"Excess Heat" Measurement in Gas-loading D/Pd System

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Abstract
A gas-loading D/Pd system has been designed to measure the "excess heat". The preliminary result has shown that the calorimetric feature of the D/Pd system is distinct from that of its twin H/Pd system. The difference between these twin systems can be attributed to the "excess heat" of the order of watts per cubic centimeter of palladium.

1. Introduction

The "heat after death" phenomenon [1] has revealed a fact that the electrolysis is not necessary for "excess heat" phenomena. Provided that enough deuterons are absorbed in the palladium crystal lattice, we may expect to see the "excess heat" in a gas-loading system as well. We are particularly interested in the gas-loading system, because we have been engaged in the gas-loading system for several years[2-7]. Early in 1993, Manduchi et al. [8] showed that the palladium samples might be soldered together during the gas-loading process with the deuterium gas. It was explained as an evidence of "excess heat" in this gas loading system. Manduchi's experiments worked out with both the Russian palladium and the British palladium samples. It seems that the only important pre-treatment in his experiment is the annealing at the high temperature (900 °C) in vacuum. This is consistent with the early basic research on the gas loading experiments. Oats and Flanagan [9] did the gas loading experiments early in 1971. They showed that the only pre-treatment was just the flame heating before use. The loading ratio was as high as 0.94 in their experiments as a matter of routine. The even surprising point was that the gas-loading was done at low hydrogen pressure (1.5 Torr, 25 °C). The key element was a heated tungsten wire which dissociated the hydrogen molecules into hydrogen atoms. These features of operating under the low pressure and heating by a tungsten wire facilitate the combination of a calorimetric system with a gas-loading system.

2. Experimental Apparatus.

The low pressure feature makes thin wall stainless steel dewar system applicable for a closed gas-loading system (Fig.1). Palladium wire of ϕ 0.34mm is wound on a quartz frame. A pair of such quartz frames with palladium wire winding are made for D_2-loading and H_2-loading, respectively. They are put into a electrical oven to be heated to 900 °C in vacuum (10^{-3} Pa). Then they are cooled gradually with the oven. Before they are put into the stainless steel dewar, a piece of tungsten filament (ϕ 0.1mm) is mounted at the center of the quartz frame. The resistance of the palladium wire is measured by the four-lead method in order to determine the loading ratio (D/Pd and H/Pd) *in situ*. The leads for palladium wire and for tungsten filament can be used for both heating the dewar vessel and measuring their resistance. A platinum thermometer is put into the gap between the tungsten filament and the palladium wire in order to measure the temperature change during the process of gas-loading and the calorimetric heating. A manometer is monitoring the gas pressure to evaluate the loading ratio, and a diffusion pump is used to pump out the air to 7×10^{-4} Pa.

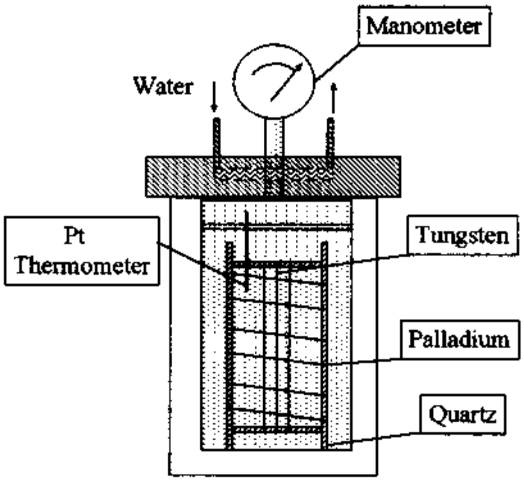

Fig.1 A Combination of a Calorimeter with a Gas-loading System

3. Calorimetric Feature.

This stainless dewar vessel is different from the glass or quartz dewar electrolytic cell in Fleischmann and Pons experiments. In stead of radiation, the heat conduction plays the dominant role in the heat transfer. Fig.2 shows the temperature, θ, in the dewar as a function of time. It can be described by the following equation

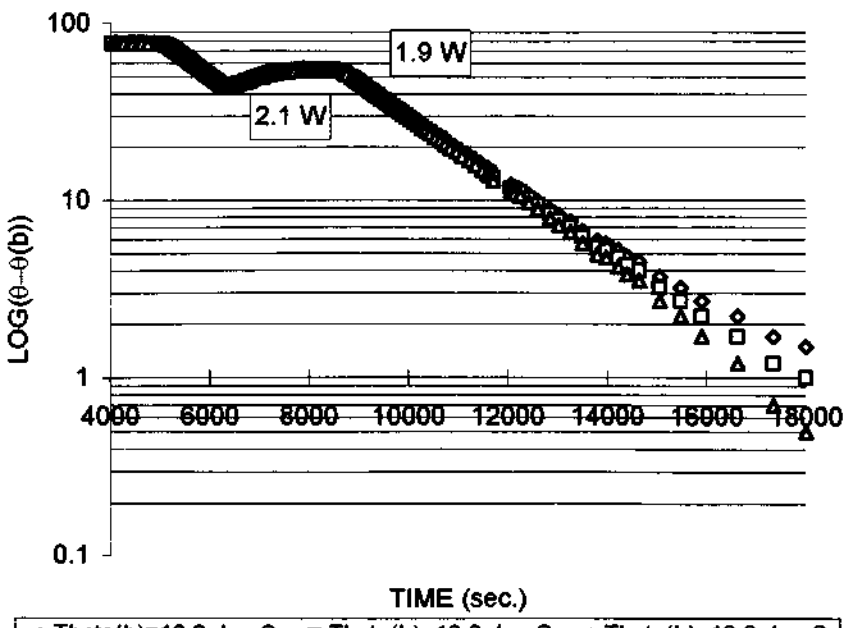

Fig.2 Calorimetric Feature of the Stainless Steel Dewar System

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$$MC_p \frac{d\theta}{dt} = -k(\theta - \theta_b) + S \quad (1)$$

when the heat source, S, is a constant source; the temperature in the platinum thermometer, θ, approaches a constant value θ_f

$$\theta_f = \theta_b + \frac{S}{k} \quad (2)$$

Here, k is the heat conduction coefficient in the Newton's Law of heat transfer. θ_b is the room temperature. The relaxation time of this system is determined by the reduced heat conduction coefficient:

$$\hat{k} = \frac{k}{MC_p} \quad (3)$$

MC_p is the equivalent heat capacity of the calorimetric system. The equation can be written as

$$\frac{d\theta}{dt} = -\hat{k}[\theta - (\theta_b + \frac{S}{k})] \quad (4)$$

When S is a constant

$$\theta = (\theta_b + \frac{S}{k}) + [\theta_0 - (\theta_b + \frac{S}{k})]e^{-\hat{k}(t-t_0)} \quad (5)$$

or

$$\log[\theta - (\theta_b + \frac{S}{k})] = -\hat{k}(t - t_0) + \log[\theta_0 - (\theta_b + \frac{S}{k})] \quad (6)$$

Here, θ_0 is the initial temperature of the dewar system at time t_o. This is the straight line section shown in Fig.2. The slope of the straight line gives the value of the reduced heat conduction coefficient, $\hat{k}$; the constant $(\theta_b + \frac{S}{k})$ may include the information for both the applied heat source, P; and the internal heat sources, Q. The applied heat source, P, is the heating power given by the experimentalist; however, the internal heat sources, Q, may include the "excess heat" which is what we are searching for.

There are two ways to determine ($\theta_b + \frac{S}{k}$):

(1) Heating method: Given a constant heating power (e.g. a constant current through the tungsten wire), the temperature, θ, approaches a constant value, θ_f .

$$\theta_f = \theta_b + \frac{S}{k} \quad (7)$$

(2) Cooling method: When we plot the Fig.2 using the experimental data, we have to have a correct value for $\theta_b + \frac{S}{k}$ in order to make a straight line for cooling part. Fig.2 shows that the least squares fit is able to find the value of $\theta_b + \frac{S}{k}$ with a precision better than 3%. 0.5 °C change in $\theta_b + \frac{S}{k}$ may make the straight line visibly distorted (i.e; bent upward or downward, if the $\theta_b + \frac{S}{k}$ is not a correct number.)

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Fig.2 shows also the sensitivity of our calorimetric system. When a small power of 2.73W is applied onto the calorimetric system through a current in the tungsten wire, the temperature goes up to a constant value of 96.0 °C. Then the current is shut off, and we see a straight line of cooling curve with no heat source (see equation (6)). Later we turn on the current in the tungsten wire again with a power of 2.55W, a clear inflection point appears to stop the straight line, and levels to a new constant value. Particularly, the curve goes up continuously at a power of 2.1W and turns horizontal at a power of 1.9W. Thus, we are confident that this system is able to detect the "excess heat" at the level of 1 W per c.c. palladium while we put 0.234 c.c. palladium wire into this calorimetric system.

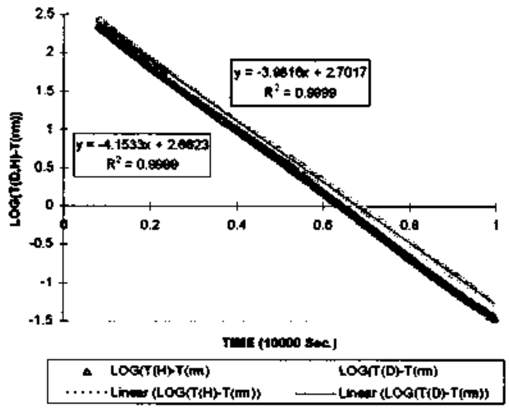

Fig.3(a) Cooling Feature for D/Pd & H/Pd Systems
(43---32 Deg.C for D; 41---31 Deg.C for H)

4. Temperature Difference between Twin Systems

A calibration is necessary to quantify the "excess heat"; however, if we are able to show that there are sharp differences in the calorimetric feature between the H/Pd and D/Pd twin systems; then, we may qualitatively show the evidence of the "excess heat".

To load the hydrogen or deuterium gas into the

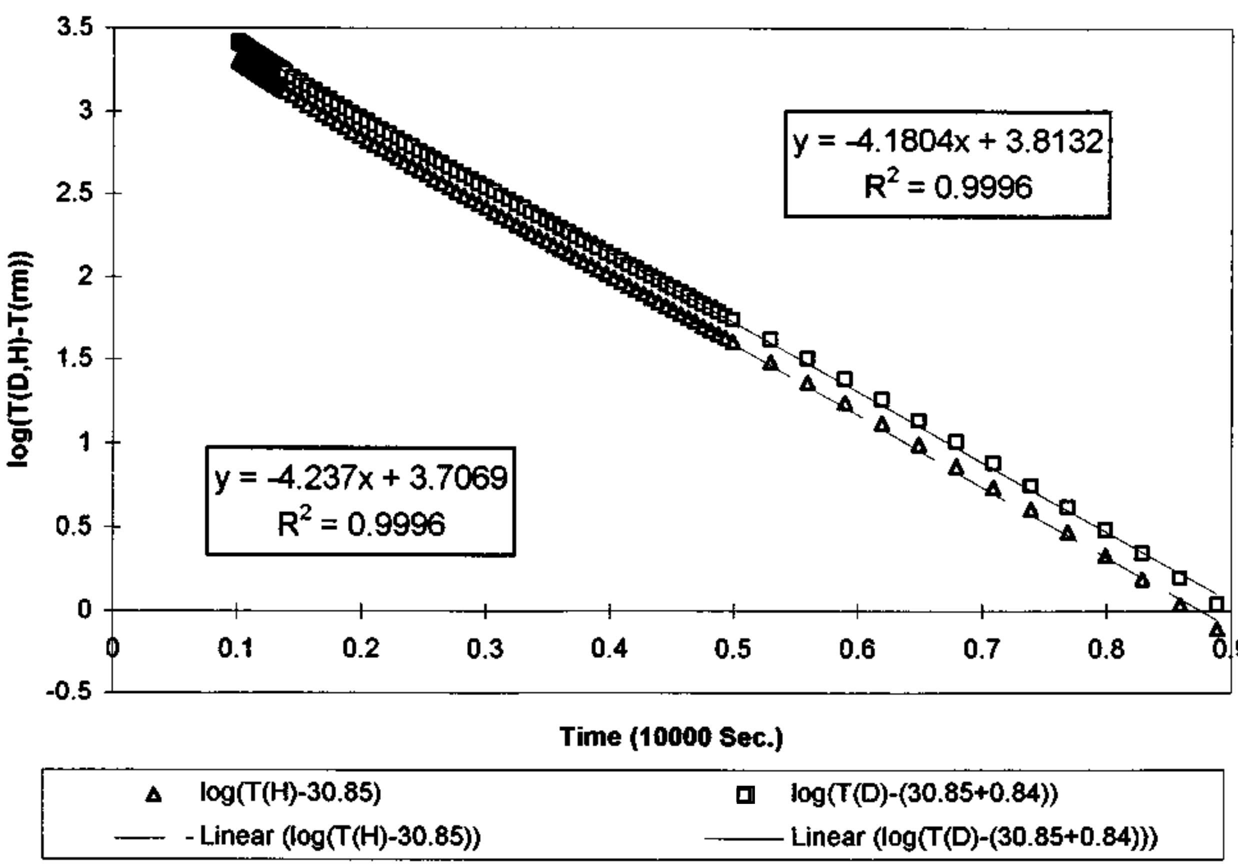

Fig.3(b) Cooling Feature of D/Pd and H/Pd Systems
(62---33 Deg.C for D; 58---32 Deg.C for H)

palladium, electrolytic cell was supposed to be necessary to provide the enough "chemical potential"; otherwise, the high pressure and low temperature was supposed to be necessary. R.F. power, a piece of incandescent tungsten wire (2000 °C), or D.C. discharge were found to be able to provide the hydrogen or deuterium atoms in stead of the electrolytic current. However, we found that even if the tungsten wire was turn-off, as long as the correct pre-treatment of the palladium wire was done, the loading ratio (H/Pd or D/Pd by atom number) might achieve 0.74 at a pressure much less than 1 atm. This is discussed in two associated papers [10] and [11]. Here, we just discuss the calorimetric feature of the gas-loading system.

In order to detect the possible "excess heat" in a D/Pd system, we intentionally built a pair of twin systems. No.1 bottle is for deuterium loading, and No.2 bottle is for hydrogen loading. These two systems have same dewar structure, and similar quartz frames. The palladium wires in both bottles are cut from same batch (ϕ 0.34mm); and have been pre-treated with same procedures. The weight of palladium wires in two bottles are 2.846g. and 2.844g., respectively. The tungsten wires are cut from same batch (ϕ 0.1mm). The resistance of tungsten wire are 4.1Ω (in D bottle), and 4.3Ω (in H bottle) , respectively. After being pumped to 10^{-3}Pa, the No.1 bottle was filled with deuterium gas up to 660 mmHg; and the No.2 bottle was filled with hydrogen gas up to 660 mmHg also. Fig.3 shows the cooling curves for bottles No.1 and No.2. A straight line is drawn to fit the experimental data using the least squares fit. From this straight line, the reduced heat conduction coefficient, $\hat{k}_D$ and $\hat{k}_H$, is calculated. To maximize the correlation coefficient R^2, we may determine the value of $(\theta_b + \frac{S}{k})$ for that straight line. Table 1 gives the corresponding values for the reduced heat conduction coefficients $\hat{k}_D$ and $\hat{k}_H$; the constants $(\theta_b + \frac{S}{k})_D$ and $(\theta_b + \frac{S}{k})_H$; and the correlation coefficients R_D^2 and R_H^2.

Table 1 Calorimetric Feature for Cooling Curve of D/Pd and H/Pd Systems

Temperature Range °C		Reduced Heat Conduction Coefficient $\hat{k}$ sec^{-1}	$\theta_b + \frac{S}{k}$ °C	R^2
41.456→ 31.173	H	$(4.15 \pm 0.02) \times 10^{-4}$	30.94	0.9999
42.785→ 31.803	D	$(3.98 \pm 0.02) \times 10^{-4}$	30.94+0.579	0.9999
58.047→ 31.741	H	$(4.24 \pm 0.04) \times 10^{-4}$	30.85	0.9996
62.161→ 32.728	D	$(4.18 \pm 0.03) \times 10^{-4}$	30.85+0.84	0.9996

It is clearly shown that the reduced heat conduction coefficients are same for both bottles with the precision better than 5%. However, the constants, $\theta_b + \frac{S}{k}$, are different for D/Pd and H/Pd

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systems. The difference between them increases, when the temperature range expands. Since the information of "excess heat" is included in this constant, $\theta_b + \frac{S}{k}$, we investigated further by another method: i.e. we heat the systems using the electrical current in the tungsten wire. Through the constant current at 0, 0.1965A, 0.359A, 0.464A, 0.601A, 0.753A and 0.801A; the D/Pd and H/Pd systems are heated to different temperatures. From these temperatures, we may have another estimate of the constant, $\theta_b + \frac{S}{k}$. Table 2 list the results of these heating experiments. The difference of $\theta_f - \theta_b = \left[\left(\theta_b + \frac{S}{k}\right) - \theta_b\right]$ gives the $\frac{S}{k} \equiv \frac{P+Q}{k}$.

In the experiments, the tungsten wires in the D/Pd and H/Pd are connected in series such that the electrical current, I_w , is same in both tungsten wires. This heating power enhances the temperature, θ , in both bottles, and eventually makes a steady state value, θ_f. We can measure the temperature difference at the steady state between the inside and the outside of the dewar, $\theta_f - \theta_b$, which should be equal to the constant $\frac{S}{k} = \frac{P+Q}{k}$. In this case, P is the heating power in tungsten wire, Q is the possible "excess power", k is the heat conduction coefficient. It is interesting to notice that when the heating power increases, the temperature difference, $\theta_f - \theta_b$, in D/Pd system increases much faster than that in H/Pd system. This could be attributed to the possible "excess power" in the D/Pd system. In order to estimate this "excess power", we need the value for heat

Table 2 Calorimetric Feature for Heating Curve of D/Pd and H/Pd System

I_w	D		H		
	P_w^D	$\theta_f - \theta_b = \left[\frac{P_w + Q_{ex}}{k}\right]_D$	P_w^H	$\theta_f - \theta_b = \left[\frac{P_w + Q_{ex}}{k}\right]_H$	$Q_{ex}^D = \frac{\left[\frac{P_w + Q_{ex}}{k}\right]_D}{\left[\frac{P_w + Q_{ex}}{k}\right]_H} P_w^H - P_w^D$
(A)	(W)	(°C)	(W)	(°C)	(W)
0.197	0.161	2.44	0.173	2.48	0.009
0.359	0.551	8.34	0.569	7.70	0.065
0.464	0.960	14.2	0.977	12.9	0.115
0.601	1.73	24.6	1.71	21.5	0.227
0.753	2.99	39.8	2.93	33.9	0.450
0.801	3.51	41.8	3.45	33.3	0.821

conduction coefficient, k_D. Since the reduced heat conduction coefficients, $\hat{k}_D$ and $\hat{k}_H$ for D/Pd and H/Pd are almost same (see table 1). It is reasonable to assume that the heat conduction coefficients, k_D and k_H, are same also. Based on this assumption, the "excess power" in the D/Pd system is calculated. It may achieve up to 0.821W (see the last column in table 2) while the heating power is about 3.5W. An additional evidence of this "excess power" in D/Pd system is the change of the resistance of the tungsten wire. Initially, the resistance of the tungsten wire in H/Pd system (4.46 Ω) is a little higher than that of D/Pd system(4.15 Ω). When the heating power is increased to ~ 3.5W, the resistance of the tungsten wire in H/Pd system (5.38 Ω) becomes smaller than that of D/Pd system (5.47 Ω). Using the resistance temperature coefficient $\alpha_W = 4.8 \times 10^{-3} K^{-1}$ for tungsten wire, we have the temperature for the tungsten wires in the H/Pd and D/Pd system, 73.5 °C and 96.9 °C, respectively. The temperature in D/Pd system is much higher than that of H/Pd system . This is consistent with the former calculation of the "excess power" inside the D/Pd system.

5 Concluding Remarks:

While a pair of twin calorimetric systems are heated by the same amount of electrical power, the systems approach different temperatures at steady state. The D/Pd system is hotter than the H/Pd system. The temperature difference may achieve the value of 8.5 °C measured by the platinum thermometer. Since the cooling curves have shown the reduced heat conduction coefficients for both systems are same, this temperature difference means that there must be an "excess heat" source in the D/Pd system.

We are using platinum resistance thermometers and the Keithley digital multi-meter to measure the temperature. The precision of the measurement is better than 0.1 °C. Hence, we are confident about the existence of this temperature difference. This is a qualitative proof of the "excess heat" in D/Pd system.

To quantify this excess heat, we need two assumptions: First the heat conduction coefficient is assumed to be same for the twin systems; second, the excess heat in the H/Pd system is assumed to be zero. The first assumption is supported by the equality of $\hat{k}_D = \hat{k}_H$; and the second assumption just makes a conservative estimate on "excess heat". If there is any "excess heat" in the H/Pd system also; then, the "excess heat" in D/Pd system should be greater than this estimate. It is at least at a level of 1W per c.c. palladium. We have observed this excess heat for more than 5 months. (from April, 20 to September 29, in 1996) in a D/Pd system with 2.846g. of palladium. It is about 10^3eV for each palladium atom, which is very difficult to be attributed to any chemical resource. This gas-loading system has the advantage of being operated at a higher temperature which is essential to enhance the heat energy efficiency as an energy source.

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