Nuclear Physics Approach

Search for Anomalous Nuclear Reactions in PdDx by Detection of Nuclear Products in Vacuum/Gas System

M.Taniguchi, N.Kaji*, A.Takahashi
Department of Nuclear Engineering, Osaka University
2-1 Yamadaoka, Suita, Osaka, Japan.
*Present address, PNC, Tokai, Japan.

Abstract

To detect charged particles from deuterated palladium for the direct evidence of anomalous nuclear reactions in solids, the heat-and-gas-release experiments have been performed. No very clear data for charged particle emission, neutron emission and helium-4 production have been obtained until now. The cause of mass-5 peak increase was discussed.

Introduction

It has past seven and a half years from the announcement of the cold fusion phenomena by Fleischmann and Pons.[1] In past years, anomalous results of helium-4 detection and mass-5 breeding with high resolution mass spectrometers have been reported by some researchers[2-5], but the phenomena show very poor reproducibility and we have had only few distinct data. Therefore, our group has started the research to obtain very clear data showing the existence of nuclear products that are supposed to be directly correlated to excess heat phenomena.

To detect the charged particles from deuterated palladium samples for the direct evidence of anomalous nuclear reactions in solids, an experimental system of vacuum chamber was designed and made. We can measure charged particles and neutrons and make mass spectrum analysis of released gas from the deuterated palladium in the vacuum chamber. The chamber is equipped with a silicon surface barrier detector for charged particle spectroscopy, an NE213 scintillation counter for fast neutron spectroscopy and a high-resolution quadrupole mass spectrometer for gas analysis. Mass-5 breeding has been reported by Itoh et al.[5] Mass-5 is supposed to be either DT^+ or DDH^+molecule and has been reported that mass-5 breeding would be thought as the tritium increase in released gas with heating deuterated palladium metals. In this work we have performed the mass-1 analysis in addition to the mass-5 analysis to make sure whether the "mass-5 breeding" is due to the DDH^+ increase by the increase of hydrogen partial pressure, or not. Two-type experiments have been performed to try to induce anomalous nuclear effects. The first one is the heating experiment of the deuterated palladium plates that were deuterated by electrolysis. In correlation with the deuterium gas release from a palladium plate with relatively high D/Pd ratio, some anomalous effects would be expected. The second experiment is the thermal cycle experiment. Palladium plates or powder samples in a vacuum chamber that was filled with deuterium gas were loaded with deuterium by thermal cycle condition, so that palladium samples absorbed and discharged deuterium repeatedly. This repeating condition was advantageous for the mass spectrum analysis of released gas.

Experimental

Figure 1 shows experimental apparatus. Deuterated palladium plate(12.5x25x1 or 25x25x1mm) were prepared as follows, for the heating experiment. Palladium plate was washed with acetone for removing organic impurities that sticked on the palladium plate surface, and some of the samples were set up in vacuum chamber and annealed with pressure less than 0.13 Pa at 900 ℃ for several hours. The palladium plate was set in the F.P. type open cell which was filled with 0.2M, LiOD/D_2O and then electrolyzed with current 1～3A over 6 hours. We determined the loading ratio of sample from the change of weight, the ratio reached around the range of D/Pd=0.74 ～0.91. We used Pd-Rh(10%) and Pd-B(0.2%) alloys as well as pure palladium plates, because Pd-Rh alloy is reported to be advantageous for attaining high loading ratio. The average of loading ratio was 0.89. Palladium plate that contains boron may be effective to produce excess heat[6] and then excess heat may be associated with nuclear reactions. Some of palladium samples were plated with copper layer of about 1 μ m thickness by electrolysis with $CuSO_4/H_2O$ solution, because copper film is expected to work as blocking layer for deuterium diffusion. Deuterated palladium sample was then set up in the vacuum chamber and the chamber was evacuated to be less than 0.13 Pa. Deuterium gas was released from the deuterated palladium sample by heating and stored in the vacuum chamber for gas analysis. The chamber was equipped with a silicon surface barrier detector(Si-SSD) which was surrounded by a copper cylinder which equipped coolant water passage for the heat protection of the Si-SSD during charged particle spectroscopy. The Si-SSD and NE213 detectors were operated to collect data while a palladium sample was releasing deuterium gas.

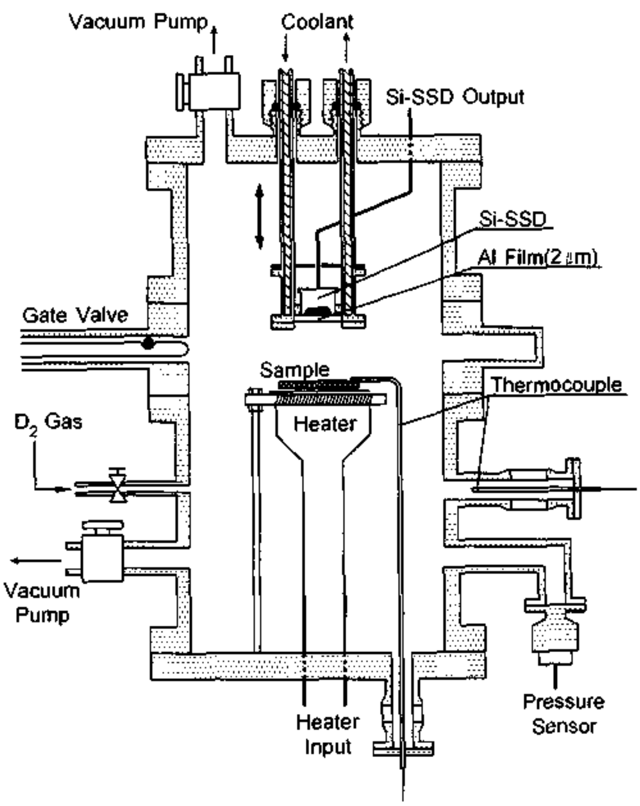

Figure 1 Experimental Apparatus

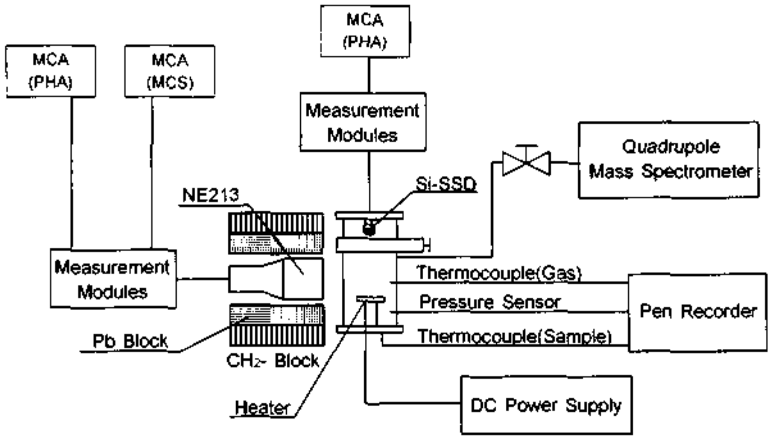

Figure 2 Experimental System

Figure 2 shows the experimental system. The output signal of the NE213 detector was led to the n-γ pulse shape discrimination circuit to reject γ-ray signals from neutron signals. The temperature of palladium sample was monitored by a CA-type thermocouple.

Palladium samples for the thermal cycle experiment were prepared as follows. In the present experiments, we used palladium plates and powder samples. Palladium powder has wide surface area, compared to its weight, and it is advantageous for gas loading. Palladium plate was washed with acetone to remove organic impurities that sticked on the palladium plate surface. Palladium plate or powder sample was set up in the vacuum chamber, which was then evacuated to 0.13 Pa and annealed at about 500 ℃ for several hours. Deuterium gas was introduced into the chamber until the pressure became about 2 atm, and applied the cyclic heating with 24 hours period for several times after keeping the sample under deuterium gas for a couple of days, and the gas was analyzed by a quadrupole mass spectrometer. During the thermal cycles, the NE213 detector was operated for neutron measurement, and then the chamber was evacuated and the sample was heated for gas release with monitoring charged particle emission.

Results and Discussion

Figure 3 shows an experimental result of mass-5 breeding ratios by the heating experiment, comparing with that by the gas loading experiment, where mass-5 breeding ratio is defined as the ratio of mass-5 peak amplitude for the sample and that of original deuterium gas. Mass-5 obviously increased and the average was 7 for the heating

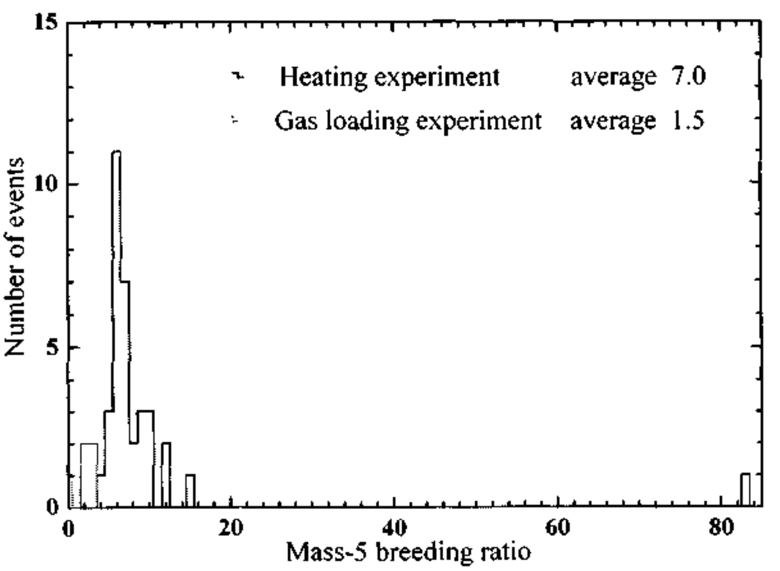

Figure 3 Events distribution for mass-5 breeding ratios

Nuclear Physics Approach

	mass-5 increse	mass-1 increse
Exp. 1-41	6.8	7.5
Exp. 1-42	5.4	12.3
Exp. 1-43	14.1	7.6

Table 1 Mass-5 and mass-1 increse

experiment and 1.5 for the gas loading experiment. Only one event by the heating experiment showed extremely high breeding ratio(82.5). Mass-5 ion is supposed to be either DT^+ or DDH^+ molecule, so that increase of mass-5 peak can be attributed to the increase of either tritium or hydrogen partial pressure in the released gas of which content is predominantly deuterium gas. The present quadrupole mass spectrometer does not have enough resolution for distinguishing the DT(5.030amu) peak from the DDH(5.036amu) peak. With the present experimental system, it was difficult to confirm the increase of tritium. Instead, we analysed the amplitude change of mass-1 peak by the quadrupole mass spectrometer to monitor the hydrogen partial pressure. Table 1 shows the ratios of mass-5 and mass-1 ion current peaks for the sample relative to the peak amplitude for the case of original deuterium gas. Not only mass-5 peaks but also mass-1 peaks increased, but amounts of these increases show no obvious correlation between mass-5 and mass-1. We analyzed only for 3 cases for the mass-1 peaks together with mass-5 peaks, so that quantitative estimation is not properly made, except pointing out that the mass-1 increase is one of the causes for the mass-5 increase. The reason of the hydrogen partial pressure increase is probably due to H impurity in D_2O(99.9 at%) or D_2 gas(99.9 at%). However, one event showed extremely high mass-5 increase (82.5 times) and the reason is not yet clarified.

There was no such data that exceeded the lowest detection limit of the quadrupole mass spectrometer for the helium-4 production possibility throughout these experiments.

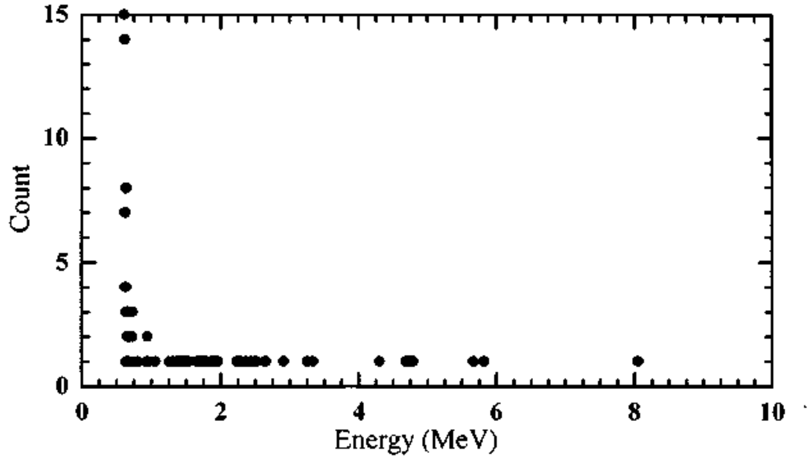

Figure 4 Si-SSD spectrum (Exp.1-33, Live time 3,580sec)

In the present experiments, charged particles which we would regard emitted from deuterated palladium samples have never been detected clearly, but some experiments showed several counts up to ~10 MeV. Because of poor statistics of counts, we can not identify whether these high energy counts were from noise signals or true signals originated from charged particles. (See Figure 4, for example)

Figure 5 shows an example of experimental results on neutron measurement for the gas loading experiment. Several other-experiments showed few neutron events slightly over 3 σ levels. In Figure 5, there are data points that exceed 3 times (broken line in Figure 5, where solid line shows average level) the standard deviation σ of the background fluctuation. However, we cannot find meaningful structure change of recoil proton spectrum of the NE213 detector since the statistics was not enough. Background neutron level was comparatively high throughout the experiments and fluctuations were so big that the increase of neutron events might be attributed to the change of background level.

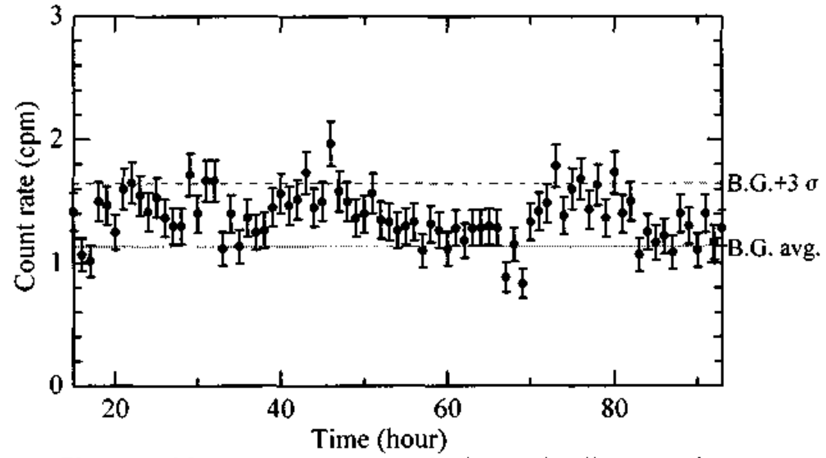

Figure 5 Neutron measurement in the gas loading experiment with Pd plate sample (Exp. 2-02)

Conclusions

We have performed the heating experiments of deuterated palladium metal samples in vacuum chamber. The deuterations of palladium were prepared by the electrolysis or the gas loading method. In the heating experiments with electrolyzed palladium samples, we observed increase of mass-5 breeding ratios, but the increase of hydrogen partial pressures was suspected to be one cause for this event. We also observed several counts of unusual energies in 3~10 MeV region of charged particle spectrum, but we could not conclude that these signals originated from charged particles, due to poor statistics. In the gas loading experiments, we observed neutron increase events but with so marginal levels that we could not confirm that these events indicated true neutrons from deuterated palladium samples. We shall run more experiments and seek for the causes of these "anomalous" events in the future efforts.

Reference

1. M.Fleishmann, S.Pons : J.Electroanal.Chem.,261(1989) P.301
2. E.Yamaguchi and T. Nishioka : Proc. ICCF-3, Nagoya, (1993) P.179
3. T.N.Claytor, et al. : Proc. ICCF-3 , Nagoya, (1993) P.217
4. D.G.Tuggle, et al. : Proc. ICCF-4 , Lahaina, EPRI TR-104188-V1, 7 (1994)
5. T.Itoh, et al. : Proc. ICCF-5, Monte Carlo, (1995) P.189
6. M.Mckubre, et al. : Proc. ICCF-5, Monte Carlo, (1995) P.17