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Research Article

Temperature Dependence of Excess Power in Both Electrolysis and Gas-loading Experiments

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Abstract

The earlier data from a "Heat after Death" electrolysis experiment and from a Tsinghua University gas-loading experiment are reviewed to show that temperature dependence of excess heat in both electrolysis and gas-loading experiments supports the straight-line behaviour in the semi-logarithmic plot discovered by Storms. Additional gas-loading data in seven Pd-tubes show that excess heat is correlated to a deuterium flux as a result of the diffusion process which is implied in this temperature dependence of excess heat.

Keywords: Deuterium flux, Gas-loading experiment, NAE, NAZ, Pumping effect, Resonant surface capture model, Temperature dependence of excess heat

1. Introduction

We presented two papers about the temperature dependence of excess heat at ICCF-21: a theoretical study and an experimental study. The theoretical study showed that this temperature dependence of excess heat [1] might reveal the resonant mechanism of excess heat: *resonant surface capture reaction* [2]. In this paper we further show that this temperature dependence of excess heat is confirmed not only by earlier Fleischmann–Pons' electrolysis experiments (the famous "heat after death" experiment (ICCF-3) [3]), but also by the earlier gas-loading experiments at Tsinghua University in three aspects: (i) "pumping effect" in a long-thin palladium wire (1999 Asti Meeting and ICCF-9) [4], (ii) the comparison of cooling curves between Pd/D and Pd/H systems (ICCF-6) [5], and (iii) the temperature cycling effect on a long-thin palladium wire (ICCF-7) [6]). Moreover, additional experiments at Tsinghua University (ICCF-

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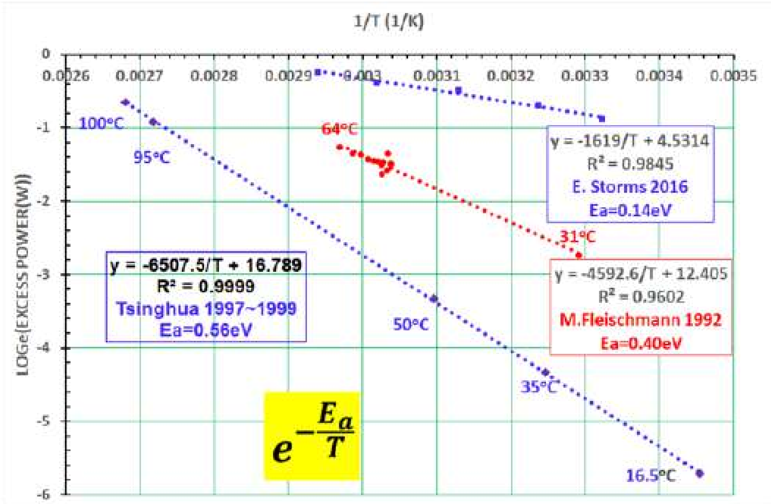

Figure 1. Three straight lines in semi-logarithmic scale plot.

18) [7] have directly shown that this excess heat effect in gas-loading experiments is related to the deuterium flux diffusion through the palladium thin wall of the Pd-tubes.

2. The Temperature Dependence of Excess Heat before Boiling Period of Electrolyte

In 1992 (ICCF-3), Fleischmann and Pons first presented their most compelling "heat after death" experiments. After a period of incubation at low current density, the electrolytic current was raised to trigger a positive feed-back effect in temperature. In their plot the excess heat power was given along with the temperature of the electrolyte [3]. Twelve pairs of data points are available directly from their plot, and are plotted in Fig. 1 (*red circles*). It is interesting to note that these data points are on a straight line again in the semi-logarithmic plot with $(1/T)$ as abscissa as first discovered by Storms 2016 [1]. The data point before triggering has been very important to confirm this straightline behavior, because the temperature of electrolyte after triggering showed somehow a little scattering This implies that although the excess heat power level was low before triggering it was still a real effect. It might be a hint that the so-called threshold effect for current density on the surface of Pd cathode might not be real. It was just the onset point of positive feed-back due to the temperature dependence of excess heat power

3. Confirmation in Gas-loading Experiments

There are three sets of data points from gas-loading experiments with the same long-thin palladium wire in D_2 gas, and a comparable data set in H gas from Tsinghua University in 1997–1999.

1. *"Pumping effect" at 100, 95, and 50°C*

A long-thin palladium wire ($\sim$250 cm$\times\phi\,34$ mm) was wound on a Quartz frame, and was sealed in a stainless vacuum Dewar filled with deuterium gas (Fig. 2). A tungsten wire ($\phi\,0.1$ mm) was used to dissociate the deuterium molecule, D_2, into atoms in order to increase the chemical potential of D atoms which was believed to be essential for loading the D atoms into palladium wire [8]. A Pt-100 thermistor monitored the temperature at the middle of the

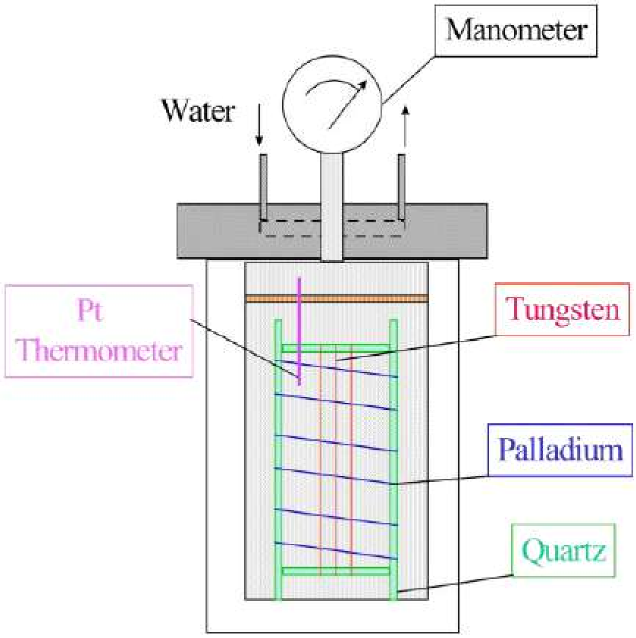

Figure 2. The early gas-loading apparatus in 1997–1999 at Tsinghua University.

palladium wire ($T_{\rm Pd}$) An intelligent DC power supply was used to heat the palladium wire to specified temperatures (100, 95, and 50°C) When we pumped out the deuterium gas after loading, the necessary power to keep the specified temperature dropped. This power reduction was a good measurement of any excess heat effect in the Pd–D system.

Figure 3 shows the necessary power (red line in upper plot) and the specified temperature of Pd wire (purple line in the lower plot). At first the temperature was kept as $95 \pm 0.2°$C for more than half a day with 2.25 ± 0.05 W DC power. When the pumping started, the power needed to maintain this temperature dropped instantly. We might expect to see the power jump up, because the degassing from Pd wire is supposed to be an endothermic process. We did see such a power jump as a short spike in the red line, but it was quickly followed by an exothermic effect−the DC power decreased until it reached a steady value. The DC power reduction is about 2.25–1.85=0.4 W for 95°C. Figure 3 also shows that this pumping effect is reproducible after one day (near 220 000 s). At 250 000 s, we changed the set temperature from $95°$C to $100°$C, and the necessary DC power needed to maintain this higher temperature increased to 1.96 W while pumping was continuing. The sudden stop of "pumping" near 300 000 s increased the necessary power to 2.48 W. The power reduction of "pumping effect" at $100°$C is about 0.52 W.

In order to exclude the possible effect of the reduction in the heat transfer coefficient, we note that Fig. 4 showed the gas pressure in the Dewar (purple line in the lower plot) while the $T_{\rm Pd}$ is fixed at 50°C for 1 day The "pumping" was done in two phases: weak pumping and strong pumping. At 40 000 s we started weak pumping with a controllable valve. In the period of 40 000–48 000 s, the gas pressure was increasing slowly; however, the necessary heating power (red line in Fig. 4 upper plot) was decreasing. Evidently, this power reduction was due to some exothermic effect inside the Pd–D system, and not by the reduction in the heat transfer coefficient. When the strong pumping started near 48 000 s, the necessary heating power dropped quickly at first; this was then followed by a slow reduction. However, the gas pressure showed totally different time-behavior: a step-wise reduction at 48 000–63 000 s. When we stopped

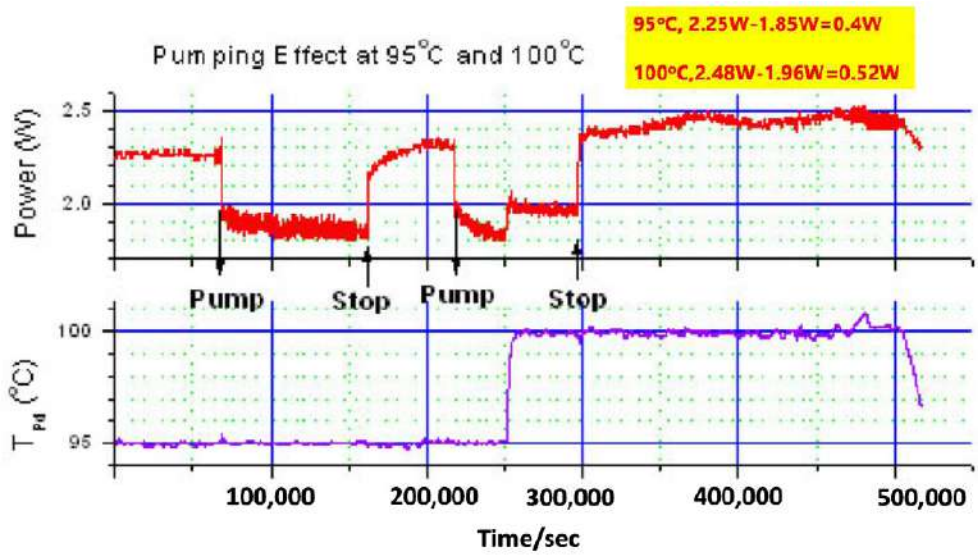

Figure 3. "Pumping effect" in gas-loading experiments near 95 and 100°C.

strong pumping near 63 000 s, the time-behavior of power and gas pressure are so different there is no way to attribute this power variation to the variation in gas heat conductivity. This exothermic effect at 50°C is about 0.24–0.204 = 0.036 W.

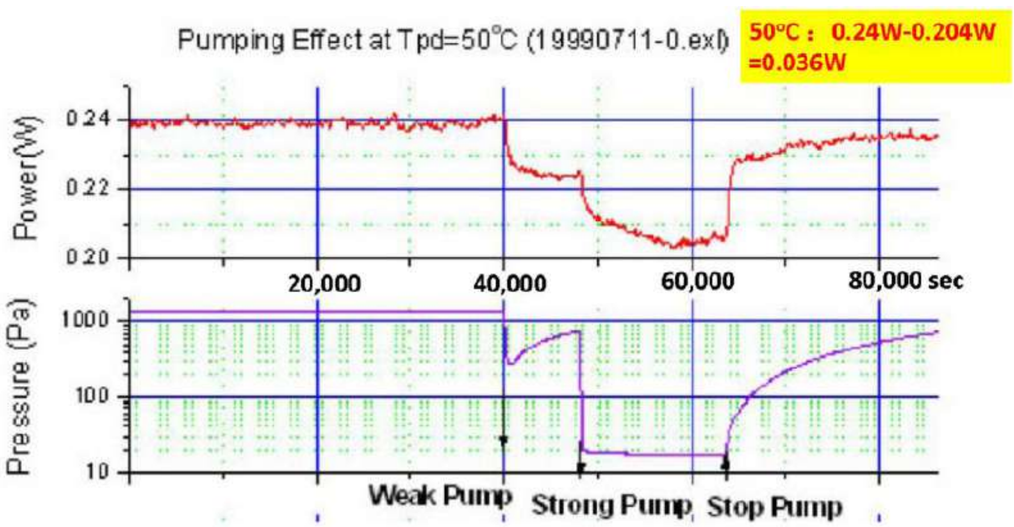

Figure 4. "Pumping effect" in gas-loading experiment at 50°C.

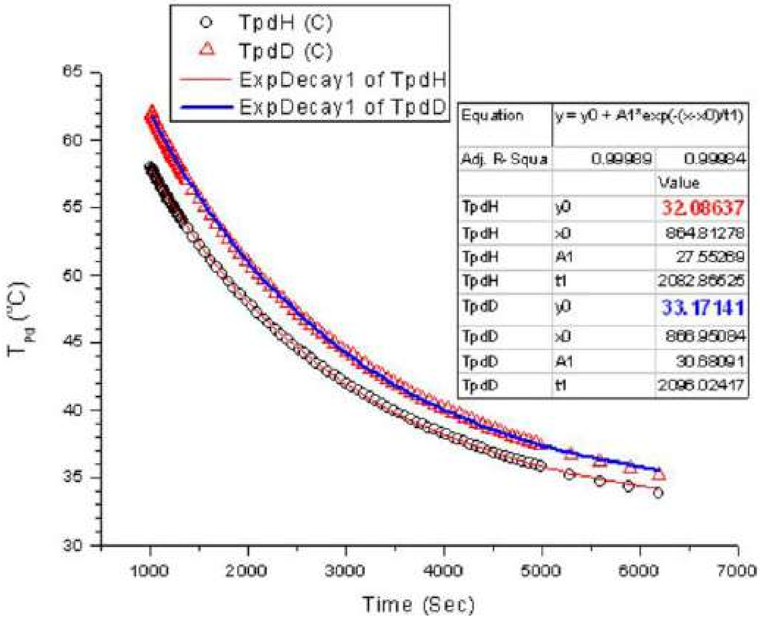

Figure 5. Comparison of cooling curves of D/Pd and H/Pd systems.

2. *Comparison of cooling curves between Pd/D and Pd/H systems near 35°C*

Although we do not know exactly at which temperature the resonant exothermic reaction occurs, we can still observe this exothermic effect during the cooling process. Particularly, we can compare the difference between the Pd/D and Pd/H system, and confirm this exothermic effect. We built two gas loading Dewars as shown in Fig. 2. The twin systems are almost the same (see Table 1) except they were filled with different gases (deuterium and hydrogen). After they were heated to similar temperatures, the heating power was shut off, and the twin systems approached different constant terminal temperatures: the temperature in the D/Pd bottle was always a little higher than that in H/Pd bottle. In Fig. 5, the red triangles show the temperature in the D/Pd bottle, and the black circles show the temperature in H/Pd bottle. They are fitted to the cooling curve very well with:

Table 1. Parameters of twin systems for gas-loading.

	D/Pd	H/Pd
Weight of Pd wire (g)	2.846	2.844
Resistance of tungsten wire (Ω)	4.1	4.3
Vacuum pressure before filling (Pa)	10^{-3}	10^{-3}
Filling pressure(mmHg)	600(D)	600(H)

$$T_{\rm Pd} = T_{\rm Pd0} + A_1 {\rm e}^{-\frac{(t-t_{\rm o})}{t_1}}. \quad (1)$$

Here, t is the time, and $t_{\rm o}$ is the starting point of their cooling curves. It is evident that when $t \to \infty$, $T_{\rm Pd} \to T_{\rm Pd0}$.

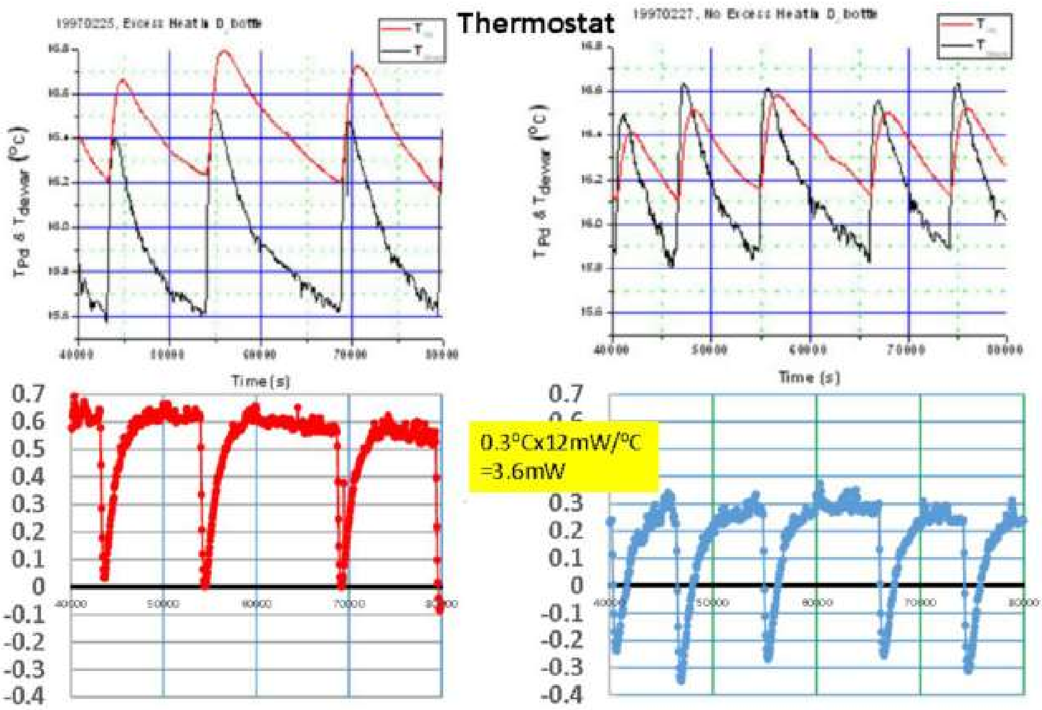

Figure 6. Temperature cycling in a thermostat for D/Pd system in different days.

However, the D/Pd and H/Pd systems approach different levels: 33.17 and 32.08°C, respectively, although they were cooling in the same room temperature. The D/Pd system is 1.09°C warmer than the H/Pd system which implies an exothermic reaction is occurring inside the D/Pd bottle even when the temperature approaches room temperature. We determined that the calorimeter constant of the H/Pd bottle was 0.012 W/°C in this temperature range; hence, the excess power was 12 mW near 35°C.

3. *The temperature cycling effect in D/Pd system around 16.5°C*

In winter, when the room temperature in our laboratory is much lower, we put the D/Pd system in a thermostated water bath. The thermostat kept the temperature of the water bath oscillating around $T_{\text{water}} = 16.0$°C (black line in Fig. 6 upper-left plot). It is evident that the temperature of the Pd wire, T_{Pd} (red line), is always a little higher than the T_{water}. The lower-left plot in Fig. 6 shows that the difference ($T_{\text{Pd}} - T_{\text{water}}$) is almost always greater than zero. It shows again the exothermic effect inside the D/Pd bottle. This difference is about 0.6°C most of the time. One might worry about the accuracy of these Pt-100 thermistors. The right plots of Fig. 6 shows the accuracy of the Pt-100 thermistors. On the next day, the colder weather triggered the thermostat more often; hence, the bath temperature was oscillating faster. It oscillated five times in 40 000 s, which was two times more than that of the previous day. According to the resonant theory of energy bands (ICCF-6) [9], if the cooling speed was too fast, the D/Pd system might not have enough chance to produce resonant reactions before it cooled down through and below the resonance energy band. Therefore, on the second day, the temperature of D/Pd system was no longer kept higher than the water bath temperature (lower right-hand side plot of Fig. 6). When $T_{\text{water}} > T_{\text{Pd}}$, the D/Pd system was heated by the water bath, the T_{Pd} was increasing until $T_{\rm water} = T_{\rm Pd}$ When $T_{\rm water} < T_{\rm Pd}$, the D/Pd system was cooled by water bath, and $T_{\rm Pd}$ was decreasing until $T_{\rm water} = T_{\rm Pd}$. As a result, while the $T_{\rm water}$ was oscillating, $T_{\rm water}$ always intercepted the $T_{\rm Pd}$ at maximum $T_{\rm Pd}$ or minimum $T_{\rm Pd}$. The upper-right-hand side plot of Fig. 6 clearly shows that our Pt-100 thermistors were working properly. Both $T_{\rm Pd}$ and $T_{\rm water}$ are accurate enough to show this tiny exothermic effect. The lower-right-hand side plot of Fig. 6 shows that the difference $(T_{\rm Pd}–T_{\rm water}) = 0.3°$C is enough to drive the D/Pd system to follow the oscillating $T_{\rm water.}$ Having compared left and right-hand side plots of Fig. 6, we might conclude that there was an exothermic effect on the first day to keep the D/Pd system 0.3°C higher than $T_{\rm water}$. Using the same calorimeter constant, 12 mW/°C, we have 3.6 mW excess heat power near 16.5°C It was a big surprise when we put these five pairs of points on the semi-logarithmic plot: they were on a straight line again (purple diamond points in Fig. 1). These experiments at low temperatures in early 1997 were designed to show an exothermic effect qualitatively only. We did not expect such a good quantitative result. Now the question is why the slope of this straight line is different from that of Storms' and Fleischmann–Pons' lines. In our theory paper [2], we showed that the slope of Fleischmann–Pons' straight line is probably related to the activation energy of deuteron in lithium deuteride. The slope of the straight line here for the gas-loading experiment might also be related to the activation energy of deuterons in tungsten deuteride. Although we have some evidence for this assumption, we would prefer to discuss this possibility when more experimental data are available. We will first show that this excess heat is related to a diffusion flux of deuterium though palladium.

4. Excess Heat is Correlated with Deuterium Flux but Not "Pumping"

To further exclude the effect of heat conductivity induced by "pumping", and show the correlation between excess heat and deuteron flux, the gas-loading experiments were continued at Tsinghua University using a Pd-tube instead of Pd-wire after 1999. The first experiment used a Seebeck calorimeter (C-80D) which is supposed to be independent of heat conductivity with the accuracy of 1 μW. The "pumping effect" has been confirmed again in a short thin wall

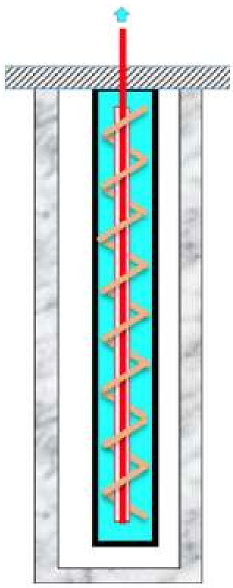

Figure 7. A new gas-loading system with a bunch of seven long-thin Pd tubes.

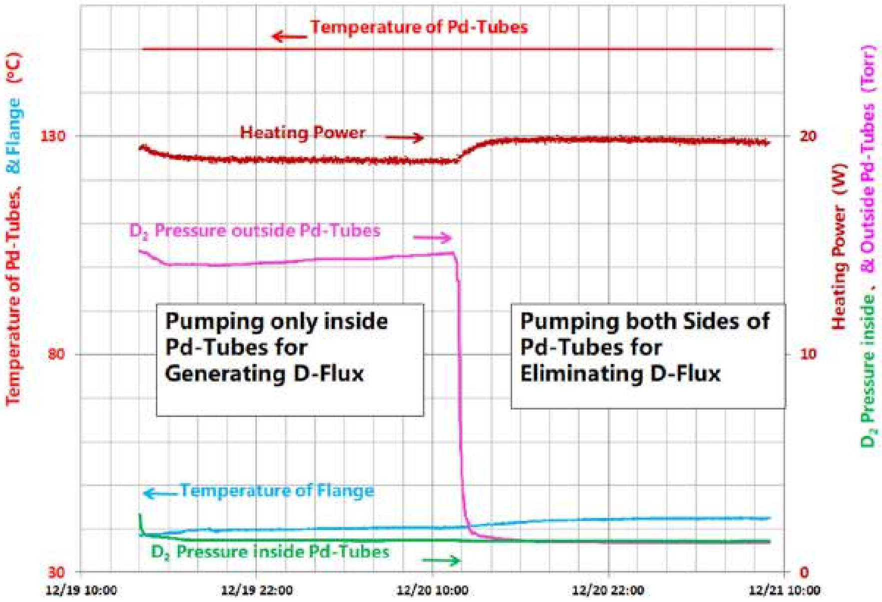

Figure 8. Excess heat is correlated with the deuterium flux but not "pumping effect".

Pd-tube ($26\,\text{mm} \times 0.1\,\text{mm} \times \phi\,4\,\text{mm}$) [10]. A peak of excess power was found near 140–150°C which was correlated with a peak of deuterium flux through the thin wall of the Pd-tube. In order to further scale-up this deuterium flux effect, a new Pd-D system was built using seven long-thin Pd-tubes ($40\,\text{cm} \times 80\,\mu\text{m}{\times}\phi\,3\,\text{mm}$) (Fig. 7) A bunch of seven long-thin Pd-tubes were sealed at their lower ends and the upper ends were connected to a mechanical pump to pump out the gas inside the Pd-tubes. The deuterium gas was filled outside the Pd-tubes in a stainless steel cylinder. An electrical heater was wound around the Pd-tubes, and the whole cylinder was put into a glass Dewar for heat insulation. The major heat conducting path was through the stainless steel flange on the top of the Dewar. An intelligent power supply was applied to keep the Pd-tubes at a specified temperature (150°C). When we pumped inside of Pd-tubes only a deuterium flux was generated from outside into the inside of the Pd-tubes. If we pumped both sides of Pd-tubes the "pumping effect" was supposed to be stronger, but the deuterium flux was reduced greatly. We might compare these two cases to judge if the heat effect was correlated with deuterium flux or correlated with "pumping". Figure 8 shows the experimental result over two days. The red line on the top shows the temperature was kept at 150°C by an intelligent power supply very closely. The green line at the bottom shows that the gas pressure inside the Pd-tubes was kept at less than 2 torr by a mechanical pump. The central pink line shows that the D_2 gas pressure outside the Pd-tubes was about 14 torr at first, and was pumped down at 12:00 AM of the second day. The necessary heating power of the intelligent power supply is shown by the brown thick line above the pink line. A very clear power jump was correlated with pumping both sides of Pd-tubes. The necessary heating power jumped from 19 to 20 W to keep the temperature at 150°C when the "pumping effect" was stronger, and heat conductivity was supposed to be smaller. Therefore, we have to attribute the DC power reduction on the first day to the deuterium flux generated by pumping inside of Pd-tubes only. Indeed the heat effect correlated with deuterium flux is more than 1 W if we consider the reduction of heat conductivity.

5. Conclusion.

Three conclusive remarks might be tentatively draw from our two presentations at ICCF-21:

(1) A straight line in semi-logarithmic scale plot is confirmed by three sets of data from gas-loading and from electrolysis experiments in different laboratories which were published well before the publication of Storms' discovery

(2) A two-step model is implied in this straightline behavior. An elastic diffusive process creates a mother state for the next inelastic nuclear transition to daughter state. Indeed this might be just the model we need for understanding the nuclear active environment (NAE) or nuclear active zone (NAZ).

(3) The additives in anomalous heat experiments might play a more important role than we thought previously. The lithium (LiOD) in electrolysis experiments was only thought to be an additive to increase conductivity of electrolyte. The lithium ($LiAlH_4$) in Ni–H system was considered only a catalyst in reactions. The tungsten wire was considered only a tool to dissociate the molecule D_2. Now we understand that: the slope of Fleischmann–Pons' straight line is very close to the activation energy of deuterium diffusion coefficient in lithium–deuteride; the slope of Storms' straight line is very close to the activation energy of deuterium diffusion coefficient in palladium–deuteride; and the slope of Tsinghua University's straight line might be close to the barrier energy of the deuterium diffusion coefficient in tungsten deuteride. Based our resonant surface capture model we might ask: What was the major fuel in these anomalous heat effects?

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