

*Reports of CFRL (Cold Fusion Research Laboratory), 1-2, pp. 1 – 12 (2004)*

## **Analysis of Nuclear Transmutation and Excess Heat Generation in Pd/D/Li Systems by TNCF Model**

H. Kozima, K. Arai<sup>1</sup>, M. Ohta<sup>2</sup>, M. Fujii<sup>3</sup> and H. Kudoh<sup>3</sup>

Cold Fusion Research Laboratory, Materials and Energy Research Institute Tokyo Ltd., Tokyo<sup>1</sup>, Department of Nuclear Engineering, Osaka University, Osaka<sup>2</sup>, Department of Energy Engineering, Yokohama National University, Yokohama<sup>3</sup>.

e-mail cf-lab.kozima@nifty.ne.jp

### **Synopsis**

Excess heat and NT data (especially Ti generation) obtained in Pd cathodes with heavy water and LiOD obtained by Iwamura et al. were analyzed on the TNCF model. Production of the observed new element Ti was explained by nuclear fission of compound nuclei formed by  $n + {}^{A}_{46}\text{Pd}$  reaction. The adjustable parameter  $n_n$  in the model was determined by the data of NT as  $10^{10} - 10^{11} \text{ cm}^{-3}$  consistent with values determined in other experimental situations. The data of the excess heat and NT were explained quantitatively and consistently in a factor of 2.

### **1. Introduction**

There have been obtained in these several years many experimental data sets showing nuclear transmutations (NT's) not only in D<sub>2</sub>O but also H<sub>2</sub>O system explained only taking into possibilities of induced decay and fission of compound nuclei formed from nuclei in solids by absorption of a thermal neutron, as shown in our previous papers.<sup>1,2)</sup> These facts show clearly main reactions in the cold fusion (CF) phenomenon are not  $d - d$  but other reactions where participate electrolyte elements, cathode metals and others including background thermal neutron from our point of view. The assumed nuclear reactions in the explanation of these NT's are, however, difficult to consider their occurrence by absorption of a thermal neutron in free space and can be taken as showing a characteristic of the CF phenomenon observed in solids.

A group in Mitsubishi Heavy Industry has been working extensively with devices specifically invented to observation of the excess heat, NT, neutron and X-ray in the standard Pd/D/Li system. Their data has shown occurrence of NT producing especially Ti with fairly large amount exceeding extent of errors in experiments.

The experimental data<sup>3,4)</sup> presented at ICCF7 and in Fusion Technology by Y. Iwamura et al. are quantitative in the excess heat and transmuted nuclei feasible to analyze them on the TNCF model and the result is given in this paper. Several parameters of experimental setup were given by them as private communications in response to our requests.

## 2. Experimental Results

A new type of experimental apparatus<sup>3,4)</sup> was developed to induce continuous diffusion of deuterium in which an electrochemical cell for calorimetry and a vacuum chamber for nuclear measurement are divided by a Pd sheet (and a multi-layer cathode sheet of composition Pd/PdCaO/Pd). Continuous X-ray ranging from 10 to 100 keV and neutron and excess heat production are observed using the apparatus. Ti atoms are detected on the surface where D atoms pass through on Pd cathodes after electrolysis. Quantitative investigation of the result showed that detected Ti atoms could not be explained by contamination.

### 2.1 Normal Pd sheet cathode

In the experiment EV27 with a normal Pd sheet cathode ( $2.5 \times 2.5 \times 0.1$  cm<sup>3</sup>), the excess energy of 1 W had lasted a day without any X-ray and neutron emission. On the counter part of the cathode to the mesh Pt anode (a disk with a diameter 1.2 cm), there appeared a circular region (part A) with dark color diameter of which was  $\approx 1.2$  cm where observed Ti by EDX, WDX, AES, XPS. Total amount of Ti atom in the part A is estimated at 21  $\mu$ g (i.e.  $2.6 \times 10^{17}$  atoms) by ICP-MS (Inductively Coupled Plasma Mass Spectroscopy) which could not be explained by contamination. In addition to Ti, Fe was detected by ICP-MS in the sample.

### 2.2 Multi-layer Pd/PdCaO/Pd cathode

The excess heat of 0.52–0.83 W, X-ray and neutron (in only a single sample in the experiment EV50) had been observed. In the part A (defined in

2.1), Fe atom with an isotopic ratio  $x \equiv {}^{57}\text{Fe}/{}^{56}\text{Fe}$  of 0.036 – 0.66 (natural abundance = 0.023) was confirmed by SIMS (Secondary Ion Mass Spectrometry).

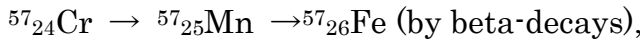
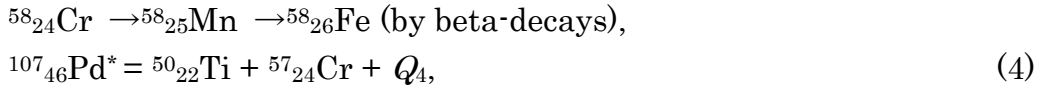
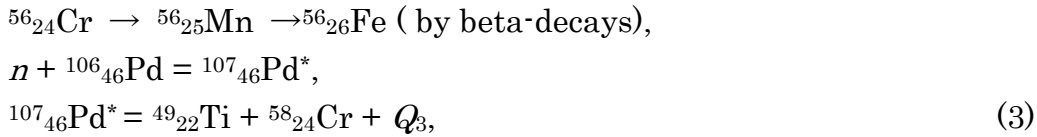
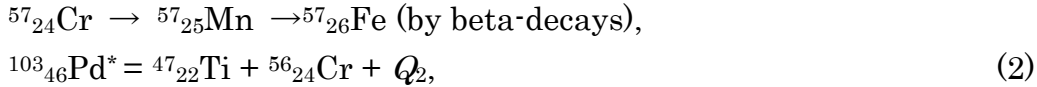
### 3. Analysis of the data on the TNCF model

The experimental data given in Subsections 2.1 and 2.2 were analyzed by the TNCF model proposed by us<sup>5)</sup> and the arbitrary parameter  $n_n$  was determined.

#### 3.1 Data of the normal Pd cathode

We analyze the experimental result given in 2.1 of the excess heat and Ti and Fe production, the main NT products in the part A was Ti.

The probable reactions in the Pd cathode relevant with these products can be assumed to be the following ones:



where the excess heat due to  $Q_1$ ,  $Q_2$ ,  $Q_3$ , and  $Q_4$  and succeeding  $\beta$ -decays are estimated at  $\approx 24.9$ , 26.2, 29.3 and 30.2 MeV, respectively, from mass differences of the corresponding reactions. We have no data of branching ratios of the reactions (1), (2) and (3), (4) which will be assumed equal each other in the following analysis.

According to the recipe of the TNCF model, we can determine the adjustable parameter  $n_n$  of the model from the amount of Ti atoms  $N_{\text{Ti}}$  using the relation between them;

$$N_{\text{Ti}} = 0.35 n_n V_n n_{\text{Pd}} V_A (X_A \sigma_A) \tau \xi, \quad (5)$$

where  $0.35 n_n v_n$  is the flow density of the thermal neutron per unit area and time,  $n_{\text{Pd}}$  is the density of Pd in the reaction region with volume  $V_A$ ,  $x_A$  and  $\sigma_A$  are the abundance ratio and the cross section of the reaction of an isotope  $^{A}_{46}\text{Pd}$  ( $\sigma_{102}$  and  $\sigma_{106}$  are 3.36 and 0.303 b, respectively), and  $\tau$  is the experimental time. The factor  $\xi$  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  $3.4 \mu\text{m}$  (reaction region) in this case according to the recipe of the TNCF model.<sup>1)</sup> (It is noticed in recent analyses<sup>6)</sup> of the data obtained by Ohmori et al.<sup>7)</sup> that  $\xi$  should be taken larger than 1 if we consider large reactivity of the trapped neutron in the surface layer resulting in anomalous nuclear reactions like micro-crater formation, decay-time shortening and induced fission.)

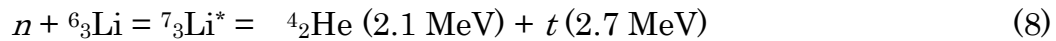
Production of Ti atoms by reactions written above should be occurring in whole period from the electrolysis to ICP-MS measurement ( $\tau_{\text{Ti}} = 3 - 5$  months) in the surface layer of the sample with a thickness of  $3.4 \mu\text{m}$  (determined by the experiment). Then, we put  $N_{\text{Ti}} = 3.34 \times 10^{10} \text{ s}^{-1}$  (assuming  $\tau_{\text{Ti}} = 3$  months) and other parameters into the expression to obtain

$$n_n = 1.42 \times 10^{11} \text{ cm}^{-3}. \quad (6)$$

This value of  $n_n$  gives the excess heat  $Q_{\text{NT}}$  by the reactions (1) – (4) and the values of the excess heat given below these reactions

$$Q_{\text{NT}} = 0.13 \text{ W}. \quad (7)$$

Another reaction



with a cross section  $9.40 \times 10^2 \text{ b}$  occurring in the surface layer with a thickness  $1 \mu\text{m}$  (we use this value assumed commonly in our model in the following analysis) contribute to the excess heat by an amount  $Q_{\text{Li}}$ . Using the value  $n_n$  given in (6), we can calculate  $Q_{\text{Li}}$  as

$$Q_{\text{Li}} = 3.06 \text{ W}. \quad (9)$$

Therefore, the excess heat expected by the TNCF model  $Q_{\text{th}}$  becomes

$$Q_{\text{th}} = Q_{\text{NT}} + Q_{\text{Li}} = 3.19 \text{ W} \quad (10)$$

to be compared with the experimental value given in Section 2

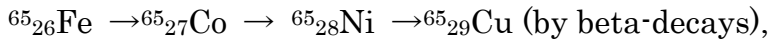
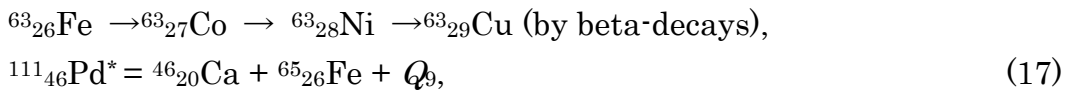
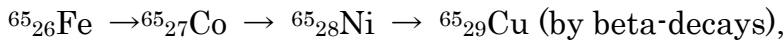
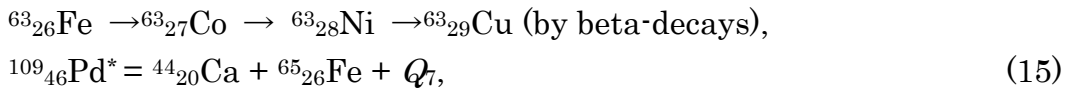
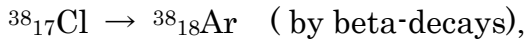
$$Q_{\text{exp}} \approx 1 \text{ W}. \quad (11)$$

The coincidence of these values by a factor 3 should be considered very good if we realize many assumptions made in the calculation.

### 3.2 Multi-layer cathode

To explain the products Fe and Cu in the multi-layer cathode, we have to take following fission reactions into our consideration instead of the reactions (1) – (4). This arbitrariness in selection for fission channels depending of the experimental result should due to the nature of the surface layers in interaction with neutron Bloch waves and will be discussed later.

The reactions relevant to the Fe and Cu production are assumed as follows;



The excess heat by  $Q_5 - Q_9$  and succeeding  $\beta$ -decays are determined by mass differences of the initial and final elements as 22.61, 27.74, 27.75, 32.34 and 32.94 MeV, respectively.

The numerical result of the isotope ratio  $^{57}\text{Fe}/^{56}\text{Fe}$  in the experiment gives us an important information about nuclear reactions in the surface layer. As long as we consider above mentioned fission reactions, this ratio is determined by branching ratio of the reactions (1), (2), (12) and (13). In the experiment EV52 where observed a value  $x = 0.66$ , we assume the reaction (12) was dominant because there was detected no Ti. The number of atoms of  $^{57}\text{Fe}$  and  $^{56}\text{Fe}$  in the sample before experiment should be  $1.13 \times 10^{13}$  and  $4.74 \times 10^{14}$  by  $x = 0.023$  and experimental value of Fe amount  $4.85 \times 10^{14}$  atoms. Denoting the number of  $^{57}\text{Fe}$  atoms produced in the experiment with duration  $\tau$  as  $\tau N_{\text{NT}}$ , we obtain a relation

$$0.66 = (1.13 \times 10^{13} + \tau N_{\text{NT}}) / 4.74 \times 10^{14}.$$

This relation gives

$$\tau N_{\text{NT}} = 3.02 \times 10^{14} \text{ atoms.}$$

We assume  $\tau$  as the electrolysis time  $5.0 \times 10^5$  s (because we have no data about the time when SIMS was performed after the electrolysis),

$$N_{\text{NT}} = 6.03 \times 10^8 \text{ s}^{-1}.$$

From this value, we can calculate  $n_n$  using a relation similar to (5) as

$$n_n = 3.12 \times 10^{10} \text{ cm}^{-3}.$$

This value of  $n_n$  determines the excess heat  $Q_{\text{NT}}$  by NT due to reactions (12) – (17), predominant reactions in them are (14) and (15);

$$Q_{\text{NT}} = 0.18 \text{ W.}$$

The reaction (8) also contribute to the excess heat  $Q_{\text{Li}}$  also in this case by

$$Q_{\text{Li}} = 0.67 \text{ W.}$$

Thus, the excess heat as a whole is expressed as follows;

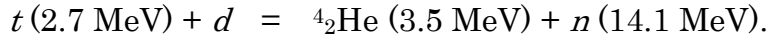
$$Q = Q_{\text{NT}} + Q_{\text{Li}} = 0.85 \text{ W}$$

to be compared with the experimental value  $Q_{\text{exp}}$

$$Q_{\text{exp}} = 0.52 \text{ -- } 0.83 \text{ W.}$$

The coincidence of these values in this case is too good perhaps by accident if we consider many unknown factors and assumed parameters in the calculation.

The neutrons with energies higher than thermal were observed in the experiment EV50 (where  $Q_{\text{exp}} = 0.83 \text{ W}$ ) could be explained by such a reaction induced by energetic triton generated in the reaction (8) as followings expected from our model



Using cross section of this reaction 0.14 b and  $n_n$  given above, we can calculate number of this reactions  $N_n$  as

$$N_n = 6.0 \times 10^5 \text{ s}^{-1}.$$

This is an enough value to be observed above the usual level of background neutrons of about few neutrons per second.

#### 4. Discussion

We could give a consistent explanation of the excess heat and NT (and neutron detection) in Pd/D/Li systems performed by Iwamura et al. on our TNCF model.<sup>8)</sup> In the explanation, we assumed some fission reactions considered improbable to occur in solids at near room temperature. To explain the experimental data, we assumed branching ratios of the relevant reactions which have not determined before due to non-existence of them in free space experiment. In other words, we can guess possible occurrence of

these fission reactions in solids and their branching ratios from experimental data obtained in CF experiments.

In view of scarce data of interactions of the neutron Bloch waves and nucleus in boundary region of solids, especially due to the local coherence of Bloch waves at a reflecting boundary,<sup>9)</sup> the events in the cold fusion phenomenon are precious keys to explore new features of thermal neutron physics in solids with promising applications related with liberation of nuclear energy and remediation of hazardous radioactivity.

### **Acknowledgment**

The authors would like to express their thanks to Dr. Y. Iwamura of Mitsubishi Heavy Industry for private communication about some details of his experiment used in this analysis. They are also indebted to J. Dash of Physics Department of Portland State University for valuable discussions in preparation of the revised version of this paper. This work is supported partially by a gift from the New York Community Trust and by the U.S. Army Research Office under grant number DAAG 55-97-1-0357.

### **References**

- (1) H. Kozima, K. Kaki and M. Ohta, "Anomalous Phenomenon in Solids Described by the TNCF Model", *Fusion Technol.* **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) Y. Iwamura, T. Itoh, N. Gotoh, M. Sakano and I. Toyoda, "Detection of Anomalous Elements, X-ray and Excess Heat induced by Continuous Diffusion of Deuterium through Multi-layer Cathode (Pd/CaO/Pd)", *Proc. ICCF7*(1998, Vancouver, Canada), 167 (1998).
- (4) Y. Iwamura, T. Itoh, N. Gotoh and I. Toyoda, "Detection of Anomalous Elements, X-ray and Excess Heat in a D<sub>2</sub>-Pd System and its Interpretation by the Electron-induced Nuclear Reaction Model", *Fusion Technol.* **33**, 476 (1998). And also private communications.
- (5) H. Kozima, "Trapped Neutron Catalyzed Fusion of Deuterons and Protons in Inhomogeneous Solids", *Trans. Fusion Technol.* **24** (*Proc. ICCF4*), 508 (1994).
- (6) H. Kozima, M. Fujii, K. Arai, H. Kudoh and K. Yoshimoto, "Analysis of

NT in Micro-craters and Excess Heat Data in W/H/Na(K) Systems by TNCF Model", *Reports of CFRL* (to be published).

(7) T. Ohmori and T. Mizuno, "Strong Excess Energy Evolution, New Element Production, and Electromagnetic Wave and/or Neutron Emission in the Light Water Electrolysis with a Tungsten Cathode", *Proc. ICCF7* (1998, Vancouver, Canada), 279 (1998). And also private communications.

(8) H. Kozima, *Discovery of the Cold Fusion Phenomenon – Evolution of the Solid State - Nuclear Physics and the Energy Crisis in 21st Century*, Ohtake Shuppan Inc., Tokyo, Japan, 1997.

(9) H. Kozima, K. Arai, M. Fujii, H. Kudoh, K. Yoshimoto and K. Kaki, "Nuclear Reactions in Surface Layer of Deuterium-Loaded Solids" *Fusion Technol.* **36**, 337 (1999).