Hydrazine-Air Fuel Cells

Hydrazine-air fuel cells emerge from the laboratory.

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Before discussing hydrazine-air fuel cells let us first relate them to fuel cells in general. When the well-informed layman hears the term "fuel cell," he most likely thinks of the hydrogen-oxygen fuel cells used for electric power aboard Gemini spacecraft (*1*). He may recall earlier reports of a tractor powered by a fuel cell (*2*). And no one can have missed the recent flurry of publicity concerning electric vehicles, culminating in the demonstration of the 160-kw Electrovan powered by fuel cells (*3*). From the research laboratories comes a still wider variety of fuel cells, some in the "test-tube" stage, others beginning to resemble power plants (*4*).

These various fuel cells show little resemblance to one another in mechanical design or appearance; they differ even in the nature of the basic electrochemical reactions used to generate electrical output. The only common factor is the technical definition of a fuel cell (*5*): "an electrochemical cell which can continuously change the chemical energy of a fuel and oxidant to electrical energy by an isothermal process involving an essentially invariant electrode-electrolyte system."

The key words are "essentially invariant electrode-electrolyte system"; they mean that the fuel-cell system can continue to deliver electricity under steady-state conditions so long as reactants are supplied at appropriate rates, and so long as products of the cell reaction and waste heat are removed at equivalent rates. For example, in a simple H_2-O_2 system, 1 gram-mole of water must be removed from the system for each gram-mole of hydrogen consumed in the electrochemical reaction, in conformity with what is perhaps the most widely known chemical equation:

$$H_2 + 1/2O_2 \rightarrow H_2O \qquad (\Delta G = -54.6 \text{ kcal}) \qquad (1)$$

On the basis of this reasoning we can sketch a diagram of a fuel-cell system symbolized as a "black box" (Fig. 1). Normally the power output is considered to be the independent variable, while the other four dependent variables must be controlled for maintenance of stable operation. Thus the operator can adjust the external load within some acceptable range of power levels, and the feed rates of the fuel and oxidant (as well as the rejection rates of the reaction products and waste heat) must so adjust as to compensate the change. Except for the very simple low-power fuel cells, such control normally requires some miscellaneous collection of additional components, known in the trade as auxiliaries, to supply and control feed of the reactants, remove by-products, and reject waste heat. The auxiliaries may also control the electrical output by imposing voltage regulation, or provide overload protection by limiting the current (Fig. 1).

In applying this general model of a fuel-cell power supply to describe the functioning of a hydrazine-air system, one can usefully break down the system into the fuel cells themselves and a set of auxiliaries.

The Hydrazine-Air Fuel Cell

The hydrazine fuel is supplied to the cell as a dilute solution of hydrazine in a circulating "anolyte" consisting of an approximately 9*N* solution of KOH in water (Fig. 2). A portion of the dissolved hydrazine in the anolyte diffuses into a porous conductive anode where it reacts electrochemically to produce nitrogen, water, and electrons according to the reaction (*6*)

$$N_2H_4 + 4OH^- \rightarrow N_2 + 4H_2O + 4e^- \qquad (2)$$

The water and bubbles of gaseous nitrogen are swept away in the circulating anolyte while the electrons flow from the anode by way of metallic current collectors to the external load circuit. The hydroxyl ions consumed in the anode reaction are supplied by a KOH electrolyte immobilized in a microporous matrix (usually asbestos) positioned between the electrodes. Oxygen required for the cathode reaction is supplied by passage of an excess quantity of air through a chamber positioned behind the porous cathode; a portion of this oxygen diffuses into the porous cathode, where it enters into a sequence of electrochemical and chemical reactions (*7*):

$$O_2 + 2H_2O + 2e^- \rightarrow H_2O_2 + 2OH^- \qquad (3)$$

$$H_2O_2 \rightarrow H_2O + 1/2O_2 \qquad (4)$$

Catalysts are supplied within the cathode to increase the rate of reaction (Eq. 4), since any decrease in concentration of peroxide increases the electrode potential available in the electrode reaction (Eq. 3); this increase follows directly from the familiar Nernst equation (*8*)

$$E = E^0 - (RT/nF)\ \ell n\ \{[H_2O_2][OH^{-1}]^2/[O_2][H_2O]^2\} \qquad (5)$$

where E is the electrode potential in fact, E^0 is the theoretical potential of an ideally reversible electrode under standard (unit activity) conditions; and R, T, n, and F represent the gas constant, temperature, number of electrons transferred, and Faraday constant, respectively. The chemical formulas in square brackets represent the activities of the reactants in and products of the electrode reaction. If one makes the activity $[H_2O_2]$ small enough, the log term becomes negative, so that the real electrode potential becomes greater than the theoretical "standard" potential. In a typical fuel-cell cathode employing catalyzed porous carbon, the concentration of hydrogen peroxide is less than $10^{-10}M$ and frequently nearer to $10^{-12}M$; such concentrations, far too low for detection by chemical analysis, are deduced indirectly from electrochemical effects.

Since decomposition of peroxide in accordance with Eq. 4 results in regeneration of oxygen within the elec-

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trode, immediately available to enter Eq. 3 as a reactant, and since the concentration of the hydrogen peroxide is almost immeasurably low, generally we may without loss combine Eqs. 3 and 4 into a net electrode reaction:

$$O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \qquad (6)$$

Comparing Eqs. 6 and 2, we see that the cathode reaction (Eq. 6) generates four hydroxyl ions that diffuse across the electrolyte space to the anode, where they are consumed (Eq. 2); the electrolyte thus remains invariant, as required for steady-state operation of a fuel cell. The four electrons liberated at the anode flow through the external load (providing useful electrical power) and, as required for electrical balance, exactly satisfy the need for four electrons to enter the cathode reaction (Eq. 6).

By adding Eqs. 2 and 6 we arrive at the net cell reaction:

$$O_2 + N_2H_4 \rightarrow N_2 + 2H_2O \quad (\Delta G = -145 \text{ kcal}) \qquad (7)$$

This reaction indicates that one mole of oxygen from the air is required for each mole of hydrazine fuel consumed, and that one mole of nitrogen gas and two moles of water are formed as byproducts that must be removed from the system.

Equation 7, combined with a little elementary thermodynamics, can supply much useful information concerning the hydrazine-air fuel cell. The theoretical "reversible" voltage of the fuel-cell reaction (which represents the best voltage that the fuel cell can possibly supply; that is, the "100-percent efficiency" point normally quoted in electrochemical literature) is given by

$$\Delta G^0 = -nFE^0 \qquad (8)$$

where E^0 is the reversible voltage for the "standard state" reaction, and ΔG^0 is the corresponding change in Gibbs free energy. Corrections for nonstandard-state operation can be readily applied by use of Eq. 5, giving the electrochemist a yardstick against which to gauge his performance. For Eq. 7, ΔG^0 is -145 kilocalories, leading to a theoretical reversible potential of 1.57 volts for the cell reaction (9).

Corrections for the fact that the reactants and products are not at unit activity in practical fuel cells are not of major importance since these corrections occur in a logarithmic term of Eq. 5; for example, a shift in the oxygen partial pressure from 1 atmosphere (roughly unit activity) to 0.1 atmosphere (considerably less than the partial pressure of O_2 at which air is exhausted from practical N_2H_4–air systems) introduces only a 33-millivolt correction to the "standard" voltage.

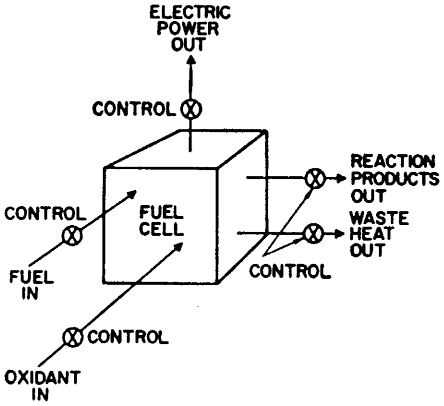

Fig. 1. Schematic diagram of a "black box" fuel-cell system.

While the Gibbs free energy, ΔG, measures the maximum potential that the electrochemist can achieve, the enthalpy change, ΔH, is of more interest to the engineer who must design heat exchangers and thermal controls for the hydrazine-air fuel-cell system. Enthalpy and free energy are related through

$$\Delta G = \Delta H - T\Delta S \qquad (9)$$

Since the entropy change ΔS is positive in Eq. 7, the free energy available for electrochemical conversion is less by an amount $T\Delta S$ than the total heat content of the fuel; for Eq. 7 this corresponds to 3 kilocalories of a total ΔH of -148 kilocalories. The value of the real enthalpy change, ΔH, depends on the initial and final states of the system—in particular on whether water leaves the system as liquid or as vapor. In normal operation of a hydrazine-air system, the by-produced water is swept away as vapor into the ambient air. For examination of the true heat balance in the system, Eq. 7 can be written in this more detailed form:

$$\begin{aligned} &4O_2\ (g,\ 0.2\ \text{atm};\ 20°\text{C}) + \\ &\qquad 16\ N_2\ (g,\ 0.8\ \text{atm};\ 20°\text{C}) + \\ &N_2H_4 \cdot H_2O\ (l,\ 20°\text{C}) \rightarrow \\ &\qquad 17N_2\ (g,\ 0.66\ \text{atm};\ 60°\text{C}) + \\ &3H_2O\ (g,\ 0.20\ \text{atm};\ 60°\text{C}) + \\ &\qquad 3O_2\ (g,\ 0.14\ \text{atm};\ 60°\text{C}) \qquad (10) \end{aligned}$$

This equation takes into account the fact that the amount of air fed into the "black box" (Fig. 1) as a reactant must be well above the stoichiometric requirement, set by Eq. 7, to avoid "gas starvation" of the cathode. In a hydrazine-air fuel-cell system the useful oxidant, oxygen, is diluted with inert nitrogen; an attempt to utilize 100 percent of the oxygen in the air would result in great losses by polarization of the cathode. The buildup of nitrogen within the pores of the electrode structure would serve as an effective barrier against diffusion by the remaining traces of electrochemically active oxygen.

Equation 10 assumes, rather arbitrarily, a fourfold excessive flow of oxygen (air). Four moles of oxygen are supplied; one is consumed electrochemically, and three are rejected in the exhaust gas. A four-times-greater amount of nitrogen flows through the system, unchanged chemically but

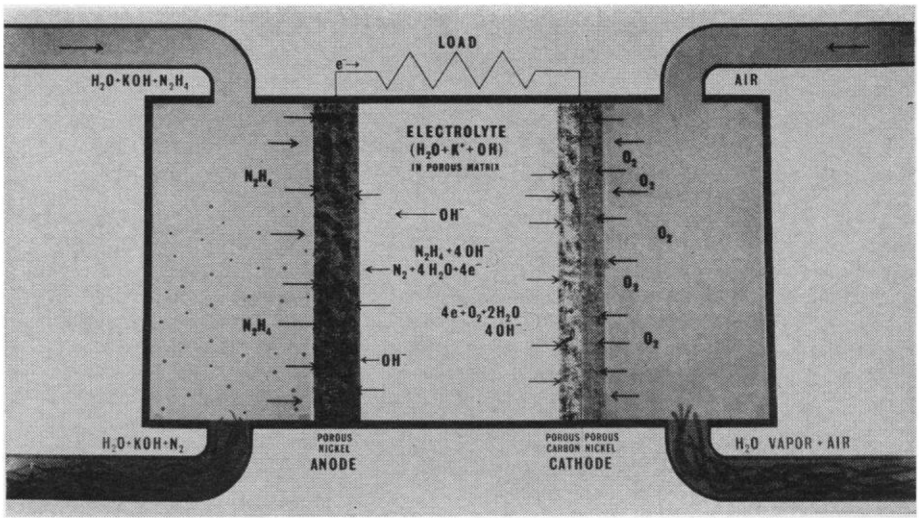

Fig. 2. Hydrazine-air fuel cell; principle of operation.

heated from ambient temperature to the temperature of the fuel-cell exhaust, an important factor in the overall heat balance.

Equation 10 is also more realistic than Eq. 7 in recognizing that the normally used fuel is not hydrazine but hydrazine monohydrate containing about 64 percent hydrazine. This practice introduces additional water, beyond electrochemical production, to be rejected (36 percent of the "fuel") if steady-state operation is to be maintained; it also lowers the energy content of the fuel (corresponding to the heat of hydration of hydrazine) by about 1.8 kilocalories per mole.

The choice of an air flow delivering four times the stoichiometric requirement of oxygen is based not on any law of nature but on engineering expediency. Adequate oxygen can be supplied at twice the stoichiometric rate for systems having minimal overload requirements, while eight to ten times the stoichiometric rate of oxygen flow may be required in a system that may be called upon for maximum power density.

With proper design of electrodes and current collector, most of the energy released can be delivered to the external load to perform useful work. The ratio of the useful electrical work to the (lower) heat value of the fuel is known as the thermal efficiency; in general it is a considerably smaller number than the electrochemical efficiency previously defined. If one subtracts the useful electrical work, delivered to the external load, from the total energy change, the residue represents waste heat that must be rejected. In small systems of low power density, simple thermal convection to the atmosphere may adequately control temperature, but, in any system delivering more than a few hundred watts at high power density, auxiliary equipment is required for heat rejection. Typically, a finned-tube heat exchanger, operating with forced-air cooling, rejects heat from a circulating anolyte.

For our analysis so far we have assumed that the electrochemical oxidation of hydrazine follows the theoretical course set by Eq. 7, or, at worst, the "practical" Eq. 10. In fact, hydrazine can enter into "noncoulombic" reactions or "side reactions" resulting in loss of fuel without generation of useful electricity. We shall return to this problem after discussion of the electrode reactions in more detail.

The Hydrazine Anode

It is interesting to note that, in practical operation of hydrazine-air fuel cells in the laboratory, the "open circuit" or no-load potentials realizable in the hydrazine-air system are closer to 1.0 volt than to 1.57 volts. In the light of experimental cell tests, this fact suggests to us and others (*10*) that Eq. 2 is oversimplified; an alternate reaction mechanism comprises

$$N_2H_4 \left(\xrightarrow{\text{catalyst}}\right) N_2 + 4H\ (\text{adsorbed}) \qquad (11)$$

$$4H\ (\text{adsorbed}) + 4OH^- \rightarrow 4H_2O + 4e^- \qquad (12)$$

in which the hydrazine is decomposed to waste nitrogen and adsorbed hydrogen atoms (the real electrochemical "fuel") by way of catalytic decomposition reactions on the anode catalyst. Since the decomposition of hydrazine to nitrogen plus hydrogen is exothermic, it reduces the free energy available to the electrochemist to −57 kilocalories per mole of hydrogen, or an equivalent theoretical voltage of 1.23 volts. This fact suggests that, from an electrochemical viewpoint, hydrazine fuel is simply a practical way to transport hydrogen in a low-volatility logistic package.

We do not have to choose between the "direct" electrochemical oxidation path of Eq. 2 and the "indirect" path of Eqs. 11 and 12; the potential of a practical electrode shows that both reaction paths exist in competition. Perry (*11*) has reported good voltage and fair stability in iron-catalyzed hydrazine anodes; it is interesting that iron was the anode catalyst employed by A. Ray in operating the world's first hydrazine fuel cell at the Union Carbide Research Laboratory in Cleveland, Ohio, in 1931. The fact that an iron-catalyzed anode operates in a hydrazine fuel cell is significant, since iron is relatively a poor catalyst for the oxidation of adsorbed atomic hydrogen in accordance with Eq. 12. On the other hand, most modern hydrazine fuel cells utilize catalysts of precious metals, especially palladium as the anode catalyst. Palladium is one of the best-known catalysts for favoring the indirect reaction route of Eqs. 11 and 12.

Selection of an optimum hydrazine anode entails more than finding a catalyst to expedite the reaction(s) of either Eq. 2 or Eqs. 11 and 12. Decomposition of hydrazine to form nitrogen plus ammonia (Eq. 13) can occur spontaneously (*12*):

$$3N_2H_4 \rightarrow 4NH_3 + N_2 \qquad (13)$$

the kinetics and detailed mechanism of reaction leading to the net result are not known. It is known that decomposition to form ammonia is accelerated as the temperature increases. At operating temperatures above about 65° to 70°C, formation of ammonia ceases to be a scientific curiosity and becomes a serious engineering problem; in addition to destroying fuel and reducing coulombic efficiency, it makes conditions unpleasant downwind from any sizable source of hydrazine-air power. An equally important factor is the concentration of hydrazine in the circulating anolyte; if the concentration is kept just barely above the minimum required to sustain the anode reaction rate (at some level of output of current), no ammonia is generated (as measured by a simple but very effective odorometric test). Holding the hydrazine concentration at this level presents very interesting problems in control dynamics that we shall discuss later.

Another side reaction competing with the electrochemical reactions in Eq. 2, or in Eqs. 11 and 12, is the exothermic decomposition of hydrazine to its elements:

$$N_2H_4 \rightarrow N_2 + 2H_2\ (\text{gas}) \qquad (14)$$

If it occurs at the anode, this reaction does not reduce the coulombic efficiency, since any traces of hydrogen produced on the anode surface are promptly consumed through the reaction

$$H_2 + 2OH^- \rightarrow 2H_2O + 2e^- \qquad (15)$$

at the anode. Since most modern hydrazine-air fuel cells employ catalysts, such as of palladium, that are highly effective in the anodic oxidation of hydrogen, traces of decomposition, in accordance with Eq. 14, occurring at the anode should not impair the efficiency of fuel utilization. However, the combined effects of the reactions of Eqs. 13 and 14 place certain restraints on the designer of the system auxiliaries: the concentration of hydrazine in the anolyte must be held down to the minimum value permitting adequate output of current, the temperature must be kept low, and components in the fuel-cell system must be carefully studied in order to avoid the fortuitous introduction (as in a metal heat exchanger) of a surface that promotes the reaction of Eq. 13 or 14.

A porous matrix is placed between the anode and the cathode (Fig. 2), the primary purpose being to restrict the diffusion of hydrazine from the vicinity of the anode to the cathode. High concentrations of hydrazine at the air cathode can result in loss of fuel by direct catalytic oxidation in accordance with Eq. 7, and even small amounts of hydrazine in the cathode region can lower the cathode potential. By careful attention to these principles, coulombic efficiencies greater than 95 percent can be achieved in laboratory cells—that is, the number of electrons entering the load circuit can be more than 95 percent of the theoretical number computed from the electrode reaction mechanisms. However, the engineer who designs systems must be a practical man, familiar with the principles of compromises or "trade-offs." Operation at very low levels of hydrazine in the anolyte reduces anode potential and thus output voltage; operation at low temperature increases the internal resistance of the electrolyte and thus reduces the useful output of power. Faced with the practical necessity of forcing a maximum power output from a system of minimum weight and volume, in today's state of the art the engineer can achieve 75- to 95-percent coulombic efficiency at the hydrazine-cell anode.

The existence of appreciable side reactions compels reappraisal of the previous thermodynamic calculations of fuel economy. Estimates of the rate of utilization of fuel can be derived very simply by multiplying the coulombic efficiency by the thermal efficiency (previously defined). Fuel may be lost through decomposition to ammonia (Eq. 13) or hydrogen (Eq. 14), or by simple evaporation of hydrazine fuel itself to the atmosphere (Eq. 16; usually negligible in a well-designed system):

$$N_2H_4 \cdot H_2O(\ell) \rightarrow N_2H_4(g) + H_2O(g) \quad (16)$$

When one estimates fuel requirements, fuel losses due to the reactions of Eqs. 13, 14, and 16 may be combined under an overall coulombic efficiency. The heat-transfer engineer, charged with responsibility for maintaining thermal balance, must employ far more refined analysis. Loss of fuel by evaporation from the anolyte (Eq. 16) does not increase the heat load on the system, since the heat energy of this portion of the fuel has been removed from the system envelope. If anything, there is a tiny loss in heat, corresponding to the heat of vaporization of the hydrazine. On the other hand, loss of fuel by way of Eqs. 13 and 14 can have significant effects on the overall thermal balance, making it quite important for the system engineer to know what portion of the fuel lost in noncoulombic processes has entered which reaction path.

Decomposition of hydrazine to ammonia (Eq. 13) generates heat at 27 kilocalories per mole, while decomposition to hydrogen (Eq. 14) generates heat at 12 kilocalories per mole. In the latter instance the fuel value of the hydrogen is not normally lost, since the typical hydrazine anode is also an effective hydrogen-consuming anode.

Performance Characteristics

The output voltage of hydrazine-air cells is mainly determined by the characteristics of the air electrode and by the concentration of hydrazine in the electrolyte. Figure 3 (top) shows the polarization curves of oxygen and air cathodes as measured against a hydrogen reference electrode; the general shape is determined by the gas-diffusion properties of the electrodes. Figure 3 (bottom) shows the voltage of the hydrazine-air cell as a function of the concentration of N_2H_4 in the electrolyte.

Auxiliary Components

We can now discuss the auxiliary components required to keep the hydrazine-air fuel-cell system in steady-state operation (Fig. 4). Some provision must be made in the system for feeding the liquid fuel at a rate commensurate with the load demand. One early approach was to feed hydrazine fuel through a metering orifice providing a uniform rate of feed; such a device is adequate for a system that always operates at a fixed level of power, but supplies too much fuel for a partial load. Another approach is to supply fuel in direct proportion to the current, since by Faraday's law each mole of hydrazine can produce 107 ampere-hours of electricity; thus there is a direct relation between amperes and moles of fuel per hour. Such a simple coulometric control ignores the complications introduced by the noncoulombic reactions of Eqs. 13 and 14. Since the rates of the self-decomposition reactions depend on temperature, concentration of electrolyte, type of electrode, age of the system, and so on, such coulometric controls require frequent adjustment for proper operation. A third alternative is to use the voltage of the system as a measure of the concentration of hydrazine; increase in the level of hydrazine increases the anode voltage (Fig. 3), providing a voltage signal that can be used to control the rate of fuel feed. The voltage of the fuel cell depends not only on concentration of hydrazine but also on the temperature and output current; compensation for these latter variables must be provided in a fully automatic system of control. Controls of this type were provided in the 300-watt hydrazine-air system shown in Fig. 5.

The most modern and sophisticated type of fuel-feed control is based on use of a hydrazine sensor that analyzes the anolyte for concentration of hydrazine and adjusts this concentration

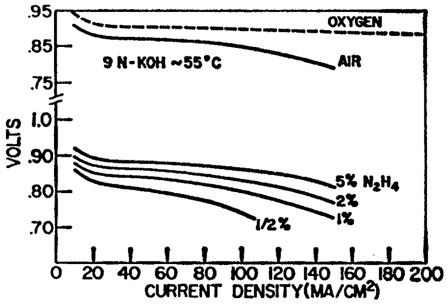

Fig. 3. Potentials of a N_2H_4–air cell at different concentrations of fuel.

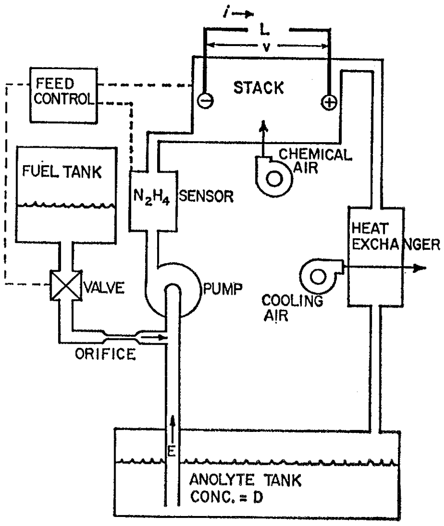

Fig. 4. Hydrazine-air system, schematic.

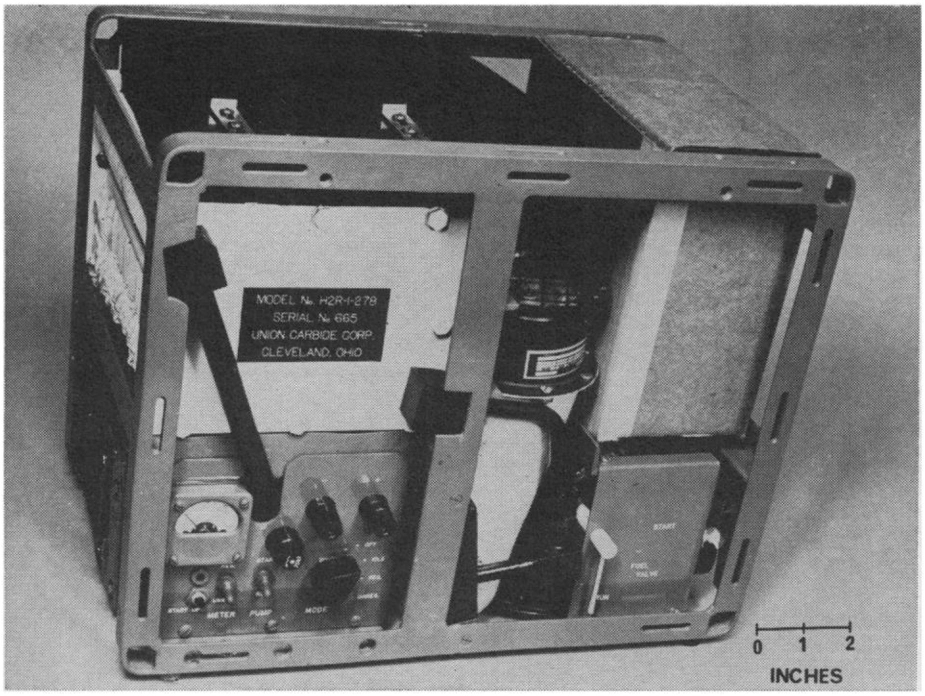

Fig. 5. Hydrazine-air fuel-cell system yielding 300 watts.

to an optimum level depending upon the temperature and output of current. This type of control was used in the hydrazine-air fuel cell powering a motorcycle (*13*). Design of a fuel-metering device for a low-powered fuel cell is made more difficult by the need to provide for very low rates of fuel consumption: a 100-watt hydrazine-air cell consumes only about 1 milliliter per minute, and reproducible control of such a tiny trickle is difficult in a portable device subjected to rough handling and frequent inclination from the horizontal.

"Chemical air," supplying the oxygen required for the cathode reaction, is supplied by a small fan. A level sensor in the anolyte tank senses the volume of anolyte; a signal from it adjusts the fan speed to maintain a constant volume of anolyte and so ensure a proper rate of removal of by-produced water. Waste heat is rejected by a liquid-to-air heat exchanger using forced-air cooling; a thermistor bridge circuit senses the temperature of the system, and a proportional-control circuit adjusts the speed of the fan of the heat exchanger to hold the temperature constant.

The anolyte tank serves as an expansion tank to accommodate small fluctuations in volume of anolyte, as a settling basin to permit separation of nitrogen bubbles from the anolyte, and as a sump to feed the suction of the anolyte pump. For proper pumping of the somewhat-frothy anolyte, a positive-displacement pump is preferred.

The heater is an optional accessory for rapid starting during cold weather. During the process of starting, the entire electrical output of the fuel cell is fed back to internal heaters. The effective resistance of the heating circuit is controlled by a power-transistor circuit with both stack-voltage and temperature-sensor inputs for control. So long as the temperature of the system is below normal, the system operates at a constant voltage corresponding to the minimum voltage required to operate the auxiliaries and controls; this arrangement automatically provides the maximum rate of self-heating of which the system is capable. As the system warms to operating temperature, control is transferred to a temperature controller which reduces the rate of generation of internal heat to a minimum level matching the rate of heat loss to the environment.

This description of auxiliaries and controls applies to a fully automatic system designed for unattended operation. Many of the control functions could of course be transferred to a human operator, with concomitant simplification in electronic circuits. At very low levels of power (below about 100 watts), one may dispense with some of the auxiliary components, and natural convection can replace forced circulation. In any case, however, the steady state (Fig. 1) must be maintained if the system is to operate as a fuel cell.

Summary

A fuel cell is an electrochemical device that continues to generate electrical power as long as reactants are supplied and products are removed at properly controlled rates. An assembly of cells is required within which the conversion of chemical to electrical energy occurs; also required is a set of auxiliary components to supply the reactants and remove the products (including waste heat) under controlled steady-state conditions. In addition to the desired energy-conversion reactions, there are deleterious side reactions that can impair fuel economy. From knowledge of these factors influencing the possible reactions, and guided by principles of elementary chemical thermodynamics, the electrochemist can select optimum conditions for cell performance. It is then the job of the engineer to design auxiliary components and controlling devices to provide the electrochemical cells with the best possible approach to these optimum conditions.

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