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Analysis of Zn and Excess Heat Generation in Pd/H_2 (D_2) System by TNCF Model

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Synopsis

Experimental data sets by X.Z. Li et al. showing generation of the excess heat and nuclear transmutation of Pd into Zn in Pd-D_2 gas and Pd-H_2 gas system are analyzed by the TNCF model. The mechanism generating Zn and the excess heat is assumed to be the n – ${}^{A}_{46}$Pd reaction followed by fission of thus formed compound nucleus ${}^{A+1}_{46}$Pd* into Zn and S and the arbitrary parameter n_n of the model is determined by the value of the excess heat in the Pd - D_2 gas system to be $\approx 10^{10}$ cm^{-3} consistently with values determined by the amount of Zn observed in the system, while the excess heat was not observed in Pd-H_2 gas system. Possible cause of this discrepancy is discussed.

1. Introduction

There have been obtained in these several years many experimental data sets showing nuclear transmutations (NT's) explained only taking into possibilities of induced decay and fission of compound nucleus formed from nucleus in solids by absorption of a thermal neutron, as shown in our previous papers.[1,2] The assumed nuclear reactions in the explanation of these NT's are difficult to consider their occurrence by absorption of a thermal neutron in free space. Furthermore, there is a riddle about branching ratio of channels the compound nucleus transforms into new nucleus: the branching ratio seems to depend strongly on situation in the solid the compound nucleus is formed. For instance, the same Pd/D system showed one of following NT's in different experiments; Pd to Ag and Pt to Au by induced β-decay, ^{4}He generation by induced α-decay, Pd to Ti and Pd to Zn by induced fission and so forth.

The excess heat and nuclear transmutation from Pd to Zn observed by X.Z. Li et al.[3,4] belongs in these data sets of NT's and have been explained [5] on the TNCF model proposed by us while there is some ambiguity on the data. Recently, at the ICCF7, they presented new data on the same system which is the present object of analysis on the TNCF model in this paper. The result of present analysis is compared with the previous one and discussed in relation with analyses of other data sets.

2. Experimental Results

The experimental data sets obtained in Pd-D_2 and Pd-H_2 gas-contact and glow discharge systems by Li et al. have been presented at from ICCF6 to ICCF7.[3,4,6,7] The former data sets were analyzed consistently [5] by us using the TNCF model. The new data on the same system have been given at ICCF7 and are summarized as follows.

2.1 The excess heat

A long and thin Pd wire (250 cm × 0.34 mm ϕ) was mounted on a quartz frame in a Dewar, the temperature in and outside of it were monitored to measure the excess energy generation in the sample. In the experiments with this apparatus, protium or deuterium gas was filled and the temperature of the filled gas was changed cyclically between 16 and 20 ℃ with a period of 6 h or 2 d. D/Pd ratio reached up to 0.3 in an experiment where the excess heat of $\leq$ 0.23 W in average and 25.9 W at maximum from the sample was measured.

The reproducibility is not good although they have measured the excess heat three times in 2 years in 2 different calorimetric systems. In an experiment to check the above result, they made an experiment without heating process and obtained the "excess heat" of 0.005 W which be compared with the above positive value of 0.23 W.

It should be noticed that they observed the excess heat only in Pd/D system but not in Pd/H system.

2.2 Zn generation

To confirm the nuclear transmutation in the sample which generated the excess heat, the authors used NAA (neutron activation analysis) of the sample. In this experiment, they used samples from Pd/D and Pd/H systems without clear identification (and we can not say which sample of Nos. 1, 3, 4, 5 belongs to which system) while in the previous paper[3)] only Pd/H system was analyzed and found Zn up to 40% in the surface layer of thickness 40 μm. (The excess heat measurements[4)] were performed in Pd/D system relative to Pd/H system with a positive result in these experiments.)

It was detected that Zn appeared in the sample used in the cold fusion experiment and the isotope ratio of $\eta \equiv {}^{64}_{30}\mathrm{Zn}/{}^{68}_{30}\mathrm{Zn}$ shifted from natural one ($\eta = 2.6$) and differed from one sample to another. (In the experiment, the isotope ratio η is investigated by NAA signal intensity ratio but not by the absolute value of abundance itself). The total amount of Zn generated is also differed from one sample to another and is 30 % in sample No.5 (while we have no description about where the Zn content is determined in the sample).

3. Theoretical Investigation

To analyze the experimental data introduced in the previous section, we assume that the excess heat and the nuclear transmutation have the same cause, i.e. the same nuclear reaction in the sample.

The nuclear reactions generating Zn in Pd/D$_2$ system are assumed as written down as follows in our present knowledge of nuclear physics which we trust its applicability for nuclear reactions in energy range up to several hundred MeV:

$$n + d = t\,(5.98\ \mathrm{keV}) + \gamma\ (6.25\ \mathrm{MeV}), \tag{1}$$

$$n + {}^{A}_{46}\mathrm{Pd} = {}^{A+1}_{46}\mathrm{Pd}^{*}, \tag{2}$$

$$ {}^{A+1}_{46}\mathrm{Pd}^{*} = {}^{64}_{30}\mathrm{Zn} + {}^{A-63}_{16}\mathrm{S} + 2n + \mathrm{Q}. \tag{3}$$

The compound nucleus ${}^{A+1}_{46}\mathrm{Pd}^{*}$ has several channels to transform including the one taken up in the above reaction (3) relevant with Zn generation. The discrimination used in this paper about the decay channels will be discussed in the next section.

The cross section of the first reaction is 5.5×10^{-4} b and is smaller than those of the third reaction with A = 102, 106, 108 or 110 leading to an unstable compound nucleus ${}^{A+1}{}_{46}\mathrm{Pd}^*$ by a factor about 10^{-4} as shown in Table 1.

Table 1, Cross section and liberated energy of n - Pd to Ru - He reactions.

Table 1. Natural abundance, absorption cross section σ (for 1/40 eV neutron) of isotopes ${}^{A}{}_{46}\mathrm{Pd}$

Isotope	${}^{102}{}_{46}\mathrm{Pd}$	${}^{104}{}_{46}\mathrm{Pd}$	${}^{105}{}_{46}\mathrm{Pd}$	${}^{106}{}_{46}\mathrm{Pd}$	${}^{108}{}_{46}\mathrm{Pd}$	${}^{110}{}_{46}\mathrm{Pd}$
Abundance(%)	0.96	10.97	22.23	27.33	26.71	11.81
σ (b)	3.363	0.523	20.25	0.303	8.504	0.227

The excess energy Q of the last reaction (3) depends on the mass number A of the initial nucleus ${}^{A}{}_{46}\mathrm{Pd}$. In the case of A = 108, which contributes most predominantly to the CF phenomenon due to the cross section and abundance, the pair product ${}^{43}{}_{16}\mathrm{S}$ decays successively through ${}^{43}{}_{17}\mathrm{Cl}$, ${}^{43}{}_{18}\mathrm{Ar}$, ${}^{43}{}_{19}\mathrm{K}$ finally to ${}^{43}{}_{20}\mathrm{Ca}$ by β-emission. In this case, the energy liberated in the process amounts to 6.815 MeV.

For the value of A = 108, the liberated energy 6.815 MeV corresponds 1.1×10^{-12} J. (1 MeV = 1.6×10^{-13} J).

The number of reactions per unit time $N_{\mathrm{n\text{-}Pd}}$ of n$-{}^{108}{}_{46}\mathrm{Pd}$ reaction is given by a following relation:

$$N_{\mathrm{n\text{-}Pd}} = 0.35 n_{\mathrm{n}} v_{\mathrm{n}} n_{\mathrm{Pd}} V \sigma_{\mathrm{n\text{-}Pd}} \xi ,$$

where $0.35 n_{\mathrm{n}} v_{\mathrm{n}} n_{\mathrm{Pd}}$ is the flow density of the thermal neutron per unit area and time, n_{Pd} is the density of ${}^{108}{}_{46}\mathrm{Pd}$ in the reaction region with volume V and $\sigma_{\mathrm{n\text{-}Pd}}$ is the cross section of the reaction. The factor ξ expresses an order of instability of the trapped neutron in the reaction region; we take $\xi = 1$ for reactions which occur in the surface layer with thickness of 1 μm according to the recipe of the TNCF model.[1)]

If we assume that the 30% Zn observed in the sample No.5 be in the surface layer with thickness 1μm of the Pd sample (250 cm $\times 0.34$ mmϕ), the number of Zn atom is calculated to be 5.5×10^{19} Zn atoms. (This assumption is based on general knowledge of surface reactions in CF phenomenon and differs from the data obtained by Qiao et al. in the previous work.3)) This number of the transmuted Zn atom accompanies the liberated energy of 6.0×10^{7} J (= 1.1×10^{-12} J $\times1.0\times10^{21}$). Thus, if the experiment lasted about one year before the NAA was performed (as in the case of the former experiment3)), this energy corresponds the excess heat generated per year;

$Q_{\mathrm{Zn}} = 6.0\times10^{7}$ J/y.

On the other hand, the maximum excess energy of 25.9 W corresponds to 8.15×10^{8} J/y (2.59×10 J/s $\times3.15 \times10^{7}$ s/y) if it lasted whole one year;

$Q_{\mathrm{heat}}(\mathrm{Max}) = 8.15 \times10^{8}$ J/y.

This value is compared with the above excess heat 6.0×10^{7} J/s expected from the amount of Zn atoms observed in NT experiment. The accordance is fairly good if we consider ambiguity in some unknown factors assumed in the calculation.

If we use the average value of the excess heat 0.23 W, we obtain a value two orders of magnitude smaller than this:

$Q_{\mathrm{heat}}(\mathrm{Av}) = 7.2 \times10^{6}$ J/y.

This value is one order of magnitude under Q_{Zn} given above which is considered fairly well with the same reason discussed in the preceding paragraph.

The relation (4) is used to determine the adjustable parameter n_{n} from the experimental data of the excess heat or the generated Zn atoms. We use here the maximum excess heat of 25.9 W and the liberated energy 6.185 MeV (1.1×10^{-12} J) per reaction. The value 25.9 J/s corresponds to 2.4×10^{13} reactions per second which is the value on the left-side term of the relation (4). Substituting corresponding values in the relation (4), we obtain the value of n_{n} at the maximum excess heat generation as follows:

$n_{\mathrm{n}} = 3.4\times10^{10}\ \mathrm{cm}^{-3}$.

This value is in the middle of values we obtained in analyses of various more than 50 experimental data sets in these several years and seems appropriate for a sample with large S/V ratio as 118 in this experiment.

4. Discussion

As shown in this paper, the experimental data sets obtained by X.Z. Li et al. in China are explained in themselves in two orders of magnitude uncertainty quantitatively and consistently with other data sets even if there are ambiguity in the description of the experimental condition and results.

In the explanation of the experimental data sets given above, we have used some assumptions on the nuclear reactions in Pd solids to interpret abnormal results from conventional viewpoint in nuclear physics: instantaneous induced fission of the compound nucleus ${}^{109}{}_{46}\mathrm{Pd}^*$ formed by a reaction n + ${}^{108}{}_{46}\mathrm{Pd}$ into Zn and S in solids. This fission reaction is almost impossible in free space to occur.

Similar anomalous reactions have to be assumed[2)] to explain experimental results of NT obtained in the cold fusion phenomenon where also observed anomalous events of the excess heat, tritium and helium generation and 2.45 MeV neutron emission. The anomalous reactions noticed in the CF materials include (1) decay time shortening and (2) induced fission of compound nucleus ${}^{A+1}{}_{Z}\mathrm{M}^*$ formed from a nucleus ${}^{A}{}_{Z}\mathrm{M}$ and a neutron n. Examples of the decay time shortening are written down as follows (Observed elements are underlined):

${}^{107}{}_{46}\mathrm{Pd}^* = {}^{109}{}_{47}\underline{\mathrm{Ag}} + e^- + \underline{\nu_e} + 1.166\ \mathrm{MeV}.$
${}^{197}{}_{78}\mathrm{Pt}^* = {}^{197}{}_{79}\underline{\mathrm{Au}} + e^- + \underline{\nu_e} + 0.719\ \mathrm{MeV},$
${}^{111}{}_{46}\mathrm{Pd}^* = {}^{107}{}_{44}\mathrm{Ru} + {}^{4}{}_{2}\underline{\mathrm{He}} + 1.0\ \mathrm{MeV}.$

Examples of the induced fission in addition to the reaction (3) are written down as follows:

${}^{197}{}_{79}\mathrm{Au}^* = {}^{57}{}_{26}\underline{\mathrm{Fe}} + {}^{141}{}_{53}\mathrm{I}^* + 97.9\ \mathrm{MeV},$
${}^{109}{}_{46}\mathrm{Pd}^* = {}^{51}{}_{22}\mathrm{Ti}^* + {}^{55}{}_{24}\mathrm{Cr}^* + 3n + Q,$

followed by β-decays of the unstable nuclei with asterisk.

If we take these experimental data sets as signals obtained by probes inserted into complex materials prepared by techniques devised for the cold fusion experiment, we have to investigate them with a viewpoint outside the conventional physics which have been established on the normal phenomena not including the events discovered in the cold fusion phenomenon.

Our trial has resulted in a new approach, the TNCF model based on the trapped thermal neutrons, and some features of the trapped neutron have been worked out; the neutron Bloch wave and its behavior at crystal boundary are typical examples of them. Nuclear reaction between the neutron Bloch waves and a nucleus in the boundary layer will be responsible to the above mentioned decay time shortening and the induced fission of the compound nuclei and should be the key to solve the problem of the cold fusion phenomenon.

The recent experimental data showing ^{4}He generation in Pd-D_2 gas system by Botta et al.[7)] has been analyzed successfully by the TNCF model assuming the time-constant shortening for α-decay of $^{109}_{46}$Pd and the result is presented in this issue.[9)] This is another evidence showing effectiveness of the TNCF model.

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References

(1) H. Kozima, K. Kaki and M. Ohta, "Anomalous Phenomenon in Solids Described by the TNCF Model", *Fusion Technology* **33**, 52 (1998).

(2) H. Kozima, H. Kudoh and K. Yoshimoto, "Nuclear Transmutation by Fission in Cold Fusion Experiment Analyzed by TNCF Model", *Cold Fusion* **25**, 34 (1998).

(3) G.S. Qiao, X.M. Han, L.C. Kong and X.Z. Li, "Nuclear Transmutation in a Gas-Loading H/Pd System" *J. New Energy* **2-2**, 48 (1997).

(4) X.Z. Li, W.Z. Yue, G.S. Huang, H. Shi, L. Gao, M.L. Liu and F.S. Bu, "'Excess Heat' Measurement in Gas-Loading D/Pd System", *Progress in New Hydrogen Energy (Proc. ICCF6)* (1996, Hokkaido, Japan), 455 (1996).

(5) H. Kozima, H. Kudoh and K. Yoshimoto, "Nuclear Transmutation by Fission in Cold Fusion Experiment Analyzed by TNCF Model", *Cold Fusion* **25**, 34 (1998).

(6) X.Z. Li, S.X. Zheng, G.S. Huang, W.Z. Yu, "New Measurements of Excess Heat in a Gas-Loaded D/Pd System", *The Best Ever! (Proc. ICCF7)* (1998, Vancouver, Canada), p.197 (1998).

(7) D.W. Mo, Q.S. Cai, L.M. Wang, S.Z. Wang and X.Z. Li, "The Confirmation of Nuclear Transmutation Phenomenon in a Gas-Loading H/Pd System using NAA (Neutron Activation Analysis)", *The Best Ever! (Proc. ICCF7)* (1998, Vancouver, Canada), p.259 (1998).

(8) E. Botta, T. Bressani, C. Fanara and F. Iazzi, "Measurement of ^{4}He Production from D_2 Gas-Loaded Pd Sample" *Progress in New Hydrogen Energy (Proc. ICCF6)* (1996, Hokkaido, Japan), p.29 (1996).

(9) H. Kozima, M. Ohta, M. Fujii, K. Arai and H. Kudoh, "Possible Explanation of ^{4}He Production in Pd/D_2 System by TNCF Model" *Fusion Technol.* **40**, 86 (2001).