

Tian, J., et al. "Excess heat" and "heat after death" in a gas loading hydrogen/palladium system. in *The 9th International Conference on Cold Fusion, Condensed Matter Nuclear Science*. 2002. Tsinghua Univ., Beijing, China: Tsinghua Univ. Press.

“EXCESS HEAT” AND “HEAT AFTER DEATH” IN A GAS-LOADING HYDROGEN/PALLADIUM SYSTEM

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ABSTRACT

“Super-absorption” could occasionally appear when an H/Pd gas-loaded system was heated by an incandescent tungsten filament. The system-pressure was kept on in 10^{-2} Pa for several hours without any aid of pumping. In this stage both “excess heat” and “heat after death” was observed once. By calibration, the feature constant of the system was $12.8^{\circ}\text{C}/\text{W}$ in maximum. When the “excess heat” appeared, the system temperature was reached 153°C at the highest whereas the stable inputting power was 0.45mW . When the “heat after death” appeared—there was no power input to the system—the temperature of the system abruptly jumped up to 761°C . In the former, 25kJ “excess heat” within 12 hours was measured which corresponded to $180\text{eV}/\text{atom Pd}$. And about 2MJ “heat after death” within 43 hours was corresponding to $13\text{keV}/\text{atom Pd}$. The maximum excess powers in each case were 3.3 and 49W and the correlated power density were 230 and $3600\text{W}/\text{cm}^3\text{Pd}$ respectively. The variation results of concentration of Li-7 and Li-6 and their abundance ratio in Pd suggested that the anomalous heat might come from a nuclear origin.

1. INTRODUCTION

The anomalous phenomena appeared in “cold fusion” have attracted many people studying on them for more than 13 years since 1989, although there are many inexplicable things in that research field. The most interesting and promising aspect in cold fusion research is that a solid, usually a kind of metal, could release a great deal of “excess heat” energy under some proper conditions especially after the metal absorbed a certain amount of deuterium or hydrogen. If the heat that the system produced was larger than the energy to keep the system working, the energy difference would be called “excess heat”. And if there were not any energy inputting to the system, the heat that still released from the system would be called “heat after death” or “after heat”.

There are two ways in the anomalous exothermic study. One of them is called “DRY” method—deuterium or hydrogen gas molecule were dissociated into two atoms on a metal’s surface and then the atoms were absorbed by the metal, so the system for this work is known to “gas-loading” system. The other is “WET”—deuterium or hydrogen atoms was separated out from heavy or light water by electrolysis and then also absorbed by the metal. Both methods have only one purpose: to observe various anomalous effects after D(H) being absorbed. However, the “DRY” method has a higher sensibility than the “WET” in heat detection because the heat capacity of a gas-loading system is much smaller than that of electrolysis system. In addition “DRY” system works without the constraint of boiling temperature, and can be easily heated to the temperature of more than 100°C . So the anomalous exothermic effect in a gas-loading system can be observed more efficiently and in a relatively wider temperature range.

Based on the previous experience, an anomalous exothermic effect with lower power density (less than one watt per cubic centimeter palladium) could be reproduced once and once again. But it has been difficult to reproduce the effect with higher power density (higher than one $\text{kW}/\text{cm}^3\text{Pd}$). At first, under what condition(s) a system can go into an anomalous stage is still unclear, and secondly the anomalous cannot appear again even if the same reaction conditions are exactly repeated. So searching the reproducible

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condition(s) for an anomalous exothermic effect with high power density would be a long-term task in cold fusion research.

Hydrogen has often been considered to be a control in either DRY or WET method since the research comes to the world. That is to say, The difference between hydrogen and deuterium interacted with the same kind of metal is regarded as a criterion by which whether a system is in the anomalous stage. This criterion seem to be reasonable for most cold fusion experiments^[1~3]. But is this criterion always right for all of the cases that anomalous phenomena appear? Does not the anomalous really exist when the hydrogen react with a metal (take Pd for example)?

Here we report an observation of an unusual anomalous phenomenon in a “DRY” system, although it has not been reproduce yet. Because it was observed in a H/Pd system, but not D/Pd system, it is worthwhile to report here in parallel with other H/Pd work reported in ICCF-9.

2. EXPERIMENTAL

2.1 Experimental system

Figure 1 shows the schematic of the experimental system. The double-Jacket stainless steel cell that we called reaction chamber has internal dimension $\phi = 10\text{cm}$ and $h = 16\text{cm}$, with useful capacity $\sim 1.3\text{dm}^3$. Palladium wire (99.99%) is made by the Institute of Aero-astronaut Ministry of China, with dimension of $\phi \times h = 0.01 \times 177\text{cm}$ ($\sim 1.4 \times 10^{-2}\text{cm}^3$). Before the experiment, the Pd wire was immersed in acetone and ethanol, respectively, for 20 minutes; then, cleaned by distilled water to get rid of impurities on the surface. Afterwards the wire was wound on a quartz frame. A platinum resistance thermometer and a piece of tungsten filament (it was taken from a small bulb of electrical light of 12 volts \times 8 watts) were put in between the frame, the former was for reading the temperature of Pd wire and the latter was for dissociating the hydrogen molecule into atoms. All of the electrical leads were introduced through an aero-connector to outside circuit. The reaction chamber was evacuated by a rotary pump (4 l/sec), and the pressure of the chamber was monitored by a vacuum gauge (RZJ-1D). The hydrogen gas was supplied by Beijing Huayuan Gas Company (Purity 99.99%). An electrical power supplier for heating Pd, W wire was a DC power (WYK-302B Model), which was made by Jintong Electronic Apparatus Limited Company, Yangzhou, China. Several high-precision digital multi-meters (DT-9203) were used for recording the voltages and currents of Pd and W circuits and the temperature of Pd and the ambient.

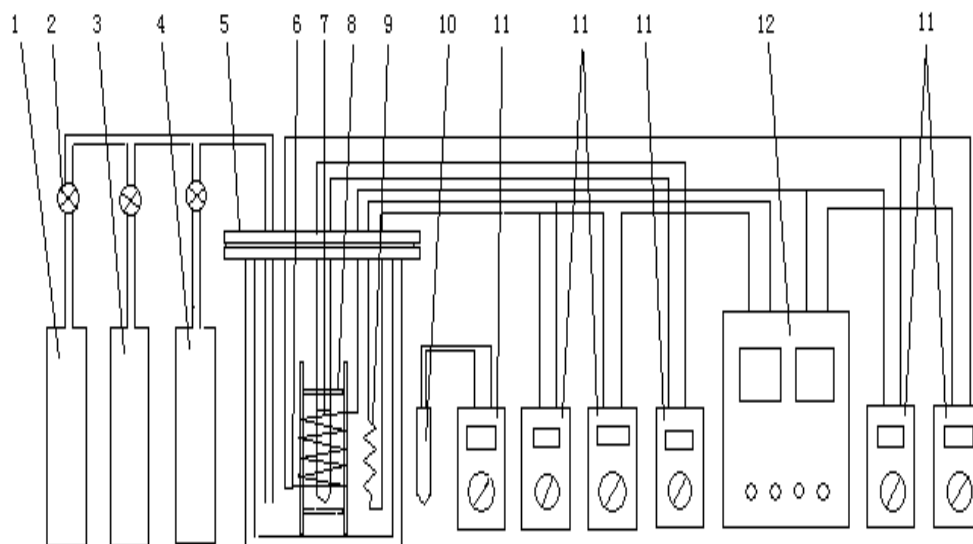


Figure 1. Schematic of H/Pd gas-loading system: 1-hydrogen gas bottle 2-adjust valve 3-mechanical pump 4-vacuum gauge 5-stainless steel reaction chamber 6-Pd wire 7-Pt thermometer for system temperature 8-quarz frame 9-tungsten filament 10-Pt thermometer for room temperature 11-4 1/2 digital multi-meter 12-dual DC power supply

2.2 Calibration

In order to determine whether anomalous effects do exist in a system, its “normal” behavior should be carefully settled in advance. So the calibration for the system of Figure 1 was carried out in vacuum first. The system was pumped to 3~5Pa, then, the Pd wire was heated by an electrical current. When the temperature of the system reached a steady-state, a pair of data (temperature and the power input) were recorded. The heating power was enhanced step by step to make the calibration curve. The same sequence was done when heating of tungsten filament replaced heating of Pd wire. The results of two calibrations are listed in Table 1. The system constants (k) were obtained by the ratio of the temperature difference (ΔT) to the corresponding variation of the input power (ΔP), i.e.

$$k = \Delta T / \Delta P \text{ (}^\circ\text{C/W)}.$$

From table1 we can see that the system behavior has a general concordance whether Pd or W wire was heated. As the system temperature increased step by step, the constant got smaller and smaller gradually. This implied that the heat lost rate of the system would be larger in higher temperature range than that in the lower temperature.

Table 1. System constant k ($^\circ\text{C/W}$) calibration results

Heater	Temperature range ($^\circ\text{C}$)			
	20~40	40~60	60~80	80~120
Pd	12.4		8.9	6.3
W	12.8	9.4	8.8	

3. EXPERIMENTAL PROCESS AND RESULTS

3.1 Heating tungsten wire for gas-loading

As described in previous investigation^[4], an incandescent tungsten filament could help hydrogen to be loaded into Pd lattice. So we evacuated the chamber at room temperature until the system pressure was less than 4 Pa. At this time, the resistance of Pd wire was of 23.20 Ohms, which was taken as R_0 . Then the hydrogen gas was introduced into the reaction chamber. The pressure read by vacuum gauge was $p=1 \times 10^5$ Pa. Then we evacuated the chamber again, but kept the system pressure at 2×10^2 Pa by co-adjusting the valves of H_2 bottle and the pumping speed. At the same time we carefully controlled the electrical power to tungsten filament and heated the tungsten filament to about 2400K. After 20 minutes the power on tungsten filament was turned off. Then let the H_2 be introduced into the chamber again ($P(\text{H}_2) \approx 1$ atm.) for 12 hours. The resistance ratio of Pd wire reached $R/R_0=1.07$ which was corresponding to a loading ratio of $x = [\text{H}]/[\text{Pd}] \sim 0.05$. This value is not too high.

3.2 “Excess heat”

When we turned on the pump again to evacuate the chamber, the unexpected phenomenon appeared: the system pressure was down to 0.01 Pa, which is the best vacuum an ordinary mechanical pump can achieve. When the pump was turn off the pressure was unbelievably unchanged. Supplied an electrical power to Pd wire ($I_{\text{pd}}=4\text{mA}$, $V_{\text{pd}}=0.1\text{V}$) for 12 hours the temperature of Pd rose by about 42°C (Figure 2). Even curiously the pressure was keeping on at 0.01 Pa for 12 hours. The “excess heat” can be easily calculated by the following equation:

$$\text{ExcessHeat} = \int_{t_1}^{t_2} \left(\frac{\Delta T}{k} - I_{\text{pd}} V_{\text{pd}} \right) dt$$

Within 12 hours the “excess heat” is about 28 kilo-Joules, which is corresponding to 180 eV/atom Pd, and the maximum power in this case is 3.2 W, which corresponds to a maximum power density of 230 W/cm³ Pd (Figure 2).

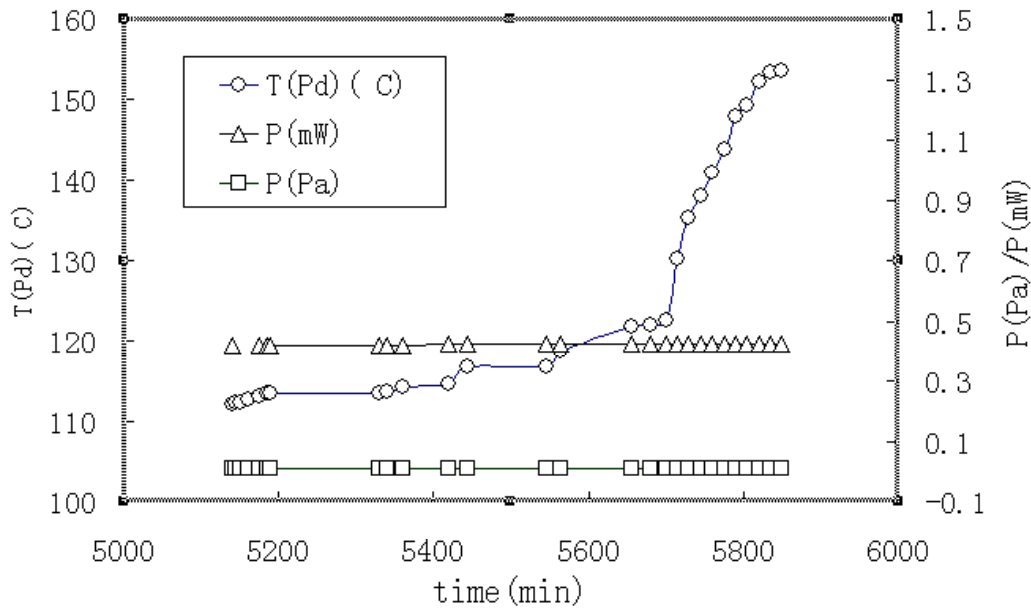


Figure 2. The variation of the temperature of Pd wire(T_{pd} ,circle), the system pressure(P -Pa,square) and the electrical power on Pd wire(P -mW,triangle) during the “excess heat” period

3.3 “Heat after death”

Raise the power supplied on Pd wire to 450 mW for 3 hours and then turn off the power. The total energy inputted by the power supplier was less than 5 kilo-Joules and the temperature of Pd was increased about 5°C at first and then decreased gradually to the original when the power was off. About 2.5 hours after power was off, the temperature of Pd wire was abruptly raise to 760°C or so and kept at that

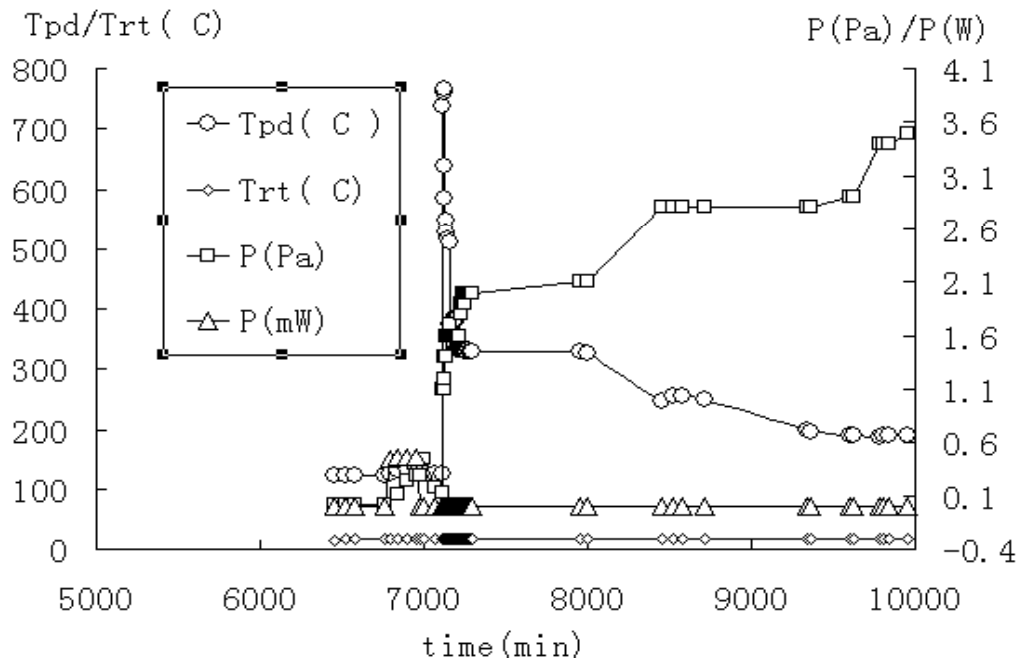


Figure 3. The variation of the temperature of Pd wire (T_{pd} , big circle), the system pressure (P (Pa), square), the electrical power on Pd wire (P (W),triangle) and the room temperature (T_{rt} , small circle) during the “heat after death” period

temperature for about one minute and then decrease rapidly to 373.6°C within about 40 minutes. After that the Pd temperature was slightly rebounded ~10°C in about 20 minutes. Then the temperature of Pd wire decreased rapidly again. When the temperature was down to 334°C, the rate of Pd temperature decreasing was nearly to zero. The temperature of Pd wire was kept on about 330°C for 12 hours. Then the temperature started decreasing slowly at rate of about 8°C/h for 10 hours. After a platform of 255°C occurred for 3 hours, the Pd temperature decreased again at about 6°C/h for another 10 hours. In the last 10 hours of experiment, the decreasing rate became less and less and finally the experiment was stopped intentionally at 190°C. The experiment had lasted for 43 hours since “heat after death” started. The total energy released is about 2 million Joules that corresponds to 13keV/atom Pd. The maximum excess heat power is about 49W and the corresponding power density is 3.6kW/cm³ Pd. Figure 3 shows the variations of Pd temperature, system pressure, and electrical power supply in this unusual period.

3.4 Variation of Li-6 Li-7 concentration in Pd and the anomaly in abundance ratio

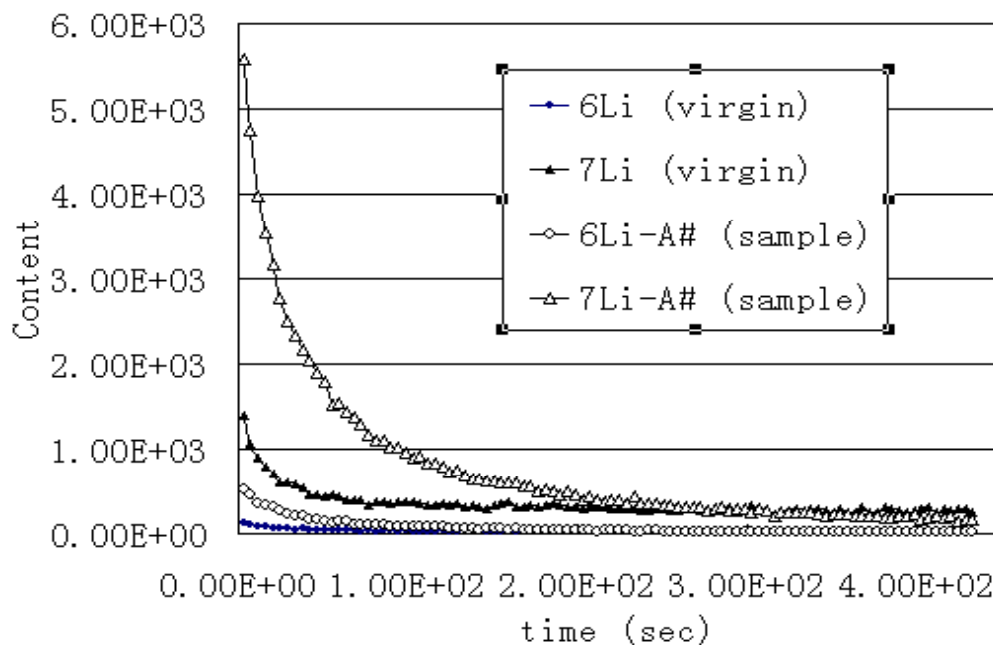


Figure 4. SIMS analysis of Li-6 and Li-7 isotopes on Pd wire surface(the solid circle and triangle show the data for the virgin Pd wire; the open circle and triangle show the data for the sample with anomalous heat.)

In order to examine the variation of isotope concentrations of Li-6 and Li-7, and their abundance ratio before and after the unusual experiment, the sample of the Pd wires used in this experiment and a virgin one were tested by a SIMS (ims-6f), which was made by CAMECA Company of France. The results are shown as in Figures 4 and 5. The figures show that the concentrations of Li-6 and Li-7 have changed a lot before and after the abovementioned process. The total amounts of Li-6 and Li-7 increased by a factor of 4 on the Pd surface in comparison with those of the virgin sample. The time axis in figure 4 and 5 is a measurement of the depth from the surface of Pd, because the ion bombardment just removes the surface layer along with the time elapse (the rate is about several Angstroms per second). Both of Li-6 and Li-7 were getting less and less when the distance from Pd surface becomes larger. The isotope concentration for Li-6 and Li-7 approaches that of virgin sample when the analysis was carried out in the deep of Pd wire (Fig.4). The abundance ratio of Li-7 and Li-6 varies not as much as the concentration itself. (Fig. 5), but it is always higher in the experimental sample than that in the virgin one on the very surface of palladium.(The abundance ratio of Li-7 and Li-6 for the natural lithium is supposed to be 92.41:7.59=12.18)

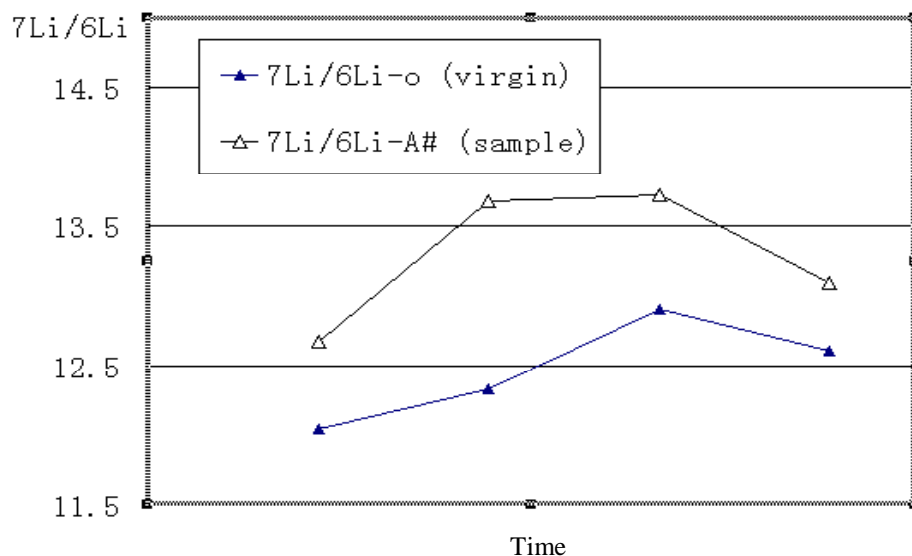


Figure 5. Variation of the abundance ratio of Li-7/Li-6 in Pd. (the solid triangles show the data for the virgin Pd wire; the open triangles show the data for the sample which produced anomalous heat.)

4. DISCUSSIONS AND SUGGESTION

4.1 Anomaly and loading ratio

High loading ratio is an important factor for the anomalous effect. But in our opinion, different ratio may be corresponding to different condition under which the anomaly could appear. Although the loading ratio in this experiment is only about 0.05, the anomaly was still observed. No anomaly had been observed in the later experiments for repetition, in which the ratios were all exceeded over 0.1 (0.7 at most). So it may suggest that high loading ratio might not be a necessary condition for the anomaly. If we choose some suitable outside conditions which would just match with the loading ratio, the anomaly might happen spontaneously. If the ratio changed, the corresponding conditions that induce the anomalous should have been readjusted.

4.2 Anomaly and system pressure

Remarkable change of system pressure had been observed before and after the “excess heat” or “heat after death” period. The pressure dropped suddenly from several Pascals to 0.01 Pa (maybe lower than 0.01 Pa because 0.01Pa is just the lowest number in that vacuum-meter). When a high power density of excess heat appeared, the pressure increased with the system temperature at exactly the same time. So we might guess that the system was in a special status -----“Super-absorption” before the anomaly began. The status is characterized by a kind of strong absorption and causes the drop of system pressure obviously.

4.3 Anomaly and tungsten filament

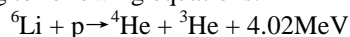
The anomaly in our experiment was unexpectedly observed. So it is still unknown that under what condition(s) the system entered the super-absorption status. But we can find by analyzing the experiment process and recordings that the incandescent tungsten filament might play a key role to induce the anomaly, because the absorption was beginning at the same time the applied power on tungsten is turning off. Although it is not clear that how the tungsten affected the absorption of H₂ by Pd and then to form a positive feedback until “super-absorption” appear, the tungsten surface in very high temperature is very favorable for H₂ to be dissociated into two atoms. Because the newly generated atomic H has a very high chemical activity, some anomalous effects could be induced during the interactions of H atoms with Pd or other atoms.

4.4 Anomaly and non-equilibrium

Taking the Pd temperature as the system temperature is only of a reference sense in our experiment. In fact the temperatures are not uniform inside the system. There is temperature gradient both in radial and in axial directions, that is, the temperature along the Pd wire is not uniform either. In addition there are also non-uniforms of electric and magnetic fields between the Pd wire and the tungsten filament when electrical power was supplied on them. Maybe these non-uniformed gradients and fields triggered the system into the anomaly. So the studies of how temperature gradient and electric/magnetic fields affect a system to enter the anomaly should be a possible direction in future research of cold fusion.

4.5 Anomaly and Li-6 /Li-7 in palladium

As a trace of impurity lithium has less concentration than 0.01%. Supposed that all of impurities in Pd wire were only lithium and all these lithium atoms would be interacted with protons completely according to following equations:



the energy calculated is just in the same order of the heat we recorded. But if the most lithium atoms became ${}^3\text{He}$ or ${}^4\text{He}$, the SIMS results could have a big contradictory to the equations. So we here suggest that the protons might have interacted with some other kinds of atoms but lithium (maybe Pd or other impurities or themselves each other) to form a fresh lithium atom, which might have a different abundance ratio from that of the natural lithium. (The standard value of abundance ratio for terrestrial lithium in handbook is 12.18). Besides the changes of Li-6/ Li-7 concentrations and their abundance ratio become smaller and smaller as the distance from Pd surface become larger. Thus it might imply that the process of forming a newly element is from the surface of Pd to the inside by diffusion of either protons or lithium newly formed.

5. CONCLUSIONS

Five points may be learned from this unusual event, which are:

- 5.1 Anomalous exothermic effect can be seen in H/Pd system as well as in D/Pd system.
- 5.2 High loading ratio of H in Pd may not be the necessary condition. Different loading ratio of H in Pd may be accorded with different condition under which the anomalous exothermic effect could take place.
- 5.3 “Super-absorption” may be one of the characteristics when the H/Pd system enters into the anomalous status.
- 5.4 An incandescent tungsten filament may play a key role in the anomalous process.
- 5.5 The heat that appear in H/Pd system may come from a nuclear origin

ACKNOWLEDGEMENTS

This work is supported by the State Commission of Science and Technology, Natural Science Foundation of China (No. 19645005 & 59995550-3), and Tsinghua University “985” Science Research Fund and Fundamental Research Fund.

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