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Observation of Anomalous Heat Release and Helium-4 Production from Highly Deuterated Palladium Fine Particles

Yoshiaki ARATA and Yue-Chang ZHANG

Cooperation Research Center for Advanced Science and Technology, Osaka University, 11-1 Mihogaoka, Ibaraki, Osaka 567-0047, Japan

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Observations were made of the anomalous production of ⁴He atoms as well as the anomalous heat release when Pd fine particles are highly deuterated inside an enclosed Pd metal vessel used as a cathode in electrolysis of D_2O . A mass analysis of the remnant Pd powders after the 2000-hr heat production revealed substantial production of ⁴He atoms.

KEYWORDS: double-structure cathode, deuterated Pd, Pd black, electrolysis, excess energy, ⁴He production

We have invented a double-structure (DS) Pd cathode for electrolysis in order to embed high-density hydrogen ions (protons/deuterons; in this paragraph we use "hydrogen" collectively for both H and D isotopes) in metallic fine powders.^{1–3)} As shown in Fig. 1, it has an outer vessel of Pd metal with a 3 mm-thick wall in which Pd fine powders (Pd black) are enclosed in a high vacuum. When it is used as a cathode in electrolysis in water, the hydrogen ions produced diffuse through the outer Pd wall and penetrate into the inner space, where the Pd powders are contained. After some 100 hrs, the inner pressure increases to more than 1000 bar due to excess hydrogen accumulation, and a high hydrogen concentration in the Pd powders is attained. The characteristic features of the DS-cathode are summarized as follows:³⁾

a) Extremely high pressurization of hydrogen gas inside the vessel in accordance with the Sievertz law⁴⁻⁶⁾ on the relation between hydrogen concentration in a metal ([H/M]) and hydrogen gas pressure (*P*): [H/M] = $a + b \log P$.

b) Extremely pure hydrogen inside the vessel because of selective filter action through the Pd vessel wall.

c) Uniform distribution of high-density hydrogen over the surface of all Pd fine powders through a "spillover effect".⁷⁾

d) Instantaneous absorption of hydrogen in nanoscale proportions with over 100% concentration due to the "atom cluster" function. $^{8)}$

e) Pumping action of Pd fine powders, absorbing the hydrogen which has been emitted from the inner wall of the Pd vessel.

These features cannot be expected in a normal cathode made of bulk Pd metal.

When this DS-cathode was immersed in heavy water for electrolysis, a significant heat release was observed continuously over more than 2000 hours.¹⁻³⁾ In a recent paper,⁹⁾ we reported a further experiment in which two identical DScathodes were immersed, one in normal water (hereafter called DS-H₂O) and the other in heavy water (DS-D₂O). The DS-cathode used had a cylinder type vessel of Pd metal, with an outer diameter of 14 mm, wall thickness of 3 mm and height of 50–70 mm. Fine powders of Pd of about 20 nm diameter with a total weight of 3 g were enclosed in a vacuum inside the vessel. A common electrolytic current was applied to the two electrodes which were connected to an electric circuit in series. The DS-D₂O revealed a continuous excess heat of as much as 5-10 W over 2000 hrs or so, while the DS-H₂O showed none (less than 1 W). Since the total energy release in D₂O amounts to 100 MJ or so, which would correspond to

burning of chemical "fuel" of as much as 100 mol, this phenomenon cannot be ascribed to any kind of chemical reaction. Thus, we searched for remnant atoms remaining inside the Pd powders after these electrolytic exposures. Here, we report the results of our element analyses, in particular, the comparison between the Pd powders remaining in the DS-D₂O and those in the DS-H₂O.

For the present element analysis, the DS-vessels were opened and a small fraction of the Pd powders (about 20 mg) were taken out of each DS-vessel and enclosed in a vacuum sample vessel. Two types of quadrupole mass spectrometers (QMS) were connected to this chamber. One (QMS-A) was for scanning masses and recording the time evolution of their intensities. The other (QMS-B) was used exclusively for high-resolution analysis of M = 4 subcomponents, in which D_2 and ⁴He can be well separated. The total volume of the vacuum space was 51. After evacuation over 10 hours to a level of 10^{-9} Torr, the chamber was closed and only a Titanium Getter pump was kept operating. Then, the sample was heated to 1300°C to release trapped molecules into the chamber. All the molecules except for inert gases were quickly adsorbed in the Getter. Since the sample was exposed once to air, we paid special attention to possible contamination by a small amount of ⁴He and other inert gases from the air.

Figure 2 presents time evolution charts of QMS-A for the most relevant mass regions (D_2 +⁴He, ²²Ne and ⁴⁰Ar). The timing of the closure of the chamber and the heating conditions (timing and current) are indicated. Figure 2(a) (without sample) shows no detectable inert gases except for ⁴⁰Ar, confirming that the sample chamber and the heating device were clean enough to make reliable QMS mass analyses. Calibration was performed by introducing a known amount of air $(1.1 \text{ cm}^3 \times 10^{-2} \text{ Torr})$ into the chamber (reaching 1.3×10^{-6} Torr), revealing ⁴He, ²²Ne and ⁴⁰Ar, as expected (Fig. 2(b)). Figure 2(c) presents the case of 20 mg of Pd powders taken from DS-H₂O, in which no detectable amount of D_2 (+⁴He) and ²²Ne is observed except for a small decaying component of D₂ immediately after heating. Finally, the time chart in the case of DS-D₂O (Fig. 2(d)) reveals a distinct component of D_2+^4He , which decays slowly and reaches a constant level. There is no detectable ²²Ne, indicating that there should be no detectable ⁴He contamination either. To search for any genuine ⁴He in the remaining M = 4component, we used QMS-B, which has a high enough resolution to resolve ⁴He (M = 4.0026) and D₂ (M = 4.0282). Coupled spectra of D_2 +⁴He were recorded every 10s in the scanning range of M = 3.95 - 4.05.



Fig. 1. (Left) Schematic view of the electrolysis apparatus with a DS-cathode. The DS-cathode is surrounded by a cylindrical Pt anode. The electrolyte (water+0.1 M LiOH) is continuously cooled by chilled water flowing through a cooling coil. (Right) Structure of the DS-cathode used for the present experiments. Fine particles of Pd (Pd black) of about 20 nm diameter are enclosed in a vacuum in a Pd metal vessel of 3 mm wall thickness, 14 mm outer diameter and 50–70 mm length.



Fig. 2. Time charts of the intensities of D_2 +⁴He, ²²Ne and ⁴⁰Ar measured by QMS-A. A) without sample, B) calibration with air (1.1 cm³×10⁻² Torr), C) 20 mg Pd powders from DS-H₂O and D) 20 mg Pd powders from DS-D₂O. The timing of the closure of the chamber and the heating conditions (timing and current) are shown.

Figure 3 shows such M = 4 coupled spectra for both DS-H₂O and DS-D₂O. There is a distinct difference between DS-H₂O and DS-D₂O. In the latter case, as time passes, the large D₂ peak decreases, and in turn, the ⁴He peak becomes relatively pronounced. The assignment of the peak to ⁴He was

confirmed by varying the ionization voltage. About one hour after the start of heating, both the D_2 and ⁴He peaks became nearly comparable in intensity. On the other hand, in the case of DS-H₂O, there is neither D_2 nor ⁴He at a detectable level. Thus, this comparative experiment demonstrated clearly that



Fig. 3. High-resolution M = 4 spectra of 20 mg of the remnant Pd powders after 2000-h electrolysis in the DS-H₂O (upper) and in the DS-D₂O (lower). Examples taken at typical elapsed times (1, 30 and 60 min) during and after heating the sample. The scanning range (M = 3.95–4.05) repeated every 10 s is shown, in which well separated D₂ and ⁴He peaks are revealed. The vertical unit scale (1×10^{-12} A) corresponds roughly to 10^{-8} cm³ ⁴He @STP. The vertical axis for the right-hand spectra is enlarged by five times compared with that for the left-hand spectra.

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the ⁴He component arose genuinely from the Pd powders of DS-D₂O. The amount of ⁴He detected for the 20-mg sample was of the order of 10^{-8} cm³ at Standard Temperature and Pressure. In the present experiment, however, we did not capture the remnant gas inside the DS-vessel; therefore, at the present stage, we have no quantitative information on the total amount of ⁴He atoms produced. Further experiments are in progress.

In conclusion, we have found clear evidence of substantial production of ⁴He atoms in addition to the excess heat from highly deuterized fine powders of Pd, which were attained by using only the DS-cathode immersed in D_2O . In contrast, there was no such evidence in the DS-cathode in H_2O . The present observation appears very unusual. The phenomenon must have occurred in such an extreme environment of highly deuterized Pd fine particles as realized for the first time in the DS-cathode. The present results indicate reactions different from the conventional nuclear reactions d+d going to ³He+n and t+p.

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