Helium and Heat Correlation

STUDY OF EXCESS HEAT AND NUCLEAR PRODUCTS WITH CLOSED D_2O ELECTROLYSIS SYSTEM

K. Yasuda , Y. Nitta† and A. Takahashi

Department of Nuclear Engineering , Osaka University , 2-1 Yamadaoka , Suita , Osaka , Japan.

† Present address , Matsushita Electric Industrial Co.,Ltd.

Abstract

Using a closed type heavy water electrolysis system , deuterium loading ratio D/Pd , output power (by using mass flow calorimetry method) and neutrons were measured in-situ simultaneously . Mass spectrum analysis of upper-cell gas and palladium cathode by a quadrupole mass spectrometer and tritium measurement in a sampled electrolyte were done by off-line techniques .

Excess heats up to approximately 4 to 5W were produced with the cold worked and copper layered (0.95 μ m) cathode . However , during excess heat , nuclear products (neutrons) were not observed over the 3 σ limit line of background level . In a few mass spectrum analyses , slight increases of helium-4 peaks were observed . However , helium-4 might not absolutely increase , because it was difficult to calibrate the mass spectrometer to deduce total amount of helium-4 from samples.

1.Introduction

The aim of this work is to study the correlation between excess heat by heavy water electrolysis with Pd cathode and deuteron-related nuclear products , i.e. , neutron , helium-4 and tritium .

The attainability of high loading of deuterium into various batches of Pd is reported to be one of the key parameters which govern generation of excess heat and nuclear products . Only SRI International[1] and IMRA-Japan[2] have ever given phenomenological relations between loading ratio (D/Pd) of deuterium and excess heat rates by in-situ measurements . However , their experiments lacked the measurements of nuclear products to see the correlation with excess heat . Miles et al.[3] have reported the correlation between excess heat and helium-4 and showed that the quantity of observed helium-4 could correspond to the assumed reaction of D + D $\rightarrow$ ^{4}He +23.8MeV as the heat generating nuclear reaction . Therefore , "cold fusion" may not be the normal D-D reaction which should produce neutron and tritium , but would be a new class of fusion in solid . Therefore , in order to prove the existence of the new class of nuclear reaction in metal / deuterium system essentially and find a solution to the mechanism of possible reaction , the simultaneous in-situ measurement of loading ratio (D/Pd) , excess heat and nuclear products with their correlations must provide us key evidences .

In the present work , D/Pd ratios , excess heats and nuclear products were tried to measure simultaneously in-situ in a closed heavy water electrolysis system . To carry out accurate calorimetry , a newly designed closed electrolysis cell which enabled heat measurement by using the

mass flow calorimetry method was manufactured.

With this system , seven experiments have been done by different cathode materials , i.e. , using several pre-conditioned palladium plates (cold worked , annealed , with or without copper surface layer) .

2.Experimental

Closed electrolysis cell

Fig.1 shows a cross sectional view of a newly designed cell . This cell has relatively large volume (738.4±4.1cc) , so that the electrolysis can be carried out with relatively high input power , i.e. , several-tens watts . The cell vessel is made of stainless steel coated with Teflon and a spiral copper cooling pipe is brazed around it . Temperatures at the inlet and the outlet point of cooling water were measured with CA thermocouples . A catalyst made of 0.5% Pd alumina pellets recombines dissociated deuterium by electrolysis with oxygen to form D_2O being fed back to electrolyte . The jointing part of plumbing was equipped with the lid , and a space between the lid and the vessel was sealed with copper gaskets . Loading ratio of deuterium could be determined by measuring the change of D_2 gas pressure of upper cell volume where air at the beginning of mounting was evacuated and replaced with D_2 gas (about 3atoms) . Even small quantity of helium-4 generated continuously or intermittently during electrolysis can be stored without loosing for many hours . By sampling the upper cell gas after the electrolysis run and analyzing it by the quadrupole mass spectrometer , the production of helium-4 can be detected .

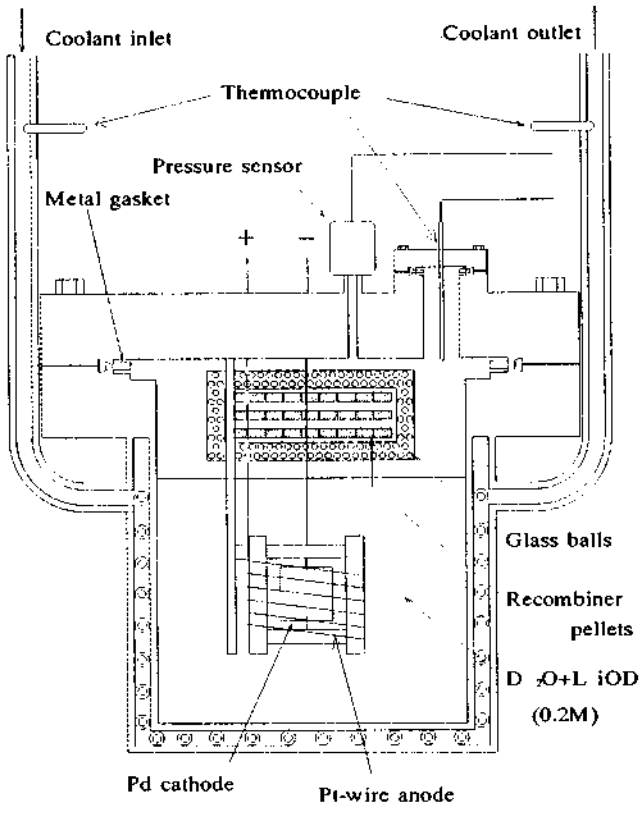

Fig.1 Closed electrolysis cell

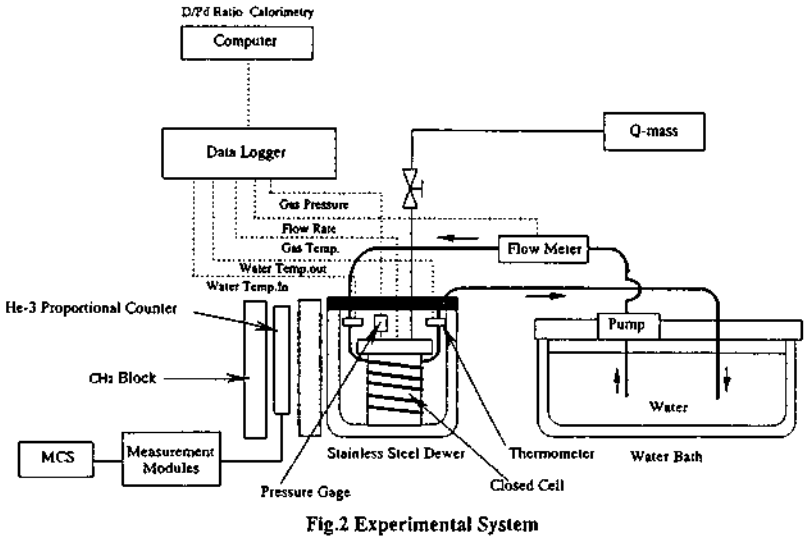

Fig.2 Experimental System

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Neutron measurement

Fig.2 shows the measuring system of the experiment , where is a heilum-3 proportional counter (LND-2531) set up inside polyethylene blocks . The counter has 8 inch (about 20.3cm) effective length and 1.93 inch (about 4.1cm) effective diameter . LLD and ULD of the single channel analyzer (SCA) were adjusted to count the signals within the ROI area (MCA-7800 , SEIKO EG&G) of ^{3}He(n,p) reaction thermal peak in pulse height spectrum to detect only neutrons . Discriminated neutron signals were counted every four minutes with multi-channel scaler (MCS) . Fig.3 shows the flow chart of neutron measurement system .

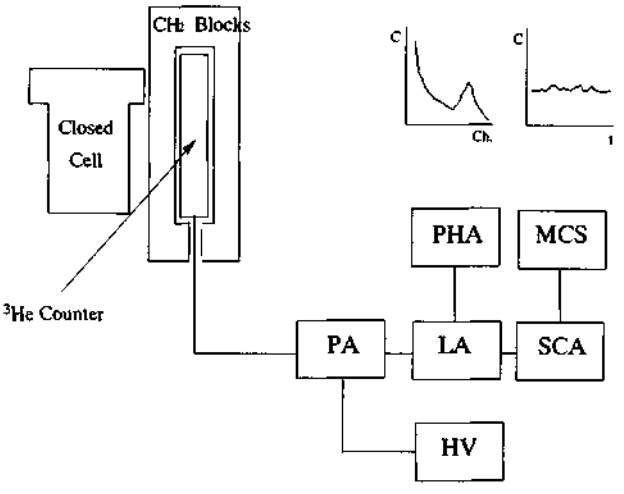

Fig.3 Block Diagram of Neutron Measurement

High resolution quadrupole mass spectrometer

High resolution quadrupole mass spectrometer (HIRESO-2SM , Vacuum Science and Engineering Co.Ltd.) was used to analyze mass spectrum (mainly mass four) of samples of upper cell gas and palladium cathodes . Minimum detectable partial pressure was about 10^{-11} Torr for helium-4 by this analyzer which corresponded to a number of atoms about 10^{14} . This analyzer has high resolution , i.e. , Δ M/M=0.02 (M : mass number) . Therefore helium-4 (mass number 4.0026) and deuterium (mass number 4.0282) can be measured as clearly separated two peaks .

On some of experiments , a hydrogen-getter-pump (SORB-AC100G , ULVAC Co.Ltd.) was used to remove deuterium from analyzed gas to increase the sensitivity for helium-4 detection .

Tritium analysis

Tritium was analyzed with the liquid scintillation counter . A 15cc liquid scintillator cocktail (AQUASOL-2) was mixed with 1cc electrolyte sampled before (and after) an experimental run for about a week or more and luminescence by β -decay was counted by the LSC method .

Experimental condition

Table 1 shows a summary of experimental conditions .

Seven experiments have been done using a 0.2M LiOD electrolyte and plate palladium cathodes (25 $\times$25$\times$1mm) . Several pre-conditioned palladium (cold worked , annealed , with or without copper surface layer[0.95 μ m]) were used for the cathodes . The Pd cathode used in Exp.3 was annealed for 10 hours at 900℃ with a heat furnace and was cooled down by the rate 10℃/minute . Pd cathodes in Exp.5 and Exp.6 had been already annealed before experiments .

No.	Pd cathode condition	Batch	Enclosed Gas	Electrolyte	Electrical current
Exp.1	Annealed	Nilaco#2	D_2(Sample1)	0.2MLiOD(350ml)	80~160(mA/cm^2)
Exp.2	Coldworked	Nilaco#2	D_2(Sample1)	0.2MLiOD(350ml)	160~320(mA/cm^2)
Exp.3	Coldworked	Nilaco#2	D_2(Sample2)	0.2MLiOD(350ml)	80~320(mA/cm^2)
Exp.4	Coldworked , Cu layer	NHE	D_2(Sample2)	0.2MLiOD(350ml)	80~320(mA/cm^2)
Exp.5	Annealed , Cu layer	NHE	D_2(Sample2)	0.2MLiOD(350ml)	80~320(mA/cm^2)
Exp.6	Annealed	Nilaco	D_2(Sample2)	0.2MLiOD(350ml)	80~320(mA/cm^2)
Exp.7	Coldworked	TNK	D_2(Sample2)	0.2MLiOD(350ml)	80~320(mA/cm^2)

Table1 Experimental Condition

3.Results and Discussions

Table 2 shows the summary of results for measurements of loading ratio (maximum values) , excess heat , neutrons , helium-4 and tritium .

No.	D/Pd ratio (Max)	Excess heat	Neutron	^{4}He(Upper-cell gas)	^{4}He(Ref. Sample)	Tritium
Exp.1	0.74	No	No	—	—	—
Exp.2	0.85	No	No	No	—	No
Exp.3	0.87	Yes (?)	No	Yes (?)	—	No
Exp.4	0.83	Yes	No	Yes (?)	Yes (?)	No
Exp.5	0.86	No	No	No	No	No
Exp.6	0.87	No	No	No	—	—
Exp.7	0.90	Yes	No	Yes (?)	—	—

(?): marginal level

Table2 Summary of Experimental Results

Figs.4 and Fig.5 show variations of excess heat and loading ratio (D/Pd) as a function of time .

In the run of Exp.3 , between 120 hours and 140 hours after the start of electrolysis , excess heat looked coming up . However , it was within the 3 σ (=3.9W) level of 99% confidence . Whenever anomalous excess heat was not produced , heat balance was at zero-line as looked like Fig.4 except the 120-140 hours interval.

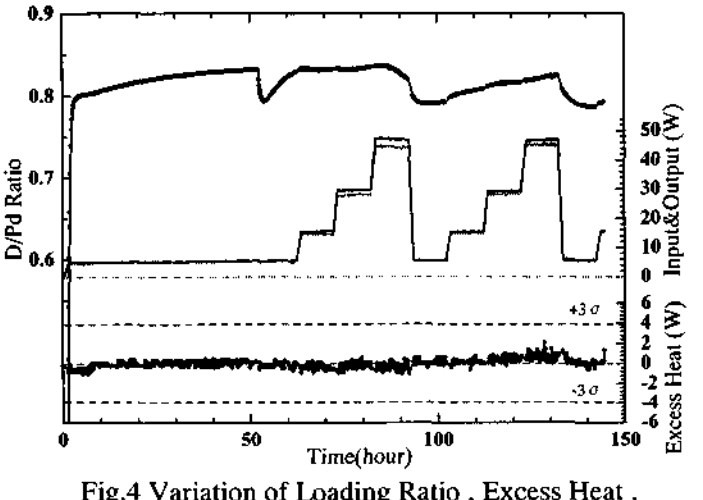

Fig.4 Variation of Loading Ratio , Excess Heat , Input and Output Powers with time (Exp.3)

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In the run of Exp.4 , excess heats were produced from the time when input current mode was changed to the stepping up mode . Maximum excess heat exceeded 3.5 σ level of the standard deviation and therefore very confident . Then D/Pd ratios were observed here to be almost constant ($\sim$ 0.83 with small variation) in spite of the drastic change of currents by the stepping up .

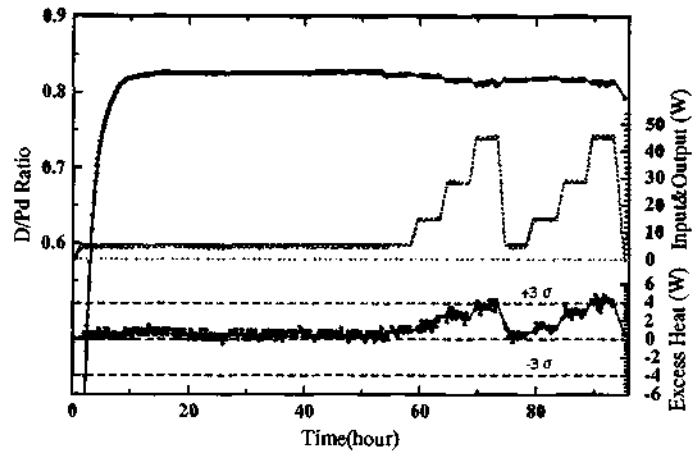


Fig.5 Variation of Loading Ratio , Excess Heat , Input and Output with time (Exp.4)

In the run of Exp.7 , electrolysis by the stepping up mode continued for about 200 hours , for about a day we stopped electrolysis and changed to the L (1A) / H (4A) mode with 6 hours duration per each of L or H mode which continued for about 240 hours . The L / H mode electrolysis technique was fully used by the open system study group of our laboratory[4] . Fig.6-1 shows variations of excess heat and loading ratio in Exp.7 .

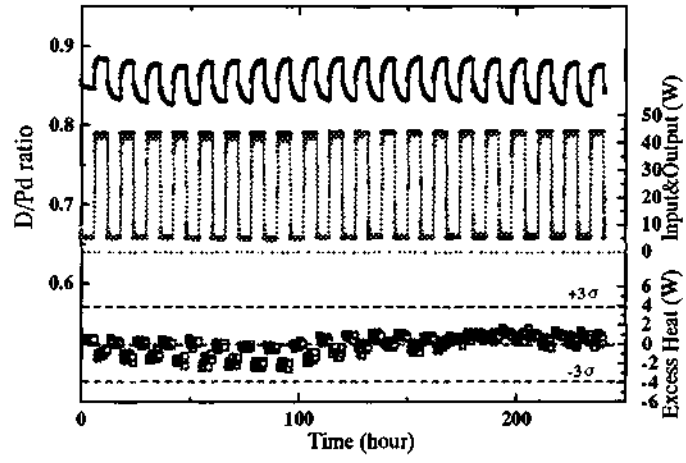


Fig.6-1 Variation of Loading ratio , Excess Heat , Input and Output with Time (Exp.7)

Excess heat was observed but not over the 3 σ level . And loading ratios were higher than those of Exp.4 as a whole but changed periodically in average , from 0.82 to 0.87 corresponding to the current changes of the L / H mode as shown with the upper most graph of Fig.6-1 . In the step-up made run of Exp.7 , excess heats with 5W max. were observed with maximum D/Pd = 0.90 (see Fig.6-2) .

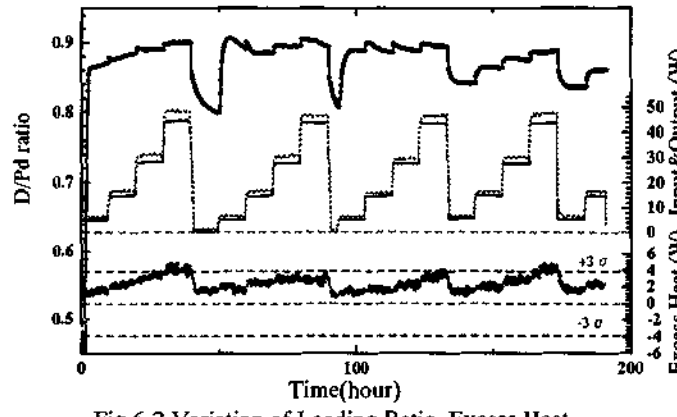


Fig.6-2 Variation of Loading Ratio ,Excess Heat Input and Output Powers with Time (Exp.7)

Helium and Heat Correlation

Fig.7 and Fig.8 show results of mass four spectra for cylinder gas samples (reference BG runs) in Exp.3 and Exp.4 . Judging from these diagrams , used original deuterium gas was regarded to be contaminated very slightly with helium-4 . However , in Exp.7 , helium-4 peak was not observed with the same deuterium gas sample of used D_2 gas cylinder , probably because the sensitivity of quadrupole mass spectrometer was getting worse than before .

Fig.9 through Fig.11 show results of mass four spectra for gas samples from the upper cell in Exp.3 ,

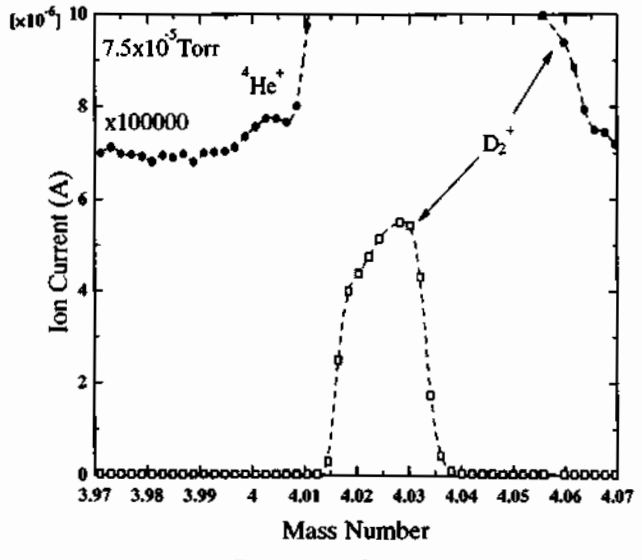


Fig.7 Mass Spectrum
(Exp.3 Gas Sample without Getter Pump)

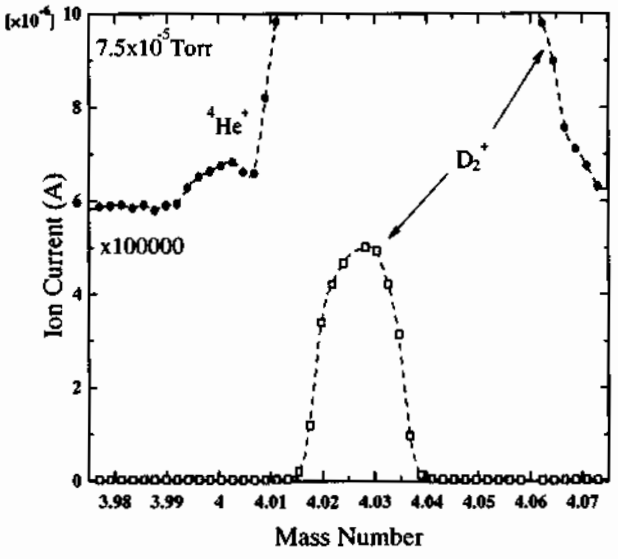


Fig.8 Mass Spectrum
(Exp.4 Gas Sample without Getter Pump)

Exp.4 and Exp.7 . In these analysis , ion currents of herium-4 peaks were observed with meaningful increases compared with ones of the original sample gas . Content of helium-4 in the sample gas from the upper cell was regarded as concentrated to be about twice through three times of the beginning D_2 gas after about 1 through 2atm D_2 gas was absorbed to Pd plate . So , amount of helium-4 peak-increases in Exp.3 , 4 and 7 should be regarded as marginal levels though Exp.4 showed the largest ^{4}He peak . It is necessary to calibrate quantitatively the mass spectrometer and fix the total quantity of helium-4 from the sample .

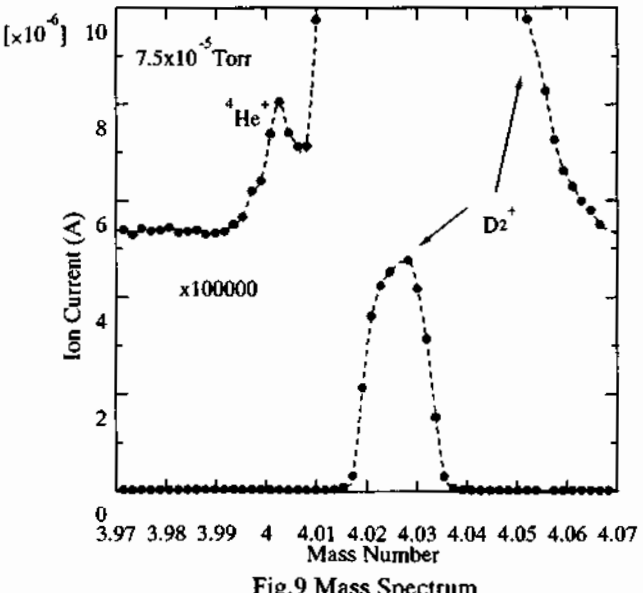


Fig.9 Mass Spectrum
(Exp.3 without Getter Pump)

Helium and Heat Correlation

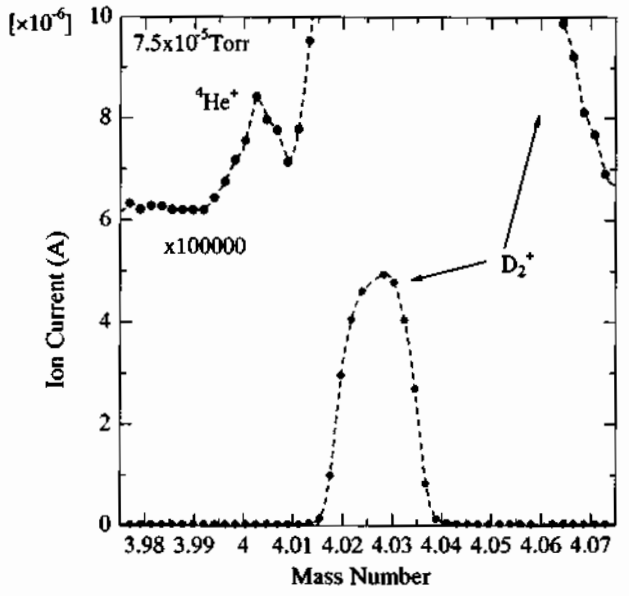


Fig.10 Mass Spectrum
(Exp.4 without Getter Pump)

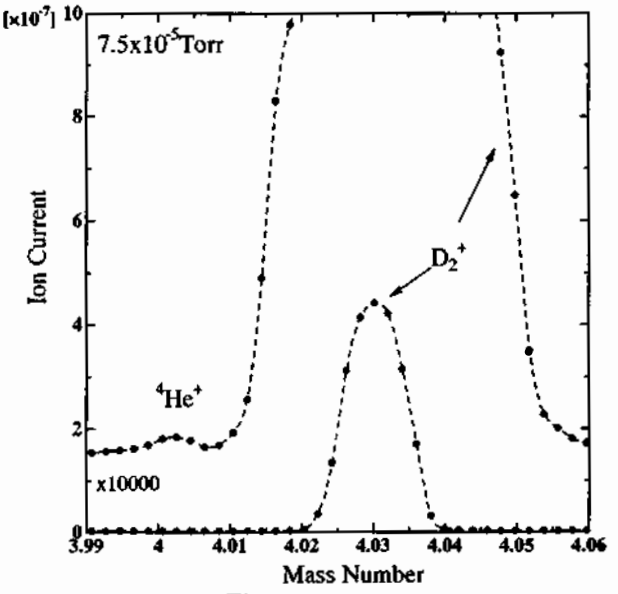


Fig.11 Mass Spectrum
(Exp.7 with Getter Pump)

In all the experiments , neutrons were not observed over the 3 σ limit line of the background level and clear increases of tritium in electrolyte were not observed either .

Fig.12 shows the MCS data of neutron in the run of Exp.4 . At four hours after electrolysis start , a count rate was observed over +3 σ level. However , since the neutron source facility "OKTAVIAN" near our experimental place was eventually running at the same time and skyshine neutrons were thought to shower , increase of neutrons might not be due to generation of neutrons from the palladium cathode .

Fig.13 shows results of tritium analysis in Exp.2 through Exp.5 . Tritium activity of electrolyte before electrolysis was defined as background and one after it as foreground . There was difference in tritium level between that of Exp.2 and those from Exp.3 to Exp.5 because electrolyte was exchanged to new one after Exp.2 ; tritium concentration (selective absorption of lighter isotope , i.e. , H and D to palladium plate) during the long electrolysis would be suggested .

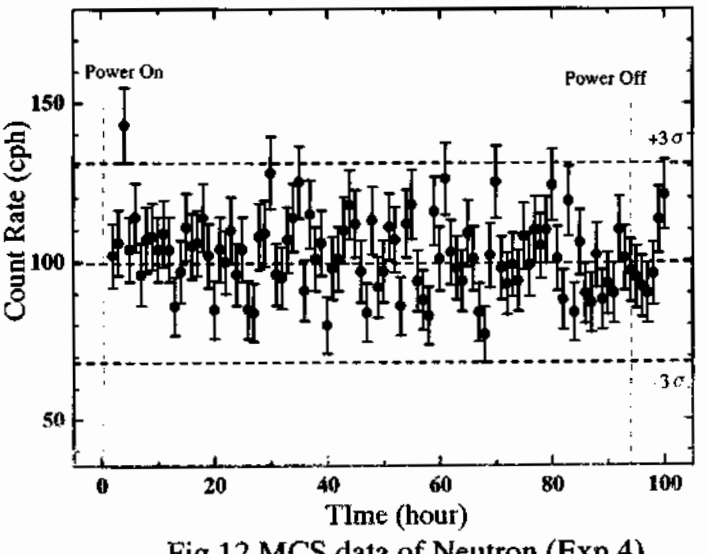
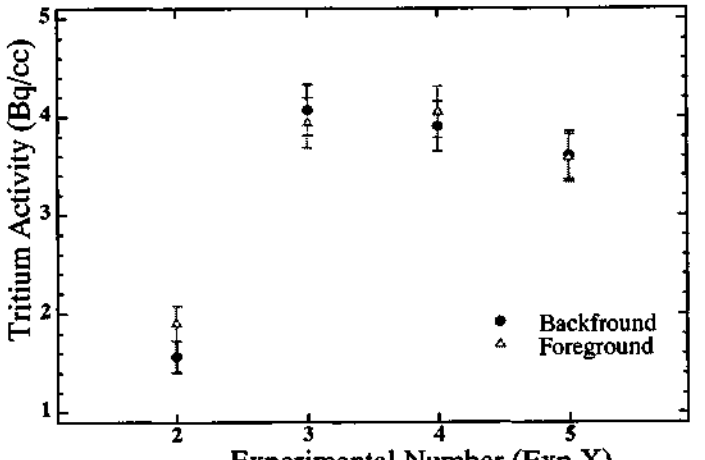


Fig.12 MCS data of Neutron (Exp.4)

Helium and Heat Correlation



Comparison of Background and Foreground for Tritium Activity (Exp.2-Exp.5)

4.Conclusions

In the run of Exp.4 and Exp.7 , excess heats up to approximately 4 to 5W were observed with very confident level. And D/Pd ratios there were observed to be almost constant in spite of drastic change of current . However , during the excess heats , nuclear products (neutrons) were not observed , and clear increases of helium-4 and any increase of tritium were not observed either . As regards to mass spectrum analysis , because it was necessary to calibrate correctly mass spectrometer and fix quantity of helium-4 , the generation of helium-4 could not be definitely concluded although visible increases of helium-4 peaks were observed for three cases of upper-cell gas . Because of limited number of experiments the correlation or non-correlation between the generation of excess heat and nuclear products could not be confirmed . Now the two cells in series connection are running simultaneously in order to establish the more confident measurement system , to study the correlation of excess heat with the degree of loading ratio D/Pd using different pre-conditioned cathodes ,and hopefully to identify the correlation of excess heat and helium-4 generation .

References :

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