Limit on Fast Neutrons from DD Fusion in Deuterized Pd by Means of Ge Detector

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Abstract

Search for fast neutrons from the electrochemically loaded Pd-D system at room temperature was made in order to study the possible d+d fusion there. A low-background high-resolution Ge detector surrounded by neutron scatterers was used to investigate the fast(1∼5 MeV) neutrons. The neutron flux was obtained by measuring yields of the γ-rays following inelastic scattering of the fast neutrons from nuclei in the scatterers. The observed spectrum shows no statistically significant excess of the γ-rays above background. The upper limit on the fusion rate was obtained as $\lambda_f < 1.6 \cdot 10^{-24}$ (ddn)fusions / [(dd pair) sec].

1. Introduction

Nuclear fusions such as d+d, d+t and others are strongly inhibitted at room temperature by the Coulomb barrier because the relative kinetic energy (temperature) is far below the barrier. The nuclear fusion at room temperature, however, might be much enhanced if the effective Coulomb barrier could be reduced slightly by possible modification of electric fields in the solid(metal) state[1-3]. This is the so-called cold fusion, in contrast to the hot fusion at high temperature. The hot fusion has been well established experimentally and theoretically since many decades in nuclear physics, and since many years in plasma physics. On the other hand there are no strong experimental evidences for the cold fusion so far.

It is crucial for experimental studies of the nuclear fusion to investigate the nuclear reaction products such as n, p, ^{3}He, and ^{3}H. Observation of just the heat (energy) excess in the macroscopic system does not prove the nuclear fusion at all since many chemical and solide-state contributions are hardly excluded as possible origines of the heat.

Many experimental surveys for the possible cold fusions have appeared, some supporting[4-8] and others disproving[9-15] the finite fusion at the room or cold temperature. Among them, search for the neutrons is one of most straightforward ways to prove the nuclear fusion since they arises only from nuclear reactions. Experimental studies of the neutron, however, are difficult because it has no charge. Recently we have suceeded in developing a new type of the high-sensitive detector for fast neutrons. It is very important to investigate experimentally the limit on the d+d fusion rate by using the new technique.

The present work reports the search for the fast neutrons from electrochemically loaded Pd-D system at the room temperature by means of the new detector. The unique point is to use a high energy-resolution Ge detector to measure the fast neutrons. The principle is to measure the energy spectrum of γ-rays following inelastic scattering of the fast neutrons. The detector is sensitive to only the fast neutrons, and the γ-ray shows up as discrete peaks, corresponding to the nuclear excitation energies, in the continuum background spectrum. Consequently the signal can be clearly identified. The details of the detector has been given elsewhere[16].

2. Electrochemistry for the Pd-D system

The Pd-D system was made electrochemically by using Pd and Pt electrodes in the electrolysis of D_2O. The electrochemical cell was a cylindrical polyethylene bottle of 70mm in height and 60mm in diameter. The cathode was a massive palladium plate, 50mm wide by 50mm long, with a thickness of 2mm. The total mass of this electrode is 59.04g. The anode was made of two sheets of platinium samples, 50mm wide by 50mm long, with thickness of 0.2mm and the spacing of cathode-anode is 10mm. The two platinium sheets were mounted parallel to each other in one electrolytic cell, as shown in the insert of Fig. 1. The platinium electrodes used in the cell were spot-welded to a 0.5 mm diameter platinium wire.

The electrolyte was 99.9% pure heavy water(D_2O) with 0.1M LiCl. The electrolytic cell holded about 176.34g of the electrolyte. The top was closed except for two holes with 3mm diameter to release gas pressure. The electrolysis was operated in a constant current of 0.7A. The applied electric voltage was about 8V. We measured the weight of the Pd plate as a function of time during the electrolysis. The weight increased gradually and finally saturated at the fixed weight as shown in Fig. 2. The increase of the weight gives the total number of the deuterium atoms in the palladium plate.

3. Neutron measurements

In the present experiment use was made of the low- background Ge detector system ELEGANTS III(s)[16]. The detector system ELEGANTS III(s), together with the Pd-D electrochemical system, is shown in Fig. 1. The Ge detector used is the low-background 171cm^3 pure Ge detector, which has been primarily used as a central detector of ELEGANTS III[17,18] for measuring rare double beta decays of ^{76}Ge. The Ge detector is surrounded by 10cm thick OFHC (oxygen free high conductive copper) and 10cm thick lead shield. OFHC is known to be quite free from radioactive contamination, namely below the observable limit of 0.2ppb. The whole system is covered by active shields of 15mm thick plastic scintillators to reject charged cosmic rays. The electrolytic cell was placed in front of the Ge detector. The 16mm thick Fe plate, as the fast neuton scatterer, was inserted between the detector and the front of the cell, and the other Fe plate was also place at the backside of the cell(see Fig. 1). The ^{65}Cu and ^{63}Cu isotopes in the OFHC shield and the ^{74}Ge and ^{72}Ge isotopes in the detector itself were also used as the scatterers of the fast neutrons.

The efficiency of the neutron counter was measured by placing a ^{252}Cf source at the position of the cell. The ^{252}Cf source provided fission neutrons with intensity of $I_n(E_n)$=104 per sec and with the averagy energy of $E_n \sim 2.5$ MeV[16], which is

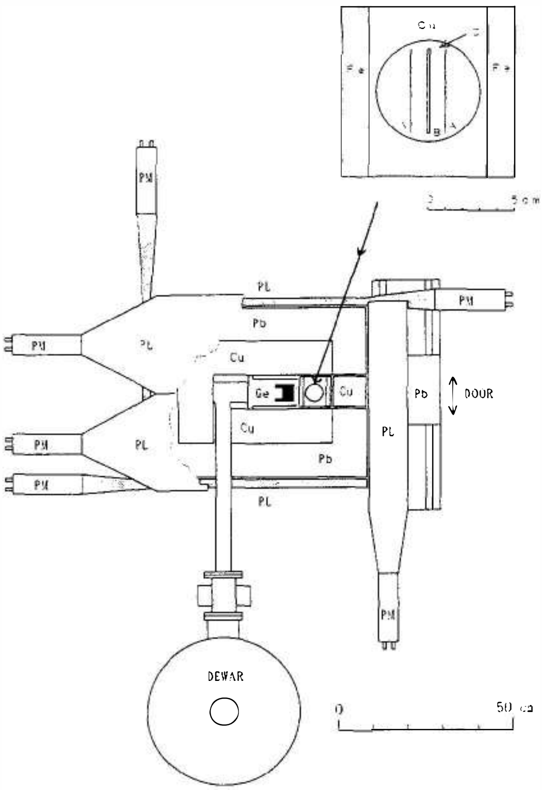

Figure 1. Top view of ELEGANTSIIIs
Ge, PL, PM, Cu, and Pb are the Ge-detector, the plastic scintillators, the photomultipliers, the copper shield(scatterer) bricks, and the lead shield bricks, respectively. The insert shows the enlarged view of the electrolytic cell(C) with the front and back iron scatterers(Fe). A and B in the cell are the Pt anode sheets and the Pd cathode plate, respectively. The enlarged view of the electrochemical cell is show in the upper part

Table1. Limits on cold fusion rates in Pd

Scatterer	γ-energy (keV)	Efficiency (x10^{-4})	Rate (dd fusion) (n/dd sec)
^{56}Fe	846.8	3.3±0.2	< 2.4 x 10^{-24}
^{74}Ge	595.8	2.9±0.3	< 3.2 x 10^{-24}
^{72}Ge	691	1.6±0.3	< 5.0 x 10^{-24}
^{63}Cu	962.1	0.7±0.1	< 6.4 x 10^{-24}
	669.6	1.0±0.1	< 5.1 x 10^{-24}
^{65}Cu	1115.9	0.3±0.1	< 1.1 x 10^{-23}

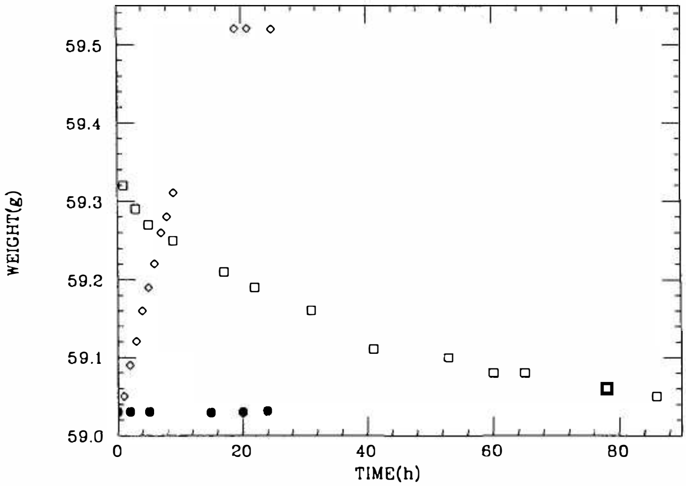

Figure 2. The weight of Pd plate as a function of time during electrolysis. Closed circles show no increase of the weight if the current is off, diamonds show increase of the weight from the begining of electrolysis and the increase of the weight saturates at the fixed weight, and squares show the decreasing of the weight after the current off.

same as the neutron energy from the d+d fusion. A 15mm thick lead plate was inserted between the ^{252}Cf source and the Ge detector to absorb γ-rays from the ^{252}Cf source. Fig. 3(A) shows the energy spectrum of γ-rays following inelastic scattering of neutrons from the ^{252}Cf source. Overall efficiencies were obtained as $k(\gamma_a)=I(\gamma_a)/I_n(E_n)$, where $I(\gamma_a)$ is the peak yield in unit time interval of t=1. The obtained values are given in Table 1. The efficiencies $k(\gamma_a)$ are of the order of 10^{-4}. In order to estimate the minimum neutron flux $S_n(E_n)$ to be detected by the present neutron counter we require the condition that the γ-ray peak yield $Y_\gamma(E_n)=k(\gamma_a)\cdot S_n(E_n)\cdot t$ in a time interval t exceeds the fluctuation of the background $I_\gamma(BG)\cdot t$ in the γ-ray spectrum. The minimum neutron flux to be measured is $S(E_n)=\sqrt{I_\gamma(BG)t}/k(\gamma_a)t$. For the 847 keV γ-ray following the first 2^+ state in ^{56}Fe, $I_\gamma(BG)$ is about 5.5×10^{-5} cps in the γ-ray peak region. The minimum neutron flux to be detected is given by $S_n(E_n)=24.7/\sqrt{t}$ with S_n and t being given in unit of sec. It is $3.2\cdot10^{-2}$ neutrons/sec for $t=6.0\times10^5$sec.

4. Results

The measurement was made for 471 hours of the electrolysis. The average number of the deuterons loaded in the Pd metal was obtained as $0.6\times3.45\times10^{23}$ from the measured excess of the Pd weight. The measured γ-ray spectrum is shown in Fig. 3(C). Before the electrolysis, we also measured the background spectrum from natural radioactivities in the detector system including the scatterers and the electrolytic cell, as shown in Fig. 3(B). There are no significant excess of counts due to the electrolysis at the peak positions of the γ-rays expected from the inelastic neutron scattering.

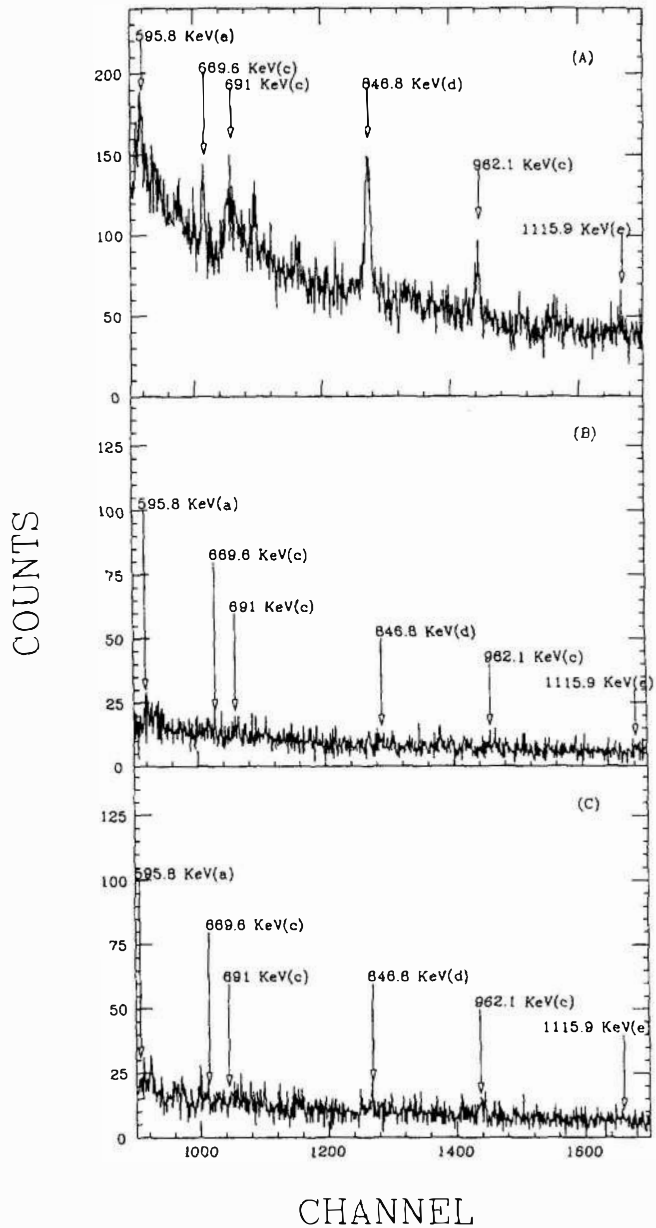

Figure 3. (A):Gamma-ray energy spectrum for the detection efficiency measurement by using the ^{252}Cf neutron source, (B):the spectrum observed before the electrolysis and (C):the energy spectrum observed during the electrolysis. The peaks a, b, c, d and e in the spectrum are the γ-rays from excited states in ^{74}Ge, ^{72}Ge, ^{63}Cu, ^{56}Fe, and ^{65}Cu, respectively.

The reduced upper limits on the fusion rate are summarized in Table 1. Combining all data listed in Table 1, the upper limit on the fusion rate is obtained as $1.6 \cdot 10^{-24}$ (ddn) fusions / [(dd pair) sec].

5. Summary & Discussions

Search for neutrons from any possible deuteron-deuteron fusion during electrolysis was made by using a low background Ge detector. It is emphasized that the present method looking for sharp γ-ray peaks is sensitive to only fast neutrons and thus useful for separating the signal from the continuum background(noise). The upper limit of $1.6 \cdot 10^{-24}$ (ddn) fusions / [(dd pair) sec] is derived. It is one of the severest limits among the recently published results. In fact there are several works which claim finite d+d fusions at the room(cold) temperature. Most of them, however, do not detect directly the fast neutrons nor the neutron energy. Since detection of low-intensity neutrons are rather hard, it is essential to use simultaneously various types of detectors for measuring the neutron energy at several different positions. The present detector, which is a very low-background/noise system sensitive to the fast neutrons from the d+d fusion, is strongly reccommended to be used for checking possible d+d fusions if any.

Acknowledgement

The authors are indebted to Dr. T. Saito for providing them with the ^{252}Cf source. They thank Dr. M. Fujiwara, Mr. K. Matsuoka, Mr. K. Kume and members of ORIS (Osaka university RI center) for kind helps and valuable discussions. The present work is supported by the Grant in Aid of Scientific Research, Ministry of Education, Science and Culture, Japan.

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