The Ninth International Conference on Cold Fusion. 2002. Beijing, China: Tsinghua University.

RELATION BETWEEN NEUTRON EVOLUTION AND DEUTERIUM PERMEATION WITH A PALLADIUM ELECTRODE

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Abstract

We observed neutron emissions from palladium after it absorbed deuterium from heavy water followed by hydrogen from light water. The neutron count, the duration of the release and the time of the release after electrolysis was initiated all fluctuated considerably. Neutron emissions were observed in five out of ten test cases. In all previous experiments reported, only heavy water was used, and light water was absorbed only in accidental contamination. Compared to these deuterium results, the neutron count is orders of magnitude higher, and reproducibility is much improved.

1. Introduction

It has been reported that palladium electrolyzed in heavy water produces excess heat.(1) However, the mechanism stills remaining unknown because research is hampered by poor reproducibility and lack of control. The very existence of the reaction is often called into question. On the other hand, based on several reliable reports of neutrons and other fusion products, many researchers assume that the mechanism involves nuclear fusion; however, many problems remain with this assumption. (2-5)

The authors have examined many of the reports available to date of neutrons and heat, and have reached the following conclusions. First, when neutrons and excess heat are observed, they usually appear after electrolysis has continued for a long time. Second, many instances have been reported in which these effects occur after the cell is replenished with new electrolyte. Third, it is known that when electrolysis is used to absorb deuterium into palladium, at first, the electrolyte contains almost pure heavy water but later, it becomes mixed with light water. (6) This substitution occurs because heavy water is hydrophilic, and light water permeates even a nominally closed cell at some stage during the process, gradually diluting the heavy water.

Based on these observations, we conclude that this reaction must require something more than the absorption of deuterium. In particular, after electrolysis has continued for a long time and the heavy water has been replenished, light water is likely to be mixed in the electrolyte. Also, after electrolysis loading has reached a certain point, any hydrogen present in the cell will migrate to the cathode and block the absorption of additional deuterium. In view of these facts, we predict that the reaction cannot occur with deuterium absorption alone, and that it requires certain triggering events.

2. Experimental

Palladium wire, 99.9% purity, 1mm in diameter and 3cm in length was used as the cathode. Tungsten lead wire, 1.5mm diameter and 150mm in length, was welded to the sample electrode. The electrolyte was composed of 100% pure heavy water from Acros Organics, and K2CO3 reagent from Kanto Kagaku Corp., which was 99.5% pure, and was adjusted to 0.2M concentration. The reagent was heated to 300ºC in an electric furnace to evaporate all water in it. The palladium sample was placed in a U-form quartz glass cell containing 100g of heavy water electrolyte. Cathode room was divided from the anode that had a platinum mesh served as the anode electrode. Electrolysis was typically performed for three hours. Immediately after that, the palladium cathode was transferred to a light water cell where light hydrogen absorption was performed. At this stage, voltage was 10V, and current was about 2A. The light water cell was made of Pyrex, and it was 10cm diameter and 20cm high, with a silicon rubber lid. [7] The palladium was introduced through a hole in the lid. The cell was also equipped with a platinum mesh anode in a configuration similar to the heavy water cell. In this phase, voltage was 40V and current reached a maximum of 8A; however, because the electrolyte temperature rose rapidly toward its boiling point, current was reduced to 1A.

Neutrons were measured with three He3 detectors placed 50cm above and apart from the cell. The detectors were calibrated with a standard Cf252 neutron source (2.58×10^4 decay/s). The background count was 0.008±0.003 c/s. The efficiency of the detectors was set at 4×10^{-5} by calibration measurement. To reduce noise, an electromagnetic shield covered the detectors. After calibration, neutrons and noise were distinguished by covering one of the detectors with a 0.5mm-thick Cd film. Neutron emission could be detected by the coincidence method with two of the detectors and the anti-coincidence method with one detector that was covered by the Cd film.

The measurement of deuterium purity in the electrolyte was estimated by mass analysis method. We have used a permeation technique through a Palladium film to estimate the deuterium purity. The electrolyte was put in an electrolysis cell and electrolyzed by several ten Volt. Another chamber is connected to vacuum system of the quadruple mass analysis. The mass number was estimated after reached a stable value. After that the concentration was decided by the mass number from 2 to 4 that means H_2, HD and D_2.

3.Experimental Results

The experiment was performed ten times. A typical result of neutron emission is shown in the Fig.1 this shows the neutron count rate as well as the input voltage, current, and electrolyte temperature during the run. In this example, voltage was raised to 85V at 3000s, and immediately after that, the detectors that were not covered by the Cd film observed 1 and 2 count rates. As shown in Fig.1, 25,800 neutrons were observed to emit from the cathode, with the count rate of 1 c/s. Neutron production peaked when voltage was raised, and 200s after that, fell to the background level again. During this period, the total neutron count was estimated as 1.57×10^6. In this example, electrolysis in light water continued for a considerable length of time and neutron emission was observed when voltage increased. In another run, neutron emissions were observed immediately after light water electrolysis commenced. Electrolyte temperature was 40ºC, and input voltage was 40V. The maximum count rate was 7.7/s, the duration time 20s and the total neutron count was estimated as 4.38×10^5.

Table 1 shows how differences in electrolysis conditions led to differences in neutron emissions. Column 1 shows sample number and column 2 is electrolyte temperature. When neutron emissions occur, they always occur after increase in voltage, either when electrolysis begins, or later on when voltage is increased to a higher level. Column 3 shows the voltage level before neutron emissions began. Voltage was zero for samples 1, 2, and 9, meaning they

produced neutrons immediately after electrolysis began. Column 4 shows the ending voltage after emissions ceased, or with samples 6 and 7, after a boost failed to produce neutrons. (Sample 3 remained at 25V during the entire run, with no boost, and failed to produce neutrons.) In some cases we tried raising voltage gradually while in others we increased it abruptly. Column 5 shows how much time was taken for each sample. Column 6 shows the total voltage increase that trigged an event, or the boost increase that failed to trigger an event, and column 7 shows the rate of increase. For example, with sample 7, we raised voltage from 70 to 90V over 15s, a total of 20V, at the rate of 1.33V per second, but no emission was detected. Column 8 shows the peak count, column 9 the total counts, and column 10 the duration of the emission event. Column 11 shows the average count rate per second, and column 12 shows the total number of neutrons extrapolated from the count, based on the calibration with the Cf252 neutron source.

It is clear from this table that in five examples, over 100,000 neutrons were observed, which is deemed a significant count. Emission performance was neither predictable nor controllable: neutron counts varied by an order of magnitude, from 10^5 to 10^6, and continued for the duration ranging from 2 to 200s. All emissions had a distinct pattern, namely a peak soon after the emission began, and a gradual decline. From the data in this table, we cannot yet establish a causal connection between neutron emissions and temperature, voltage, or other control parameters, but in samples that did produce neutrons; the degree of the total voltage increase does appear to correlate with the peak, average, and total neutron counts. Higher voltage correlates with higher neutron counts.

Figure 2 shows the relationship between the neutron emission rate and the deuterium purity in the solution. All the electrolytes were examined after finished the test experiment. However, all of the electrolytes were kept in a tightly stopped up bottle for several months. And after that they were measured by mass analysis. A dependence of the neutron emission rate on the deuterium purity is seen in the graph. However, the dependency was not good, so we conclude there are still other, unknown factors effecting neutron emission. Another fact that the electrolyte purity varied. It was measured at around 95% + - 3%. We assume this variation is caused by water contamination occurring during the preparation of the electrolyte, such as when the electrolyte is exposed to the atmosphere. Or it may be caused by different amounts of water in the reagent, or other factors.

Neutron emissions during light water absorption following heavy water absorption are very difficult to explain by the models proposed heretofore, which involve d-d fusion reactions. These other models assume that neutron emissions occur when heavy water alone is absorbed, and the emissions must be accompanied by excess heat and tritium production. The reaction we observed came about after alternating absorption of deuterium followed by hydrogen, and the reaction appeared to be highly reproducible, reliably generating high neutron emissions. We conclude that the models proposed heretofore based upon d-d reactions are inadequate to explain our present results, which involve hydrogen nuclear reactions.

4. Discussion

4.1 Fusion in Solids

Many theoretical models have been proposed and hypothesized to explain the anomalous result. That no visible fusion rates (e.g., d+d fusion rates) can take place in stationary states of metal deuterides is almost proved. [3] Possibilities may exist in transient states of deuteron motion in solids. If we require a very drastic enhancement of any fusion rates that can reach the observed heat level, the following three conditions should simultaneously be fulfilled:

1. A dynamic mechanism should exist for forming close clusters of more than two deuterons within 0.02nm (comparable to a de Broglie wavelength of 1eV deuteron) space in solids.

2. Quantum mechanical tunneling through the Coulomb barrier, i.e., barrier penetration probability should be enhanced very much, should be more than 10^{-6}; namely, we need super screening. To depress the Coulomb repulsive potential very much, squeezing of quasi-free electrons in metal deuterides during the transient dynamics should happen local points where deuteron clustering are taking place.

3. Anomalous enhancement should exist of inter-nuclear formation rates of the virtual compound nucleus, of the order of 10^6 times for the S value, compared with the known S value (~100 keV・b) of d + d fusion. This condition requires the existence of a third "heavy" particle (i.e., a proton or a deuteron, in addition to d+d) to drastically increase the "strong interaction." Hence, we come to the simultaneous multibody fusion of hydrogen isotopes.

If observed excess heat were out of the scope of nuclear reactions, we need to require conditions 1 and 2 only. In other words, condition 3 is essential to meet the nuclear origin of excess heat, but it is not considered in most models. The necessity of condition 3 is understood in the following explanation. The process of the fusion reaction can generally be divided into the following four steps:

1. the process of atomic motion is solids to form close pairs and clusters of hydrogen isotopes

2. barrier penetration with probability exp (-G), where G is the Gamow integral by the Wentzel-Kramers-Brillouin (WKB) approximation

3. the formation of an internuclear virtual compound nucleus by strong interaction

4. the splitting of the compound nucleus into branches of decay channels by the final state interactions. Further discussion can be finding other article[8].

4.2 H+D+D Fusion

The possible decay channels of ^{5}Li* (21.88 MeV) are shown in ref [8].

H+D+D→ ^{5}Li*(21.88 MeV), Spdd = 4×10^5 keV・b ;

Excited levels and decay schemes in reference [8], decay channels via lower excited states of ^{4}He* may take place because ^{4}He is a composite particle of nucleons (neutrons and protons). We do not know the branching ratios for the three decay channels. No gamma transition is expected. Levels shown with broken lines in ref. [8] are thresholds for particle emissions in parentheses, which is also the case in ref. [8] through 12.

4.3 D+D+D Fusion

Typical decay channels of ^{6}Li* (25.32 MeV) are shown in ref. [8]. Major decay channels are speculated by the spin-parity selection rules for the S-wave (1+, 3+) and the P wave (0-,2-) states of ^{6}Li* (25.32 MeV: Ī π) to be d(15.9 MeV: 1+) + α (7.9 MeV) and (4.75 MeV)+^{3}He (4.75 MeV). However, proton emission channel is also possible. If the 4 - state (P wave) dominates, gamma transitions to lower excited states of ^{6}Li* are also possible, as shown typically by ref [8]. Subbranches of the intermediate state excitations are shown in ref[8]. Similar subbranches are possible for the proton mission channels, as shown in Fig.10. In gamma-transition channels, typically as in reference[8], the lifetime of ^{6}Li* (25.32 MeV) is estimated as large as 10^{-15}s, which is much lager than the light traveling time 3×10^{-18}s for a 1-nm domain. Therefore, there may be energy transfer via QED photons to plasmon by nuclear/electromagnetic interaction, and no specific gamma rays [19.62 and 23.8 MeV] will be emitted. Instead, 19.62 and 23.8 MeV could be deposited as heat from plasmon oscillation (lattice vibration). No a priori knowledge is available to define branching ratios for many

channels. [8] The situation is complicated.

4.4 H+3D Fusion

With decay channels [8] the situation is more complicated. However, we expect that ^{3}He (16.77 MeV) + ^{4}He (12.57 MeV) and their sub-branches [2] may be major decay channels. Level schemes are given in ref [8]. Because excited levels of ^{3}He are not known, branch is an assumption. Other channels of deuteron, proton, neutron, and triton emissions are also possible. Channels to neutron and triton emissions associate continuum charged-particle spectra.

5. Conclusion

We have confirmed clear neutron emissions during hydrogen absorption into the palladium electrode followed after deuterium absorption. The neutron count and duration of the emission and the time during electrolysis was fluctuated considerably. Neutron emissions were observed in five test cases out of ten. Compared to experiments in which only deuterium was absorbed, repeatability was good and the neutron count was high. The multi-body fusion model in metal adequately explains the phenomena. Especially, the H+D+D fusion model seems to be the predominant model to understand the experimental facts.

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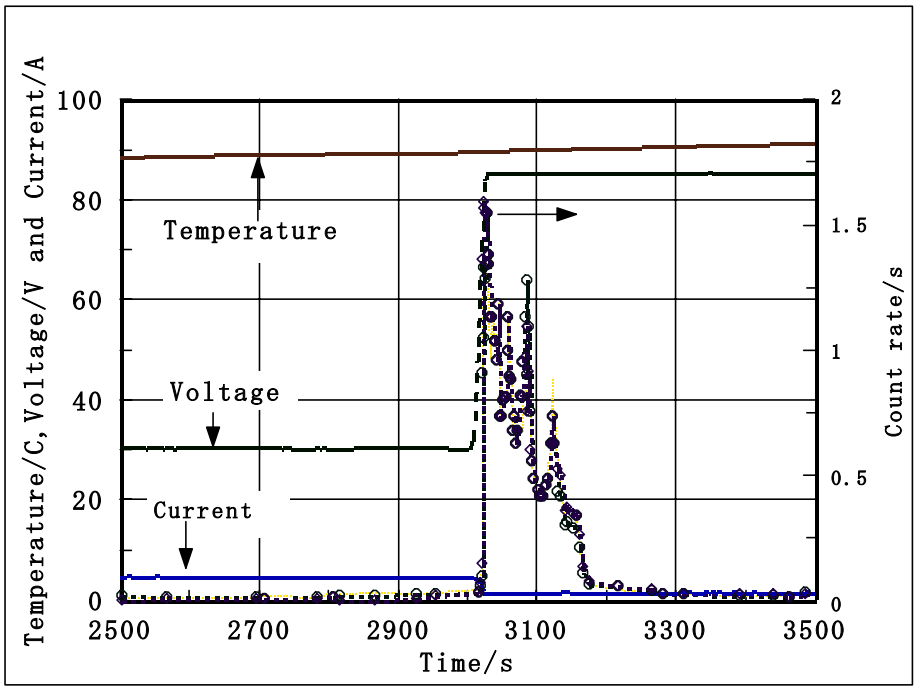

Figure 1: Neutron counts after absorbing deuterium and hydrogen. After 3000 s, electrolysis voltage was raised to 80 V, and neutron emissions suddenly occurred. (Data from sample 4.)

Sample No.	Cell Temp. (°C)	Starting Voltage	Ending Voltage	Duration of boost (s)	Voltage Boost (V)	Voltage change (V/s)	Peak Count	Total counts	Duration of Burst (s)	Count rate (c/s)	Total neutron burst
No. 1	**40**	**0**	**40**	**15**	**40**	**2.67**	**7.70**	**17**	**50**	**0.17**	**438600**
No. 2	**26**	**0**	**30**	**10**	**30**	**3.00**	**3.08**	**5**	**2.6**	**1.92**	**129000**
No. 3	**40**	**0**	**25**	**5**	**25**	**5.00**	**0**	**0**		**0**	**0**
No. 4	**90**	**30**	**83**	**20**	**53**	**2.65**	**1.54**	**61**	**200**	**0.305**	**1573800**
No. 5	**95**	**50**	**90**	**40**	**40**	**1.00**	**0.05**	**3**	**100**	**0.03**	**77400**
No. 6	**90**	**40**	**90**	**20**	**50**	**2.50**	**0**	**0**		**0**	**0**
No. 7	**90**	**70**	**90**	**15**	**20**	**1.33**	**0**	**0**		**0**	**0**
No. 8	**90**	**20**	**90**	**40**	**70**	**1.75**	**0.910**	**5**	**135**	**0.037**	**129000**
No. 9	**60**	**0**	**0**	**200**	**0**	**0**	**0.025**	**5**	**200**	**0.025**	**129000**
No. 10	**80**	**72**	**92**	**15**	**20**	**1.33**	**0.460**	**1**	**195**	**0.005**	**25800**

Table 1: Changes of the factors and neutron emission for various measurements.

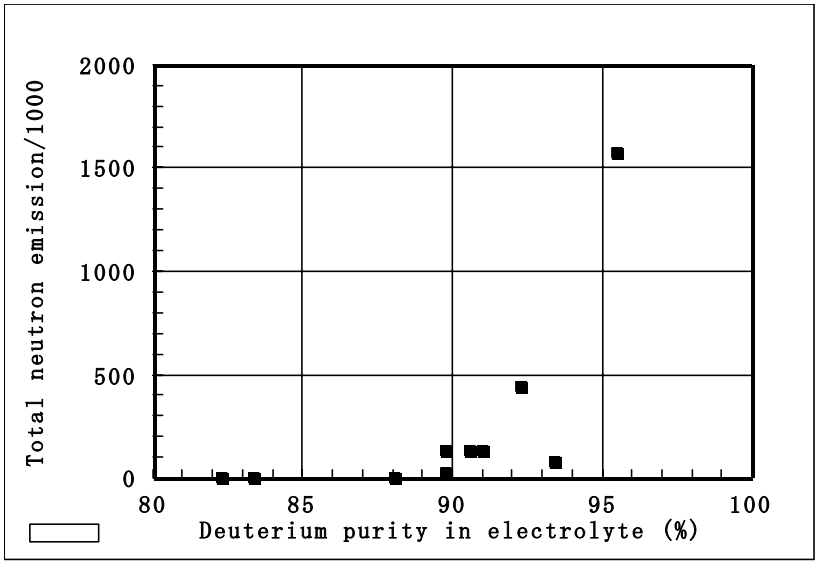

Figure 2: Relationship between neutron emission and deuterium purity of electrolyte.