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Comment on the Paper

A Comment on the Paper "Transmutation of Palladium Induced by Laser Irradiation," by J.-P. Biberian, L. Forsley and P. Mosier-Boss published in the *J. Condensed Matter Nucl. Sci.,* 37, pp. 9–22 (2023), ISSN 2227–3123.

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Abstract

The experimental result reported by Biberian et al. published in *J. Condensed Matter Nuclear Science* (**37**, pp. 9 – 22 (2023)) is analyzed from our point of view. We have found several ambiguities in their report and in this paper we propose further experiments to clarify the problems related to the mechanism of nuclear transmutations in the cold fusion phenomenon.

Keywords: Cold fusion phenomenon, SEM-EDX, nuclear transmutation, fission reaction.

1. Introduction

Since the announcement of the discovery of the cold fusion phenomenon (CFP) or nuclear reactions in the Pd-D system by Fleischmann et al. in 1989 [1], CFP research has expanded to a far larger range of events over almost all phenomena including generation of various new nuclides by the nuclear transmutation of element nuclei in the original sample, and emission of light nuclides (such as neutron, proton, triton, and ${}^4{}_2$He) in addition to the original excess heat observed in various transition metal hydrides and deuterides. The systems where CFP has been observed has extended to transition metals and their binary and ternary compounds including hydrogen and/or deuterium.

The experiment performed by Biberian et al. [2] is a work with a new material and a new technique originated by Mastromatteo [3] using a sample supplied by him. Biberian et al. performed the experiment using H_2 and D_2 instead of H_2 in the experiment by Mastromatteo and analyzed the Pd samples using the SEM-EDX technique after experiments. They concluded that they found elements generated as the result of a fission process which is induced by hydrogen absorbed in a thin-film sample of palladium under the excitation of a 5 mW laser pointer (650 nm of wavelength).

In this paper, we investigate the result of the experiment performed by Biberian et al. [2] and propose further experiments to confirm several ambiguities found in the result reported in their paper.

Table 1. Results of EDX analysis at Points 1 – 9 [Biberian 2023 (Section 3, pp. 11 - 13)].

Point	Elements (%)
1	N (10.44), O (68.22), Na(6.44), Al(0.42), Si(0.24), P(0.07), S(1.46), Mn(0.14), Fe(2.94), Ni(8.83), Zn(0.53), Pd(0.25)
2	N(14.55), O(67.06), Na(2.17), Ca(0.68), Mn(0.42), Fe(1.05), Ni(6.08), Zn(0.40), Mo(0.29), Pd(7.28)
3	N(11.09), O(52.11), Na(5.60), S(2.12), Ca(0.46), Mn(1.13), Fe(5.34), Ni(15.08), Pd(7.04)
4	N(18.59), O(58.75), Na(1.58), Fe(0.70), Ni(2.98), Pd(17.40)
5	O(41.68), Na(2.87), Pd(55.45)
6	N(13.88), O(56.39), Na(2.00), Fe(0.75), Ni(1.57), Pd(25.41)
7	O(4.40), Pd(95.60)
8	O(3.58), Pd(96.42)

Table 2. Results of EDX analysis at Point (arbitrary chosen on the sample surface) [2 (Section 4, p. 13)].

Point	Elements (%)
arbitrary	O (0.01), Si (0.12), Ca (0.09), Pd (99.78)

2. Experimental Results

Biberian et al. [2] performed experiments similar to that performed by Mastromatteo [3] using two samples provided by him. The samples they used had the following structure: a silicon wafer with a thermal oxide layer (about 500 nm), then a thin layer of titanium (30 nm) for adhesion of the palladium film (500 nm).

The 5 mW, 650 nm red laser (no brand name) is not focused, therefore it covers an area of about 4 mm^2. The quartz tube chamber containing the sample is evacuated with a mechanical pump down to about 2×1 mbar, then hydrogen gas is introduced at a pressure of about 2 bars (absolute pressure). Experiments have been performed with hydrogen and deuterium with two identical samples. Each sample is 1 cm^2 silicon chip with a 250 nm thin film of palladium deposited at their surfaces. Both experiments lasted about three months.

2.1. Elemental Chemical Analysis of the Sample under Hydrogen

In the experiment with the first sample in hydrogen atmosphere, several spots appeared at the center of the sample. The largest spot was about 180 μm in diameter, with several smaller ones around it. EDX analysis of the chemical composition was performed at points 1 to 9 in and around the largest spot as explained below.

Points 1 to 3 are at the central part of the largest spot, 4 to 6 at the peripheral part of the largest spot, and 7 to 9 at points outside of the largest spot in an area where no spot exists.

2.2. Elemental Chemical Analysis of the Sample under Deuterium

The experiment with the second sample in deuterium atmosphere was performed under the same conditions as for hydrogen. However, contrary to the hydrogen sample, no hot spot was observed and the elemental analysis on the surface has shown only palladium as seen in the following data (at a point on the surface).

3. Analysis of the Experimental Results

It should be mentioned that the temperature of the sample where the cold fusion phenomenon occurs is a key factor for the occurrence of CFP. This has long been noted (cf. for example [4]). The use of laser beam irradiation on the sample in the experiments by Biberian et al. [2] belongs to this type of experimental technique for the effective excitation of

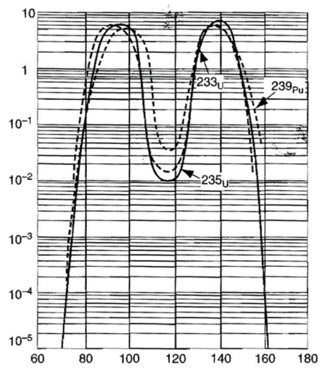

Figure 1. Mass fission yield curves for ^{233}U + n (thermal), ^{235}U + n (thermal) and ^{239}Pu + n(thermal). Yield (in log scale) vs. Atomic Number are shown [11].

nuclear reactions in solids. It is desirable to measure the temperature of the samples in future experiments to know the direct effect of temperature on the occurrence of CFP more quantitatively, rather than the indirect effect of laser irradiation.

The analysis of experimental data obtained in transition metal hydrides and deuterides should be performed by a model based on the premises (consistent with each other) and also with common knowledge established in modern science. In this analysis, we use the TNCF model, a phenomenological model proposed in 1994 [5] and developed up until now [6], [7], [8].

3.1. Elemental Chemical Analysis

The data of elemental chemical analysis shown in Table 1 for the protium system in Sec. 2.1 and that in Table 2 for deuterium system in Sec. 2.2 pose several questions to understand. These questions will be explained in the following two subsections.

3.1.1. *Percentage Abundances of New Elements detected after the Experiments by Biberian et al. [2 Biberian 2023].*

In the various experiments performed since the discovery of the cold fusion phenomenon (CFP), we have some data of nuclear transmutation including data explicable only by the nuclear fission. One of these data, we have analyzed that obtained by Miley et al. [9] as explained in our book [10 (Section 2.5.3 Nuclear transmutation by fission)].

In the analysis of the data obtained in the multi-layer cathodes used by Miley et al. [9], we used the mass fission yield curves for ^{233}U + n (thermal), ^{235}U + n (thermal) and ^{239}Pu + n (thermal) shown in Fig. 1 ([10 (Fig. 2.6)]).

As we see in the data shown in Fig. 1, the two peaks appear in the mass fission yields curves for common fission reactions of ^{233}U, ^{235}U and ^{239}Pu. If we can extend this characteristic of the fission reaction to lower mass number cases as observed by Miley et al. [9 (Ni)] and Biberian et al. [2 (Pd)], we may observe fission product elements with mass numbers A distributed symmetrically at $A_0 = A_{Pd}/2 = 47.13$ with the average mass number of Pd isotopes; $A_{Pd} = 94.25$. We know that the element with the average mass number A_{av} close to 47.13 is titanium (Ti) and that with $A_{av} = A_{Pd}/4$ (= 23.6) is Mg, that with $A_{av} = 3A_{Pd}/4$ (= 70.7) is Ga and Ge.

Table 1 shows the above presumption about the mass fission yields does not apply to the data obtained by Biberian at al. [2]. We do not know how reconcile this dilemma between the probable assumption and the data tabulated in Table 1.

In addition to the problem of the mass distribution of detected elements, there is a question about the total mass investigated by EDX in the experiment by Biberian et al. [Biberian 2023]. We can only know the ratio of numbers of elements in percentage as shown in Tables 1 and 2 and not the numbers of element nuclei in a definite volume. Therefore, we could not determine the balance of total mass before and after the reaction in the surface region of the samples.

We use therefore a simple assumption that the addition of mass numbers of elements detected and tabulated in Tables 1 and 2 gives a qualitative measure of the average mass numbers at the point the data was taken. The data at Points 1 and 8 in Table 1 are simple examples to understand. For instance, the data at Point 8 (in the Light Area in Fig. 2 of [2 Biberian 2023]) gives us the following result of analysis. Using $A_{\rm pd}$ = 94.25. and $A_{\rm O}$ = 15.47, we obtain average mass number of elements at Point 8 as A_8 = 90.87. This value is close to the value $A_{\rm pd}$ = 94.25 before the measurement where the surface is covered by Pd only.

On the other hand, a similar analysis at Point 1 (in the Central Area in Fig. 2 of [2]) gives us following result; A_1 = 22.60 compared to the original Pd surface $A_{\rm pd}$ = 94.25.

This result raises us the question: Where has the missing mass gone (if our assumption made above is acceptable)? To answer this question, it will be necessary to know more details about the sample and the experimental process such as temperature of the sample during measurement, components of outgas from the chamber throughout the experiment, the three-dimensional structure of the hot spots, and so on.

3.1.2. *Localization of Nuclear Reactions in CFP*

Self-organization of the optimum superlattice for the nuclear reactions in the surface region of the sample had been pointed out to explain localization of nuclear reactions in CFP [6]. The experimental data of the large hot spot at the central part of the sample surface observed in the experiment by Biberian et al. [2] clearly shows another example of localization of nuclear reactions in CFP. Some discussions on this point will be given in the next subsection in relation to the difference between experimental data obtained in the protium and deuterium systems.

3.2. Effects of Protium and Deuterium

There are several experiments where the occurrence of CFP in Pd-H systems was observed even though it has long been known that the Pd-D system is more appropriate for CFP experiments. One of the typical examples of successful experiments in Pd-H system is explained in our analysis of experiments in critical and supra-critical conditions given by us in 2017 [12].

To investigate the protium and deuterium systems in CFP, we have to rely on a reference system for analysis of experimental data. We use the TNCF model proposed by us in 1994 at ICCF4 and developed since then [5], [7].

First of all, we want to cite the data of diffusion of hydrogen isotopes in transition metals. The data given in Figs. 2 and 3 show the typical data of diffusion of hydrogen isotopes in Pd showing the behavior of diffusion coefficients D_H and D_D in Pd. In the low temperature region below about 100 °C, the diffusion coefficient D_D of deuteron in Pd is larger than D_H, showing the inverse mass dependence, while in the upper temperature region the dependence reverses to the normal type.

We have pointed out that this complex dependence of diffusion coefficients D_H and D_D may be a cause of the characteristics of CFP in PdD and PdH systems [6], [10]. Considering the experimental condition of the experiments

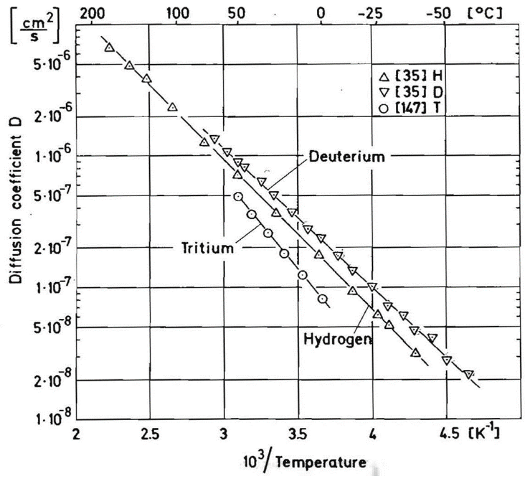

Figure 2. Diffusion coefficients of isotopes of hydrogen in Pd [13]. (Numbers in brackets refer to references in this paper).

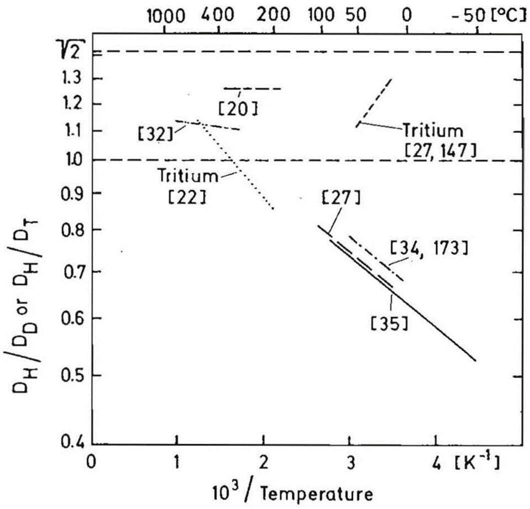

Figure 3. Ratios of diffusion coefficients of isotopes of hydrogen in Pd [13]. (Numbers in brackets refer to references of this paper).

done by Biberian et al. [2], the temperature of the sample may be heated up more than 100 °C and an optimum superlattice of Pd and H in PdH system is formed easily than in PdD system.

The appearance of the largest hot spot at the center of the sample in the experiment by Biberian et al. [2] may be also related to this characteristic. The temperature of the sample in the geometry of their experiment is necessarily higher in the central part than in the peripheral part due to the heating by laser beam and the larger loss of heat in the periphery even if the heating is homogeneous over the sample surface. Then, it is likely the formation of an optimum superlattice for CFP will occur more easily in the central part than in the periphery.

4. Conclusions

The experiment performed by Biberian et al. [2] has shown the occurrence of CFP related to or induced by nuclear fissions in a PdH system with specific sandwich structure with the irradiation of a laser beam. The characteristics of the obtained data suggest many interesting features of CFP observed hitherto by many researchers and it is desirable to continue the experiment to obtain more detailed data with refinement of experimental conditions and measurement apparatuses. We would like to add our opinion on the use of CR-39 technique which needs careful checking to deduce a definite conclusion, as we have pointed out before [14].

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