

OPERATIONAL CHARACTERISTICS OF HIGH-TEMPERATURE
FUEL CELLS

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INTRODUCTION

High temperature fuel cells have not received as much attention in the United States as their low temperature counterparts. As a result, from the viewpoint of a total system, the high temperature fuel cell is far less developed. The reason for this is simple. High temperature fuel cells are not as attractive for most military and space applications as low temperature fuel cells.

In Europe, especially in France, England, and Holland, a much larger portion of fuel cell development is directed toward commercial applications. The high temperature fuel cell is being more extensively investigated because it has a much more attractive economic potential. At the Institute of Gas Technology, interest in fuel cells is very definitely directed toward commercial goals. Emphasis on both high and low temperature fuel cell research is aimed at the development of inexpensive systems. This paper on the High-Temperature Molten Carbonate Fuel Cell reports on work which has been in progress at IGT since 1960. Details of much of this work already appear in the literature.¹⁻⁵

The principle advantages of the high temperature fuel cell are the elimination of expensive electrode catalysts from the system and the ability to operate with a variety of fuels and unpurified air.

However, raising the temperature to a typical figure of 900°F or above in a fuel cell which employs a water electrolyte would require a relatively high pressure system. This would be undesirable from both a technical as well as an economic viewpoint.

To avoid this difficulty, two solutions are possible. The electrolyte can be either 1) a salt that melts at a high temperature and which has a very low vapor pressure at this high temperature, or 2) an ion conducting solid used at still higher temperature (about 2000°F). Operating at this latter very high temperature requires the use of noble metals for stability rather than for catalytic purposes. This we want to avoid. For this reason, the fuel cell work at IGT is focused on the use of a molten salt electrolyte operating in the range of 900 to 1300°F.

The IGT molten carbonate fuel cell is patterned after the concept of Broers.⁶ In this system, a eutectic mixture of two or three alkali metal carbonates is mixed with an inert material such as a metal oxide. This forms a ceramic tile-like structure at room temperatures which becomes a non-Newtonian fluid or paste at the operating temperature.

In the IGT system, the paste electrolyte is held between a silver film cathode about 10 microns thick and a porous fiber nickel anode. The cathode is supported by a stainless steel grid. A complete fuel cell element of this type with an active area of one square foot is shown in Figure 1. These elements can be stacked in various ways to produce batteries of any voltage desired.

At the present time, the fuel for the IGT cell is reformed natural gas which is prepared by steam reforming in the presence of commercially available catalysts.

PERFORMANCE CHARACTERISTICS

Performance characteristics of high temperature molten carbonate fuel cells are usually inferior to those obtained in low temperature systems. Since the elevated temperature should improve reaction rate kinetics, an explanation for the poorer performance must be sought in terms of electrode properties and mass transfer processes. Low temperature fuel cell electrodes can be prepared with very high real to geometric surface areas. This may be achieved using high surface area carbons, nickels and precious metal blacks. In the high temperature cells, such active surfaces are unstable. They tend to sinter to form relatively low surface area electrodes.

A second factor affecting performance levels is the nature of the gas-liquid-solid interface at the electrode. In low temperature cells, it has been possible to achieve a relatively effective interface in terms of mass transfer properties, for example, a thin film of electrolyte on the bulk of the electrode, using either a double porosity electrode structure, waterproofing, ion exchange membranes, or matrices. In molten carbonate fuel cells, there has always been evidence of macroscopic flooding on electrodes which have been in operation for some time - e.g. over several hundred hours.

Broers⁷ and we at IGT⁸ have observed evidence for diffusion control in the liquid film for this type cell. Quantitative measurements in terms of film theory have not been possible because of lack of information on the solubility of hydrogen, oxygen, carbon monoxide, carbon dioxide, and water vapor in carbonate melts. Nevertheless, more effective use of electrode surface is clearly warranted.

To see if improved interfacial characteristics could be achieved, experiments at IGT have been conducted with a variety of matrix materials with different surface areas and particle sizes. The use of high surface area metal oxides has led to the development of cells which show no evidence of macroscopic flooding after prolonged operation. They have yielded performance characteristics of the type shown in Figure 2. Earlier performance levels are also shown in this Figure for comparison.

All tests at IGT are performed on hot pressed electrolyte discs. Those discs are prepared by pressing electrolyte powders at 8000 lbs/in² and 950°F. To prevent extrusion of the electrolyte between sections of the dies, aluminum discs are inserted in the pressing unit. The discs are designed to take advantage of the difference in coefficient of thermal expansion between the steel and aluminum to insure a seal tighter than that which could be achieved by mechanical tolerances alone. A thin palladium-silver foil is placed between the electrolyte powders and the aluminum to facilitate release of the electrolyte discs from the die assembly.

A series of experiments on different density fiber nickel electrodes shown in Figure 3, have yielded polarization characteristics shown in Figure 4. Higher density electrodes, such as 43 and 62 percent nickel, show a high level of polarization and the appearance of limiting currents. Electrodes having a density of 15, 20, and 33 percent yield equivalent performance within experimental error. These data were taken with cells having an electrode area of three square centimeters. Results have since been duplicated with cells having surface areas of 32 square centimeters. Cell lifetimes of 850 hours at the indicated performance level is the best achieved so far. These cells are still being tested. From these results, it appears that further optimization of electrolyte and electrodes may yield even better performance levels, placing a whole new perspective on the use of this type fuel cell system.

In the next sections the impact of these new polarization curves on heat transfer and overall system efficiency will be outlined.

HEAT TRANSFER

Considering the voltage characteristic shown in Figure 2, it is apparent that the maximum voltage-current performance is desired. The volume, weight, and cost of the fuel cell system is decreased. It is of interest, however, to examine the engineering implications especially as to how heat transfer is affected by the different voltage-current characteristics.

At IGT, we have considered the three dimensional heat transfer problem in a fuel cell battery.⁹ Figure 5, is a simplified diagram of the fuel cell. Heat transfer in such a cell depends mainly on such factors as battery dimensions, the voltage-current characteristic, the degree of fuel and air conversion, and the physical properties of the materials used to construct the cell. The complete problem is too broad to discuss here. However, in Figure 6 the temperature distribution within such a fuel cell is shown for the voltage-current characteristics of Figure 2. In all cases, the operating cell potential would be 800 millivolts.

It can be seen that the maximum temperature rise from the center of the battery to the assumed isothermal walls (in the case of the 1963 voltage-current characteristic) is only about 7°F. However, when the performance of the cell is improved (Best 1965), the maximum temperature rise increases to about 480°F. In practice, of course, the walls of the fuel cell would not be isothermal. Nevertheless, one can deduce from the experimental data that the low performance fuel cell would require the addition of heat to keep it at operating temperature. On the other hand, it would be necessary to remove heat from the system having the high performance characteristic.

This has been partly verified by constructing the 32-cell stack of elements shown in Figure 7. The performance of this battery was based on the 1963 characteristic. As expected, it was necessary to add heat to the system. The improved performance characteristics suggests another possibility. This is to incorporate reforming catalysts near the electrodes so that waste heat from the fuel cell can be used to sustain the endothermic reforming reaction. At the same time, this would provide a means for cooling the cell.

A further variation in cell performance is seen in Figure 8. Here the effect of conversion on temperature distribution is shown. In the direction of air flow (Z), it can be seen that as the conversion of oxygen is increased, two things happen. The first effect is a rise in the maximum temperature in the battery. This would be expected since less gas is available to remove heat. The second effect is somewhat less expected. The flow of gas through the cell causes a temperature distribution in the cell that is not symmetrical. Physically, this situation can be interpreted as follows: At a high rate of gas flow, heat is being removed from the section of the battery near the gas inlet and is being redistributed to the section near the outlet. In the case of a low rate of gas flow, complete symmetry in the Z direction can be expected.

The purpose of these heat transfer considerations was to determine some feeling for the size and shape of fuel cells which will be built at IGT within the next few years. More important, however, it permits assumptions on the nature of heat inputs in the system. It also establishes what the overall efficiency of such a system might be.

EFFICIENCY

In the high temperature molten carbonate fuel cell, a typical operating voltage that is compatible with the materials from which the cell is built is about 0.7 to 0.8 volts. From the previous discussion, we have seen that cells operating at this voltage level can be thermally self-sustaining. Thus, the overall efficiency of the system depends on the efficiencies of fuel and air conversion, and the reforming requirements. Air conversion is especially important in high temperature fuel cells where the heat content of the oxidant is substantial. In Figure 9, the overall efficiency of an external reformer-molten carbonate fuel cell is shown as a function of the efficiency of fuel and oxygen conversion. The oxidant is air.

For purposes of this example, it was assumed that 1) the external reformer was 60 percent efficient and 2) that no gas-to-gas heat exchange is possible but heat exchange from gas-to-boiling liquid is feasible. These assumptions are based on the need for unreasonably large gas-to-gas heat exchangers especially for low oxygen conversion. While such heat exchangers could be built, they would cost more than the fuel cell itself. However, from a practical viewpoint, steam generation is more feasible, and heat recovery for this purpose has been assumed.

It can be seen that at low oxygen conversion, the overall efficiency is not greatly affected by the efficiency of fuel conversion. It does become more important as the efficiency of the oxygen conversion improves. The reason for this is that when the fuel conversion is high, but the oxygen conversion is low, it is necessary to maintain a separate fuel supply to preheat the air. At low fuel conversion, the air can be preheated by burning spent fuel. The maximum efficiency for the external reforming system without gas-to-gas heat exchange will probably be between 35 and 39 percent.

In Figure 10, a similar analysis is made for an internal reforming system. In this case, the heat for the reformer is supplied by the polarization and entropic heats from the fuel cell. Again, the concept of gas-to-gas heat exchange for preheating feeds has been assumed to be impractical. The overall efficiency for this system is somewhat higher as might be expected. A figure of about 40-43 percent seems to be possible with present technology.

Finally, it should be noted that oxygen conversions higher than 60 percent may be possible in the future. In such a case, the efficiency of the overall system will be even higher. However, it is not likely to be greater than 50 percent unless low cost gas-to-gas heat exchangers can be developed.

ECONOMICS

The only real basis for economic evaluation of fuel cells at the present time is the cost of materials. Lack of knowledge and/or experience with manufacturing components and systems prohibits any more extensive analysis. To a certain extent, storage battery hardware can serve as a guide but this breaks down when confronted with a new component like the paste electrolyte in the high temperature fuel cell.

In an early publication³ we estimated the material costs of molten carbonate fuel cells and related these costs to the performance of IGT cells. Applying a similar method, we have once again presented our performance data in this fashion, (Fig. 11). Material costs are essentially the same as in the earlier publication. It can be readily seen that progress has been made which gives rise to considerable optimism for the economics of this type fuel cell. Typical performance characteristics indicate that material costs of about \$45 per kilowatt can be achieved and the best performance data reduces that figure to less than \$20 per kilowatt.

With respect to further improvement, we might look to using copper in place of silver at the cathode and to reducing the amount of stainless steel used for the current collector in the cell. Lifetime remains the big factor in the case of the molten carbonate fuel cell. Only more intensive experimental work can find a solution to this problem.

ACKNOWLEDGMENT

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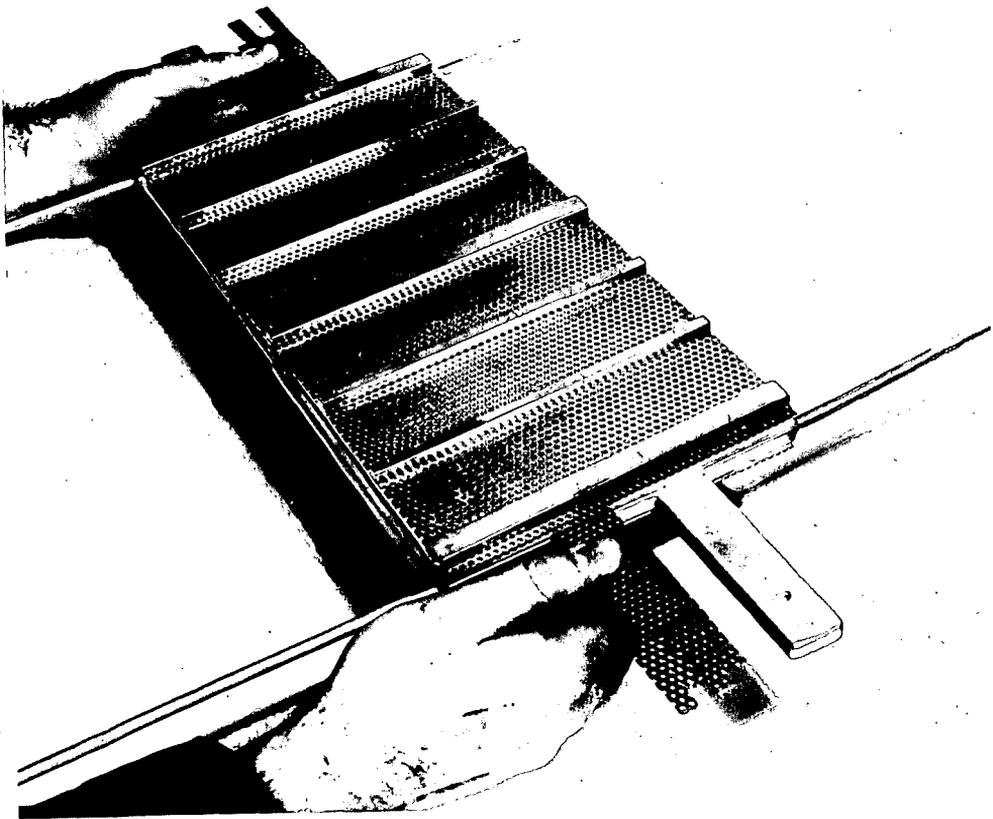


Fig. 1.-SINGLE ELEMENT OF IGT HIGH-TEMPERATURE FUEL CELL BATTERY

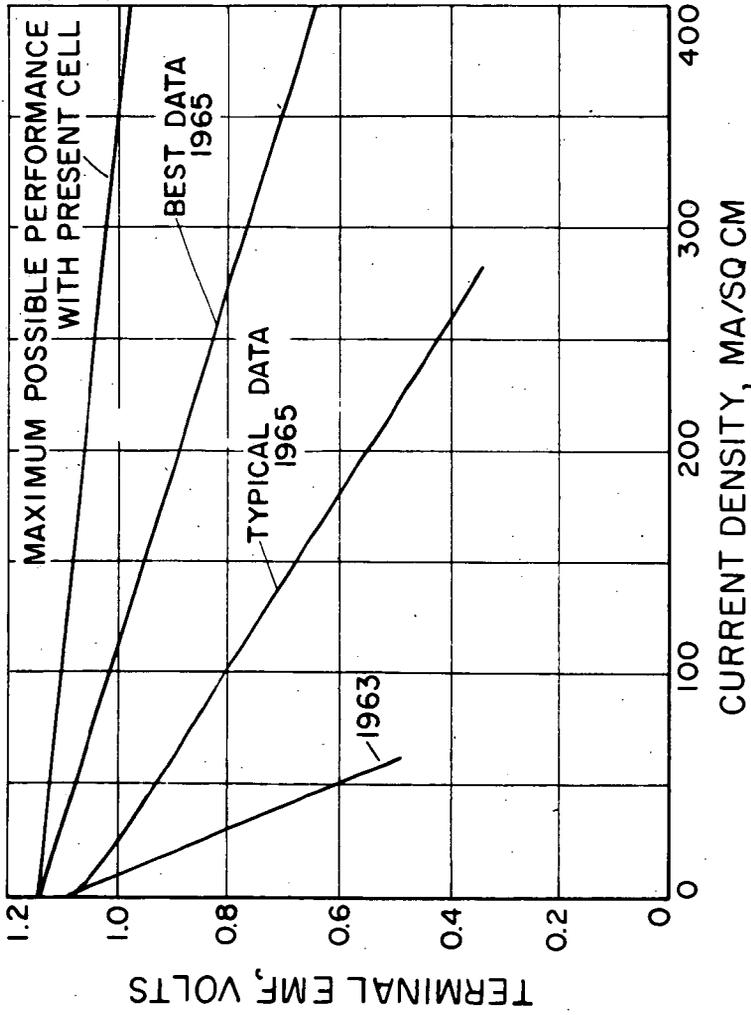


Fig. 2.-PERFORMANCE CHARACTERISTICS OF IGT MOLTEN CARBONATE FUEL CELL



15 %



20 %



33 %



43 %



62 %

Fig. 3. -DIFFERENT DENSITY FIBER NICKEL ELECTRODES USED IN IGT MOLTEN CARBONATE FUEL CELL (120X)

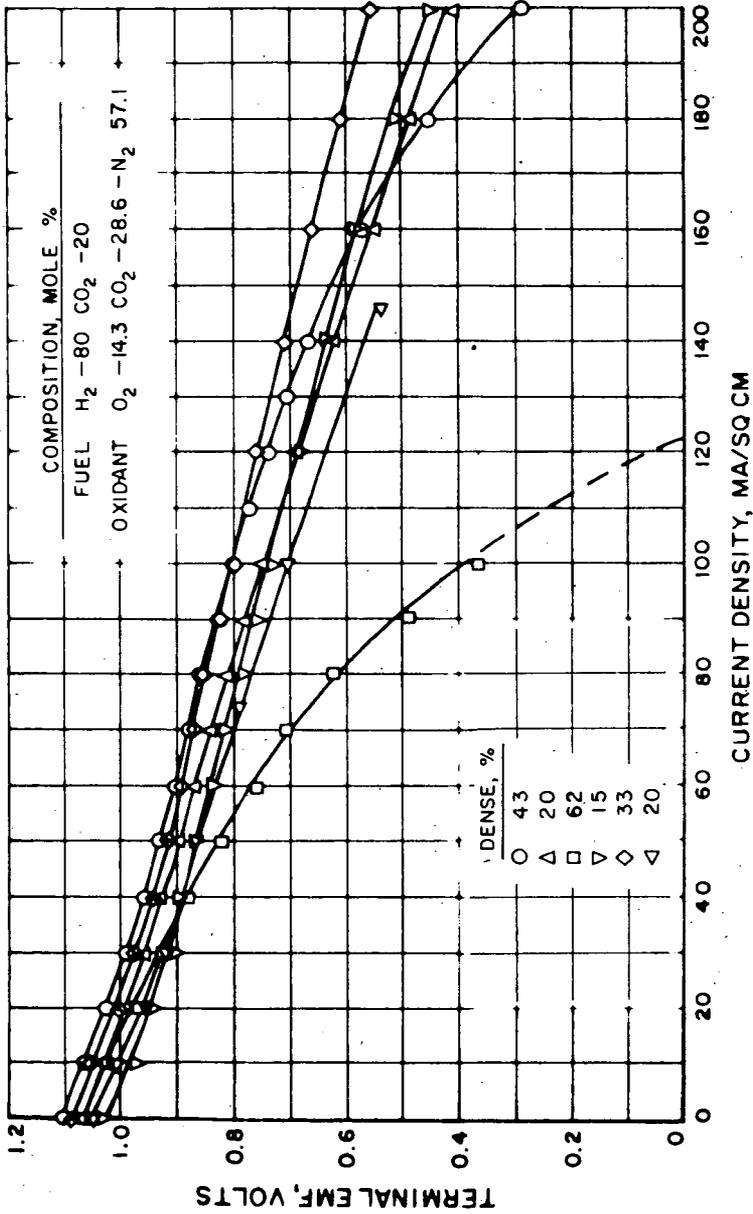


Fig. 4. -COMPARISON OF PERFORMANCE CHARACTERISTICS OF DIFFERENT DENSITY FIBER NICKEL ELECTRODES AFTER SEVERAL HUNDRED HOURS OPERATION

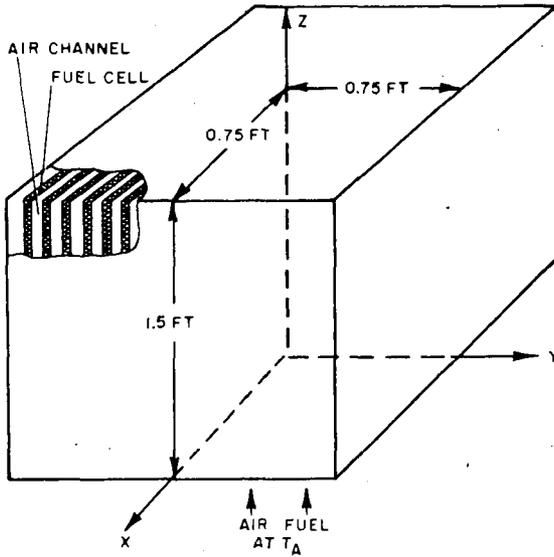


Fig. 5.-MODEL OF FUEL CELL BATTERY USED IN HEAT TRANSFER ANALYSIS

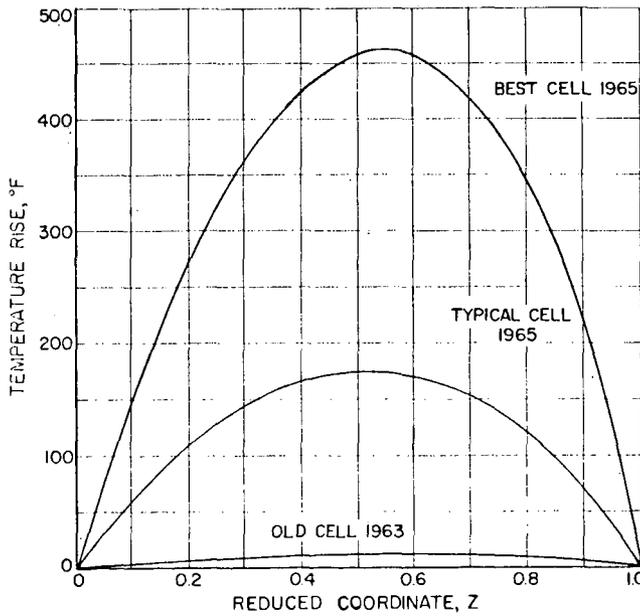


Fig. 6.-EFFECT OF POLARIZATION CHARACTERISTICS ON TEMPERATURE DISTRIBUTION. DIRECTION OF GAS FLOW.

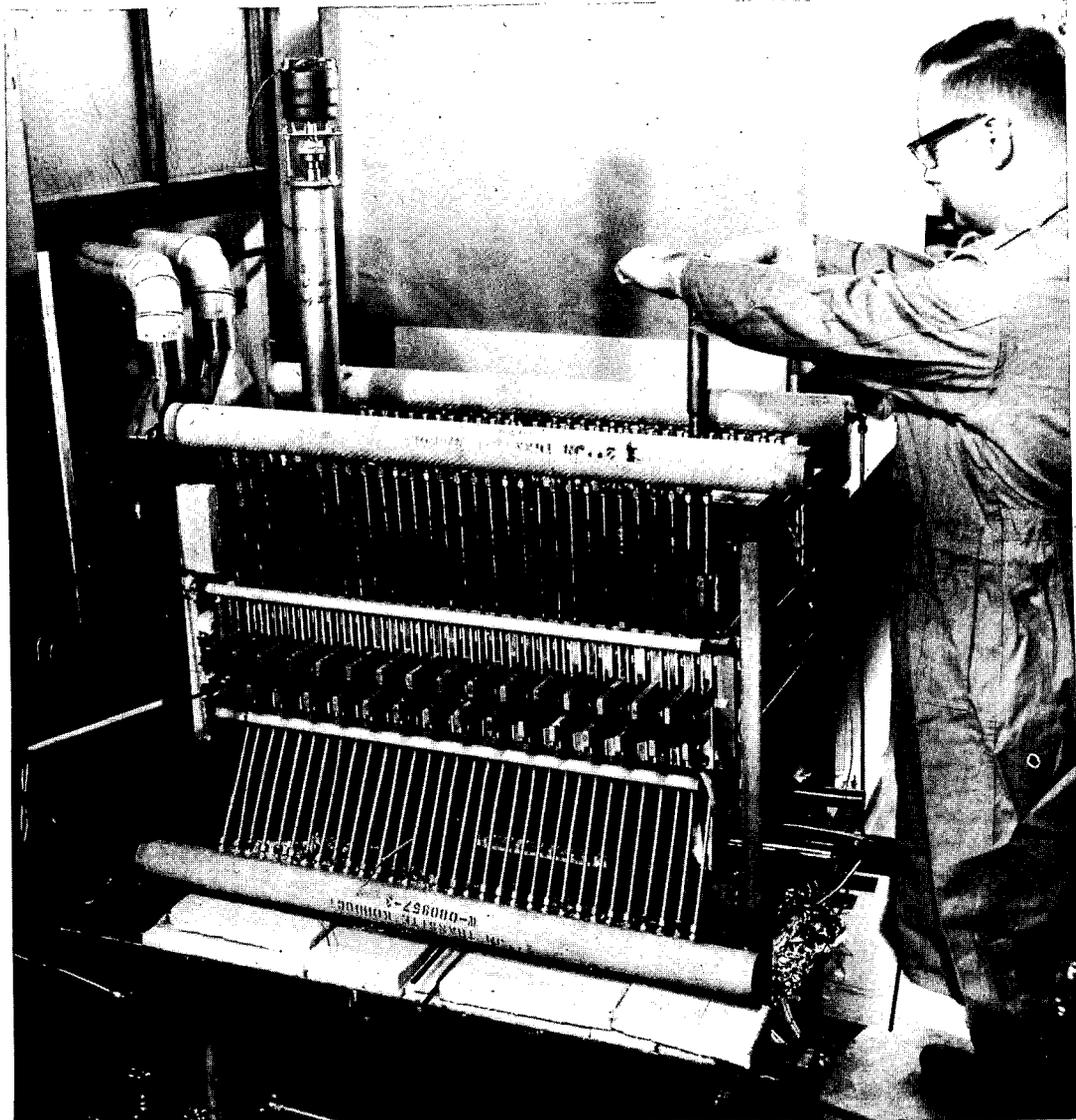


Fig. 7.-IGT 32-CELL HIGH-TEMPERATURE BATTERY

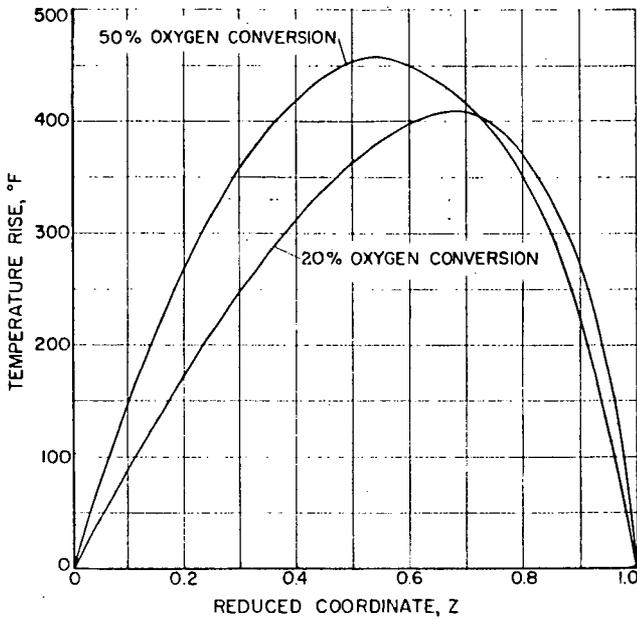


Fig. 8. -EFFECT OF AIR CONVERSION AT HIGH PERFORMANCE LEVELS ON TEMPERATURE DISTRIBUTION IN DIRECTION OF GAS FLOW

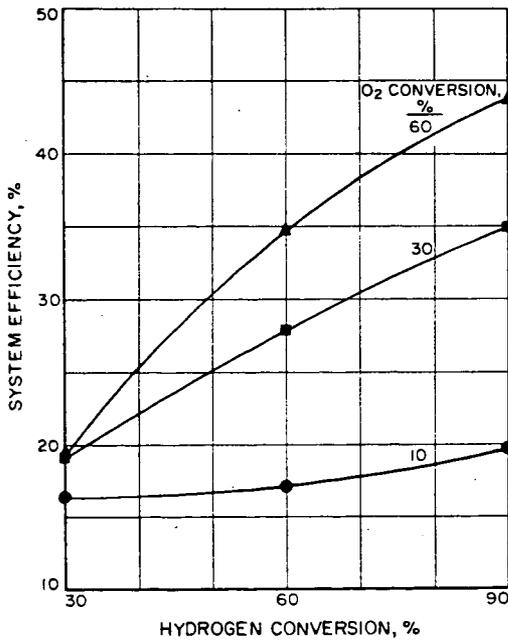


Fig. 9. -OVERALL SYSTEM EFFICIENCY AS A FUNCTION OF FUEL AND OXIDANT CONVERSION WITH AN EXTERNAL REFORMER

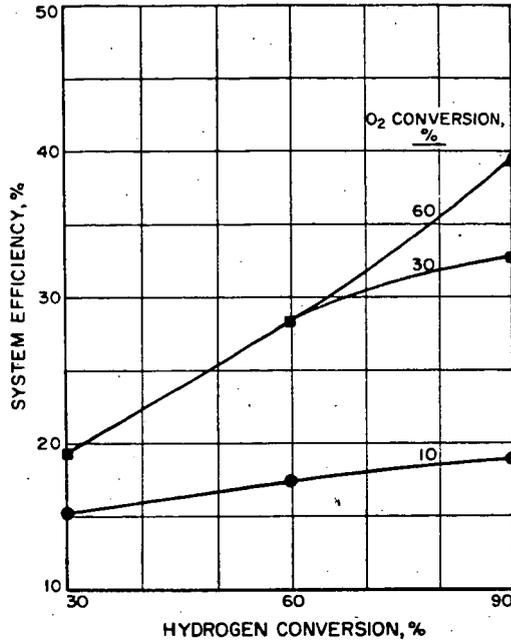


Fig. 10. -OVERALL SYSTEM EFFICIENCY AS A FUNCTION OF FUEL AND OXIDANT CONVERSION WITH AN INTERNAL REFORMER

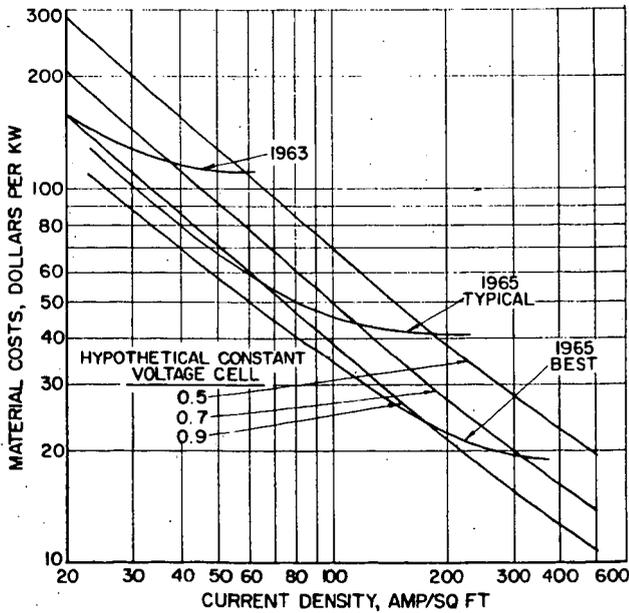


Fig. 11. -ECONOMICS OF MATERIALS USED IN HIGH TEMPERATURE MOLTEN CARBONATE FUEL CELLS