



Research Article

A Change of Tritium Content in D₂O Solutions during Pd/D Co-deposition

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Abstract

In this study electrochemical co-deposition of Pd/D on nickel electrodes was performed to determine whether a nuclear fusion reaction occurs in the palladium deposit. Co-deposition was performed with a palladium salt/D₂O solution. The content of tritium in D₂O solution was varied depending on the electrolysis procedure during co-deposition. A comparison between the co-deposition of Pd/D and the simple electrolysis of D₂O was performed to investigate the change of tritium concentration in the D₂O solution. © 2014 ISCMNS. All rights reserved. ISSN 2227-3123

Keywords: Electrolysis, Palladium co-deposition, Tritium

1. Introduction

An early report on experiments with the Pd/D system suggested that fusion of deuterium atoms occurs within the Pd-lattice when the system is under prolonged cathode polarization [1]. Among the observed results attributed to nuclear events, a significant amount of tritium was found. Questions were raised concerning the origin of the tritium [2–4,10].

In this study, the electrochemical co-deposition of Pd/D on nickel electrodes was performed to determine whether a nuclear fusion reaction occurred in palladium metals that were co-deposited from a palladium salt/D₂O solution [6–9]. The content of tritium (³T) in the D₂O solution was varied depending on the electrolysis procedure of Pd/D co-deposition. A comparison between the co-deposition of Pd/D and the simple electrolysis of D₂O with a Pd foil cathode was performed to investigate the change of tritium concentration in the D₂O solution.

2. Experimental

Two types of palladium electrodes on porous Ni cathodes of 3 mm O.D. were prepared by electrodeposition in the cell design shown in Fig. 1 [5]. One type is PdCl₂–LiCl–D₂O solution and the other type is Pd(en)Cl₂(palladium

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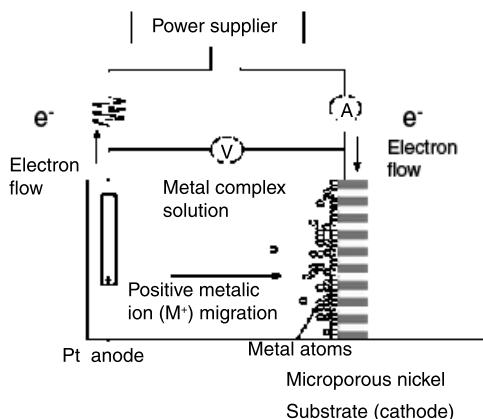


Figure 1. Electrolytic co-deposition cells.

ethylenediamine chloride)–LiCl–D₂O solution. Palladium foil electrodes (7 × 20 mm) were subjected to D₂ evolution from D₂O + 0.3 M LiCl electrolysis. The co-deposition experiments were carried out in a closed system with a recombination catalyst. The resulting solutions were examined for tritium. Samples of liquid electrolyte were measured using liquid scintillation counting. In all cells, measurements were made for the activity of ³H in solution. The resulting liquid was analyzed the same way as the electrolyte samples were. Platinum gauze anodes (99.9% pure), used in all cases, were washed and allowed to dry in air.

All of the cells were first run at low-current density. After a few days, current density was increased up to 300 mA. Samples of electrolyte were withdrawn for tritium analysis from the cell. Tritium analysis was performed on the alkaline electrolyte by in situ Liquid Scintillation Counting (LSC). A 1 ml sample of electrolyte is added to 2 ml of Ready safe, Beckman LSC cocktail in a vial. The prepared solutions were counted for 1 min in a Beckman LS 6000 TA scintillation Counter. The results are given as counts per minute (cpm) per ml of samples.

3. Results and Discussion

3.1. Pd/D co-deposition

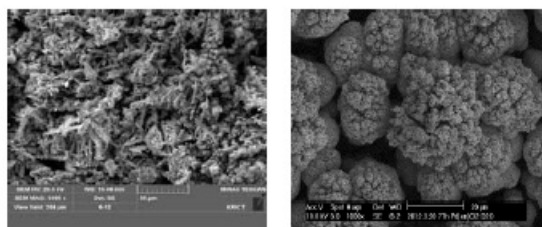
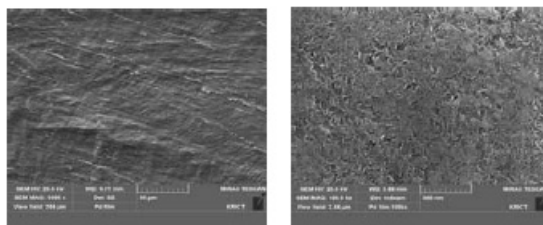
Pd/D co-deposition with two different conditions was performed. One is Pd/D co-deposition with PdCl₂ and the other is Pd(en)Cl₂. The same type cathode (porous Ni) and anode (Pt gauze) was used and the current density conditions are only a little different. Experimental conditions and results are summarized in Table 1. Pd/D co-deposition using PdCl₂ was conducted from 1 up to 300 mA of current density. The tritium level was decreased from the initial level of 596–235 cpm after the reaction. Pd/D co-deposition using Pd(en)Cl₂ was conducted from 1 up to 200 mA and the tritium level was decreased from 489 to 160 cpm. Figure 2 shows SEM images of the surfaces of cathodes after Pd/D co-deposition. This is an unexpected result. The tritium content should have remained constant or increased [11]. These results suggest that tritium was consumed or adsorbed in the electrodes. The surface of a cathode co-deposited with PdCl₂ (Fig. 2(a)) was somewhat rough like branches of acicular shape, while the shape of cathode co-deposited with Pd(en)Cl₂ (Fig. 2(b)) was like aggregates of spherical globules.

Table 1. Change of tritium concentration of Pd/D co-deposition.

Conditions	PdCl ₂ 0.03 M LiCl 0.3 M D ₂ O 100 mL Ammonia water 1.8 mL	Pd(en)Cl ₂ 0.03 M LiCl 0.3 M D ₂ O 100 mL Ammonia water 1.8 mL
Electrodes (cathode/anode)	Porous Ni/ Pt gauze	Porous Ni/ Pt gauze
Current	1–50 mA (4 h)-200 mA (7 h)-300 mA (1 h)-1 mA	1–10 mA (6 h)-50 mA (1 h)-100 mA (1.5 h)-150 mA (1 h)-200 mA (1 h)-1 mA
Time	97 h	77 h
Before	596.00 cpm	489.00 cpm
After	235.00 cpm	160.00 cpm

3.2. D₂O electrolysis

Electrolysis was performed with 0.3 M lithium chloride in D₂O. The cathode was palladium foil, and the anode was platinum gauze. The experimental conditions and results are summarized in Table 2. D₂O electrolysis was conducted from 20 up to 400 mA of current density. As a result, an increase in the tritium level was seen from the initial tritium level of 10.00 up to 167.00 cpm after the reaction. Despite the short duration of electrolysis and the initial low level of tritium, the tritium level increased a great deal. Figure 3 shows SEM images of the surface of the cathode after D₂O electrolysis. Band split vertically appeared on the surface of cathode (Fig. 3(a)). The change of the surface was observed clearly at high magnification. Small defects appeared, along with what looks like small uniform patterns.

**Figure 2.** SEM image of Pd/D co-deposition cathodes. (a) PdCl₂ (b) Pd(en)Cl₂.**Figure 3.** SEM image of D₂O electrolysis cathode. (a) 1000× and (b) 100 000×.

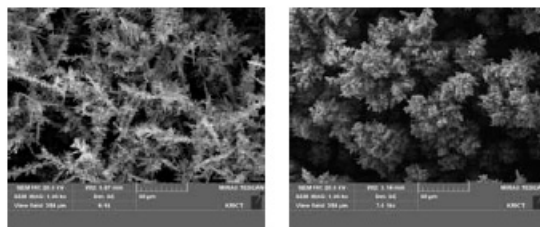


Figure 4. SEM image of Pd/D co-deposition and D₂O electrolysis cathode. (a) PdCl₂ and (b) Pd(en)Cl₂.

3.3. Pd/D co-deposition and D₂O electrolysis

Pd/D co-deposition under two different conditions was performed. One condition is Pd/D co-deposition with PdCl₂ and the other is with Pd(en)Cl₂. The Pd/D co-deposition was conducted at a current of 1 mA for 1 week, and D₂O electrolysis was conducted by increasing current density. The same type of cathode (porous Ni) and anode (Pt gauze) was used and the current density conditions were the same. Experimental conditions and results are shown in Table 3. Pd/D co-deposition with only low current does not change the tritium concentrations. Tritium levels were not changed with either of the Pd/D co-deposition cathodes at a current of 1 mA. But the tritium level after D₂O electrolysis increased greatly. The tritium level using PdCl₂ increased from the initial at 28–314 cpm. The tritium level using Pd(en)Cl₂ increased from the initial level of 30–258 cpm. Figure 4 shows SEM images of the initial of 28–314 cpm after reaction surface of the cathode after Pd/D co-deposition and D₂O electrolysis. The surface of 30 cpm to the final level of 258 cpm the cathode was fine and weak forms compared with Pd/D co-deposition cathodes (Fig. 2). The surface shape of cathode co-deposited with PdCl₂ (Fig. 4(a)) was somewhat rough, with long branches of acicular shape; while the surface shape of the cathode co-deposited with Pd(en)Cl₂ (Fig. 4(b)) was short with sparse shapes.

4. Conclusions

Pd/D co-deposition with only low current did not change the tritium level at the concentrations studied in this work. Contrary to our expectations, Pd/D co-deposition with higher current, up to 300 mA, decreased the tritium concentration of D₂O solutions, when highly tritiated D₂O was used. When D₂O with lower levels of tritium was used, both palladium foil and Pd co-deposited electrodes increased tritium content during D₂O electrolysis

Table 2. Change of tritium concentration of D₂O electrolysis cells

Conditions	LiCl 0.3 M D ₂ O 100 mL
Electrodes (cathode/anode)	Pd foil/ Pt gauze
Current	20–250 mA (2h)-20 mA-200 mA (0.5 h)- 400 mA (0.5 h)
Time	65h
Before	10.00 cpm
After	167.00 cpm

Table 3. Change of Tritium concentrations of Pd/D co-deposition and D₂O electrolysis

Conditions	PdCl ₂ 0.03 M LiCl 0.3 M D ₂ O 100 mL Ammonia water 1.8 mL	Pd(en)Cl ₂ 0.03 M LiCl 0.3 M D ₂ O 100 mL Ammonia water 1.8 mL
Electrodes (cathode/anode)	Porous Ni/ Pt gauze	Porous Ni/ Pt gauze
Current	1 mA	1 mA
Time	168 h	168 h
Before	40.00 cpm	24.00 cpm
After	35.00 cpm	26.00 cpm
Conditions	LiCl 0.3 M D ₂ O 100 mL Porous Ni/ Pt gauze	LiCl 0.3 M D ₂ O 100 mL Porous Ni/ Pt gauze
Electrodes (cathode/anode)	Porous Ni/ Pt gauze	Porous Ni/ Pt gauze
Current	20–250 mA (2 h)-20 mA- 200 mA (0.5 h)-400 mA (0.5 h)	20–250 mA (2 h)-20 mA- 200 mA (0.5 h)-400 mA (0.5 h)
Time	65 h	65 h
Before	28.00 cpm	30.00 cpm
After	314.00 cpm	258.00 cpm

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