Nuclear Physics Approach

Search for tritium in Pd + D systems by a gas proportional chamber

Nobuharu Yoshikawa

Institute for Nuclear Study, University of Tokyo, Tokyo,188,Japan

Takayohi Aoki, Yoshitugu Kurata and Hiroshi Ebihara

Isotope Center, University of Tsukuba, Tsukuba,Ibaragi,305,Japan

Kenya. Mori

Technical Center, Tanaka Kikinzoku Kogyo K.K., Hiratsuka,kanagawa,254,Japan

Abstract
Tritium concentration in deuterium gas from Pd + D systems was measured by a gas proportional chamber. This type of detector was able to observe directly a beta ray spectrum of tritium and give a genuine number of tritium in studying gas phase system. For this purpose, the gas proportional chamber was made and was operated in low background. The detection limit was 20 Bq in one liter of deuterium gas. Tritium searches in deuterium gas phase of several different systems were done by this detector. The clear spectrum of beta ray originated from tritium decay could not be observed due to low concentration of tritium in the examined deuterium gas phase.

1.Introduction

There were several reports[1,2,3] to claim the generation of tritium in the gas phase of Pd + D systems. In the reports, the methods used for tritium detection were the ion chamber or the mass analyzer system. But in those measurements, some possibilities to measure other kinds of events could not be excluded, so that the different type measurements to confirm the generation of tritium caused by nuclear reaction were essential. If the measurement of beta ray spectrum from tritium disintegration could be possible, it would give the direct evidence of existence of tritium. The gas proportional chamber was made for this purpose and could measure the energy spectrum of beta ray of tritium. Tritium concentrations in deuterium gas released from Pd and Titanium alloy powder were studied to search for the evidence of cold fusion. The detector of this type was used in our laboratory for the tritium searches in the gas phase of the electrolysis cells, in which the generations of excess heat were observed[4]. In this work, the detection limit was confirmed by the calibrated tritium gas. During operation of the systems, neutron measurements were done by ^{3}He counters.

2.Detection method of tritium

(a) Detector and measuring method

The chamber was formed by two cathode planes of 100 μm mesh and an anode plane with five 20 μm sense wires and six 100 μm potential wires. The spacing between sense and potential wires was 5 mm. The gap between cathode and anode was 5 mm. The sensitive area was 50 mm × 140 mm and the thickness was 10 mm. The chamber was shielded by aluminum and copper plates and lead blocks. Cosmic rays were suppressed by plastic scintillation counters which were placed just above

the chamber to cover the whole surface.
Pulse signals from the sense wires summed to one output were amplified and fed to a pulse height analyzer. Digitized signals were stored in a personal computer and analyzed by the KODAQ code[5]. The signals due to cosmic rays were rejected by anti-coincidence technique. Sampled gas was put into the chamber by a syringe. It was confirmed that the hydrogen gas didn't disturb the energy resolution of chamber but higher voltage was needed. The counter gas was Ar + 10%CH_4.

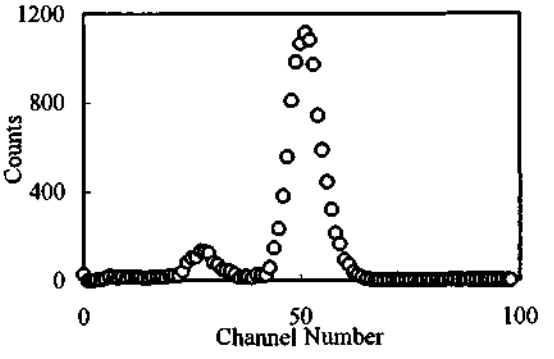

Fig.1 X-Ray spectrum of ^{55}Fe

(b) Energy and efficiency calibration
The energy calibration was done using 5.9 keV X ray from ^{55}Fe source. As the window of the chamber was a thin plastic film, X-ray could penetrate it. The X-ray spectrum was shown in Fig.1. There were the 5.9 keV full energy and 3.0 keV escaped peaks and both peaks were used for the energy calibration.
The calibration of detection efficiency was done by several different volumes of deuterium gas which contained a little tritium gas.
The deuterium gas used for the calibration was obtained by the electrolysis of heavy water in which a very small amount of tritium water was added.

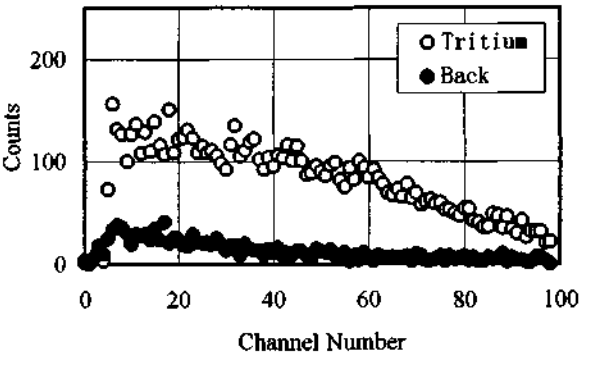

Fig.2 Tritium and Background Spectra

The tritium spectrum was shown in Fig.2, together with background spectrum. Counts of detector as a function of input gas volume were shown in Fig.3. The linearity of counts to input gas volume was fairy good. The tritium concentration in deuterium gas was estimated in the following way; the deuterium gas was once more reduced to the heavy water using a Pt black and measured by a liquid scintillation counter. Using this value, the efficiency was estimated to be 42 ± 8 %.

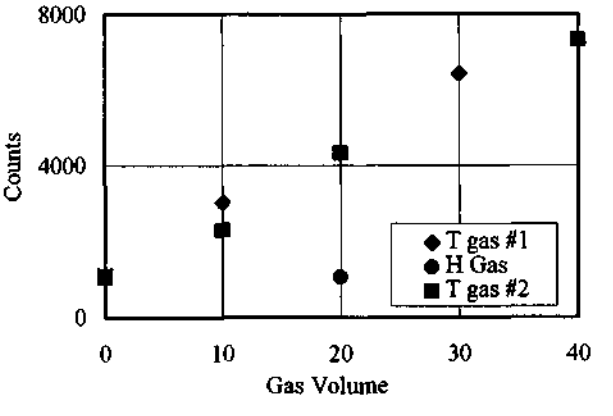

Fig.3 Tritium counts per input gas volume

3.Detection method of neutron
The neutron monitors were set near the Pd + D systems. Two ^3He counters(Model Rs-P4-0810, Reuter-Stokes Inc) were used. Each counter was surrounded with a cylindrical plastic moderators. Output signals of counters were digitized and recorded in the personal computer.

4.Tritium search
(a) Deuterium gas from palladium wires in gas phase
Palladium wires of 2 mm diameter and 10 cm length were used. The wires were annealed at 800 °C

in a vacuum for 12 hours, and the surfaces of wires were modified by a method of sand blast. Those were set in a vacuum duct(3.5 cm diameter and 21 cm length) which was connected with a standard volume of 2 liter. After evacuation of the duct, deuterium gas was supplied from the standard volume. The maximum pressure was 3 atm. Normally, after one week, D/Pd loading ratio; n = 0.65 was achieved. Then wires were heated up to 230 °C by a heater which was wound on the outside of the duct. Within 10 minutes, the temperature rose and gas released. After holding the high temperature for several tens of minute, the system was cooled down to the room temperature. The relation of temperature and pressure was shown in Fig.4. Hereafter this type experiment was referred to as external heating. The gas samples were taken by a syringe through a gas sampling port at the high temperature and the room temperature. After injection of the sample gas into the chamber, a measurement was done for 10 minutes. The gain of the chamber was checked by the X-ray source before and after the measurement. This measurement was repeated 4 times.

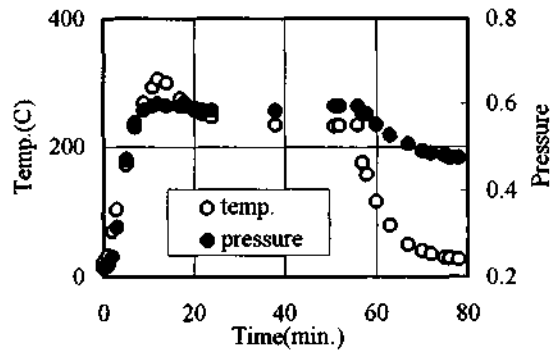

Fig.4 Temperature and Pressure

In the Pd + D system, a cyclic absorption and desorption was tried but the complete cycle was not achieved. Titanium alloys were also used for hydride material in the same setup of the palladium wire. Several absorption and desorption cyclic processes were succeeded. Titanium alloys were mechanically powdered to dimensions less than 100 μm and heated up to 300 °C in a vacuum for 12 hours. After cooled down to the room temperature, deuterium gas was filled up to 3 atm. It was heated up to 230 °C for desorption and cooled down to 0 °C for absorption. This cycle was repeated several times. The D/Pd ratio; n was 0.3 - 0.4. Compared with the palladium wires, this value was not high.

(b) Deuterium gas from Pd wires loaded by the electrolysis

In the gas phase experiment(external heating), there were two disadvantages, one was that the loading ratio was not so high, another was that long time was needed for heating, because the heater was wound on the outside of the chamber to avoid the existence of any materials in the gas phase. To improve these two points, deuterium gas was loaded in Pd wire(28 cm length,2 mm diameter)by a electrolysis. The wires were coated with thin Au layers by electroplating, before

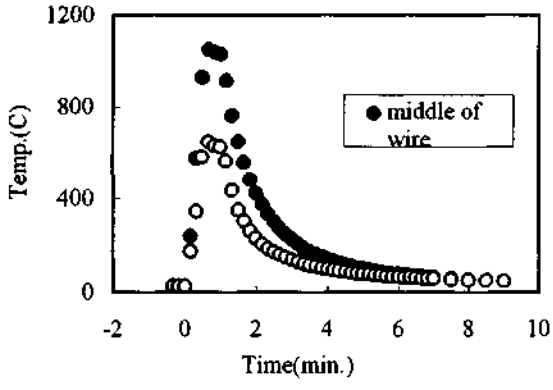

Fig.5 Temperature of Pd wire

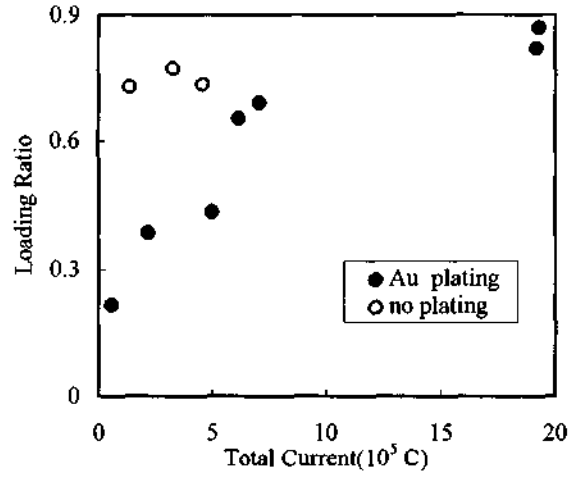

Fig.6 Loading ratio by electrolysis

and after the electrolysis. Those wires were set in a vacuum duct and high currents (first 1 min.,80 A and next 1 min., 60 A) were supplied to the wires. The temperatures at center of wire(closed circles) and near the electric contact(open circles) were shown in Fig.6. Hereafter this type experiment was referred to as internal heating. The wire was heated up to 1000 °C, then deuterium gas was abruptly released within 35 seconds. The released deuterium gas from the wire was stored in a balloon which was immersed in a water vessel. By the water level, the exact gas volume was known. Deuterium gas was sampled from the balloon and measured by the chamber in the same way of the external heating experiment. In Fig.6, loading ratios were plotted as a function of total current which was used in the electrolysis. Closed circles were those of gold plating and open circles were no plating . Thickness of gold was from 0.026 μm to 1.16 μm. After electrolysis, further gold was plated in the 5 wires of total 7 wires with thickness of about 1 μm.

5.Summary of measurement

(a) Tritium

Many gas samples were collected from the external and internal heating experiments. Those were measured by the chamber in the same manner . Before and after measurements of the sampled gas,

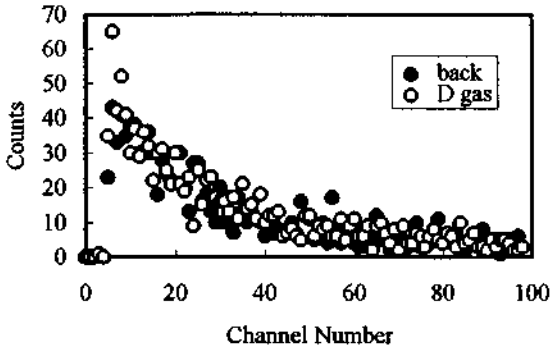

Fig.7 Spectra of sampled gas and background

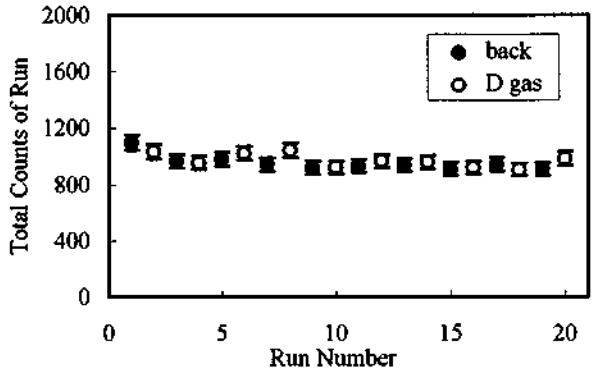

Fig.8 Summary of internal heating

background spectra were also measured. When sampled gas was input into the chamber by the syringe, the counter gas was stopped and the gain of the chamber was adjusted to the same as the background measurement by increasing the counter voltage. The measurement time for one run was 10 minutes. Before and after measurement of spectrum, the gain was checked by X-ray spectrum. This run was repeated 4 times, after measurement, the sampled gas was purged by the counter gas. Several background runs were taken after the

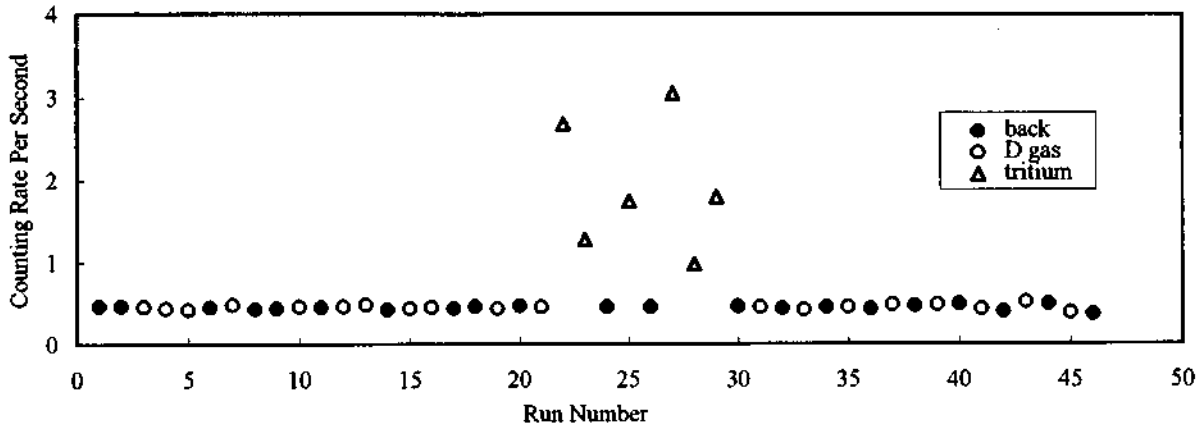

Fig.9 Summary of measurements

sampled gas was fully purged. The spectrum of sampled gas in the internal heating experiment was shown in Fig.7, together with background spectrum, where both spectra were the sum of 4 runs. As shown in the spectra, statistics were not enough. To compare with sampled gas and background runs, spectra were integrated to obtain better statistics. In Fig.8, obtained values of internal heating experiment were shown. The differences between background and sampled gas runs were very little. If sampled gas contained 1 Bq tritium, these values should increase to 50 % more. The other results obtained until now were shown in Fig.9. The data from run 1 to run 20 were the experiments of electrolysis cells, in which excess heats were observed. The data from run 21 to run 30 were those of the calibration gas. The total tritium number in the calibration gas was from 1 to 7 Bq. The runs more than 30 were the results of the external heating experiments.

b)Neutron

In the external and internal heating experiments, the two ^{3}He counters were set near the systems. The output signals of counters were recorded in every one minute, it started before heating and ended when the systems were completely cooled down. Clear increases of neutron were not observed. Results of the internal heating system in the highly loaded cases were shown in Table 1.

Table 1

Run number	Loading ratio	Coulomb[a)]	thickness of gold(μm)[b)]	neutron (1) counts /min.	neutron (2) counts/min.	neutron (3) counts/min.
1	0.65	6.18×10^5	0.638(0)	2.03 ±1.42 2.70 ± 1.64	1.00 ±1.00 0.5 ± 0.707	2.33 ±1.52 2.63 ±1.62
2	0.69	7.08×10^5	0.0408(1.05)	2.26 ±1.50 2.23 ±1.49	3.00 ±1.73 2.00 ±1.41	2.36 ±1.53 2.03 ±1.42
3	0.87	1.93×10^6	0.026(1.0)	2.10 ± 1.44 2.00 ± 1.41	4.52 ± 2.12 1.00 ± 1.00	2.26 ± 1.50 2.13 ± 1.46
4	0.82	1.92×10^6	0.055(1.0)	2.06 ± 1.43 2.46 ± 1.57	2.50 ± 1.58 5.00 ± 2.23	2.30 ± 1.51 2.20 ± 1.48

a) total current used for electrolysis

b) thickness of gold plated by electroplating, after loading, gold was once more plated, which was given in parentheses

1) average count of 30 minutes measurement before heating, upper column was detector A, lower was detector B

2) average count of 2 minutes measurement under heating 3) average count of 30 minutes measurement after heating

6.Conclusion

The gas proportional chamber was proved as the suitable detector for tritium hunting, especially in gas phase systems. The detection efficiency and detection limit were confirmed by calibration gas which contained tritium of 1 to 7 Bq. The detection limit was 0.5 Bq per 20 ml gas volume. External and internal heating methods were applied to the Pd + D systems. Deuterium gas from these systems were measured by the chamber. Clear differences between the deuterium gas and the background spectra were found until now . Further investigation for various systems should be needed in our laboratory, although it is important to do joint-works with other laboratories where anyone have samples for studies of this type .

References

1) T.N. Claytor et al., Proceedings of the 3rd International Conference on Cold Fusion p217
2) Y.Iwamura et al., Proceedings of the 4th International Conference on Cold Fusion p160
3) D.G.Tuggle et al., ibid., p221
4) T.Aoki et al., ibid., p214
5) K.Omata et al., INS-T-496