Measurements of D/Pd and Excess Heat during Electrolysis of LiOD in a Fuel-Cell Type Closed Cell Using a Palladium Sheet Cathode

Masafumi Kobayashi, Norio Imai, Norifumi Hasegawa, Akiko Kubota, Keiji Kunimatsu
IMRA JAPAN CO.,LTD.
3-6 Techno Park 2 Chome,Simonopporo,Atsubetsu-ku, Sapporo 004 Japan

ABSTRACT

Measurement of D/Pd and excess heat was carried out during electrolysis of LiOD in a fuel-cell type closed cell using two batches of palladium sheet cathodes. We applied the "saw-tooth" current mode and the following "L-H " current mode which was employed originally by Takahashi. Excess heat of 10-30% of input power was observed in experiment-A using a Pd sheet cathode that was one of the same batch used by Takahashi. But in experiment-B, the palladium sheet that was one of the different batch did not produce any measurable excess power. The saw-tooth mode and the L-H mode operation had no effect to enhance D/Pd.

1. Introduction

Takahashi,Iida,Takeuchi and Mega have reported [1] the production of excess power that exceeded 100 W/cm^3 in an open type electrolytic cell using a Pd sheet cathode. They suggested that the "saw-tooth mode" and the "L-H mode" operation might enhance D/Pd ratio more than 1.0 although measurement of D/Pd was not conducted in their study. We developed a fuel-cell type closed cell and the method of in-situ determination of D/Pd ratio during electrolysis.

The porpose of the present study is firstly to replicate Takahashi-type experiment in the fuel-cell type closed cell and secondly to investigate effect of saw-tooth and the L-H mode operation on the loading ratio. The results of simultanious determination of excess heat and D/Pd are reported.

2. Experimental

Fig.1 shows schematic view of the fuel-cell type closed electrolysis cell. A stainless steel pressure vessel was used with a PTFE cup fitted inside the vessel. Electrolyte was 150 ml 1M LiOD. The vessel filled with about 9 atm.D_2 was submerged in a water bath kept at constant temperature. A cold worked pure Pd plate (1mm thick 25 $\times$ 25mm) from Tanaka Kikinzoku Kogyo K.K. which was one of the same batch used by Takahashi was used in Experiment-A. A second similar Pd sheet which was prepared by the same method but is one of a different batch was used in Experiment-B. The Pd sheet

FRONTIERS OF COLD FUSION

cathode was supported by PTFE retainers as shown in Fig.1. Two sheets of gas diffusion type electrode (anode) were placed parallel to the Pd cathode. The minimum anode-cathode distances was 10mm(or 6mm) for both sides. Loading ratio, D/Pd, was calculated by measuring D_2 gas pressure and temperature. The electrolyte temperature was monitored by two thermocouples covered with PTFE tubes. Thermocouple-1(T-1) was located close to the upper part of Pd cathode. T-2 was placed at the middle point between the cathode(Pd) and anode. The calorimetric determination of excess heat was conducted by comparing the temper9ature rise in the electrolyte observed by Ni and Pd cathode respectively for a given input power assuming there is no excess heat generation for Ni cathode. We started the "pre-loading phase" with the "saw tooth current mode" between 0.25A and 4.0(5.0)A repeated every twenty minutes, which was continued for 7 days. From the 8th day we switched to the L-H current mode operation by changing the current between 0.25A and 4.2(5.0)A every six hours.

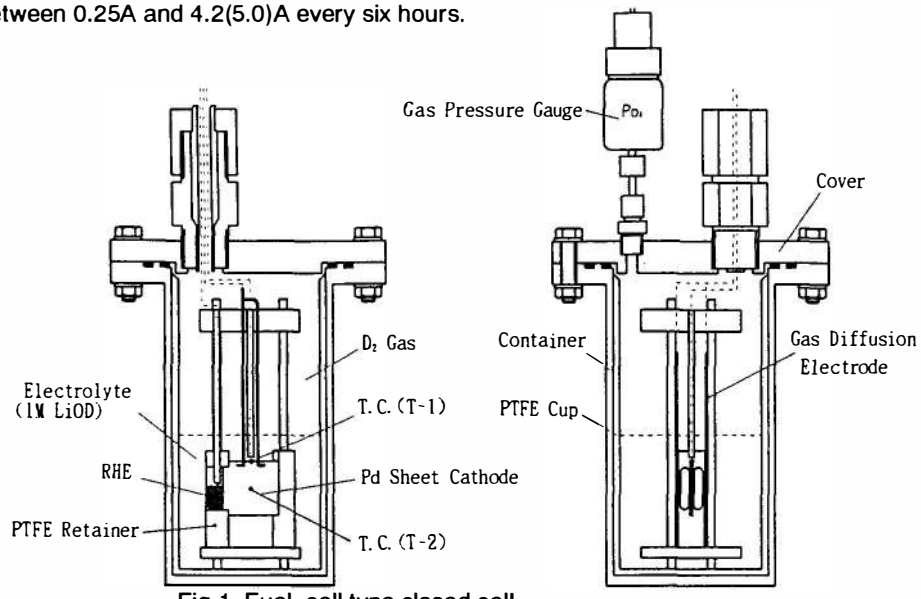

Fig.1 Fuel-cell type closed cell

Experimental conditions are shown as follows.

Table 1. Experimental conditions

	Experiment-A	Experiment-B
cathode material	Pd sheet 1st batch	Pd sheet 2nd batch
electrolyte	1M LiOD 150ml	1M LiOD 150ml
current	0.25A-4A 0.25A-4.2A	0.25A-5A
saw tooth cycle	20 min.	20 min.
saw tooth mode duration	1 week	1 week
L-H cycle	6 hr.	6 hr.
calibration	Ni sheet cathode	Ni sheet cathode
water bath temperature	20°C	20°C

3. Results and Discussion

Fig.2 shows D/Pd as a function of time during the electrolysis (Experiment-A). After

initiation of the saw-tooth mode operation, the loading ratio reached an almost constant value of 0.78 within two hours. The loading ratio remained constant for a week throughout the saw-tooth mode operation. On switching to the L-H mode operation, D/Pd varied between 0.77 and 0.79 during Low and High current mode period respectively. This suggests that the L-H current mode operation caused releasing and re-loading of deuterium. From the 13th day we changed the current of the L-H mode to 2A and 3.5A. And further, we changed the current to 1A and 3A from the 15th day. Then,D/Pd stayed almost constant at about 0.80.

Fig.3 shows solution temperature(T-1) as a function of time during the electrolysis (Experiment-A).The temperature does not reach its steady value during the saw-tooth current mode operation for a given current density due to its too short cycle time, while the electrolyte shows its steady temperature for the given current densities during the L-H current mode operation as shown in Fig.3.

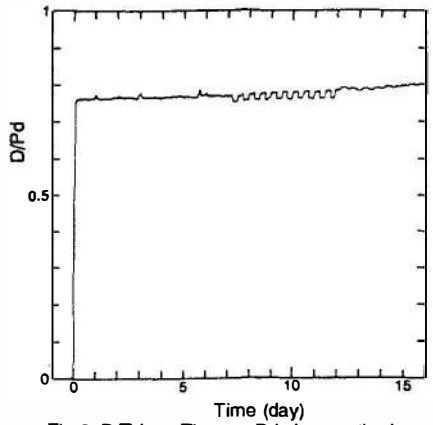

Fig.2 D/Pd vs. Time on Pd sheet cathode in 1M LiOD at 293K (Experiment-A)

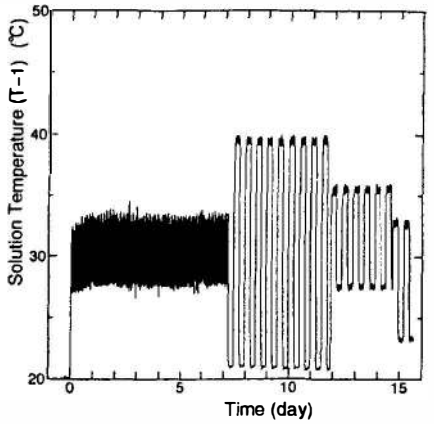

Fig.3 Solution Temperature (T-1) vs.Time on Pd sheet cathode in 1M LiOD at 293K (Experiment-A)

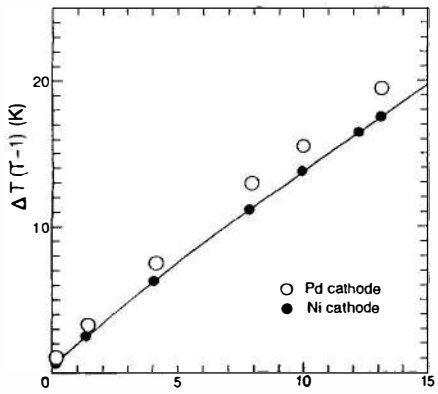

Fig.4 ΔT vs. Input Power on Ni or Pd sheet cathode in 1M LiOD at 293K (Experiment-A)

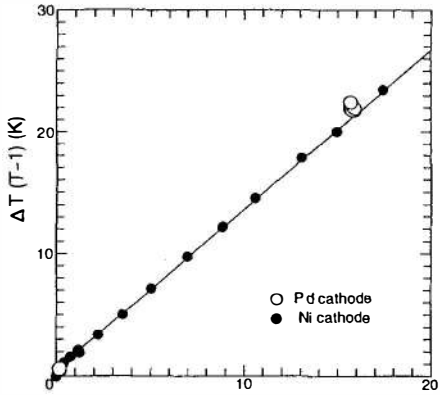

Fig.5 ΔT vs. Input Power on Ni or Pd sheet cathode in 1M LiOD at 293K (Experiment-B)

Fig.4 and Fig.5 show the relation between the change in solution temperature and input power for Experiment-A and B respectively. The relation is compared between the Ni and the Pd cathode . From Fig.6 and Fig.7 we conclude that excess heat of 10-30% with

respect to the input power was observed (Experiment-A), while there was no measurable excess heat for Experiment-B

In view of the low loading ratio around 0.80 in the present study, we further investigated effect of the way of holding the Pd sheet cathode by PTFE retainers on the loading ratio. The retainers hold the Pd cathode at the two edges making the edges of Pd umpolarized during electrolysis. The umpolarized edges may lead to release of deuterium during its loading. Fig.6 shows the H/Pd as a function of overvoltage ofserved in 1M LiOH in the absence and presence of the PTFE retainers. The Pd was hanging in solution by two Pd wires in the absence of the PTFE retainers. The higher H/Pd values were observed without retainers, which suggests that the unpolarised edges of Pd in the PTFE retainers are in fact releasing hydrogen while loading is going on at other parts of the Pd cathode.

We further investigated effect of the saw-tooth and the L-H mode operation on the loading ratio by observing the H/Pd under galvanostatic condition after the two succesive operations. Fig.7 shows the dependence of H/Pd on overvoltage observed at constant currents with and without the saw-tooth and the L-H mode operations before the galvanostatic steady state loading ratio measurement. No effect of preloading by the saw-tooth and the L-H mode operations was observed as shown in Fig.7.

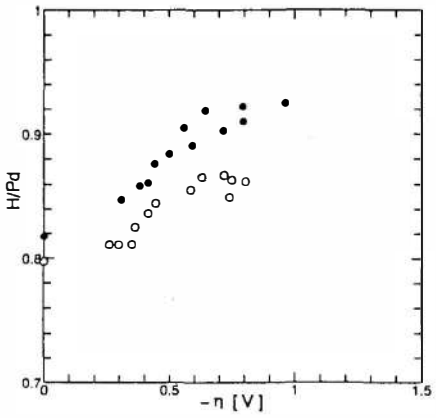

Fig.6 H/Pd vs. overvoltage on Pd sheet cathode with(○) or without (●) retainers

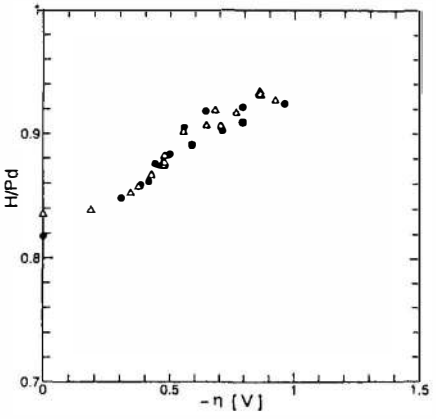

Fig.7 H/Pd vs. overvoltage on Pd sheet cathode with (△) or without (●) the saw-tooth and the L-H mode operation

4. Summary

The D/Pd ratio of 0.79～0.81 was observed on two batches of Pd sheet cathodes using the saw-tooth and the L-H mode operation. The low loading ratio suggests that the two modes operation has in fact little effect on improving the D/Pd.

Excess heat of 10～30% with respect to the input power was observed by only one of the Pd batches. The detection of the excess heat despite for the low D/Pd suggests that the two modes operation may lead to the higher local D/Pd than the average D/Pd on the Pd sheet cathode.

It is necessary to carry out further studies with better electrolysis geometry so that excess heat measurement can be conducted under higher D/Pd ratio.

5. References

[1] A.Takahashi, T.Iida, T.Takeuchi and A.Mega : Excess Heat and Nuclear Products by D_2O/Pd Electrolysis and Multibody Fusion, Submitted to International Journal of Applied Electromagnetics in Materials,1992.

[2] E.Storms , Fusion Technology, **433**,20,(1991)