MEASUREMENTS OF EXCESS HEAT AND NUCLEAR PRODUCTS IN Pd-D₂O SYSTEM USING TWIN OPEN TYPE ELECTROLYSIS CELLS

H. Fukuoka, T. Ikegawa¹, K. Kobayashi², A. Takahashi
Department of Nuclear Engineering, Osaka University,
2-1 Yamadaoka, Suita, Osaka, 565, Japan
¹Present affiliation, HITACHI Co.Ltd.
²Present affiliation, University of Tokyo

Abstract

Measurements of excess heat, X-rays and neutrons to study possible correlation between excess heat and nuclear products during the electrolysis of LiOD heavy water electrolyte using Pd cathodes were carried out. Two open type electrochemical cells with two sets of X-ray and neutron detection systems were formed to be twin type. Therefore, two experiments being carried out by two cells at the same time were compared at any time and false signals in coincidence for two cells were able to be rejected as accidental noises. No excess heats were observed out of seven runs after ICCF5. But weak excess X-rays and neutron burst were observed at the same time in one case. And only soft X-rays were detected during electrolysis of heavy water using Pd cathodes in two cases and one case for electrolysis of light water.

1. Introduction

Since “cold fusion” phenomenon was reported by Fleishmann and Pons¹, excess heat and nuclear products during heavy water electrolysis were reported by many groups. However, it was difficult to reproduce this phenomenon in most of the groups². We reported slight excess heat (5-7%) with weak neutron emission in energy over 3MeV at ICCF5³. The aim of this study was to reproduce our last results and to perform more reliable measurements to search the correlation between excess heat and nuclear products. If nuclear reaction with heat generation takes place in solid without neutrons, high energy (several MeV) charged particle emissions are usually expected. Therefore, emissions of characteristic X-rays and bremsstrahlung X-rays will be caused by slowing-down of the charged particles in material. Since most of these emissions will be characteristic X-rays, charged particles can be detected indirectly by the detection of the characteristic X-rays. Pd has been used as the cathode of the heavy water electrolysis in our group. Because characteristic X-rays
of Pd Kα and Kβ are 21-23keV, it is necessary to measure the X-rays of this energy range. A CdTe solid state detector was chosen to detect these X-rays because of its high efficiency in the energy less than 90keV and its compatibility to be used at room temperature.

CdTe detector picks up electromagnetic noise or mechanical noise easily will give a bad influence upon the accurate measurements. Figure 1 shows the X-ray spectrum of the bare CdTe detector compared with that of calculated spectrum with CdTe detector covered with molybdenum foil during a heavy water electrolysis run with Ni cathode. If the origin of this spectrum was due to X-rays, the spectrum of CdTe detector covered with molybdenum foil should look as the lower spectrum in Figure 1, because X-ray attenuation cross section of molybdenum is like Figure 2. But actually measured X-ray spectrum using the molybdenum covered detector during the heavy water electrolysis was obtained as shown in Figure 3, from which we could find that the origin of this spectrum was not totally X-rays. We introduced them two-dimentional pulse-shape and pulse height analysis to reject the noise signals. Figure 4 shows background X-rays in two dimensional spectrum which was plotted for rise time of pulse as the axis of abscissa and pulse height on the vertical line. The
signals of which rise time were \( \sim 2 \mu \text{sec} \) were accepted as X-ray signals, and the signals <1 \( \mu \text{sec} \) were rejected as noise signals. Figure 5 shows background spectrum before rise time discrimination and background spectrum after rise time discrimination. This rise time discrimination could reject most of noise signals and slower rise-time events of X-rays. The electrical circuit of rise time discrimination of CdTe signals is shown in the next section.

On the other hand, calorimetry and neutron measurements were performed, too. The electrochemical cells and measurement systems were twin type, and two type experiments were able to be practiced at the same time, for example heavy water electrolysis and light water electrolysis using Pd cathodes for both cells. The advantage of this twin experimental system was that simultaneous events happening in both cells could be rejected as the noises. Using this system, the accurate comparative experiments of cold fusion could be performed for foreground/background or /blank runs.

2. Experimental

The twin open-type cells and measurements systems were designed as shown in Figure 6. The system included two open-type electrolysis cells called F cell and B cell, two X-ray spectroscopy systems using CdTe detectors, and two fast neutron spectroscopy systems with NE213 detectors.
One power supply was connected two cells in series and supplies the same electric current. Coolant ($\text{H}_2\text{O}, 20 \pm 0.1^\circ\text{C}$) flow was connected to two cells also in series. Figure 7 shows the structure of open type cell. Inner cell wall was made of glass and outer cell wall was made of acrylic resin. The plate cathode ($25 \times 25 \times 1\text{mm}$) was hold by Teflon electrode holder which rolled up the anode wire (1mm φ). Coolant flowed in the coolant pipe which was made of glass and temperature distribution of the electrolyte was kept uniform by stirrer. The effluent gas ($\text{D}_2$, $\text{H}_2$ and $\text{O}_2$) which were generated by the electrolysis went out through the gas outlet. The electrolyte temperature was measured by K-type thermocouples. The output power of electrolysis was linear function of the thermocouple outputs, and the coefficients of the linear function were determined by the calibration run using Ni cathode in every experiment. The excess energy was determined by the difference between output power and input power to the electrolyte. The input power was expressed by ($\text{cell voltage} - 1.5$) × electrolytic current) because of gas generation.

Figure 8 shows the X-ray measurement system. The CdTe detector ($2 \times 2\text{mm}$) was placed in front of the cell. Using this system, noise components in CdTe signals were distinguished by the rise time and separated from real X-ray signals. The measurement range of the system was about 6-90keV in
energy. The detection efficiency of $K_{\alpha}$ and $K_{\beta}$ X-rays was 0.076% and the measurement system could detect one event per minute when Pd cathode emitted 1300 photons of this energy per minute. X-ray events are counted every 4 minutes besides spectroscopy. For F cell at Run 7, high resolution Si-SSD which was cooled with Peltier device was used instead of CdTe detector to detect lower energy X-rays than 6keV.

The $n-\gamma$ pulse separation technique was applied to the neutron measurement using an NE213 fast neutron spectrometer, which was placed in front of the cell, as shown in Figure 9. The $\gamma$-ray component and noise component could be separated completely by this diagram. The measurement range of neutron is 0-9MeV in energy. But one of the NE213 detectors (for B cell) could not perform right measurement in low energy ($<3\text{MeV}$) because of containment $\alpha$-particle emitters which are included in this NE213 detector, so it was used exclusively for higher energy. Neutron events were also counted every 4 minutes in addition to spectroscopy.

At most of experiments, the electrolysis current density was changed periodically from LOW (160 or 280mA/cm$^2$) to HIGH (608 or 640 mA/cm$^2$) or the reverse in every 3 or 6 hours (L-H mode) in order to give dynamic change of deuterium accumulation in Pd metal. But for a week at the beginning of electrolysis, the electrolysis current density was kept to be 160mA/cm$^2$ constantly (Constant Mode) in order to keep Pd cathodes from cracking, except for Runs 1-3. The electrolyte was 0.2M LiOD/heavy water solution.

3. Results and Discussions
Table 1 shows the summary of the experiments. In the table, Yes means that anomalous events were observed.

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<th>No</th>
<th>Period</th>
<th>Cell</th>
<th>Cathode</th>
<th>Treatment</th>
<th>Particular treatment</th>
<th>Electrolyte</th>
<th>Electrolyte current (mA/cm²)</th>
<th>Excess heat</th>
<th>X-ray</th>
<th>Neutron</th>
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Table 1  The summary of experiments

Figure 10  Typical example of heat balance (at Low-High mode)

Figure 11  X-ray energy spectra using CdTe detector
Figure 10 shows the typical data of heat balance when the current density was changed from 160mA/cm² (L-mode) to 640mA/cm² (H-mode) or the reverse in cycle. The excess power during thermal equilibrium was fluctuated around zero and not exceeding 99% confidence level (+3σ and -3σ).

Figure 11 shows the X-ray energy spectrum of Run 1 during heavy water L-H mode electrolysis at the first week of the electrolysis. This spectrum rising up in the low energy region is regarded as somewhat similar to electromagnetic noise signals, but it might be soft X-rays of the nuclear bremsstrahlung by charged particles. The similar spectrum was observed at Run 2 during light water electrolysis. These results may suggest the possibility of the production of low energy (less than 200keV) charged particles which generate no characteristic X-rays but do bremsstrahlung X-rays. At Run 6, the different X-ray spectrum was obtained at first week of Constant Mode electrolysis for the heavy water electrolysis using Pd cathode which was pre-processed by chemical etching, as shown in Figure 12. The MCS data is shown in Figure 13, where we see X-ray events caused as bursts. The events might be noise signals because...
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noise signals always appeared as bursts. Figure 14 shows the MCS data at Run 5 during the Constant Mode heavy water electrolysis using annealed (900°C, 10 hour) Pd cathode. Only for the F cell which electrolyte was LiOD heavy water solution, neutron and X-ray increased simultaneously around the time 17hr. Much larger X-ray burst-events after that event were probably noise signals because there were two simultaneous burst-events for the B cell which electrolyte was LiOH light water solution. Figure 15 shows the expanded-scale data for the neutron and X-ray coincident event. Because the time variation of neutron count rate is not identical to that of X-ray's, the origin of these events would not be the same reaction if these events were signals of some nuclear reactions. But there is possibility that the triggering conditions of two events will be the same solid-state or chemical conditions of the Pd cathode. Corresponding spectra of X-ray and neutron could not be identified, because counts in spectrum channels didn't exceed the statistical error level.

The helium-4 analysis of the gas in used Pd sheet at Run 5 for the F cell by quadrupole mass spectrometer was performed with the gas extraction condition Pd sheet was kept 1150°C for 10 hour after electrolysis. Helium-4 peak was appeared slightly but that was very marginal and uncertain level, because of the problem of leaking of air into the gas storage part of the spectrometer oven during a long heating up process for the Pd sheet.

4. Conclusion

In order to study the correlation between excess heat and nuclear products, the measurements of heat balance, X-rays and neutrons during heavy water and/or light water electrolysis using Pd cathodes were performed. In this work, the reliable twin system was used. Above all, the X-ray measurement system for the correlation study was improved.

As a result, no excess heat beyond 3σ level was observed. But for only heavy water electrolysis, X-ray and neutron event occurred simultaneously. Other three X-ray events were observed out of seven runs for both of heavy water electrolysis and light water electrolysis. And all of them occurred at the first week of the electrolysis. The energy spectra of X-ray were not characteristic X-ray's but looked like the bremsstrahlung's. But these X-ray spectra were also similar to noise signal's. The simultaneous X-ray measurements by a bare detector and a detector covered with metal foil as described at introduction will be needed in future to prove that these X-rays were not noise signals but genuine soft X-ray signals.

References

3) A. Takahashi, et al., 1995, Proc. 5th Conf. on Cold Fusion,
4) E. B. Saloman, et al., 1988, Atomic Data and Nuclear Data Tables, 38, 1