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SEARCH FOR NEUTRON EMISSION DURING THE ELECTROLYSIS OF HEAVY WATER

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ABSTRACT

A liquid scintillator detector NE 213 with pulse shape discrimination technique was used to observe neutrons during the electrolysis of heavy water with a palladium cathode. From the measured fore and background counting rates, a neutron emission rate of $(8.2 \pm 2.9) \times 10^{-3}$ n/(sec.g.) Pd was determined implying $(2.9 \pm 1.0) \times 10^{-24}$ fusions/ [(dd pair).sec.] as compared to $\sim 10^{-23}$ fusion/ [(dd pair).sec.] reported by Jones et al. using titanium electrode.

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PESQUISA SOBRE EMISSÃO DE NÊUTRONS DURANTE A ELETRÓLISE DE ÁGUA PESADA

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RESUMO

Um cintilador líquido NE 213 junto com a técnica de discriminação em forma de pulso foi utilizado para observar a emissão de nêutrons durante a eletrólise de água pesada com catodo de paládio. A taxa de emissão de nêutrons observada foi de $(8.2 \pm 2.9) \times 10^{-3}$ n/(seg.g.) Pd indicando $(2.9 \pm 1.0) \times 10^{-24}$ fusões/[(par dd).seg.] comparado com o valor obtido de $\simeq 10^{-23}$ fusões/[(par dd).seg.] por Jones e outros utilizando o eletrodo de titânio.

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INTRODUCTION

Generally speaking the process of nuclear fusion is always associated with experiments which involve enormous magnets, powerful lasers and plasma and temperatures as high as 100-150 million degrees Kelvin. Under these extreme conditions the hydrogen atoms are known to fuse forming helium and giving off energy. The experiments recently reported by Jones et al[1] and Fleischmann and Pons[2] are however concerned with the production of fusion reaction in an electrochemical cell containing heavy water and a cathode made from palladium or titanium metal. The two common reactions considered here are:

$$d + d \longrightarrow {}^{3}He\ (0.82\ MeV) + n\ (2.45\ MeV) \qquad (1)$$

$$d + d \longrightarrow {}^{3}H\ (1.01\ MeV) + p\ (3.03\ MeV) \qquad (2)$$

While the experiments of Jones et al[1] reported the observation of neutron emission during the electrolysis with energy spectrum peaking around 2.45 MeV, Fleischmann and Pons[2] have reported in addition to the neutron and gamma-ray emission observation of subtantial quantities of heat released during the electrolysis which was measured by calorimetric method.

It appears from these experiments that a hitherto unknown mechanism based on the interaction of deuterium atoms with palladium (titanium) metal lattice plays an important role in catalysing the fusion reaction. Considering an extraordinary importance of this process as a future energy source many laboratories in various parts of the world quickly started work to confirm the original results and evaluate net energy out put of the reactions[3-7].

The present experiment was designed to observe the neutron emission during the electrolysis of heavy water and try to measure the energy spectrum of these neutrons. For this purpose we used a liquid scintillator NE 213 and an electronic system with pulse shape analysis in order to distinguish between neutron and γ-ray signals. The experiments whose results we report here were realized during the period, April 14 - 20, 1989.

EXPERIMENTAL SETUP AND RESULTS

An schematic of experimental setup is shown in fig. 1. The electrochemical cells were constructed from silica tubes (65 mm long and 25 mm diameter) closed flat at one end. Polyethelene stoppers with rubber o-ring and having small holes for the passage of electrode terminals kept the cells almost air tight. The escape of oxygen and deuterium gases liberated during the electrolysis occured through a pin-hole provided in the stopper. A platinum foil (0.25 mm thick) folded into a cylindrical form (20 mm diameter and 50 mm long) and welded to a platinum wire (1 mm diameter) was used as anode, while a palladium metal sheet (40 mm x 6 mm x 1 mm) weighing $\simeq$2.75g and welded to a platinum wire served as a cathode. The palladium metal was 99.9% pure and was used as such without any pretreatment. The stock solution of the electrolyte was made by following the prescription given in ref. 1. The purity of heavy water used was 99.8% and approximately 20 ml of solution was used in each cell. Three cells were used simultaneously for the experiment. Statilized D.C. power supplies (separate for each cell) operating in the constant current mode provided a current between 100 and 250 mA for each cell.

The neutron spectrometer recently constructed and tested at the reactor physics division of IPEN-CNEN/SP has been crucial as well as a source of motivation for carrying out the present experiment.The neutron detector consisted of a NE 213 liquid scintillator encapsulated in a standard type VH-1 aluminum (2" x 1.5") with glass window. The detector was optically coupled with a RCA-8850 photomultiplier tube.More details about the neutron spectrometer and its performance are described elswhere(8). The important features of the spectrometer are: ~ 6% intrinsic detection efficiency (measured with a calibrated ^{252}Cf source: (En= 2.1$\pm$ 1 MeV) and a pulse shape discrimination for gamma rays better than 8 x 10^{-4} (γ/n). The neutron detector as well as the electrolytic cells were placed inside a shield constructed from borated paraffin (20 cm thick) and lead (10 cm thick) blocks. The cells were arranged as shown in the fig. 1 at a distance of approximately 2 cm from the detector. This distance was considered safe enough to avoid the convection heating of the detector from the heat generated during the operation of the cells. The

performance of the liquid scintilator is known to be affected by temperature fluctuations.

Each electrolysis run lasted for about 6 hours during which the pulse height spectrum of recoil protons from the interaction of neutrons with scintilator material was stored in a multichannel pulse height analyser. Before each run control experiment was carried out to obtain the background spectrum. Two types of control experiments were performed : a) original cells were replaced by similar cells containing the same electrolytic solution however in ordinary water and the electrolysis continued; b) original cells were maintanied but the electrolysis was turned off. As a further check on the neutron background in the experimental area two ^{3}He long counters were placed at about 4 meters from the cells contineously monitoring the background. No significantly large fluctuations (other than the statistical) in the background were observed during the period of the experiment. After each electrolysis run the palladium cathode was cleaned by scraping off the gray iron deposite and the loss of electrolyte was made up by adding the necessary volume of the solution.

The energy calibration of the spectrometer was periodically checked with γ-ray sources of ^{137}Cs and ^{60}Co as well as with a neutron source of Am-Be. The pulse height spectrum accumulated during the electrolysis of heavy water is shown in figure 2. The data correspond to a total run time of 48 hours. The energy calibration implied that the signals from 2.45 MeV neutrons should appear in this spectrum within the channels ranging from 17 to 40. The background spectrum for the same total time period is also shown in this figure. By comparing the energy spectra from gamma and neutron source it was determined that at least a part of the background came from γ-ray events despite a high degree of pulse shape discrimination achieved.

As can be seen from fig. 2 the counting statisties obtained in the present experiment is not sufficient to permit the unfolding of the recorded recoil proton spectrum into neutron energy spectrum, which is usully done by using a response function either generated experimentally or obtained through a Monte Carlo calculation. It was thus not possible to determine the exact energy distribution of emitted neutrons. The

spectra presented in fig. 2 were therefore integrated over energy region where 2.45 MeV neutrons are expected (channels 17 through 40) resulting in the signal plus background rate of 5.65 $\pm$ 0.34 per hour while the background rate was 3.21 $\pm$ 0.26 per hour. The errors quoted here are only statistical errors. From these results we deduce a net signal rate 2.44$\pm$ 0.43 per hour in the present experiment. The total neutron detection efficiency including the geometrical acceptance (the geometrical factor was calculated) is approximately (1.0 $\pm$ 0.3) x 10^{-2}, this results in the neutron emission rate of (6.8 $\pm$ 2.4) x 10^{-2} n/sec. Since a total of 8.25 g of palladium were used in there cells, we obtain the emission rate to be (8.2 $\pm$ 2.9) x 10^{-3} n/(sec.g) Pd.

CONCLUSIONS

The results of the present investigation indicate that within the statistical limits of the experimental data slightly more number of neutrons are detected during the operation of the electrolytic cells containing heavy water than can be accounted for by the background events. With the assumption that d:Pd ratio in the deutrated electrode is unity (this ratio was not measured in our experiment), the present result corresponds to a fusion rate (according to equation 1) of (2.9 $\pm$ 1.0)x10^{-24} fusions/[(dd pair) sec] or to an upper limit of 3.9 x 10^{-24} fusions/[(dd pair) sec]. This value is more than two and a half times lower than that reported by Jones et al[1]. The present value of (8.2 $\pm$ 2.9) x 10^{-3} n/(sec.g)Pd is more than five orders of magnitude smaller than what has been claimed by Fleischmann and Pons[2].

To summarize, we conclude that the present experiment although provides some evidence that a small flux of neutrons is detected during the electrochemical infusion of deuterium into palladium metal, the observed rate of the reaction (equation 1) is quite small.

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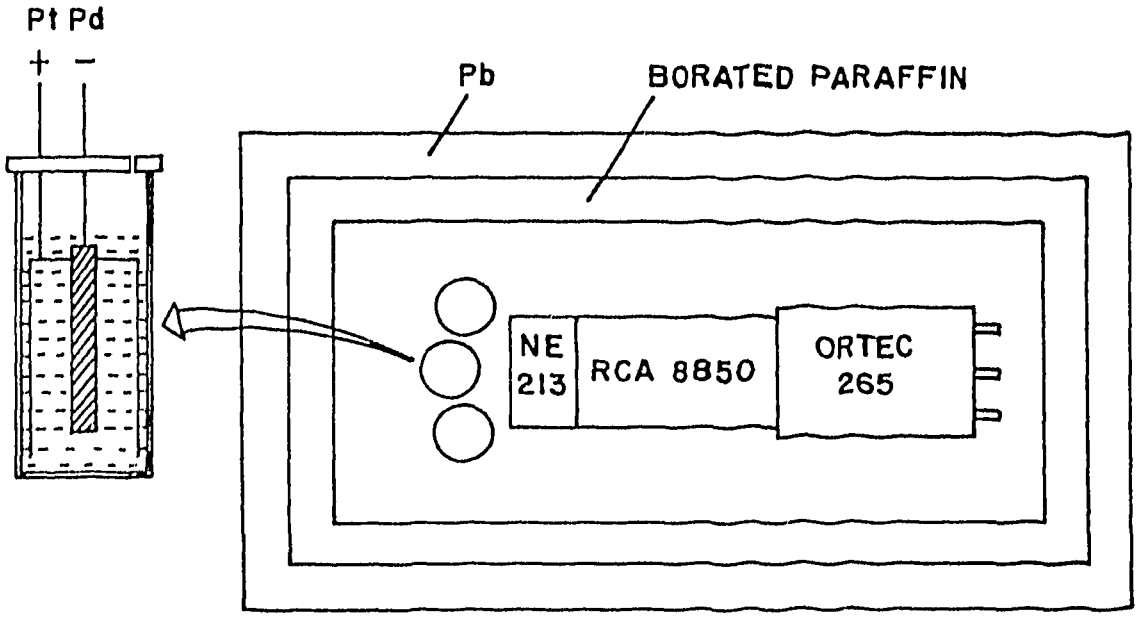

FIGURE 1 - An schematic of experimental arrangement showing the electrochemical cell and the neutron detector.

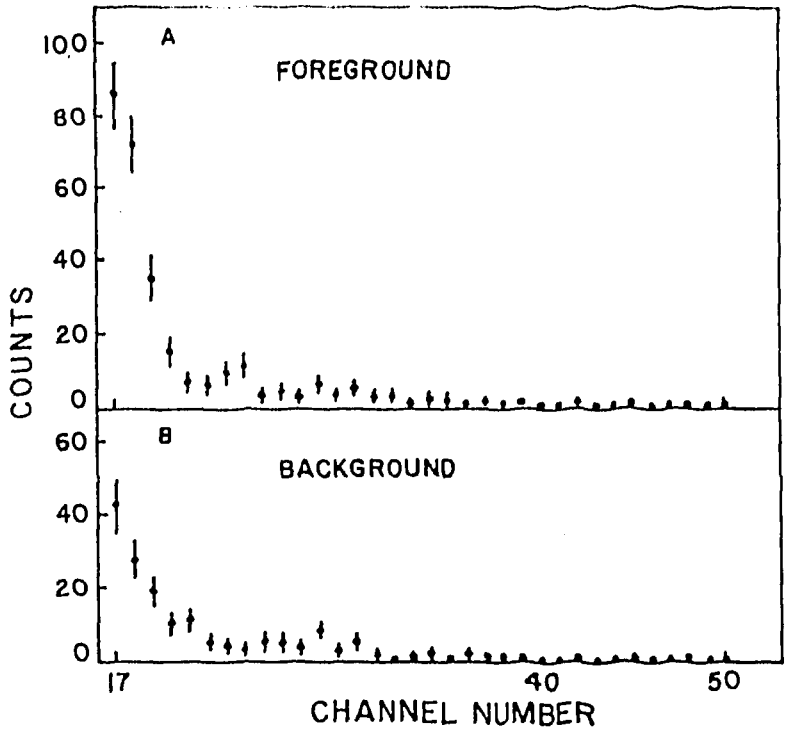

FIGURE 2 - Pulse height spectrum of recoil protons detected by the liquid scintilator NE 213 A) recorded during the period of electrolysis B) recorded during the measurement of background.