

**A COAL-BURNING SOLID-ELECTROLYTE FUEL CELL POWER PLANT**

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**ABSTRACT**

A plant is proposed which combines a coal-gasification unit with a high-temperature solid-electrolyte fuel cell battery to produce an efficient power generation system. The special requirements imposed on the gasifier by its coupling with a fuel cell battery are discussed and two avenues of investigation - one experimental, one analytical - are proposed. The results of a computer simulation of the overall plant are presented and preliminary projections of plant economics and characteristics are reported. The many research and development problems, which must be solved before such plants are technically and economically feasible, are pointed out, and the progress made towards their solution is discussed.

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### Introduction

The direct conversion of the energy of coal into useful electrical power has long commanded the serious attention of many investigators. This paper describes how in principle a coal gasification unit can be combined with a high temperature solid electrolyte fuel cell battery to produce just such a direct energy conversion system. Before the details of this system are discussed, however, it may be well to review some of the properties and characteristics of the solid-electrolyte fuel cell and to cite some of its operating characteristics for reference a little later on.

### Solid Electrolyte Fuel Cell

The basic component of the Westinghouse solid electrolyte fuel cell is the zirconia-calcia or zirconia-yttria electrolyte. This material is an impervious ceramic which has the unique ability to conduct a current by the passage of  $O^{2-}$  ions through the crystal lattice. The ease with which these ions pass through the electrolyte is measured by the electrical resistivity of the electrolyte. Values of this resistivity for both types of electrolyte as functions of temperature have been published in several places. (9,5)

Fuel cells have been made by applying porous platinum electrodes to this material. (16) The operating principles of the resultant cells have been discussed (1), and optimized batteries constructed from such cells have been described. (4) In brief these batteries operate at about  $1000^{\circ}C$  and consist of short, cylindrical electrolyte segments of about 30 mils thickness shaped so that they can be fitted one into the other and connected into long tubes by bell-and-spigot joints, as shown in Figure 1. The overall length of an individual segment is 1.1 cm, with a mean diameter of 1.07 cm. The segment weighs 2 g and occupies a volume of  $2.0 \text{ cm}^3$ . Electrodes are applied to the inside and the outside of these segments which then have an overall resistance of about  $0.2 - 0.3 \Omega$ . The inner electrode of one segment is attached to the outer electrode of the adjacent

segment, in this way connecting the individual segments electrically in series. Gaseous fuel passes through the center of the resultant, segmented tube, and oxygen or air is supplied on the outside. Figure 2 shows two 15-cell batteries constructed on this principle.

The performance of this and similar batteries has been evaluated for a variety of fuels, using either air and pure oxygen as the oxidizing agent. With  $H_2$  fuel and pure  $O_2$ , a three cell battery has produced an open circuit voltage of 2.9 volts. The current density at maximum power was  $750 \text{ ma/cm}^2$ . At the maximum power point the battery produced 2.1 watts; and each cell segment 0.7 watt - about the same as an ordinary flashlight battery. (2,3)

#### System Configuration

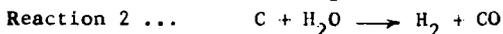
In order to utilize such batteries in the production of electrical power from coal, it is necessary to devise a reasonable overall system for the purpose. As far as the present type cell is concerned, a gaseous fuel is required for cell operation. In order to provide such a fuel to the fuel cell batteries, the scheme shown in Figure 3 has been devised.

Essentially, coal (indicated as carbon on the figure) is introduced into a reactor, into which recirculating gases from cell bank 1 also enter. Cell bank 1 consists of a number of fuel cell batteries of the kind just described. The gases from this cell bank consist primarily of  $CO$  and  $CO_2$  (along with  $H_2$  and  $H_2O$ ) in some definite ratio. In the reactor, some  $CO_2$  and  $H_2O$  is reduced to  $H_2$  and  $CO$  by reacting with the coal. This results in a gas composition leaving the reactor which has higher  $CO/CO_2$  and  $H_2/H_2O$  ratios than that of the entering gas. The gas leaving the reactor is then cleaned and passed, without cooling, into the fuel cell bank 1. There it serves as fuel for the cell unit, combining with oxygen which has been ionically transported through the cell electrolyte, resulting in power generation. The gases leaving this cell bank are split into two streams. One stream is recirculated to the reactor at a given recycle rate, completing the major system loop. The second stream is sent to fuel cell bank 2, which completes the combustion of  $CO$  to  $CO_2$  and  $H_2$  to  $H_2O$ .

The reason for this system configuration is that it allows all of the oxygen which enters the system to react with the coal to pass through the electrolyte and thereby contribute to the electrical energy output of the system. In addition, by limiting the degree of oxidation in cell bank 1, the concentrations of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  entering the reactor are only slightly removed from their equilibrium values. This means the bed operates under more nearly reversible conditions, so that overall the maximum electrical energy output from the coal-oxygen reaction is recovered. In order to obtain such recovery, however, an over-size coal reactor, excessively large cell banks relative to the power produced and high recycle rates would have to be employed. A compromise must be effected between the desire to obtain high recovery and the desire to avoid large and expensive equipment. Much of the work to date on this system has been geared to locate those optimum conditions which best effect this compromise.

#### Reactor Considerations

Because the reactor must contribute to an overall system optimum, certain of its design features must be tailored specifically to the task of gasifying coal in a rather unusual way. In most conventional gasifiers, oxygen is introduced at some point directly into the reactor. The fuel-cell power plant has been so designed, however, that oxygen is introduced into the reactor only in the form of  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ , and  $\text{CO}$ . The coal may be considered to be gasified primarily by the two reactions:



which proceed at rather slow rates compared to the direct oxidation reaction:



Moreover, reactions (1) and (2) are endothermic, as compared to reaction (3) which is quite exothermic, meaning that heat will have to be supplied to the reactor where (1) and (2) occur. The problem of supplying this heat considerably complicates the overall reactor design.

Because reactions (1) and (2) proceed at relatively slow rates, it is advantageous to take steps to increase the rate of these reactions. Two such steps which may be taken are the maintenance of a high reaction temperature or the use of a highly reactive fuel. The temperature level of the reactor is limited, however, due to the fact that the reactions which occur there are endothermic. This means that heat must be supplied to the reactor from an external source (e.g. the fuel-cell banks where exothermic oxidation reactions are taking place and internal battery heating due to  $I^2R$  losses is occurring). This transfer may be accomplished either directly through the walls of the gasifier or indirectly in the sensible heat of the recirculating gases. However; some finite temperature gradient will be required to transfer this heat, constraining the reaction temperature to be less than the source temperature. If the source is the cell banks, an upper temperature limit is imposed due to the mechanical and structural properties of the cells. This is reflected in an upper temperature limit on the reactor of about  $1000^{\circ}\text{C}$ .

At this temperature, many of the fuels, which appear attractive for fuel cell use because of the clean nature of their gasified products, become unusable because of their low reactivity. On the other hand, those fuels which are reactive at this temperature produce quantities of tars, pitches, and other impurities which may have a deleterious effect on cell operation. The presence of such undesirable gasification by-products is increased even further in the absence of a direct supply of oxygen in the reactor. The issue is complicated by the fact that cell performance is most efficient when the fuel contains high concentrations of either CO or  $\text{H}_2$ . In the reactor, however, large quantities of CO and  $\text{H}_2$  inhibit reactions (1) and (2), retarding their reaction rate. This effect compounds the problem of striking an effective compromise between a reasonable reactor size and highly efficient cell operating conditions.

In order to study the effect of such factors as these on the overall system performance, two avenues of investigation have been pursued. A small test reactor has been built to which a stream of gases containing CO,  $\text{CO}_2$ ,  $\text{H}_2$  and  $\text{H}_2\text{O}$  can be added, simulating the recycle gases in the

proposed design. Secondly a systems study of the proposed plant has been undertaken, culminating in a computer simulation of both reactor and cell banks. The purpose of this study was to demonstrate the engineering feasibility of such a plant, to assess its economic desirability and to point out areas where further development effort would be most beneficial.

#### Test Reactor

The test reactor is shown in Figure 4. It consists of a 1-3/8" i.d. inconel tube located in a commercial four-zone furnace. This reactor itself is of conventional fixed bed design with the exception of two small fuel cells which are located near its inlet and outlet. (The inlet cell is barely visible in the figure.) The open circuit voltages of these cells are used to determine the compositions of the inlet and outlet streams, thereby providing a measure of the extent of gasification which occurs in the reactor. The use of the cells as measurement devices allows a rapid and convenient analysis of the gases to be obtained immediately upon their exit from the fuel bed, without cooling or tampering with the gas stream in any way. Initial runs using this setup have been entirely satisfactory, and a report from this laboratory on the observed gasification kinetics will be presented shortly.

In addition to providing kinetic data for the various coals of interest, the test reactor serves several other purposes. It is being used to study not only the inhibiting effect of high CO and H<sub>2</sub> concentrations on the gasification, but to examine the effects of operating very near to the equilibrium gas compositions, which yield the highest cell efficiency. The problems of cleaning and filtering the reactor output gases are being examined, and the effects of such materials as fly-ash, pitches, and tars on cell performance are being explored. Overall, the test reactor is used to supply necessary engineering information for the computer simulation in order that optimum system operating conditions and general feasibility can be determined.

### System Simulation

Some preliminary results of this simulation may be of interest at this time. The simulation includes both fixed and fluidized bed conditions, operated either isothermally, or adiabatically. It includes both the reactor and cell banks 1 and 2. In the reactor the surface reactions between carbon and steam and  $\text{CO}_2$  are assumed to be controlling. In the gas phase, the "water gas" equilibrium were assumed to hold. Rate expressions of the Langmuir-Hinshelwood type were adopted for the surface reactions, and published data for the specific constants were employed in the simulation. (7,10,11,14) In the cell banks equilibrium conditions were assumed to prevail as far as the gas composition was concerned, with the generated voltages modified to account for both the losses due to internal cell resistance and those which occur due to cell polarization.

Figure 5 illustrates how the gas composition varies in its passage through the main system loop, as predicted by the computer simulation. Note that as  $\text{CO}_2$  and  $\text{H}_2\text{O}$  react with the carbon, their percentages decrease, while the percentage of  $\text{CO}$  and  $\text{H}_2$  increase. In the gas space immediately adjacent to the coal bed provision has been made for some volatile material to enter the gas stream; this is indicated by the sharp change in composition indicated by the dotted lines. In the cell banks as oxidation occurs, the percentage  $\text{CO}$  and  $\text{H}_2$  decrease while  $\text{CO}_2$  and  $\text{H}_2\text{O}$  increase. In the example shown on Figure 5, the hydrogen (H) to carbon ratio, H/C, is 1.0. The oxygen (O) to carbon ratio at the inlet to the reactor is 1.67 and the system is considered to be isothermal at  $1200^\circ\text{K}$ .

In addition to predicting the change in gas composition as it moves through the system the simulation provides the size reactor needed to generate sufficient gaseous products to attain an assigned power level, if a particular fuel, an inlet O/C ratio, a recycle rate (moles of  $\text{CO}$  and  $\text{CO}_2$  flowing through cell bank 1 per mole of carbon C, consumed in reactor), a percent fuel burnup, and a system temperature are specified.

Figure 6 shows how the reactor size required to gasify a g mole/sec varies with conversion, for an inlet  $n_{\text{O}}$  value of 1.67 and a temperature

of 1200°K. Plotted on the same figure is a curve which represents the total theoretical power output of the system per g. mole / sec gasified - also as a function of conversion. Two cases are illustrated on the figure for reactor size: one corresponding to plug flow conditions in the reactor; the other corresponding to perfectly mixed conditions. The reactivity of the carbon in the bed itself was taken to be that for retort coke, and published rate data for this material was used in the construction of Figure 6<sup>(10,11)</sup>.

Figure 7 shows how a normalized gasifier size depends upon degree of conversion. Here the size of the reactor in grams per 100 theoretical watts produced by the system is plotted vs the oxygen to carbon ratio,  $n'_0$ . This figure is obtained from the data on Figure 6 by dividing reactor size by power produced, each per g. mole gasified per second. The case presented in Figure 7 is for isothermal operation at 1200°K. Two inlet ( $n'_0$ ) values, 1.25 and 1.67, are illustrated for both plug flow and perfectly mixed reactors. The gas flow in the cells is assumed to be plug flow in all cases. The size of a reactor for a particular configuration and inlet condition is obtained by locating the desired ( $n'_0$ ) value on the abscissa and reading the ordinate for the appropriate curve. To demonstrate the effect of temperature on reactor size, Figure 8 shows a plot of reactor size vs. the reciprocal absolute temperature for an inlet ( $n'_0$ ) of 1.67 and an outlet ( $n'_0$ ) of 1.25.

In addition to such results obtained for isothermal operation, the simulation predicts system size and behavior under adiabatic conditions. In this case the net heat release by the exothermic reactions and  $I^2R$  losses in the cell banks contributes to increasing the sensible heat of the circulating gases. The heated gases then pass through the reactor, losing heat to the endothermic reactions which occur there. The simulation has shown that, depending on conversion and recycle rate, the temperature rise in the cells may exceed, balance, or be less than the temperature drop in the reactor, and careful design will be required to adjust effectively the various system parameters in order to attain a proper thermal balance.

### Discussion

On the bases of the investigations both with present-day laboratory equipment and with the overall system simulation certain key problems have emerged which require solution before the proposed plant can be considered as a feasible power generation system. There is considerable incentive to seek out these solutions, however, because of the many desirable features such a plant would have, such as high overall efficiency, even for small sized plants, inherently simpler operation due to the lack of moving parts, and compact overall size.

Some of the problems connected with the suggested coal gasifier have been discussed briefly in a previous section. Essentially these problems center around the fact that heat will have to be supplied to the reactor and that the gasification temperature is not spectaculaty high. Both of these factors indicate that a rather large scale reactor may be required compared to the idealized size obtained from the iso-thermal simulation. However Jolley, Poll and Stantan reporting on the fluidized gasification of non-caking coals with steam in a small pilot plant have shown that reasonably good heat transfer through the walls of the gasification chamber can be attained with as little an overall temperature driving force as 100°C. (8) This is encouraging in that it means that much of the heat which is generated in the cell banks can be transferred directly to the reactor by conduction through separating walls. Moreover, the temperature gradient required to do this is not excessive.

In addition to this external heating of the gasifier, some measure of internal heating will be accomplished by means of the recycle stream, which serves as a heat-carrying fluid as well as a gasification agent. Preliminary indications are that the recycle molar flow rate will be three to six times larger than the amount of coal gasified per unit time. This means that a good portion of the endothermic heat of reaction can be provided by the recirculating gases.

The final solution to this problem remains still to be engineered, but for the reasons given above, there is some indication that a reasonable

answer may be found, resulting in a reactor size per theoretical watt approaching that given in Figure 7.

The fuel cell batteries themselves are not finished products to be sure, and still require considerable development effort. Essentially this effort is directed towards extending the life of the devices, minimizing associated electrode polarization losses, and reducing cell costs. Considerable progress has been made on these problems and reports concerning this progress have been published by this laboratory.<sup>(6)</sup>

If these and other problems can be solved, the overall plant described in this paper provides a very attractive method for the gasification and utilization of coal. Because it uses air as the oxidizing agent, this raw material cost is negligible. However, because only oxygen enters the major system process stream, the gasification step proceeds as if pure industrial oxygen had been used, with no nitrogen dilution of the gasified products. It is reported that the cost of oxygen represents about 40-60% of the raw material costs of gasification processes using industrial oxygen.<sup>(15)</sup> Thus, if viewed only as a gasification unit, the process offers the possibility of reducing a major portion of the raw material costs.

In addition to this savings, however, the attractiveness of the proposed scheme comes from the fact that the plant utilizes the gasified coal products in an extremely efficient manner. It has been estimated that the overall efficiency of the proposed plant in converting coal to electricity will be about 60%.<sup>(13)</sup> Present day stations operate at efficiencies of about 38-42%, with very large installations required to attain the higher figure.<sup>(12)</sup> Since fuel costs represent about 40-50% of the cost of generating electricity by present-day techniques, the improved efficiency of the proposed plant represents a highly desirable improvement.

In order to attain this efficiency however, certain demands on system performance will have to be met. Table I lists the projected system characteristics and operating conditions based on an economic optimization of the system as currently envisioned.<sup>(13)</sup> On the basis of this study, it has been suggested that: 1) if bell-and-spigot cells with

an internal resistance of  $0.25 \Omega$  can be made and fabricated into banks for \$0.10 a cell; and 2) if a minimum payout time of 5 years can be used to recover the investment, return a profit, pay taxes, and provide maintenance (the cell life must obviously exceed 5 years), then coal-burning solid-electrolyte fuel-cell power systems can produce electrical energy at 5 mills/kw-hr; the system efficiency will be 60%, and the current density in the cell banks, will be 700 milliamps/cm<sup>2</sup>.

TABLE I

<u>Projected System</u>	<u>Characteristics</u>
Overall Efficiency	60-70%
Cell Banks	
Current Density	700 ma/cm <sup>2</sup>
Cubic Feet per Kilowatt	0.3 ft <sup>3</sup> /kw
Reactor	
Volume per Kilowatt	0.1-5.0 ft <sup>3</sup> /kw
System	
Recycle Ratio (moles recirculated per mole gasified)	3-6
Percent Fuel Burnup	95-98%

Cells have already been built which have an internal resistance of  $0.3 \Omega$ . The cost of materials for these cells is about \$0.004 for zirconia and \$0.03 for platinum, and effort is being expended to find suitable replacements for platinum to further reduce this raw material cost. Cells have operated at 750 milliamps/cm<sup>2</sup> for short periods of time on H<sub>2</sub> fuel. These facts indicate that solutions to the cell problems may be in the making. If they can be attained, along with reasonable solutions to the coal reactor problems, the proposed plant offers much promise as a power generation station of the future.

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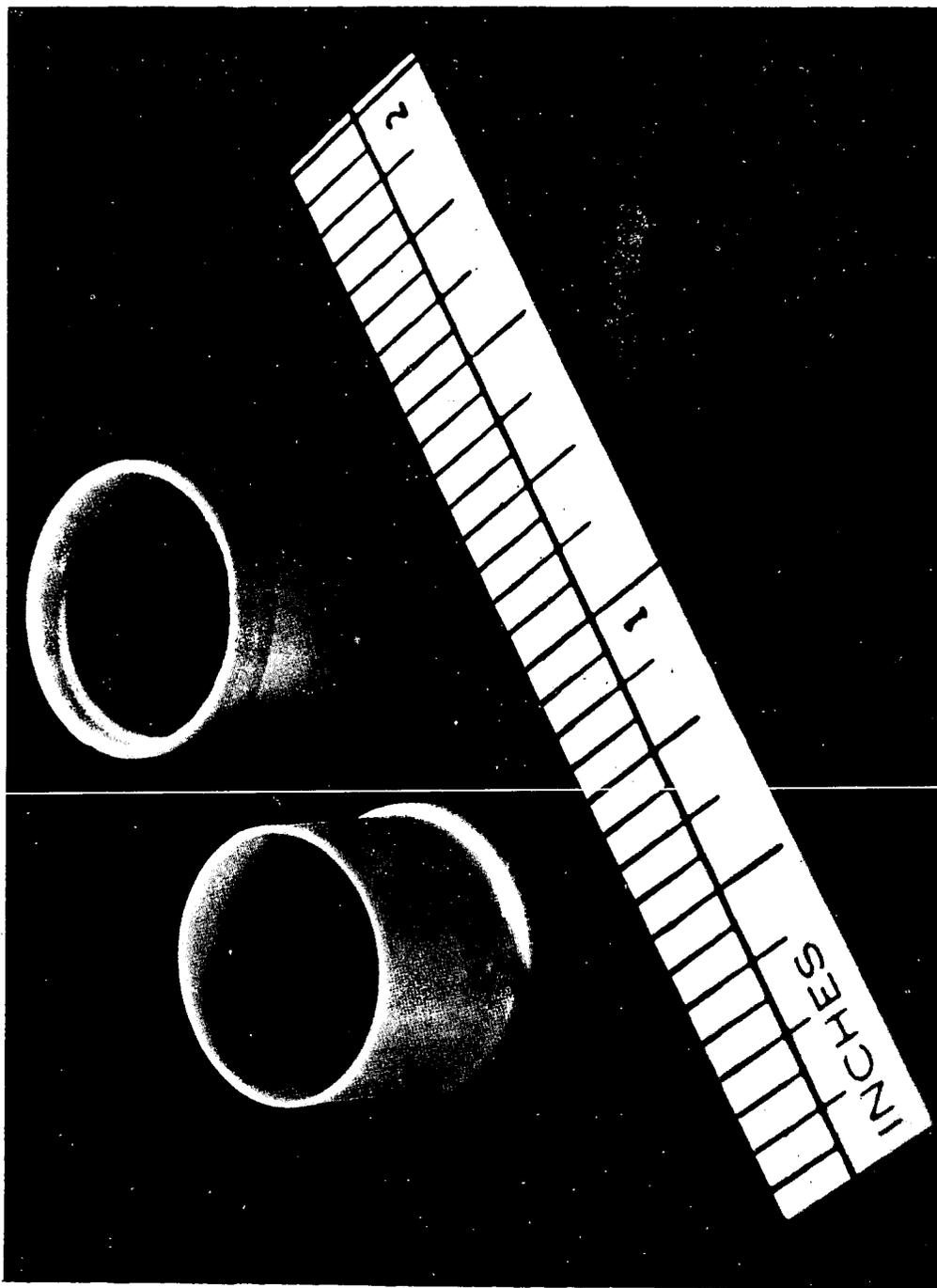


Figure 1 - Bell-and-Spigot Segments

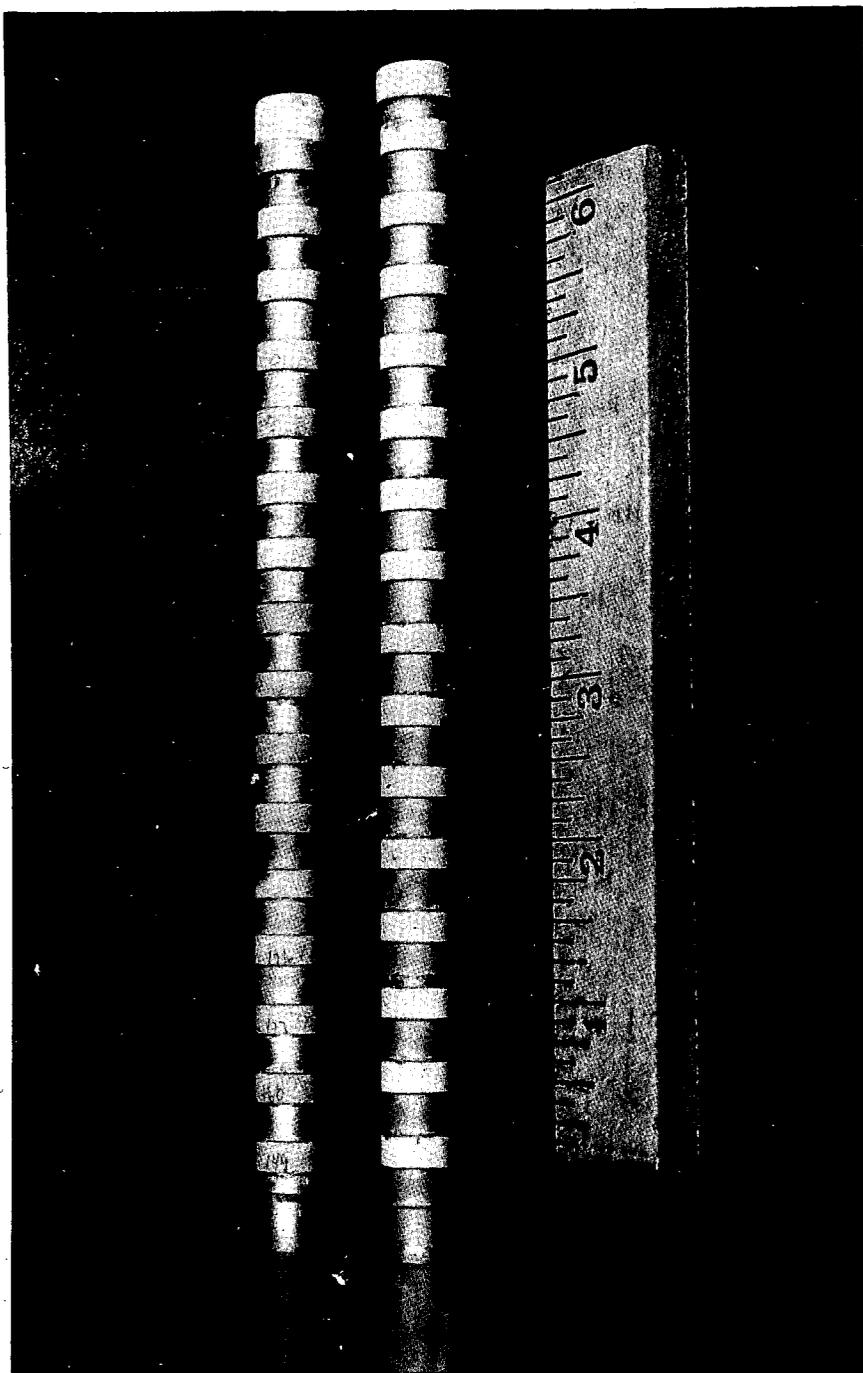


Figure 2 - Two 15 cell solid electrolyte fuel cell batteries without leads.

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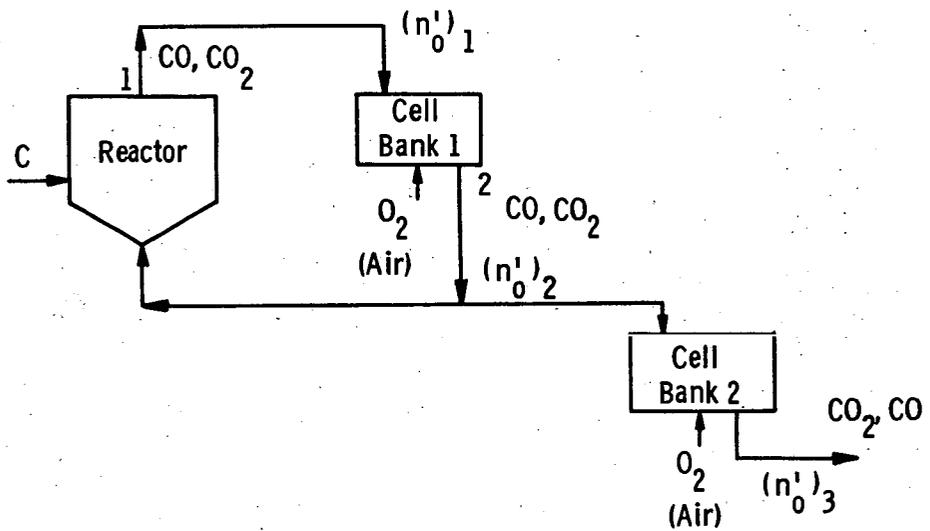


Fig. 3 — Simplified coal burning fuel cell system

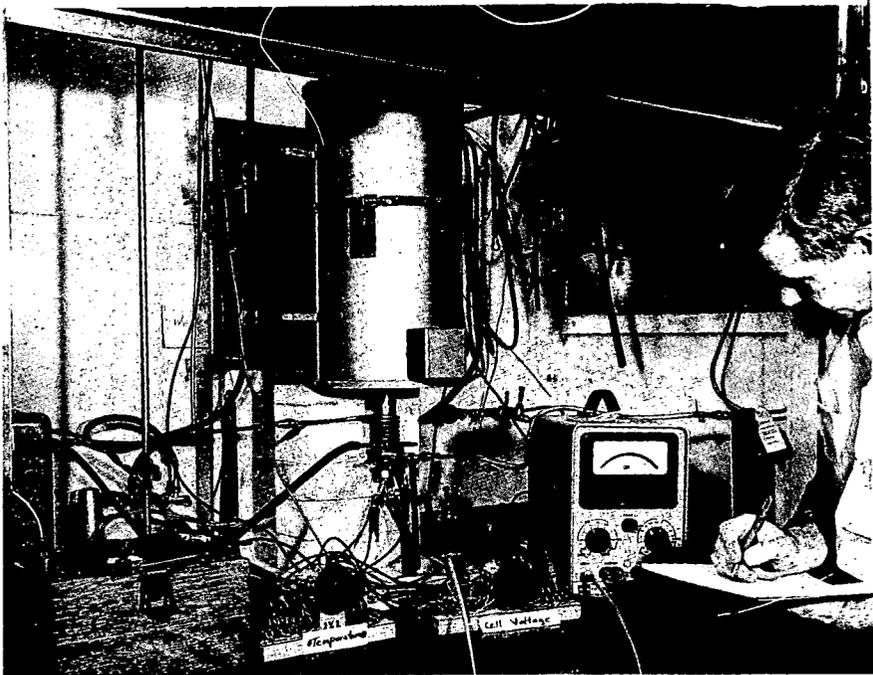


Figure 4 - Test Reactor

CURVE 567690-A

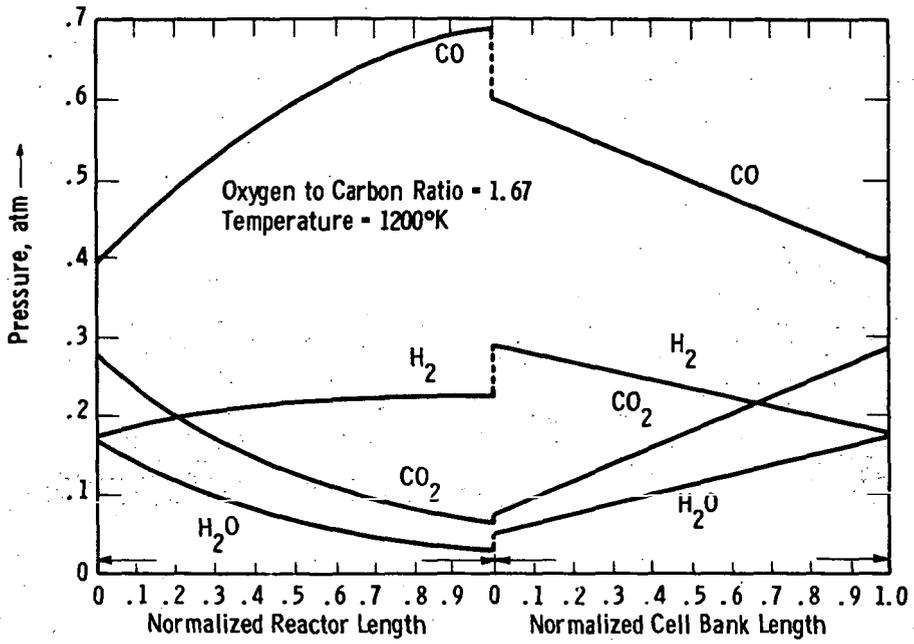


Fig. 5-Simulated reactor and cell bank concentration profiles

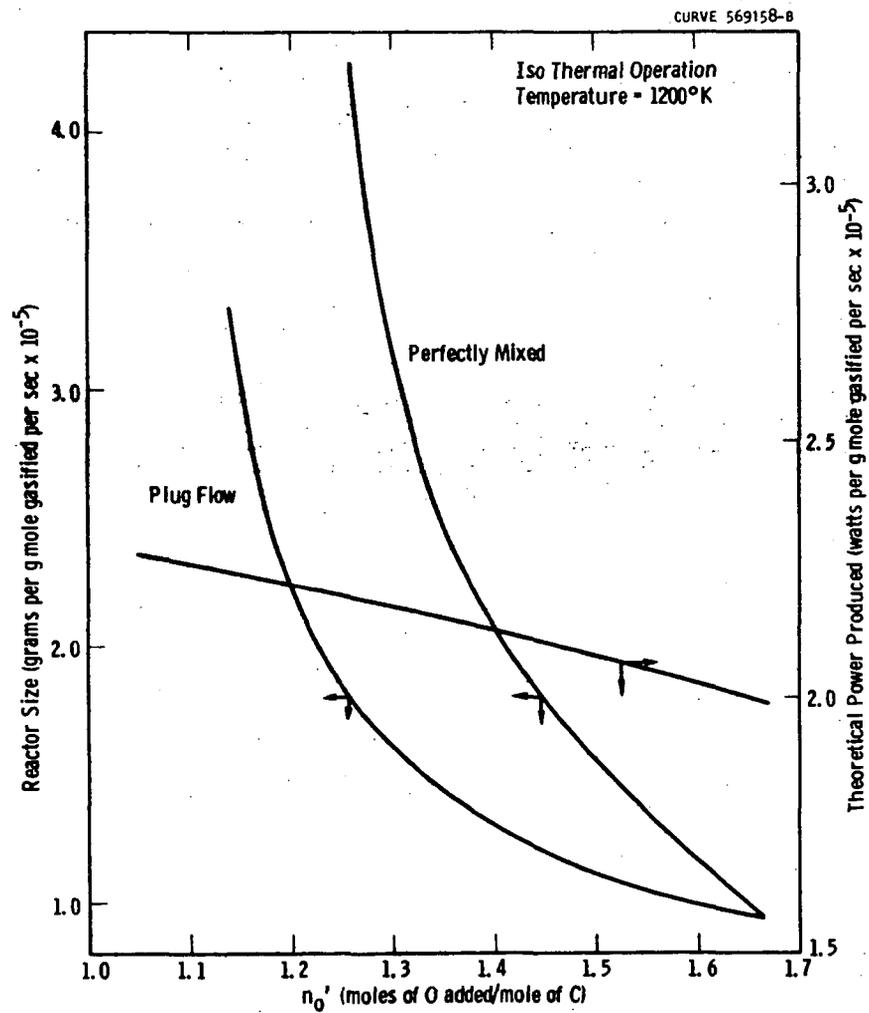


Fig. 6—Reactor size and system power vs conversion

CURVE 569159-8

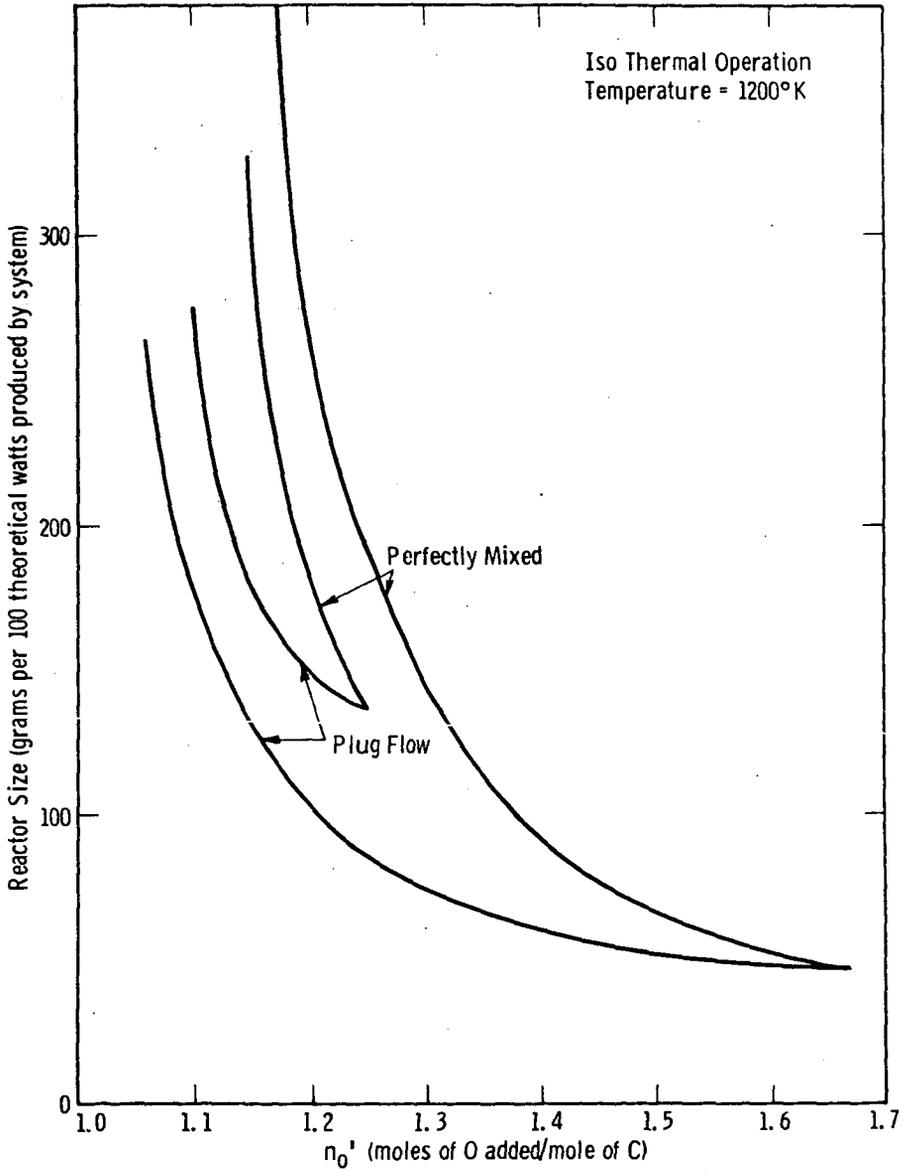


Fig. 7—Effect of conversion on normalized reactor size

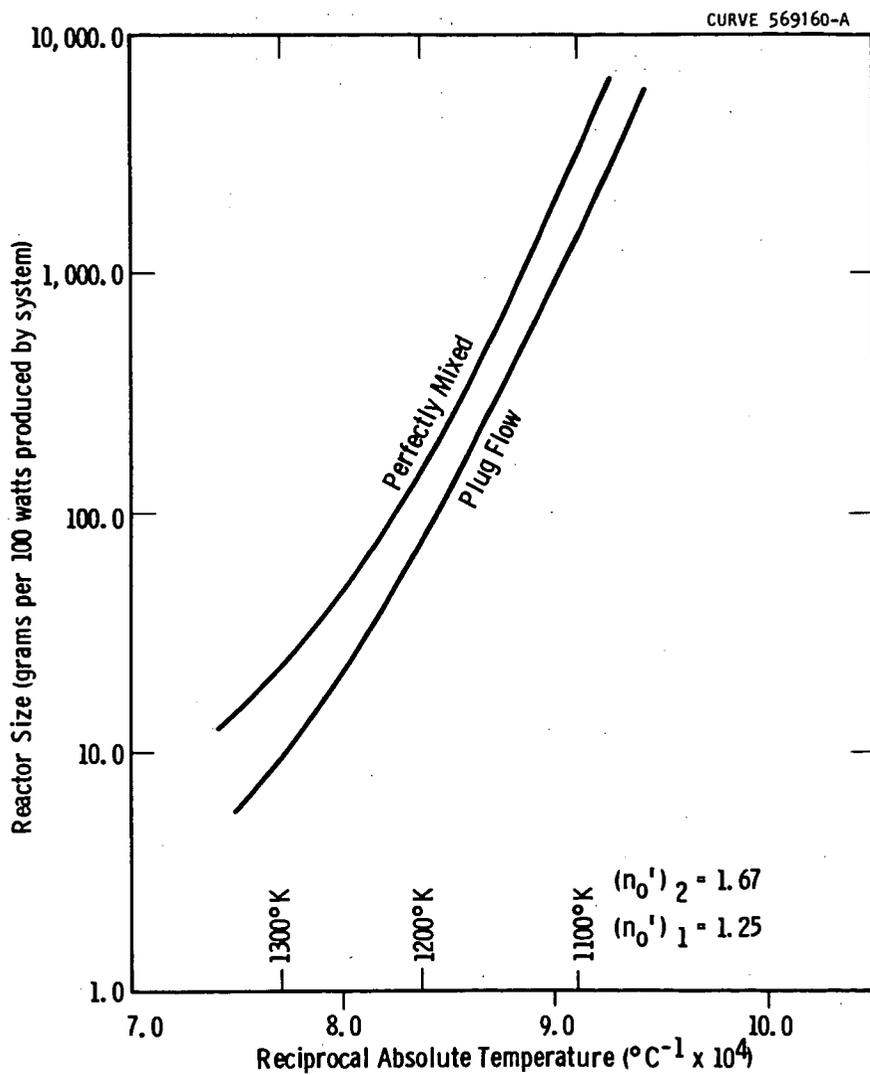


Fig. 8—Influence of temperature on reactor size