

## **Nuclear Physics Approach**

### INVESTIGATION OF NUCLEAR EMISSIONS IN THE PROCESS OF D(H) ESCAPING FROM DEUTERIZED (HYDROGENIZED) PdO-Pd-PdO AND PdO-Pd-Ag SAMPLES.

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#### ABSTRACT

Emission of proton-like and neutron-like events was observed in the process of escaping deuterium from deuterized PdO-Pd-Ag and PdO-Pd-PdO samples. The ratio of the proton-like and neutron-like event fluxes was estimated as  $N_p/N_n \approx 1$ . The charged particle emission was also observed in the process of escaping hydrogen from hydrogenized PdO-Pd-Ag and PdO-Pd-PdO samples. The emitted charged particles may be identified as protons and  $\alpha$ -particles. Investigation of charged-particle emission was carried out by 3 methods: (1) plastic scintillation counter; (2) Si-SSD; (3) CR-39 plastic track detector. The results obtained by these independent methods are in good agreement with each other. An effect of the weak thermal neutron flux on the processes of cold fusion in the samples loaded with D(H) was also investigated. It was observed that the flux of neutrons emitted from deuterized PdO-Pd-Ag samples exposed by thermal neutrons was approximately 300 times as large as in the case of unexposed samples.

#### 1. INTRODUCTION

As was shown in /1/, heat bursts and neutron emission take place as a result of escape of deuterium from electrolytically deuterized PdO-Pd-Au samples. This effect was reproduced in many series of experiments performed by dr. A. Lipson's group from the Institute of Physical Chemistry, Russian Academy of Sciences. The PdO-Pd-Ag and PdO-Pd-PdO samples demonstrate the same properties as PdO-Pd-Au ones. These are the samples that were used by us for investigating nuclear emission. The experimental task was to study channels of nuclear reactions that take place in Pd samples electrolytically loaded with D(H).

The PdO-Pd-Ag and PdO-Pd-PdO samples were prepared by vacuum annealing of the Pd foil 30  $\mu$ m thick in the Institute of Physical Chemistry of the RAS. The PdO-layer 200 Å thick was produced by annealing in the oxygen while the Ag-layer was created by deposition in the electrolyte. The sample area was 21 x 16 mm<sup>2</sup>. The technology of the sample preparation was described in /1/.

#### 2. METHODS

The neutron detector consists of 8 scintillation counters. Each counter contains of a plastic scintillator 5x5x30 cm<sup>3</sup> covered with the 0.5 mm-thick Cd foil and is supplied by 2 end-view FEU-85 photomultiplier tubes (PMT) connected to the coincidence circuit (Fig.1).<sup>3</sup> The detector was surrounded by CH moderator blocks (5 x 5 x 30) cm<sup>3</sup> covered with the Cd foil. A fast neutron emitted as a result of the nuclear reaction in the sample can interact in 1 of

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8 scintillation counters. Then it spends its energy in collisions with atoms of C and H of the scintillator and moderator and is slowing down to thermal energy  $E = 0.026$  eV. At the final stage, the thermal neutron is captured by Cd with emission of gamma-quanta that also can be detected by the same counters. The efficiency of the neutron detector measured with the neutron (Po-Be) source placed in the central detector cell was  $\eta \approx 1\%$ . The measured background rate was  $N_b = (0.050 \pm 0.005)$  1/s. The charged-particle emission was investigated by 3 methods: (1) Plastic scintillation detector; (2) Si-SSD; (3) CR-39 plastic track detector.

The plastic scintillation detector is 1 mm thick and 30 mm in diameter and has a photomultiplier (PMT). A sample was placed at a distance of 1.5 cm from the scintillator (Fig. 2). There was Al foil 12  $\mu\text{m}$  thick between the sample and the scintillator to prevent the effect of light on the PMT. The detector was calibrated by  $\alpha$ -particles from Pu-239 source and by secondary protons from interactions of neutrons of (Po-Be) source with scintillator nuclei. Si-SSD with the active area of 1.25 cm and the active thickness of 100  $\mu\text{m}$  was screened by the 12  $\mu\text{m}$  Al-foil. The distance between the sample and the detector was 3 mm. The detector was calibrated by  $\alpha$ -particles of the Pu-239 source.

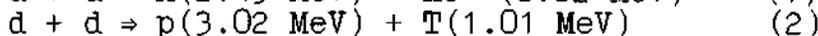
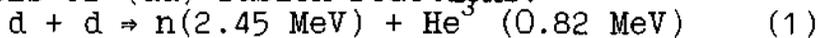
The track detector was the 600  $\mu\text{m}$ -thick plastic film. Charged particles stopping in outer detector layer produce tracks that became visible after etching. The 6N NaOH solution at a temperature  $t = 70^\circ\text{C}$  was used for etching for 7 hours. The track shape presents information on the type of the particle and its energy. For calibration, the CR-39 film was exposed to  $\alpha$ -particles and protons of known energies.

In the process of electrolysis of the 1M solution of NaOD in  $\text{D}_2\text{O}$  or KOH in  $\text{H}_2\text{O}$ , the PdO-Pd-Ag and PdO-Pd-PdO samples were used as cathodes. The Pt was taken as an anode. The current density was  $15 \pm 20$  mA/cm<sup>2</sup>. The duration of the electrolysis was  $5 \pm 15$  min. The sample loaded with D(H) was washed in pure  $\text{D}_2\text{O}(\text{H}_2\text{O})$  and dried by the filter paper. Then, it was placed near one of the detectors mentioned above. The measurement time was about 1 hour. Furthermore, the sample was loaded with D(H) again. Each sample was used for 15-20 cycles of electrolysis and then was changed.

### 3. RESULTS

#### 3.1. Measurements using neutron detector and plastic scintillation detectors.

The experimental task was to estimate the yields of two possible channels of (dd) fusion reactions:



The PdO-Pd-PdO sample was loaded with D(H) according to the procedure mentioned above and then was placed at the distance 1.5 cm from the scintillator, that was screened by the 12  $\mu\text{m}$  Al foil. (Fig. 2). To stimulate the escaping gas from the sample, it was heated up to  $40^\circ\text{C}$  by the hot air during the first 300 s of the experiment. The controlling experiment was carried out with an unloaded sample during the same time and under the same conditions. As a result of the experiments, it was found that the spectra of emission from the deuterized, hydrogenized and unloaded samples which were measured by the scintillation detectors

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differed from each other. (See Figs. 3a, 3b). The detector lower energy threshold corresponded to proton energy  $E_p \approx 0.5$  MeV.

To estimate the yield of reaction (2), the number of events with the energy 0.5–2.5 MeV for the hydrogenized sample was subtracted from that for the deuterized sample. Thus, the proton flux from reaction (2) was estimated as  $N_p = (0.75 \pm 0.08)$  1/s into  $4\pi$  steradian. The neutron emission was also studied. However, it did not differ from the background level  $N_b = (0.050 \pm 0.005)$  1/s. If we take into account the neutron detector efficiency  $\eta = 1\%$ , the minimum neutron flux accessible to registration was on the level 5 n/c into  $4\pi$  steradian. Therefore, another method was used to estimate the neutron flux from reaction (1). It was assumed that a neutron from reaction (1) can, at first, interact in the thin plastic scintillator. Then the second interaction takes place in the scintillators of the neutron detector. The neutron flux can be estimated as a number of such double coincidences. The detection efficiency was estimated as  $\approx 4 \cdot 10^{-4}$ . The difference between spectra of deuterized and hydrogenized samples in the case of signal coincidence from 2 detectors is presented in Fig. 3c. The neutron flux was estimated by number of events in the energy range 0.5–2.45 MeV as  $N_n = (0.6 \pm 0.2)$  1/s into  $4\pi$  steradian. Thus, we may conclude that the ratio of neutron and proton fluxes from reactions (1) and (2) is  $N_n/N_p \approx 1$ .

### 3.2. Measurements with Si-SSD.

To verify the results obtained, the measurements with Si-SSD were carried out. A PdO-Pd-Ag or PdO-Pd-PdO sample was loaded with D(H) according to the procedure mentioned above. Then, the loaded sample was placed near the Si-SSD. The charged-particle spectra obtained with different samples are presented in Figs. 4a, b. In this case, the measurement with the same sample in 1 hour after the first experiment (when the main part of D(H) escaped the sample) was used for control. The spectrum of the charged particles detected by Si-SSD is in good agreement with the spectrum for the same samples obtained with plastic scintillation detector. The disadvantage of both methods was impossibility to determine the type of charged particles. Therefore, measurements using CR-39 plastic track detector were performed, which allowed us to obtain information on the type of the emitted particles.

### 3.3. Measurements with CR-39 plastic track detector.

A sample was electrolytically loaded with D(H). Then the PdO side of the sample was kept in contact with the 1 half of the track detector. The unloaded sample was kept in contact with the other one-half of the track detector. In both cases the time of contacting was 0.5–1 hour. In total, the detector was exposed for 15 hours (15–30 cycles of sample loading). Then it was etched in 6N solution of NaOH at 70° C during 7 hours. The results of measurements with using a microscope are presented histograms (Figs. 5a,b). They show the number of tracks per  $\text{cm}^2$  in a certain range of track diameters. The background was estimated as a number of tracks per second from the unloaded sample. The effect-to-background ratio was found to be 2–4 depending on the type of a particle. Particles were identified by calibration measurements, showing that track diameters less than 8  $\mu\text{m}$  corresponded to

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protons, while track diameters of  $8+15 \mu\text{m}$  corresponded to  $\alpha$ -particles. Track diameters exceeding  $15 \mu\text{m}$  correspond to heavier charged particles (Li, Be-nuclei). Thus, it is possible to conclude that the samples were emitting protons.  $\alpha$ -particle emission is also possible. According to /3/,  $\alpha$ -particles may result from fusion reaction of 3 and more particles (p or d). The events with track diameters more than  $15 \mu\text{m}$  were investigated separately. The density of such events for deuterized PdO-Pd-Ag sample was  $(66 \pm 11) 1/\text{cm}^2$  against the background of  $(8 \pm 3) 1/\text{cm}^2$ . It seems probably that this effect was caused by emission of heavier particles (Li or Be nuclei).

### 3.4. Effect of thermal neutrons on particle emission from deuterized PdO-Pd-Ag samples.

As was shown in /2/, neutron emission caused by (dd) fusion reaction in the  $\text{KD}_2\text{PO}_4$ -crystals is enhanced by an influence of weak thermal neutron flux. We made an attempt to check a possibility of increasing the fusion reaction yield for PdO-Pd-Ag samples electrolytically loaded with D(H) by exposing them with thermal neutron flux. Cf-252 was used as a neutron source  $E_n \approx 2.3$

MeV. Its intensity was  $A \approx 180 \text{ n/s}$  into  $4\pi$  steradian. The source placed in the Pb container was installed into the central cell of the neutron detector. There was a (CH) moderator between the neutron source and the sample as well as behind the sample. The PdO-Pd-Ag sample was electrolytically loaded with D(H). The PdO side of the loaded sample was in contact with the CR-39 track detector. The control CR-39 detector was in contact with the unloaded sample. Then the assembly was placed at a variable distance  $r$  from the Cf-252 source. The efficiency of the neutron detector with the central cell filled with CH was  $\eta \approx 2 \cdot 10^{-4}$ . The intensity of background in the presence of the Cf-252 source was  $N_b = (0.085 \pm 0.005) 1/\text{s}$ . The

dependence of the fast neutron flux emitted by deuterized PdO-Pd-Ag sample as a function of the distance  $r$  between the sample and the source is presented in Fig 6. In contrast to the neutron flux from the unexposed sample which was estimated as  $N_n = (0.6 \pm 0.2)$

$\text{n/s}$  into  $4\pi$  steradian (3.1), the neutron flux from the exposed sample attained  $N_n = (200 \pm 20) \text{ n/s}$  into  $4\pi$  steradian. Thus, the fast neutron flux from the deuterized sample increases by a factor of  $3 \cdot 10^2$  in the presence of the external thermal-neutron irradiation. The track density for protons in CR-39 was estimated as  $4 \cdot 10^5 1/\text{cm}^2$ . It is  $\sim 10$  times more than the case of unexposed sample. The estimated proton flux was  $N_p = (160 \pm 20) 1/\text{s}$  into  $4\pi$  sr. So the ratio of proton and neutron fluxes in the case of thermal neutron exposed sample was  $N_p/N_n \approx 1$ .

### 4. CONCLUSION

As the result of the experiments, we may make the following conclusions.

(1) Emission of proton-like and neutron-like events was observed in the process of escaping D from deuterized PdO-Pd-Pd and PdO-Pd-Ag samples. The ratio of the fluxes of the proton-like and neutron-like events is  $N_n/N_p \approx 1$ . It is possible to conclude that the fusion reactions (1) and (2) take place in these samples.

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(2) Charged-particle emission was observed in the process of escaping H from hydrogenized PdO-Pd-Ag and PdO-Pd-PdO samples. These particles may be identified as protons and  $\alpha$ -particles. We failed to determine the nature of this emission. The emission of neutron-like events was not observed in the case of hydrogenized samples.

(3) We observed an effect of the weak thermal neutron flux on the emission from the deuterized PdO-Pd-Ag samples. The gain factor for the fast neutron emission  $3 \cdot 10^2$  as compared to the unexposed samples. Possible explanations for the mechanism of this effect was proposed by Kozima /4/ and Hagelstein /5/.

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### REFERENCES

1. A.G. Lipson, B.F. Lyakhov, B.V. Deryagin, and D.M. Sakov, Letters to JTP, 1992, v. 18, 20, p. 58 (in Russian); B.F. Lyakhov, A.G. Lipson and D.M. Sakov, Proc. of the 1-st Russian Conf. on CF (Sept.28 - Oct.2, 1993, Abrau-Durso, Novorossiisk) p.154 (in Russian).
2. A.G. Lipson and D.M. Sakov, Proc. of 5-th ICCF (April 9-13, 1995, Monte-Carlo, Monaco) p. 571.
3. A. Takahashi et al., Proc. of the 1-st Russian Conf. on CF (Sept.28 - Oct.2, 1993, Abrau-Durso, Novorossiisk) p. 76
4. H. Kozima and S. Watanabe, Proc. of 5-th ICCF (April 9-13, 1995, Monte-Carlo, Monaco) p. 347.
5. P.L. Hagelstein, Trans. Fusion Tech.; 1994, v. 26, No 4T, 461

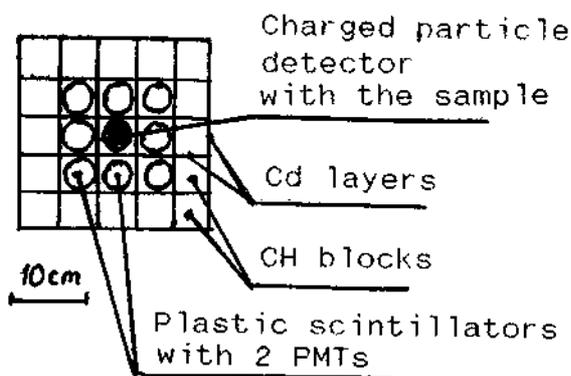


Fig.1. Diagram of the experimental set-up.

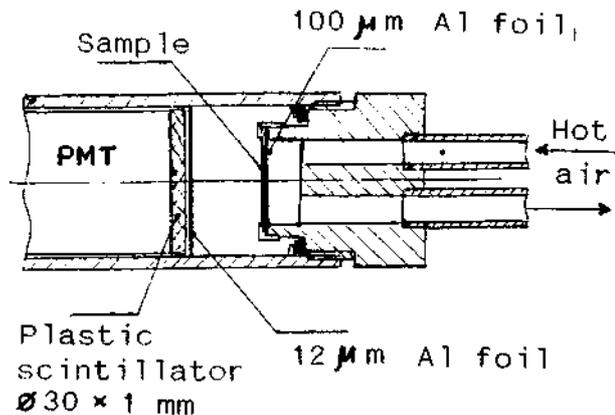


Fig.2. Plastic scintillation detector.

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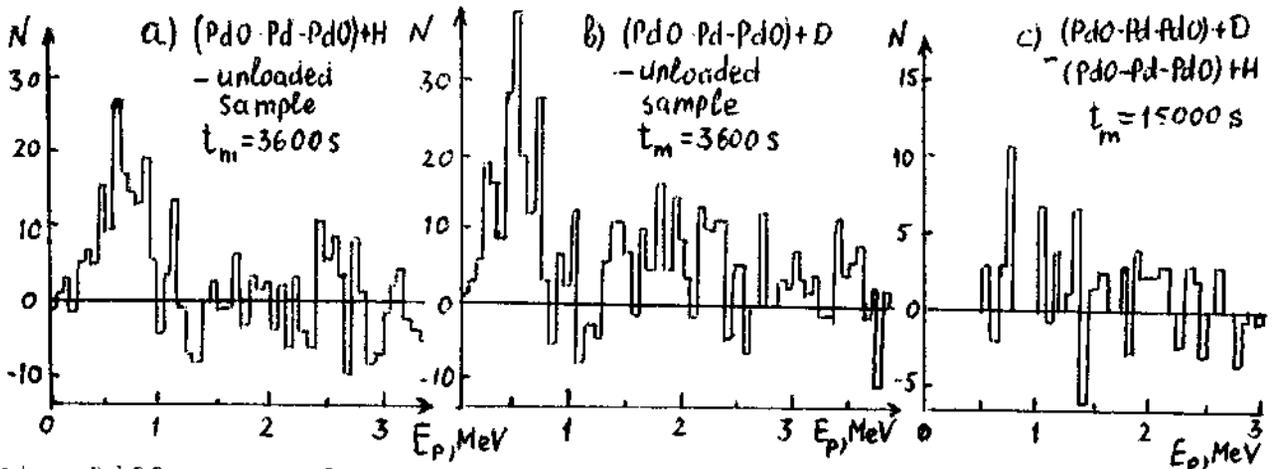


Fig.3. Difference of energy spectra measured by plastic scintillation detector for (a) hydrogenized and unloaded PdO-Pd-PdO samples; (b) deuterized and unloaded PdO-Pd-PdO samples; (c) deuterized and hydrogenized samples in the case of coincidence with the neutron detector.

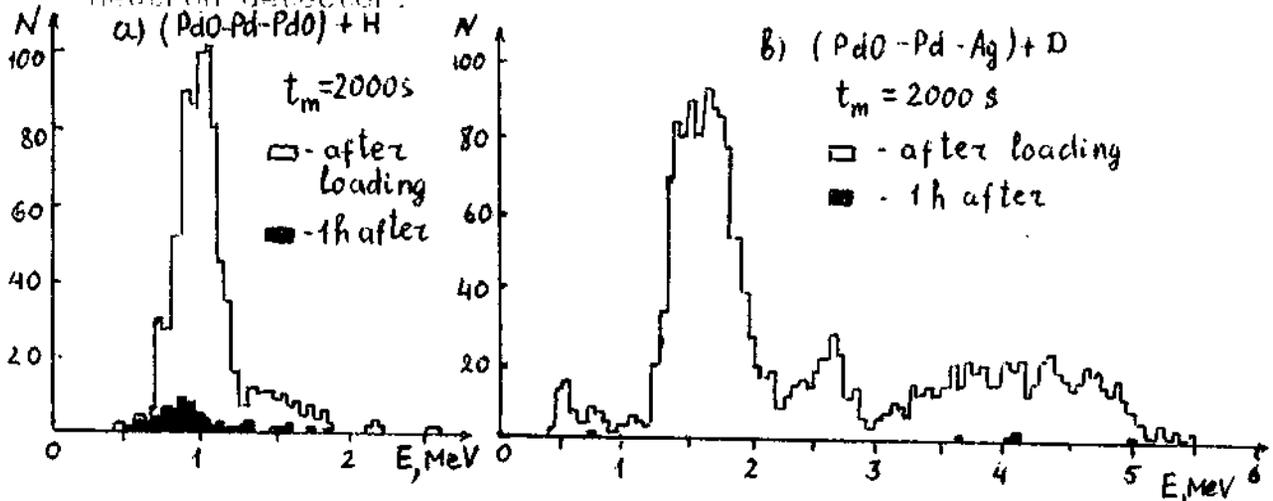


Fig.4. Energy spectra measured by Si-SSD for (a) hydrogenized PdO-Pd-PdO sample; (b) deuterized PdO-Pd-Ag sample (measurement time is  $t_m = 2000s$ ).

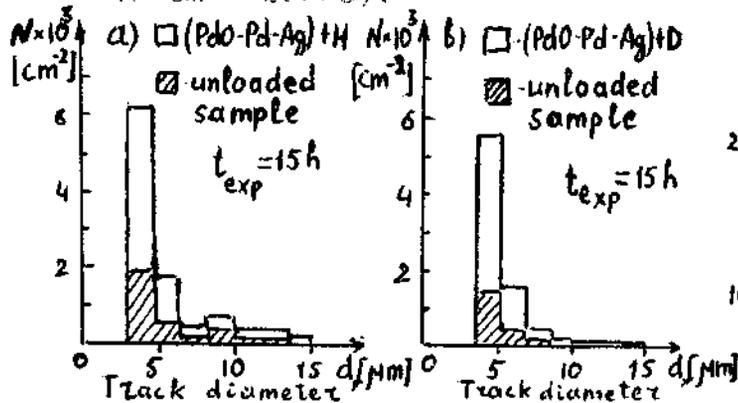


Fig.5. Diagrams for track density in CR-39. (a) deuterized PdO-Pd-Ag sample; (b) hydrogenized PdO-Pd-Ag sample (exposition time is  $t_{exp} = 15h$ ).

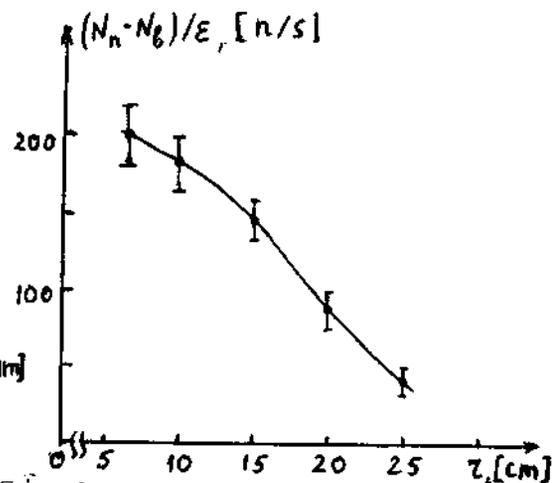


Fig.6. Neutron flux emitted from PdO-Pd-Ag sample vs thickness  $r$  of the moderator between the sample and the neutron source.