

Anomalous Heat Generation/Absorption in Pd/Pd/LiOD/D₂O/Pd Electrolysis System

Chi-Meen WAN, Swe-Kai CHEN, Chiao-Jiunn LINN,
Chi-Yung LIANG, Chun-Jung LIN, Shuh-Biao CHU,
and Chi-Chiao WAN

ABSTRACT

The annealed Pd cathode, which was deuterium-loaded for 3500 min, showed an anomalous heat generation. Anomalous heat absorption was regularly repeated from 4000th to 7500th min of deuterium loading in the same cell. The current density was 2700 mA/cm². Total heat production was 0.3 to 3.3 MJ. Heat production of Pd per unit volume was 3.2 to 35.2 kJ/mm³. Total absorption was 22 kJ and absorption density was 217 J/mm³ in a typical absorption.

1. Introduction

Anomalous heat was reported by Pons and Fleischmann (1). Although it has been verified from many places, the reproducibility is still very poor. In the laboratory at National Tsing Hua University, Taiwan, the results are very similar. In more than 3 years, small excess heat has been characterized repeatedly(2). An anomalous heat in Pd/Pd/LiOD/D₂O system was observed once at 2 pm on 28 December, 1991. There is another kind of phenomenon, anomalous heat absorption, which has not been reported, occurs repeatedly. Whether there is any relation between heat generation and absorption is very valuable to study, it is the purpose of this report to introduce the phenomena.

2. Experimental

Both cathode and anode are palladium wires with diameters of 3.2 & 0.35 mm, respectively. The surface area of the cathode is 2 cm². Quartz tube with diameter 50 mm and height 100 mm is used as cell container. The containing D₂O with 0.1M LiOD in the cell is 350ml, which is kept

at constant level with an automatic D2O feeder. The charging current density is monotonically and stepwisely increased to 2700 mA/cm² by a power supply until anomalous heat is generated.

The electrolysis cell is emerged in a water bath with a constant reference temperature. This temperature is kept by an Endocal RTE-220 Refrigerated Bath/Circulator (NESLAB; NH, USA). The RTE-220 bath is with a flow of 15 liters/min at 0 feed head and has a boost heater, which will come on if the bath temperature is 2.5 °C below the setpoint.

3. Results

I. The Phenomenon of Anomalous Heat Generation

Figs.1(a)-(b) show the records of cell and bath reference temperatures and DC power vs. time, respectively. These figures show the anomalous heat. In order to assure that the anomalous heat was generated in the electrolysis cell, rather than from the bath, a similar experiment was performed. The bath temperature was suddenly increased to 100 °C to observe the response of the cell. The results is shown in Figs.2(a)-(b). The main difference between Figs.1 & 2 is the temperature change. There is a temperature decrease for bath on the left side of the peak in Fig.1(a), and for cell on the left in Fig.2(a). The temperature decrease before (ie, on the left of) a temperature peak is explained to be a resistance to temperature rise caused by another heat source. By this mechanism, one can assure that the temperature rise in Figs.1(a) & 2(a) were caused by a heat source in the cell and the bath, respectively. The generated heat in the cell shown in Fig.2(a) is at least $15^{\circ}\text{C} \times 500\text{ml} + 6.4^{\circ}\text{C} \times 350\text{ml} = 77,240 \text{ Cal} = 324,408 \text{ J}$. This would heat the water in the bath from 3509th to 3523-th min and from 85.4 to 100.4 °C (the 0.4 °C variation was an error recorded by computer). That is, the increasing rate of the bath heated by the cell is 1.07 °C/min.

On the other hand, it can be calculated from Fig. 3(a) that the boost heater heats the bath water from 85.4 to 100.4 °C and from 3914 to 3933 min. The heating rate is 0.79 °C/min. The heating rate by the cell is 35.7% higher than that by the boost heater. This means that the anomalous heat generated in the cell is at most 1086 W. (The heat generation for the boost heater is 800W.) Since the anomalous heat generation was from 3510th to 3560th min, the total heat generation in the cell is estimated to be at most $1086 \text{ W} \times 50 \text{ min} \times 60 \text{ sec} = 3258 \text{ kJ}$. The size of palladium cathode is 3.2 mm in diameter, 20 mm in length. The volume of the cathode is 101.6 mm³. The unit volumic heat is thus from 3.2 to 35.2 kJ/mm³ if one assumes that the heat was totally generated by the cathode.

II. The Phenomenon of Anomalous Heat Absorption

Figs.3(a)-(b) show the records of anomalous heat absorption in the electrolysis cell. Fig.3(a) shows the temperature distribution in the cell (the upper three tangled curves) and reference temperature in the bath (the lower curve). The DC power, voltage, and current in the cell are shown in the upper, middle, and lower curves, respectively.

It can be seen from Fig.3(a) that the cell temperature decreases from 95 to 78 °C within 10 min and then recovers to 95 °C. The reference temperature can rapidly drop by 4 °C. These temperature drops occurred once every 500 to 600 min. The highest drop is 15 °C. The anomalous heat generation and absorption happened in the same cell and the time interval between the heat generation and the 1st heat absorption is about 300 min. The typical heat absorption shown in Fig.3(a) is estimated to be

$$15\text{ }^{\circ}\text{C} \times 350\text{ ml} = 5250\text{ Cal} = 22,050\text{ J.}$$

The voltage and current were steadily decreased by a small rate until a temperature drop suddenly occurred and then increased to the original values (Fig.3(b)). The DC power drop for each cycle is about 10 W, ie, in 110 ± 5 W. For each drop, the dropping rate is more rapid than the temperature recovering rate. The dropping rate is at least $15^{\circ}\text{C}/5\text{min} = 3^{\circ}\text{C}/\text{min}$. The unit volumic heat absorption is $22050\text{ J}/101.6\text{ mm}^3 = 217\text{ J}/\text{mm}^3$.

4. Summary

In this experiment, a Pd/Pd/LiOD/D₂O electrolysis system was used. The diameters of Pd for cathode and anode were 3.2 and 0.35 mm, respectively. In the case of quartz container, both anomalous heat generation and absorption were observed. The total heat production for anomalous heat burst in quartz container is calculated to be from 0.3 to 3.3 MJ and the unit volumic heat of Pd cathode is from 3.2 to 35.2 kJ/mm³. For heat absorption, the total heat involvement is 22kJ and 217J/mm³. The current density was 2700 mA/cm².

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5. References

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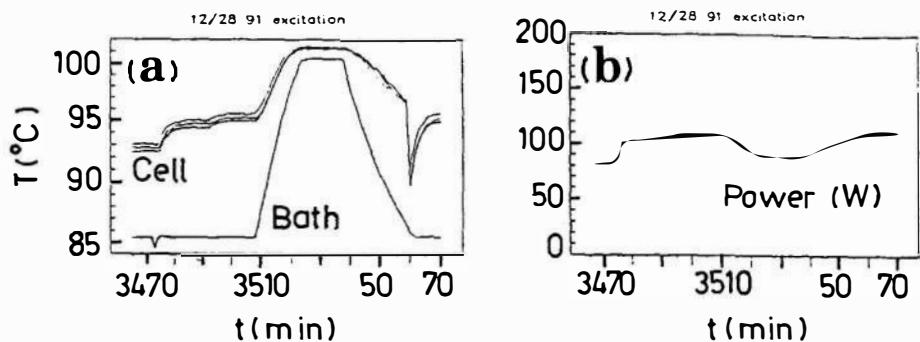


Figure 1. Records of System Temperature Increase (a) and Applied DC Power (b). (by heat generated in cell.)

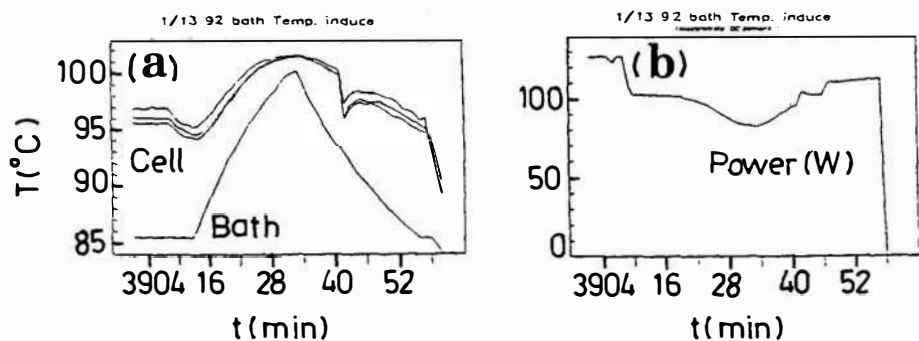


Figure 2. Records of System Temperature Increase (a) and Applied DC Power (b). (by boost heater.)

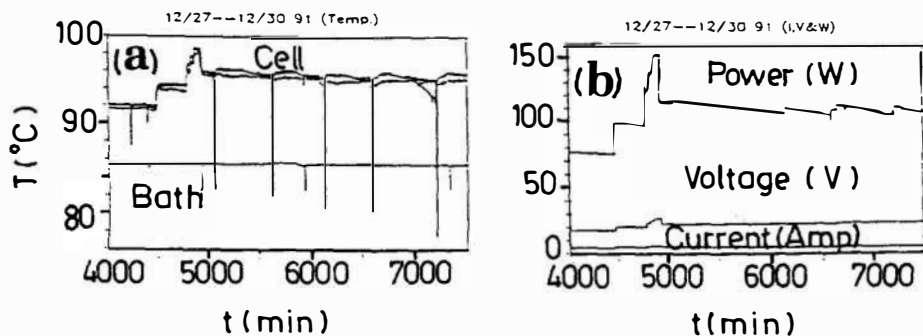


Figure 3. Records of System Temperature Drop (a) and Applied DC Power, Voltage, and Current (b) During Heat Absorption Inside the Cell.