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Preparation of Pd-Pt alloy on Alumina for the Hydrogen Isotopes Separation

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1. Introduction

Development of a simple and efficient hydrogen isotope separation technique is important for fusion reactors and tritium handling facilities. For the purpose, cryogenic distillation methods have been good method because of its efficiency and mass treatment. The new gas chromatographic method in which Pd metal were used as column packing material showed high separation efficiency, simple apparatus and moderate operation condition. They work at above room temperature and the retention time is relatively short within several minutes [1-3].

Especially Palladium-platinum (Pd-Pt) alloys have been reported very effective column for hydrogen isotope separation. It is known that small amount of Pt causes to decrease the heat of absorption when hydrogen atoms diffuse in the Pd crystalline lattice [4].

The preparation of palladium-platinum alloys was done by arc melting [4-8] in previous researches. Generally the metal powder itself is not advisable because these precious column materials are more economical to be supported by inert materials like alumina or molecular sieves. In this view point, M. Hara et al. adopted barrel sputtering method to support Pd-Pt alloy on the surface of alumina powder [9]. But this sputtering method needs specially designed barrel sputtering machine and seems to be difficult to achieve high loading of Pd-Pt metal on a supporting material.

Therefore, the preparation of Pd-Pt alloy on alumina support has been studied. And an impregnation method in the presence of PVP was confirmed excellent method for the Pd-Pt alloy on alumina for hydrogen isotope separation.

2. Experiment

Preparation processes of Pd-Pt alloy on alumina through impregnation were disclosed. And the prepared materials were subjected to characterization for hydrogen absorption

2.1 Preparation of Pd-Pt alloy on alumina

The used support material was α-alumina (Aldrich) which has low porosity and good thermal stability, and the particle size was 100 mesh. The procedure is as follows. Some amount of $PdCl_2$ and $PtCl_2$ were dissolved in 400 ml of ethanol water (1/1, w/w) solution and the PVP (MW=10,000) was added. After adding the α-alumina powder to the mixture, the suspension was stirred for enough mixing. The prepared mixture was left alone at roomed temperature without mixing for a day. The liquid components were distilled out in a rotary evaporator, and the residual solid was dried at 120℃ for 30 min and then calcined in air at 400 ℃ for 2 hr for the complete decomposition of PVP. The dried solid was pulverized in a mortar. The powder was placed into quartz tube in tubular furnace and was heated in H_2 flow for the perfect reduction of the metal precursors and metal oxides at 120℃ for 30 min and 300℃ for 2hr successively. Finally, the powder was annealed at 800℃ for 5 hr in Ar flow to enhance the crystallinity of the alloy on α-alumina.

By this manner, three Pd contents were made for the alumina support: 6, 17, and 29% (w/w) which was analyzed by ICP-AES. And the crystallinity was examined by XRD.

2.2 Measurement of Pressure-Composition Curve

To evaluate hydrogen absorbing behavior, the desorption isotherm was measured for the Pd-Pt alloy on α-alumina using a high-vacuum system. Figure 1 shows the system diagram. The main components were a sample vessel equipped with electric furnace, ionization vacuum gauge, turbo-molecular pump, reference volume, thermocouples, gas reservoirs, and etc. Five gram of sample powder (including 0.85 g of Pd metal)

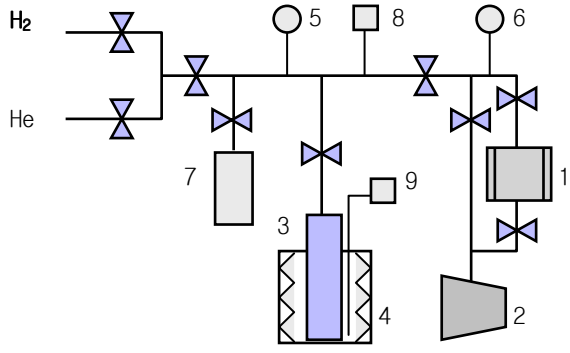

Figure 1. Schematic diagram of high vacuum system measuring desorption isotherms of hydrogen; 1. Turbo-molecular pump, 2. Rotary pump, 3. Sample vessel, 4. Electric furnace, 5. Pressure transducer, 6. Ionization vacuum gauge, 7. Reference volume, 8. Manifold thermocouple. 9. Vessel thermocouple.

System was evacuated first below 10^{-6} torr and then the sample was degassed at 300 ℃ for 2 hr. After the sample was cooled down at room temperature, hydrogen

was introduced into the sample vessel and then the system was degassed 300℃. The sample was activated by repeating hydrogen absorption and Desorption cycles at given temperatures and then the system was evacuated below 10^{-6} torr. After the activation, the sample was cooled down at 80℃ and then hydrogen gas was introduced into sample vessel. To obtain desorption isotherm, the sample was evacuated step by step. The conventional constant volume method was used to obtain the Pressure-Composition curve.

3. Results and Conclusion

Figure 2 shows diffraction patterns of Pt-pt alloy on α-alumina. The patterns consist of the peaks of α-alumina (marked by α) and Pd-Pt alloy, but there is not any other component peak. From this result it is confirmed that Pd-Pt crystalline clusters were formed very well on the surface of α-alumina particles.

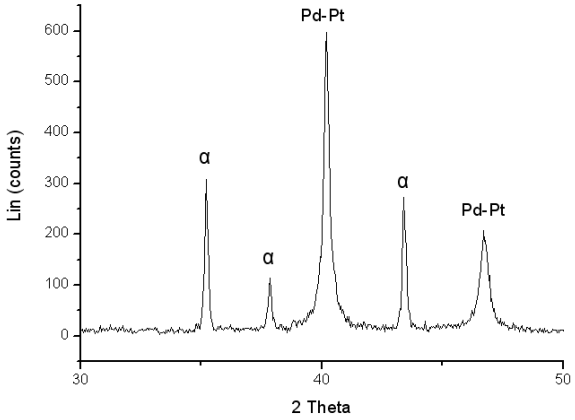

Figure 2. XRD patterns of 17 wt% Pd-Pt alloy on α-alumina.

Figure 3 shows hydrogen desorption isotherms by 17 wt% Pd-Pt alloy on α-alumina. A plateau appeared in a region from [H]/[M]=0.05 to 0.38, and the plateau increase with increasing hydrogen concentration slightly. In comparison with the isotherm for Barrel sputtering Pd-Pt alloy on alumina, the equilibrium pressures is a little lower, but the overall shape of curves is almost the same. Therefore the difference between two equilibrium curves is attributed to the test condition and different sample preparation method.

From these results, Pd-Pt alloy on α-alumina prepared by impregnation method adding PVP has good property for hydrogen absorption and desorption. And the produced alloy can be used as profitable working material in gas chromatographic technique for hydrogen isotope separation.

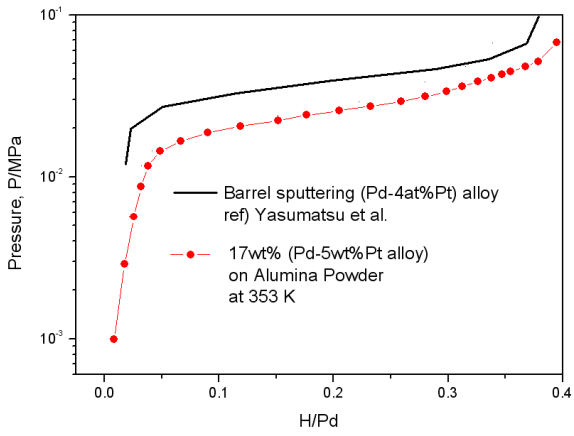

Figure 3. Desorption isotherm of 17 wt% Pd-Pt alloy on α-alumina. ref) T. Yasumatsu et al., J. of Alloys and compounds 293-295 (1999) 900.

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