Analysis of Neutron Irradiation Effects in Pd-LiOD (H) System

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Abstract

Experimental data of the excess heat and gamma spectrum from Pd/D(H)/Li system were analyzed on the TNCF model. The density n_n of the trapped neutrons was determined as 3.0×10^9 cm^{-3} by the excess heat data. The several peaks in the gamma spectrum were identified with the photons generated by fusion reactions between the trapped neutrons and nuclei in the cathode, isotopes of Pd and Li and occluded deuterons or protons. Large peaks at 2.2 and 6.25 MeV in foreground and also in background runs were interpreted as due to contamination of hydrogen and deuterium.

1. Introduction

The cold fusion phenomenon in various systems with hydrogen and/or deuterium has been widely and thoroughly investigated in these almost eight years and the essential parts of the phenomenon has been confirmed.

In the experimental data of the cold fusion, gamma spectrum has recently been observed with high precision[1-3]. In the analysis of the data obtained in Ni + K_2CO_3 system[2], we have given a consistent explanation[4] of gamma rays at $E_\gamma = 511$ keV and a nuclear transmutation of ^{39}K into ^{40}K.

In a recent paper[3] on the excess heat and gamma spectrum an excess power of 1.5 to 2.5 W and clear gamma peaks in a range up to 7.5 MeV in Pd/D(H)/Li system were observed

In this paper we will give a consistent interpretation of several peaks in the gamma spectrum in relation with nuclear reactions between nuclei in the sample and the trapped neutrons, using the TNCF model. In the analysis we will use a conclusion obtained in the previous analysis[4] that the fusion cross section in the volume decreases to 10^{-2} of that in the surface layer which has been assumed the same as in vacuum.

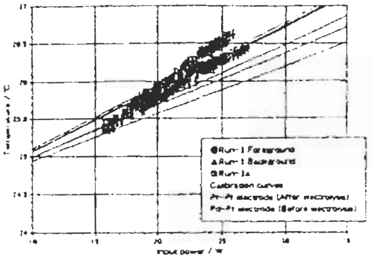

Fig.9 Correlation between temperature and input power of Run-3 and Run-3a
Solid curves are calibration curves.

Fig.1.

2. Experimental Facts

In the experiment[3] where the effect of neutron irradiation on Pd/D(H)/Li - Pt system was measured, a palladium sample was used which had shape of a spherical half-shell with an inner radius of 1.25 and an outer radius of 1.30 cm. The foreground(FR) and the background (BR) runs were with electrolytes LiOD + D_2O and LiOH + H_2O, respectively.

A reference run (RR) was on Pt/D/Li–Pt system. Neutron energy spectrum, gamma spectrum and the excess heat with and without thermal neutron irradiation was measured, from which we will take up the latter two cases in this analysis.

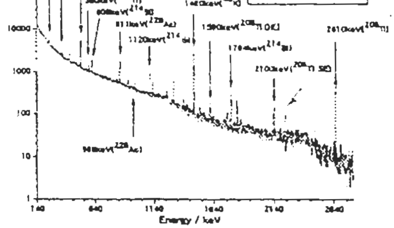

Fig.2 Gamma Spectrum over the energy range 140–2700keV
(^{252}Cf unirradiation case)

Fig.2.

1) Excess Heat

The excess heat had been determined by measurement of the electrolyte temperature T versus input power P_{in}. They concluded that the excess powers of 1.5 to 2.5 W were generated in the FR case from their data including **Fig.1** (their Fig.9). Though the authors neglected the deviations from the calibration line in cases of BR and RR cases below $P_{in} \leq 23$ W, we can see the excess heat generation not only in FR but also other runs (BR and RR). We will use the excess heat data to determine the density n_n of the trapped neutrons in the next section, not forgetting the above comments.

2) Gamma spectrum

The gamma spectrum up to 7.5 MeV was measured in the FR and BR cases without and with artificial thermal neutron irradiation from a ^{252}Cf source. The spectra in Figs.2 and 3 (their Figs.7 and 8) show many peaks of which up to 2.1 MeV has been identified by the authors as the reactions which generate the corresponding photons. It should be noticed here the existence of the positron annihilation peak at 511 keV which has been measured also in another experiment[2].

Other peaks we will take up from Figs.2 and 3 are those at 2.22, 5.49, 5.72, 6.15, 6.25, 7.09 MeV. Though the authors neglected a peak at 2.22 MeV in **Fig.2** (without ^{252}Cf) and discarded the peaks at 2.22 and 6.25 MeV in **Fig.3** as they had the same intensities in FR and BR cases, it is necessary to give a consistent explanation of the existence of these peaks in the experimental data with other peaks in the spectrum.

We can see that the base levels of the spectra in Fig.3 for FR and BR cases were clearly different. If this investigation is correct, we have to give an explanation of the existence and the intensity difference of the peaks at 2.22 and 6.25 MeV in FR and BR without and with irradiation.

3. Experimental Data Analysis

The TNCF model[5-8] assumes an existence of trapped neutrons in the cold fusion materials. The neutrons had been supposed to be fairly stable and only fused with foreign nuclei which are in the boundary layer where the neutrons suffer large perturbations and become unstable (in the classical sense). An analysis[4] of the experimental data where observed the nuclear transmutation at room temperature[2], however, showed that the stability of trapped neutrons against a fusion reaction in the volume of the material is higher by a factor of 10^2 compared with the boundary.

Therefore, we will use the same relation in the volume we have used for the reactions in the surface layer, with a factor $\xi = 0.01$ introduced in an analysis[4] of the data[3] at room temperature.

We will treat the excess heat and gamma generation in Pd/D(H)/Li system as follows with a factor ξ to take into account the effect of the stability in the volume.

The number of events N_x in a time τ of the fusion reaction between the trapped neutron and the nucleus x is given by a following relation with $\xi = 0.01$ in the volume and $\xi = 1$ in the surface layer:

$$N_x = 0.35\, n_n v_n \rho_x V \sigma_{nx} \tau \xi. \qquad (1)$$

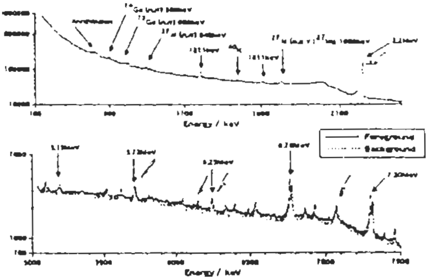

Fig3 Gamma spectrum over the energy range 140–2500keV and 5000–7500keV (^{252}Cf irradiation case)

In this relation, $0.35 n_n v_n$ is the neutron flux per unit area and time, ρ_x is the density of the nucleus x in the volume V, σ_{nx} is the cross section of the fusion reaction.

Considering abundance and fusion cross section of the nucleus, the effective reactions in the Pd/D(H)/Li cathode could be taken as follows:

$$n + p = d + \gamma(2.22\text{MeV}), \qquad (2)$$
$$n + d = t + \gamma(6.25\text{MeV}), \qquad (3)$$
$$p + d = {}^3\text{He} + \gamma(5.49\text{ MeV}), \qquad (4)$$
$$n + {}^6\text{Li} = {}^4\text{He}\ (2.11\text{MeV}) + t\ (2.7\text{MeV}), \qquad (5)$$
$$n + {}^7\text{Li} = {}^8\text{Li} + \gamma(2.03\text{MeV}), \qquad (6)$$
$$n + {}^{104}\text{Pd} = {}^{105}\text{Pd} + \gamma(7.09\text{MeV}), \qquad (7)$$
$$n + {}^{105}\text{Pd} = {}^{106}\text{Pd} + \gamma(9.56\text{MeV}), \qquad (8)$$
$$n + {}^{108}\text{Pd} = {}^{109}\text{Pd} + \gamma(6.15\text{MeV}), \qquad (9)$$
$$n + {}^{110}\text{Pd} = {}^{111}\text{Pd} + \gamma(5.75\text{MeV}), \qquad (10)$$

The natural abundance of the isotopes appeared in the reactions (5) to (10) are 7.42, 92.59, 10.9, 22.3, 26.5 and 11.8%, respectively. The cross sections of the reactions (2), (3) and (5) to (10) are 0.3, 5.5 x 10^{-4}, 940, 0.05, 0.52, 143.0, 6.1 and 0.23 barn at room temperature, respectively.

We have to take into account a following photo-disintegration of deuteron in the analysis of the experimental data:

$$\gamma + d = p + n. \qquad (11)$$

The threshold energy of this reaction is about 2.22 MeV and the energy dependence of the cross section[9] is shown in Fig.4 with a broad peak at

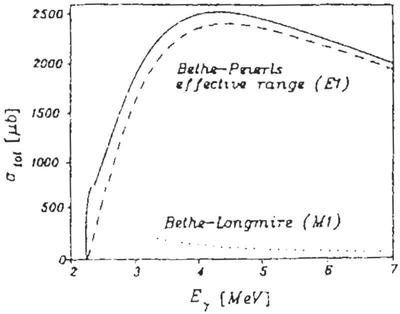

Fig.4.

about 4 MeV of 2.5 m barn.

The excess heat generated by the reactions (2), (3) and (5) to (10) was calculated using n_n as an adjustable parameter and assuming; 1) all the liberated energy was transformed into heat in the system, 2) D/Pd = 1 in the volume and 3) thickness of the Li surface layer on the cathode is 1 μm.

The experimental value of the excess power 2 W gave a value 3.0 x 10^9 cm^{-3} for the density of the trapped neutron:

$$n_n = 3.0 \times 10^9\ cm^{-3}.$$

This value $n_n = 3.0 \times 10^9\ cm^{-3}$ of the density of the trapped neutrons is in the middle of the values determined by us in the analyses of cold fusion data[7] and in consistent with values obtained in similar experimental conditions.

The peaks at 6.76 and 7.30 MeV in Fig.3 which have been out of our investigation might be attributed as due to stray photons generated by the following reactions in Pb blocks used in the experiment for protection:

$$n + {}^{204}Pb = {}^{205}Pb + \gamma(6.73MeV), \quad (12)$$
$$n + {}^{206}Pb = {}^{207}Pb + \gamma(6.74MeV), \quad (13)$$
$$n + {}^{207}Pb = {}^{208}Pb + \gamma(7.37MeV). \quad (14)$$

Natural abundance of the isotopes ^{204}Pb, ^{206}Pb and ^{207}Pb are 1.48, 23.6 and 22.6%, respectively.

If this speculation is true, interpretation of the gamma spectrum is useful to confirm the behavior of trapped neutrons in solid materials around us.

4. Discussion

The most difficult problem in the analysis of the data as shown in Figs.2 and 3 is an interpretation of the peaks at 2.22 and 6.25 MeV. These sharp and strong peaks have the same energies as those of photons generated in $n + p$ and $n + d$ fusion reactions, respectively. Therefore, it is natural to take these peaks as those due to the reactions (2) and (3).

Then, almost the same intensities of the peaks in FR and BR show that majority of these reactions were occurring in FR and BR with the same frequency. So, we have to conclude that there were fairly much contamination of H_2 (D_2) or H_2O (D_2O) in the experimental system (perhaps except samples) and the reactions (2) and (3) occurred between irradiated neutrons and them. It should be also noticed that a small but sharp 2.22 MeV peak is seen in Fig.2 without ^{252}Cf irradiation.

In the following analysis of the gamma spectrum, we take the peaks at 2.22 and 6.25 MeV are mainly due to contaminated nuclei p and d and contain components due to the reactions occurring in the sample.

Then, the reactions (2) to (4) and (6) to (10) written down in the previous section give possible peaks in the gamma spectrum at 2.22, 6.25, 5.49, 2.03, 7.09, 9.56, 6.15 and 5.75 MeV. Surprisingly enough, all these peaks do really exist in the observed spectrum shown in Fig.3 except the one at 9.56 which is outside of the scale and weakness of the last two peaks due to the abundance and cross sections of ^{108}Pd and ^{110}Pd. This fact shows clearly that there are fusion reactions between the thermal neutron and nuclei in the sample as assumed in the TNCF model. On the other hand, as has been pointed out above, there is a peak at 2.22 MeV due to the reaction (2) even in the unirradiation case (Fig.2). This fact shows the existence of the trapped neutrons in the sample even if no thermal neutron was irradiated artificially.

The reactions (2) (or (3)) and (5) to (10) could occur in FR (and BR) case if there are enough thermal neutrons either trapped or irradiated. The calculation of n_n given above showed that numbers of events of reactions (5) and (6) are comparable to those of (7) to (10). We evaluated the number of events of the reaction (2) (or (3)) which was similar to those of (7). Therefore, the intensity of the peak at 7.09 MeV in Fig.3 gives us a possible intensity of the peak at 2.22 (or 6.25) MeV due to the reaction (2) (or (3)) in the sample, which is too small to influence the observed peak there. This scenario gives an explanation of the peaks at 2.22 and 6.25 MeV due mainly to the contamination of hydrogen and deuterium including signals from reactions (2) and (3).

In addition to the consideration given above, we want to mention a fact of a proton generation by the reaction (11) in the FR and a deuteron generation by the reaction (2) in the BR cases. In the FR case, the proton generation through the photodisintegration (11) by gamma generated in reactions (7) to (10) amounts to 4 x 10^6 /s. On the other hand, in the BR case, the deuteron generation through the fusion reaction (2) by trapped or irradiated thermal neutrons amounts to 10^{10} /s. These values of p and d generation may influence the signal due to the presence of minor hydrogen isotopes in FR and BR cases. The annihilation peak in Fig.3 at 511 keV shows the existence of reactions producing nuclei which decay by positron emission. In the case of Ni-K_2CO_3 system[2], the responsible reactions were written down as follows:

$$^{62}\mathrm{Ni} + n = {}^{63}\mathrm{Ni} \rightarrow {}^{63}\mathrm{Cu} + e^- + \nu_e, \quad (15)$$

$$^{63}\mathrm{Cu} + n = {}^{64}\mathrm{Cu} \rightarrow {}^{64}\mathrm{Ni} + e^+ + \nu_e, \quad (16)$$

In the case of Pd/D(H)/Li system we are now considering, there are no such a reaction to produce positron in the sample and electrolyte. So, we can consider the same reactions used in the previous analysis[4] have been working in surrounding materials of stainless steel with Ni or materials with Cu in it. Then, starting from ^{62}Ni, natural abundance and fusion cross section of which are 3.66% and 15 barn, respectively, or from ^{63}Cu, natural abundance and fusion cross section of which are 69.1% and 4.5 barn, respectively, we can reach the nucleus ^{64}Cu which emits a positron as written in the above reactions. The small annihilation peak in Fig.3 could be understood as such.

Analysis of fine experimental data provides an insight into the physics of the cold fusion phenomenon as had been said. The data on the excess heat and gamma spectrum obtained by Okamoto et al.[3] are new one of those fine data in the field of cold fusion. The experimental data not only show the real existence of the cold fusion phenomenon in the Pd/D(H)/Li system but also figure out the existence and the role of trapped neutrons using an artificial neutron source.

The analysis of the data on the TNCF model given here tells us that there is a consistent plot in the excess heat generation and the gamma emission in the electrolytic cold fusion system, though there were no descriptions of the controversial reproducibility and necessity for preparatory drive.

We want to express our hope again that the cold fusion phenomenon in those various materials from metals to oxides could be explained by a phenomenological model, the TNCF model using a single parameter n_n, the density of the trapped neutrons. Perhaps through these treatments we will arrive at the final, decisive key to explain the cold fusion phenomenon and develop a new energy source to resolve the serious energy problems of our culture lying in front of us.

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investors will see them. Yes, of course I'm talking about this, the only popular journal dedicated solely to promoting cold fusion.

It takes visibility to attract money. You have to create an awareness of your product in the minds of your potential customers (investors). With a product this is done via new product releases, reviews of the product in industry journals, and with advertising. With research and development, it's done with published scientific papers. But you know all that, so what's keeping you from your word processor, and the ensuing fame and fortune?

The next time one of the exposé TV shows decides to cover cold fusion, who are they going to contact? It isn't going to be some relatively unknown person or group.