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NEUTRON SPECTRA AND CONTROLLABILITY BY PdD/ELECTROLYSIS CELL WITH LOW-HIGH CURRENT PULSE OPERATION

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Abstract: Neutron spectra with two components (2.45 and 3-7 MeV) have been repeatedly observed by pulse electrolysis of D_2O-Pd cell. Tritium production with (T/n) ratio 10^5 was also confirmed with low-high current operation. These results are consistently explained with the products and byproducts in competing process of d-d and d-d-d fusions in PdD lattice.

INTRODUCTION

Major results obtained upto now in cold fusion studies can be summarized[1] as follows; a) weak neutron emission, b) tritium production with anomalously large T/n ratio, c) excess heat and d) very poor reproducibility. Trying to solve the puzzle of these anomalous nuclear effects, the authors have proposed a model of competing process between d-d and muliti-body deuteron fusions in PdDx FCC lattice with circumstantial evidences by experiments[2-3]. This paper describes the results of our further experiments and the detailed analysis based on the competing process, so as to solve the mechanism of "cold fusion" and to find a way to control the sporadic phenomena.

MODELING

We are conceiving a competing process of d-d, d-d-d and d-d-d-d fusions in PdDx FCC lattice with high D-loading (x>0.85; when all octahedral sites are occupied with D, x=0.857) under non-stationary conditions like the vibrationally excited state of D-in-lattice[2,4]. Occurrence of this kind of nuclear process seems very hard within known knowledge of physics. However, it could occur in non-stationary condition of PdDx FCC lattice when there existed a mechanism strongly enhancing the

Coulomb barrier penetration for the two-body (d-d) reaction to reach a detectable reaction rate (10^{-22} f/s/cc, for example). The barrier penetration is anyway hard problem yet to be solved, but the super-radient plasma oscillation model by Preparata[5] may be an idea. To explain the present experimental results shown later, we restrict the model to the competing process of d-d and d-d-d fusions. The symbol D denotes deuteron in lattice and the symbol d does free (high energy) deuteron, in the following. Major decay channels and fusion products of D-D-D fusion are as follows:

1) D + D + D → d (15.9 MeV) + ^{4}He (7.9 MeV)

2) D + D + D → t (4.75 MeV) + ^{3}He (4.75 MeV)

The branch 1) may be a major channel because of high Q-value, but we do not know the branching ratio of the branch 2).

Produced charged particles with high kinetic energies generate the following byproducts in their slowing down processes in PdDx lattice:

3) d(15.9 MeV to slow down) + D → n (mainly 3-7 MeV) + ^{3}He ; (n/d) ratio = 10^{-6}
→ t + p ; (t/d) ratio = 10^{-6}

4) ^{4}He(7.9 MeV to slow down) + D → n (0-1.8 MeV) + p + ^{4}He ; (n/^{4}He) ratio $\simeq 10^{-8}$

5) t(4.75 MeV to slow down) + D → n (>14.1 MeV) + ^{4}He ; (n/t) ratio $\simeq 10^{-7}$

When D-D-D fusion becomes predominant[4], the 3-7 MeV neutrons become a major component of neutron yield assuming the branching ratio of the branch 2) to be about 0.1. The overall (T/n) yield ratio would then be about 10^5, and the overall (^{4}He/n) yield ratio would be 10^6. When we observe particle spectra in experiment, there should be monochromatic peaks at 15.9 MeV by deuteron, 7.9 MeV by alpha-particle, 4.75 MeV by triton and 4.75 MeV by ^{3}He. (There would be 23.8 MeV alpha-particle, if D-D-D-D fusion occurred.) To confirm this competing model, spectroscopies of neutron and charged particles and observations of (T/n) and (^{4}He/n) ratios are therefore key issues.

EXPERIMENTAL

Detail of pulse electrolysis and measuring system is written elsewhere[2-4]. In the present experiment, we used a cubic (10x10x10 cm^3) cell with 1000 ml of D_2O + 0.3 mol/l LiOD, 20 mm diam. 30 mm long Pd cathode (cold worked), Pt anode and external cooling channel (spiral glass pipe) of light water flow, contained within an organic glass case with 10 mm thick wall. We tried various function modes for pulse electrolysis; sawtooth, square wave (on-off) and low-high current pulse. For 35 days from the beginning of D-loading, we used sawtooth current (max.=1.4A, min.=0.1A) with 1050 sec period (Exp.79 and Exp.80). For next 6 days, we increased maximum current to 3A (Exp.81). For a next week, we changed repetition periods; 5200 sec (Exp.83, 2 days), 270 sec (Exp.84, 2 days) and 1050 sec (Exp.85, 4 days).

For next 21 days, we tried a low-high current operation with 12 hr period (Exp.86) ; 6 hr low current (0.1A) and 6 hr high current (2.8A). Variations of current, voltage and cell temperature are shown in Fig.1 for typical cycles. For next 9 days, we ran the cell with 1050 sec period sawtooth of 5A maximum current (Exp.87). In the following 8 days, two background runs were made; 1) taking out Pd cathode and stopping electrolysis, and 2) using Pt cathode (instead of Pd) with the sawtooth operation of Exp.87. No differences were found in neutron count rates and spectra by NE213 detector for these two BG runs.

To monitor nuclear products, we measured neutron count rates and spectra by a 5-inch diam. 2-inch thick NE213 liquid scintillation detector with n-gamma pulse shape discrimination. A He-3 detector was also used for relative monitoring. Variation of tritium activity in the cell was monitored by sampling 1 ml of electrolyte every week, chemical processing, solving it in a 10 ml AQUASOL-2 liquid scintillator and measuring beta-activity using standard LSC systems.

NEUTRONS AND THEIR SPECTRA

Evolutions of neutron count rates and spectra were compared with BG count rates and spectra, for Exp.79 through Exp.87. In the beginning period of D-loading (Exp.79), we observed slight excess neutrons (about 20 % over BG level) after 15 days from the start and for 3 days. After this, no excess neutrons were found until Exp.82 which was the start of long-time excess neutron emission. This trend is similar to our previous experiments[2-4]. By changing the period of sawtooth pulse in Exps.83-85, we had largest excess neutrons for 1050 sec period in these three runs. Evolution of neutron counts in Exp.86 is shown in Fig.2 (see also Fig.1). In almost every sweep, higher excess neutrons are given for the high current interval than the low current interval. As the sweep number increases, neutron counts increase for both current intervals. Decrease in neutron counts after the sweep number 35 may be attributed to the contamination of SS-constituents which were dissolved in electrolyte when we inserted a SS-tubed thermocouple at the 35th sweep (electrolyte color changed to brown). This result of Exp.86 may give us a hint for how-to-control "cold fusion"; 1) low-high current pulse operation with long (e.g., 12 hr) period, after sufficient D-loading, is effective, 2) metal ion impurity is poisonous. In Exp.87, we came back to the sawtooth operation with 1050 sec period and observed maximum excess neutrons (1.6 times the BG level in average) in this series of experiments. Unfortunately, the cell was once broken after Exp.87 by accident.

To compare measured neutron spectra with model calculations, we employed the folding technique, instead of the unfolding, because of poorer statistics. Measured pulse height distributions by NE213 were converted to recoil proton spectra using

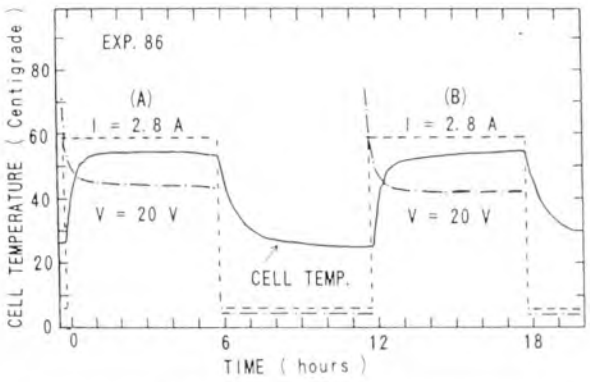


Fig. 1 Low-high current pulse operation for EXP. 86

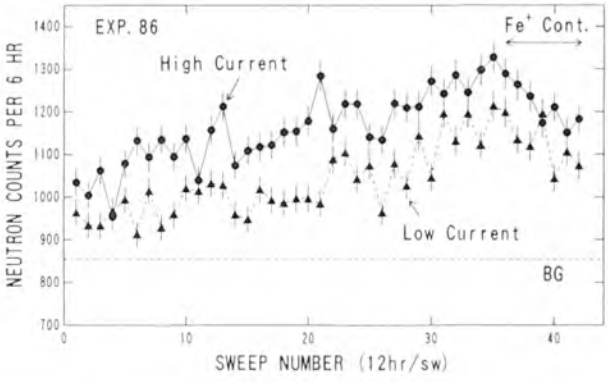


Fig. 2 Evolution of neutron counts in EXP. 86

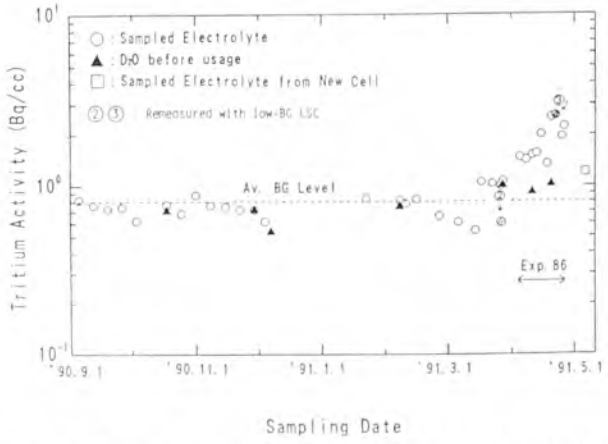


Fig. 5 Evolution of tritium level in electrolyte

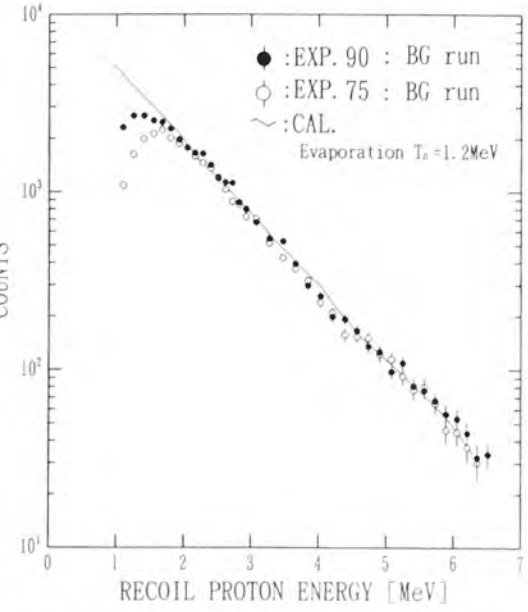
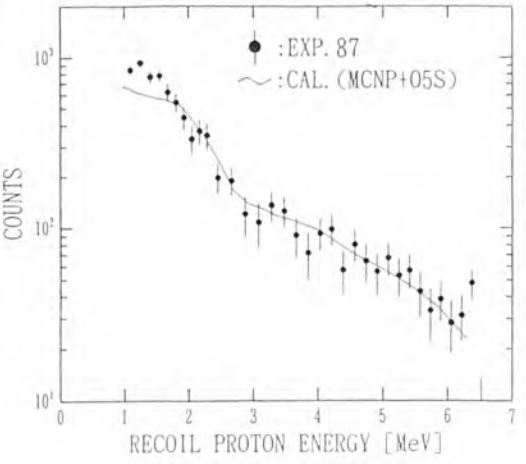


Fig. 3 Background neutron spectra for NE213 recoil proton spectra



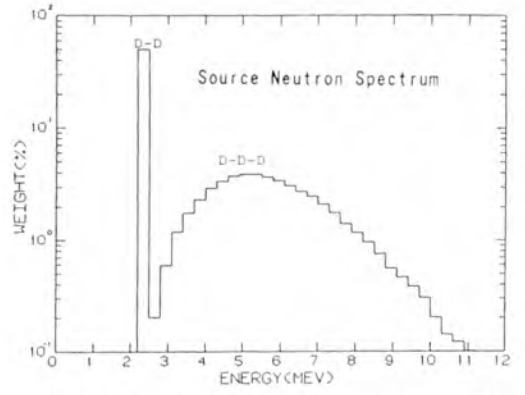


Fig. 4 Upper: NE213 recoil proton spectra for EXP. 87 excess neutron
Lower: Source neutron spectrum used in calculation

experimentally determined light efficiency curve. For model calculations, assumed source neutron spectra were an evaporation spectrum by spallation reaction of cosmic rays for background runs, and a combined spectrum of 2.45 MeV (d-d) and higher energy broad component (see Fig.4 lower curve) (d-d-d). Effect of neutron moderation by the cell, the detector and the polyethylene shield was estimated by MCNP. Response functions of the NE213 detector were calculated by O5S. MCNP and O5S are well known codes in neutron physics. Then, recoil proton spectra were calculated by the folding method. We observed that background spectra were the same all the time, as shown in Fig.3 and agreed well with the calculated evaporation spectrum using nuclear temperature 1.2 MeV. We observed similar excess neutron spectra in Exp.82, Exp.86 and Exp.87, having two energy components. As shown in Fig.4, calculation with the competing model of d-d and d-d-d fusions agreed very well with the measured spectra, except the low energy range less than 1.7 MeV where the deuteron dissociation by 7.9 MeV alpha-particles was not included in calculation.

TRITIUM

Results of tritium measurement are shown in Fig.5 which includes our previous experiments. Activities are constant and in averaged BG level before Exp.86, for about 7 months, and significantly increase in Exp.86. We re-measured some of samples (2, 3 in Fig.5) after about 1 month using a low background LSC system. Measured pulse height distributions of the samples 2 and 3 agreed completely with that of standard tritiated water sample. We found the activity in sample-3 was about 5 times the BG (D_2O before usage) level. We estimated total T-atoms accumulated in the electrolyte to be 4.2×10^{11} t-atoms. We conceive that accumulated tritium in the Pd cathode diffused into electrolyte by the thermal cycle in Exp.86 and therefore the origin of tritium production should be all fusion reactions occurred in the series experiment with the present Pd cathode. Thus, estimation of the corresponding neutron yield is given by integrating excess neutrons in Exp.79 through Exp.86, to be 6.3×10^{6} neutrons. We obtain the (T/n) ratio 6.7×10^{4}. Some of tritium atoms still remain in the Pd cathode and some would escape in gas phase, so that the ratio will be somewhat greater than that value. The competing model of d-d and d-d-d gives 10^{5} with considerable agreement with the experiment.

References: 1) J. Bockris, Fusion Tech., 18, 11-31, (1990), 2) A. Takahashi, et al, Fusion Tech., 19, 380-390, (1991), 3) A. Takahashi, et al., J. Nucl. Sci. Tech., 27, 663-666, (1990), 4) A. Takahashi, et al., "Neutron Spectra from D_2O-Pd Cells with Pulse Electrolysis", Proc. Provo Meet. Anomalous Nuclear Effects in D/Solid Systems, Oct. 1990, Procvo USA, 5) G. Preparata, Fusion Tech., 20, 82-92, (1991)

EDITORIAL NOTE TO THE PAPER "NEUTRON SPECTRA AND ..."
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The decision to publish these Proceedings as soon as possible did not allow any revision of the English presentation of the text originally submitted. This paper contains a few misprints and improprieties, for which we apologize with the readers.