Excess Energy and Nuclear Products

CORRELATION OF EXCESS HEAT GENERATION AND NEUTRON EMISSION IN Pd-LiOD ELECTROLYSIS

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Abstract

The correlation of the excess heat generation and the neutron emission in Pd-LiOD electrolysis has been investigated by use of a pair of experimental systems consisted of the same equipments in principle and operated coincidentally. Three pairs of electrolysis runs have been performed on the coincidental experiment systems, resulting clear time correlations between the excess heat generation and the excess neutron emissions. The reproducibility of the anomalous phenomena occurred in the present electrolysis could be accomplished completely in the series of experiments by a pretreatments of the Pd electrodes and the special purity control of the electrolyte used.

Introduction

The correlation between the excess heat generation and the excess neutron emission has been discussed based on the data obtained by the background runs with the light water and the foreground runs with the heavy water which were operated reciprocally under the same electrolysis conditions. In the course of the discussion, some uncertainty arouse on the neutron detection that came from the different date of the operation between the background runs and the foreground runs. To avoid such an uncertainty, especially in the excess neutron evaluation, two sets of the experimental systems have been assembled with the possibly same equipments such as the excess heat monitoring systems and the neutron detection systems of NE213 liquid scintillation counters.

In the present work, by use of the experimental systems, the background runs with the light water and the foreground runs with heavy water have been carried out conincidentally under the same conditions which gave us the positive results with high reproducibility to confirm the time correlation between the excess heat generation and the excess neutron emission and to check the reproducibility of the anomalous phenomena occurred in the deuterium-Pd system.

Experimental

The blockdiagram of the coincident experiment systems used are illustrated in Fig.1. The systems consists of the same equipments in principle. However, there might be some little differences in their characteristics especially in the excess heat monitoring systems and the neutron counting systems.

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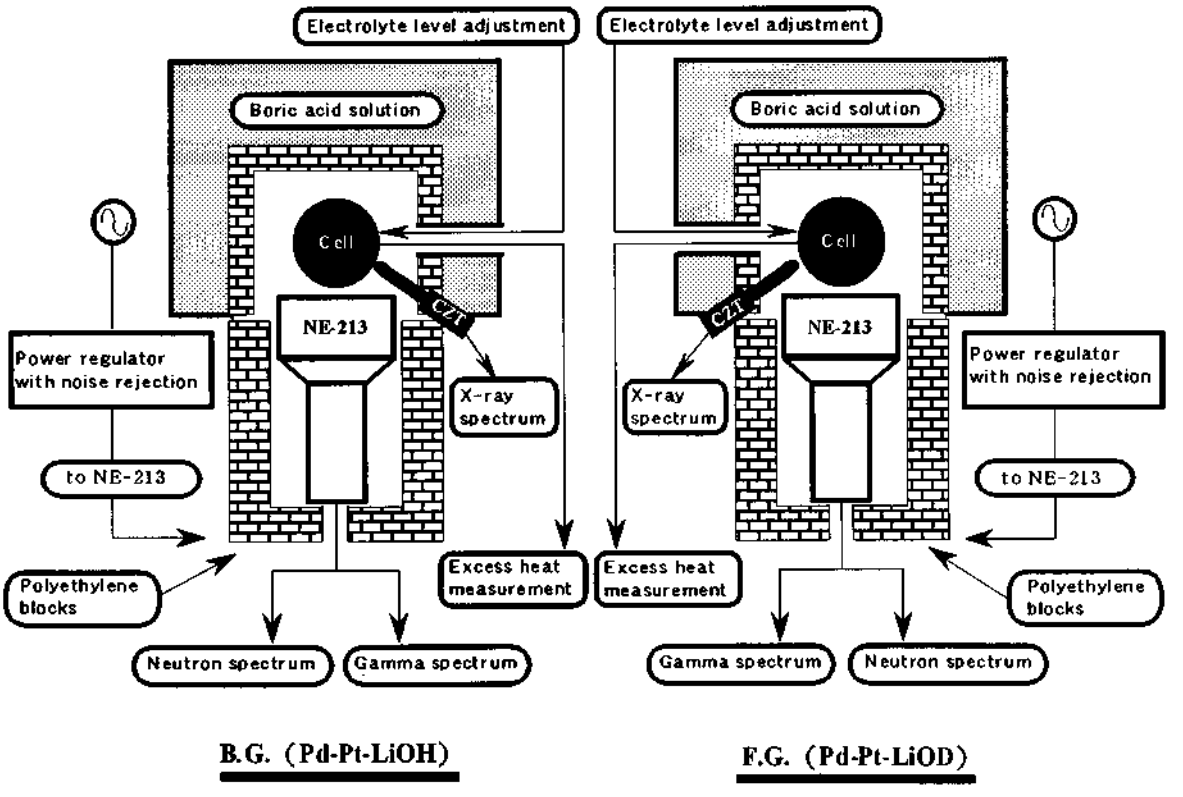

Fig.1 Coincident operating systems

The machine factors on the two systems have been checked before and after the every electrolysis operation. The details of the NE213 neutron counting systems were described in the previous papers.[1]

The schematic view of the electrolysis cell system is illustrated in Fig.2. The details of the electrolysis cell are same as described in the previous papers except the automatic electrolyte supplier. By this, the level of the electrolyte could be adjusted at a constant level throughout the electrolysis.

The ambient temperature was controlled at 24 ± 1℃ and the temperature of the cooling water was kept at 24 ± 0.1℃. The fluctuation of the electrolyte temperature caused by the supplying the electrolyte became negligible throughout the operations.

The data acquisition were performed on a personal computer in every thirty seconds

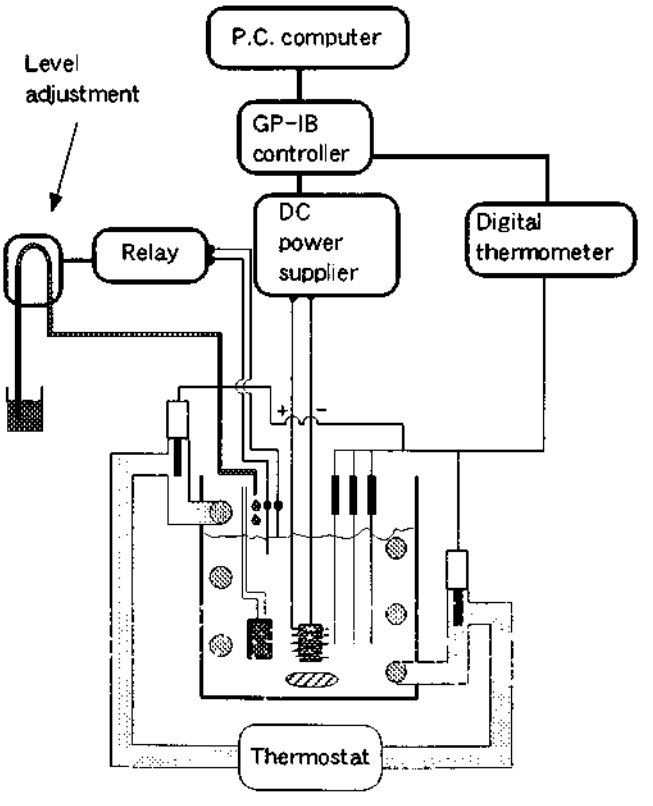

Fig.2 Electrolysis cell and Calorimetry system

Table1 Experimental conditions

		Cathode size	Pretreatment	Electrolysis mode
Run 1	B.G.	Pd 0.8 mm × 10 mm × 25 mm	Mechanical polishing 1 mm → 0.8 mm ⇩ Vacuum annealing 850 ℃, 10 hours	B.G. (LiOH) F.G. (LiOD) 1 M High / Low 800 / 20 (mA/cm²) 3 / 3 (hours)
	F.G.			
Run 2	B.G.			
	F.G.			
Run 3	B.G.			
	F.G.			

for the electrolyte temperature and in every hour for the neutron counting, the γ ray counting and the X ray counting, respectively.

The experimental conditions are listed in Table 1. The Pd cathodes used were four nine purity Pd plates manufactured by Tanaka Kikinzoku Co. Ltd. and supplied from NHE Center of Institute of Applied Energy as the standard material in New Hydrogen Energy Project. Before use, the surfaces of Pd plates were mechanically polished to the mirror surface, so 0.1 mm from the initial surface of the Pd electrodes was lost for each sides. The electrolyte used was 1 mole/dcm^3 LiOD or LiOH. The light water and the heavy water were distilled three times before use. The electrolysis was performed under the square pulse mode with 3 hours repetition, and with high current density of 800mA/cm^2 and low current density of 20mA/cm^2 as shown in Table 1. The electrolysis operations were carried out for four weeks or more. The excess power evaluation was performed by means of inner standard calibration method by use of a standard electrical resistance placed in the cells, and the details were described in the previous papers. [2]

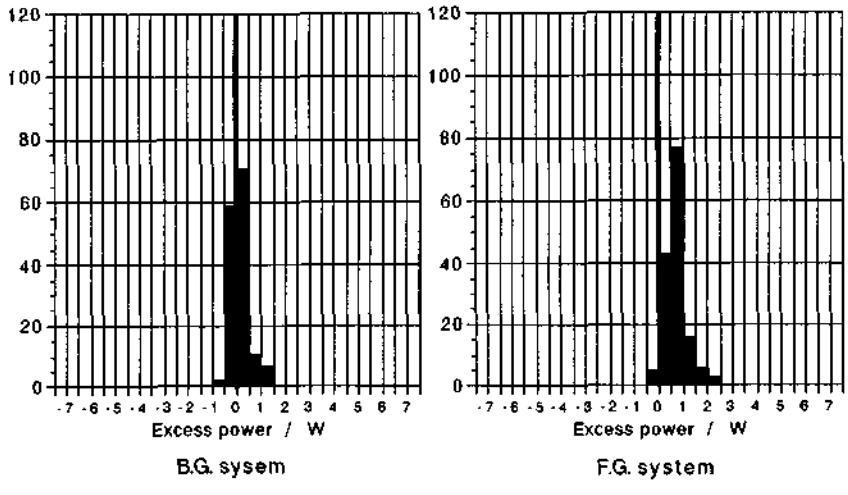

Fig.3 Histgram of the excess power for Run 1

Results and discussion

Excess power

The excess power obtained in the present series of the experiments are demonstrated by the

statistical histgrams in Fig.3, Fig.4, and Fig.5 for Run 1, Run 2, and Run 3, respectively. In these figures, the left hand side histgrams represent the results obtained in the background runs with the light water and the right handside histgrams represent the results obtained in the foreground runs with the heavy water.

The distributions of the histgrams indicate that the foreground runs with the heavy water gave the clear excess power generation except Run 2, while the background runs did not give any excess power as reported previously. The excess power generation can replicate in Run 1 and Run 3, but did not in Run 2. The reason why Run 2 did not give the excess power will be discussed later in this paper.

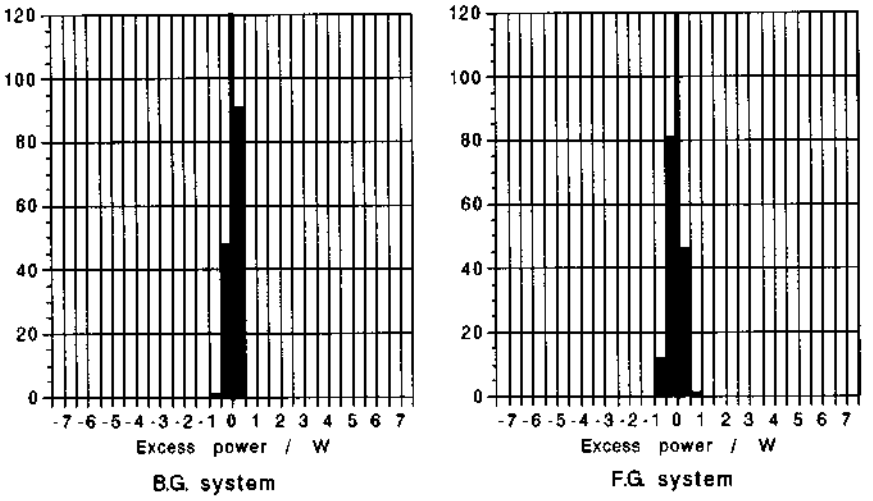

Fig.4 Histgram of the excess power for Run 2

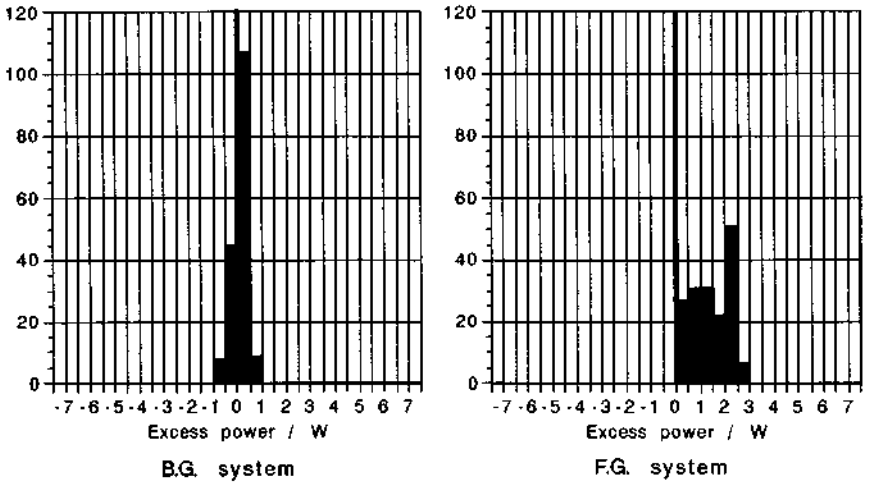

Fig.5 Histgram of the excess power for Run 3

Excess neutron

The neutron energy spectrum evaluated by the neutron ratios between the neutron counting rates in the background runs and the foreground runs are shown in Fig. 6. The error bars represent 3 σ . When the neutron ratios were over unity, it was concluded that the excess neutrons were detected in the corresponded bit of the PHA. It can be said from these spectrum that the excess neutrons were observed in Run 1 and Run 2, but did not observed in Run 2. The fact well corresponds with the results for the excess power as mentioned above. The reason why the Run 2 did not give any excess neutrons will be also discussed later in the paper. In Fig.7, the time correlation between the excess power generation and the excess neutron emission are demonstrated for Run 3. The time correlation is very clear from the start of the electrolysis to the termination of the operation. The intensities of the neutron ratios increased with increase of the magnitudes of the excess power. The clear time correlation between the two events is firstly reported in the new hydrogen energy project and other related researches.

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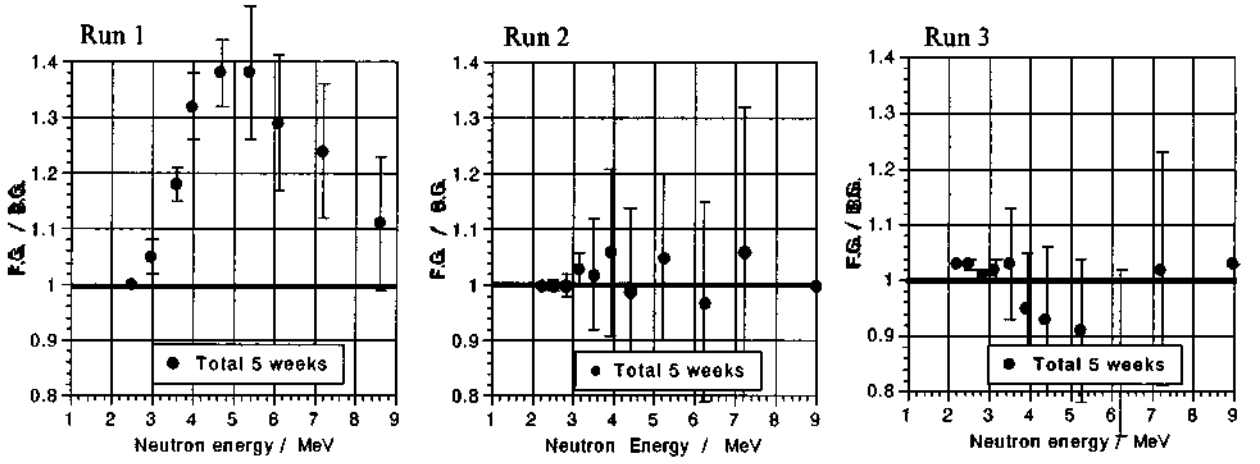

Fig.6 Neutron energy spectra of Run 1,Run 2 and Run 3

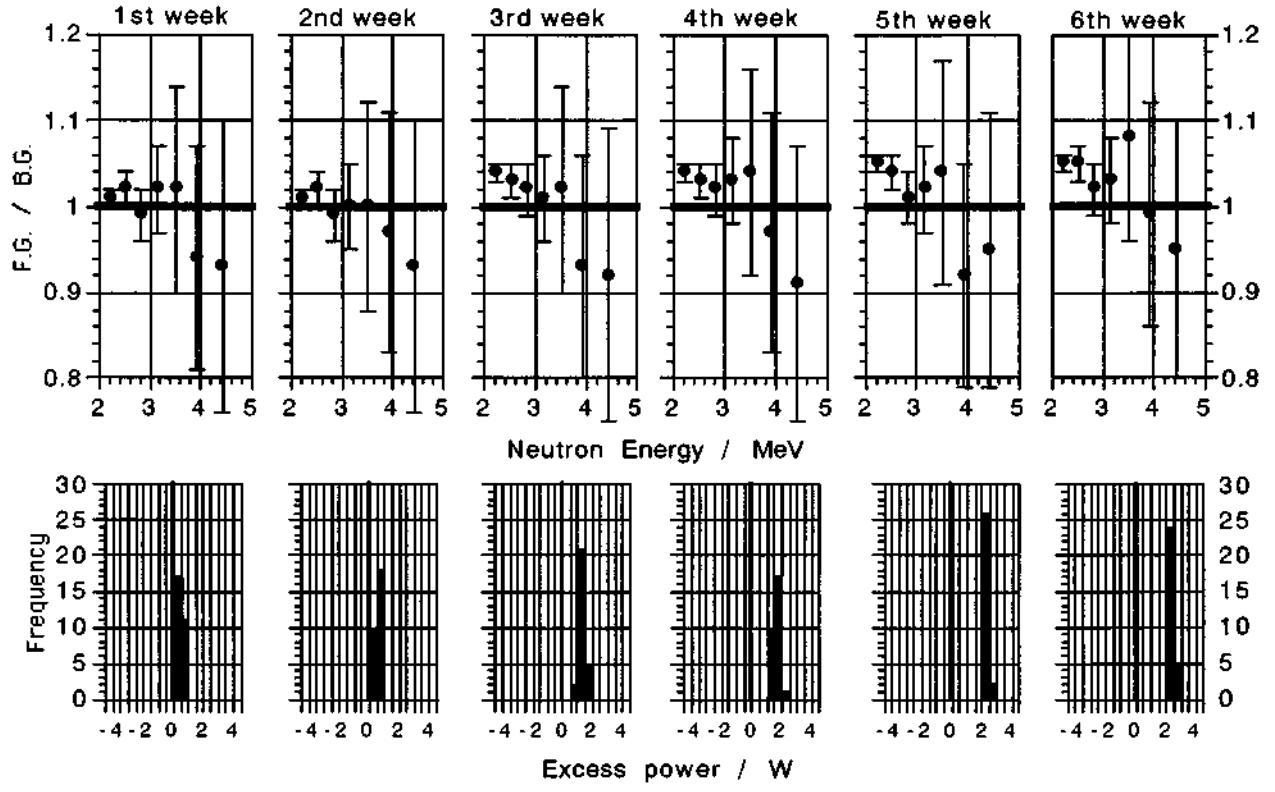

Fig.7 The time correlation between the excess power generation and the excess neutron emission

Cell voltage (open circuit voltage)

As discussed in the previous papers,[3] the cell voltage was recognized as one of the key factors to elucidate the mechanisms of the anomalous phenomena occurred in LiOD-Pd electrolysis process. The cell voltages have been also measured in the present series of experiments. The data are shown in Fig. 8 for the three runs.

The cell voltages changed with some nonlinear fractions in Run 1 and Run 3 comparing with the curves observed in the background runs in all of the runs, while Run 2 gives almost straight linear curves even in the foreground run. The similar nonlinear fractions have been observed in the previous experiments by the authors, resulting the excess heat generation and the excess neutron emission. [1] It can be said that the previous findings have been strongly confirmed by the present results obtained

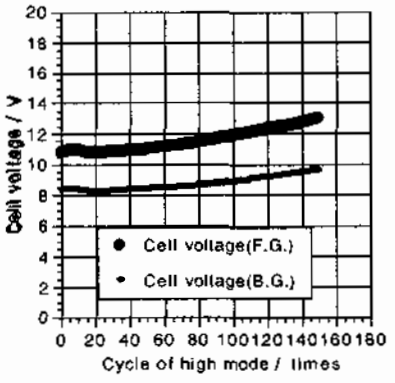

Fig.8-a Cell voltage for F.G and B.G. of Run 1

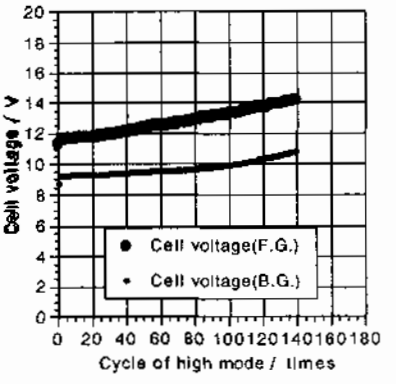

Fig.8-b Cell voltage for F.G. and B.G. of Run 2

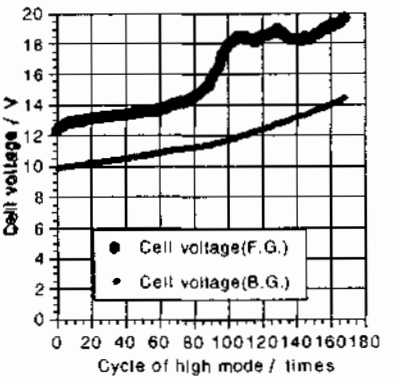

Fig.8-c Cell voltage for F.G. and B.G. of Run 3

from the couple of the coincidental electrolysis operations. The nonlinear fraction in the Run 3 is tremendously anomalous as has been never found in the present authors studies.

Combining the three findings of the excess power generation, the excess neutron emission and the nonlinear fraction of the cell voltage, it can be concluded that the larger deviation with larger nonlinear fraction from the curves obtained in the background runs gave larger excess power and more clear excess neutron emission.

At the last part of the present work, the authors should describe the reason why Run 2 gave no anomalous events. In fact, there was a different procedure in Run 2 from the other two runs. The Pd electrodes in Run 2 were placed in the electrolyte for two weeks before the electrolysis operation start to check the machine factors. Only this point is unique point for Run 2 in the all of the experimental conditions. It can be said that the Pd electrode should be employed for the electrolysis immediately after the cooling of the Pd electrodes in annealing chamber.

Conclusion

A pair set of the experimental systems to avoid the uncertainty caused from the different date of the background runs and foreground runs has been assembled to study the correlation between the excess power generation and the excess neutron emission in LiOD-Pd electrolysis. By use the systems and the well controlled conditions, the time correlation was observed clearly. The well controlled conditions was found to replicate the anomalous phenomena based on the deuterium. The anomalous nonlinear fraction observed in the cell voltage in the heavy water electrolysis was confirmed as one of significant factors to elucidate the new hydrogen energy process. The further investigations should be carried out to clarify the mechanisms of the anomalous phenomena including the nonlinear fraction occurred in the cell voltage.

Acknowledgments

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[3] M.Okamoto, T.Kusunoki, Y.Yoshinaga, H.Ogawa and M.Aida, "Excess Heat Generation, Neutron Emission and Cell Voltage Change in D_2O-LiOD Systems", Trans. Fusion Tech., **26**, 176～179 (1994)