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Study of Concentration of Helium and Tritium in Electrolytic Cells with Excess Heat Generation

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ABSTRACT

Concentrations of helium and tritium in gas and liquid phases in calorimetric cells with excess heat generations were measured. Values of factor F (nuclear ash) defined as ratio of amount of heat released by particular nuclear reactions to the excess heat, were calculated to be $F(\text{He in gas phase}) \approx (4 \pm 16) \times 10^{-3}$ and $F(\text{T in liquid phase}) \approx (8 \pm 6) \times 10^{-10}$, and upper limit of $F(\text{T in gas phase}) \leq 1 \times 10^{-6}$. These extremely small values suggested either that (1) the nuclear reactions might occur in deep inside of the Pd cathode, or (2) the major amount of the detected excess heat could not match with the heat expected from the nuclear reactions if the reactions occurred on the surface of Pd cathodes.

1. INTRODUCTION

It is well known that excess heat generations are found in many Pd/D₂O systems.¹ Searches for nuclear reaction products (proton, neutron, gamma-ray, tritium, helium, etc.) have been started by many groups²⁻⁴ considering in minds that the amounts of the excess heat can not be explained by the chemical reactions but by the nuclear reactions. However, the origin of the excess heat generation is still in dispute. It is interesting to keep searching for the nuclear products of helium and tritium in gas and liquid phases of the calorimeter cells with the excess heat generation.

2. EXPERIMENTAL SETUP

The calorimetric systems were employed to observe the excess heat, and the helium and tritium atoms in the gas and the liquid phases. The whole system was composed of the calorimetric cell with a lower gas sampling port, a catalyzer glass tube with a recombined D₂O sampling port, and of a gas volume meter with an upper gas sampling port. They were connected each other by silicone rubber tubes and are schematically shown in Fig. 1.

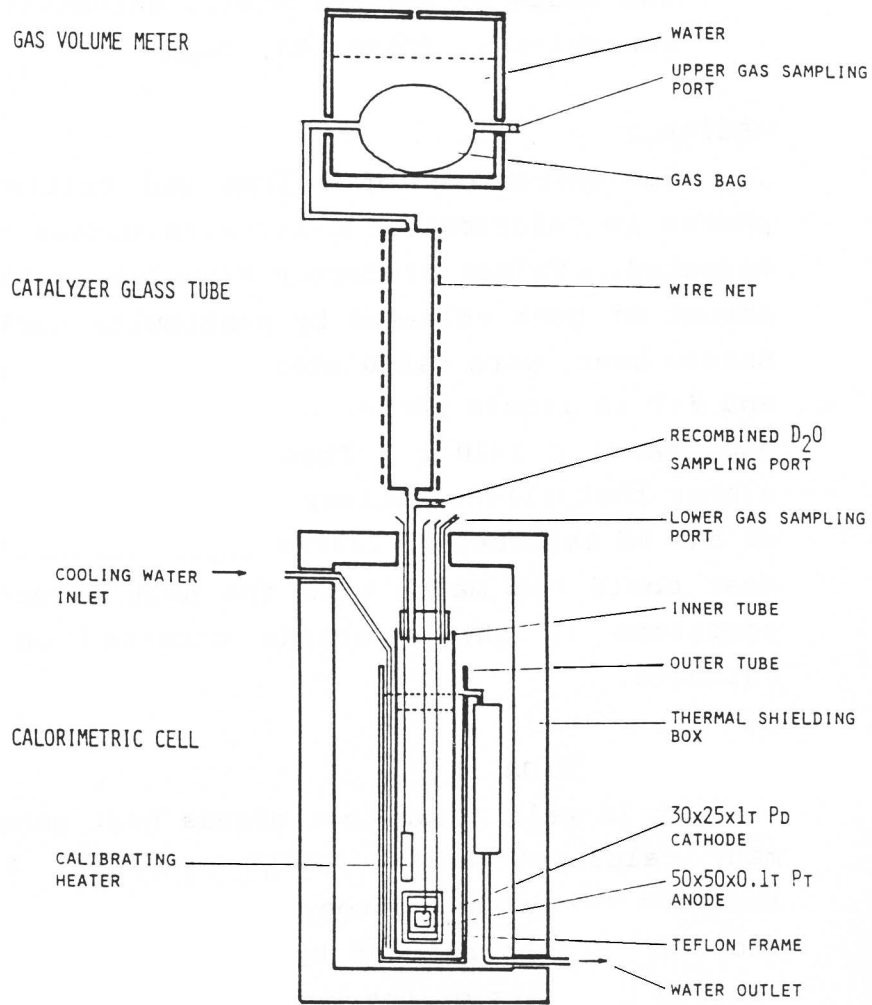


Fig. 1. Schematic diagram of the softly closed calorimetric cell.

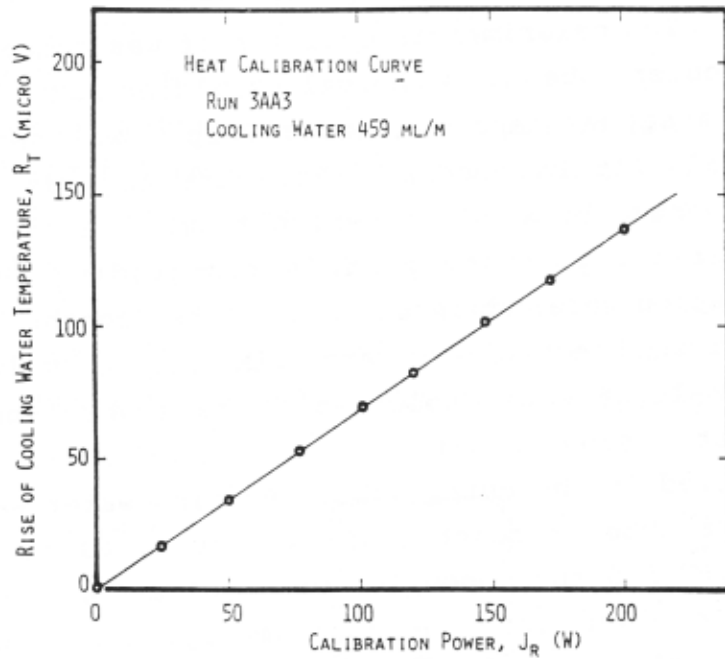


Fig. 2. Typical heat calibration for calorimetry.

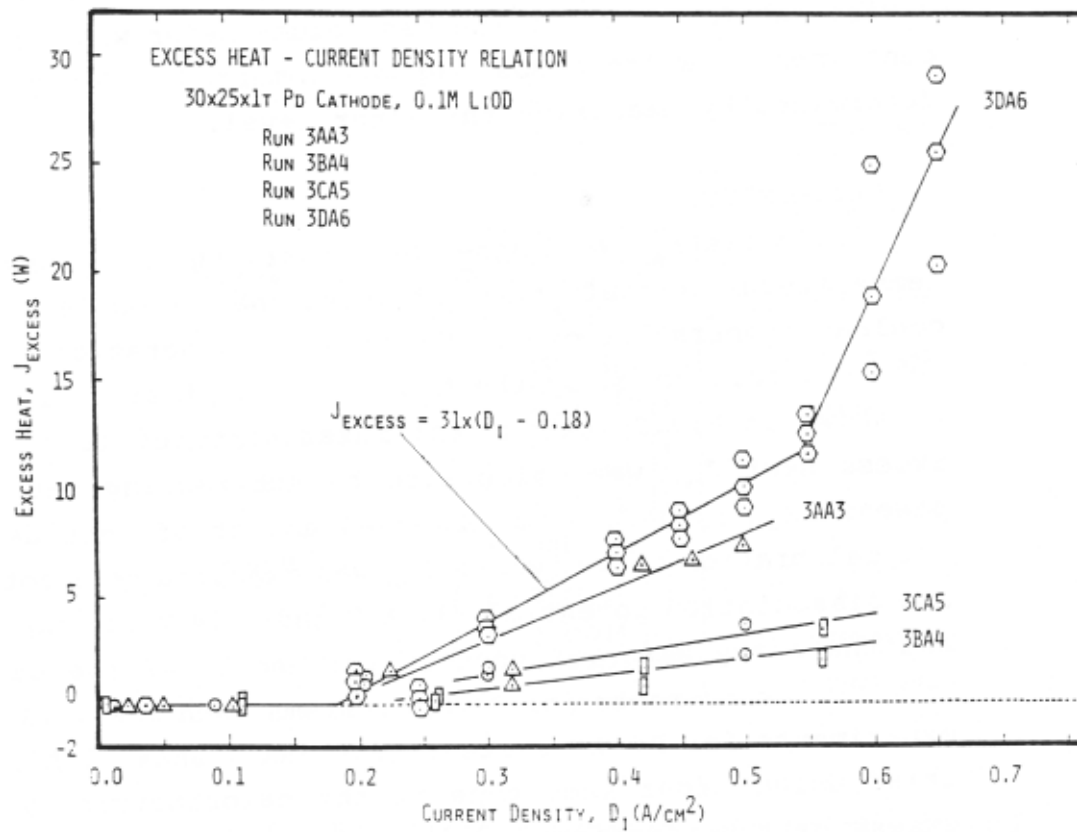


Fig. 3. Excess heat generation vs. current density.

The calorimetric cell itself was composed of an inner tube, an outer tube and a thermal shielding box. A cathode of $30 \times 25 \times 1$ t Pd sheet (Tanaka Kikinzoku Corp.) was sandwiched between two $50 \times 50 \times 0.1$ t Pt anodes. Both the cathode and the anodes were supported by a 10 mm teflon frame. A calibrating heater, the lower gas sampling port, a gas guiding tube and electrolytic solution were put in the inner tube together with the cathode and anodes. Electrolytes were 0.1M LiOD heavy water solution or 0.1M LiOH light water solution. The inner tube was shielded by a plastic stopper with a gas guiding tube. The inner tube was settled in the outer tube. Running water was introduced to the outer tube at constant flow rate to cool down the inner tube. Two thermocouples were put at an inlet and an outlet of the coolant for the calorimetry, which was monitored by a personal computer. The outer tube was hold in a thermal shielding box with heat insulating materials. The catalyzer glass tube contained platinum black powders and was wrapped in a wire net for safety. The gas volume meter was a plastic water container in which a gas bag was immersed. The gas volume was determined by measuring the water level.

3. CALORIMETRY

Calorimetry was made by observing differences between temperatures of coolant at the inlet and the outlet. Rise of the coolant temperature due to Joule power generations in the inner tube was calibrated by the heater. A typical calibration curve is shown in Fig. 2 with an estimated error of less than ± 0.5 W. Excess heat J_{ex} , was calculated by subtracting net input Joule power, $(E_{cell} - E_{diss}) \times I$, from observed amount of heat determined by the calibration curve, where E_{cell} was measured cell potential, E_{diss} was the dissociation potential (1.48 V and 1.54 V for the light water and the heavy water, respectively) and I was the cell current.

Two experiments for Pd/H₂O systems (Run 3AA7, 3BA8) and four experiments for Pd/D₂O systems (Run 3AA3, 3BA4, 3CA5, 3DA6) were tried using four same type of the calorimetric systems. The excess heat generations were found not for the two H₂O systems but for all the four D₂O systems. A small amount of the excess heat was dependent on densities of the cell currents D_I as shown in Fig. 3. One can see a threshold current density of about 0.18 A/cm². For Run 3DA6, the excess heat generation is given roughly as $J_{ex}[W] \approx 31 \times (D_I[A/cm^2] - 0.18)$.

4. HELIUM GAS

Concentration of helium gas was measured by a gas chromatographic method during electrolysis. A small volume of molecular sieve 5A grains were packed in two 3 mm 6×10 m long stainless steel pipes. Argon carrier gas was led to the pipes at constant flow rate of 16 ml/min. Gas samples of 1 ml were ejected by a syringe into the one of the pipes. Disturbances due to helium and other impurity gases were sensed by a thermal conductivity detector, whose signals were stored by a recorder as a function of retention time. Gas sampling was made at the upper gas sampling port of the volume meter by extracting the inner gas in the bag into the syringe.

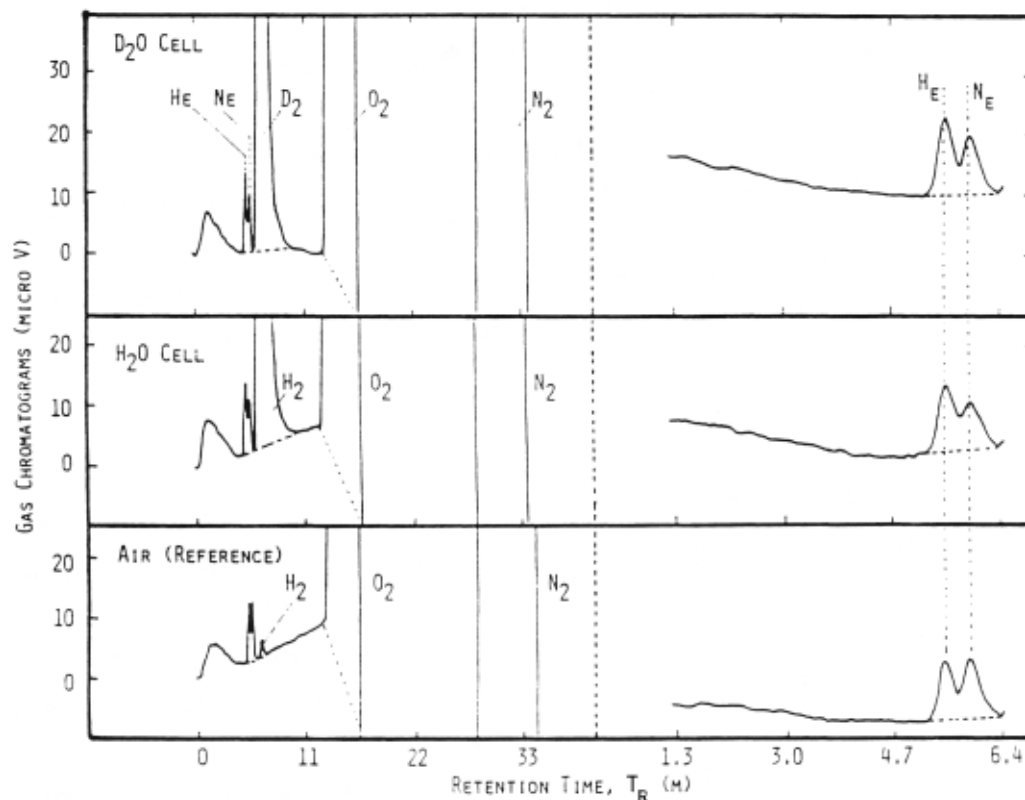


Fig. 4. Gas chromatograms for the D_2O cell with the excess heat generation, the H_2O cell without the generation and for the air as reference.

Measurements were done for the one D_2O cell with the excess heat generation of (4.8 ± 0.5) W and for the two H_2O cells without the generation. Fig. 4 shows typical gas chromatograms for the D_2O cells and the H_2O cells, and a chromatogram of air in the experimental room, which played a role of concentration references (He 5.24×10^{-4} , Ne 1.81×10^{-3} , O_2 20.9, N_2 78.0 volume %). Taking the measured helium gas concentrations and the gas volume determined by the volume meter into account, production rates (He atoms/sec) were calculated as $(4.5 \pm 1.5) \times 10^{10}$ for the D_2O cell and $(4.1 \pm 1.5) \times 10^{10}$ and $(3.9 \pm 1.5) \times 10^{10}$ for the two H_2O cells. Therefore, assuming the excess heat generation had relation with the helium production, the net helium production rate for the (4.8 ± 0.5) W excess heat was $(0.5 \pm 2.1) \times 10^{10}$ He atoms/sec.

5. TRITIUM WATER

Samples of 1 ml recombined heavy water were collected at the sampling port of the calorimetric D_2O cell with the constant excess heat generation of (3.4 ± 0.5) W for every 16 days. Concentrations of the tritium in the samples were measured by liquid scintillation counting method. A net increase of (1.0 ± 0.7) dpm/ml in disintegration rates over the 16 days was found after collecting long term drifting of the scintillation counter. This value led to $(5.6 \pm 3.9) \times 10^9$ T atoms in 600 ml electrolyte. Therefore, tritium production rate under the condition of (3.4 ± 0.5) W excess heat generation was $(4.0 \pm 2.8) \times 10^3$ T atoms/sec.

6. TRITIUM GAS

A proportional gas chamber was developed to measure energy spectrum of beta-rays emitted from the tritium atoms. The chamber consisted of an anode with five $20 \mu\text{m}$ sense wires and six

100 μm potential wires, and of two cathodes of 100 μm mesh. The effective area was 50 mm \times 140 mm and the thickness was 10 mm. The chamber was shielded by lead blocks and surrounded by aluminum and lucite plates. Pulse signals from the sense wires were amplified and were accumulated by a pulse height analyzer (hereafter PHA). The signals due to cosmic rays were rejected by anti-coincidence plastic scintillation counters which covered the surface of the chamber. Energy calibration spectrum was measured using 5.9 keV X-rays from Fe source, and is shown in Fig. 5 with a background spectrum.

The tritium concentration in gas phase of Run 3CA5 D₂O cell with the excess heat generation was measured. The gaseous sample (20 ml) was taken from the lower gas sampling port of the cell. It was put into the chamber, after the counter gas (Ar + 10% CH₄) flow was stopped. Successive 600 sec spectrum measurements were performed 5 or 6 times, after readjustments for small gain downward shift were made by increasing the bias with the help of the ⁵⁵Fe X-ray, and these spectra were finally summed up.

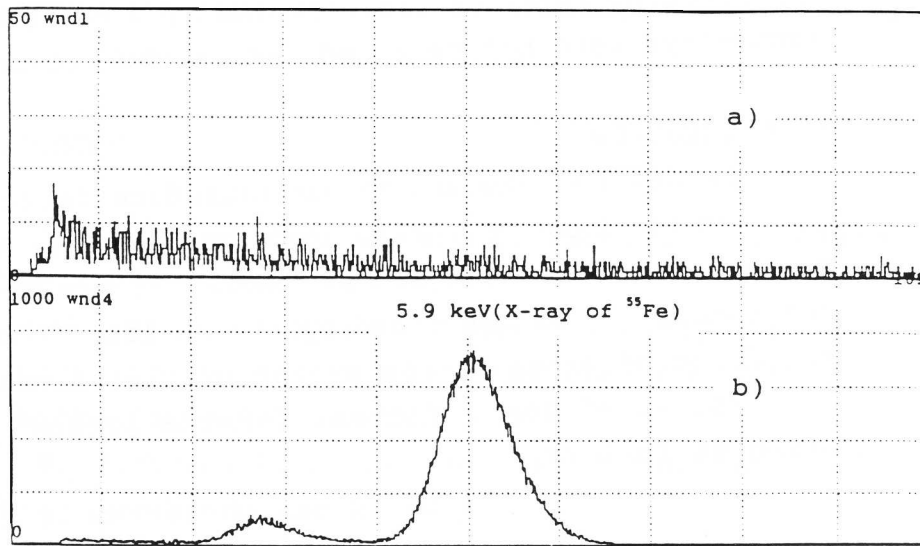


Fig. 5. Spectra of background (a) and X-ray of ⁵⁵Fe (b).

The energy spectra were integrated in the range from 0.2 keV to 12 keV to obtain averaged counting rates. Taking detection efficiency of 0.5 into account, the disintegration rate was (0.90 ± 0.06) dps/20 ml.

For comparison with this concentration, similar measurements were done for the H₂O cell without the excess heat generation, and for pure D₂ gas, pure H₂ gas and the air. However, quite similar values as for the D₂O cell were obtained for these references.

Therefore, subtracting the values of the references, the expected net disintegration rate of the D₂O cell was calculated to be (0.00 ± 0.08) dps/20 ml, where the excess heat generation was (3.5 ± 0.5) W and the cell current was 7.5 A ($D_1 = 0.5$ A/cm²).

7. NEUTRON AND GAMMA-RAY

Measurements were made for fast and slow neutrons and gamma-rays by using a 5 cm diam. \times 5 cm long liquid scintillation neutron detector, a ³He gas neutron detector and a 5 cm diam. \times 5 cm long NaI(Tl) gamma-ray detector, respectively, along with the PHA system. The detection

experiments were done for the D₂O cell with the excess heat generation of (4.8 ± 0.5) W and also for the same cell without the generation at the current density less than 0.15 A/cm^2 . Since detection counting rates for the both experiments agreed within statistical errors, net increases in the counting rates of the fast and slow neutrons and also of the gamma-rays were not observed during the excess heat generation.

8. DISCUSSION

It may be interesting to introduce factor, F(nuclear ash) to discuss the relation between the quantities of the nuclear ashes and the excess heat. It was defined as ratio of the amount of heat released by the nuclear reactions relevant to the detected ashes to the amount of the excess heat. Values of the F factors for the present experiments were given as follows,

$$F(\text{He in gas phase}) \approx (4 \pm 16) \times 10^{-3}$$

$$F(\text{T in liquid phase}) \approx (8 \pm 6) \times 10^{-10},$$

and upper limit of $F(\text{T in gas phase}) \leq 1 \times 10^{-6}$.

In these calculations, the postulated nuclear reactions were taken as



and $\text{D} + \text{D} = {}^3\text{T} + \text{H} + 4.03 \text{ MeV}.$

Comparing with the factors, it seemed the factor for helium was larger than that for the tritium productions. However, there still remained a possibility that the increased helium concentration was made not by the nuclear reaction but by some kind of helium pumping processes, because the productions were also observed in the same order for the H₂O cells without the excess heat generations. Such processes were assumed to be a thermal transpiration process and a helium leakage process.

Anyhow, the extremely small values of the factors could not explain the amounts of excess heat.

However, it was noteworthy that, if the nuclear reactions were to be made inside of the Pd cathode, residual helium and tritium atoms in the cathode could hardly escape out of the cathode to the gas and the liquid phases so that the meaningful amounts of helium and tritium in these phases were not detected, as shown in the present experiment. Therefore, it should be concluded the small F values suggested either that (1) the nuclear reactions might have to occur in the deep inside of the Pd cathodes, or that (2) the major amount of the detected excess heat could not match with the amount of the heat expected from the nuclear reactions, if the reactions occurred on the Pd cathode surfaces.

9. REFERENCES

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