Wave Nature of Deuterium Flux Permeating through the Palladium Thin Film with Nanometer Coating Layers --- (I) Experimental Observation

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Abstract. Wave nature of deuterium flux permeating through the palladium thin film is revealed using nanometer coating layers. Three sets of experimental data [1,2,3] agree with wave in multiple-layer theory quantitatively or qualitatively. Other than granular particle diffusion model and surface catalyst model, the wave nature of deuterons inside the coating layers must be included in order to explain the experimental phenomena.

1. Introduction

The anomalous deuterium flux permeating the thin wall of palladium tube showed a peak while the temperature of Pd tube was monotonically decreasing through 150°C to 120°C [1]. It was a hint that other than the diffusion there must be some different mechanism governing the deuterium flux permeating the thin Pd film, otherwise diffusion coefficient was supposed to be a monotonic function of temperature. When deuterium molecule dissociated into 2 atoms, and entered the Pd surface as if a granular particle, it would be reflected by the single surface layer only. However, if its behavior was similar to that of a wave; then, it would feel more layers behind the surface layer. The reflecting wave from the surface would be determined not only by the surface layer, but also by the layers behind the surface. Then the reflection of wave should depend on the interference of several reflecting waves from several layers, and manifest itself a peak-wise behavior. This distinct feature might be tested by a deuterium flux permeating through a palladium thin film with multiple nanometer coating layers.

2. Experimental results

Fig.1 shows the schematics of the apparatus. We loaded deuterium gas to Pd film through coating layers while pumping on other side, and then heating the Pd film. Thus deuterium flux formed in Pd had to go through multi-coating layers on the surface of Pd first. By the analysis of the experimental data, we can identify relationship between deuterium flux and coating layers on Pd film.

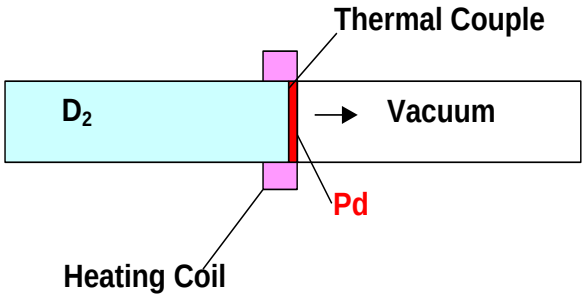

Fig. 1 – Schematics of the deuterium flux apparatus.

The following plots show some results of observation. Under the same conditions, the Pd film with

coating layers (Pd-TiC-Pd) has greater deuterium flux than Pd film without coating layers has unexpectedly.
Fig. 2 and Fig. 3 show a set of comparative results of the experiment. These two experiments have almost the same gas-loading procedure, and the only difference between these two experiments is the sample of Pd film. The first sample is a Pd film (ϕ20mm × 0.1mm) without coating layer, while the second one was coated with a layer of titanium carbide (Pd / TiC). The substrate is a Pd film of ϕ20mm × 0.1mm, while the Pd / TiC coating layer is 20/2nm. In two experiments, we were first using mechanical pumps to evacuate both sides of the Pd film, and then turned off the pumps. After that we loaded deuterium gas into Pd film from the side with coating layer. At that time deuterium gas pressure reached 80kPa, right in the measuring range of pressure gauge. We heated Pd films in a stepwise way, (changing the heating power every 80 minutes, such that the temperature rose to 120°C thermal equilibrium first; then to 200°C thermal equilibrium, and then dropped back to 120°C thermal equilibrium and room temperature thermal equilibrium)(see temperature curves in Fig. 2 and Fig. 3).

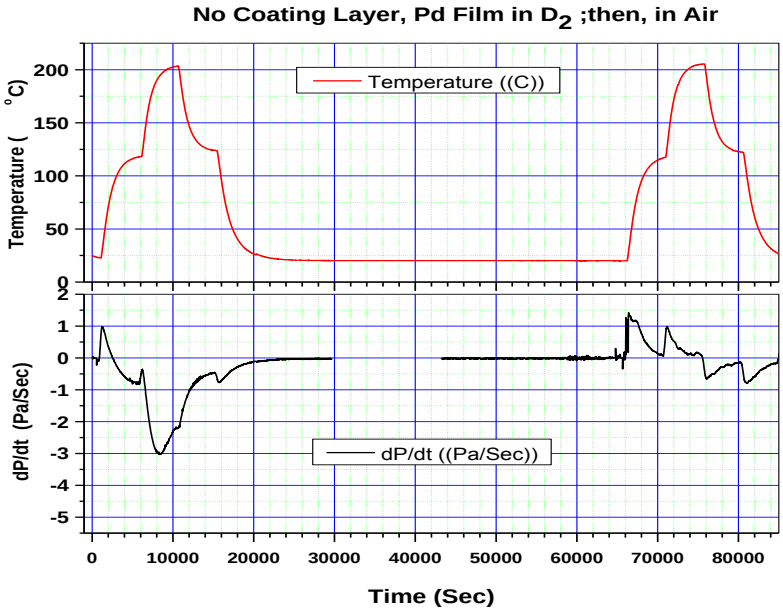

Fig. 2 – Pd film without coating layer.

At the same time we used the multi-channel data acquisition system (Keithley 2700) to record the pressure data. In order to analyze data of pressure change, we calculate deuterium gas chamber pressure derivative to get pressure change rate. The negative peak of pressure change rate represents the rapid decrease of deuterium gas pressure (lower plots in Fig. 2 and Fig. 3 for dP/dt). After deducting the pressure changes caused by temperature, we use the curve to watch deuterium flux.
In order to compare with loading deuterium gas, we loaded air instead of deuterium gas with the same procedure. After heating and cooling in air, we also got temperature and pressure change rate in air as well (Fig. 2 and Fig. 3).
On both figures data after 45,000 seconds are loading air procedure. The positive and negative peaks of dP/dt curves indicate pressure change rate accompanied with temperature changes. We can see from the loading air part, every time Pd film temperature changes, there will be a correspond peak on the pressure change rate curve.
The heating and cooling corresponds separately to the positive peak and negative peak, regardless of the Pd film with or without coating layer. What's more, at the loading air part the positive peak shape of pressure change rate curve is almost the same as the negative peak shape, and peaks height are roughly equal. This shows the air inside chamber acts as the ideal gas isochoric change following the gas state equation. The air dos not permeate through the Pd film. However, loading deuterium gas procedure is completely different. After thermal equilibrium pressure change rate is larger than that of loading air, and positive and negative peak are not asymmetric. A wider and higher negative peak indicates more reduction of deuterium gas pressure. Deuterium flux appears. The puzzle is that Pd film with coating might have much more deuterium flux than Pd film without coating at the same temperature, although Pd film with one coating layer is thicker than Pd film without coating layer. This phenomenon conflicts with classical diffusion theory. (In Fig. 2 the peak of deuterium flux reaches -3.0Pa/s, while in Fig. 3 it reaches

-5.0Pa/s).

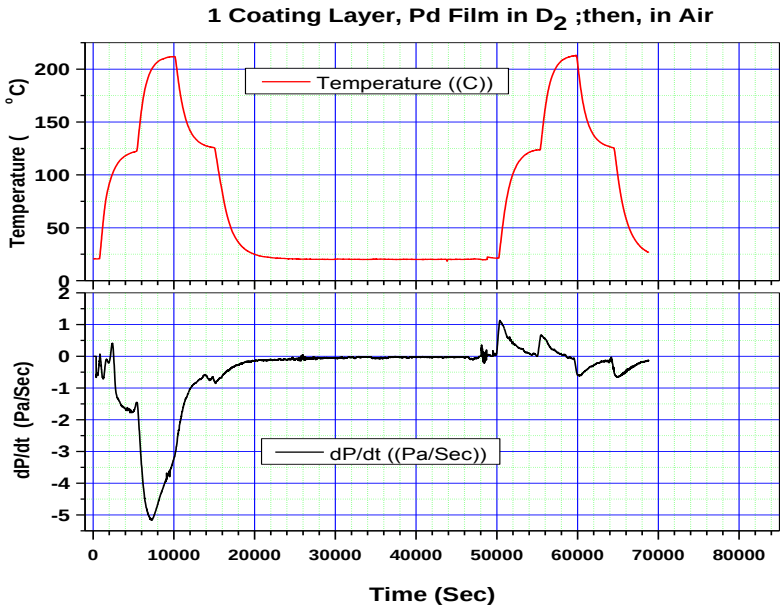

Fig. 3 – Pd film with coating layer.

Further more, we use different numbers of coating layer Pd film to run the same loading procedures. The results show the deuterium flux with different coating layer numbers is not monotonic. Fig. 4 indicated that at the different number of Pd/TiC layers on Pd film with the same experimental conditions, the peak value of deuterium flux changes. All the experiments run with the same deuterium gas pressure (80kPa), and the substrate of Pd film was ϕ20mm × 0.1mm, each Pd/TiC layer thickness was 20/2nm. We repeated experimental procedures as abovementioned experiments. Different coating layer number Pd film reaches the peak deuterium flux at slightly different temperature. We found that the deuterium flux through the multilayer films was not monotonic with the number of coating layers, and there comes the peak in a certain layer.

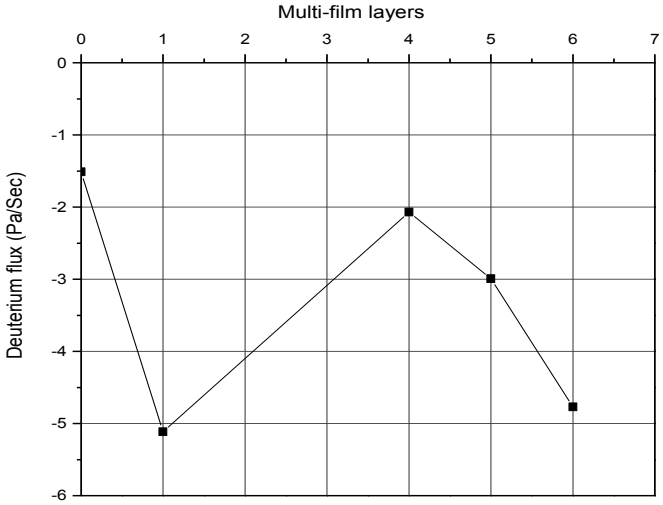

Fig. 4 – Deuterium flux in different coating layer number.

3. Discussion

In 1858, reference to the Fourier thermal equation in 1822, Fick established the quantitative formula to describe the matter migration from high-density area to low-density areas. [4] With low concentration, during unit time and through unit area, material quality (flux) determined by the concentration gradient:

$$\vec{J} = -D\vec{\nabla}n \quad (1)$$

Here D is constant coefficient, called the diffusion coefficient. The concentration n can be expressed as the number of diffusion atoms per unit volume, or other physical quantity represents amount of material like gram atoms. (diffusion flux also takes the appropriate units) Equation (1) is often called as Fick's first law. Equation (1) shows that, keeping the Pd film with the same pressure, the diffusion of deuterium flux through the Pd film usually monotonously decreases when thickness of Pd film increases.

The work of this experiment shows that, accompanied with the variation of the number of coating layers The peak of deuterium flux appears. That is to say, the deuterium flux reaches peak value at the certain number of coating layers. According to the wave nature of deuteron motion, although individual reflected waves on each scattering body is not zero, the summation of reflected waves may be smaller than that of single scattering body. In another words, with the appropriate number of coating layers the different reflected waves might have destructive-interference which would lead to a very small reflectivity. Deuterium flux peak would occur at this condition.

Based on above discussion, permeating characteristics of Nano-system materials can not be fully explained by traditional diffusion theory. The wave nature of deuteron motion can qualitatively describe some anomalies in D-Pd/TiC/Pd multi-layer systems.

Acknowledgments

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

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