



# Tritium aging effects on separation factors in palladium-hydrogen system



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## ABSTRACT

The separation factors of the palladium-hydrogen system were experimentally measured at different temperatures ranging from 248 to 303 K after aging for up to 3.5 years by tritium. The results showed a remarkable hydrogen isotope effect in the aged palladium-hydrogen system compared to the raw system. The hydrogen-deuterium separation factor of the palladium-hydrogen system aged by tritium was correlated to temperature and hydrogen/deuterium ratio in the solid phase of palladium hydride.

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## 1. Introduction

Palladium is widely employed in the storage, extraction, purification, and separation of hydrogen isotopes because of the resistance to oxidation and poisoning, fast kinetics of absorption and desorption, and the ability to retain helium generated by tritium decay for more than 16 years [1,2].

The aging effects of palladium by tritium have been reported by many research groups, focusing on helium distribution in the palladium matrix [3–7], helium release phenomena [2,8,9], thermodynamics and kinetics of the palladium-tritium system during tritium storage [10–12], and the changes of the crystal structure and mechanical properties of palladium [13–15]. The results confirmed that the aging effects of palladium by tritium can be attributed to the helium generated from tritium decay, especially in the formation of helium bubbles.

The aging effects of palladium by tritium are also expected to have an influence on the separation factors of hydrogen isotopes, which describe the equilibrium characteristics of mixed hydrogen isotopes in metals. Although the separation factors of hydrogen isotopes have been experimentally and theoretically studied [16–24], they have not been systemically investigated for the

palladium-hydrogen system, aged by tritium, due to radiation.

In this work, the hydrogen-deuterium separation factors ( $\alpha_{H-D}$ ) in palladium-hydrogen system after aging for up to 3.5 years by tritium were experimentally measured between 248 and 303 K. The effects of temperature and hydrogen-deuterium (H-D) ratio in the solid phase of palladium hydride on  $\alpha_{H-D}$  were analysed and discussed.

## 2. Experimental

The size of palladium powder used in this study varied between 40 and 60 mesh. The morphology of powder showed an aggregate formed by crystallites (Fig. 1). The specific surface area of the powder was measured by the BET method as 0.438 m<sup>2</sup>/g. The purity of palladium powder was >99.95% (impurities: Pt ~ 0.005%; Rh < 0.001%; Ir < 0.001%; Au < 0.001%; Ag ~ 0.0006%; Cu ~ 0.0006%; Al < 0.001%; Ni < 0.001%; Fe < 0.001%; Si < 0.001%). 12.0 g palladium powder was placed in a tritide bed, which was activated by four cycles of absorption-desorption using deuterium with a purity of >99.97% at 550 K before tritium loading. The tritide bed was designed with the internal dimensions of  $\Phi 12$  mm  $\times$  78 mm; wall thickness: 2.5 mm.

The sample was loaded with an amount of tritium to T/Pd ratio of ~0.645; the purity of tritium was 98.3%. The sample had been stored at room temperature for 3.5 years. The <sup>3</sup>He content in the Pd

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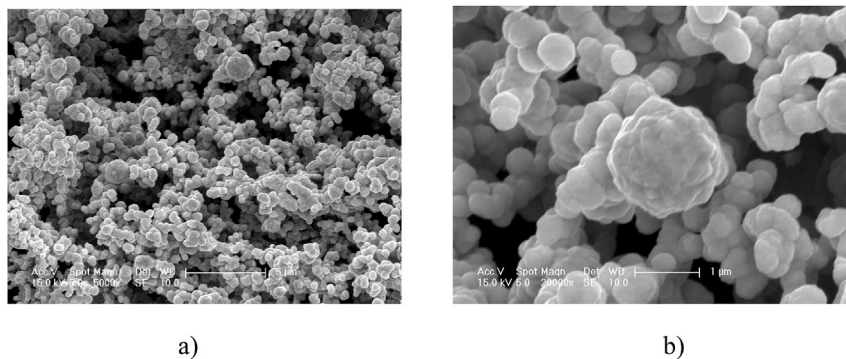


Fig. 1. SEM of palladium powder. a) 5000 × (testing scale: 5 mm); b) 20000 × (testing scale: 1 mm).

matrix can be estimated from the initial tritium stoichiometry and aging time by applying the radioactive decay law, i.e.  $\text{He}/\text{Pd} \approx 0.115$ . Before the measurements of  $\alpha_{\text{H-D}}$ , tritium was desorbed from the aged tritide bed at 523 K, which is lower than the  $^3\text{He}$  desorption temperature and has no obvious effect on major properties in aged palladium powder [2,25]. The fresh palladium sample was prepared as the control experiment.

The experimental system of  $\alpha_{\text{H-D}}$  measurement is shown in Fig. 2. The main experimental error was due to the stoichiometric ratio in preparing the mixture of protium and deuterium using the PVT method, in which the pressure error was 0.005 kPa for the 25 kPa PMA manometer (Philips, B1), and 0.5 kPa for 2.5 MPa PMA manometer (Philips, B2). The temperature error was  $\pm 0.1$  K for Pt100 (E1, E2). The hydrogen isotope abundance error was 0.1% for GAM400 (IPI).

The initial H-D mixture was prepared using the PVT method; the H-D ratios in the standard pressure vessel (213.8 ml) were 1:9, 3:7, 5:5, 7:3, and 9:1. Then, the H-D mixtures were inlet into the aged (unaged) palladium bed up to an atomic ratio of (HD)/Pd of 0.3. After thermodynamic homogenisation of palladium hydride,  $\alpha_{\text{H-D}}$  was measured by analysing the exhausted mixture gas from the palladium hydride bed using a quadrupole mass spectrometer at 248 K, 258 K, 268 K, 278 K, 288 K, and 303 K.

The valve A10 was always open during the measurement of  $\alpha_{\text{H-D}}$ ,

while the valves A1–A3, A5–A6, A8–A9 were closed. A4 and A7 were opened after vacuumising the interrelated parts in the experimental system.

The steps of  $\alpha_{\text{H-D}}$  measurement are as follows: 1) close A8; 2) increase temperature of the isothermobath to 248 K, and keep the temperature of the isothermobath in balance; 3) vacuumise the operating system and interrelated parts, confirming that A1–A2, A5–A6, A9 are closed; 4) open A8, A3, A4, A7, and exhaust the H-D mixture from the palladium bed into the experimental system; record corresponding equilibrium pressure, which is regarded as the desorption pressure; 5) close A8, open A1, exhaust the H-D mixture (sample gas) from the experimental system into GAM400 for analysis; and finally 6) repeat steps 3–5 in order to reduce the experimental error at the same temperature.

By increasing the temperature of the isothermobath,  $\alpha_{\text{H-D}}$  values at 248 K, 258 K, 268 K, 278 K, 288 K, and 303 K were measured. Fig. 3 shows one of the experimental recordings of  $\alpha_{\text{H-D}}$  measurements, including the values of the temperature and pressure when sampling the H-D mixture from the palladium bed.

The wastage of gaseous mixture of protium and deuterium was 0.016 mmol for each H-D ratio analysed by GAM400, while the total wastage was not more than 0.23 mmol for each measurement cycle (the initial amount of protium and deuterium was 14.1 mmol in the palladium bed). At low temperatures (248 K), the pressure in the palladium bed was so low that there was insufficient exhausted H-D mixture gas to give a satisfactory result by GAM400; thus, the measurement had to be repeated four times in order to obtain a relatively more precise  $\alpha_{\text{H-D}}$  value.

The isotope separation factor between protium and deuterium is defined as follows:

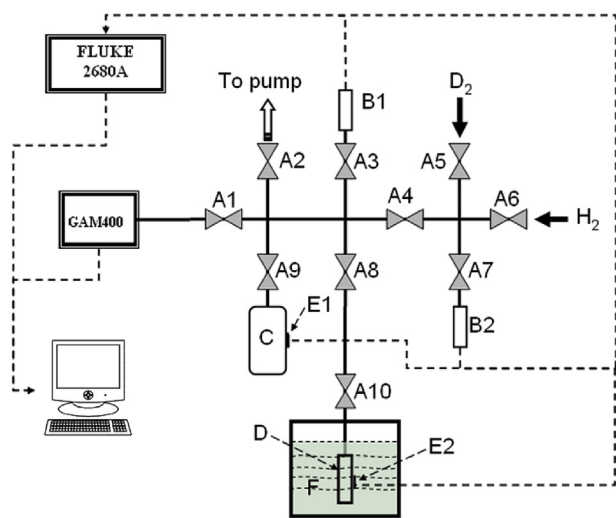


Fig. 2. Schematic diagram of experimental system for  $\alpha_{\text{H-D}}$  measurements. A1–A10 — valves; B1 — 25 kPa manometer; B2 — 2.5 MPa manometer; C — standard pressure vessel; D — palladium bed; E1–E2 — Pt100; F — isothermobath.

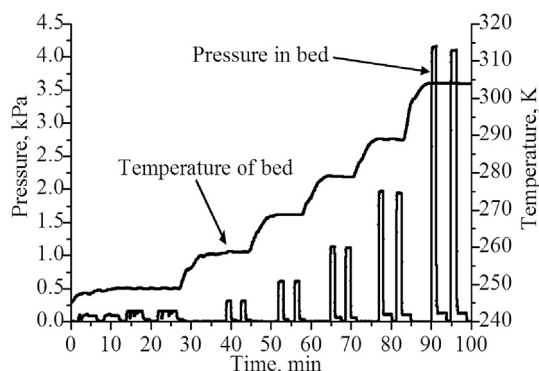


Fig. 3. An experimental recording of  $\alpha_{\text{H-D}}$  measurements.

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