

# MEASUREMENT OF EXCESS HEAT AND APPARENT COINCIDENT INCREASES IN THE NEUTRON AND GAMMA-RAY COUNT RATES DURING THE ELECTROLYSIS OF HEAVY WATER

CHARLES D. SCOTT, JOHN E. MROCHEK,  
TIMOTHY C. SCOTT, GORDON E. MICHAELS,  
EUGENE NEWMAN, and MILICA PETEK  
*Oak Ridge National Laboratory, Chemical Technology Division  
Oak Ridge, Tennessee 37831-6226*

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*Excess heat and apparent increases in the neutron and gamma-ray count rates have been observed in a series of tests performed at Oak Ridge National Laboratory to study the electrolysis of heavy water in the presence of palladium cathodes. For these tests, LiOD at a concentration of 0.1 to 1 N in D<sub>2</sub>O was used in an insulated glass electrochemical cell in which the temperature was controlled and heat was removed by flowing water in a cooling jacket. Results of two of the tests, one of which lasted for over 1900 h, are reported. In the latter test, an internal D<sub>2</sub>-O<sub>2</sub> recombiner was incorporated into the cell to give a closed system without off-gas.*

*Excess power, usually in the range of 5 to 10%, was detected for periods of many hours. Some of these events were initiated and could be extended by system perturbations. On three separate occasions, the mean neutron count rate exceeded the background by statistically significant values; one of these was apparently coincident with an extended period of excess heat generation. Increases in the gamma-ray count rates were apparently also coincident with two of the periods of excess neutrons.*

## INTRODUCTION

Several research groups have reported the measurement of excess energy during the electrolysis of heavy water containing LiOD as the electrolyte and in the presence of a palladium cathode.<sup>1-5</sup> In some cases, anomalous neutron count rates have been reported for such systems,<sup>5-8</sup> and increases of the tritium content of the electrolyte solutions have also been measured.<sup>9</sup> Researchers at Oak Ridge National Laboratory (ORNL) have carried out a series of electrochemical tests with the emphasis on careful measurements of as many parameters as possible. These included the simultaneous and continuous monitoring of both the heat balance and the neutron and

COLD FUSION

TECHNICAL NOTE

**KEYWORDS:** *excess heat, cold fusion, closed system, internal recombiner*

gamma-ray count rates. Flow calorimetry was used in which flowing water in an insulated jacket around the electrolysis cell served to control electrolyte temperature and remove heat. In one extended test of over 1900 h, an internal catalytic system was used to recombine the electrolytically generated D<sub>2</sub> and O<sub>2</sub>, resulting in a closed system without the need for release of any off-gas.

Excess heat and apparent increases in the neutron and gamma-ray count rates were observed.

## MATERIALS AND METHODS

Although there has been almost a continuous evolution of the design of the electrochemical system used in these tests, two primary electrolysis cell designs were used. The first was an open system in which the electrolysis gases, D<sub>2</sub> and O<sub>2</sub>, were allowed to continuously exit the cell. The second concept utilized an internal recombiner that catalyzed the recombination of D<sub>2</sub> and O<sub>2</sub> to form heavy water, which then returned to the electrolyte solution. The latter was a closed system that did not require replenishment of D<sub>2</sub>O except when samples were withdrawn.

### Open Electrolysis Cell

The open electrochemical cell that did not recombine the electrolysis gases was fabricated from Pyrex glass with a nominal inside diameter of 4 cm and an active internal height of 12.5 cm (Fig. 1). It had an ~4-cm-high internal gas space into which a N<sub>2</sub> purge gas could be introduced and removed. A 2.5-cm-thick Teflon cap was attached to a No. 40 Teflon O-ring connector and secured to the cell body by a clamp and a rubber O-ring. The cap accommodated the entrance of electrodes, a glass tube enclosing a thermocouple for measurement of electrolyte temperatures, a glass-encased resistor for internal heat calibration, and a polyethylene tube for introduction of makeup D<sub>2</sub>O and removal of electrolyte samples. The cell was surrounded by an enclosed glass annulus with ~1 cm of open space through which water was forced to flow

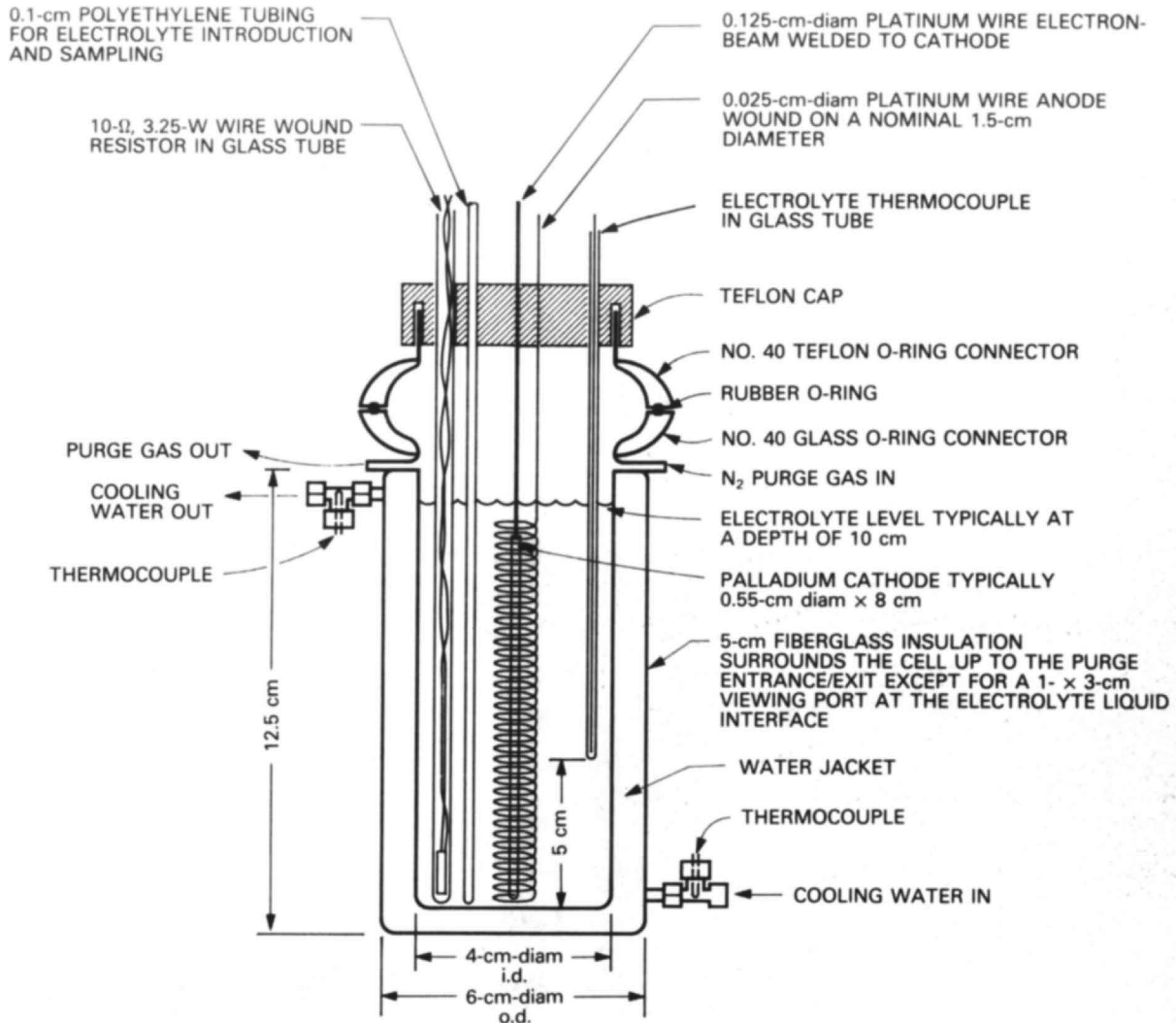


Fig. 1. Open-system cell with cooling-water jacket used for electrolysis tests.

by a metering pump. Thermocouples inserted in the cooling water inlet and outlet streams allowed the determination of temperature difference and, hence, heat flow. Fiberglass insulation that was 5 cm thick covered the entire exterior of the cell and top except for a 1- x 3-cm area that could be opened to observe the electrolyte level.

**Closed Electrolysis Cell**

The design concept for the closed cell was similar to that described above, except that the cell body was fabricated from nominal 1.5-in. Pyrex glass pipe, and the 1.25-cm-thick Teflon top was sealed with a conventional glass pipe flange (Fig. 2). As in the open system, a 1-cm annular water jacket was used for heat removal, and 5 cm of foam insulation surrounded by an additional 5 cm of fiberglass insulation was used to reduce unmeasurable heat loss. Typically, a 10-cm height of electrolyte solution was used, which resulted in a 9-cm height of gas space.

Total recombination of the evolved D<sub>2</sub> and O<sub>2</sub> was carried out within the gas space of the enclosed electrolysis cell. The catalytic recombiner was a 375-cm coil of 32-gauge platinum wire that had been electrochemically coated with ~10 wt%

palladium black (see Fig. 3). The wire was wrapped around six Teflon-sheathed screws that were attached to the top flange and extended down into the gas space. Teflon tubing, connected through the flange to the gas space by conventional tubing compression fittings, exited through a heavy water bubbler so that any formation of off-gas could be detected. Similar gastight fittings were used for electrode connections and entrance of a Teflon electrolyte charging tube and a thermocouple sheathed in stainless steel.

Operation of the recombiner was initiated at the beginning of an experiment by imposing a 0.7-A (2.4-V) electrical current over a period of 2 to 4 h, after which no detectable off-gas from the system was observed. Following initiation, the recombiner electrical current was required only if the cell current had been stopped for an extended period and was then restarted.

The cooling jacket did not extend over the entire outside surface of the cell because space was needed for the glass pipe flanges. Thus, the recombiner section operated at a relatively high temperature as compared with the electrolyte. This resulted in heat loss at the top of the cell in spite of the 4 in. of insulation. A future design change will rectify this problem.

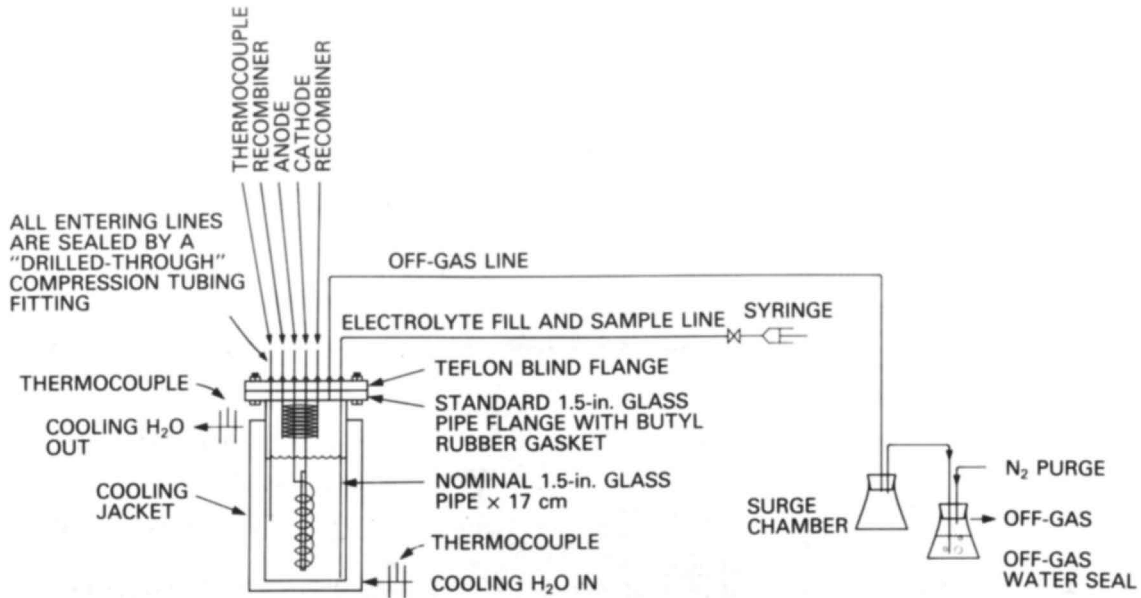


Fig. 2. Electrolysis cell and auxiliary equipment used for closed-system tests. The entire system is insulated with 4 in. of fiberglass insulation.

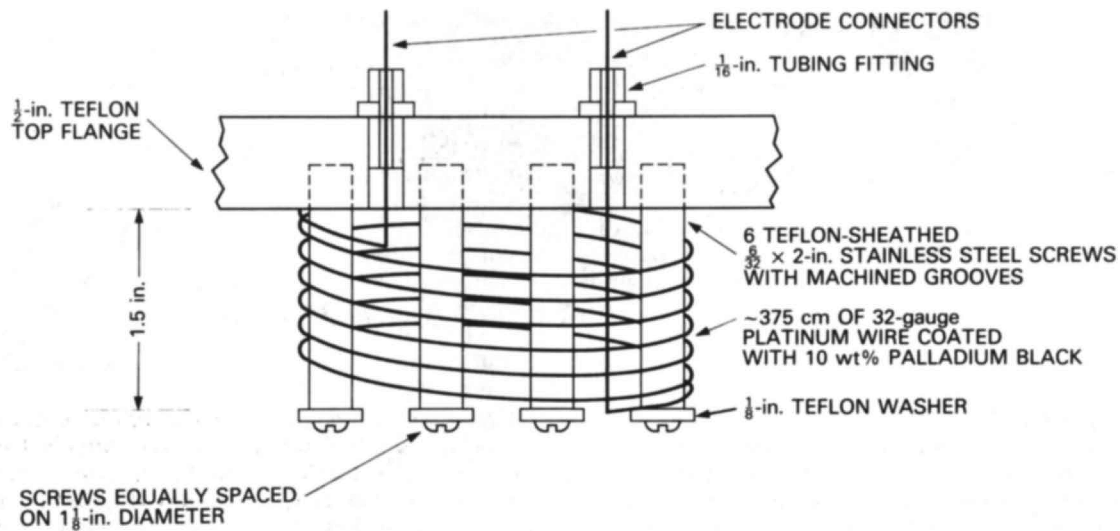


Fig. 3. Configuration of the recombining used in the closed-system electrolysis cell.

**System Design**

The cooling water was supplied by a water reservoir with the temperature controlled to within  $\pm 0.1^\circ\text{C}$ ,<sup>a</sup> along with a positive-displacement pump<sup>b</sup> that controlled the coolant flow rate with an accuracy of 0.5 to 1% (Fig. 4). Nitrogen purge gas, when used, was monitored by a rotameter, while D<sub>2</sub>O was added and electrolyte was sampled by a syringe

pump connected to a Teflon tube that entered through the flange and extended to the bottom of the cell.

The electrical power<sup>c</sup> supply operated at a constant current that was measured by a Keithley 171 microvolt DMM meter<sup>d</sup> with an accuracy of  $\pm 1$  mA. Electrolyte and coolant water inlet and outlet temperatures were measured by calibrated thermocouples (temperature differences were generally in the range of 2 to 5°C), and the overall electrode voltage was determined to within 0.001 V by a Keithley 181

<sup>a</sup>Polystat Model 1194-00, Cole Parmer Instrument Company, Chicago, Illinois.

<sup>b</sup>Duplex Pump Model RP2, Penn Systems, Inc., Broomall, Pennsylvania.

<sup>c</sup>Princeton Applied Research, Model 371, Potentiostat/Galvanostat.

<sup>d</sup>Keithley Company, Cleveland, Ohio.

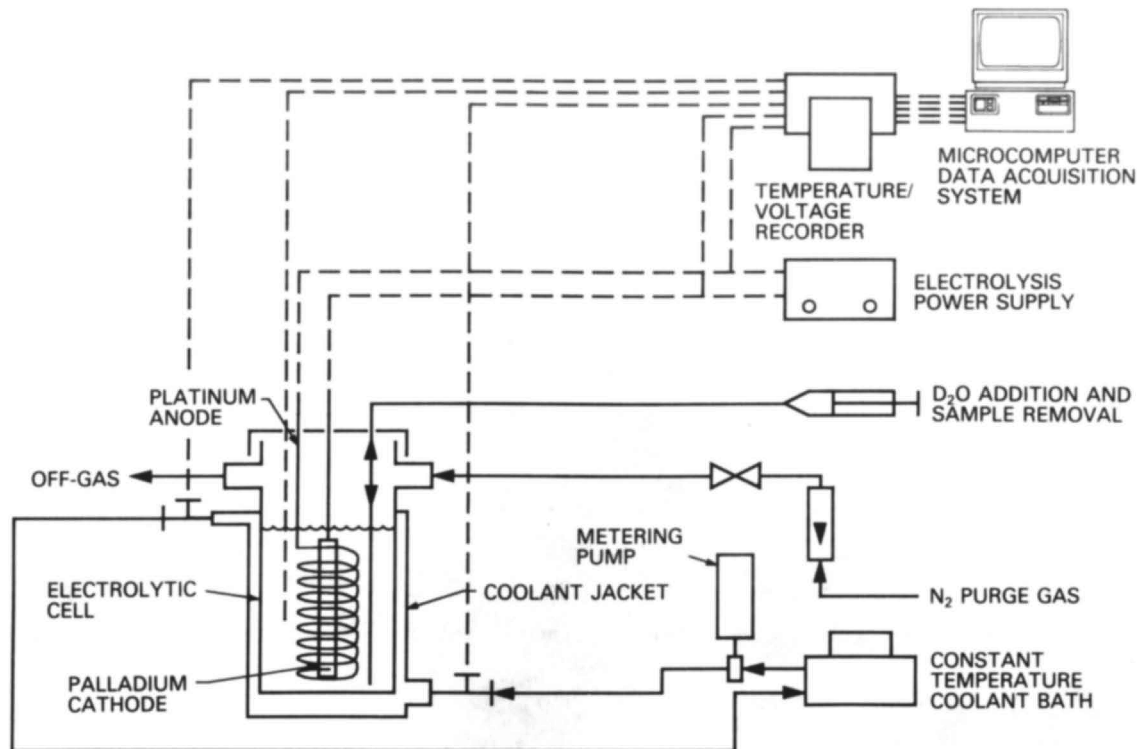


Fig. 4. Integrated system components used in the electrolysis tests.

nanovoltmeter.<sup>d</sup> These system parameters, along with all measured temperatures, were recorded on a strip-chart recorder typically every 10 s, and average values based on ten data points were recorded on a microcomputer data acquisition system every minute. Calibration of the thermocouples, including curve fitting by fourth-order polynomials, allowed temperature measurements to be made to within 0.05 to 0.1°C. The data acquisition system also calculated a heat balance at 1-min intervals with an estimated experimental error calculated to be typically 0.2 W.

#### Radiation Detection

The experimental apparatus was contained within a 2-ft-thick concrete enclosure in an attempt to reduce the neutron background. Neutron levels were measured by a 2.5-cm-diam  $\times$  2.5-cm NE-213 scintillator<sup>e</sup> placed immediately adjacent to and at the midpoint of the insulated electrolysis cell (Fig. 5). Necessary electronics were used to allow pulse-shape discrimination with a neutron threshold of  $\sim 1.2$  MeV. This arrangement also allowed the measurement of gross gamma rays with an energy level greater than  $\sim 0.3$  MeV. The NE-213 scintillator is sensitive to both neutrons and gamma rays; during calibration, it was determined that there was a gamma-ray contribution to the neutron peak of  $\sim 4.1\%$  of the gamma peak, or  $\sim 20\%$  of the neutron peak. All reported neutron count rates were corrected for this contribution. The overall counting efficiency was  $1.46 \times 10^{-3}$  as determined by a  $^{252}\text{Cf}$  source, and the neutron detector had a typical sensi-

tivity at three standard deviations equivalent to  $3 \times 10^{-24}$  fusion/deuterium pair per second.

A separate gamma-ray spectrometer utilizing a 7.6-cm-diam  $\times$  7.6-cm NaI detector<sup>f</sup> was also used. The detection head was placed immediately next to the midpoint of the insulated cell and at an orientation  $\sim 130$  deg to the NE-213 detector. Although there was no efficient provision for interacting possible neutrons with protons in order to detect the resulting gamma rays, the 1 cm of water in the cooling jacket of the cell should result in some indication. A multichannel analyzer that covered an energy range of 0.14 to 5.20 MeV was used, and the system had an overall detection efficiency of  $5.75 \times 10^{-5}$  as determined by a  $^{252}\text{Cf}$  source with a polyethylene converter. The NaI system had somewhat less sensitivity than the neutron detector to possible fusion reactions because of the higher gamma-ray background. The neutron and gamma-ray spectra were periodically recorded on magnetic media by a small computer system.

The tritium concentration in the electrolyte was measured at definite intervals by removing a small volume of the electrolyte and counting with a liquid scintillation system. The accuracy of each measurement was  $\pm 200$  Bq/l.

#### Materials

The heavy water used in the experiments was deuterium oxide  $\text{D}_2\text{O}$  (99.9 at. % deuterium) with a low tritium content,  $\sim 2000$  Bq/l.<sup>g</sup> The electrolyte was prepared by dissolving

<sup>d</sup>Nuclear Enterprises, Inc.

<sup>f</sup>Harshaw Chemical Company, Solon, Ohio.

<sup>g</sup>Aldrich Chemical Company, Inc., Milwaukee, Wisconsin.



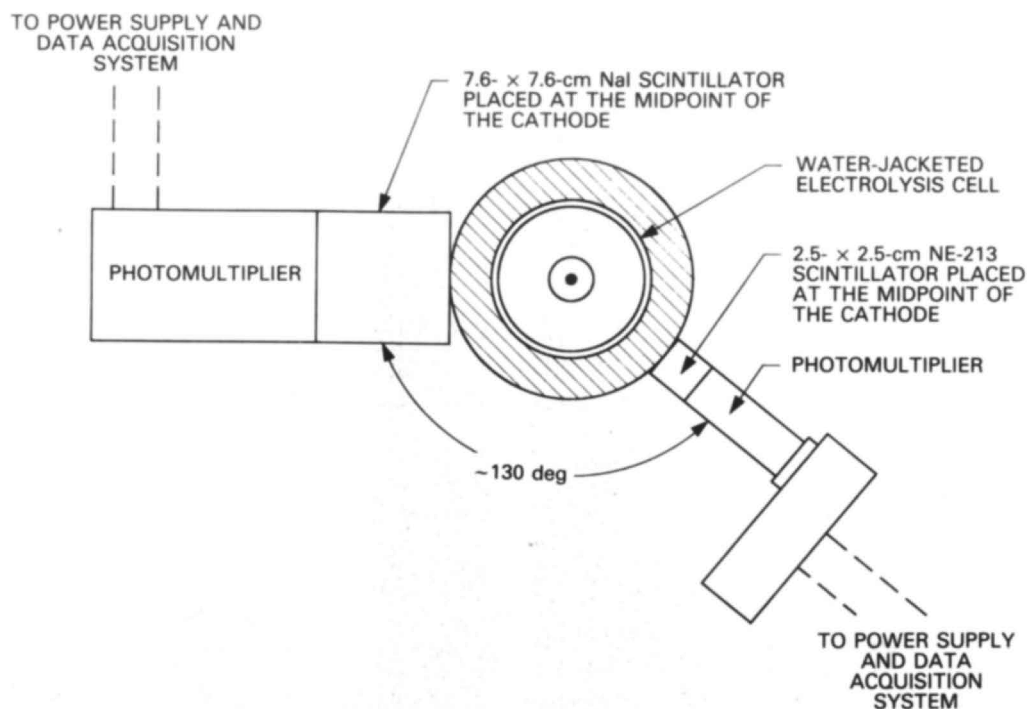


Fig. 5. Placement of scintillation detecting systems used for detection of neutrons and gamma rays.

reagent-grade natural lithium in the  $D_2O$  at a nominal concentration of 0.1 to 1.0  $N$ .

The cathode material was 99.9% palladium<sup>b</sup> that had been cast under argon and then swaged to the desired diameter. This cylindrical material was cut to the desired length; a 0.13-cm platinum connecting wire was electron beam welded to the top; and the metal assembly was then annealed at 900°C for 2 h in vacuum. All of the anode material and the recombiner were fabricated from 99.9+ % platinum wire in the size range of 24 to 32 gauge.<sup>1</sup> The anodes consisted of wire coils made by wrapping the wire around the exterior of a skeletal glass mandrel (four 0.15-cm-diam glass rods with cross bracing) that surrounded the cathode and provided an electrode spacing of 0.3 to 0.5 cm.

#### Operating Procedures

The tests were initiated by loading the prepared electrolytic cell with ~125 ml of the electrolyte, starting the purge gas at ~1 ml/s for the open cell, and turning on the electrical current. A purge gas was not used with the closed system, and  $D_2-O_2$  recombination was initiated by electrically heating the wire coil prior to the accumulation of a large amount of electrolysis gas in the headspace. This initiation heat was not required after the catalytic reaction started, at least in the current range of 0.71 to 4.26 A.

The temperature of the electrolyte was controlled by the cooling water temperature and flow rate and was usually maintained in the range of 28 to 38°C; however, some controlled excursions were imposed up to 70°C and down to 24°C. Makeup  $D_2O$  was added to the electrolyte either continuously with a syringe pump or batchwise every 8 h to maintain a

constant electrolyte inventory for the open-system test, and electrolyte samples were taken periodically (every 2 to 3 days). The gamma-ray and neutron count rates were measured continuously and recorded every 4 h.

#### Energy Balance

An energy balance expressed as power out versus power in was determined for each test, based on the following assumptions:

1. The electric current is 100% efficient for the electrolysis of  $D_2O$ .
2. The system operates at quasi-steady state with a constant inventory of deuterium in the cathode.
3. Except where internal recombination is used, all of the  $D_2$  and  $O_2$  exit the electrolysis cell without recombination.
4. The exiting gases, including a purge gas (if used), are saturated with  $D_2O$  that is at equilibrium with heavy water at the temperature of the electrolyte.
5. There is no heat loss to the ambient environment.

The second assumption is obviously not correct when a major portion of the formed  $D_2$  is being adsorbed by the cathode, but this is true for only a short period in the early phase of the test. Since the volume of makeup heavy water required to be added to maintain a constant inventory in the electrolysis cell was approximately equal to the volume electrolyzed and evaporated for the open systems, it was assumed that very little recombination of the electrolysis gases occurred. Conversely, no addition of  $D_2O$  or electrolyte was required for the closed systems with recombination except to replace electrolyte samples that were withdrawn. Although

<sup>b</sup>Materials Research Corporation, Orangeburg, New Jersey.

<sup>1</sup>Engelhard Corporation, Iselin, New Jersey.

the system was well insulated, some heat loss obviously occurred to the ambient, but this would result in a conservative estimate of the recovered heat. Necessary chemical and physical properties for D<sub>2</sub>O were obtained from a reference handbook.<sup>10</sup>

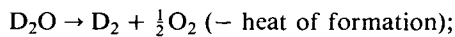
*Open System*

The resulting energy balance for the open systems can be represented by

POWER IN: (volts) × (amperes);

POWER OUT:

ELECTROLYSIS (typically >30%)



FORCED COOLING (typically >60%)

(cooling water temperature increase) × (flow rate);

LATENT HEAT (typically <2%)

(heat of D<sub>2</sub>O vaporization).

*Closed System*

The closed system with recombination allows a much simpler energy balance with fewer assumptions:

POWER IN:

(volts × amperes);

POWER OUT:

(cooling water temperature increase) × (flow rate).

**EXPERIMENTAL RESULTS**

As in the case of earlier tests at this laboratory,<sup>4,5</sup> excess power was detected during certain periods. In addition, an apparent coincidence of a higher neutron count rate and an increase in the gamma-ray count rate were observed in one test.

**Operating Parameters**

During the tests, conditions of quasi-steady state were often maintained for many hours at a time; however, significant changes in the operating parameters were made at various times (Figs. 6 and 7). These changes were carried out in an attempt to determine conditions that would initiate or maintain the generation of excess power and/or the products of nuclear interactions. Parameter changes included cathode current density, electrolyte (LiOD) concentration, and the electrolyte temperature that was brought about by a change in the cooling water flow rate and temperature.

The same type of systematic increase in the cathode current density was imposed during the closed-system test; however, there were also periods of current density cycling in which rather severe system instability was imposed (Fig. 7). These included the interval of 1005 to 1170.5 h in which the cathode current density was cycled between 100 and 400 mA/cm<sup>2</sup>

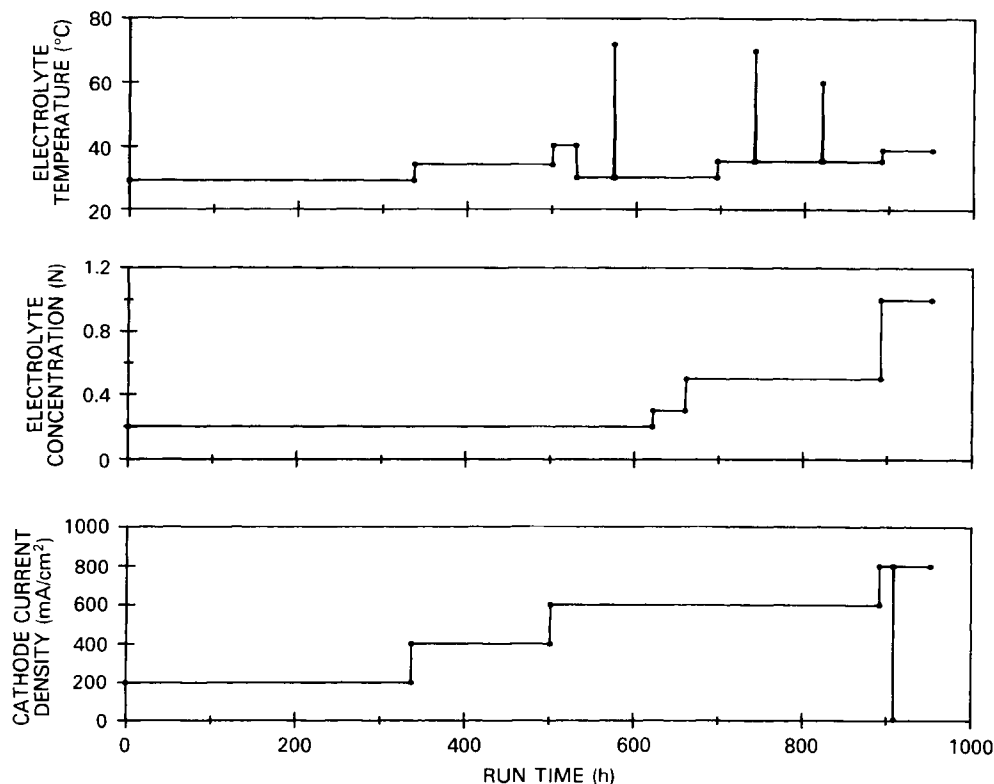


Fig. 6. Major variations of the controlled operating parameters (cathode current density, electrolyte concentration, and electrolyte temperature) during the open-system test.

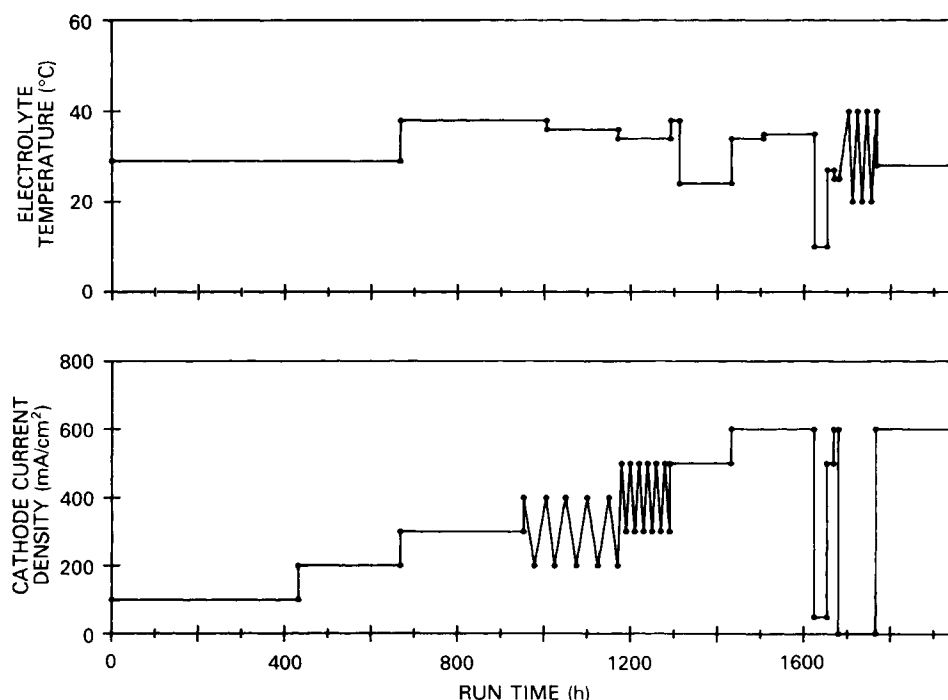


Fig. 7. Major variations of the controlled operating parameters (cathode current density and electrolyte concentration) during the closed-system test.

for a cycle period of 66 min; this was followed by a second interval of cycling between 300 and 500 mA/cm<sup>2</sup> with a period of 5.7 min.

There appeared to be a small, but progressive, decrease in the LiOD concentration during each test, apparently due to the deposition of this material on surfaces in the vapor space. Such deposits resulted in a slow increase in both the cell voltage and the power input. To accommodate this effect, the LiOD concentration was periodically checked and occasionally modified with makeup electrolyte when samples were taken. These changes were usually small, but they could result in measurable decreases in the cell voltage and power input.

#### Excess Energy

Several periods of apparent excess energy have been observed in tests in both the open and closed systems. Some of these were seemingly spontaneous when the system was operating at a quasi-steady state, while others of longer duration were initiated and maintained by system perturbations.

#### Open System

During the first 540 h of operation, there were successive increases in the cathode current density up to a level of 600 mA/cm<sup>2</sup>, but the power balance (rate of energy out minus rate of energy in) was essentially zero within the calculated experimental error. The calculated experimental error was predominantly due to the uncertainty in the temperature measurement ( $\pm 0.1^\circ\text{C}$ ) and the cooling water flow rate (1%).

At  $\sim 540$  h, heat in excess of that provided by inlet electrical power was apparently initiated by a decrease in the electrolyte temperature (Fig. 8). During the next 300 h, the power excesses ranged as high as 11% (well outside the experimental

error of  $\sim 3\%$ ); but, after a few hours, it tended to fade away. Increases in the electrolyte concentration or changes in the electrolyte temperature appeared to again enhance the energy excess. The electrolyte temperature variations seemed to be the most effective means for maintaining excess power with either a reduction in the temperature or rapid cycling of the temperature to 60 to 70°C and back to the original value. Ultimately, the energy balance became negative when the cathode current was increased to 800 mA/cm<sup>2</sup>. This was probably due to increased heat loss to the environment. A short cycle of the cathode current to zero and back to the original value also had no lasting effect.

#### Closed System

It became obvious during the closed-system test that there was a loss of heat to the environment, primarily through the top flange. This loss was apparently due to the relatively high temperature associated with the recombination and the lack of positive heat removal by the cooling water on the top flange. (The temperature of the outside of the top flange was usually in the range of 50 to 60°C.) Based on subsequent calibration tests, it was estimated that 1 to 3% of the generated heat was lost to the environment and was not recovered by the cooling water, even after additional insulation had been added to the system at  $\sim 150$  h into the test. This heat loss was not introduced into the heat balance calculations; therefore, the reported values of heat removal were very conservative. Although more precise experimental measurements were made in this test, the major contributors to the experimental error were still the measurement of temperatures with calibrated thermocouples ( $\pm 0.05^\circ\text{C}$ ) and the control of cooling water flow rate ( $\pm 0.5\%$ ).

The power balance was essentially even or somewhat negative during the first 740 h of operation (Fig. 9). Following

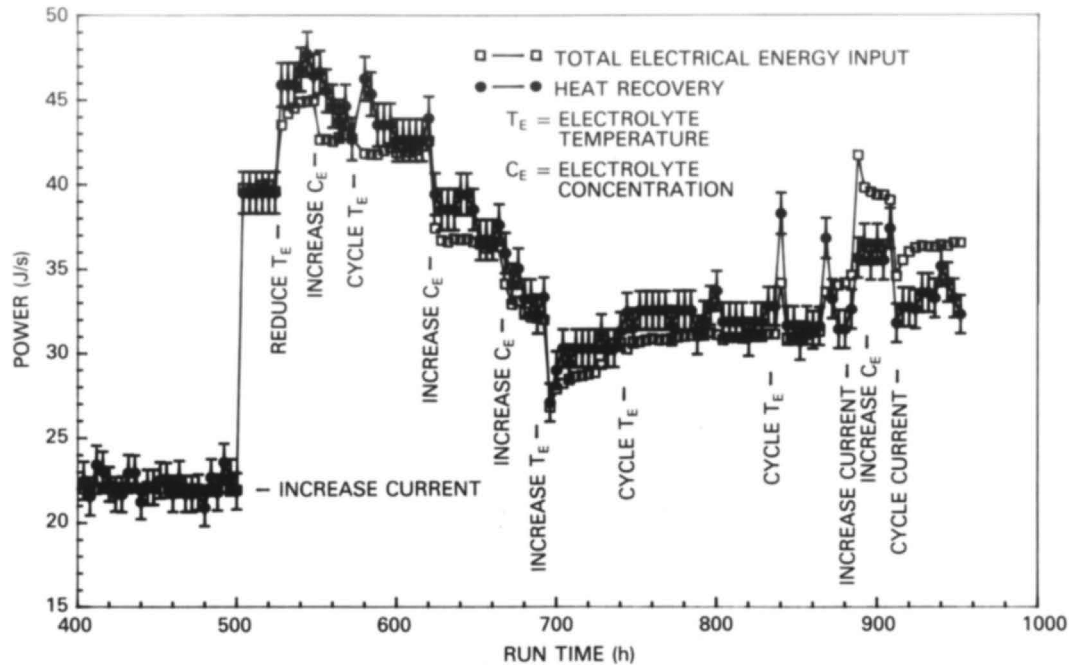


Fig. 8. The power balance during periods of excess power initiated by changes in operating parameters during the open-system test. Each point represents the mean value of 30 measurements during a 4-h interval, and the error bars represent calculated uncertainty in the experimental values based on independent calibration of temperature and flow metering systems.

that interval, there were two major periods of spontaneous excess power, each of which lasted for several hours (Fig. 10). These were relatively modest excesses with maximum values of  $\sim 6\%$  but well above the calculated experimental error of  $\sim 1$  to  $2\%$ .

Cycling of the cathode current appeared to have little effect on the generation of excess energy, but an extended period of excess power was initiated at  $\sim 1320$  h by a decrease in the electrolyte temperature from  $38$  to  $24^\circ\text{C}$  (Figs. 9 and 11). During this period, the power excess was generally in the range of  $5$  to  $10\%$ , with an estimated experimental error of  $1$  to  $2\%$ .

For this period of over  $300$  h of excess energy, the cathode current density was increased to  $600$  mA/cm<sup>2</sup> and, for a short time, was decreased to  $50$  mA/cm<sup>2</sup> with only limited effect. The electrical current was then stopped for  $\sim 100$  h while additional heat calibration tests were made. When the cell current was again initiated, the excess energy persisted at about the same level.

At  $\sim 1780$  h, the cathode current was stopped momentarily, and the heavy water electrolyte ( $\text{D}_2\text{O}$ -LiOD) was replaced by a comparable light water system ( $\text{H}_2\text{O}$ -LiOH). To ensure that most of the  $\text{D}_2\text{O}$  was removed, the electrolyte inventory was completely replaced twice. As expected, removal of the heavy water resulted in a decrease in the power excess to approximately power balance, but this required over  $100$  h (Figs. 9 and 12). Perhaps this means that the energy-producing process was a bulk phenomenon that required a significant replacement of  $^2\text{H}$  with  $^1\text{H}$  within the palladium cathode to "turn off" the excess heat generation.

#### Detection of Products of Nuclear Interactions

During the open-system test, no anomalous count rates were observed for neutrons or gamma rays on either the NaI or NE-213 detection systems. In particular, the neutron count

rate from the NE-213 detector remained at about the level of the predetermined neutron background ( $40$  neutron count/ $24$  h) for the entire duration ( $1300$  h) of the test. However, in the closed-system test, there were small, but significant, increases in both the fast neutron and the gamma-ray count rates from the NE-213 system and, at some energy levels, in the gamma-ray count rates from the NaI scintillation system (Table I and Fig. 9).

#### Neutrons

Unexplained increases in the fast neutron count rate were observed in three significant periods of the closed-system test (Fig. 9). The first of these occurred during a  $72$ -h period around  $400$  h from the start of the test in which the mean neutron count rate increased  $57\%$  above the previously determined background level and  $27\%$  above the immediately preceding mean count rate. The  $95\%$  confidence level of the higher neutron count rate, as shown in Fig. 9, indicates that this was statistically significant. This observation was made during a time of apparent modest excess power that was at a level well within the experimental error.

The second important interval of increased neutron count rate was initiated and continued for  $96$  h during the second cyclic operation of the cathode current density in which a cycle period of  $5.7$  min was used. This was the most significant increase in the mean neutron count rate, resulting in a level that was  $76\%$  greater than the background and  $54\%$  greater than the immediately preceding level. This was again a statistically significant increase.

The longest period of apparent increase in the neutron count rate lasting  $\sim 300$  h started a few hours after initiation of the extended period of excess power that began at  $\sim 1330$  h from the beginning of the test as an apparent result of the decrease in electrolyte temperature (Fig. 9). The maximum mean value was  $69\%$  greater than the background level and

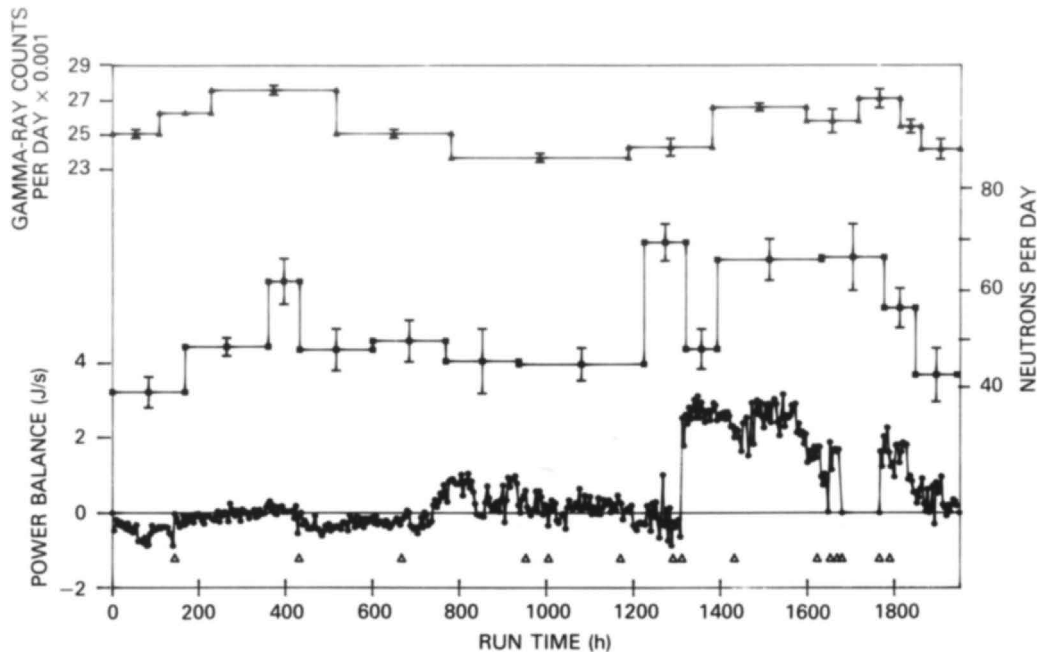


Fig. 9. Important experimental measurements during the closed-system test. The lower curve is the power balance, with each point representing the mean value of 30 measurements during a 4-h interval; the middle curve represents the mean value of the daily neutron count rate over the period indicated; and the upper curve represents the mean value of the daily gamma-ray count rate over the period indicated for the energy range of 2.64 to 3.14 MeV from the NaI system. The error bars in the upper two curves are the 95% confidence levels of the indicated mean values of the counting rates. The open triangles represent significant changes in the operation of the system, including 144 h—increase insulation; 431, 667, and 953 h—increase cathode current density from 100 to 200 to 300 to 400 mA/cm<sup>2</sup>, respectively; 1005 h—initiate cyclic cathode current density varying from 200 to 400 mA/cm<sup>2</sup> with a 66-min period; 1170 h—initiate cyclic cathode current density varying from 300 to 500 mA/cm<sup>2</sup> with a 5.7-min cycle; 1291 h—maintain cathode current density constant at 500 mA/cm<sup>2</sup>; 1312 h—reduce the electrolyte temperature from 38 to 24°C; 1312 h—increase cathode current density to 600 mA/cm<sup>2</sup>; 1623 h—reduce cathode current density to 50 mA/cm<sup>2</sup> and electrolyte temperature to 10°C; 1653 h—increase cathode current density to 500 mA/cm<sup>2</sup>; 1669 h—increase cathode current density to 600 mA/cm<sup>2</sup>; 1694 to 1766 h—carry out energy calibration tests; 1766 h—initiate electrolysis at a cathode current density of 600 mA/cm<sup>2</sup>; and 1790 h—replace electrolyte with LiOH-H<sub>2</sub>O.

38% higher than the immediately preceding mean value of the neutron count rate, a statistically significant increase. As in the case of the excess power, this neutron count rate began to decrease when the heavy water was replaced with light water and essentially reached background level by the end of the test. This higher level of the neutron count rate persisted even during the time when the electrical current was turned off for heat flow calibrations. Incidentally, Sanchez et al.<sup>8</sup> also observed high neutron levels for a few hours after the electrical current had been turned off in their experiments. There was no indication that the extended observed increases in the neutron count rate were the result of a general environmental increase.

### Gamma Rays

Variation in the count rates of gamma rays determined by both the NE-213 scintillation system and the NaI system (at least up to energy levels of 3.6 MeV) also appeared to have small, but statistically significant, increases. Since the NaI spectra involved higher count rates and better statistics, a more detailed analysis was possible. For that system, the gamma-ray spectra from each 4-h-long counting period were divided into ten equal energy segments, with each segment spanning an energy range of ~0.520 MeV (Table I). The most significant increases in the gamma-ray count rate occurred in

the energy range of 2.64 to 3.14 MeV; therefore, mean values of the count rate for this range have been included in Fig. 9, where they can be easily compared with the neutron count rates and excess power. There were apparent coincidences of increased gamma-ray count rates with the first spontaneous neutron increase around 400 h of operation and for the final neutron increase that occurred during the extended period of excess power starting at ~1320 h (Table I and Fig. 9).

Surprisingly, there were no statistically significant increases in the gamma-ray count rate in the 2.12- to 2.63-MeV energy region (the energy level where gamma rays are emitted from neutron interactions with protons); however, all other energy bins less than that range showed an apparent increase during the extended period of excess power. There was no indication of increased gamma-ray count rates above an energy level of 3.14 MeV, at least up to 5.2 MeV.

These extended periods of increased gamma-ray count rates could not be explained by environmental variations, such as the local barometric pressure or ambient temperature, and there was no apparent change in the instrumental threshold or preamplifier gain. Characteristics of the changes in the gamma-ray energy spectra are also not consistent with interference from electrical noise or from an external radiation source since the variations are relatively long term and do not occur as narrow peaks.

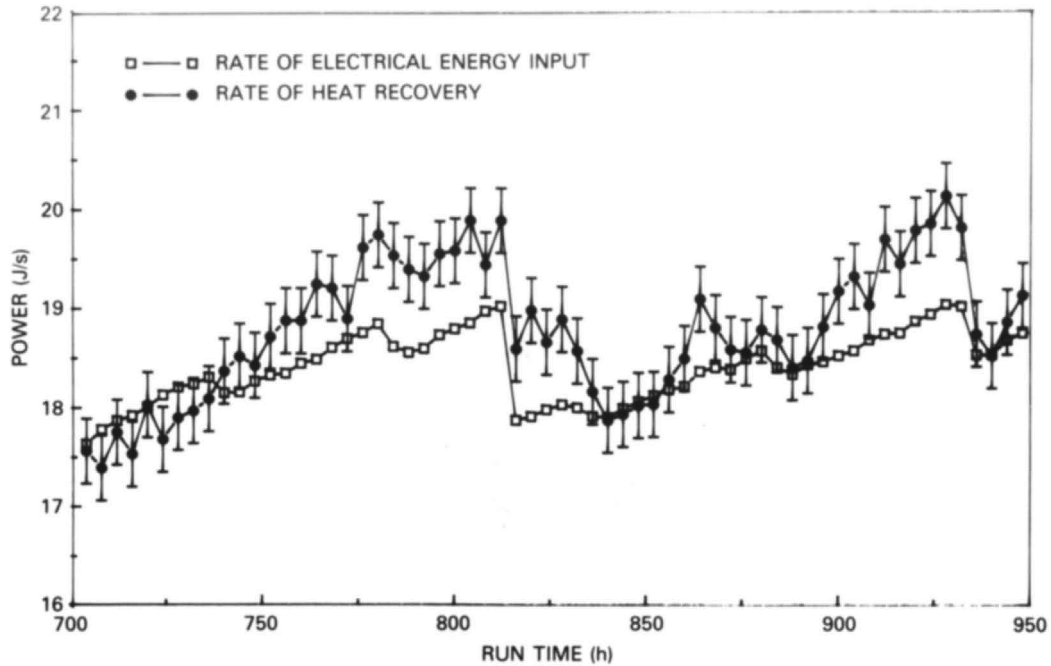


Fig. 10. Power balance in the closed-system test during the period when two major periods of spontaneous excess power were observed. The variation in the rate of energy input was primarily due to changes in cell voltage that resulted when the electrolyte concentration was periodically adjusted. Each point represents the mean value of 30 measurements during a 4-h interval, and the error bars represent the maximum calculated uncertainty in the experimental values based on independent calibration of temperature and flow metering systems.

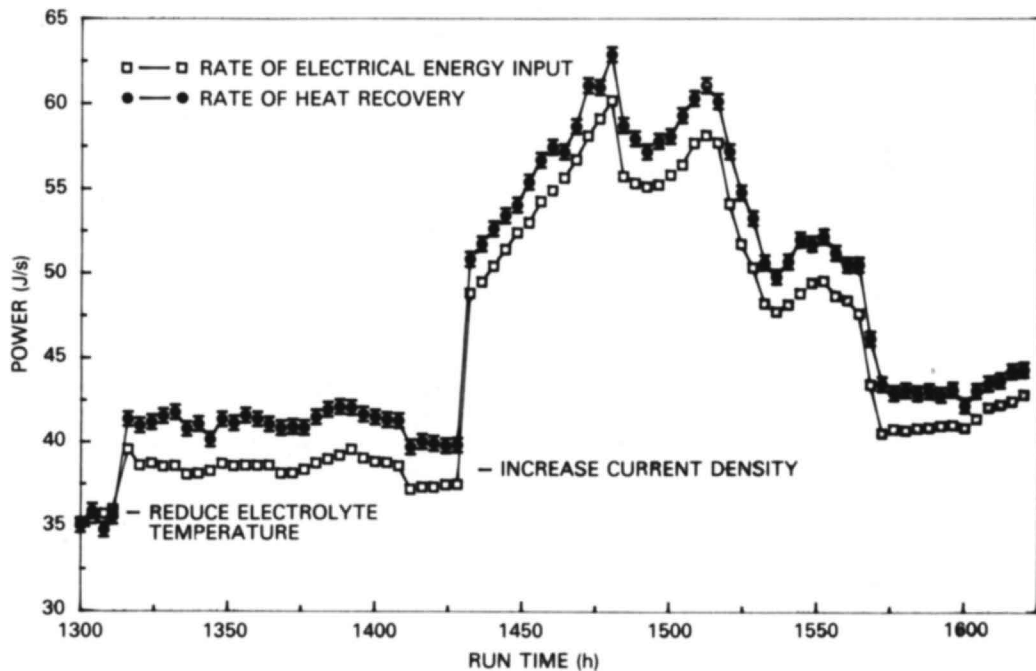


Fig. 11. Power balance in the closed-system test during the period where extended excess power was initiated by the reduction in electrolyte temperature. Variations in the rate of energy input were due to changes in the electrolyte temperature, increase in cathode current density, and periodic adjustments to the electrolyte concentration. Each point represents the mean value of 30 measurements during a 4-h interval, and the error bars represent the maximum calculated uncertainty in the experimental values based on independent calibration of temperature and flow metering systems.

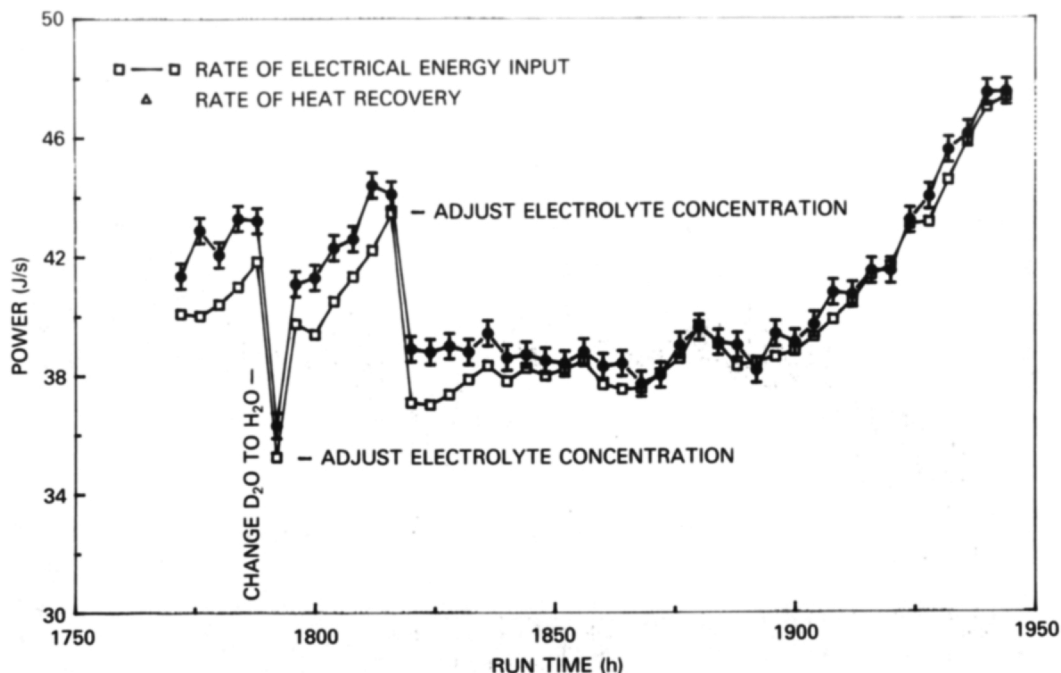


Fig. 12. Power balance in the closed-system test during the period when the electrolyte was changed to light water. Variations in the rate of energy input were primarily due to variations in the electrolyte concentration. Each point represents the mean value of 30 measurements during a 4-h interval, and the error bars represent the maximum calculated uncertainty in the experimental values based on independent calibration of temperature and flow metering systems.

TABLE I

Change of Gamma-Ray Count Rates as a Function of Run Time and Energy Level\*

Detector and Energy Level (MeV)	Mean Count Rate (count/4 h) <sup>a</sup>		
	1088 to 1188 h	1509 to 1609 h	1948 to 2048 h
NE-213 detector >0.3	29 ± 2.1	43 ± 2.4	28 ± 2.5
NaI detector			
0.14 to 0.57	785 233 ± 791	792 446 ± 497	712 468 ± 510
0.58 to 1.09	192 750 ± 285	194 227 ± 150	177 009 ± 230
1.10 to 1.60	78 523 ± 168	79 169 ± 288	72 249 ± 112
1.61 to 2.11	18 562 ± 55	19 127 ± 157	17 204 ± 52
2.12 to 2.63	11 156 ± 44	10 995 ± 94	10 252 ± 30
2.64 to 3.14	3 872 ± 31	4 446 ± 80	3 805 ± 24
3.15 to 3.66	554 ± 11	568 ± 28	565 ± 10
3.67 to 4.18	463 ± 6	468 ± 25	470 ± 10
4.19 to 4.69	421 ± 8	427 ± 7	427 ± 7
4.70 to 5.20	385 ± 7	385 ± 17	395 ± 9

\*The first time interval was just before the initiation of excess power for an extended period; the second interval was during the period of excess power; and the third interval was after the test had been terminated.

<sup>a</sup>Mean gamma-ray count rates during the time period noted with the included errors representing the 95% confidence limit on the mean values.

### Tritium

Analysis of the electrolyte showed no evidence of increased tritium concentration greater than the starting material, within an accuracy of 200 Bq/ℓ.

### DISCUSSION

Although periods of induced excess power were observed during the open-system test, the most significant results were

obtained during the closed-system test. Perhaps the most important feature of the closed system is that there is no need to assume that electrolysis gases do not recombine; thus, there is less inherent uncertainty in the experimental results. Spontaneous periods of excess power were observed; even more significant, however, was the demonstration that excess power could be initiated by introducing system perturbations. A decrease in the electrolyte temperature appeared to be most efficient for this effect. One of the consequences of decreasing the electrolyte temperature is an increase in the deuterium content of the cathode.<sup>11</sup> Therefore, it may be advantageous to work at low temperatures and high current densities when searching for deuterium-deuterium (D-D) reactions.

Of equal importance were the several periods of apparent increases in the neutron count rate. In one case, this also appeared to be spontaneous, but in two other cases they appeared to be related to system perturbations such as cathode current cycling or electrolyte temperature change. The apparent coincidence of the increase in neutron count rate with most of the extended period of excess power was of even greater interest.

The maximum increase in the fast neutron count rate, taking into account the efficiency of the detection system, was  $\sim 0.23$  n/s, a value that is statistically important but relatively modest. Assuming the increase is due to neutron emissions from the electrolysis experiment and assuming spherically homogeneous emissions, this value corresponds to  $1.6 \times 10^{-23}$  fusion/D-D pair. The detected rates are too low, by many orders of magnitude, to explain the observed energy excesses in terms of conventional D-D fusion theory.

The apparent increases in the gamma-ray count rates appeared to be coincident with increases in the neutron count rates. These gamma-ray increases were quite modest and somewhat puzzling. With low levels of increased neutrons and the very low efficiency of the gamma-ray detectors, it was not surprising that gamma rays from the neutron/proton interactions (2.2 MeV) were not observed. However, apparent increases in gamma rays at other adjacent energy levels may indicate that some type of nuclear interaction was occurring. But again, the energy balance between the excess heat observed and an assumed nuclear process differs by many orders of magnitude.

The several periods of excess power were unequivocal and could not be explained by experimental inaccuracies or artifacts. Apparent increases in the neutron and gamma-ray count rates were quite modest and perhaps could be explained by some sort of unknown variations in the background levels of the two separate monitoring systems. However, the apparent coincidence of increases in two independent detectors, and especially the coincidence with an extended period of induced excess power and the concurrent decreases when light water was added to the electrolysis system, gives more credence to the results.

The replacement of LiOD-D<sub>2</sub>O with LiOH-H<sub>2</sub>O resulted in an ultimate reduction of excess power and a decrease of the neutron and gamma-ray count rates to background values. These observations suggest that deuterium is a necessary component of the electrolyte for positive results; however, the length of time required for the reductions was well over 100 h. This finding could be interpreted as demonstrating that the possible interactions are bulk phenomena, which occur throughout the cathode matrix, rather than surface phenomena, which would have disappeared very rapidly after removal of the source of deuterium.

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