Measurement of Neutrons in Electrolysis at Low Temperature Range

Masatoshi FUJII, Masami CHIBA, Kenji FUKUSHIMA, Motomi KATADA, Tachishige HIROSE, Kenichi KUBO, Hiroaki MIURA, Shinya MIYAMOTO, Hiromichi NAKAHARA, Yuki NAKAMITSU, Tsutomu SEIMIYA, Toshiaki SHIRAKAWA, Keisuke SUEKI, Hideyuki TORIUMI, Torahiko UEHARA, Toshihiro WATANABE, Tokyo Metropolitan University

ABSTRACT

We observed three neutron trains continuing 2 or 3h as the excess flux during an electrolysis of deuterated alcoholic electrolyte at low temperature range with Pd cathode for 1878h. Those were 22 counts/2h and two times of 27 counts/3h in the average background counts of 4/h. We could not find any excess in the other time intervals between 28.6ms and 1878h. The production rates of the 22 and 27 counts were $6.3X10^{-24}$ and $4.5X10^{-24}$/d-d/s during 2 and 3h, respectively. The confidence levels of neutron emission for 22/2h and 27/3h were 88% and 59%, respectively, estimated from the statistics. And for the total of the three trains, the confidence level became 98%.

1. Introduction

We focused our attention on the result of De Ninno et al.[1] who observed neutron emission changing temperature of titanium in pressurized deuterium gas. Neutrons emerged from the titanium in a particular temperature of 240 K. This temperature seems to be corresponding to the phase-transition of titanium hydrides. The change could make a cracks with a high electric field between the gaps which accelerated deuterons.

So, we carried out the first experiment for searching neutrons from an electrolysis changing the temperature between 200 K and 300 K in order to create the dynamic condition in palladium cathode. Additionally, the advantage of the electrolysis at the low temperature range is not only the capability of changing the temperature, but also increase of an absorbed amount of hydro-

FRONTIERS OF COLD FUSION

gen into palladium [2-4]. Furthermore, our detection system was constructed in attention with a time interval as wide range as possible, since we do not know how the neutron emission occurs in the time structure.

2. Methods

2.1 Neutron detection system

Neutron detection system was consisted of ten ^3He proportional gas counters (Reuter-Stokes, USA), which were inserted in a paraffin moderator shaped as a cylinder with a height of 30 cm, outer and inner diameter of 38 cm and 10 cm, respectively. Each couple of the ^3He counters was linked to a preamplifier. The pulse height data were recorded using a CAMAC PHADC. The event time was also stored to analyze the time structure of the neutron emissions. The pulse height data can be read out every 28.6 ms, which is limited by the CAMAC readout time.

Detection efficiency of neutrons was defined as number of detection per number of emitted in the cell which was measured by the ^{252}Cf neutron source placed at the palladium electrode position. The detection efficiency was 3.3 % selecting within -3σ to $+2\sigma$. The detection system was placed in Nkogiriyama underground laboratory, Institute for Cosmic Ray Research, University of Tokyo, which was covered with sandstone of 100 m water equivalent. The average of neutron background rate was about 4/h.

2.2 Electrolysis

The cell used in this work was made of quartz glass of 22 mm in inner diameter and 150 mm in length. The anode was a coiled shape made of a platinum wire of 0.5 mm in diameter and 500 mm in length. The palladium cathode electrode of 5 mm in diameter and 20 mm length was placed in the center of the platinum wire. We used an electrolyte as 2N DCl(37 wt% with 99% D; Aldrich Chemical Company, Inc.) of deuterated methanol (99.8 atom% D; ISOTEC Inc.) solution, whose freezing point was lower than 200 K.

The electrolysis was continued unexchanging the electrode but exchanging the electrolyte for about 2 weeks, because methanol was resolved into formaldehyde and formic acid which froze under the low temperature condition. The electrolysis was carried out with constant current mode which was repeated between -113.2 mA/cm^2 for 170 min. and -0.14 mA/cm^2 for 10 min. The total electrolysis duration was 1878 h, and the temperature of the electrolysis bath were kept at about 210 K.

The amount of absorbed deuterium was estimated by the measurements of thermogravimetory (TG) and temperature programed desorption (TPD) spectra of mass number 2. In our electrolysis condition, the average ratio of deuterium was Pd:D=1:0.7.

3. Analysis and results

We studied the frequency distributions of the neutron count during the electrolysis of each time interval. The expected frequency distribution was deduced from the Poisson distribution substituting the average and the total counts of the time interval.

Searching from the time interval from 28.6 ms to 240 min, almost all the time interval frequency distributions were fit to the Poisson, but some anomalous events were observed in 120 and 180 time intervals (Fig. 1). They were one event of 22 counts/2h, which occurred at 31.01 days after the beginning of the electrolysis and two events of 27 counts/3h at 62.14 and 82.17 days, respectively. The pulse height distributions of the three events were similar to that of the 252Cf source data (e.g. fig. 2 shows the distribution of 22/2h train). Thus the counts of these events can be decided as the neutron signals. The neutron signals of these

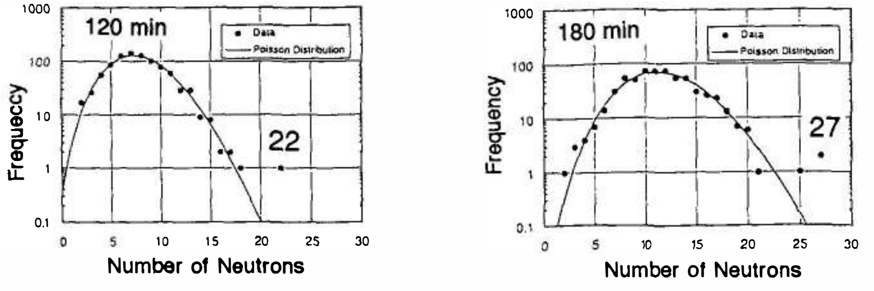

Fig. 1 Frequency distributin of 120 and 180 min

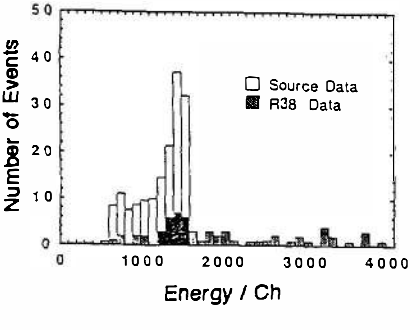

Fig. 2 Energy spectram

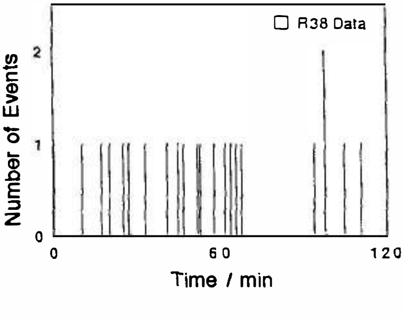

Fig. 3 Time distribution

events were distributed over the time duration (e.g. fig. 3 shows the time distribution of 22/2h train). This time distribution indicates that these emission phenomena is not both a continuous process during the electrolysis, and burst process.

Table 1. Summarize of the excess neutron emission.

No.	tp/d	td/h	C	P	m	n (PXm)	EX10^{-24} /d-d/s
1	31.01	2	22	0.00013	939	0.12	6.3
2	62.14	3	27	0.00066	626	0.41	4.5
3	82.17	3	27	0.00066	626	0.41	4.5
Sum of three events		8	76	---	---	0.021	4.9
Total observation		1878 90/d	7437 3.96/h	---	---	--	---

tp:passage time, td:duration time, C:counts of neutron, P:probability, m:sampling number, n:frequency of appearance, E:emission rate.

A statistical analysis of these three events were summarized in table 1. These production rate of the 22 and 27 counts were 6.3X10^{-24} and 4.5X10^{-24}/d-d/s during 2 and 3h, respectively. The confidence levels of neutron emission for 22/2h and 27/3h were 88% and 59%, respectively. For the total of the three trains, the confidence level became 98%.

Since the anomalous neutron emission intermittently occurs and continued a few hours, we will consider that the possible process of the neutron emission phenomena maybe related to the accumulation of stress and the creep mechanism of cathode material.

We would like to express our appreciation to Prof. K.Yamakoshi of The University of Tokyo to a facilities for a usage of the Nokogiriyama underground laboratory. The present study was partially supported by a Fund for Special Research Project at Tokyo Metropolitan University and a Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture of Japan.

4. References

[1] De Ninno, A. et al., 1989, Europhys.Lett., 9, 221.
[2] Harper, J.M.E. et al., 1974, Phys.Lett., 47A, 69.
[3] Burger, J.P.et al.1975, Solid State Commun. 17, 277.
[4] Liu, D.R. et al., 1988, Acta. Metall., 36, 2597.