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COMMERCIAL PHOSPHORIC ACID FUEL CELL SYSTEM TECHNOLOGY DEVELOPMENT

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U.S. DEPARTMENT OF ENERGY
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Fourteenth Intersociety Energy
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ABSTRACT

A review of the current commercial phosphoric acid fuel cell system technology development efforts is presented. In both the electric utility and on-site integrated energy system applications, reducing cost and increasing reliability are the technology drivers at this time. The longstanding barrier to the attainment of these goals, which manifests itself in a number of ways, has been materials. The differences in approach among the three major participants (United Technologies Corporation (UTC), Westinghouse Electric Corporation/Energy Research Corporation (ERC), and Engelhard Industries) and their unique technological features, including electrodes, matrices, intercell cooling, bipolar/separator plates, electrolyte management, fuel selection and system design philosophy are discussed.

BACKGROUND

For a number of years the Phosphoric Acid Fuel Cell has been the leading candidate fuel cell power plant system for terrestrial applications. Initially, development funds came primarily from electric and gas utility sources, from fuel cell manufacturers, and from the U.S. Army. The emergence of the Nation's energy program provided new direction coupled with increased government support for the development of commercial fuel cell power plant systems. In the commercial field, the phosphoric acid fuel cell was clearly identified as the near-term or first generation system aimed at both utility electric generation as well as on-site integrated energy systems. Currently, three major contractors or contractor teams are active in the terrestrial phosphoric acid fuel cell field: United Technologies Corporation (UTC), Westinghouse Electric Corporation/Energy Research Corporation (ERC), and Engelhard Industries. This paper is a report of the status of these major phosphoric acid fuel cell development efforts, with emphasis on the differences among the contractors and the unique technological features.

Similarities exist in the basic cell technology of the three major contractors in that all three employ carbon or carbon paper electrode structural elements, platinum-supported-on-carbon catalyst layers and silicon carbide/Teflon matrices. However, beyond this, the various technologies become very diverse.

At present, only UTC, the technology leader, is working toward commercializing fuel cell power plants for both electric utility and on-site, integrated energy (OS/IES) applications. The electric utility program is in the system-technology demonstration or operational feasibility phase with a 4.8 MW module test scheduled to be conducted on the Con Ed network in mid-1980. The OS/IES program is in the engineering and design phase of a prototype 40 kW system. This is to culminate in a prototype system verification test in mid-1980.

The 4.8 MW demo test represents an earlier phase of cell development than the 40 kW prototype and, therefore, differences exist in the basic cell configurations of the two systems. However, it is expected that the general characteristics of the cell and stack technology to be developed for commercialization will be very similar for both applications. Both will employ similar catalyst layers, two-phase liquid cooling systems, fibrous carbon/graphite cell elements and reactant seal and supply systems. Though similar, some optimization of various aspects of the technology will be required to address the differences in design power level and operating conditions of the two applications. The present electric utility system (4.8 MW) operates at 375°F and 50 psia over a profile from 1/4 to full-rated load; for the ultimate commercial power plant, current thinking by UTC is to utilize a large area cell operated at approximately 400°F and up to 120 psia. On the other hand, the OS/IES is to be operated at atmospheric pressure, 375°F, over a load profile from open circuit to full-rated load.

Of the competitive suppliers, ERC has been in the OS/IES development effort the longest. In 1970, ERC contracted with the Army to develop a methanol fueled phosphoric acid power plant; a fully automatic 1.5 kW demonstration unit was delivered in 1978. DOE (then ERDA) contracted with ERC in 1977 to apply the experience gained under the Army program to the somewhat different requirements of OS/IES. In 1978, ERC teamed up with Westinghouse, a marriage that added system analysis, system design, and marketing capability to ERC's electrochemical technology/engineering background.

The Westinghouse/ERC program is at the point where most of the enabling electrochemical component technology has been developed. Presently, the electrochemical components are being optimized for cost and life, basic design and development is starting for other power plant components and an initial system design effort is starting. The most unique characteristic of the Westinghouse/ERC power plant is the DIGAS (DIstributed GAS) cooling concept. In this cooling scheme, the air feed stream is split inside the manifold into a reactant stream and a coolant stream. The two streams flow independently through the fuel cell stack and are merged in the exit manifold.

Westinghouse's approach is to develop the OS/IES to be able to market not just a fuel to electric conversion device, but a total energy system that will supply all the space conditioning, water heating, and electric power requirements for a residential or commercial application.

Engelhard contracted with DOE in 1976 to study several fuel cell applications and to develop fuel cell stack technology. Their previous experience included building and marketing H_2,O_2 fueled laboratory-size demonstration fuel cells. They also have an Army funded fuel cell development contract. Presently, Engelhard is concentrating on stack component development and is beginning to define attractive application/fuel combinations in preparation of an OS/IES system conceptual design effort. The component development effort has resulted in several unique approaches to fuel cell design. One area of innovation is their bipolar plate, the device which separates and directs the flow of reactants as it provides electrical conduction between individual cells. The conventional bipolar plate is a single piece of conductive material with reactant flow channels either molded or machined on either side. Engelhard's approach is to split the bipolar plate into three sections, an impervious cen-

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ter section (for reactant separation), with an open-cell foam section on either side (for reactant flow). All sections are made from organic precursors that are graphitized then densified. This process is very compatible with mass production and promises considerable cost savings over one-piece, bipolar plates which must be individually molded or machined. Engelhard has also developed a low cost cooling plate that can be made of conventional materials and fabricated using conventional furnace brazing techniques. The coolant is a dielectric liquid. This cooler offers potential cost reduction over other liquid and ebullient liquid coolers.

Phosphoric acid fuel cell technology and development efforts for both the electric utility and OS/IES applications are directed toward reducing cost, increasing performance, increasing life, and improving reliability. These, in turn, can be reduced simply to cost and reliability goals. In the technology area, the most vexing problem, the longstanding barrier to the attainment of these goals has been materials. In the cell catalyst layer, the carbon support must resist oxidative corrosion while the catalyst must resist dissolution and sintering at the operating temperature, pressure, and potential. Both cell and fuel processor catalysts must possess sufficient tolerance to fuel contaminates. The carbon/graphite cell and stack materials must possess just the right combination of structural properties. The seals must seal well, be durable, and not contaminate. Metal parts throughout the system must possess the right structural and thermal properties, and also be durable. Finally, all materials must not be too costly and be easy to manufacture. It has been very difficult to find and to develop materials that could meet, simultaneously, the cost and reliability goals. The approach to solving these problems has involved technology/development efforts guided by system trade-off studies.

Beyond materials, a difficult problem has been the ability to provide the cell stack with sufficient electrolyte to attain a cost effective stack operation. In what follows some of these issues and their resolution by each of the major contractors will be discussed.

UNITED TECHNOLOGIES CORPORATION

CELL STRUCTURE - The advanced cell structure concept commonly called the "paper" or integral ribbed subtrate is UTC's approach to lowering cell system cost and, simultaneously, increasing electrolyte capacity. In part, the cost reduction is brought about by the substitution of a 2-3 day process in place of the conventional 2-week molding and curing process which produces a complex bipolar/separator plate. Aside from being complex, the molded processing method places a limitation on the size of the plate that can be fabricated. Eliminating the size constraint on the cell structure thereby provides the possibility to go to a larger and more cost effective cell.

The unique features of the integral ribbed substrate concept are: 1) replacing with a simple impervious flat plate the complex bipolar/separator (which normally contains a ribbed reactant flow field on either side perpendicular to each other); 2) transferring the function of each solid ribbed flow field onto a ribbed porous element which, after deposition of catalyst on one side, behaves like an electrode and which is also designed to act as an electrolyte reservoir. Figure 1 illustrates the differences between the molded cell and the ribbed substrate cell approach. The ribbed substrate is fabricated from a blend of carbon fibers and resin which are 1) made into a preform, 2) graphitized, and 3) milled flat and ribbed in a single pass cut on a milling machine.

In addition to this method of forming the ribbed substrate, an advanced method with even lower cost potential, is being pursued under DOE contract.[1*] The key difference is that instead of forming the initial structure from a blend of fibers and resin, the process begins with a fabric-like material. Currently, a needled rayon precursor has been developed and successfully tested.

HIGHER TEMPERATURE AND PRESSURE OPERATION - Quantitative data on the performance increases due to higher temperature (375° to 425°F) and to higher pressure operation (50 to 120 psia) have been obtained. At initial time, an expression was obtained for the effect of reactant pressure change (15 to 150 psia) upon IR-free cell voltage for a cell operating at a constant 300 ASF.

$$\Delta E_{cell} = b_s \log P_1/P_2 \qquad (1)$$

where P is reactant pressure and b_s is the sum of the anode Nernst (thermodynamic) effect and cathode Tafel slope (kinetic) at the given temperature.

At 120 psia, 400°F, 375 ASF, 2" x 2" subscale cells have achieved an initial performance increase of up to 70 mV compared to older type cells operating at 50 psia, 375°F, and 300 ASF. However, as expected, the higher temperatures and, to a lesser extent, pressures also cause an acceleration in the decay rates. In the UTC power plant system, increasing pressure without increasing temperature is not possible because of the interdependence of cell coolant and reformer conditions. Progress has been made at reducing the sharp cell performance decay rate occurring during high temperature and pressure operation. The rate of decay has been reduced from 60 mV per 1000 hr. to approximately 12 mV per 1000 hr., which is the 4.8 MW decay rate at 50 psia. (See Figure 2)

From the overall power plant standpoint, the optimum pressure and temperature depend not only on stack technology and development, but also on the cost and performance implications upon balance of system (turbocompressors, heat exchangers, piping, etc.).

INTERCELL COOLING - Two-phase water cooling is the method used. Thin-walled, 2-pass copper tubes with stainless steel headers are the latest design. The acid environment requires the copper tubes to be coated with a thin Teflon protective film; however, this reduces heat transfer somewhat. The chief disadvantage of these intercell coolers is their relatively high cost. The probability of cooler failure due to penetration of the film and subsequent corrosion of the copper has not yet been established. To eliminate this potential failure mode and to reduce cooler cost, a graphite cooler technology effort was undertaken. Unfortunately, the one concept that met the technology goals failed to be any cheaper than the metal cooler.[1]

ELECTRODES - Primary focus has been on the cathode. Stabilization of the carbon support of the Pt catalyst via heat treatment has been one important effort. This is especially important for the 40 kW system which experiences swings to much higher potentials. Investigation indicated that above 800 mV cathode losses attributable in great measure to oxidative corrosion of the C support occur. Other cathode treatments by UTC are aimed directly at the Pt catalyst to enhance activity without sacrificing stability. Recent work indicates progress in this area.[1] Work is being done to develop and optimize the catalyst layer for high temperature and pressure operation. The support carbon is being stabilized to inhibit corrosion, the wetproofing is being optimized to minimize diffusion losses, and techniques are being developed to improve the activity of platinum.

With total Pt electrode loading reduced to approximately 0.75 mg/cm^2, the Pt cost has been reduced to

*Numbers in parentheses designate References at end of paper.

much less importance than years ago when unsupported electrocatalysts were used. However, the recent sharp rise in Pt cost coupled with ≥ 50% lower power density operation for the 40 kW system [3] versus the 4.8 MW system increases the importance of electrocatalyst cost particularly for the OS/IES system.

WESTINGHOUSE/ERC

Probably the most important innovation of the ERC technology program is the DIGAS (DI stributed GAS) cooling method, which is illustrated in Figure 3. The DIGAS method distributes process air to both the cathodes (via bipolar plates) and to special cooling plates. The total amount of air, that required for reaction plus that required for cooling, is supplied to the air inlet manifold. The air then flows through the cell via either the cathode flow (cathodic) channels of the bipolar plate or the channels of a special cooling plate. All of the air is collected in a common exit manifold and flows to an external loop where heat is removed, the oxygen depleted air is purged and fresh air is added such that steady operation is maintained.

Since the cathodic channels of the bipolar plates and the cooling plate channels are parallel paths for air, the flow split is determined by the relative cross-sectional area, hence pressure drop, of the cathode and cooling channels. A typical design is a 10:1 split with three times the stoichiometrically required amount (3 stoich) of cathode gas going to the cells and 30 stoich cathode gas going to the cooling plates.

Table I displays ERC's comparison of DIGAS cooling to the two other commonly accepted cooling methods, process gas cooling and liquid cooling, performed under contract to DOE.[4] The estimates are based on selected configurations of the three cooling methods. No effort was made to optimize any of them; however, all systems are reasonable and are consistent for comparison purposes. For any system chosen by a particular manufacturer, some optimization of the values presented in Table I may be expected. Each of the system feature comparisons will now be discussed.

CONSTRUCTION SIMPLICITY - DIGAS cooling is relatively simple. Depending on stack operating conditions, at regular intervals a cooling plate is added to the stack of cells in the same manner as a bipolar plate is stacked. No special manifolding, connections or seals are required.

Process gas cooling is the simplest of all. There are no special cooling plates or stacking procedures needed. Several times the stoichiometric reaction requirement of cool air is supplied to the stack. All the air flows through the cathodic channels of the bipolar plate. Upon exiting the stack, the air can be vented or recycled as in the DIGAS method.

Liquid cooling is by far the most complex and expensive. It involves passing a liquid coolant through special cooling plates inserted at regular intervals in the stack. The coolant may remain in the liquid phase at all times using only its sensible heat to cool the stack or the coolant may partially vaporize using latent heat for part of the cooling load. The coolant must be either separately manifolded or supply lines must be connected to each individual cooling plate. Standard heat transfer materials such as copper or aluminum are corroded by phosphoric acid and must be sheathed if they are to be used. Standard, inexpensive fuel cell materials such as graphite or graphite/resin composites present difficulties in sealing and making connections. Finally, the liquid itself and coolant lines provide a possible shorting path in the cell (shunt currents which result in parasitic losses) which must be minimized.

ELECTROLYTE LOSS - Since the vapor pressure of phosphoric acid (hence electrolyte loss) is a weak function of temperature, the main consideration here is the amount of gas flowing past the electrodes to carry off electrolyte. In both DIGAS and liquid cooling methods, a small excess of reactants flow past the electrodes, while in a process gas cooled stack all the cooling air must pass the electrodes. Process gas cooling has the highest electrolyte loss which translates to the shortest life or the shortest mean time between electrolyte replenishment.

RELIABILITY - For both process gas and DIGAS, reliability is high since there is no special cooling fluid and no complex manifolding or connections. A leak in the stack liquid cooling system would at a minimum cause a shutdown while the leak was repaired or the defective cooling plate replaced. If an organic cooling fluid were used, a leak could conceivably poison the catalyst and make replacement of the entire stack necessary. Most air leaks could be ignored.

EXTERNAL HEAT EXCHANGE - Heat recovery is by far best with a liquid or two-phase liquid cooling system. Heat recovery is possible, though more difficult, with either of the recirculating air cooling systems.

COST COMPARISON - This is an estimate based on materials used and construction complexity; it reflects the materials and construction problems of liquid cooling.

TOTAL DIFFERENTIAL TEMPERATURE - This concerns temperature gradients across a fuel cell stack. The total temperature differential, ΔT, between any two points in the stack contains two components, a ΔT in the stacking direction (perpendicular to both reactant flows) and a ΔT in the flow direction (parallel to one reactant flow). It is important to maintain the stack temperature within a fairly narrow band. If the temperature drops too low, CO poisoning of the catalyst becomes a problem, and if the temperature is too high, the operating limit of the stack materials is exceeded. A commonly accepted operating range is between 350 and 400°F. Since every cell is cooled equally in the process gas method, there is no stacking direction component of the total ΔT. Also, since the coolant directly contacts every cell, the inlet area is cooled more than in other methods; this accounts for the large ΔT in the flow direction. For both the DIGAS and liquid cooling methods, the same number of cells between cooling plates is assumed. This sets the stacking direction ΔT at 15°F for both systems. The difference in flow direction ΔT results from the differences in liquid and gas heat transfer characteristics.

TOTAL AUXILIARY POWER - These requirements are mainly a function of the amount of coolant circulated and the cross-sectional area of the coolant passages. As expected, the power requirement for pumping a liquid coolant is quite small. Since all the cooling air flows through the cathodic channels of the bipolar plate, the power requirement is quite large for the process gas method. Power for DIGAS lies between the two. The same amount of air must be circulated through the stack as in the process gas method, but a large fraction passes through the large area cooling plate passages.

PRESSURE DROP ACROSS CELL - This is a function of the air flow through the cathodic channels of the bipolar plate. The same amount of air is required for both DIGAS and liquid cooling, hence the same pressure drop. The high pressure drop for process gas cooling reflects the high air flow rate required through the cathodic channels.

Based on the above considerations, ERC has concluded that the DIGAS concept offers the best compromise between reliability, life-cycle cost, and heat recovery.

Westinghouse has recently made a more detailed study of DIGAS cooling. Under DOE contract[5] they developed a lumped parameter fuel cell stack simulation code that calculates reactant gas composition, current-voltage characteristics, and heat transfer characteristics for an air cooled fuel cell stack.

in the model, the cell area is broken down to a grid of finite elements so that power and heat generation can be calculated as functions of temperature and reactant composition, as each varies from point to point in a cell.

As discussed previously, one of the major drawbacks of DIGAS cooling is a higher total differential stack temperature than that of liquid cooling. Westinghouse is presently redesigning the DIGAS cooling plates in an effort to reduce the differential temperature. Results of the simulation show that a 28°F differential temperature can be achieved with proper design. This matches what can be achieved with liquid cooling.

Westinghouse is expanding their computer model to include simulation of the other subsystems in the fuel cell power plant and total energy system. The model includes performance and economic calculations so that trade-offs can be made that will optimize the total energy system as a whole rather than optimizing just the fuel cell power plant itself.

ENGELHARD INDUSTRIES

The unique features of the Engelhard OS/IES effort currently are in the following areas: 1) development of a fuel conditioner that will operate on methanol (methyl fuel); 2) novel bipolar plate approaches; 3) liquid intercell cooling; and 4) matrix and electrolyte transport reservoir technology. In the current two-year contract with DOE[6] the electrocatalysis task is solely funded by Engelhard. Therefore, the details of any unique developments are proprietary.

FUEL PROCESSING - Methanol will be initially used in the Engelhard OS/IES; natural gas, the choice in both the UTC and Westinghouse/ERC programs, will receive emphasis in later phases of the work. Methanol selection was based upon its projected availability from coal in the 1990 time frame. A subscale fuel processor is being readied for parametric investigations.

BIPOLAR PLATES - In the stack area, two novel bipolar approaches appear promising. Under subcontract, Pfizer is developing chemically resistance carbon (vitreous, carbon and graphite) structures starting with reticulated vitreous carbons or cloths. One of these structures, the so-called B element, is impermeable to gas transport and will serve as the bipolar plate connecting two adjacent cells. See Figure 4. (In the UTC ribbed substrate integral cell concept, this is UTC's separator plate.) The other structure, the so-called A element, is porous to permit fuel or oxidizer transport to the electrochemically active electrode areas (which could be on the A elements). Graphite structures using Prizer's chemical vapor deposition techniques are being utilized to help produce the required properties in both A and B elements. The resulting bipolar plate consists of a B element sandwiched between two A elements. Electrolyte storage can be accommodated in the A element.

In the second approach, thin graphite plates are separated by a gas-impermeable layer of a chemically resistant material (such as graphite or plastic) that permits satisfactory electrical and thermal conductivity. Grooves in each plate will permit reactant transport. If the graphite plates are porous and connected suitably to the matrix, they can provide an electrolyte reservoir for matrix replenishment or for overflow volumes resulting from changing conditions (volume tolerance). Several promising material combinations are under test. The Engelhard/Pfizer bipolar plate approach has some aspects in common with the UTC ribbed substrate integral cell approaches previously discussed.

INTERCELL COOLING - A brief discussion of the liquid intercell cooling approach employing a dielectric fluid, the Engelhard method, was discussed previously in connection with the Westinghouse/ERC cooling approach. The Engelhard design makes use of baffles to provide good heat transfer control yet designed for ease of fabrication. Not yet finally determined is the suitability of the dielectric fluid selected (Monsanto Therminol 44) or the ability of the aluminum cooler to hold up for the five year stack life goal under the corrosive acid environment of the fuel cell stack.

MATRIX - The matrix effort emphasizes materials capable of operating at temperatures up to 400°F and construction that results in good electric and thermal conductivity, good transport (flow) properties, and satisfactory reactant crossover resistance. A thin SiC-fluorocarbon matrix is laminated to each electrode. Improved electrolyte transport, needed when operating conditions change, is provided by sandwiching an electrolyte transport member between the two laminates. This electrolyte transport member also facilitates electrolyte replenishment to the matrix from a storage area.

CONCLUDING REMARKS

As part of the Nation's energy program three major contractor or contractor teams (UTC, Westinghouse/ERC, and Engelhard), active in the terrestrial phosphoric acid fuel cell field, are receiving government

Table I - Comparison of Cooling Schemes for Phosphoric Acid Fuel Cell System

System Features	DIGAS Cooling	Process - Gas Cooling	Liquid Cooling
1. Construction Simplicity	Simple	Simple	Complex
2. Electrolyte Loss	Low	High	Low
3. Reliability	High	High	Low
4. External Heat Exchange	Fair	Fair	Good
5. Cost of Cooling Subsystem, % Stack Cost	5	5	25-50
6. Total Differential Temp °F	45	60	20
ΔT_{cell} in stacking direction °F	15	0	15
ΔT_{cell} in flow direction °F	30	60	5
7. Total Auxiliary Power Req't., %	2	5	1
Stack Auxiliary Power Req't., %	0.5	3	<0.5
Balance of System Auxiliary Power Req't., %	1.5	2	0.5
8. Pressure Drop Across Cell, in. H_2O	0.4	3.5	0.4

support to develop commercial fuel cell power plant systems. At present, only UTC, the technology leader, is working toward commercializing fuel cell power plants for both electric utility and OS/IES applications. The other contractors are a few years behind in their OS/IES development efforts.

While similar in many respects, significant technological and system differences exist among the three contractors. These include intercell cooling, bipolar/separator plate, electrolyte management, materials, fuel selection, and system design philosophy. Phosphoric acid fuel cell technology/development efforts for both the electric utility and OS/IES applications are directed toward reducing cost and increasing reliability. The longstanding barrier to the attainment of these goals, which manifests itself in a number of ways, has been materials.

In addition, for the electric utility application, UTC is pursuing higher temperature and pressure operation (400°F and up to 120 psia). The initial performance gains due to higher temperature and pressure must be balanced against possible shorter life as well as the cost and performance implications upon the balance of the system.

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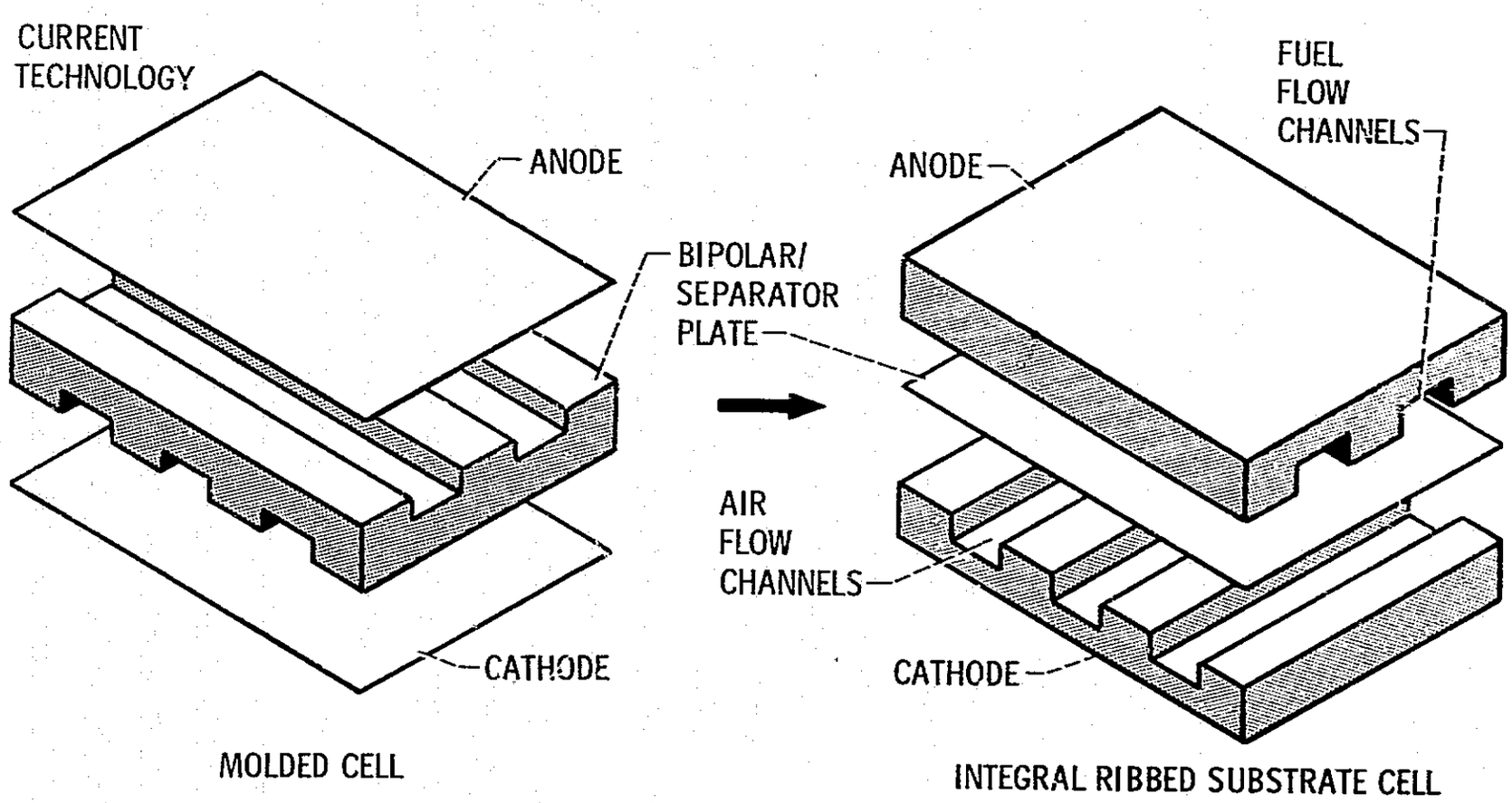

Figure 1. - Integral ribbed substrate cell characteristics.

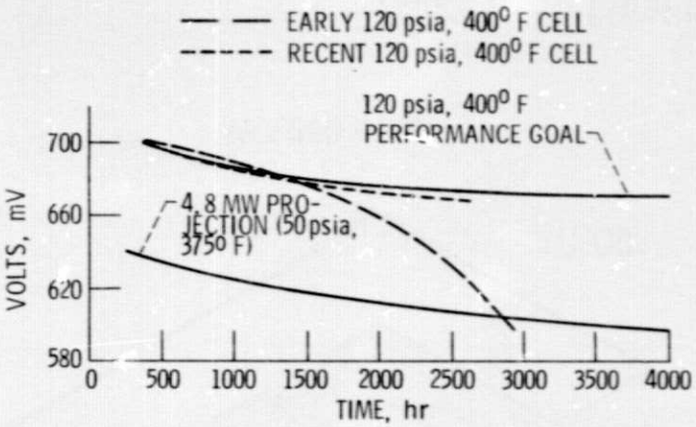

Figure 2. - Improvement in high temperature and pressure cell performance stability.

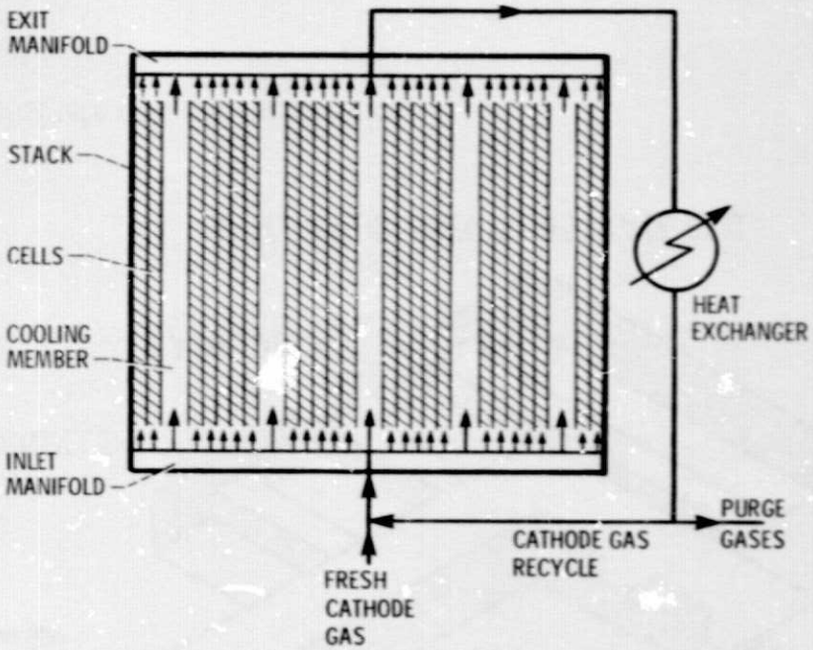

Figure 3. - Distributed-gas (DIGAS) cooling system.

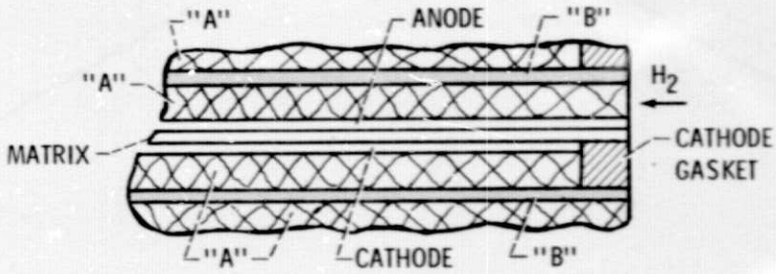

Figure 4. - A-B-A bipolar plate construction.

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16. Abstract

A review of the current commercial phosphoric acid fuel cell system technology development efforts is presented. In both the electric utility and on-site integrated energy system applications, reducing cost and increasing reliability are the technology drivers at this time. The longstanding barrier to the attainment of these goals, which manifests itself in a number of ways, has been materials. The differences in approach among the three major participants (United Technologies Corporation (UTC), Westinghouse Electric Corporation/Energy Research Corporation (ERC), and Engelhard Industries) and their unique technological features, including electrodes, matrices, intercell cooling, bipolar/separator plates, electrolyte management, fuel selection and system design philosophy are discussed.

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