J. Condensed Matter Nucl. Sci. 4 (2011) 119–131

Research Article

Studies on Anomalous Phenomena of D/Pd Systems using a Gas-loading Process – A Stride Towards Neutrino Detection

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Abstract

A brief review of 20 years of experiments at Tsinghua University confirms anomalous phenomena during gas loading in D/Pd systems. A scale-up of the experiment would make it feasible to test the hypothesis that neutrinos are emitted during these phenomena (the "neutrino conjecture").

Keywords: Deuterium/Palladium system, Gas-loading, Neutrino detection, Pumping effect

PACS: 89.30.Jj ,89.90.+n, 88.20.fn, 88.85.mh

1. Introduction

Condensed matter nuclear science research has been conducted for 20 years at Tsinghua University. It started from both electrolysis and gas-loading experiments, but later we worked on gas-loading experiments mainly because of the following theoretical consideration: If the charged particles react, there must be some charged nuclear products, due to the conservation of electrical charge. Hence, we should try to detect charged particles instead of neutron or gamma radiation, in order to confirm this anomalous phenomenon. The gas loading cell is much more favorable for charged particle detection. The first successful experiment used CR-39 to detect charged particle emission from a palladium foil after thermal cycling at high deuterium pressure [1]. At the same time the first theoretical efforts were initiated to explain the tunneling through the Coulomb barrier, and the puzzle of "excess heat without commensurate radiation" [2] ("thunder without lightening").

After 1995, our gas-loading experiments were switched from low-temperature, high-pressure to high-temperature, low-pressure in order to improve reproducibility (i.e. switched from (77 K, 9 atm.) to (180°C, 0.8 atm.)). Our motivation was mainly

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- Pons' presentation in ICCF-4, "Heat after death" [3].
- The paper by Oats and Flanagan on gas loading at high temperature with high reproducibility [4].
- Manduchi's gas loading experiments at high temperature [5].

It appears that gas-loading has higher reproducibility with less dependence on materials.

Our gas-loading experiments started from the goal of high loading ratio in terms of a hot tungsten wire. It led to the studies of

- The pumping effect [6].
- A simple calorimeter configuration [7].
- Nuclear transmutation in a gas-loading H/Pd system [8].
- Correlation between deuterium flux and heat flow at temperatures higher than the boiling point of heavy water [9]
- Gas-loading systems with international collaborations [10–13].

The selective resonant tunneling model has successfully explained the puzzle of excess heat without strong radiation [14], corrected the mistakes in D–T hot fusion data [15], and further improved the formula for six major hot fusion cross-sections [16]. Indeed, these hot fusion data justified the selective resonant tunneling model. Moreover, this model explained the "heat after death" phenomena, three-deuteron fusion reactions [17], and the asymmetry in cooling and heating processes, this led us to find the correlation between deuterium flux and heat flow directly. Based on this model, a conjecture on neutrino emission from palladium deuteride has been proposed [18], and a self-sustaining heat generator using deuterium flux has been outlined [19].

The recent gas-loading experiments at room temperature showed that (1) neutrino emission should be detectable in a palladium tube system with 2 W excess heat running for 7 days, (2) a 30 m long palladium tube (diameter 3 mm, 0.1 mm wall thickness) might be enough to build the first demonstration for a self-sustaining heat generator.

2. Nuclear Tracks in CR-39 (1989–1998)

After the Santa Fe workshop in 1989, it was clear that the neutron or gamma radiation from palladium deuteride must be very weak. Hence, the confirmation of the nuclear nature of this anomalous phenomenon should be based on charged particle detection with low noise and good long-term stability. A CR-39 Solid State Nuclear Track Detector was suggested by the Institute of Nuclear Technology of Tsinghua University. The Institute of Atomic Energy of China kindly provided the high quality CR-39 detectors, and the Institute of High Energy Physics, Chinese Academy of Science, calibrated them using its accelerator.

A graduate student devoted one year to running the gas-loading experiment (thermal cycling in a liquid nitrogen dewar). The positive result was published in the Provo workshop (1990) and in the first AIP Proceedings on this subject [1]. It led to the Chinese involvement in the International Advisory Committee, with help from Professor Scaramuzzi, who was an advocate of gas-loading technology at low temperature. Figures 1(a), (f), and (g) are nuclear tracks from the palladium foils after deuterium loading. Figure 1(d) and (e) are controls using hydrogen instead of deuterium gas, or using deuterium gas with no palladium foil. Figure 1(b) is a comparison with a CR-39 chip radiated by an americium radioactive source, which emitted α-particles of $\sim$5 MeV energy. The numbers in parenthesis are the batch numbers. Figure 1(c) shows the surface effect on charged particle emission. The surface cleaning procedures using aqua regia might have caused the negative result. The reproducibility was still poor, although batches 6 and 7 both produced positive results. Price [20] also found a negative result. Thus, reproducibility was the key issue at the beginning of the gas loading studies.

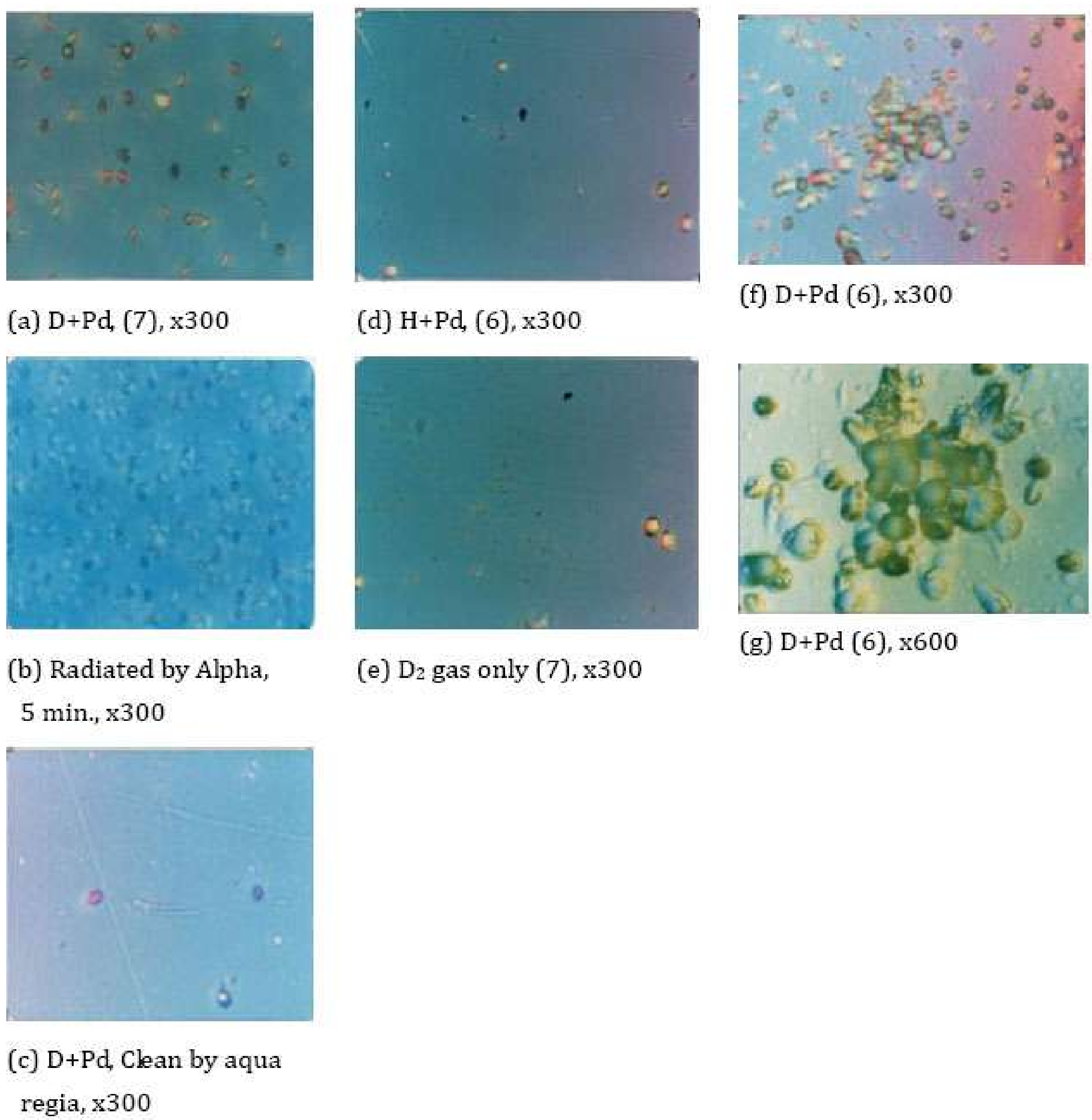

Figure 1. (a) D_2 + Pd, (7), ×300. (b) Radiated by α-particle source (Am) 5 min, ×300. (c) Clean by aqua regia, ×300. (d) H_2 + Pd,(6), ×300. (e) D_2 gas only, (7), ×300. (f) D_2 + Pd,(6) ×300. (g) D_2 + Pd, (6), ×600.

3. Gas-Loading Pd Wire in a Dewar – Pumping Effect

High loading ratio was reportedly good for reproducibility. Materials problems were considered to be a key issue for high loading ratio by several researchers. However, Italian scientists [5] published their successful gas-loading work

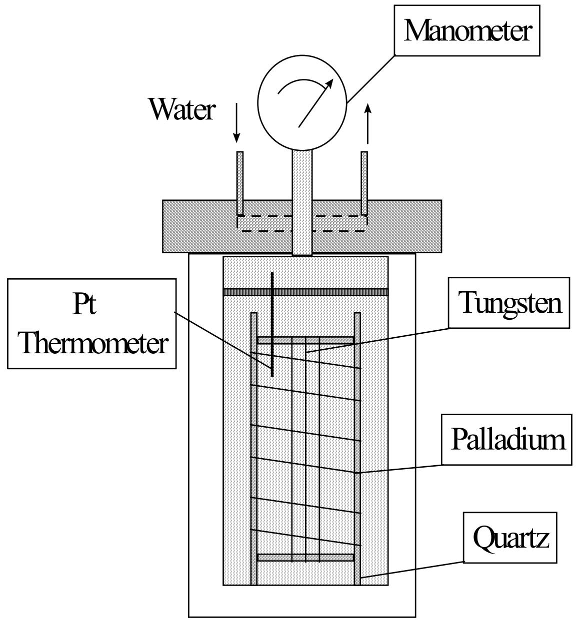

Figure 2. Gas-loading dewar with a long-thin Pd wire and a tungsten heating wire.

with materials from various resources. Oats and Flanagan [4] also claimed that high loading ratio might be achieved using their gas-loading cell without any concern about materials. Hence, a gas-loading cell was constructed in 1995 (Fig. 2) at Tsinghua University.

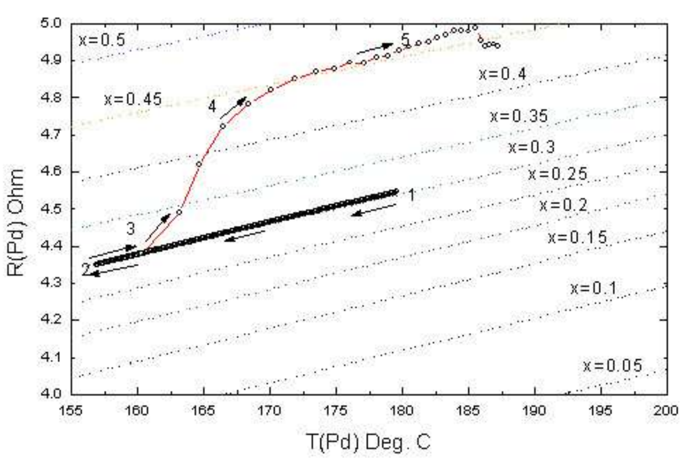

Figure 3. Pumping at 160°C induced a jump in loading ratio(constant current mode).

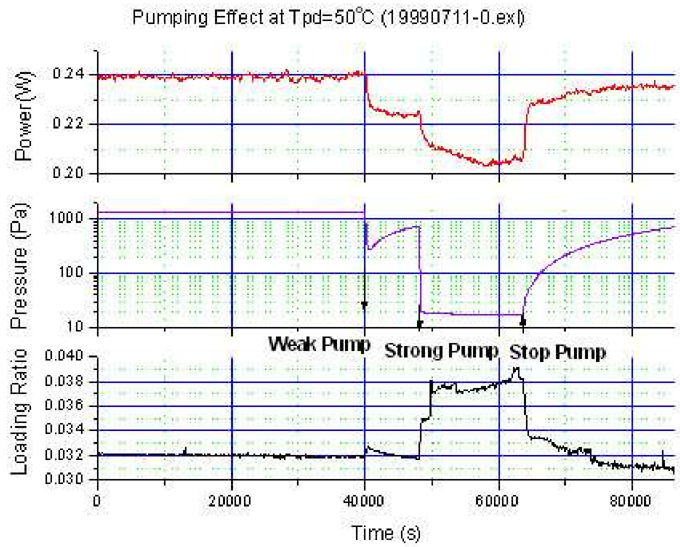

Figure 4. Necessary heating power to keep constant temperature is not a function of pressure, but directly related to pumping rate.

A long-thin palladium wire (250 mm long × 0.34 mm diameter) was wound on a quartz frame and sealed in a stainless steel dewar. A tungsten wire (0.1 mm diameter) was wound on the same quartz frame to provide a high

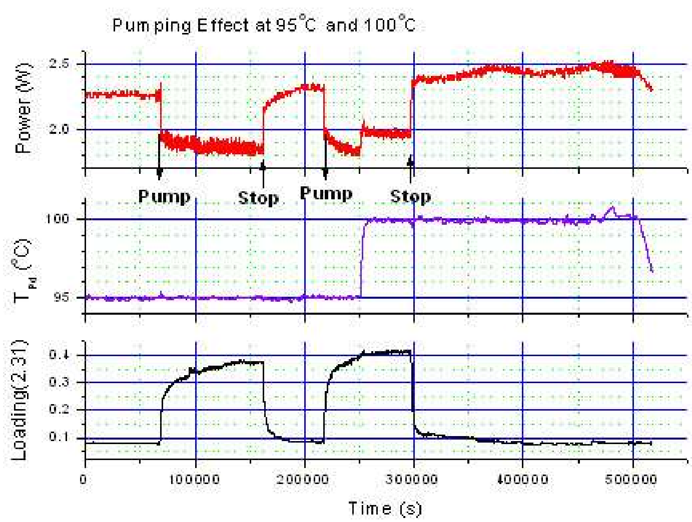

Figure 5. Excess heat under constant $T_{(Pd)}$, 1998,12,13–17. (5_days7.xls).

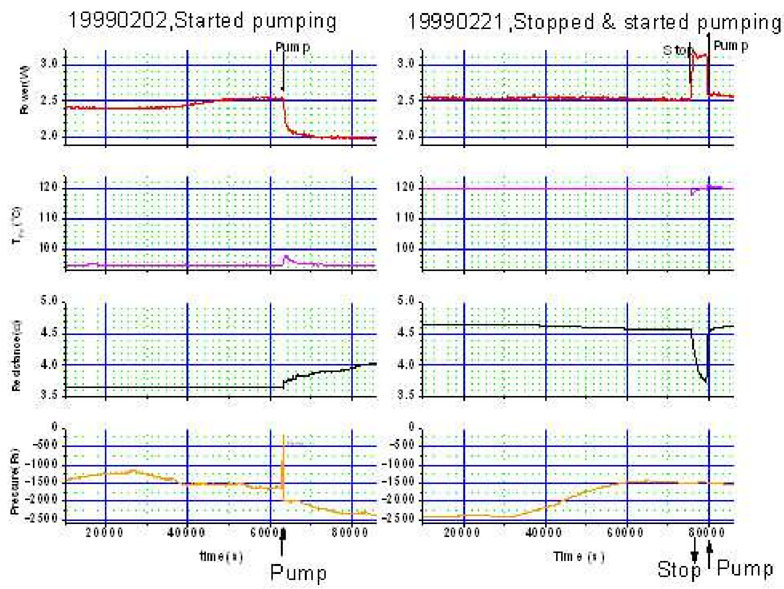

Figure 6. Pumping effect after 20 days at 120°C.

chemical potential of hydrogen atoms from the dissociation of hydrogen molecules. It was designed mainly for the study of the loading ratio; however, its operation led to an unexpected result: the pumping effect. It helped us to discover a correlation between deuterium flux and heat flow.

When we heat the palladium wire, we might anticipate that the loading ratio $x = \mathrm{D/Pd}$ would drop if the pressure is reduced. However, the experiments showed that the loading ratio did not drop. Absorption occurs at high temperatures even if the pressure is lower than one atmosphere. In our gas-loading experiments, a pumping effect appeared in the D/Pd system. When the palladium wire was heated by electrical current its resistance was recorded to monitor the gas loading ratio. The resistance of a palladium wire depends on both its loading ratio and temperature. Figure 3 shows the resistance of the palladium wire, R(Pd), as a function of its temperature, T_{Pd}, and loading ratio. Each dotted line shows the resistance as a function of T_{Pd} for a specific value of x. When we kept the electrical current in the Pd wire, I(Pd), constant and changed the temperature of the dewar wall (T_{w}), then T_{Pd} changed to keep the thermal flow in balance with the electrical power input. The loading ratio was not changed when T_{Pd} dropped from point 1 (180°C) to point 2 (157°C) and rose from points 2 to 3 gradually. Starting the pump at point 3 introduced a jump in x from 0.3 to 0.45 (points 3 to 4). The time between two adjacent data points was only 10 s, so it was a quick jump. This jump was followed by a temperature rise due to the enhancement of the electrical power input in constant I(Pd) mode (points 4–5).

This behavior appeared every time the pump was started. It was first thought (mistakenly) that this jump in resistance was induced by a positive feed-back mechanism as follows. The pumping reduced the gas pressure, so its thermal conductivity was reduced, and T_{Pd} increased. The increase of T_{Pd} would cause further increase of the resistance; hence, the electrical power input would be enhanced and it would cause further increase of T_{Pd}.

However, this feedback mechanism is not supported by the time behavior of the Pd wire resistance. Figure 3 shows that the resistance jumped much faster than the temperature rise at the onset (point 3). It was first a jump in loading

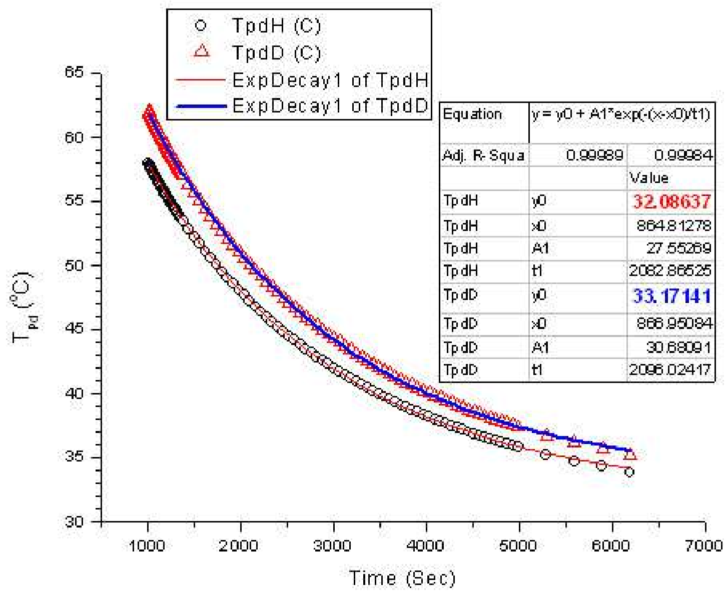

Figure 7. Exponential cooling behavior for D/Pd and H/Pd systems.

ratio, followed by an increase in temperature. Nevertheless, we were most interested in whether there was any "excess heat" accompanying this jump in loading ratio. Thus we have to eliminate first the temperature effect on the electrical resistance of Pd wire, and the effect of temperature on heat radiation.

New experiments were run in a constant temperature mode to avoid the temperature effects on electrical resistance and on heat radiation. Even if we assume that the heat transfer coefficient does not change at this pressure, we have to distinguish the heat of the electrical power input from the "excess heat" (if any). A computer-controlled power supply was used to keep T_{Pd} constant. A tiny Pt thermistor was attached to the palladium wire to monitor T_{Pd}, which could be preset to a fixed value using a computer program. In this way, we eliminated temperature effects during the pumping; however, we still observed the drop in heating power, P_h, needed to keep T_{Pd} constant.

When we started pumping the deuterium dewar at 40,000 s, the power necessary to keep $T_{Pd} = 50°C$ dropped quickly (Fig. 4). One might suspect that the drop in deuterium gas pressure reduces the heat conductivity and hence, the necessary electrical power. This explanation is not correct.

When the pressure in the dewar increased slowly from 250 to 800 Pa the heating power, P_h did not increase with the pressure at all. When we increased the pumping rate again at 48,000 s, P_h dropped suddenly again. It is clearly shown that the P_h is a function of pumping rate but not a function of pressure in the dewar. Particularly, when we stopped the pumping at 63,000 s, P_h jumped up quickly, although the gas pressure was still very low in the dewar. These data exclude the possibility of explaining the jump of P_h as a jump in heat transfer coefficient. Indeed, the sudden change in P_h is possibly an evidence of "excess heat". Assuming a constant heat transfer coefficient, this drop of P_h corresponds to an "excess heat" of 0.035 W, it is about 15% of the input power (0.24 W) . The volume of the Pd wire is about 0.237 cm^3; hence, the power density is on the order of 0.15 W/cm^3 at 50°C.

Based on the high temperature electrolysis experiments, we might anticipate that the "excess heat" would be enhanced at higher temperature. Figure 5 showed similar data at 95°C and 100°C. When T_{Pd} was kept constant, the

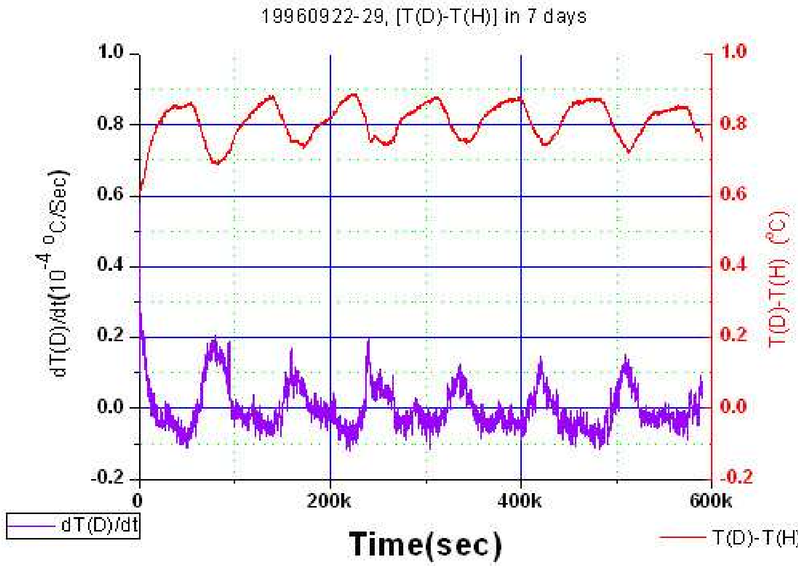

Figure 8. Temperature difference between D/Pd and H/Pd systems.

pumping at 69,000 s introduced a resistance jump and a power drop again. Since the T_{Pd} was kept constant, the change in resistance was attributed to loading, and the change in power was due to the "excess heat". When the electrical power dropped from 2.28 to 1.84 W, the Pd temperature was still kept at 95±0.5°C. If the heat transfer coefficient did not change during the pumping; then, the "excess heat" power was about 0.44 W, which is 19% of the power input. When the pumping was stopped at 1,60,000 s, the necessary power jumped back to 2.3 W. This effect was reproduced

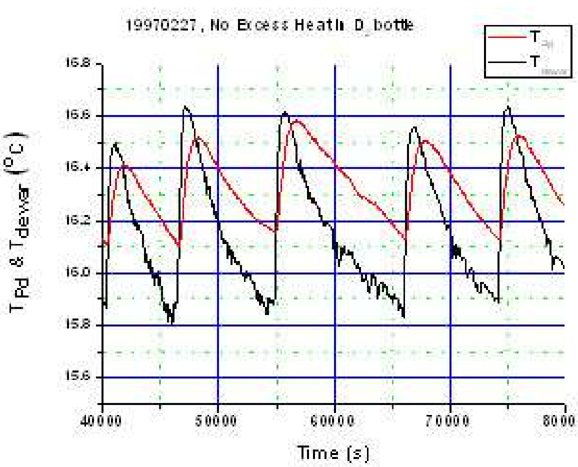

Figure 9. Temerature of palladium wire and dewar when there was no excess heat.

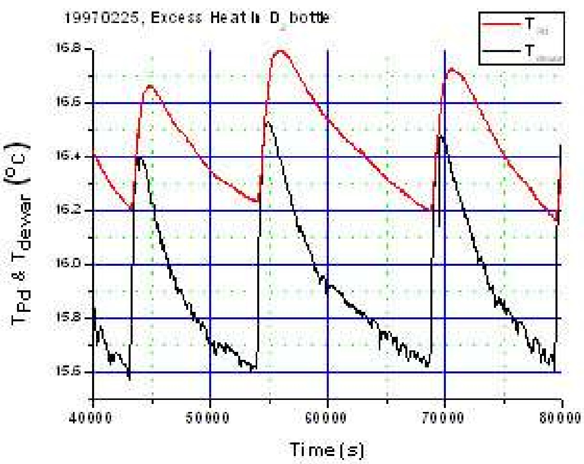

Figure 10. Temerature of palladium wire and dewar when there was excess heat.

the next day, at 2,18,000 s the necessary power dropped again to 1.84 W due to the pumping. There were two jumps in power at 2,50,000 and 2,97,000 s, respectively. They were different in nature. The first power jump at 2,50,000 s was due to the computer program setting. We required that $T_{\rm Pd}$ should be raised from 95°C to 100°C at 2,50,000 s; the computer increased the power input to meet this requirement. However, at 2,97,000 s we stopped pumping, and the power jumped up instantly to keep $T_{\rm Pd} = 100$°C. The power densities for this "excess heat" near 95°C and 100°C are about 1.8 and 2.1 W/cm^3, respectively.

We have extended this experiment to $T_{\rm Pd} = 120$°C, and kept pumping for as long as 20 days. The $P_{\rm h}$ value was kept low until the pumping was stopped after 20 days. The corresponding "excess heat" power was 0.6 W at 120°C (greater than 2.5 W/cm^3). Figure 6 shows this pumping effect after pumping 20 days. This experiment was started on February 2, 1999 near 95°C, the pumping reduced the heating power from 2.5 to 2.0 W (the plot on the top-left). After pumping 20 days, the temperature had been raised to 120°C by increasing heating power to 2.5 W (the plot on the top-right). On February 21, 1999 near 120°C, stopping the pumping enhanced the necessary heating power to 3.1 W; then, the pumping again reduced the heating power back to the 2.5 W level. The computer-controlled temperature was maintained at the preset level (second row of plots). The resistance of the palladium wire increased every time we started pumping (third row of plots). The fourth row clearly shows that the pressure did not change much after pumping 20 days, because there was very low deuterium gas left in the vacuum vessel. However, the pumping effect still existed vigorously. It might imply that pumping effect was related to the property of palladium wire (near the surface layers). It is not related to a reduction of the heat transfer coefficient.

4. Difference Between D/Pd and H/Pd Systems

It was a prevailing trend in 1990's to test the difference between D/Pd and H/Pd systems. Our long-thin palladium wire loading vessels provided a convenient calorimeter for this purpose, because we put the long-thin palladium wire frame inside a dewar to get higher temperature with much lower heating power. We made a pair of twin loading systems. They were heated by tungsten wires with same electrical current. The parameters for these twin systems are listed in Table 1.

Figure 7 shows the cooling feature of these twin systems. The values of $T_{\rm Pd}$ are changing with time. Both cooling curves may be fit with an exponential,

$$y = y_0 + A_1 \exp[-(x - x_0)/t_1]. \tag{1}$$

Here, y is the temperature of palladium wire, and the x is the time. This exponential behavior implies that this sytem may be described by a heat transfer equation with three parameters (C, k, S):

$$C\frac{\mathrm{d}T_{\mathrm{pd}}}{\mathrm{d}t} = -k(T_{\mathrm{pd}} - T_{\mathrm{room}}) + S, \tag{2}$$

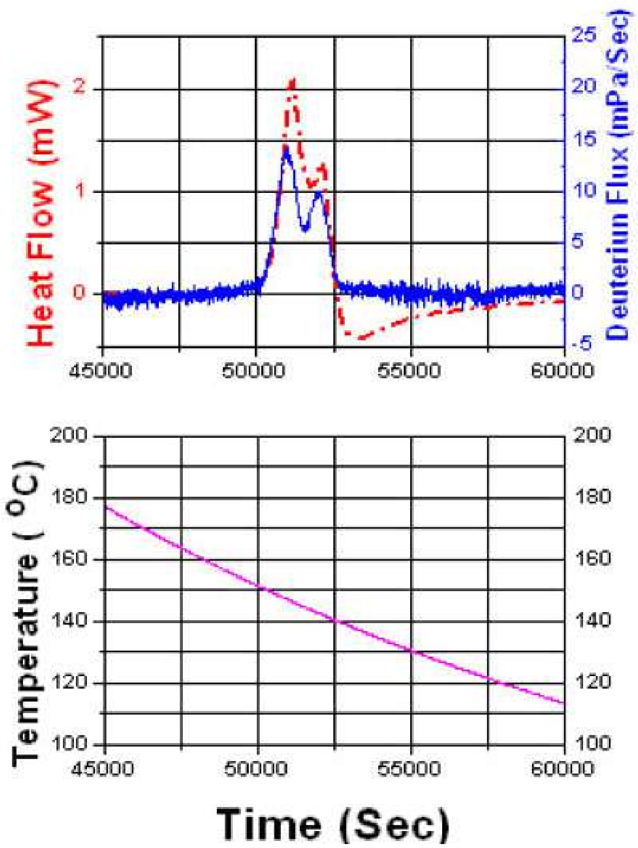

Figure 11. Correlation between heat flow and deuterium flux.

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)

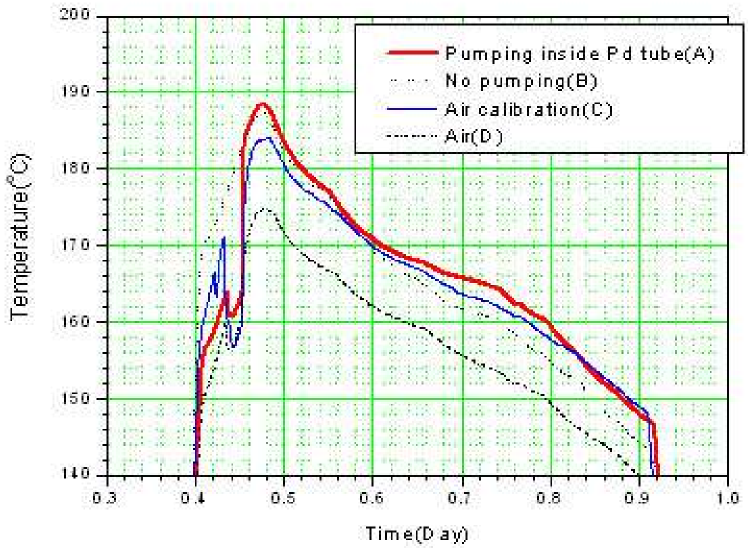

Figure 12. Comparison among 4 cooling cases shows the excess heat correlated with pumping effect.

Here C is the heat capacity of the system, k is the heat transfer coefficent and S is the heat source (if any). From the parameters of the fitting curves, we find that

$$y_0 = T_{\text{room}} + \frac{S}{k}, \qquad t_1 = \frac{C}{k}. \tag{3}$$

In Fig.7, the triangles are for the D/Pd system, and the circles are for the H/Pd system. The difference in y_0 implies different S for D/Pd and H/Pd systems, respectively, because T_{room}, k, and C are almost the same for D/Pd and H/Pd systems. Qualitatively, we may say that the D/Pd bottle is hotter than H/Pd bottle by 1.1°C. From Fig. 4, we know that 0.24 W may raise the T_{Pd} from room temperature (30°C) to 50°C; therefore, 1.1°C might correspond an excess heat source of 12 mW in the D/Pd system.(roughly, 50 mW/cm^3) Usually, the first question one may ask is the calibration of the thermistors. Can we eliminate the possibility of a thermistor problem? (Was the temperature difference just caused by thermistor error?). Fortunately we measured the temperature difference between D/Pd and H/Pd systems for 7 days (Fig. 8). It was not constant, but varies with the derivative of the temperature ($\mathrm{d}T_{\text{Pd}}/\mathrm{d}t$). The temperature difference, $[T(\text{D})\text{-}T(\text{H})]_{\text{Pd}}$ (the thick line and the right ordinate) reaches a maximum when ($\mathrm{d}T_{\text{Pd}}/\mathrm{d}t$) (the thin line and the left ordinate) reaches a minimum.This is consistent with the selective resonant tunneling model. It seems that the D/Pd bottle is always warmer than H/Pd bottle. This is not an error in the thermistors. It should be noticed that the time scale is expanded to 6,00,000 s, the temperature is almost in equilibrium with the environment at every instant. One more piece of evidence is from the D/Pd bottle itself. Figures 9 and 10 show the temperature of palladium wire and the temperature of the dewar when we put the dewar in the water bath of a thermostat. Due to the heater in the thermostat, which acted intermittently, the temperature of the dewar varied around a pre-set value. When the D/Pd bottle had no heater inside, T_{Pd} was supposed to vary with the dewar. When there was no excess heat inside the D/Pd bottle, the temperature of palladium wire increased only if $T_{\text{dewar}} > T_{\text{Pd}}$; and the temperature of palladium wire decreased only if $T_{dewar} < T_{Pd}$. This is clearly shown in Fig. 9: The T_{Pd} (solid line) went up whenever T_{dewar} (dotted line) was higher than T_{Pd}, and T_{Pd} (solid line) turned downwards whenever T_{dewar} (dotted line) was lower than T_{Pd}. Hence the interception points of the T_{Pd} and T_{dewar} curves are always at the peak of the solid line or at the bottom of the valley of the solid line (i.e. $dT_{Pd}/dt = 0$ at $T_{Pd} = T_{dewar}$). When Fig. 9 shows that the calibration of thermistor T_{Pd} and T_{dewar} was correct, Fig. 10 shows that there had to be excess heat inside the D/Pd bottle on Feb. 25, 1997, because T_{Pd} was increasing even if T_{Pd} was higher than T_{dewar}.

5. Correlation Between D_2 Flux and Heat Flow

A decisive experiment was conducted in 2000 in collaboration with the Department of Chemistry at Tsinghua University where the high precision calorimeter (C-80D) was located. The most important feature of the C-80D calorimeter is that it is based on the Seebeck effect; hence, it eliminated any concern about the change of the heat transfer coefficient during the pumping. The C-80D, imported from France (SETARAM), has such high precision that it is able to detect one microwatt heat flow from its reaction vessel. When we put a piece of sealed palladium tube inside the reaction vessel of the C-80D calorimeter, and pumped out the deuterium gas from outside of the Pd tube, the deuterium gas inside the Pd tube permeated through the thin wall of the Pd tube. The pressure in the vessel was determined by the balance between the deuterium flux and the pumping rate. The change in the deuterium flux is detected by the deuterium pressure in the vessel.

We found that the heat flow in C-80D was accompanied by a change of deuterium pressure in the vessel while the pump was working continuously. In Fig. 11, the thick line shows the heat flow recorded by the C-80D calorimeter, and the thin line shows the derivative of the pressure. The two peaks in heat flow were correlated with the two peaks in the deuterium flux. This phenomenon was observed when the heater in the C-80D was turned off, and the temperature of the vessel was cooling down slowly through 150°C to 140°C. The selective resonant tunneling theory predicted that the excess heat would be observed preferably in the slow cooling process, because the slow cooling down process might elongate the time in the resonance. We did observe this asymmetry in the cooling and heating processes.

Figure 12 shows the scale-up of this pumping effect. The length of the palladium tube was lengthened from 26 to 200 mm, and single Pd tube configuration was changed to a bundle of 5 Pd tubes. Under the same situation (room temperature, pattern of heating power, gas pressure, etc.) the temperature of Pd tube was monitored during the cooling processes. There were 4 different cooling cases: A, Cooling in the deuterium gas and pumping inside the Pd tube; B, Cooling in the deuterium gas without pumping; C, Cooling in the air and with 1 W inner heater on the Pd tube for calibration; D, Cooling in the air with inner heater off. It is evident that the temperature of Pd tube was the highest in case A; and temperature of the Pd tube was the lowest in case D although the heat transfer coefficient of deuterium gas was supposed to be greater than that of air. The temperature of the Pd tube for case B was lower, it implied that the pumping inside the Pd tube was very important for this excess heat. The calibration using inner heater on the Pd tube (case C) gave a rough estimate of the power of excess heat. It was on the order of 1 W, and it lasted 10 h.

6. A Stride Towards Neutrino Detection

The neutrino detector at the Neutrino Science Research Center in Japan requires a minimum neutrino flux for its operation. On the surface of the big liquid scintillator sphere, there has to be at least 10^6 neutrinos per cm^2 per second. For this 14 m diameter sphere, it requires a source of 10^{12} neutrinos/s at the center of the sphere. If each neutrino accompanies 10 MeV reaction energy; then, 1 W reaction energy just provides the necessary neutrino flux for detection. We know that most of the reaction energy is carried away by neutrinos [18], only the heavy charged particles leave their energy in the palladium deuteride—the excess heat. The excess heat is from the recoil energy of the neutrino emission. Hence, we might expect even many more neutrinos from 1 W excess heat ($\sim 10^{15}$ neutrinos/s). If the detection of neutrinos is successful; then, nuclear energy without nuclear contamination will have been demonstrated.

If we scrutinize the "excess heat" data in the past 20 years, the highest "excess power" density with long enough time was the "heat after death" in 1994 [3]. In average, the "excess power" density was estimated to be 3.7 kW/cm^3 in 3 h. Hence each atom of reactant released $\sim$220 eV only. This is on the order of recoil energy. It confirms that most of the reaction energy is carried away by neutrinos. Indeed this is the physics basis of "nuclear energy without nuclear contamination".

Acknowledgments

This research is supported by the Ministry of Education, The Ministry of Science and Technology (Fundamental Research Division), Natural Science Foundation of China (#10145004, #10475045), The China Association for Science and Technology, and Tsinghua University (Basic Research Fund (985-III)), National Basic Research Program of China (2009CB226113).

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