

D₂ release process from deuterated palladium in a vacuum

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Abstract

To enhance reproducibility of the phenomena taking place in deuterated palladium, we studied in detail the change in surface temperature, electrical resistance, and D₂ pressure during the release of D₂ from deuterated palladium in a vacuum. As a result, we categorized the temperature changes into three different types that were independent of coating materials. In almost all experiments, the resistance decreased and the D₂ pressure initially increased briefly and then gradually decreased in the D₂ release process. We also tried to simulate the temperature changes by calculating the balance between Joule heat and heat dispersion.

1. Introduction

Nuclear products of d-d reactions, such as ⁴He, and an increased temperature have been observed when D₂ is released from deuterated palladium by the vacuum method¹⁾. To make these phenomena more reproducible and to make the reaction mechanism clear, we studied in detail the change in surface temperature, electrical resistance, and D₂ pressure during the D₂ gas loading into palladium, as well as the release of D₂ from deuterated palladium in a vacuum. Measurement of electric resistance during loading and release of D₂ was confirmed to be useful in monitoring the loading ratio. We measured the changes in surface temperature, electrical resistance and D₂ pressure during the D₂ release process and found that the temperature changes could be categorized into three different types. We also tried to simulate the changes in surface temperature by calculating the balance between Joule heat and heat dispersion such as enthalpy of D₂ release, radiant heat, and heat conduction.

2. Experiment

Figure 1 shows the samples preparation process. Samples were 30 x 30 x 1mm Pd plates. They were slightly etched by aqua regia and preheated at 400°C for 2 hr in a vacuum before being loaded with D₂ gas in a chamber at room temperature or an elevated temperature. The loading ratio (D/Pd) was 0.60 - 0.65. One side of each deuterated plate was coated with manganese oxide or silicon nitride, and the other side was coated with a metal film (Au or Ag). The sample was then placed in a vacuum chamber and heated by injecting currents while the chamber was evacuated by a turbo-molecular pump. Sample temperature, electrical resistance, and D₂ pressure were measured by thermocouples, the four-point probe method, and a vacuum gauge, respectively, as shown in Fig. 2.

3. Results and discussion

Availability of electrical resistance measurement

Figure 3 shows typical curves of electrical resistance and D₂ pressure during loading to Pd. Electrical resistance increases as D₂ pressure decreases during loading, and reaches a constant value after loading. The end of loading process can be determined by measuring electrical resistance. Figure 4 shows the dependence of the electrical resistance ratio (R/R₀) on the loading ratio (D/Pd) calculated from variations of D₂ pressure and Pd weight. As shown in the figure, the R/R₀ increases with an increase of D/Pd, and the

electrical resistance measurement was available to monitor the loading ratio in D₂ loading and release process.

Change in surface temperature, electrical resistance and D₂ pressure

Figures 5 a), b), and c) show electrical resistance and D₂ pressure during D₂ release caused by flowing electric current through the sample in a vacuum. In almost all experiments, after current started to flow, the resistance decreased, while the D₂ pressure initially increased briefly and then gradually decreased in the D₂ release process. However, we found that the temperature changes could be categorized into three different types and are independent of coating materials. In the first type, the temperature increases exponentially with time and reaches a constant value determined by the balance between Joule heating and heat dispersion. In the second type, the temperature rises above the balanced temperature after remaining low for several hours, and the pressure simultaneously falls. This change is similar to that reported before¹⁾. In the third type, there are some peaks in temperature that do not correlate with pressure. After these experiments, almost all samples were found to be bent. We also found that the surface temperature of Pd without loading increases as a simple exponential function of time and reaches a constant value, as shown in Fig. 5 d). This temperature change was the same in almost all samples in reheating by flow of electric currents after D₂ release.

Simulation of surface temperature changing

To find an explanation for the temperature changes, we tried to simulate the surface temperature change phenomenon by heat balance between Joule heat, enthalpy of D₂ release, radiant heat, and conduction of heat. The heat balance in this case is expressed as Eq. (1).

$$Cm(dT/dt) = [\text{Joule heat}] - [\text{radiant heat}] - [\text{conduction of heat}] - [\text{enthalpy of D}_2 \text{ release}] \\ = I^2 [R_0 \{1+a(T-T_0)\} (1+x) + R_s] - \varepsilon \sigma (T^4 - T_n^4) S - k_0 (1+10^{-3}) (T - T_n) \\ - c \Delta H_d (x / t_b) \exp(-E_a / k_B T) \quad (1)$$

C: specific heat, m: weight of Pd, R₀: resistance at temperature T₀, a: temperature coefficient of resistance, R_s: series resistance in the circuit, ε: emissivity, σ: Stephan-Boltzmann constant, T_n: chamber temperature, S: surface area of Pd, k₀(1+10⁻³): temperature dependence thermal conductivity, ΔH_d: enthalpy of release, x: loading ratio (=x₀ - ∫ x dt), t_b: gas release time, c exp(-E_a/k_BT): temperature dependence of release process (c=1.48x10⁵, E_a=0.434 eV), k_B: Boltzmann constant.

Numerical solution using the Runge-Kutta-Gill method was used to simulate the temperature change. Figure 6 shows the typical results of simulation for changes of temperature, loading ratio (D/Pd), and electrical resistance ratio (R/R₀) under the condition of variable initial D/Pd and enthalpy of release (ΔH_d). It is found that the temperature changes depend on initial loading ratio (D/Pd), although reached temperatures determined by the heat balance are same, as shown in Fig. 6 a). In the case of Pd without loading (D/Pd = 0.0), the temperature change of Pd can be simulated, although the constant value that was calculated was different from the experimental result (Fig. 5 d). However, reproducing other temperature changes of deuterated Pd was difficult even though the D₂ release process was added to the experiments. We cannot explain these temperature variations. To make an accurate simulation, we must study the behavior of D₂ on the surface and measure the contact resistance change between the Pd and electric current probes in bending Pd. In addition to the above experiments, the same experiments should be performed for H₂ and the results compared with those of D₂.

4. Conclusion

We studied in detail the changes in surface temperature, electrical resistance, and D_2 pressure during the D_2 gas loading and D_2 release processes. As a result, we confirmed that the electrical resistance measurement can be used to monitor the loading ratio in D_2 loading and release processes. We also found that the electrical resistance and D_2 pressure initially increased briefly and then decreased gradually in D_2 release process in almost all experiments, while the temperature changes could be categorized into three different types and were independent of coating materials. We also tried to simulate the surface temperature changes by calculation of heat balance between Joule heat and heat dispersion. We concluded that the surface temperature change of Pd without loading could be simulated, but other temperature changes were difficult to simulate. We found that to explain temperature changes, the contact resistance change between Pd and electric current probes must be measured in bending Pd and the results of experiments for H_2 should be compared with those of D_2 . We retained measurements of nuclear reaction products during the release of D_2 for future studies.

Reference

1) E. Yamaguchi and T. Nishioka, "Direct Evidence for Nuclear Fusion Reactions in Deuterated Palladium," *Proceedings of the Third International Conference on Cold Fusion*, Nagoya, October 21-25, 1992, p. 179, Universal Academy Press, Inc. (1993).

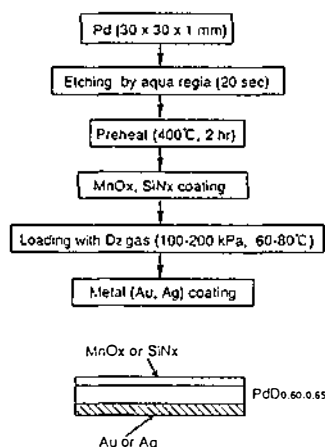


Fig. 1 Preparation of samples

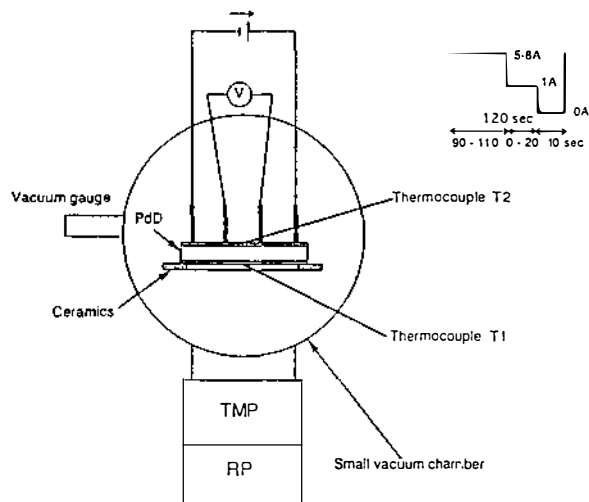


Fig. 2 Measurement system

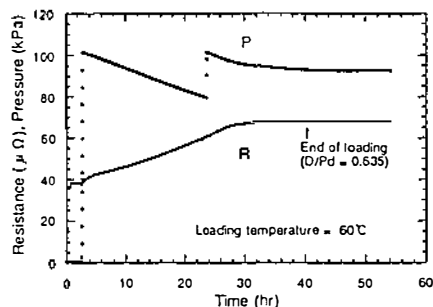


Fig. 3 Typical curves of electric resistance and D_2 pressure in loading to Pd

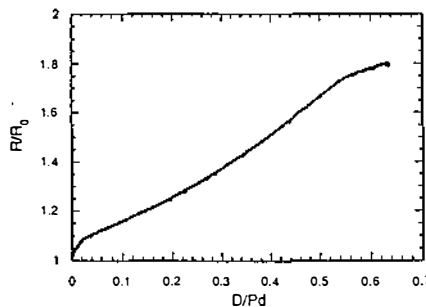


Fig. 4 D/Pd vs. R/R_0

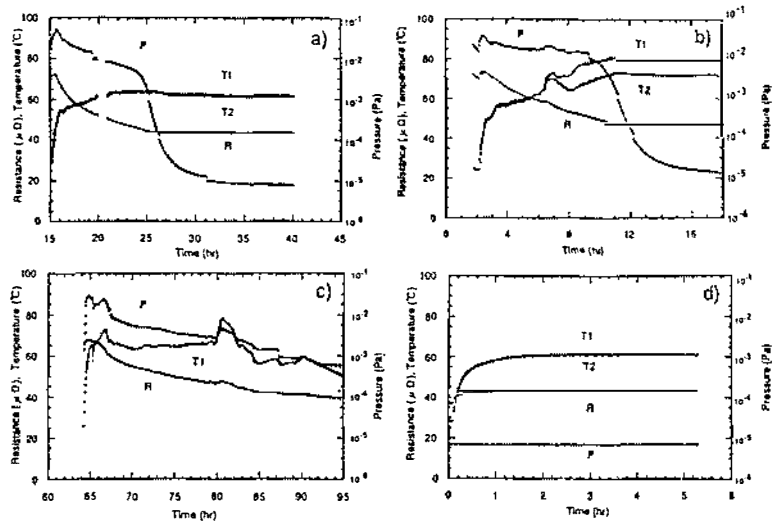
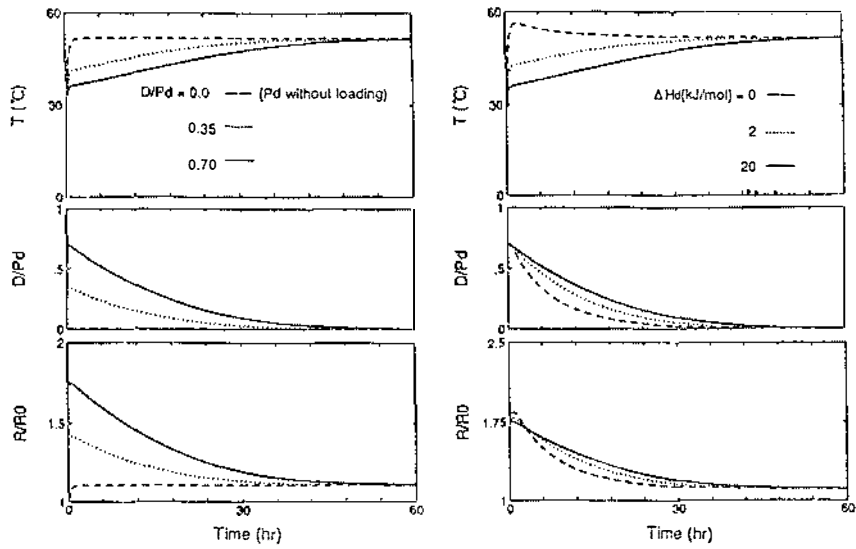


Fig. 5 Typical curves of the surface temperature, electrical resistance and D2 pressure during D2 release process in a vacuum.
 (a) Type 1 (Sample: Pd), (b) Type 2 (Au/Pd/Au),
 (c) Type 3 (MnOx/Pd/Ag), (d) Pd without loading.



a) Dependence of temperature change on initial loading ratio (ΔH_d is 20 kJ/mol)
 b) Dependence of temperature change on ΔH_d (Initial loading ratio is 0.70)

Fig. 6 Results of simulation by electric current flow at 7A