J}{K \text{ gmol CO}_2^{\circ}}\right)$,

T = cell temperature, (Kelvin),

X = mole fraction,

P = total pressure of anode or
cathode gas chamber (atm).

The change in standard free energy for the IRMCFC overall reaction can be written as an empirical function of the cell operating temperature (Shah 1988, 10):

$$\begin{aligned} \Delta G^{\circ} = & G_{(H_2O),a}^{\circ} + G_{(CO_2),a}^{\circ} - G_{(H_2),a}^{\circ} - \frac{1}{2} G_{(O_2),a}^{\circ} - G_{(CO_2),c}^{\circ} \\ = & 4186 [-46.005 + 0.01305 (T-1000)] , \end{aligned} \quad (18)$$

where: ΔG° = change in standard free energy, $\left(\frac{J}{\text{gmol CO}_2^{\circ}}\right)$,

T = cell temperature, (Kelvin).

Combining equations (17) and (18):

$$\Delta G = \Delta G^\circ + RT \ln \frac{[P_a X_{(H_2O)} X_{(CO_2)}]}{[(P_c)^{3/2} X_{(H_2)} (X_{(O_2)})^{1/2} X_{(CO_2)}]} \quad (19)$$

Electrical work is the product of charge and the voltage at which the charge is transferred. The overall reaction for the reversible IRMCFC can be based on a virtual transfer of one gram-mole of carbonate ions. Each carbonate ion carries two gram-moles of electrons. The charge per gram-mole of electrons is given by Faraday's number.

$$\begin{aligned} W_{REV} &= [\text{charge transferred}] [\text{voltage}] \\ &= [(1 \text{ gmol CO}_3^{2-}) (2 \frac{\text{gmol electrons}}{\text{gmol CO}_3^{2-}}) (F \frac{\text{coulombs}}{\text{gmol electrons}})] [E_{REV} \text{ volts}] \\ &= 2FE_{REV} \text{ joules} , \end{aligned} \quad (20)$$

where: F = Faraday's number

$$\begin{aligned} &= (6.022045 \text{ E}+23 \frac{\text{electrons}}{\text{gmol electrons}}) (1.6021892 \text{ E}-19 \frac{\text{coulombs}}{\text{electron}}) \\ &= 96,485 \frac{\text{coulombs}}{\text{gmol electrons}} . \end{aligned}$$

Nernst Potential: The Nernst potential is the force that drives reversible electrode reactions in equilibrium conditions. This force, or reversible voltage, is generated by the overall cell reaction, and is a function of the local temperature, pressure, and reactant concentrations. The Nernst potential is the maximum voltage that can be generated from the overall cell reaction. Combining equations (15), (19), and (20) yields a formula for the Nernst potential:

$$E_{REV} = -\frac{\Delta G^\circ}{2F} - \frac{RT}{2F} \ln \frac{[P_a X_{(H_2O)} X_{(CO_2)}]}{[(P_c)^{3/2} X_{(H_2)} (X_{(O_2)})^{1/2} X_{(CO_2)}]} ,$$

where: E_{REV} = Nernst Potential, (volts), (21)

X = equilibrium mole fractions,

P_a = equilibrium anode chamber pressure,

P_c = equilibrium cathode chamber pressure.

As reactants are utilized, their concentrations change. Since the Nernst potential is strongly dependent upon the concentrations of reactants, it varies with the degree of utilization. An IRMCFC was simulated with fuel utilizations ranging from nearly 0 to 99 percent. (In this study, fuel utilization is defined as the fraction of the inlet fuel stream's oxygen

demand that is satisfied in a single pass through a cell.) Cell temperature, fuel feed (flowrate and composition), and oxidant feed were the same for each simulation. For each fuel utilization, the reactant exhausts were brought to equilibrium and the Nernst potential was calculated. At the cell outlet, the final reaction was assumed to be reversible. These data are plotted in Figure 2, which shows how the Nernst potential varies with fuel utilization.

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Figure 2. Nernst Potential (Constant Reactant Flow)

2.4 Irreversibilities

One condition for the reversible operation of a fuel cell was virtually zero current density. Of course, no work can be extracted at zero current density. Practical IRMCFCs efficiently generate work by drawing a significant current at reduced voltage. (Increased voltages are obtained with IRMCFC stacks, i.e., many cells connected together electrically in series.) When a large current is drawn, concentration gradients form in the gas streams and the cell does not operate reversibly. Consequently, portions of the Nernst potential, the fuel cell's driving force, must be used to overcome these irreversibilities. These portions of the Nernst voltage, which are internally sacrificed to drive the cell current, are commonly called voltage polarizations or overpotentials. The irreversibilities that the Nernst potential must overcome are typically categorized into three processes:

Ohmic Polarization (η_o): Ohmic resistance is the sum of electronic, ionic, and contact resistances. The ohmic overpotential is consumed as it overcomes resistance to the flow of carbonate ions through the electrolyte and resistance to the flow of electrons through the electrodes and the terminal connections.

Concentration Polarization (η_c): The transport of species to and from the electrochemical reaction sites must occur through the gas streams and the electrodes. Diffusion and convection limitations cause the effective concentrations at the electrochemical reaction sites to be less than their bulk concentrations in the gas chambers. This reduces the electrochemical activity at the electrode. The portion of the Nernst voltage used to overcome these irreversibilities is labeled the concentration overpotential.

Activation Polarization (η_a): Irreversible electrode reactions can result in losses at the electrode-electrolyte interface. To prevent electrons from being transferred too slowly to sustain the flow of the local cell current, part of the electrode voltage is consumed to maintain an adequate charge transfer rate. The electrode potential used to overcome this activation barrier is called the activation overpotential (Kinoshita, McLamon and Cairns 1988, 11-13; Appleby and Foulkes 1989, 21-26).

Each local Nernst potential within a cell is reduced to the cell's output voltage by the sum of the local overpotentials. The magnitudes of these local overpotentials are strongly dependent upon the local current density. Figure 3 illustrates how local ohmic, concentration and activation overpotentials reduce the local Nernst potential as a function of the local current density (Liebhafsky and Cairns 1968, 93-100). The activation overpotential rapidly increases as the current density initially rises from zero, but then its sacrifice of the local Nernst potential remains essentially constant. The Nernst voltage consumed by ohmic resistance increases linearly over the entire range of current densities. At the highest current densities, consumption of the Nernst voltage by the concentration overpotential becomes significant. As the local current density approaches some limiting value, the concentration overpotential consumes nearly all of the local Nernst potential.

When combined, the overpotentials reduce the local Nernst potential to the cell's output voltage. Figure 3 also shows that a cell's output voltage decreases as its local current densities increase.

2.5 Output Voltage

The output voltage of an IRMCFC is equal to the potential difference between its anode and cathode. Since the cell electrodes are very good conductors, their surfaces must be virtually isopotential. That is, the voltage of each electrode must be the same everywhere within the cell. However, within an IRMCFC, local Nernst potentials drive nearby electrode reactions at different rates, producing different current densities. Each local Nernst potential is reduced to the cell's single output voltage by local overpotentials that are proportional to the local current density. Therefore, a cell's output voltage can never exceed its minimum local Nernst potential. In fact, a cell's output voltage is always less than its minimum local

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Figure 3. Local Overpotentials

Nernst potential unless it happens that the local current density is virtually zero where the Nernst potential is a minimum. So, although the Nernst potential varies within a cell, all power is generated at a voltage less than or equal to the minimum local Nernst potential (assuming steady state).

A general expression for the output voltage of an IRMCFC can be expressed as

$$E_{cell} = (E_{REV})_{min} - (\Sigma\eta)_{local} \leq (E_{REV})_{min} , \quad (22)$$

where: $(E_{REV})_{min}$ = minimum local Nernst Potential,
 $(\Sigma\eta)_{local} = (\eta_a + \eta_o + \eta_c)$, overpotentials local to $(E_{REV})_{min}$.

Since local conditions within a practical cell vary greatly, locating the point at which the minimum Nernst potential is generated is not straightforward. As reactants are utilized within a stack, gas compositions change and different current densities are generated.

Temperature variations develop. Along the directions that fuel and oxidant flow, the concentrations of reactants decrease, while concentrations of products increase. A drop in the total pressure also occurs as reactant streams flow through the cell.

These directional gradients make conditions within a cell strongly dependent upon its internal flow geometry, i.e., the arrangement of reactant flows across the faces of individual cells. Internal flow geometries, which are cross-flow or countercurrent, result in an extremely complicated distribution of conditions within a cell. The simpler co-current flow geometry makes analysis of conditions within a cell more manageable.

As reactants are utilized within a cell with co-current internal flow, their concentrations decrease along the same direction. Reactant concentrations and pressure are a maximum at the inlet and a minimum at the outlet. Since the Nernst potential decreases with reactant concentration and pressure, the minimum value in a cell with co-current internal flow is generated at the outlet. If conditions were such that the very last electrochemical reaction at the outlet proceeded reversibly (zero current density), no overpotentials would have to be overcome there ($\Sigma\eta=0$), and the exact output cell voltage could be calculated with equation (22) using outlet conditions.

In reality, however, the last electrochemical reaction at the outlet of a co-current IRMCFC is not reversible. The actual cell output voltage is reduced from the outlet Nernst potential by various overpotentials. Activation and concentration polarizations combine to reduce the output voltage by increasing the anode voltage and decreasing the cathode voltage. Ohmic polarization further reduces the output voltage according to Ohm's law, creating a voltage drop proportional to the outlet current density. For a specific cell operation, the sum of these outlet overpotentials can be estimated from experimental data. Therefore, the output voltage of an IRMCFC with co-current internal flow can be written

$$E_{\text{cell}} = (E_{\text{REV}})_{\text{outlet}} - (\Sigma\eta)_{\text{outlet}} \quad (23)$$

Since a single cell produces a large current at a low voltage, it is necessary to connect multiple cells electrically in series to attain a practical voltage. Individual cells connected in such a way are referred to collectively as a fuel cell stack. The output voltage of an IRMCFC stack is the sum of its cell voltages:

$$E_{\text{stack}} = \Sigma E_{\text{cell}} \quad (24)$$

3 MCFC Stacks

3.1 Structure of an MCFC Stack

Figure 4 illustrates the structure of an MCFC stack. Conductive, bipolar separator plates connect individual cells in a stack both structurally and electrically. Made of stainless steel, each bipolar separator plate physically separates the fuel gas stream of one cell from the oxidant gas stream of the adjacent cell. One side of each separator plate channels a fuel stream so that it flows over a porous anode, while the flip side channels an oxidant stream over a porous cathode. Each bipolar separator plate also collects current, connecting adjacent cells of a stack electrically in series. From the anode, electrons are conducted through the bipolar separator plate and into the cathode of the adjacent cell. There, they react with the oxidant gas stream and carbonate ions are formed. The carbonate ions diffuse through the electrolyte and into the anode, where they react with the fuel gas stream, releasing electrons into the anode. Electrons are conducted in this manner through all the cells, establishing direct current (dc) through the stack. An external circuit connects a load between the two endplates of the stack, completing the circuit.

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Figure 4. Structure of an MCFC Stack

3.2 Internal Flow Geometry

Figure 4 depicts an MCFC with the fuel and oxidant streams flowing perpendicular to each other within the cell. This internal flow geometry is known as cross-flow. Other possible internal flow geometries are co-current and countercurrent. In general, a countercurrent internal flow geometry produces the most uniform current densities and temperature distributions within a cell. A co-current internal flow geometry, in general, results in the least uniform current densities and temperature distributions.

3.3 Power Conversion

The dc power that an MCFC stack produces can be used, without conversion, for some applications, such as transportation and chemical processing. However, for most purposes it needs to be converted to ac (alternating current) by a power conditioning unit. If an MCFC power plant is composed of multiple stacks, they could be connected electrically to the power conditioning unit in series, in parallel, or independently. If connected in series, each stack would have the same current; if connected in parallel, each stack would have the same voltage.

3.4 Distribution of Reactants to Cells Within a Stack

Since cells in a stack are connected electrically in series, the same current must flow through each one. To achieve the stack current, each cell must transfer an equivalent number of carbonate ions across its electrolyte. Therefore, it is ideal if the fuel and oxidant feed streams are evenly distributed to each cell within a stack. Today's manifolds, however, have not been able to achieve this, and cells are typically supplied with unequal reactant flows. Such an irregular distribution of reactants results in decreased stack voltage, especially at higher utilizations. To illustrate this, consider Figure 5.

The two-cell stacks in Figures 5(a) and 5(b) are each fed with 100 moles of fuel per hour. To achieve the required stack current, each of the two cells must react 45 moles of fuel per hour. Since 90 of the 100 moles of fuel per hour must be reacted, the target stack fuel utilization is 90 percent. The composition of the fuel and oxidant which is fed to the two-cell stacks is the same that is described in Figure 2. Figure 5(c), a curve identical to Figure 2, gives the Nernst potential as a function of fuel utilization for both stacks.

Ideally, fuel and oxidant is evenly distributed among each cell in a stack. Such is the case in Figure 5(a). Fifty moles of fuel per hour is distributed to each cell in the stack. Each cell is also provided with 10 percent excess air enriched with stoichiometric carbon dioxide. To achieve the required current, each cell reacts 45 moles of fuel per hour. Consequently, both cells also attain the target fuel utilization of 90 percent. Figure 5(c) shows that each cell which utilizes 90 percent of its fuel generates a Nernst potential of 0.7474 volts. Adding the two cells' potentials, the maximum output voltage the stack could generate is 1.4948 volts.

(a) Ideal Flow Distribution

(b) Irregular Flow Distribution

(c) Effect of Flow Variability

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Figure 5. Voltage Reduction Due to Flow Variability

The stack in Figure 5(b) is not ideal. The fuel feed is distributed between the two cells with 8 percent variability. One cell receives too little fuel, 46 moles per hour, while the other receives a surplus, 54 moles per hour. For simplicity, a proportional amount of carbon dioxide enriched air (10 percent excess) is also provided to each cell. To achieve the required current, each cell reacts 45 moles of fuel per hour. The fuel utilization of the cell that received too little fuel, 97.8 percent, is much higher than the target utilization. Conversely, the cell that received a surplus of fuel has a fuel utilization much lower than the target utilization, 83.3 percent. The voltages of the two cells can be obtained from Figure 5(c). The cell with surplus fuel generates an elevated voltage of 0.7897 volts. However, the cell with too little fuel generates only 0.6496 volts, a decreased voltage. As can be seen in Figure 5(c), the voltage loss is greater than the voltage rise. The combined voltages produce a maximum stack output voltage of 1.4393 volts, which is 0.0555 volts lower than the ideal case. Consequently, the maximum power that the fuel cell stack could generate is decreased by nearly 4 percent.

For higher fuel utilizations, the net effect of irregular flows is a voltage loss. Although some cells in a stack with irregular flows produce increased voltages, they cannot compensate for the decreased voltages generated by the remaining cells. The consequences of flow variability get much more severe as fuel utilization approaches 100 percent. Unfortunately, this serves to discourage operation at such utilizations.

3.5 MCFC Stacks Developed in the United States

Currently, there are three companies in the United States that are developing MCFC stacks: MC-Power Corporation (MCP), International Fuel Cells (IFC), and Energy Research Corporation (ERC) (Hooie et al. 1992, 18-20). Although the MCFC stacks that they are developing are made of similar materials and have the same basic structure, they differ in three basic ways: how fuel is reformed; the stack operating pressure; and the way reactant streams are distributed to individual cells (manifolding).

MCP Stack: Fuel is externally reformed in a vessel separate from the MCP stack. The MCP stack is usually pressurized, operating at 2 to 3 atmospheres (200 to 300 kPa). The MCP stack is internally manifolded. Four manifolds, one in each corner, extend down through the stack. Reactant streams flow into the cells from the fuel and oxidant intake manifolds and are channeled across the face of each cell in a complex flow geometry. Spent fuel and oxidant flow from the cells into the exhaust manifolds and exit the stack. MCP has tested a 19-cell, 1.06 m² (11.4 ft²) stack, which is nominally rated at 20 kW.

IFC Stack: The IFC stack is externally reformed and is typically operated at nearly 6 atmospheres (608 kPa). The IFC stack is externally manifolded. Intake manifolds for fuel and oxidant are sealed to adjacent sides of the stack. Exhaust manifolds are sealed to the opposite sides of the stack. IFC has tested a 20-cell, 0.74 m² (8 ft²) stack, which is nominally rated at 20 kW.

ERC Stack: The ERC stack is externally manifolded and operates near one atmosphere (101 kPa). A catalyst impregnated within the current collectors (bipolar separator plates) allows methane to be internally reformed. Since the endothermic reforming reaction consumes some of the heat that is generated within a stack, this helps to control stack temperature. ERC has tested a 234-cell, 0.37 m² (4 ft²) stack, which is nominally rated at 70 kW. An IRMCFC stack was the design chosen for analysis in this study.

4 Configurations of Multiple MCFC Stacks

4.1 Parallel Stack Configuration

In conventional fuel cell systems, multiple stacks have been arranged in parallel with regard to the flow of reactant streams. As illustrated in Figure 6(a), the initial oxidant and fuel feeds are divided into equal streams, which flow in parallel through the fuel cell stacks.

(a) Parallel Flow of Reactant Streams Through Stacks

(b) Series Flow of Reactant Streams Through Stacks

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Figure 6. Conventional and Networked Flow Configurations

Conventional fuel cell configurations avoid high reactant utilizations because they produce low voltages. In conventional configurations, the single-pass utilization (excludes recycle) of each reactant stream is accomplished entirely within one stack. Therefore, when high utilizations are attempted, the low voltage that is generated adversely affects the total power production. This limits the reactant utilization that can be practically attained.

As previously discussed, manifolding problems also limit the practical reactant utilization of conventional MCFC systems. Ideally, fuel and oxidant streams are distributed

equally among individual cells in a stack. Today's manifolds, however, have not been able to achieve this, and cells are typically supplied with unequal reactant flows. This causes the composition of outlet reactant streams to be variable among the cells. At high utilizations, this variability leads to a reduction in stack voltage. Therefore, conventional systems avoid such high utilizations.

4.2 Series Stack Configuration

"Networking" is ducting reactant streams such that they are fed and recycled through fuel cell stacks in series. Figure 6(b) illustrates how the reactant streams in a fuel cell network flow in series from stack to stack. By networking fuel cell stacks, increased efficiency, improved thermal balance, and higher total reactant utilizations can be achieved. Networking also allows reactant streams to be conditioned at different stages of utilization. Between stacks, heat can be removed, streams can be mixed, and additional streams can be injected.

4.2.1 Flow Geometry Between Stacks in Series

Fuel and oxidant streams can flow co-currently or countercurrently through stacks networked in series, as shown in Figure 7(a) and (b). The flow geometry of reactant streams between stacks should be distinguished from the internal flow geometry of reactant streams across the face of an individual cell, which can be co-current, cross-flow, or countercurrent.

(a) Co-Current Flow of Fuel and Oxidant Through Stacks

(b) Countercurrent Flow of Fuel and Oxidant Through Stacks

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Figure 7. Flow Geometries Between Networked Stacks

Since the overall IRMCFC reaction is exothermic, the temperatures of the fuel and oxidant streams generally increase with reactant utilization. In addition, electrochemical potential, and therefore current density, decreases with reactant utilization. When fuel and oxidant streams flow co-currently between stacks, their temperatures increase together and their electrochemical potentials decrease together. This tends to create non-uniform temperature and current density distributions between stacks networked in series. However, when the fuel stream flows countercurrent to the oxidant stream, temperature and current density distributions are more uniform. A uniform temperature distribution allows stacks to be operated nearer their maximum operating temperature. This reduces the total voltage polarization, which generally decreases as temperature increases.

4.2.2 Stacks in Series Approach Reversibility

MCFC stack networks produce more power than conventional configurations because they more closely approximate a reversible process. To illustrate this fact, consider Figure 8, which compares the maximum power which could be generated by three different IRMCFC systems: A, B, and C.

Figure 2, the graph of Nernst potential versus fuel utilization, was duplicated three times in Figure 8. Fuel utilization is directly proportional to the charge transferred across the electrolyte. Therefore, the shaded areas of the graphs represent power, i.e., the product of voltage and current. Since reversibility is assumed at the outlet of each stack, no voltage losses are deducted from the Nernst potential. Therefore, each shaded area represents the maximum power that each cell could actually generate.

System A in Figure 8 is composed of a single stack. Three stacks are arranged in series in System B. System C features many, or "n," stacks configured in series. For comparison, each system is assumed to have the same total stack membrane area. That is, the area of each stack in System B is one third the area of the stack in System A. Similarly, the area of each stack in system C is one "nth" the area of the single stack in System A. For simplicity, each stack is considered to contain only one cell.

Each of the three systems is fed the fuel and oxidant streams described in Figure 2. Since each system achieves the same total fuel utilization (90 percent) across the same total area, each stack has the same average current density. Irreversible voltage loss is mainly a function of current density and stack temperature. Since these parameters are equivalent in each stack, it is assumed that each stack would suffer the same voltage losses. Each cell's outlet Nernst potential would be reduced by the same amount (equation [23]).

In System A, 90 percent of the fuel is utilized in a single stack, and all the current is generated at a single voltage. The power that this system can achieve is represented by the graph's shaded region.

In System B, three stacks in series each utilize 30 percent of the fuel. The current generated by each stack in System B is one third of the current generated in System A. Each stack in System B produces a different voltage. At the exit of the first stack, a high Nernst

System A - One Stack

$$P_{MAX} = I E_{REV}$$

$$P_{MAX_1} = (0.75) I$$

System B - Three Stacks

$$P_{MAX_3} = (0.98 + 0.86 + 0.75) \frac{I}{3}$$

$$P_{MAX_3} = (0.87) I$$

System C - "n" Stacks

$$P_{MAX_n} = (V_1 + V_2 + \dots + V_n) \frac{I}{n}$$

$$\lim_{n \rightarrow \infty} P_{MAX_n} = P_{REV}$$

Where: E_{REV} = Nernst Potential

$$I = nFN_f U_f$$

P_{REV} = Power generated by the system if every point in each stack was reversible and all the free energy was converted to electrical work

P_{MAX} = Power generated by the system if the outlet of each stack was reversible

Additional power generated by networking IRMCFC stacks in series

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Figure 8. Stacks in Series Approach Reversibility

potential is generated because 70 percent of the fuel is still unreacted. Likewise, at the exit of the second stack, 40 percent of the fuel remains unreacted, generating another improved Nernst potential. Only 10 percent of the fuel remains at the exit of the third stack, yielding the same Nernst potential that the single stack in System A produced. The three-stack network can produce more power because two-thirds of the total charge is transferred at increased voltages. Comparing the shaded areas of the graphs illustrates the additional power that can be produced by arranging stacks in series.

In System C, many stacks are connected in series. Very small currents are generated at still higher voltages. As the number of stacks in series is increased, the maximum achievable power quickly approaches the power that a reversible system would generate, i.e., complete conversion of the available free energy. (A reversible system is reversible at every point in each stack, not just at the stack outlets.) The shaded area in the graph nearly fills the entire area under the curve - the reversible power.

Each system in Figure 8 converts an equivalent amount of free energy (90 percent fuel utilization) into heat and electrical work. The key difference, however, is that the systems with MCFC stacks networked in series transfer charge at higher voltages, thus converting more of the free energy directly into electrical work and less into heat. As the number of stacks in series is increased, a reversible process is approached, which would convert all the free energy into work and none into heat. Although heat that is produced from free energy can be reconverted into electrical work (e.g., via a steam turbine), an MCFC stack's direct conversion of free energy is intrinsically more efficient. Therefore, networking MCFC stacks in series results in more efficient power production even when waste heat is recovered.

Although each stack added to a series network will improve the system's efficiency, the incremental benefit obtained with each additional stack diminishes. A finite number of stacks could adequately, but not exactly, approach a reversible process. In a practical network, the number of stacks would be limited by economic, space, and design constraints.

In a similar paper study, Liebhafsky and Cairns (1968, 111-116) compared two arrangements of tubular, calcia-stabilized solid oxide fuel cells. In one arrangement, hydrogen and air were supplied to a single, 30-cm cell. In the other arrangement, the same cell was segmented into three, 10-cm cells that were ducted such that the same reactant streams flowed through them in series. Each arrangement had a total fuel utilization of 90 percent and each cell had the same average current density. Each cell in the series arrangement accomplished one third of the total fuel utilization. Calculations showed that the series arrangement produced 5 percent more power than the single cell, and that further sectioning would produce greater improvements. It was concluded that the increase in irreversibility associated with changes in gas composition has nothing to do with electrode kinetics, but is rooted in the Nernst equation.

4.2.3 Reactant Conditioning Between Stacks in Series

When MCFC stacks are networked in series, reactant streams can be conditioned between the stacks at different stages of utilization.

The composition of reactant streams can be optimized between stacks by injecting a reactant stream (Figure 9), or by mixing the existing reactant streams. As previously discussed, the uneven distribution of reactants to cells within a stack produces variability in the compositions of outlet reactant streams. This leads to a reduction in stack voltage. However, when stacks are networked in series, reactant streams can be thoroughly mixed between cells. This reduces the variability in reactant composition and helps to minimize the stack voltage loss.

Between stacks networked in series, heat can be removed from the reactant streams (Figure 9). This may be done to control stack temperature. Or, the heat may be recovered for a combined cycle or for cogeneration. The heat in a network reactant stream can be transferred to a cooler stream in a heat exchanger, or hot and cold reactant streams can be mixed directly.

Figure 9. Conditioning Reactant Streams Between Stacks in Series

Methane can be injected into fuel streams between stacks networked in series (Figure 9). Since the reforming of methane into hydrogen is endothermic, its careful distribution among stacks in series is expected to improve the thermal balance of the system by allowing waste heat to be more evenly consumed throughout the total utilization of reactants. Improved thermal balance should allow stacks to be operated nearer their maximum temperature, reducing ohmic voltage losses. However, injecting portions of the fuel feed between stacks in series decreases the Nernst potential of every stack except the last one, since less fuel passes through each stack. (The amount of fuel that passes through the last stack does not change.) System optimization will require evaluation of whether the benefits of improved thermal balance outweigh the reduction in Nernst potential associated with such fuel redistribution.

4.2.4 Higher Total Reactant Utilizations

The optimum total reactant utilization of stacks networked in series is higher than that of conventional, parallel stacks. Conventional designs avoid high utilizations because they produce low voltages. In conventional configurations, the total utilization of reactants is accomplished in one stack. Therefore, when high utilizations are attempted, the low voltage that is generated adversely affects the total power production. In networks, however, the utilization of reactants is accomplished incrementally, and the low voltage associated with high utilization is restricted to stacks that produce only a portion of the total power.

High utilizations are also avoided by conventional MCFC systems because of flow variability. At high utilizations, flow variability can lead to a reduction in stack voltage. When stacks are networked in series, however, the composition of reactant streams can be optimized between stacks by injecting a new stream or by mixing the existing reactant

streams with one another. This reduces the variability in reactant composition and helps to minimize the stack voltage loss, allowing higher utilizations to be safely approached.

Another study (Walhood and Ferguson 1992) maximized the efficiency of conventional and series-connected fuel cell systems by optimizing cell voltage and current density. The study found that the optimum fuel utilization in the series-connected system was higher than that in the conventional system. Most importantly, the higher fuel utilization and lower current density of the series-connected system combined to give more efficient performance than the conventional system.

5 IRMCFC Power Systems

5.1 Requirements and Options of an IRMCFC System

When designing an IRMCFC power system, several requirements must be met. An IRMCFC system must properly condition both the fuel and oxidant gas streams. Methane must be reformed into the more reactive hydrogen and carbon monoxide. Carbon deposition, which can plug gas passages in the anode gas chamber, must be prevented. To supply the flow of carbonate ions, the air oxidant must be enriched with carbon dioxide. Both oxidant and fuel feed streams must be heated to their proper inlet temperatures. Each IRMCFC stack must be operated within an acceptable temperature range. Excess heat generated by the IRMCFC stacks must be recovered and efficiently utilized. Acceptable environmental performance, such as suppression of nitrogen oxides, must be ensured. The following is a discussion of these requirements as well as some options for meeting them.

5.1.1 Steam Reforming

Within an IRMCFC stack, near the anode inlet, the methane reforming reactions consume steam faster than hydrogen oxidation produces it. Generally, the methane is completely reformed before 50 percent of the hydrogen has been oxidized. This creates a deficiency of steam in the fuel near the IRMCFC anode inlet and an excess of steam at the outlet. The steam needed at the anode inlet of an IRMCFC for methane reforming can be provided by recycling a portion of the anode exhaust. Alternatively, steam from other sources, such as a heat recovery steam generator, can be injected into the inlet fuel stream.

5.1.2 Preventing Carbon Deposition

Carbon deposition must be prevented in an IRMCFC stack because soot can plug gas passages in the anode gas chamber. If the fuel stream is maintained at a high temperature and supplied with sufficient steam, solid carbon will not be formed. Steam can be recycled through the anode from the anode exhaust or injected into the inlet fuel stream from some other source. Temperatures within the normal operating range of an IRMCFC are usually adequate to prevent carbon deposition if ample steam is present in the fuel.

5.1.3 Carbon Dioxide Enrichment

Air, which is used as the oxidant for an IRMCFC system, must be enriched with carbon dioxide. In the cathode, each reacting oxygen molecule requires two molecules of carbon dioxide to form the carbonate ion. In effect, the carbonate ion simply transfers carbon dioxide across the electrolyte from the oxidant stream to the fuel stream. Therefore, it is logical to enrich the air oxidant with carbon dioxide by recycling a portion of the IRMCFC anode exhaust. If an efficient separation technique existed, dilution of the oxidant stream could be avoided by recycling pure carbon dioxide from the anode exhaust.

5.1.4 Achieving Stack Inlet Temperatures

Both oxidant and fuel streams must enter the IRMCFC stack within an acceptable temperature range, perhaps even at some specific design temperatures. Generally, both the fuel (natural gas) and oxidant (air) feeds are near ambient temperature and need to be heated. Heating can be accomplished by mixing the feeds with hot gases, such as the exhaust from a stack or combustor, or by passing the feeds through heat exchangers.

5.1.5 Management of Stack Temperature

Due to certain structural limitations of an IRMCFC stack, its operating temperature must be kept within an acceptable range. A lower temperature limit, the freezing point of the molten carbonate electrolyte, ≈ 825 K (≈ 1000 °F), must not be approached. Near this freezing point, the ionic resistivity of the electrolyte is unacceptably large. An upper temperature limit, ≈ 970 K (≈ 1290 °F), must not be exceeded because of problems with various stack materials. High temperatures, for example, accelerate electrode corrosion and electrolyte mobility (leakage) and evaporation (Kinoshita, McLarnon and Cairns 1988, 24). Since high temperatures are favored for reactant conditioning and stack efficiency, it is the upper temperature limit that is more restrictive.

The overall reaction that takes place within an IRMCFC stack is exothermic. To maintain an acceptable operating temperature, excess heat must always be removed from the stack or consumed by endothermic fuel reforming. To accomplish this, three methods are conceivable: internal heat exchange, carrying heat away with reactant streams, and methane reforming.

The conventional method of IRMCFC stack temperature control relies on the reactant streams to carry heat out of the stack. Once the hot reactant streams exit the stack, their excess heat can be transferred to a cooler stream by direct mixing or through a heat exchanger. The flowrates of the reactant streams must be increased until their heat capacity is large enough to carry away enough heat to sufficiently cool the stack. Often the flowrate of air is increased to achieve heat removal, which dilutes the oxidant reactants with the inert gas nitrogen. The drawback of this method is that such dilution of reactant streams results in lower generated voltages and decreased efficiencies.

A second temperature control method is the incorporation of internal heat exchange tubes within an IRMCFC stack. In such an arrangement, coolant (e.g., steam) pumped through the tubes would carry excess heat away from the stack. In tubular solid oxide fuel cells (SOFC), internal heat exchange is used to preheat the air feed stream. A significant advantage of this method is that heat removal is achieved without dilution of the reactant streams. Although conceivable, this method of heat removal is not popular in conventional IRMCFC stack design, possibly due to size constraints.

Another method of temperature control for which the IRMCFC was specifically designed is methane reforming. The endothermic methane reforming reaction directly consumes heat generated within an IRMCFC stack. Conventionally, the entire methane fuel

stream is fed immediately into the IRMCFC stack inlet. When this is done, the reforming reaction is nearly complete at the point where 50 percent of the fuel has been utilized (Kinoshita, McLarnon and Cairns 1988, 82), and only the inlet side of the stack is cooled. However, if fractions of the methane feed were injected along the total length of the anode gas chamber, the stack could be cooled more evenly.

5.1.6 Waste Heat Utilization

There are two basic ways in which waste heat can be recovered from hot reactant streams in an IRMCFC power system: direct mixing and heat exchange.

Direct mixing is an intrinsically efficient method of heat transfer. A popular method of heating cool feed streams is mixing them with hot, recycled reactant streams.

Heat exchangers can be used to transfer heat between process streams that have reasonable temperature differences. For example, heat exchangers can be used to raise steam with waste heat. In various bottoming cycles, the steam could be used for additional electric power production, district heating, absorption refrigeration, and air conditioning.

5.1.7 Nitrogen Oxides

One drawback of using air as an oxidant is that it introduces nitrogen into the power system, and nitrogen oxides, or NO_x , can be formed. In conventional applications, nearly all the NO_x formed is nitric oxide, NO , with the remainder being nitric dioxide, NO_2 . Nitrous oxide, N_2O , rarely constitutes an appreciable portion of the NO_x formed in conventional applications. Combustion-generated nitrogen oxides are typically classified into three categories according to the mechanism by which they are formed: fuel NO_x , prompt- NO , and thermal NO_x (Casleton, Kothari and Williams 1990).

Fuel NO_x is formed when a fuel containing nitrogen compounds (such as coal) is oxidized. If natural gas is used as fuel, this dominant source of NO_x is eliminated.

Prompt- NO is thought to be formed when nitrogen interacts with hydrocarbon fragments in the fuel. In an IRMCFC, however, nitrogen exists only in the cathode and is completely separated from the fuel by the electrolyte. Consequently, prompt- NO cannot be generated in IRMCFC stacks. However, if the anode and cathode exhaust from an IRMCFC stack are mixed in a combustor, then conditions could exist for prompt- NO to be formed.

When temperatures are high enough, atomic nitrogen and oxygen can be generated from the air stream. When sufficient quantities of these atomic species are present, they can react with molecular nitrogen and oxygen to form thermal NO_x . Apparently, the generation of thermal NO_x can be effectively prevented by keeping temperatures below 1811 K (2800 °F). The maximum operating temperature of an IRMCFC, ~970 K (~1290 °F), is well below this temperature, so the formation of thermal NO_x within a stack is not a problem. However, the combustors in an IRMCFC system typically operate at higher temperatures, and care must be taken to control their maximum temperature. Typically, the hottest area in a

combustor is near the fuel inlet, where the fuel feed has not yet been diluted with excess oxygen.

5.2 IRMCFC Network

Figure 10 shows an IRMCFC network. An IRMCFC network is composed of stacks arranged such that reactant streams are fed and recycled through them in series. The arrangement of stacks in series, as well as a unique recycle scheme, allows an IRMCFC network to meet all the requirements of an IRMCFC power system, while achieving high efficiency. Many advantages of networking, such as increased efficiency, improved thermal balance, higher total reactant utilization, and reactant conditioning between stacks (heat removal, methane injection) have already been discussed. The following is a description of the network recycle scheme as well as a discussion of the disadvantages of an IRMCFC network.

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Figure 10. IRMCFC Network

5.2.1 Anode-Anode Recycle

In a series of IRMCFC stacks, nearly all the methane is reformed. The exhaust from the last stack's anode is a high temperature mixture of the products of combustion (steam and

carbon dioxide) and unreacted fuel (hydrogen and carbon monoxide). Initially, the fuel feed is at ambient temperature and does not contain steam, which is necessary to reform methane and prevent carbon deposition. Therefore, it is beneficial to recycle a portion of the exhaust and mix it with the network's fuel feed (Figure 10, Stream 1). The high temperature anode exhaust can heat the fuel feed towards the first stack's inlet temperature, as well as provide it with the steam it needs to reform methane and prevent carbon deposition. Ideally, carbon dioxide would be efficiently separated from the anode exhaust, so only steam and unreacted fuel would be recycled to the anode inlet. This would prevent the fuel feed from being diluted with carbon dioxide.

5.2.2 Anode-Cathode Recycle

The oxidant feed is usually air, which does not have a carbon dioxide to oxygen ratio of two, which is the stoichiometry of the electrochemical reaction. Since the anode exhaust is rich in carbon dioxide, it is beneficial to recycle a portion of it to mix with the oxidant feed (Figure 10, Stream 5). Ideally, carbon dioxide would be separated from the unreacted fuel and steam that is also present in the anode exhaust, and then recycled alone to the cathode. If carbon dioxide separation proved inefficient, then steam could be condensed out of the anode exhaust, leaving only the unreacted fuel to dilute carbon dioxide in the anode-cathode recycle. If both separating carbon dioxide and condensing out steam proved inefficient, it is still worthwhile to recycle an unrefined portion of the anode exhaust to the cathode.

If some unreacted fuel is recycled with the carbon dioxide to the cathode, it may oxidize within the cathode chamber, consuming oxygen and releasing unwanted heat. To prevent this, a combustor is placed in the anode-cathode recycle loop to burn any fuel before the recycle is mixed with the oxidant feed.

5.2.3 Cathode-Cathode Recycle

In a series network, stack temperatures can be controlled by recycling cooled oxidant through the cathodes to carry away heat. The portion of air that is required to sufficiently cool the stacks is continuously recycled through the stack network. In the recycle loop, the exhausted air is first combined with the anode-cathode recycle stream in a combustor (Figure 10, Stream 4). There, the leftover oxygen in the cathode recycle oxidizes the unburned fuel in the anode recycle. The resulting stream (Figure 10, Stream 3) is passed through a heat exchanger, transferring the waste heat it carried away from the IRMCFC stacks and the combustor to a heat recovery system. Finally, the recycle (Figure 10, Stream 2) is combined with the low temperature air feed. Sufficient heat is retained in the recycle stream at the heat exchanger to achieve the appropriate stack inlet temperature.

5.2.4 Network Recycle Scheme Disadvantages

For recycling to improve the performance of an IRMCFC network, it must provide benefits that outweigh its inherent disadvantages. If carbon dioxide is not separated from the anode-anode recycle, the concentration of carbon dioxide in the anode is increased, which reduces the Nernst potential. The Nernst potential is similarly reduced by the anode-cathode

recycle if steam is not condensed out, since recycled steam dilutes reactant concentrations in the oxidant. These reductions in the Nernst potential reduce the power output of the IRMCFC network. In addition, part of the power which is generated by the network is consumed by the equipment necessary to circulate the recycle streams. Such circulation equipment, along with the additional ducting that is required by recycling, also increases the capital cost of the IRMCFC network.

5.2.5 Disadvantages of an IRMCFC Network

Given the same initial feed streams, the flowrate of reactants through stacks networked in series is much larger than the flowrate of reactants through stacks in a conventional system. Conventional fuel cell systems divide the initial feed streams among many stacks arranged in parallel. However, the initial feed streams in an IRMCFC network are not divided, but fed directly into the first of a series of many stacks. Perhaps the greatest disadvantage of IRMCFC networks is that this increased flowrate creates larger pressure drops.

The pressure drop in a stack is approximately proportional to the reactant stream velocity for laminar flow, and to the square of this velocity for turbulent flow. Any difference between the velocities of the fuel and oxidant reactant streams creates a widening pressure differential across the fragile cells. High flowrates worsen this problem. In addition, the pressure differential between reactant streams grows larger as the pressure drops of stacks in series accumulate. Assuming the flow is laminar, the pressure drop for stacks in series could be expected to increase by a factor of the number of stacks squared (e.g., three stacks in series would have a pressure drop nine times that of three stacks in parallel). The typical pressure drop for an individual stack varies with operating pressure but typically is 0.50 to 1.74 kPa (2 to 7 "H₂O).

One way to reduce the pressure drop of stacks in series would be to increase the size of each stack's flow channels. This would decrease the velocity of any given flowrate. However, bigger flow channels would increase the overall size of a stack, which is already severely limited by shipping constraints. In addition, such a redesign could increase both development time and capital cost.

Another potential disadvantage of an IRMCFC network is the interdependence of the stacks in series. A problem with one stack could alter the performance of succeeding stacks. Furthermore, bypassing or isolating a problematic stack in a network could be difficult. In the conventional parallel configuration, stack performance is not so interrelated.

6 Simulations of IRMCFC Power Systems

Computer simulations were done to evaluate the performance of various IRMCFC networks and to compare them to conventional IRMCFC power systems. Simulations were accomplished with the public version of ASPEN (Advanced System for Process Engineering) as installed at METC. ASPEN is an extremely powerful and complex tool, and it is far beyond the scope of this report to describe it in a comprehensive fashion. However, the following is a brief discussion of the method in which ASPEN models an IRMCFC stack.

6.1 METC ASPEN IRMCFC Stack Model

The METC ASPEN IRMCFC stack model gives the user great freedom in defining the process he wishes to simulate. However, some details of the internal operation of the stack are not as flexible as others. For instance, the internal flow geometry of the stack is always assumed to be co-current. In addition, the internal voltage losses, ohmic, activation and concentration polarizations, are not individually calculated by ASPEN. Instead, the user is required to specify the total voltage polarization at the stack outlet.

Input Parameters: ASPEN must have the composition and flowrate of the IRMCFC stack's inlet fuel and oxidant streams as well as a sufficient definition of their thermodynamic state (e.g., temperature and pressure). This information can be directly defined by the user, or ASPEN can calculate it from upstream process data.

Once the inlet reactant streams are known, the IRMCFC stack must be described. The number of cells in the stack must be given. Either the stack current or single-pass fuel utilization must be defined. The total outlet voltage polarization must be provided. The ASPEN model assumes that there is sufficient heat transfer between the fuel and oxidant streams such that they exit the stack at a common outlet temperature. Specification of this outlet temperature is optional. If left unspecified, ASPEN assumes that the stack is adiabatic. If the outlet temperature is provided, ASPEN equilibrates the exhaust gases at this temperature and calculates the corresponding heat duty.

Solution Algorithm: Hydrogen, carbon monoxide, and methane are all potential fuels for an IRMCFC. Through reforming reactions and the water gas shift, one mole of methane is converted into 4 moles of hydrogen. The water gas shift also exchanges one mole of carbon monoxide for one mole of hydrogen. ASPEN calculates the total fuel available in terms of the equivalent moles of hydrogen according to the preceding stoichiometry.

There is a direct relationship between the stack current and fuel utilization. Since one of these parameters is specified by the user, the other is calculated by ASPEN. Next, the total number of equivalent moles of hydrogen that are utilized is calculated by ASPEN. Since the stack area is not an input to the model, ASPEN does not calculate current density.

For each equivalent mole of hydrogen that is utilized, one mole of carbonate ions is transferred across the electrolyte. Therefore, ASPEN transfers one mole of carbon dioxide and one-half mole of diatomic oxygen from the oxidant stream into the fuel stream for each equivalent mole of hydrogen utilized.

Nearly all the reactions that take place within the IRMCFC, such as the water-gas shift, methane reforming, and the formation of nitrogen oxides, reach equilibrium rapidly. Therefore, ASPEN separately equilibrates the fully utilized fuel and oxidant streams at the stack outlet temperature by minimizing Gibbs free energy. If the stack outlet temperature was not specified, then ASPEN iterates until an outlet temperature is found that makes the stack adiabatic.

Once the fuel and oxidant streams are in equilibrium, ASPEN calculates the outlet Nernst potential. Each cell's output voltage is then computed by subtracting the user-supplied total outlet voltage polarization from this Nernst potential. Since the cells in a stack are connected electrically in series, the stack output voltage is the sum of the cell output voltages.

The stack power output is computed by ASPEN as the product of stack current and stack voltage. Finally, by subtracting the stack power output from the total enthalpy change in the reactant streams, ASPEN calculates the heat duty of the stack (Shah 1988, 19-22).

For a given total outlet voltage polarization, the performance predicted by the ASPEN IRMCFC model is generally conservative. The outlet Nernst potential that is calculated is the minimum possible for a given fuel utilization. Since the model assumes a co-current internal flow geometry, the conditions at the outlet of the stack generate the smallest local Nernst potential of anywhere within the stack. For a given fuel utilization, the Nernst potential that ASPEN calculates is the smallest possible for any internal flow geometry.

6.2 Comparison of Conventional and Networked IRMCFC Systems

The key difference between conventional and networked fuel cell systems is the configuration of the stacks. In conventional systems, reactant streams flow through stacks in parallel. Networked reactant streams flow through stacks in series. To compare the performance of these two configurations, two IRMCFC systems were simulated with ASPEN¹. The parallel-flow, conventional system is diagrammed in Figure 11. The series-flow, networked system is diagrammed in Figure 12.

Each system's primary electrical power output is generated by three IRMCFC stacks. A steam turbine bottoming cycle generates a secondary amount of electrical power. Since the capacity of the first commercial fuel cell power generation plants is projected to be small (1 to 10 MW), the capital cost of any bottoming cycle must remain low. Therefore, a relatively simple, single-pressure steam bottoming cycle was added to both fuel cell systems.

¹For ASPEN input and report files, contact J.G. Wimer, DOE/METC, Morgantown, WV 26507-0880

Figure 11. Conventional IRMCFC System

As can be seen from the diagrams, the layout/hardware of the two systems is identical with the exception of their stack configuration. Many beneficial features that are unique to networks, such as reactant conditioning between stacks, were not included in the networked system. This was done in order to isolate simply the parallel-flow versus series-flow contrast.

IRMCFC Stacks: Each system contains three IRMCFC stacks, all of which operate at atmospheric pressure. In addition, the operating temperature of all the stacks is kept within the same limits, 839 K (1050 °F) and 972 K (1290 °F). Voltage losses due to activation, concentration and ohmic polarizations were assumed to be the same for the IRMCFCs in both systems. For each IRMCFC, 0.3 volts was subtracted from its outlet Nernst potential. This is a conservative estimate, representing a higher outlet voltage overpotential than would be expected. In both systems, fuel and oxidant flow co-currently between stacks.

Recycle Scheme: The conventional and networked systems also have the same recycle scheme. A portion of the spent fuel and oxidant is mixed, combusted, and then recycled

Figure 12. Networked IRMCFC System

through the cathode in order to provide the necessary amount of carbon dioxide to the stacks. In addition, enough air is recycled through the cathode to cool the stacks, transferring excess energy to the heat recovery boilers. During the transfer of heat, enough energy is left in the oxidant recycle to heat the fresh air feed to the designated cathode inlet temperature, 860 K (1088 °F). A second portion of the spent fuel is recycled through the anode to provide enough steam to prevent carbon deposition and internally reform methane. This anode-anode recycle also heats the fresh fuel feed to the designated anode inlet temperature, 860 K (1088 °F).

Reactant Supply: Both systems are fueled with the same flowrate of pure methane. Each system is also provided with enough air to accomplish cooling of the stacks via the recycle heat exchanger. The amount of air that is required by each system is different, since it depends on the heat generated within the stacks.

Exhaust: The fuel and oxidant exhaust streams from each IRMCFC system are burned together in a combustor. The hot products of combustion are cooled in heat exchangers to 380 K (224 °F) and sent up the stack. In the heat exchanger, heat is transferred to the heat recovery boilers.

Steam Bottoming Cycle: The layout, hardware, and steam conditions of the two bottoming cycles are identical. However, since the two IRMCFC topping cycles dump different amounts of waste heat into their heat recovery systems, the flowrates of steam in their bottoming cycles differ. Condensed water is compressed from 8.466 kPa (2.5" Hg) to 16.33 MPa (2368 psia) before it is heated to a saturated liquid in the feedwater heater. A 5 percent pressure drop occurs across the feedwater heater. Next, the saturated liquid is evaporated and the resulting steam is superheated to the turbine inlet temperature, 813.15 K (1004 °F). The evaporator and the superheater each have a pressure drop of 5 percent. After these pressure drops, the steam, now at 14.0 MPa (2030 psia), is expanded through the turbine to a pressure of 8.466 kPa (2.5" Hg). (These conditions meet the requirements of Asea Brown Boveri's Model V turbine.) The isentropic efficiency of the turbine is adjusted to produce an outlet vapor fraction of 0.87. The cycle is completed as the turbine exhaust is condensed.

The heat supplied to the steam cycle is transferred from the recycle and exhaust streams of the fuel cell network. As shown in Figures 11 and 12, all the feedwater is first heated by the network exhaust stream. The resulting flow is split into two uneven flows. The smaller flow is evaporated and superheated by transfer of the remaining heat in the exhaust stream. The bigger flow is evaporated and superheated by transfer of heat from the fuel cell network recycle stream. Figures 13 and 14 graph stream temperatures as a function of the heat transferred to the steam cycle.

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Figure 13. Heat Recovery and Transfer: Networked IRMCFC System (3-Stack, Co-flow)

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Figure 14. Heat Recovery and Transfer: Conventional IRMCFC System (3 Stack, Co-flow)

Parasitic Power Losses: In both the conventional and networked systems, the work required by the steam cycle compressor is supplied by (deducted from) the work produced from the steam turbine. The work required by other parasitic devices, such as blowers and the power conditioning unit, are not taken into account for either system.

Stack Current and Power Conversion: The stacks are assumed to be connected to a power conditioning unit in series. Therefore, within each system, every stack utilizes the same amount of fuel and achieves an equivalent current.

Total Fuel Utilization: Distinct from the fuel utilization of individual stacks is the concept of total fuel utilization. Total fuel utilization reflects the combined fuel utilization of every fuel cell in the network, as well as the effect of recycling fuel through the network. It does not, however, include the effect of fuel burned in recycle or exhaust combustors. Since it has been stipulated that stacks within each system achieve the same current, each stack also accomplishes one third of the total fuel utilization.

Total fuel utilization affects system performance in many ways. As total fuel utilization is increased, stack current increases, but the final output voltage decreases. The

amount of heat that must be removed from the stacks also increases with total fuel utilization. For each system, the total fuel utilization was optimized for maximum system efficiency. Figure 15 shows the effects of total fuel utilization on conventional and networked system efficiencies, given equivalent stack currents within each system.

Figure 15. Effect of Total Fuel Utilization on IRMCFC System Efficiencies

As can be seen in Figure 15, problems arise in both systems as their total fuel utilizations are lowered. (These problems can be easily solved by changing or relaxing system design constraints.) When the total fuel utilization of the networked system is lowered to 71 percent, fuel conditions begin to favor the formation of solid carbon, which can plug gas passages in the anode. At slightly higher utilizations, enough carbon dioxide and steam is produced and recycled through the anode to prevent carbon deposition by the Boudouard reaction (7).

When the total fuel utilization of the conventional system is lowered to 66 percent, unreacted fuel, recycled from the anode exhaust, is mixed with the cathode air feed. Consequently, it may oxidize within the cathode chamber, consuming oxygen and releasing unwanted heat. The fraction of anode exhaust that is recycled is proportional to its carbon dioxide content. As total fuel utilization is decreased, the fuel to carbon dioxide ratio in the

anode exhaust increases. When the amount of fuel recycled with the carbon dioxide surpasses that which the recycle combustor can oxidize, unburned fuel is mixed into the cathode feed.

Comparison of Performance: Each system's performance at optimum total fuel utilization is summarized in Table 1.

The change in enthalpy between the streams entering and exiting each system is identical. However, by more closely approaching a reversible process, the networked IRMCFC system converts more of the free energy into electrical work than does the conventional IRMCFC system. The efficiencies of the fuel cell stacks, based on the lower heating value (LHV) of natural gas, are 44.90 percent for the stacks placed in series and 35.38 percent for those in a parallel-flow configuration.

Free energy that is not converted to electrical work becomes heat. Therefore, the conventional configuration of fuel cells generates more heat than the networked system, which makes its job of heat removal more challenging.

Table 1. Performance Comparison of Networked and Conventional Systems

	Networked IRMCFC System			Conventional IRMCFC System		
	Stack #1	Stack #2	Stack #3	Stack #1	Stack #2	Stack #3
Fuel Utilization	0.305	0.438	0.780	0.784	0.784	0.784
Total Utilization (Optimum Value)	0.95			.87		
Current ($\times 1\text{E}+6$ Amps)	4.576	4.576	4.576	4.190	4.190	4.190
Voltage ($E_{\text{REV}} - \Sigma\eta$, $\Sigma\eta=0.3$ V)	0.5905	0.5124	0.3708	0.4227	0.4227	0.4227
Power (MW)	2.702	2.345	1.697	1.771	1.771	1.771
Total IRMCFC Power (MW)	6.7433			5.3135		
Steam Cycle Power (MW)	2.7313			3.2360		
Total System Power (MW)	9.4746			8.5495		
IRMCFC Power (%)	71.2			62.1		
Steam Cycle (%)	28.8			37.9		
IRMCFC Network Efficiency, LHV	44.898			35.378		
Total System Efficiency, HHV	56.846			51.296		

More power is produced by the steam cycle of the conventional system than by that of the networked system. This is because the fuel cells in a parallel-flow configuration discharge more heat to the bottoming cycle than do the networked cells. Fuel cells convert free energy directly into electrical work, without the intermediate step of heat. Transfer of heat to a bottoming cycle in order to convert it to electrical work is far less efficient than the direct conversion of energy in the fuel cells. Therefore, the amount of heat discharged to the steam bottoming cycle should be minimized.

The total power produced by the networked IRMCFC system is greater than that of the conventional system. The overall efficiency of the networked system is 56.85 percent, while that of the conventional system is 51.30 percent. It is expected that the networked system will have a much higher efficiency once it is more fully optimized.

6.3 Series Network Optimization

Total Outlet Voltage Polarization: Specifying a realistic total voltage polarization at the stack outlet is crucial since this parameter has such a drastic impact on predicted performance. Two simulations were done of the IRMCFC network and steam bottoming cycle depicted in Figure 12. In the first simulation, the total outlet voltage polarization of each stack was specified to be 0.3 V, while 0.2 V was specified in the second simulation. Table 2 illustrates the impact of this parameter on the system's performance.

Table 2. Effect of Total Outlet Voltage Polarization on Performance

	Networked IRMCFC System	
	0.3	0.2
Total Outlet Voltage Polarization, $\Sigma\eta$ (V)	0.3	0.2
IRMCFC Network Efficiency, LHV	44.898	54.634
Total System Efficiency, HHV	56.846	62.517

Flow Geometry Between Stacks: The IRMCFC network diagrammed in Figure 12 was simulated with fuel and oxidant flowing both co-currently (as diagrammed) and counter-currently between stacks. The outlet temperature of each stack was graphed in Figure 16. Stack temperatures are much more uniform in the countercurrent case. A uniform temperature distribution allows stacks to be operated nearer their maximum operating temperature. An increase in stack temperature reduces the total voltage polarization, which generally decreases as temperature increases. Therefore, the countercurrent system should be slightly more efficient. Unfortunately, the ASPEN IRMCFC model does not yet have the capability to calculate the total voltage polarization as a function of temperature. Consequently, the quantification of further performance comparisons between the two cases must be postponed.

until the ASPEN model is improved. The countercurrent case is also expected to produce more uniform stack current densities.

Number of Stacks in Series: Although each stack added to a series network improves the system's efficiency, the incremental benefit obtained with each additional stack diminishes. There is a finite number of stacks that could adequately approach a reversible process while meeting economic, space, and design constraints.

Pressure: As shown by equation (21), the Nernst potential generated by the overall cell reaction increases with the pressure ratio:

$$\frac{(P_c^{3/2})}{P_a} , \quad (25)$$

where: $P_c \Rightarrow$ equilibrium cathode chamber pressure,
 $P_a \Rightarrow$ equilibrium anode chamber pressure.

Figure 16. Effect of Flow Geometry on Temperature Distribution Among Stacks

The operating pressures of stacks in a series network should be optimized to attain the maximum system efficiency. An additional advantage is that reactant streams at elevated pressures require smaller piping and vessels.

Stack Current and Total Fuel Utilization: There is nothing that requires the stacks within a fuel cell network to achieve the same current. In all probability, maximum system efficiency will be obtained when networked stacks utilize different amounts of fuel, achieving different currents. Total fuel utilization should also be optimized for a network in which stacks achieve different currents. Once found, this value is likely to differ from the optimum value shown in Figure 15, in which stack currents were equal.

Reactant Conditioning: Between stacks, at different stages of utilization, the reactant streams can be conditioned for optimum system performance. Such conditioning could include mixing, heat exchange, and methane injection.

Additional Optimization: Other parameters that could be adjusted for optimum performance include the ratio of carbon dioxide to oxygen in the cathode and the ratio of fuel feed to air feed. Separation processes, which could be introduced to improve the system's performance, include removal of carbon dioxide or steam from the anode recycle stream.

7 Conclusion

7.1 Summary

To support a discussion of fuel cell networks, some related concepts were reviewed and the operation of an individual molten carbonate fuel cell was explained. The chemical reactions associated with an internally reforming molten carbonate fuel cell (IRMCFC) were each described and then combined into the overall IRMCFC reaction. Applicable thermodynamic and electrochemical principles were discussed. Finally, the structure and operation of fuel cell stacks were explained.

Key to the concept of networking is the arrangement of multiple fuel cell stacks with regard to the flow of reactant streams. Conventional fuel cells systems have been designed such that reactant streams flow in parallel through fuel cell stacks. In a fuel cell network, however, reactant streams are ducted such that they are fed and recycled through stacks in series. The series flow of oxidant and fuel streams between networked stacks can be either countercurrent or co-current. Electrically, networked stacks can be connected to a power conditioning unit in series, in parallel, or independently.

Arranging fuel cell stacks in series offers several advantages over conventional fuel cell systems. Stacks networked in series more closely approach a reversible process, which increases the system efficiency. Higher total reactant utilizations can be achieved by stacks networked in series. Placing stacks in series also allows reactant streams to be conditioned at different stages of utilization. Between stacks, heat can be consumed or removed (methane injection, heat exchange), which improves the thermal balance of the system. The composition of streams can be adjusted between stacks by mixing exhaust streams or by injecting reactant streams.

When designing an IRMCFC power system, several requirements must be met. An IRMCFC system must properly condition both the fuel and oxidant gas streams. Methane must be reformed into the more reactive hydrogen and carbon monoxide. Carbon deposition, which can plug gas passages in the anode gas chamber, must be prevented. To supply the flow of carbonate ions, the air oxidant must be enriched with carbon dioxide. Both oxidant and fuel feed streams must be heated to their proper inlet temperatures. Each IRMCFC stack must be operated within an acceptable temperature range. Excess heat generated by the IRMCFC stacks must be recovered and efficiently utilized. Acceptable environmental performance, such as suppression of nitrogen oxides, must be ensured.

The recycle scheme of an IRMCFC network allows it to meet all the requirements of an IRMCFC power system, while achieving high efficiency. Spent fuel and oxidant streams are mixed, combusted, and then recycled to enrich the air oxidant with carbon dioxide. Enough oxidant is recycled through the IRMCFC stacks to cool them. Excess heat is transferred from the oxidant recycle to the heat recovery boilers. During the transfer of heat, enough energy is left in the oxidant recycle to heat the fresh air feed to the designated

cathode inlet temperature. A second portion of the spent fuel is recycled to the fuel feed to provide enough steam to prevent carbon deposition and internally reform methane. This anode-anode recycle also heats the fresh fuel feed to the designated anode inlet temperature.

Two ASPEN simulations compared the performance of conventional and networked fuel cell systems with identical recycle schemes and steam bottoming cycles. Each simulated system was composed of three IRMCFC stacks operating at the same temperature and pressure and with the same voltage losses. When the total fuel utilization of each system was optimized for maximum efficiency, the efficiency of the fuel cell stacks networked in series was nearly 10 percent greater than that of the stacks arranged in parallel (44.898 percent versus 35.378 percent, LHV). When the power generated by each system's steam bottoming cycle is considered in addition to its fuel cell power, the gap in efficiency narrows to 5.5 percent. The efficiency of the total networked system is 56.846 percent, while that of the total conventional system is 51.296 percent.

The fuel cell network that was simulated was not fully optimized. Optimization of flow geometry, operating pressure, stack fuel utilization and current, reactant conditioning, and other parameters is expected to yield further significant increases in total system efficiency.

7.2 Recommended Future Work

Much more analysis of fuel cell networks should be performed with ASPEN. The existing natural gas-based simulations should be optimized. A simulation of an integrated gasification fuel cell (IGFC) plant should be developed. Various cogeneration possibilities should be simulated, such as absorption refrigeration or district heating. Finally, the ASPEN fuel cell model itself should be improved. Instead of requiring that the total outlet voltage polarization be provided by the user, ASPEN should calculate voltage losses as a function of certain operating parameters.

One disadvantage of fuel cell networks is the possibility of an increased pressure drop. It should be determined how to eliminate or minimize any increase in pressure drop. Also, the impact of pressure drop on system performance should be assessed.

A more detailed analysis of fuel cell networks is needed before such systems are considered for commercialization. A market study must be performed to identify potential customers, their power demand, and cogeneration preferences. Once these questions are answered, the construction of a proposed plant should be planned. The proposed plant's footprint, modularity, capital cost, and construction schedule must be determined. Certain aspects of the proposed plant's operation, such as start-up, shut-down (emergency and planned), and load variances should be designed. In addition to the plant's capital cost, its operating costs, including fuel, maintenance, and replacement, must be projected in order to calculate the cost of electricity. Other issues, such as the availability, life, and environmental performance of the plant should also be addressed.

Based on electrochemical principles, the concept of networking applies to any device with an electrochemical membrane, such as a fuel cell. Therefore, the electrochemical membrane separation device (EMSD), which has been developed for hot gas cleanup in future power plants, can also benefit from networking. As in the case of fuel cells, it has been shown that networking EMSDs in series improves their efficiency (Wimer et al. 1992). Simulations of EMSD networks operating within an entire electric power generation system need to be accomplished.

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