Excess Energy and Nuclear Products

Excess Heat Production and Nuclear Ash in PdO/Pd/PdO Heterostructure after Electrochemical Saturation with Deuterium

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Abstract

Heat production nuclear ash and electrophysical processes in PdO/Pd/PdO (Au/Pd/PdO) heterostructures electrochemically saturated with deuterium (hydrogen) have been investigated. It was shown in after electrolysis period a strong heat flash with duration of 2-7 s and energy density of 60-100 J/cm^2 was observed for Pd/PdO:D(H) sample placed in air atmosphere. The thermal energy of each flash was approximately 2-5 times higher than the energy supplied to the sample during electrolysis. Neutron- and γ-emissions accompanying the heat production have been investigated.

1. Introduction

The results of 6-years cycle of investigations that was trained on determination of the nature of excess heat and nuclear ash generation in Pd/PdO(Au/Pd/PdO) heterostructure electrochemically saturated with hydrogen (deuterium) will be presented. In contrast to traditional (for CF-experiments) systems that are massive Pd samples saturated with D [1-3] the system under consideration has some essential advantage in terms of obtaining or reproducible and controllable results:

a. short saturation time and absence incubation period. that precedes to excess energy generation;

b. low current density in electrolysis process: control of H(D)-content in Pd-sample at any time;

c. high reproducibility of excess (in contrast to applied in electrolysis) energy in each cycle of saturation of Pd-sample with H(D);

d. possibility of H(D) concentration in a small volume onto Pd-PdO interface, that is cause of high loading ratio (x) as well as local high elastic energy density in Pd lattice i.e. non-equilibrium phonon generation [4].

Therefore, application of thin Pd/PdO heterostructure as a cathode in CF-experiments can bring some advantage to achieve minimum distance between deuterons (protons) and, simultaneously, high concentration of non-equilibrium phonons. Both effects lead to increase of nuclear reactions probability in Pd-lattice with D(H)participation.

2. Experimental

Electrolysis was carried out in a glass cell was subdivided cathodic and anodic spaces (electrolyte volume of about 250 cm^3). The electrolytes used: 1 M KOH or H_2O for Pd/PdO:H_x production: 1 M NaOD in D_2O for Pd/PdO:D_x production. Current density used of about j=10-30 mA/cm^2.

Samples: cold-rolled Pd-foils with thickness 30-50 μm and working area of about S= 4.5 cm^2 for PdO/Pd/PdO (S=2.2 cm^2 for Au/Pd/PdO).

Samples preparation: 1) annealing in vacuum at temperature 600°C for 3 hours with slow cooling (1°C/min) to room temperature; 2) metered annealing and oxygen flame at a temperature of about 1000°C That leads to PdO layer formation having a thickness of about 200-500 Å; 3) electrochemical deposition of Au on the one side of PdO/Pd/PdO heterostructure: thickness of gold coating of about 2000 Å.

Research methods

l. H(D)-concentration in the samples was determined by vacuum thermodesorption technique at 400°C and $P=10^{-6}$ mm Hg.

2. A gas thermometer was applied to record the afterelectrolysis thermal effect. The schematic diagram of that gas thermometer is presented in Fig.1. The quantity of heat (Q), evolved by the sample in the pulse regime, was recorded by a U-shaped mercury pressure gauge with an analytical volume of the exposure chamber (2), V=69 cm^3 which was filled either by air or by O_2 or Ar under atmospheric pressure. The value Q of the Pd-samples was estimated by integrating over the pressure-time (P-τ) curve within a range limited by the pulse origin moment, and by the moment of intersection by it of the time axis τ. Thereafter, for the purpose of quantitative estimation, a comparison of pulses, being obtained on thermal bursts with calibration pulses from a nichrome spiral (8) was carried out. The measurement error of Q did not exceed ±20%.

3. A specially developed set up on the basis of electromagnetic displacement transducer with sensitivity of about $2.5*10^{-6}$ cm was applied to determine in situ changes in the length of thin Pd samples m the hydrogenation-dehydrogenation processes.

4. A block of 7 proportional NWI-62 neutron counters ($B_{10}F_3$) placed into a tank containing vacuum oil and covered by Cd sheet l mm thickness, was used to neutron detection in deuterated Au/Pd/PdO samples (neutron detection efficiency: $\varepsilon_n=3.9*10^{-2}$).

5. To detect a γ-radiation upon thermal effect in afterelectrolysis period semiconductor γ-detector (pure Ge) GEM-20180P by- EG&G ORTEC was used (efficiency of γ-detection $\varepsilon\gamma=3*10^{-3}$).

3. **Experimental results**

Excess energy

On the electrolysis process, at the electrolysis time 1<40 min in Au/Pd/PdO τ<40 min in Au/Pd/PdO heterostructure the one-side bending of sample is observed (Fig.2). The degree of bending is depend upon dectrolysis time. This bending is a cause of deuterium (H) storage on the Pd-PdO interface [5]. At electrolysis time τ>40 min the straighten of sample takes place due to deuterium (H) breakdown into the sample bulk.

The free energy (elastic deformation) stored up in the sample due to electrolysis may approximately be estimated with the formula [6]

$$F = \int \psi_2(U_{\alpha\beta})\, ds$$

where $U_{\alpha\beta}$ is 2-dimension deformation tensor; $\psi_2=Eh(\xi/l)4$ here E - is Young modulus of Pd, h - sample thickness; ξ - sample mean deflection, l - length of the sample. For $h=3*10^{-3}$ cm, l=4.5 cm, s=4.5 cm^2 and electrolysis time τ=30 min we have ξ=5 mm (Fig. 2). And then F=76 cal/cm^2. It should be noted that the energy value applied upon electrolysis time was E_{el}

of electrolysis energy applied about one order of magnitude.

When the electrolysis is interrupt at pause (τ<40 min for h=50 μm) after the drying of sample there takes place the thermal burst (in air, Ar , atmosphere or vacuum). Which is accompanied by the heating of sample up to temperature a of 900 °C (Fig.3a).

In the case of electrolysis carried out for 5 hours (Fig.3a, curve 4), the monotonous increase in the pressure of the gas thermometer is due to movable excess hydrogen which easily leaves the Pd lattice have been took place.

When the hydrogenated Pd/PdO samples are placed into the chamber at t=20°C. the dependence ΔP- τ is of the extremum character (Fig.3a, curves 1,2,3). The retardation of the thermal effect (when O_2 or air is present in the chamber) is limited by the sample drying time. and may range from a few seconds to several minutes. The pressure increase being accompanied by the visible heating of the Pd sample (e.g red or white glow) up to temperatures higher than 900°C (the thermocouple reading) during 3 to 4 seconds, and by its axial bending. Then the chamber is evacuated, which is due to the consumption of O_2 for the reaction with hydrogen in the Pd sample, and for condensation of water vapor.

The Pd/PdO system. which has been electrolytically deuterated for the same time intervals, behaves itself somewhat differently (Fig. 3b). When a sample is put into the analytical volume at room temperature, strong deformations take place in it, as well as in hydrogenated samples; however, no thermal burst is observed. When the sample is preheating by the aid of an electric lamp up to a temperature of 35 to 40°C the behaviour of the Pd/PdO:D samples becomes similar to that of the Pd/PdO:H samples at room temperature. In this case, the character of a thermal pulse is more prolonged, the growth time about 7 to 10 sec, the vanishing time about 40 sec), while the integral thermal effect is approximately by a factor of 1,3 to 1,4 higher than that in the Pd/PdO:H system.

The integral mean values of heat evolution, Q both for Pd/PdO:H system (at t=20°C) and for the Pd/PdO:D system (at t=40 °C), were estimated by carrying at calibration measurements with a nichrome spiral. For 100 thermal bursts, these are equal to Q_H =15,1 ±3,5 cal/cm^2 , Q_D =20,3 ±4,8 cal/cm , respectively. The reproducibility of the effect is extremely high, so that one sample gives not less than 150-200 thermal bursts in succession with consecutive hydrogenation cycles. In Fig.4 are presented in the form of histograms the results obtained for 100 bursts in succession on one of the Pd/PdO:H samples. In 5 cases from 100 there were observed very powerful bursts attaining the value of 40 to 45 cal/cm^2 . Let us note that the energy transmitted to the sample directly on electrolysis W_E =8,0 ±1,5 cal/cm^2 . Therefore, the observable thermal effect $Q_{H,D}$ may be considered as being anomalous.

The magnitude of thermal effect can be increased essentially when specific surface of heterostructure samples will be strongly increased too. For this purpose it was Pd-black (r^{-4}=10 cm) deposited by electrochemical method onto PdO surface. At saturation time τ =30 min, the mean Q value is increased of about 2 times (in contrast to samples without Pd black) and became of about 35 ±3.6 cal/cm^2 (Fig.5). The reproducibility of this thermal burst at room temperature is equal to 100 %.

Nuclear ashes

To determine correlation between thermal and nuclear processes in Au/Pd/PdO:D(H) heterostructure the simultaneous detection of excess heat production and nuclear radiations was carried out. The registration was began from the moment

of sample's drying and was finished to the moment of total gas desorption from the sample.

The experiments have shown no statistically noticeable emission of neutrons is observed on the Au/Pd/PdO:H control samples (with the thermal effect), while the distribution of neutron events is well consistent with the Poisson distribution of the natural (cosmic) background (Fig.6, curve 2).

In the same time thermal effect (and deuterium desorption) from the sample Au/Pd/PdO:D_x is accompanied by a neutron emission (Fig.7). The maximum of neutron counts is observed in the beginning of D-pressure jump, when strong plastic deformation of sample is taking place. In this time interval the neutron bursts are observed with multiplicity i=6-17 (in the time gate of about 1 ms) or with intensity n=100-500 n/(s*cm^2) Pd (Fig.6, curve 1). The residual plastic deformations that occurs on the D_2O condensation process are accompanied by weak neutron emission too. The neutron emission decay to background level takes place after 5-6 min (from the beginning of D-gas desorption). After finishing of the process the loading ratio value is $x \sim 10^{-3}$ [7]. However, neutron emission can be prolongated during 15-20 min, if Au/Pd/PdO sample will be loaded by mechanical stress (by hanging up to the one end of the sample a weight of about M=40 g). The rate constant of dd-reaction in Au/Pd/PdO:D_x heterostructure $\lambda_{DD} = 2*10^{-21}$ s^{-1} per dd-couple (for x=0.72) and $\lambda_{DD} = 10^{-18}$ s^{-1} per dd-couple (for $x=10^{-3}$). It should be noted that the sample with $x \sim 10^{-3}$ posses of residual elongation of about 1/3 from elongation at x=0.72, accordingly with the date obtained from electromagnetic displacement's transducer measurements. It has been discovered that this elongation is due to quasimetallic clusters of hydrogen [7].

The same samples of Au/Pd/PdO:D_x and Au/Pd/PdO:H_x (x=0,72) after electrolysis procedure was used for γ-emission tests in the energy range of 2.0-10.0 MeV. In the spectrum of Au/Pd/PdO:D_x samples (Fig. 8) there are 3 plainly expressed maxima with positions $(E_\gamma)_1 = 2.225 \pm 0.005$ MeV; $(E_\gamma)_2 = 3.8 \pm 0.5$ MeV; $(E_\gamma)_3 = 6.3 \pm 0.3$ MeV. The narrow line $(E_\gamma)_1$ (half width of about 10 keV) could be ascribed to p+n reaction, because its position is in a good agreement with γ -line (E=2,225 MeV) obtained upon PE-irradiation by thermolized neutron from Cf^{252} -source (Fig.9). The $(E_\gamma)_3$ maximum can be give rise to γ-quanta from d+n reaction (E_γ=6.25 MeV). In fact this peak increases of about 2 times upon the sample irradiation by thermalized neutrons. The nature of $(E_\gamma)_2$ maximum is not clear. However as it was proposed earlier [8] the E_γ =3.8 MeV peak can be exhibition of the first excited state of He nucleus that may form with phonon participation [9].

In contrast to deuterated heterostructure the samples of Au/Pd/PdO:H generate only one - maximum with position of about $E_\gamma = 4.6 \pm 0.3$ MeV. Under the irradiation of the sample by thermalized neutrons the maximum E_γ =4.6 MeV is increasing in intensity more than 5 times and shifted on 0.3 MeV into the high energy band. The nature of this γ-emission is unknown now.

Therefore excess energy generation in Au/Pd/PdO:D(H) heterostructure is accompanied by γ-emission. The γ-emission intensity depends upon the thermal neutron background [10].

Moreover, as it shown by A.Roussetski (this volume of ICCF-6 Proceedings) desorption process in heterostructure of Pd/PdO type (both H and D saturated) is accompanied by emission of charged particles (protons and possibly α-particles).

4.Conclusion

Simultaneous generation of excess energy and weak nuclear radiation in Au/Pd/PdO:D_x (H_x) heterostructure as well as strong plastic deformation in it are indicate on the possibility of phonon laser action in system of this type. Really, the main process that determines heterostructure's properties in afterelectrolysis time is an intensive exothermic desorption of hydrogen (D) from the sample. This effect, from other side, is a main condition to phonon laser operation on Pd-PdO interface in the high loading ratio zone (clusters of quasimetallic hydrogen [7]). In this zone coherent multiphonon excitations can initiate an anomalous energy transfer and, simultaneously lead to coherent neutron transfer reactions [11]. As result, both excess heat and weak nuclear radiation are observed. In this connection the short electrolysis process for heterostructure saturation with H(D) can be considered as pumping up procedure of phonon laser.

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Captions to figures

Fig. 1. Experimental set up with gas thermometer and neutron detector: 1 - PE(Co) ; 2 - chamber with atmosphere ; 3 - gas thermometer (Mcleod) ; 4 - sample ; 5 - neutron counters ; 6 - moderator ; 7 - Cd; 8 - heater.

Fig. 2. Au/Pd/PdO sample: initial (a) and deformed (b) after electrochemical hydrogenation during τ =30 min ; ξ - is mean deflection.

Fig. 3a. Thermal bursts for Au/Pd/PdO:H_x : saturation time : 10 min - curve 1 ; 20 min - curve 2 ; 30 min - curve 3 ; 5h - curve 4 ; calibrating pulse from nichromium coil - curve 5.

Fig. 3b. Thermal burst for Au/Pd/PdO:D_x saturation time : 30 min - curve 1 ; 5h - curve 2 ; calibrating pulse - curve 3.

Fig. 4. Distribution of the heat production events (n_Q) with respect to the heat evolution energy Q for Au/Pd/PdO:H_x -system.

Fig. 5. Distribution of n_Q with respect to Q for [Au/Pd/PdO+Pd-black]:H_x system.

Fig. 6. Distribution of the number of neutron events (n_i) with respect to the multiplicity of neutron pulses (i) for Au/Pd/PdO:D_x heterostructure for $\Delta\tau$=120 s time intervals preceding the heat burst (N=100 experiments) - curve 1 ; Poisson distribution for neutron events with N=100 - curve 2.