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Charles D. Scott, Elias Greenbaum, Gordon E. Michaels, John E. Mrochek,
Eugene Newman, Milica Petek, and Timothy C. Scott

Chemical Technology Division
Oak Ridge National Laboratory*
Oak Ridge, Tennessee 37831-6226

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ABSTRACT

Preliminary tests have been made with electrolytic cells utilizing 0.2 N LiOD in D_2O as the electrolyte and a palladium cathode surrounded by a wire-wound platinum anode operating at cathode current densities of 100 to 400 mA/cm^2. The cathodes were swaged to diameters of 2.8 or 5.5 mm with 8.5 cm of active length. The electrolyte temperature was controlled, heat was removed by flowing water in a cooling jacket, and the cell was insulated. Cooling water and electrolyte temperatures were measured by thermocouples, and neutron and gamma-ray spectra were recorded. The electrolyte level was periodically monitored and replenished with D_2O. Tests up to 2 weeks in duration were made with no sustained release of energy in excess of the electrical power input, although there was one period of 12 h when an unaccountable heat excess was observed. In another test, an anomalous neutron flux was measured during the first few hours that was 3.5 standard deviations above the background.

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KEYWORDS; Cold fusion, electrolysis, palladium cathode

1. INTRODUCTION

There have been recent experimental indications from several research groups that excess energy has been observed during the electrolysis of heavy water in a LiOD electrolyte solution with a palladium cathode.[1-3] In some cases, neutron flux anomalies have been observed and an increase in tritium content of the electrolyte solution has also been measured.[3-5]

A group of researchers at Oak Ridge National Laboratory, which includes chemists, physicists and chemical engineers, has initiated an investigation with the goal of experimentally studying possible thermal or nuclear reactions occurring during electrolysis under the above conditions. The experimental system utilized positive heat removal by circulating cooling water, a design concept that is amenable to system scaleup and continuous operation.

2. MATERIALS AND METHODS

2.1. Electrochemical Cell

The electrolysis cell was fabricated from Pyrex glass with a nominal ID of 4 cm and an active internal height of 12.5 cm (Fig. 1). There was also an internal head space of ~4 cm in height which was continuously purged by N_2 gas. A Teflon cap was used to partially seal the system and support electrodes, an electrolyte thermocouple encased in glass, a glass-encased heating source for internal calibration, and a polyethylene tube for introduction of make-up water and removal of electrolyte samples. The cell was encased in a cooling jacket with forced water flow and thermocouples in the inlet and outlet streams. Two inches of fiber glass insulation was used on the entire exterior of the cell except for a 1- x 3-cm opening in the insulation that was used to observe the electrolyte level.

Cathode rods of 99.95% palladium were prepared by casting under argon and then swaging to the final dimensions of 0.28- or 0.55-cm diam x 8 to 9 cm. Electrical contact was via a 0.050-in. Pt wire, electron-beam welded to the Pd cathode. The anodes were 0.02-in.-diam Pt wire

that was wound around a glass skeletal mandrel which surrounded the cathode with an annular electrode separation of 0.4 to 0.5 cm. The cell was operated at a cathode current density of 100 to 400 mA/cm^2.

2.2. System Design

Heat removal was determined by the temperature increase of the forced flow of cooling water through the jacketed cell. A circulating coolant bath with temperature controlled to within ±0.1°C was used with an external positive displacement pump that controlled the coolant flow rate with an accuracy of 2 to 4% (Fig. 2). Rotameters were employed to monitor the coolant and N_2 purge gas flow rates. Heavy water replenishment and electrolyte sampling were performed intermittently by means of a manually operated syringe.

The potentiostat/galvanostat was operated in constant current mode with an accuracy of ±1 mA. Electrolyte and coolant water inlet and outlet temperatures were measured by calibrated thermocouples with an accuracy of ±0.1°C, and the overall electrode voltage was measured to an accuracy of ±0.01 V. These system parameters were recorded on a strip chart recorder every 6 s.

2.3. Radiation Detection

The experimental system was located within a 2-ft-thick concrete enclosure in order to reduce background radiation. Neutrons were measured by a NE-213 scintillator placed immediately adjacent to the insulated cell (Fig. 3). This detector employed pulse-shape discrimination to differentiate between neutrons and gamma rays and the threshold energy for fast neutron detection was set at 1.2 MeV. The overall neutron detection efficiency of 1.46×10^{-3} was determined by a ^{252}Cf source. Gamma-ray contribution to the neutron peak was 4.1% of the gamma peak or ~20% of the neutron peak. The reported neutron flux was corrected for this contribution. This neutron detector had a typical sensitivity at 3 standard deviations of 3×10^{-24} fusions/deuterium-pair-s.

A separate NaI gamma-ray spectrometer with a polyethylene converter to generate capture gamma rays was also used for detection of neutrons. This system was lead-shielded and contained in an airtight enclosure to further reduce the background gamma-ray spectrum. A multichannel analyzer with a window set over the expected capture gamma-ray region was used; the system had an overall detection efficiency of 5.75×10^{-5} as determined by a ^{252}Cf source. However, this system had somewhat less sensitivity due to the higher gamma-ray background. The tritium concentration in the electrolyte was measured periodically by an external scintillation system.

2.4. Chemicals

The heavy water was 99.5% D_2O with an initial tritium content of 1×10^3 to 1.3×10^4 Bq/mL. The electrolyte was prepared by dissolving reagent-grade, natural lithium in the D_2O at a nominal concentration of 0.2 N.

2.5. 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 and turning on the electrical current. The electrolyte temperature was controlled by the cooling water temperature and flow rate and usually maintained in the range of 28 to 32°C with some controlled excursions to 70°C.

The various temperatures and the cell voltage were recorded every 6 s on an analog recorder and at 1-h intervals as digital displays. Heavy water (D_2O) was replenished every 8 h in order to maintain a constant electrolyte concentration, and electrolyte samples were taken periodically (every 2 to 3 d). The gamma-ray and neutron spectra were measured continuously and recorded on magnetic media every 4 h.

2.6. Energy Balance

An energy balance 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) all of the D_2 and O_2 exit the electrolysis cell without recombination; (4) the exiting gases, including the purge gas, were saturated with D_2O that was at equilibrium with heavy water at the temperature of the electrolyte; and (5) there was no heat loss to the ambient environment.

The second assumption will obviously not be correct during the initial phase of the experiment when a major portion of the formed D_2 is being adsorbed by the cathode. Since the amount of make-up heavy water required to maintain a constant inventory in the electrolysis cell was approximately equal to that lost by electrolysis and evaporation, there was apparently very little recombination of the product gases. There was obviously some heat loss to the ambient although the system was well insulated, but this would result in a conservative estimate of the recovered heat.

The resulting energy balance can be represented by:

ENERGY IN:

Voltage (V) x Current (A)

ENERGY OUT:

1. ELECTROLYSIS (Typically >30%)

   D_2O --> D_2 + 1/2 O_2 (-Heat of formation)

2. FORCED COOLING (Typically >60%)

   (Temperature increase) x (Flow rate)

3. LATENT HEAT (Typically <2%)

   (Heat of D_2O vaporization)

Chemical and physical properties for D_2O were obtained from reference handbooks.[6,7]

3. RESULTS AND DISCUSSION

The results of these tests are very preliminary and certainly do not confirm "cold fusion" as described by Pons and Fleischmann. However, during one short time period there was an apparent neutron increase, and during a period of a few hours there was apparently a significant amount of excess heat generated.

3.1. Neutron Detection

In one test (CF-1) in which a palladium cathode 0.28-cm diam x 8.5 cm was used at a current density of 100 mA/cm^2, the neutron background averaged 13.1 neutrons/4 h with a standard deviation of 5.3 neutrons/4 h. During the first few hours of this test there was a 4-h observation period, in which the neutron detector registered 32 counts/4 h (Fig. 4) exceeding the background count rate by 3.5 standard deviations. A frequency histogram of the neutron count data (Fig. 5), shows that the variability of our neutron observations for this run fits a binomial distribution with the possible exception of the anomalously high datum of 32 counts/4 h.

3.2. Excess Heat

There was no sustained excess energy measured during either of the two tests. However, there were several short periods of time with apparent excess energy and one period of 12 h in one test when the energy recovered was in excess of the instantaneous input energy by as much as 50% (Fig. 6). That energy excess occurred during the second test (CF-2) with a 0.55-cm diam x 8-cm palladium cathode operating with a current density of 100 mA/cm^2.

3.3. Additional Observations

The periodic addition of D_2O to maintain the heavy water inventory resulted in fluctuations of LiOD concentration and, thus, unsteady state operation since cell voltage changed with electrolyte concentration. A series of tests were also made at different electrolyte temperatures over the range of 30 to 70°C without apparent effect. Small tritium increases were measured in the electrolyte, but these did not exceed the expected increase due to the evaporation and electrode processes.

4. CONCLUSIONS

Preliminary tests of the electrolysis of D_2O utilizing LiOD electrolytes and palladium electrodes have not confirmed the "cold fusion" phenomena. However, there has been one apparently anomalous neutron flux measurement and periods of up to 12 h of apparent excess energy. None of these results have been reproduced nor can they be explained by conventional nuclear or chemical theory.

5. ACKNOWLEDGMENTS

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FIGURE CAPTIONS

Fig. 1. Electrolysis cell used in cold fusion study.

Fig. 2. Schematic of experimental system.

Fig. 3. Top view of the placement of the neutron and gamma-ray monitoring systems.

Fig. 4. Corrected neutron flux as a function of time in test CF-1 with a 0.28-cm diam x 8.5-cm palladium cathode in 0.2 N LiOD operating at 100 mA/cm^2.

Fig. 5. A frequency histogram of the neutron flux in test CF-1 indicating the anomalous result of 32 neutrons/4 h.

Fig. 6. Energy balance of test CF-2 with a 0.55-cm diam x 8.5-cm palladium cathode in 0.2 N LiOD operating at 100 mA/cm^2.

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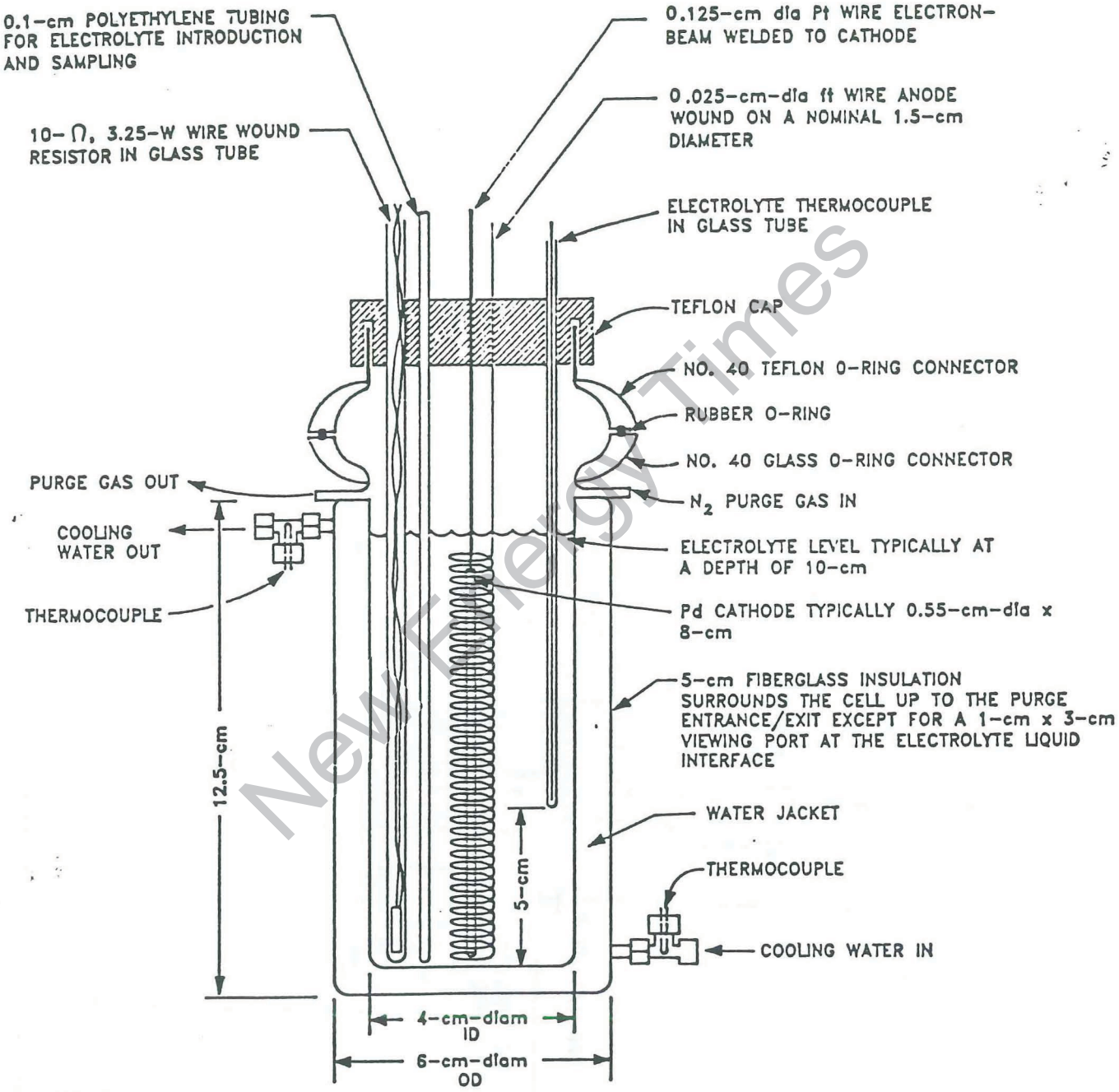
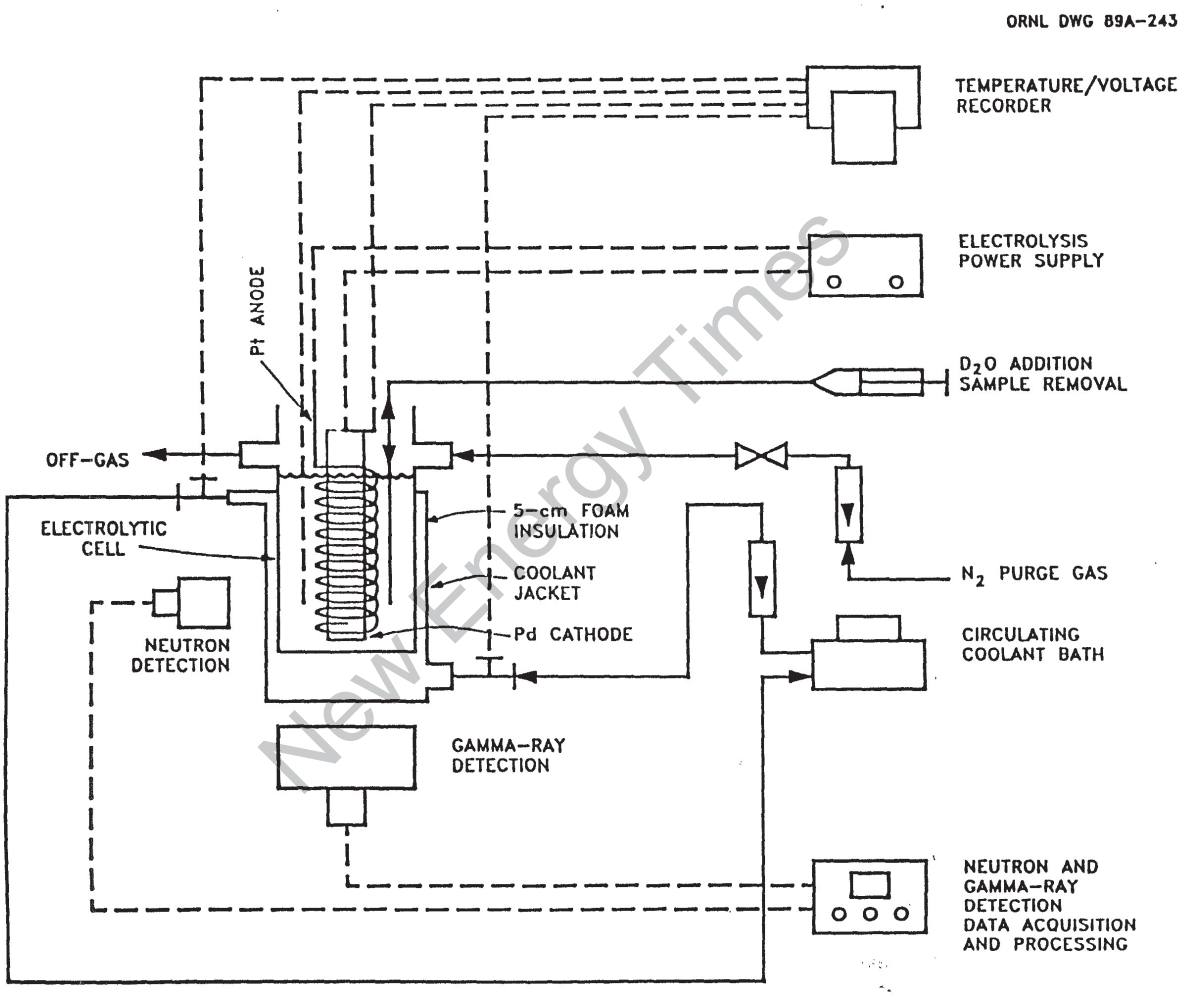


ELECTROLYSIS CELL USED IN COLD FUSION STUDY



SCHEMATIC OF COLD FUSION EQUIPMENT

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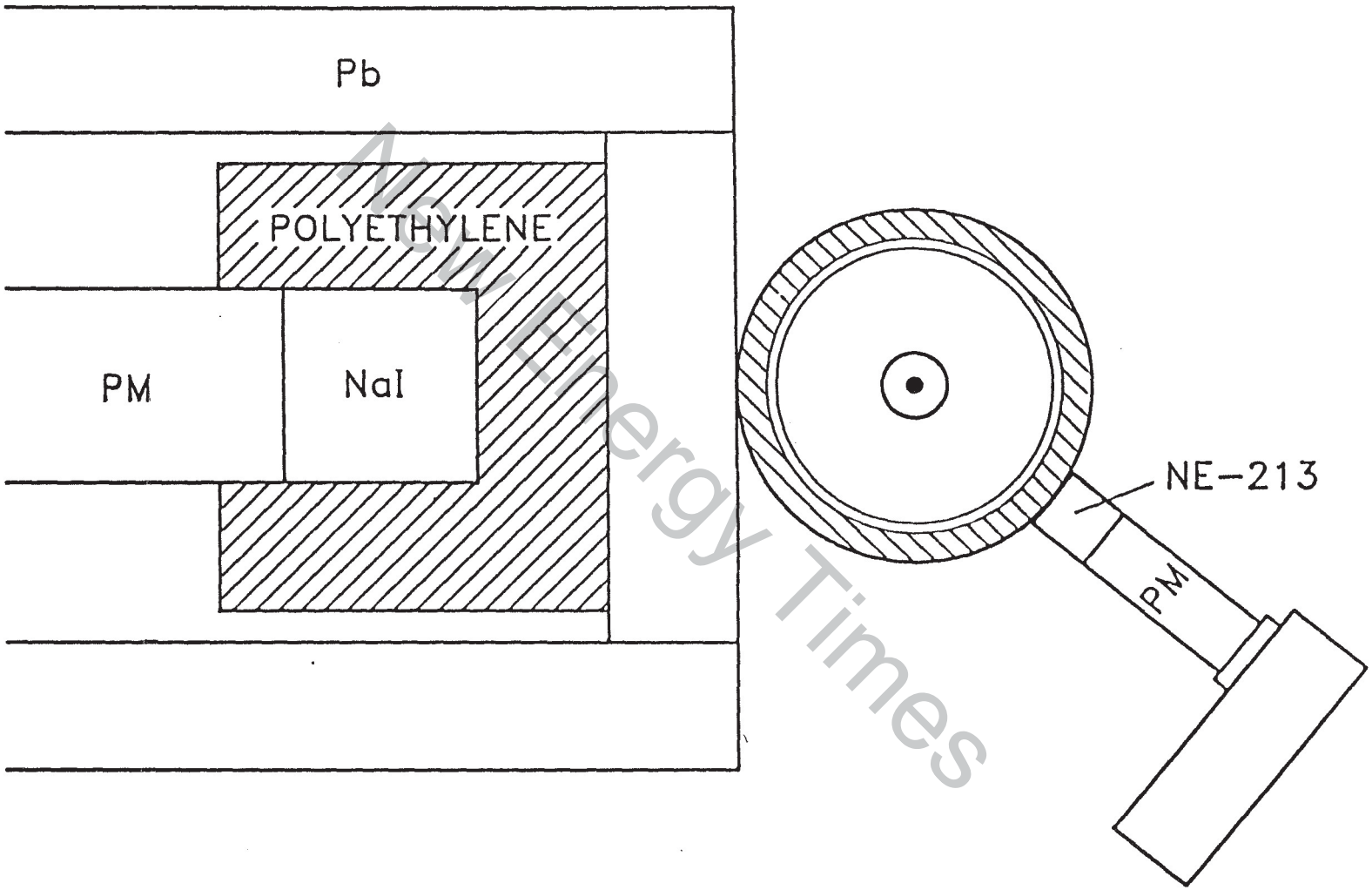

DETECTION SCHEMATIC
(TOP VIEW)