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PROGRESS IN GAS-LOADING D/Pd SYSTEM

—The feasibility of a self-sustaining heat generator—

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Great progress has been made after 14 year of experiments with the gas-loading D/Pd system. 6 watts of "excess heat" were generated in a gas-loaded D/Pd system for 9 hours continuously. This experiment has been repeated 6 times already in various configurations. The "excess power" density in the Pd disk is more than 100 W per cubic centimeter, which is about the power density in a fuel rod of a thermal neutron fission reactor.

1 Introduction

Gas-loading has been applied (instead of electrolysis) to load deuterium into the palladium lattice at Tsinghua laboratory since 1989[1-12]. There are four advantages to the gas-loading D/Pd system: safety, sensitivity, low cost, and a higher operating temperature. Among these four advantages, the most important one is that the operating temperature can be higher than 100ºC. Not only is Carnot efficiency higher, but the higher temperature of working media led to discoveries about the reaction in the temperature domain above the boiling point of heavy water, which could not have been made with unpressurized liquid systems.

In ICCF-9, a ***correlation*** between deuterium flux and heat flow was first reported [1] in the temperature range of 140-150ºC after 16 repetitions in a high precision Calvet calorimeter (C-80D). After ICCF-9, based on that correlation, a new gas-loading D/Pd system was built to make use of this discovery, with the ultimate goal of developing a self-sustaining heat generator. Two improvements were made to keep the excess heat continuous:

(1) A constant deuterium gas supply to keep the pressure difference across the Pd thin film.
(2) An electrical heater was used to heat the Pd film, and to calibrate the calorimeter.

The key issue has been the discovery of the anomalous behavior of the deuterium flux. Usually, the deuterium flux permeating the Pd film was considered as a monotonic function of the temperature (T_{Pd}). The deuterium flux was supposed to increase dramatically with the temperature. However, it was discovered that at certain temperature, T_r, the deuterium flux reached a peak value and then declined. In other words, the deuterium flux dropped in an anomalous way when temperature was just over T_r. This drop in deuterium flux was accompanied by a drop in heat flow, conforming to the correlation between heat flow and the deuterium flux that we observed previously [1]. Consequently, a negative feed-back mechanism was established in Pd film when the temperature reached the higher temperature side of the flux peak at T_r, i.e. when $T_{Pd} > T_r$, the heat flow decreased when T_{Pd} increased; hence, T_{Pd} would decrease back until T_{Pd} reached a steady state. The new apparatus was designed based on this concept in mind.

2 Apparatus

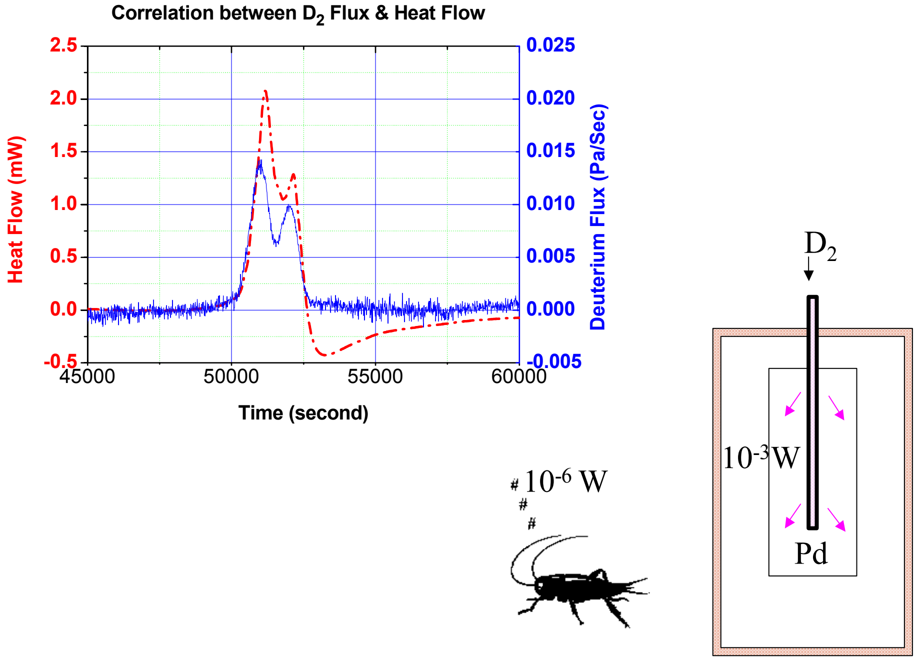


Figure 1. Calvet calorimeter confirmed the correlation between deuterium flux and heat flow

A schematic of the previous apparatus [1] is shown in Fig. 1 (right lower corner). The high precision Calvet calorimeter was selected for excess heat measurement. It was sensitive to a heat flow in the order of micro-watt, which is about the variation of metabolic heat of a singing insect. When the deuterium flux through the thin wall of the Pd tube reached a peak at T_r (the thin solid line in the upper left corner of Fig. 1), the heat flow reached a peak as well (shown by the thick dash-dot-dash line). The peak value was in the order of 2 milliwatts. The high sensitivity of Calvet calorimeter was well suited to confirm the correlation between this heat flow and the deuterium flux, but there was no direct measurement of temperature of the Pd tube. It was assumed that the temperature of the Pd tube, T_{Pd}, was determined by the environment. (That is, the heater and the reaction vessel were assumed to be isothermal). Indeed we knew nothing about the temperature distribution on the Pd tube, and we did not know how to control this distribution of temperature in the Calvet calorimeter.

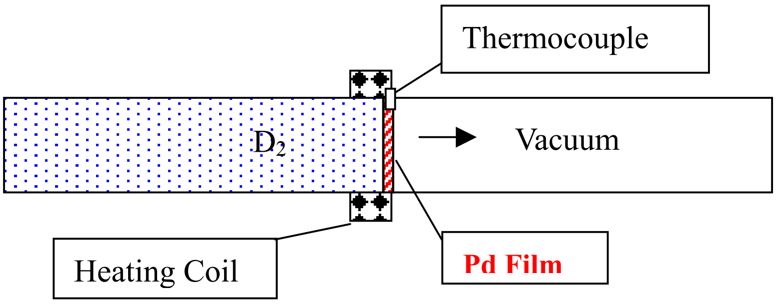


Figure 2. New apparatus to detect and control the temperature of Pd disk from edge

Fig. 2 shows the schematics of the new apparatus designed to improve the detection and control of the temperature of the Pd film. A palladium disk (ϕ20mm×0.1mm) was inserted between two stainless steel cells. An electrical heater was wound around the Pd disk to heat the disk from its edge. A thermocouple was attached to the edge of the Pd disk to detect its temperature at edge. The left cell was filled with deuterium gas (about 1 atm.), and the right cell was evacuated by a mechanic pump to about 150 Pa.

3 Results

When only the circumference of a round Pd disk was heated, the temperature gradient pointed to the direction of the radius; i.e. the temperature at edge was higher than that at the center of the Pd disk. Once the temperature approached T_r, a heat source appeared first at the edge of the palladium disk which "ignited" the Pd disk from the edge to the center. As a result, the temperature gradient reversed its direction suddenly; i.e. the temperature at center was higher than that at edge. Due to the abovementioned negative feed-back mechanism, this new distribution of the temperature was quite persistent. Even if the heating power was reduced later to lower the temperature at the edge of Pd disk, the temperature at center remained higher than T_r.

Fig. 3 shows schematically the temperature distribution of the Pd disk in the period of heating-up (lower contours) and cooling-down (upper contours), respectively. When the heater started heating the edge of Pd disk, the temperature distribution curve was of concave shape first (see the middle of Fig. 3). When the heating power was increasing, the edge temperature was raised to the region of heat generation (the dotted line region where $T_{low} < T_{Pd} < T_{upper}$); then, the heat flow would quickly raise the temperature of inner part of the Pd disk. This resulted in an inversion of the temperature gradient. The concave curve turned into a convex curve as shown by the two curves in the middle of Fig. 3.

This inversion appeared as a hysteresis in plot of T_{Pd} versus heating power in Fig. 4. When the electrical heating power was raised, the temperature at the edge of Pd disk, T_{Pd}, increased also. A jump of T_{Pd} appeared at about 110°C in Fig. 4. It implied that there was an inner heating power in the Pd disk. When the external heating power was raised further, T_{Pd} curve went up smoothly again. When the electrical heating power reached its maximum, and returned; the T_{Pd} decreased smoothly until the electrical heating power reduced to zero where the T_{Pd} dropped with time.

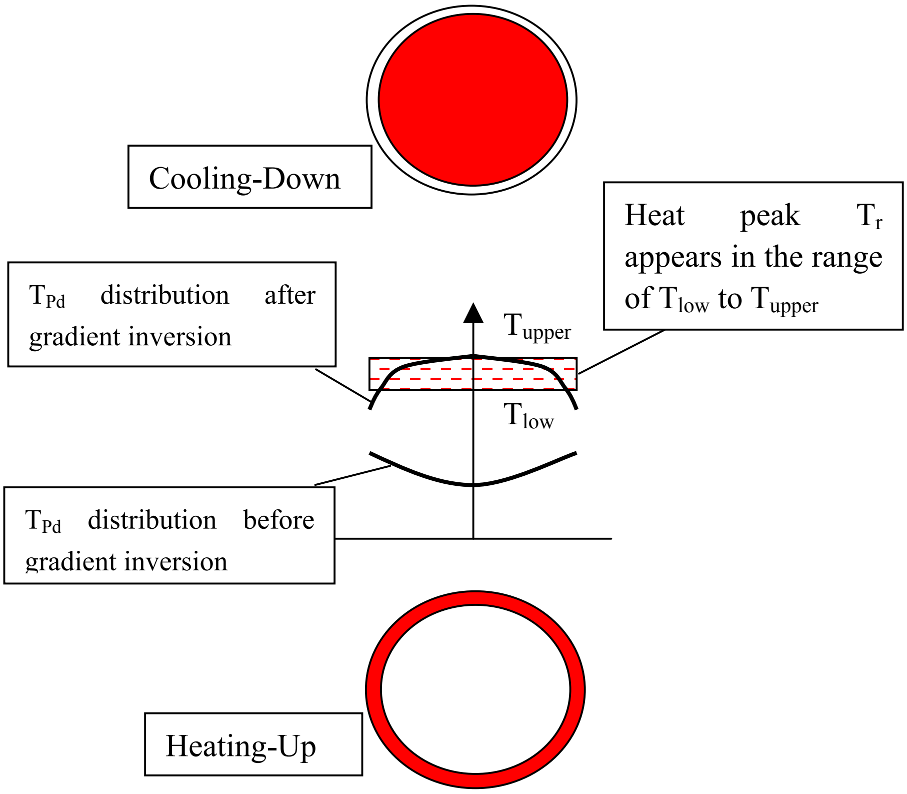


Figure 3. Temperature distribution on the Pd disk during the heating-up and cooling-down phase

In a comparison blank run, we replaced the Pd disk with a copper disk. Fig. 5 shows the plot of T_{Pd} versus heating power for the case of copper disk (the black crosses). In order to show the temperature variation with time, for each step of electrical heating power, the edge temperatures at each time step were shown by a series of crosses aligned in a vertical line. It approached its equilibrium value gradually. There was a distinct difference between the Pd and Cu behavior. For the copper disk there was no temperature jump or wide hysteresis behavior as that for Pd disk. For the Pd disk the cooling-down curve was far above the heating-up curve when T_{Pd} is less than 80ºC. Nevertheless, for the copper disk the cooling-down curve was close to the heating-up curve even if T_{Pd} is less than 80ºC.

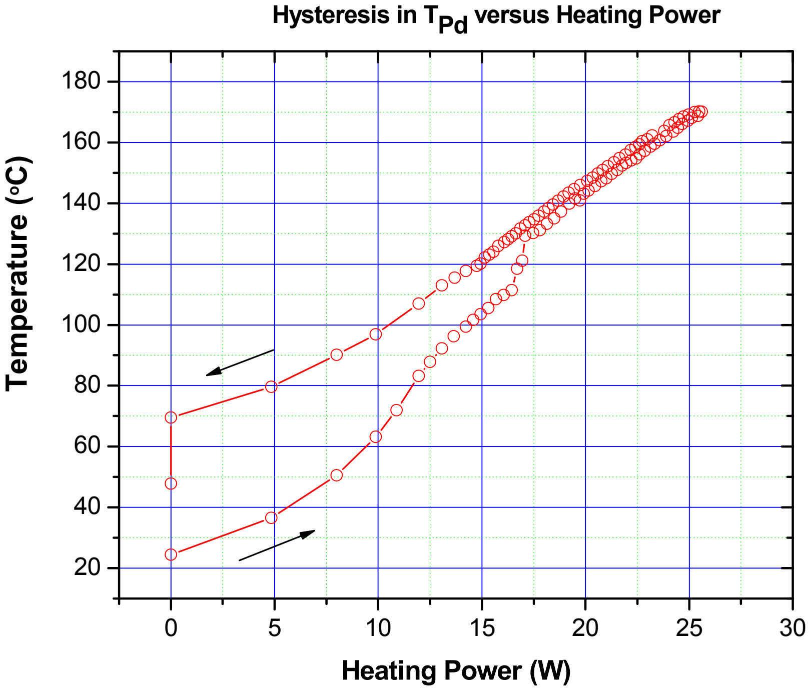


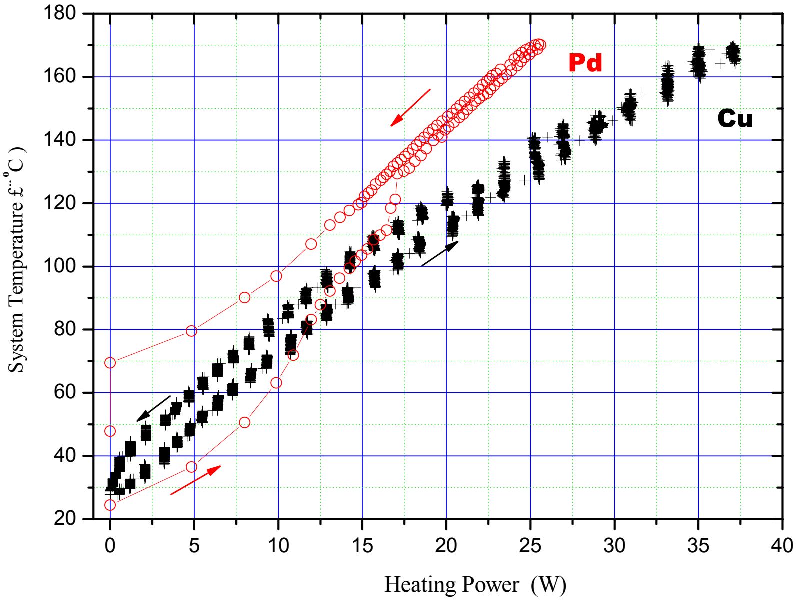
Figure 4. Hysteresis in the plot of T_{Pd} versus heating power

4 Discussion

A quantitative analysis have been done as follows. Because the equilibrium time was very long for the system, the heating and cooling process was a time-consuming if we waited to reach the equilibrium at every power-step. Instead of waiting, we tried to analyze the experimental data when the system did not reach its equilibrium with the environment. For this purpose an integration method was applied.

The basic calorimetric equation was

$$\underbrace{C_M}_{\text{Heat Capacity}} \frac{dT}{dt} = \underbrace{-k(T - T_{room})}_{\text{Heat Transfer}} + \underbrace{IV}_{\text{Joule Heating}} + \underbrace{Q_x}_{\text{?Excess Heat}} \qquad (1)$$



Here C_M is the heat capacity of the system in order to consider the non-equilibrium feature (i.e. $dT/dt \neq 0$). k is the heat transfer coefficient; T and T_{room} are the temperature of the system and the room, respectively. I and V are the electrical current and voltage of the heating power, respectively. Q_x is the excess heat source (if any).

Usually, the derivative term on the left hand side of equation (dT/dt) would introduce the major error due to the temperature fluctuation; hence, the integration form of equation was derived as

$$C_M(T_f - T_i) = -k\int_i^f (T - T_{room})dt + \int_i^f IVdt + \int_i^f Q_x dt \qquad (2)$$

Here, T_i and T_f are the initial and final temperature of the system, respectively. In order to find the heat transfer coefficient, k ; we may integrate all the data for the heating-up and cooling-down process. Then $T_i = T_f$ which will eliminate the unknown C_M first. For the copper disk, Q_x is assumed to be zero also. Having integrated the joule heating power, we obtained

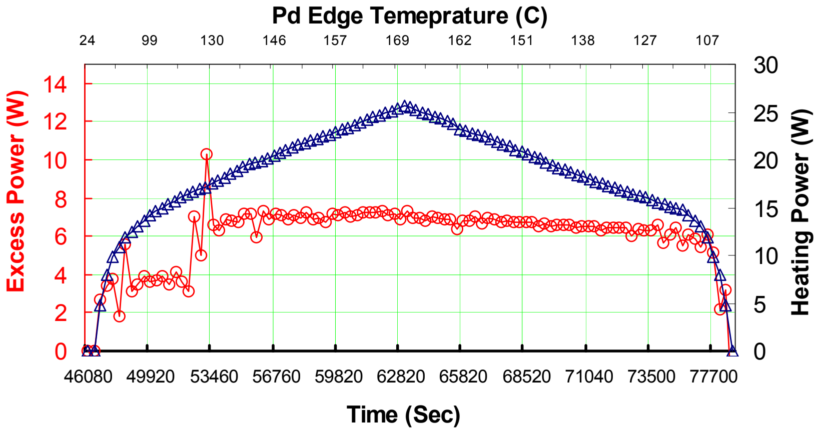


Figure 6. Excess power and input electrical power

$$k = 0.222\ W / {}^{o}C, \tag{3}$$

Then, we select the maximum temperature as T_f , and the heat capacity of the system can be obtained from the same integration equation for copper disk as

$$C_M = 120 \pm 1\, J / {}^{o}C \tag{4}$$

Indeed, the heating-up and cooling-down were two sets of independent data, and we can use them to calculate the heat capacity independently. The difference of the heat capacity for heating-up and cooling-down is less than 1 J/°C. This is a good confirmation of our assumption on the constancy of *k* and C_M.

Since the replacement of copper disk by palladium disk does not change the calorimetric feature of the system much, we further assume that the heat transfer coefficient and the heat capacity are same for the Pd disk and Cu disk system. We can estimate the power of the excess heat for the Pd disk system as

$$\frac{\int_1^2 Q_x dt}{(t_2 - t_1)} = \frac{C_M (T_2 - T_1) + k \int_1^2 (T - T_{room}) dt - \int_1^2 IV dt}{(t_2 - t_1)} \tag{5}$$

Here t_1 and t_2 are the starting and ending time of each power step. Fig. 6 shows the excess power as a function of time. The excess power (open circles) are about the same order of magnitude as that of the external electrical heating power (triangles). The maximum excess power is more than 10 W while the electrical heating power is less than 9 W. Indeed the excess power density in the Pd disk is more than 100W/cm^3 which is greater than the power density in a fuel rod of the thermal neutron fission reactor.

The total excess heat released in 9 hours was 192 kJ. Based on the total number of deuterium atoms permeating the Pd disk (2.6×10^{20}), we estimate the average energy released from each deuterium atom was 4.6 keV. Even if we average over all the palladium atoms (0.3 g. of Pd ~0.003 mole), it gives 0.44 keV per Pd atom. In other words, this amount of excess heat is about 57 MJ/Mole for Pd which is much greater than any heat of formation, heat of solution, and chemical bond energy. We conclude that there was a non-chemical origin for such a large amount of excess heat.

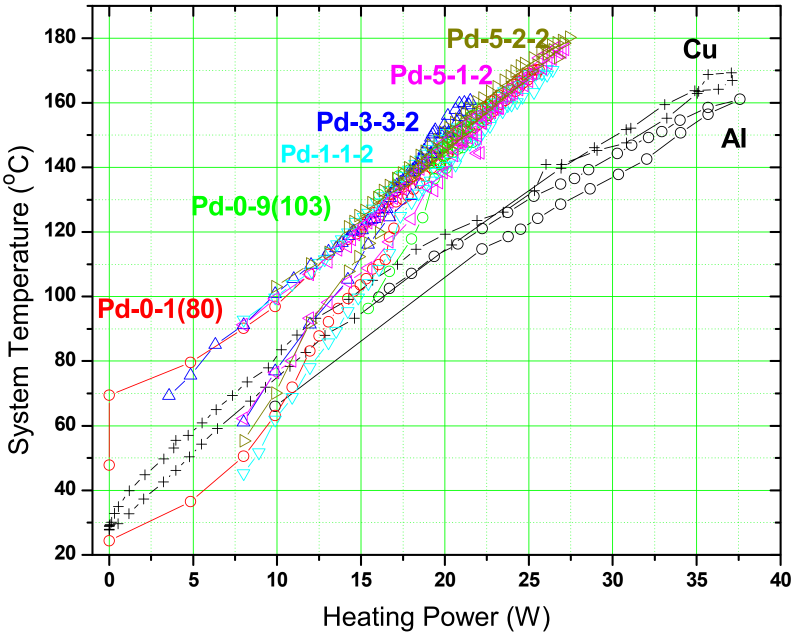


Figure 7. Reproducibility of temperature jump in Pd film

This hysteresis behavior of the system temperature versus heating power has been repeated in six different configurations shown by Fig. 7. We changed the pressure difference across the Pd disk from 80 kPa (red open circles) to 103 kPa (green open circles). We put varying layers of a thin coating on the surface of Pd disk, including one layer of coating (light blue triangles), 3 layers of coating (dark blue triangles), and 5 layers of coating (pink and dark green triangles). These layers are composed of titanium carbide (~2nm) and palladium (~20nm) alternatively. The aluminum disk data (black open circles) are added as a control also, and it was very close to the copper disk data (black crosses). Hence, the excess heat in Pd disk is confirmed, and reproducible.

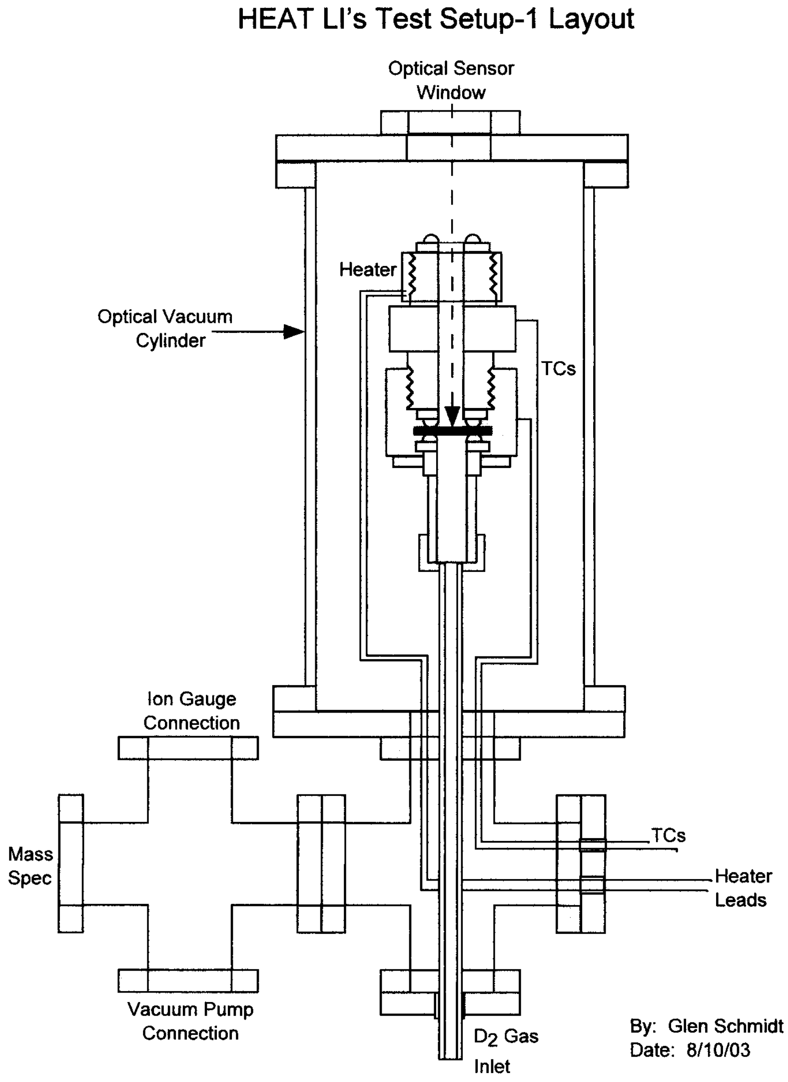


Figure 8. New set of apparatus with bell jar for thermal insulation.

5 The feasibility of a self-sustaining heat generator

This behavior implied that it might be possible to construct a self-sustaining heat generator, if we can generate enough excess power in the Pd disk, with insulation good enough to maintain this temperature with zero external heating power.

Fig. 8 shows schematics of a new set of apparatus provided by Dr. Schmidt at Institute for Engineering Reach and Applications. The whole D/Pd system was installed in a bell jar which was evacuated by an ion pump and a turbo-molecular pump. This vacuum greatly improved the thermal insulation of D/Pd system. 40 Watts of electrical heating power would heat the D/Pd system to 400ºC, i.e. the heat transfer coefficient k ~ 0.1 W/ºC which was less than 0.222W/ºC of present D/Pd system. The preliminary results in this new set of apparatus will be reported later.

Acknowledgements

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References

[1] Xing Z. Li, Bin Liu, et al.,' "Super-Absorption"—Correlation between Deuterium Flux and Excess Heat— ', *Condensed Matter Nuclear Science*, Proceedings of ICCF-9, May 19-24, 2003, Beijing China, edited by Xing Z. Li, (Tsinghua University Press 2004), p.202.

[2] Xing Z. Li, Bin Liu, et al., '"Pumping Effect" –Reproducible Excess Heat in a Gas-loading D/Pd System—', ibid. p.197.

[3]Xing Z. Li, et al. "Anomalous Nuclear Phenomena and Solid State Nuclear Track Detector," *Nucl. Tracks. Radiat. Meas.*, **22**, 599 (1993). See also: Shi.Y. Dong, Xing Z. Li, et al., "Precursor to 'Cold Fusion' Phenomena and the Detection of Energetic Charged Particles in Deuterium/Solid System," *Fusion Technology*, 20, 330 (1991).

[4] Xing Z.Li, Wei Z.Yu, et al., " 'Excess Heat' Measurement in Gas-loading D/Pd System," *Journal of New Energy*, Vol.**1**, no.4, Fall 1996, p.34.

[5] Xing Z.Li, Shu X.Zheng, Hai F.Huang, Gui S.Huang, Wei.Z.Yu, "New Measurement Of Excess Heat in a Gas-Loading D/Pd System," *Proceedings of 7-th International Conference on Cold Fusion*, April 19-24, 1998, Vancouver, Canada. Edited by ENECO, Inc. Salt Lake City, Utah USA, p.197.

[6] Feng S. Bu, Xing Z. Li, et al., "Loading Ratio Study in a Gas-loading System," *Proceedings of 6-th International Conference on Cold Fusion*, Vol.**1**, edited by M.Okamoto, October 13-18,1996, Japan, (Published by New Energy and Technology Development Organization, and Institute of Applied Energy), p187.

[7] Gui S. Huang, Xing Z. Li, " A Possible Phase Transition in a Gas-loading D/Pd System," ibid. Vol.1, p198.

[8] Xing Z. Li, et al., "Super-Absorption—The Effect of Crystal Lattice on Enhancement of Nuclear Reaction—", *Proceedings of 2001 Chinese Physical Society Fall Meeting,* Sept. 20-23, 2001, Shanghai, China. P.98 (in Chinese)

[9] Xing. Z. Li, J. Tian , M. Y. Mei and C. X. Li, "Sub-barrier Fusion and Selective Resonant Tunneling," *Phys. Rev.* ,**C 61**, 024610 (2000).

[10] Xing.Z.Li, "Nuclear Physics for Nuclear Fusion." *Fusion Science and Technology,* **41**, 63 (2002).

[11] Jian Tian, Xing Z. Li, et al., "Anomalous Heat Flow and Correlation with Deuterium Flux in a D/Pd Gas-loading System", *Condensed Matter Nuclear Science*, Proceedings of ICCF-9, May 19-24, 2003, Beijing China, edited by Xing Z. Li, (Tsinghua University Press 2004), p.353.

[12] Wei Wu, Xing Z. Li, et al., “Anomalous Heat Effect during Permeation of Deuterium Gas through the Palladium Tube, ” ibid. p.412.