Preparation of Pd Electrodes and Their Hydrogen Loading Ratios

Toshiyuki SANO, Toshihisa TERASAWA, Tamio OHI, and Shinji NEZU

IMRA MATERIAL R&D CO., LTD.
5-50 Hachiken-cho, Kariya, Aichi 448
JAPAN

ABSTRACT

We prepared a series of palladium-based rods with various compositions and processing histories as cathode for water electrolysis. These rods were evaluated in terms of hydrogen loading ratio (H/Pd). The hydrogen loading ratios of Pd-Ag and Pd-Ce alloys are compared with that of a pure Pd rod. The hydrogen loading ratios of Pd rods subjected to annealing and/or cold-working (swaging) are also compared with that of a cast Pd rod. The results show that the alloying markedly reduces the loading ratio, and neither annealing (350-650 °C) nor swaging (up to the processing ratio of 98 %) produces a distinct effect.

1. Introduction

The lack of reproducibility of excess heat generation in heavy water electrolysis might be ascribed to unknown factors which are closely related to the material nature of the palladium cathode. It has been pointed out that attaining a high loading ratio is the key to give rise to excess heat generation [1]. Correlation between the rate of excess heat generation and the hydrogen loading ratio reported recently [2].

It has been known that the amount of hydrogen absorbed by a Pd electrode polarized cathodically is a function of the hydrogen overpotential [3,4]. On the other hand, the relation between palladium characteristics and the amount of absorbed hydrogen is not well understood.

In this study we prepared a series of palladium-based rods with various compositions and processing histories as cathode for water electrolysis, and measured the hydrogen

FRONTIERS OF COLD FUSION

loading ratios in order to investigate the effect of alloying, cold-working, and annealing.

2. Experimental

2-1. Preparation of electrodes

Palladium grain (99.99 %, 3-5 mm in diameter) was used as a mother alloy. Button-shaped mother alloys of Pd-Ag and Pd-Ce were prepared by arc-melting of the palladium grain and the respective metals (Ag: 99.9 %; Ce: 99.7 %). The mother alloys were cast into rods (8 mm in diameter) by high frequency induction melting (1650 °C) under an argon atmosphere. Using these rods, the following four groups of rods were prepared by annealing and/or swaging when necessary:

(1) Rods cast only.
(2) Cast and subsequently annealed rods.
(3) Cast and subsequently swaged rods.
(4) Cast, then annealed, and subsequently swaged rods.

In swaging, the percentage of a reduction in a cross section (processing ratio) was 75 % a swaging process. The final diameters of swaged rods were 4, 2, or 1 mm. The annealing temperatures were 350, 450 or 650 °C. The samples were held at an annealing temperature for 1 hour, and then cooled to room temperature at the rate of 1 °C/min.

2-2. Measurement of Hydrogen Loading Ratio

The rods (1-8 mm in diameter) were washed by acetone before each measurement using a ultrasonic cleaner, and then electrolyzed in H_2O at 25-30 °C (electrolyte temperature) using a cell shown in Figure 1. It should be noted that an evolved hydrogen gas is separated from an oxygen gas by a separator. A platinum black reference electrode was used to measure the hydrogen overpotential. A platinum rod which had the same surface area as a test rod was used as control electrode. All the measurements reported here were made galvanostatically at the current density of 300 mA/cm^2. The amount of the hydrogen absorbed in a test electrode was determined from the difference in evolved gas volume between the test electrode and the Pt control electrode in a subsequent run. Gas volume measurements were made using a mass flow meter (KOFLOC, Model-3710). Output from the mass flow meter was monitored every 5 seconds and integrated into the amount of hydrogen.

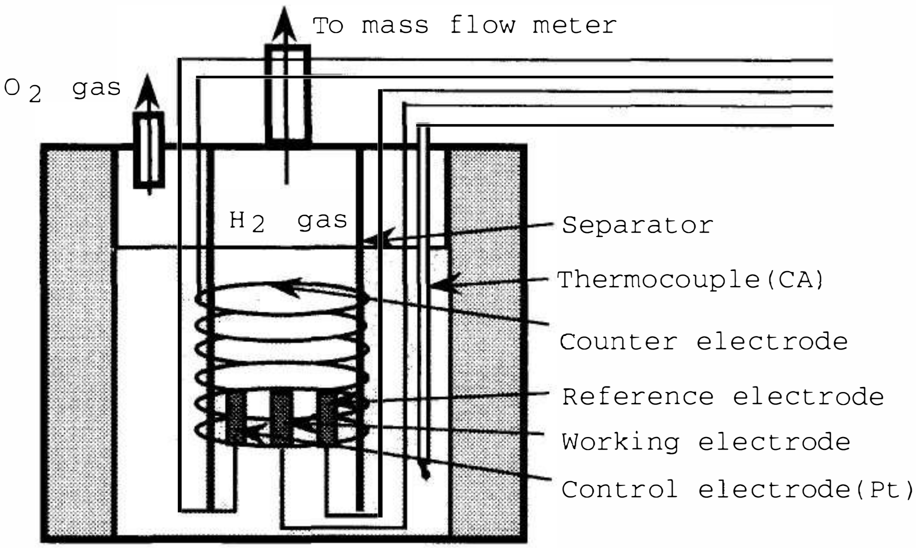

Figure 1. Electrolysis cell used for hydrogen loading ratio measurement.

3. Results and Discussion

Figure 1 shows the hydrogen loading ratios of Pd-based alloys with various Ag or Ce contents. In this experiment, all the electrodes (2 mm in diameter) were prepared by swaging of cast rods (8 mm). As is shown, the loading ratio markedly decreased with the increase of the Ag or Ce content.

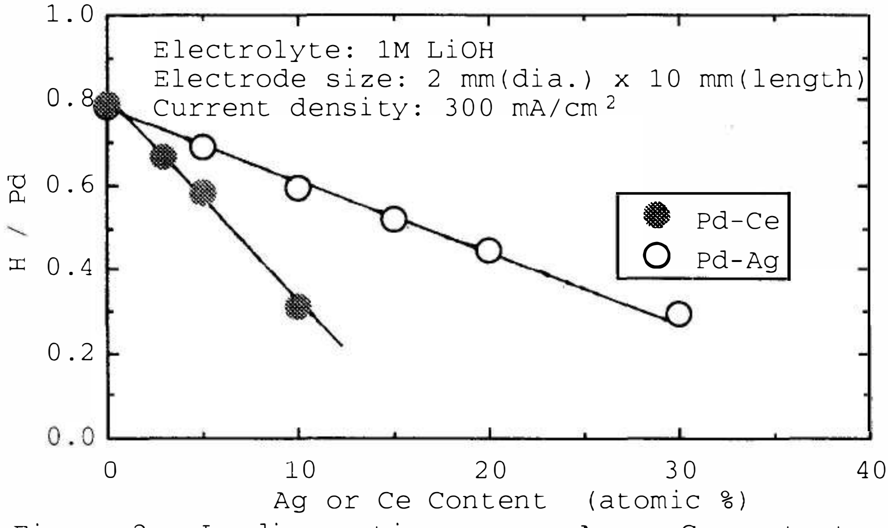

Figure 2. Loading ratios versus Ag or Ce content.

Table 1 shows the hydrogen loading ratios of swaged and/or annealed Pd rods. Although a little variation is seen, a distinct effect of those processing could not be observed under the condition employed in this study. The variation may suggest that other unknown factors are involved in the loading.

Table 1. Hydrogen loading ratios of swaged and/or annealed Pd rods.

Annealing Temp. / °C	Processing Ratio 0 %	75 %	94 %	98%
not annealed	0.776	0.780	0.785	0.767
350	0.746	0.773	0.776	0.778
450	0.782	0.767	0.731	0.759
650	0.774	0.762	0.746	0.748

5. References

(1) M. McKubre et.al.,Conference Proceedings Vol. 33 "The Science of Cold Fusion", p419, SIF, Bologna 1991.
(2) K. Kunimatu et.al.,Proceedings of The Electrochemical Society of Japan 1992 Fall Conference, p77.
(3) T. Maoka and M. Enyo, Surf.Tecnol.,9, 147 (1979).
(4) T. Maoka and M. Enyo, Electrochim. Acta, 26, 607 (1981).