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EXPERIMENTAL CORRELATION BETWEEN EXCESS HEAT AND NUCLEAR PRODUCTS

A. TAKAHASHI, T. INOKUCHI, Y. CHIMI, T. IKEGAWA,
N. KAJI, Y. NITTA, K. KOBAYASHI and M. TANIGUCHI
Osaka University, Yamadaoka 2-1, Suita, 565 Japan

Abstract

A comparator of twin system was developed to study possible correlation between observed excess heat phenomenon and nuclear products. Simultaneous on-line measurements were done for foreground(Pd cathode) and background(Ni cathode) cells to monitor input/output powers, neutron spectra and X-ray spectra. Slight (5-7%) excess powers were observed with 99 % confidence level, only for Pd-cathode-cell, with weak neutron emission in the energy over 3 MeV. Burst events by X-ray detectors were analyzed.

1. Introduction

The aim of this work is to study experimentally the possible correlation between "excess heat phenomena" observed by cold fusion (CF) experiments with heavy water electrolysis$^{1,2)}$and theoretically modeled deuteron-related nuclear reactions in metal/deuterium systems$^{3)}$. However, it is difficult to establish the methodology for studying the correlation, because of the lack of reproducibility of the CF effect by experiments and also because of the lack of fully-reasonable theoretical models.

In this work, the authors have classified intrinsic conclusions of various theoretical models on nuclear or non-nuclear products, into 5 scenarios as mentioned in Section-2. The first scenario is the case of high energy charged particle emission as principal nuclear products and characteristic X-ray emission as the secondary products. Weak neutron emission with specified spectrum is of interest in the scenario. The second scenario presumes no high-energy charged-particle emissions, but does production of He-4 or other isotopes. Following two major scenarios, techniques and tools of measurement were prepared for calorimetry, neutron spectroscopy, X-ray spectroscopy and high-resolution mass spectroscopy.

Due to the "non-reproducible nature" of cold fusion phenomenon, simultaneous and parallel runs of test(foreground) and blank(background) experiments, using identical experimental set-ups except test materials (Pd vs. Ni, heavy water vs. light water, etc.), are needed to clarify in-situ excess power, X-ray emission and neutron emission taking place only for the foreground(FG) run. A comparator of twin system as mentioned in Section-3 was developed for this purpose. If we could observe any meaningful difference in FG and BG runs for output powers, X-ray and/or neutron emission from the comparator experiment running simultaneously for FG and BG systems, the "cold fusion" effect will become very reliable.

2. Major Scenarios

One clear thing that CF experiments since 1989 have clarified is that excess heat is not by known d-d fusion reactions, because of no observations of correspondingly intense neutron (2.45 MeV) emission. Therefore, if the excess heat were due to nuclear effect, we should have some kind of "new class of nuclear reactions in solids". So many theoretical models and ideas have been proposed[3)] to resolve this issue, e.g., lattice-induced fusion, virtual-neutron transfer reaction, lattice-induced multibody fusion, proton-induced fission, alkali-proton reaction, neutron hallow nuclear reaction, etc.. The authors group has proposed multibody fusion models with intrinsic nuclear products and particle spectra[4,5)]. At present, we do not know the ultimate working model. In spite of essential differences in models by different authors, it seems possible to classify the cases of resultant nuclear products, particle spectra and secondary reactions, into a small number of scenarios as shown in Table-1, since we find common factors in kind of particles, energies, secondary effects, chemical and material conditions, among the variety of intrinsic conclusions on "nuclear ash" by many different theoretical models.

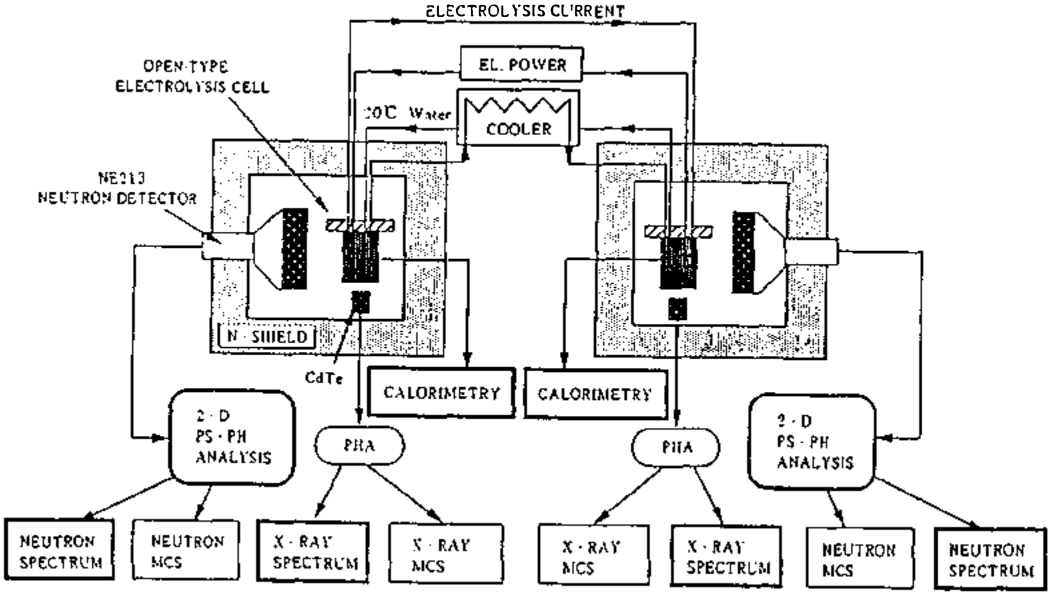
Scenario-1: Nuclear excited energy of "aneutronic" reaction is considered to be released, by anyway, as kinetic energies of charged particles (α, p, t, h, d) of reaction products. Usually we consider that the emitted charged particle energies are in 1-20MeV region, and therefore one watt excess power corresponds to 10^{11} to 10^{12} reactions per second. Primary high energy charged particles interact with lattice atoms (i.e., Pd and deuterons) and electrons via ionization-and-recombination, slowing-down with knocked-on atoms and cascade displacements, and so on, to deposit thermal (vibration)energy to lattice. In the process, X-ray emission (PIXE) and secondary neutron emission can take place. X-rays will be produced by the characteristic K_α and K_β processes, the electron(scattered by high energy charged particle) bremsstrahlung and the nuclear bremsstrahlung.

For example, if 5-10 MeV α-particles are major primary products, we look for 21-22.4 keV Peaks of K_α and K_β X-rays from Pd. The α-particles will dissociate deuterons in PdDx by D(α,n) reactions. There are also D(d,n) reactions by knocked-on deuterons. Secondary neutrons by these reactions should have continuous spectra in the 0-10 MeV region (main component in 3-5 MeV region). Neutron yields by these processes are estimated, by D(α,n) and D(d,n) cross sections and averaged slowing-down spectra of α and deuterons, to be 10^{-8} to 10^{-10} neutrons per α-particle. Therefore, when we see 1 watt excess power by the Scenario-1, we should have neutron yield of 10-10,000 n/source/sec from the cell.

Scenario-2: No neutrons and no high energy charged particles are produced in this scenario. We consider a special mechanism like "direct energy transfer from excited compound nucleus to lattice vibration"[3)] as proposed by Schwinger. Low energy charged particles (α = ^{4}He, h = ^{3}He) with less energy than 100 keV may be emitted by the primary reaction. Soft X-rays with exponentially decreasing intensity from several keV to about 50 keV may be only possible radiation from a cell. Production of secondary neutrons is negligible. Mass spectroscopy for ^{3}He, ^{4}He and other isotopes is the key tool of experiments. However, minor channel reaction by Scenario-1 might happen[4,5)] to yield very weak emission of neutrons and weak emission of high energy charged particles.

Table-1: Scenarios of correlation between excess heat and nuclear products

Scenario	Excess H.	Primary Reaction	Secondary Reaction	Measurements
#1	Yes	Several Mev Charged Particles (α,p,t,h,d)	D(α,n):(n/α)=ca.10^{-9} D(d,n):(n/d)=ca.10^{-6} X-ray:22.4keV for Pd	Heat,He-4 C.P.spectrum n-spectrum X-spectrum (n/t) ratio
#2	Yes	Nuclear-Lattice Energy-transfer Fission Low-Energy C.P.	Ionization Displacement Recombination Soft X-ray	Heat,He-4 Isotope shift Fission Products X-spectrum
#3	Yes	Chemical Mechanical	Fracto-fusion D(d,n)	Heat 2.45Mev n
#4	No	Very few Rad. n,gamma	negligible	n-spectrum gamma-ray
#5	No	None	None	None



Foreground (Pd cathode) Background (Ni cathode)

Fig.1: A twin system of CF experiment with heavy water electrolysis, calorimetry, 2-D neutron spectroscopy and X-ray spectroscopy

Other scenarios: In Table-1 we show all the scenarios under consideration. Scenario-3 draws a possibility of "exotic" chemical or mechanical process for excess power generation, probably associating weak emission of nuclear particles and radiations. Scenario-4 belongs to S. Jones' idea. Scenario-5 belongs to Morrison/Huizinga's idea, and it will be finally proved when every thing in the CF study is proved to be due to some kind of systematic errors.

3. Comparator of Twin System

A twin system of CF experiment was designed and made as shown in Fig.1, using two identically made open-type heavy water electrolysis cells with Takahashi type electrodes[6), two X-ray spectroscopy systems with CdTe detectors, two fast neutron spectroscopy systems with NE213 detectors, and two calorimetry monitors. One system serves for foreground (FG) runs, and the other does for background (BG) runs. In this work, the left cell for FG employs a Pd sheet cathode (25x25mm x 1mmt), and the right cell for BG does a Ni sheet cathode (25x25mm x 1mmt).

One power supply for electrolysis is commonly used for two cells and always the same electric current was supplied by the series connection from the FG to BG cell. Coolant (light water regulated to 20 +- 0.1 degree C by a chiller/heater) flows also by the series connection from the FG to BG cell. Two sets of electrolysis cell, CdTe detector (2x2x1 mm) and NE213 detector (13cmdiam.x5cmt) are installed in each of two cavities with 30x30x30 cm size, symmetrically made in a 1x2x1.5 m polyethylene pile for neutron shield.

The only difference between FG and BG systems is the difference in cathode material for the present work; i.e., Pd for FG and Ni for BG. Except this, conditions are set up to be same for electrolysis runs and on-line measurements of calorimetry, X-ray and neutrons. Therefore the twin system works as a "comparator" to detect on-line any meaningful differences in output powers and radiations between FG and BG runs.

Calorimetry: Difference of temperatures between inlet and outlet ports of coolant is as small as about 0.2 degree C for 100 watts joule heating by electrolysis, so that the flow calorimetry of the system[6) is not accurate enough to detect small amount of excess power. Instead, the calorimetry method using calibrated cell temperatures versus input joule heating powers can work with considerable accuracy (+- 0.65 watt for 50 watt input as discussed later) for a large dynamic range of input power variation (0-100 watt). Therefore, we employed the latter technique in the present work. Inner container of a electrolysis cell is made of 1mm thick pure silicate glass, and outer container for thermal insulation is made of 2mm thick polyacrylite glass. Room temperature was air-conditioned to keep 20 +- 0.5 degree C during the experiment. The upper parts of cooling coils made of silicate glass in the cells were covered with 30mmlong x 5mmt teflon pipes to isolate the effect of heat conduction change due to the change (20 mm max.) of D_2O level in the 4-5 days cycle for adding heavy water to the cells.

Calibration lines for FG and BG cells were obtained, using Ni cathode for both cells, for 5 steps (0-4 A) of electrolysis currents with 3 hours duration each step. Very straight calibration lines (input power vs. cell temperature) were obtained. Difference of calibration lines between two cells was small. Here, temperature data we retaken as average of two points, measured by two thermocouples for each cell, in which we set up a magnetic stirrer at the bottom for mixing and homogenizing electrolyte (heavy water with 0.2 mol/l LiOD).

X-ray spectroscopy: Considering the possibility of X-ray emission in 0-50 keV region by Scenario-1, a CdTe detector was chosen because of small size (2x2x1 mm) and relatively good energy resolution (+- 2 keV) in the energy region less than 80 keV. Using an Am-241 calibration source, capability of spectroscopy for Ex > 6 keV was ascertained. Attenuation of 20 keV X-ray by heavy water layer and cell wall (silicate) was also measured with the Am-241 source. Making a hole through the outer container of cell, we set the CdTe detector on surface of the inner glass wall, from where we had 1mm silicate glass plus 15 mm electrolyte layer reaching to the Pd cathode surface. By the experiment with Am-241, we found the attenuation of about 1/30 of 21-22 keV X-rays which were supposed to be emitted from the surface of Pd cathode. Taking into account the efficiency (including geometrical factor) of CdTe detector, we may detect 22 keV X-rays, if intensity is more than 10^8 photons per second. In the later part of present work, we introduced 2-dimensional pulse-shape vs. pulse-height analysis for CdTe signals, to reject completely noise signals which gave "bremsstrahlung-like" spectra with burst events in the early part of experiments. Time-evolution of X-ray counts in 15-25keV bin was monitored by MCS systems with 4 min time width, both for FG and BG runs.

Neutron spectroscopy: The n-γ pulse shape separation technique has been utilized from the beginning of our CF study since 1989[7]. In the present work, we always used two sets of 2-D pulse-shape vs. pulse-height analyzers for FG and BG runs, so as to separate completely γ-ray events and noise events from neutron events for several months long measurements, where the stability of NE213 system became important. Because of the 2-D analyses, we can extract also γ-ray spectral data any time we want, as well as neutron spectral data. The n-γ separability in the 2-D contour map was so good that we had a wide "count-zero zone" between the γ- and the neutron-contour. For MCS counting of neutron events, we set the low energy threshold of 2 MeV for FG run, and 3 MeV for BG run, while spectroscopies were done in the 1.5-7.0 MeV range of recoil proton energy.

The reason why we chose the D_2O/Ni cell (not light water / Pd cell) for BG runs is as follows: Background neutrons are mostly produced in materials near the NE213 detector and in the detector itself by the cosmic ray induced spallation reaction and D(γ,n) reaction which emit Maxwellian-like continuous energy neutrons with about 2 MeV nuclear temperature. Therefore, from the neutron detection point of view, heavy water gives more BG neutrons than light water. The present twin condition will give exactly the same neutron BG conditions on-line both for FG and BG runs.

Experiments were done for 1) cold worked Pd sheet (NHE No.1-CW) and 2) annealed Pd sheet (900 degree C, 1 hour annealing). Experimental procedure for NHE No.1-CW and other cathodes are shown in Table-2. The pulsed L/H mode electrolysis technique[6] was fully used in the present work.

To want to have information on D/Pd ratio in the L/H mode operation, we used a separate closed cell system[8] to measure in-situ D/Pd ratios as a function of electrolysis current.

4. Results and Discussions

D/Pd ratios measured by the closed cell mostly showed about 0.90 for the L-modes (0.5 A, typically) and around 0.80 for the H-modes (4 A, typically), drew maximum for 0.5-1.0 A current and decreased for higher currents than 1 A. However, exceptionally, some Pd samples (annealed) showed increasing D/Pd ratios for the higher currents to reach 0.95. For the L/H modes general, we speculate that a slowly varying dynamic condition of repeating absorption and desorption is realized to change D/Pd ratio around 0.85. Relation of excess heat and exceptionally high D/Pd at higher current for some samples is not clear at present.

Figure-2 shows the results of powers, X-rays and neutrons in the beginning 60 hours (broken line shows interpolation of data lost by the power-off accident of the Campus). No excess power and no excess neutrons were found. However, unusual burst events of X-rays for the FG (Pd) cell was recorded in 4 successive cycles of current-off intervals of electrolysis. Pulse height spectra corresponding to these bursts show exponentially decreasing distribution from 6 keV to about 20 keV; this spectrum can be typically that of electromagnetic noise signals, but it might have some true information of X-rays since we saw coincidences with the L/H mode cycles. Possibility of soft X-ray emission by the nuclear bremsstrahlung by low energy charged particles (e.g., 46 keV α -particles[5)]) should be studied in the future work.

In the following 150 hours (60-210 hours) after Fig.2, we saw no excess power for both of FG and BG runs, and no excess X-ray counts above natural BG level, though we could recognize slight increase (10-20 % at most over BG level) of neutron MCS counts and spectrum of FG run had a "structure" over 3 MeV, while BG(Ni) or cosmic neutrons has no such structure. After 18 days, we started to see a trend of excess power in the FG(Pd) cell and the trend continued for about 260 hours. Fig.3 shows the results of input/output and excess power levels for FG and BG runs, for the last 80 hours of this period. Clear excess power was observed for the FG(Pd) cell, while the BG(Ni) cell showed only fluctuated data around the "zero" excess power line. No 21-22 keV characteristic X-rays were detected by the CdTe spectroscopy in the period, though broad peaked spectra in 6-30 keV region with burst events were sometimes observed for both of FG and BG runs. Neutron (exactly speaking, recoil proton) spectrum had typical structure over 3 MeV but MCS counts were near background, while the natural BG spectrum taken with the same detector in 2 weeks after stopping electrolysis had no such structure as shown in Fig.4.

It took about an hour to reach equilibrium temperature after changing the current mode as we saw in the calorimetry calibration runs. So, we omitted calorimetry data for the beginning 1.5 hours after the mode change, and we sampled up data for later time interval than 1.5 hours, for the discussion of excess power generation. To estimate experimentally the total error width covering both of random and systematic errors, we took statistics of power balance for all the "excess power" data for BG(Ni) runs (including Ni data for annealed Pd cathode too): The results are shown in Fig.5, where solid curve shows Gaussian normal distribution with the standard deviation σ = 0.65 watt. This σ is regarded as the overall error of the present calorimetry during the long experimental period (few months), assuming the Ni cathode cell (BG cell) has no excess heat emission. The distribution of excess power data for NHE No.1-CW, compared with BG(Ni) are shown in Fig.6. Excess power data exceeding the 99 % confidence

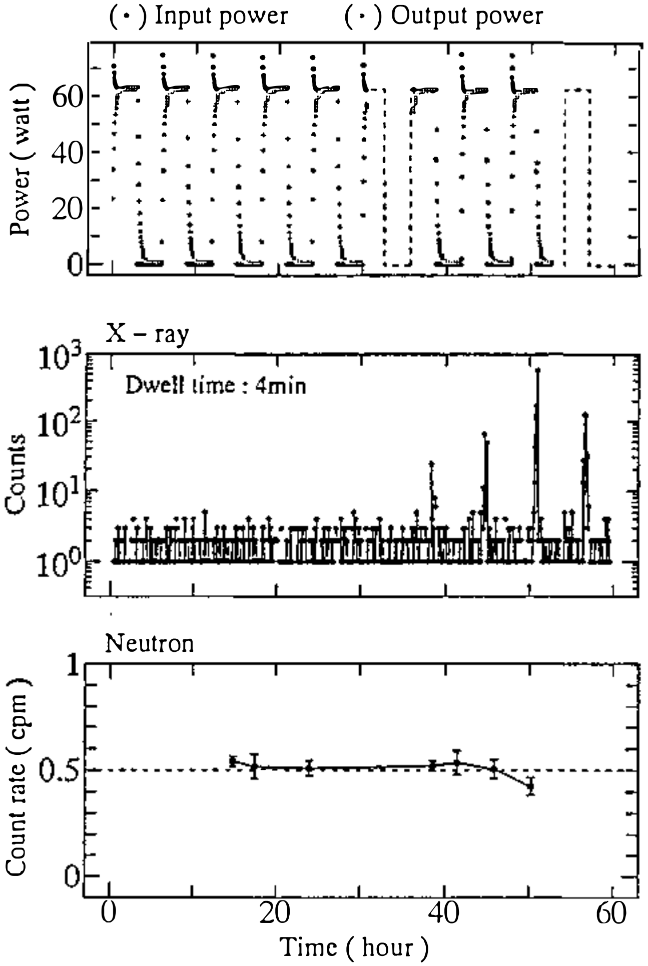


Fig.2: Results of FG run in the beginning 60 hours, for input/output, n and X–ray

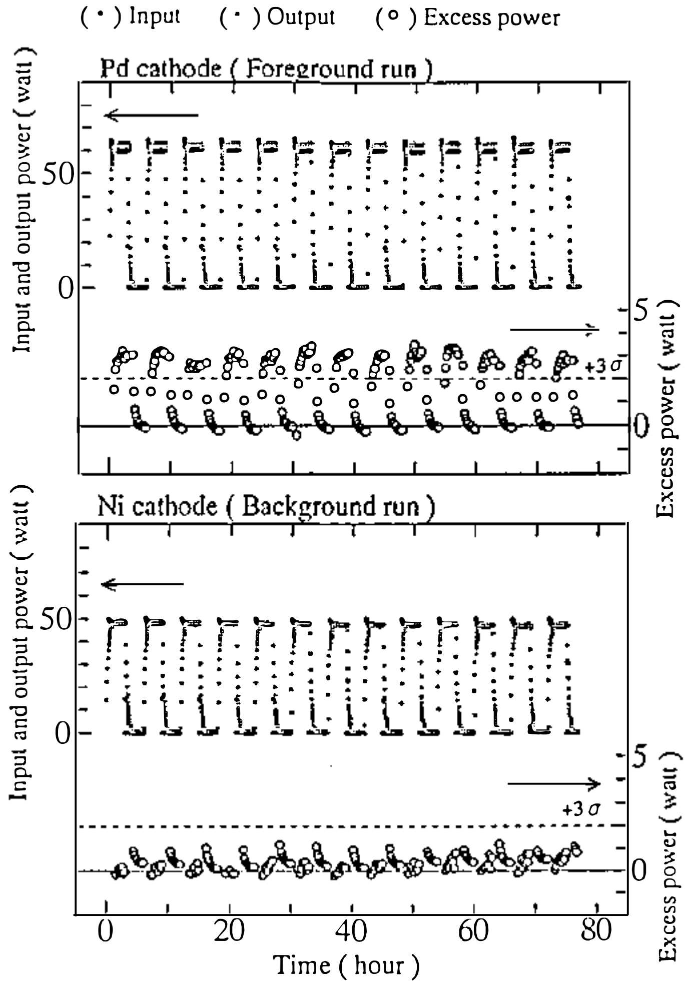


Fig.3: Results of FG and BG runs in the later period of experiments, for excess power

Table-2: Procedure of elctrolysis experiments

Date	Electrode		Mode	Mode
'94~'95	Pd	Ni	(hour)	Current
10/18~ 10/25	Tanaka 2nd batch	Nilaco No.1	On/Off (3-3)	4.5A/0.0A
10/25~ 10/28	〃	〃	On/Off (12-12)	4.5A/0.0A
10/31~ 11/3	NIIE No.1 Cold Work	〃	On/Off (3-3)	4.5A/0.0A
11/4~ 11/10	〃	〃	On/Off (3-3)	4.5A/0.0A
11/10~ 11/17	〃	〃	Low/High (6-6)	0.5A/4.5A
11/18~ 11/28	〃	〃	Low/High (3-3)	0.5A/3.0A
11/29~ 12/2	〃	〃	On/Off (3-3)	3.0A/0.0A
12/2~ 12/6	〃	〃	Saw-Tooth (20 min.)	0.5A~3.0A
2/7~ 2/13	NIIE No.2 Annealed	〃	On/Off (3-3)	4.5A/0.0A
3/10~ 3/17	〃	〃	Step mode (3)	—

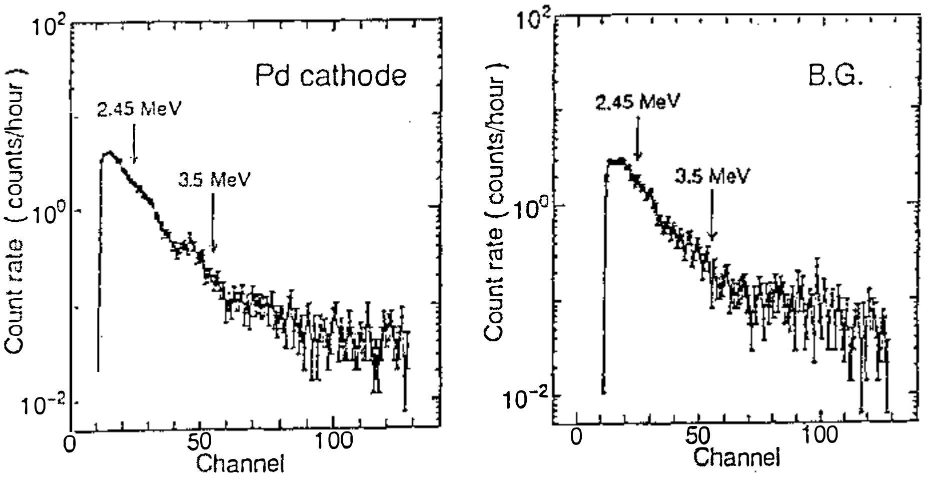


Fig.4: NE213 recoil-proton spectra, correspoding neutron spectra by unfolding, for FG(Pd) electrolysis run (left figure) and BG (elctrolysis off) run (right figure)

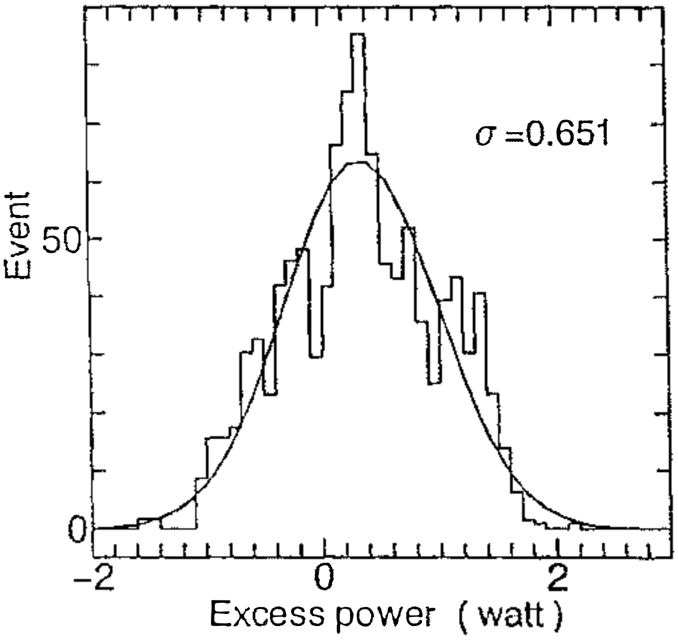


Fig.5: Distribution of power-balance data for BG(Ni) runs

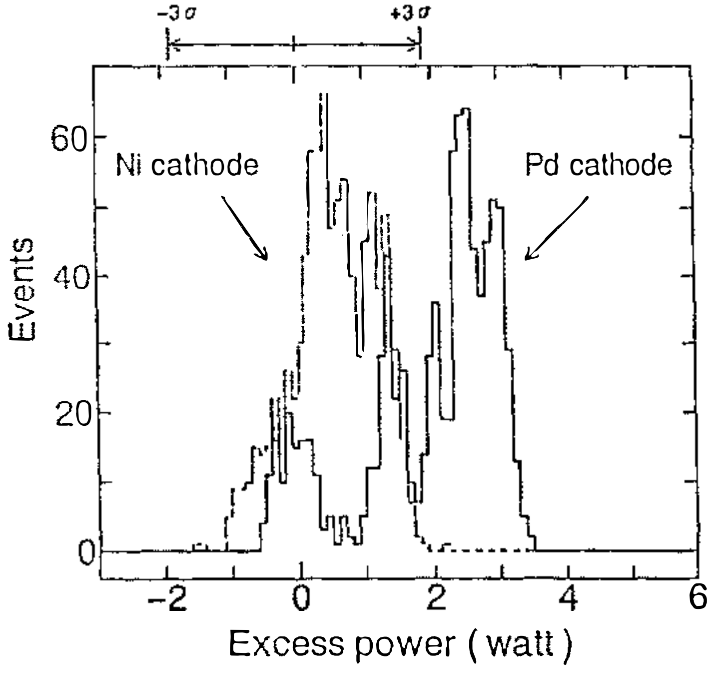


Fig.6: Distributions of excess power (power balance) data, compared for FG(Pd) and BG(Ni) runs

level (3 σ = 3x0.65 = 2.0 watt) are very clearly seen for the FG(Pd) cell. We had 2.3 to 3.5 watt excess power. However, excess ratio (output/input - 1.0)x100 is only 5-7 %.

The broad-peaked X-ray spectra frequently observed as bursts in 6-30 keV region was proved to be due to some noise (non-X-ray) events, based on the 2D analysis (pulse shape vs. pulse height) for CdTe signals, in the later part of the present experiment. The second series experiment for annealed Pd cathode was done using the gated condition for X-ray contour of 2-D X-ray analysis, and upto now no such broad peaked spectra in the 6-30 keV region were observed. The experiment with annealed Pd cathode is still under data processing, however we can say that a trend of excess power only for the FG(Pd) cell is also being observed.

5. Conclusion

To study the origin of excess heat and correlation with nuclear products, experimental works with a newly designed twin systems for calorimetry, X-ray and neutron 2-D spectroscopy were carried out. The twin system was proved to be very reliable and useful for the correlation study.

We observed 5-7 % (2.3-3.5 w) excess power beyond the 99 % confidence level only for the heavy water electrolysis cell with Pd cathode, while the reference heavy water cell with Ni cathode running simultaneously (in parallel with the FG run) gave no excess power data within error band 1 σ = 0.65 watt. Slight indication of neutron emission in spectra over 3 MeV was sometimes observed, but it was not tightly correlated to excess power generation, and neutron emission rates were very weak as 1 n/source or less. No characteristic X-rays, 21-22 keV for Pd were observed during the present work, though unusual burst events with continuous spectrum under 20 keV was once observed for the FG(Pd)-cell run.

We might conclude that Scenario-1 was denied because we did not see 21-22 keV X-ray when we saw excess power for the FG(Pd) cell. However, 1 mm thickness of Pd cathode plate can largely attenuate the characteristic X-rays if produced at the central zone and we should estimate scattered X-ray spectra in that case. The level of observed excess heat is still weak, so that we might not reject unforeseen systematic errors. Therefore, to draw definite conclusion on Scenarios, we need further a number of experiments with different cathode materials and different electrolysis conditions so as to statistically assure the excess heat, X-ray and neutron data.

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