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ANOMALOUS HEAT EVOLUTION FROM A SOLID-STATE ELECTROLYTE UNDER ALTERNATING CURRENT IN HIGH-TEMPERATURE D_2 GAS

ELECTROLYTIC DEVICES FOR ENERGY GENERATION

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A coin-shaped proton conductor made from metal oxides of strontium and cerium can be charged in a hot D_2 gas atmosphere to produce excess heat. Anomalous heat evolution was observed from the proton conductors charged with alternating current at 5 to 45 V at temperatures ranging from 400 to 700°C. The anomalous heat produced temperature increases as much as 50°C. Excess heat was estimated as a few watts in most cases, totaling up to several kilojoules.

INTRODUCTION

The cold fusion reaction can be identified sometimes by a very low and weak level of neutron emissions, or occasionally by a high level of tritium production. But in most cases, the reaction produces a low or high level of excess heat, and this is the most readily observed signature of the reaction. Reproducibility is generally still poor. The reaction cannot be controlled because the mechanism is not understood. It is hoped that a successful theory will emerge, such as the one described by Chubb and Chubb,[1] who discuss a solid-state deuterium-deuterium fusion reaction that yields ^{4}He.

The proton conductors are used in this work as a gas phase electrochemical system, in contrast to the liquid electrochemical systems that are widely used to study the reaction, usually employing heavy water and palladium electrodes. One of the principal experimental difficulties with the liquid phase systems is that it takes an extremely long time to load the deuterium into

the metal. On the proton conductor, the reaction begins relatively quickly, usually within a half hour after the experiment has started.

Takahashi et al.[2] observed a large amount of heat generation by employing cyclic changes of polarization (unipolar alternating current or pulsing) with a palladium electrode in heavy water. This suggests that a perturbation such as changing electric currents might be advantageous. Yamaguchi and Nishioka[3] reported clear evidence of ^{4}He production from a palladium plate covered with MnO_2 film on one face and gold film on the other. The sample was believed to contain a comparatively low concentration of deuterium in this work, as opposed to the high loading that is considered essential to the reaction with palladium. Yamaguchi heated the sample up to several hundred degrees Celsius with an electric heater. This may mean that a quick rise in temperature is advantageous, although the solubility of deuterium is lower at elevated temperatures. The heat and the reaction products were extremely difficult to reproduce and showed no controllability. These examples suggest that some kind of triggering mechanism may be helpful in this reaction. We assumed that the reaction might occur when a chaotic state is caused by the movement of protons in the lattice. We have therefore used proton conductor ceramics at various temperatures and applied alternating electric fields as a trigger to cause the reaction.

The cold fusion reaction is widely believed to be related to nonstationary behavior of hydrogen and deuterium in the metal and electrolyte interface. In research heretofore, only palladium, palladium alloys, and titanium have been commonly used as host metals. There have been many difficulties in controlling the reaction in these metals. These difficulties are often linked with the following: absorption and diffusion of hydrogen and deuterium, temperature changes introduced by electrolysis, and other factors. Cold fusion reactions are expected to be transitory and unstable under these circumstances; they begin and end spontaneously after long loading periods. We have attempted to get around these problems and control the reaction by charging at a high temperature in D_2 gas. We discovered materials that load in a stable fashion, generating relatively stable and long-lasting reactions.

EXPERIMENTAL

Samples were made from a mixture of metal oxide of strontium, cerium, yttrium, and niobium. The procedures were developed by Iwahara et al.[4,5] and Yajima et al.[6] These powdered oxides were first mixed and then sintered in an electric furnace at 1400°C in air for 16 h. The samples were pulverized, again mixed, alcohol was added, and the samples were put in a pressing machine and formed into round plates 20 mm in diameter and 1 mm thick. These plates were again sintered at 1300 to 1480°C in air for 16 h. Both sides of the sintered sample were then coated with porous platinum film by painting with a platinum organic compound and deposition in 700°C. The resulting platinum film thickness was 0.15 to 0.3 μm. The film was porous and had a very rough surface; hydrogen gas easily passed through the gaps.

Figure 1 shows a schematic representation of the experimental system. The sample was heated to a constant temperature with an electric heater covered with stainless steel. Electric power was supplied from a stabilized power source. The electric field of constant voltage [Electric power of Proton Driving (EPD)] was supplied from a function generator via a power amplifier. Pressure was measured by a capacitance manometer with 0.1-Torr accuracy. Temperature was recorded with 0.1°C accuracy. All parameters including EPD (voltage and electric current), heater power, sample temperature, gas pressure, and cylinder wall temperature were recorded through a data logger to a floppy diskette. The reaction cell is made of a stainless steel cylinder 40 cm long and 20 cm in diameter, with walls 5 mm thick.

The upper part of the cell is shown in Fig. 2. The sample is held on both sides with 0.3-mm-thick platinum plates, which are in turn sandwiched between 0.1-mm-thick nickel plates. Five thermocouples with thin stainless steel are pressed directly on the upper nickel plate. The nickel plates make electrical and thermal contact with the ceramic sample and the thermocouples. This part is fixed on the place of the heater part. Spiral heater wire is also connected to the bottom part of the sample. The EPD power was supplied through copper wires 1.6 mm in diameter, covered with a ceramic insulator. The sample holder is surrounded by nickel plate reflectors. The holder is fixed with four supports made of 6-mm-diam stainless steel, covered by an alumina insulator. Two nuts attached under the support rods press the nickel plate, the alumina spacer, and the sample to make tight contact with the thermocouples. The components are welded to the cell cover flange that has several electric connectors. The connectors introduce the thermocouple, electric power lines for the heater, and electric field supply for the sample.

Experimental procedures are shown as follows:

1. The reactor cylinder is evacuated by a rotary and diffusion pumps (with a liquid N2 trap) to 2×10^{-5} Torr.
2. Sample temperature is raised to 400 to 700°C.
3. Gas is introduced into the cylinder at 0.1 to 50 Torr.
4. The sample is charged with EPD at 5 to 45 V alternating voltage, with frequency set between 10^{-4} and 1 Hz depending on the sample temperature and thickness.

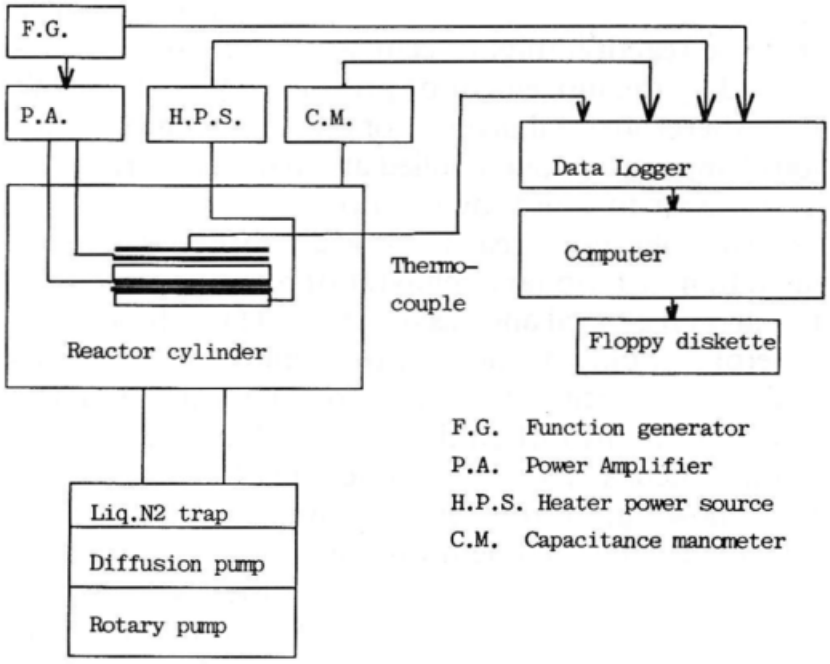

Fig. 1. Schematic representation of the measurement system.

RESULTS

The relationships between EPD input and temperature were calibrated with a ceramic sample conductor at various temperatures and gas pressures. Figure 3 shows a typical calibration curve. Deviations are caused by differences in thermal conductivity. Changes in gas pressure caused very small deviations. The next relationship obtained is between input EPD (E) and the temperature change of T in the temperature range from 300 to 700°C as follows:

$$E = A \exp(BT). \tag{1}$$

Figure 4 shows a typical excess heat evolved in an experiment with 1 Torr of D_2 gas. With only heater power supplied, the sample temperature rose to 470°C and stabilized. After 1 h EPD input of ±45 V, 0.005 Hz was supplied. At 0.7 h, the sample temperature rose to 480°C. Input EPD usually varies no more than 10%; however, here the input current varied from (67 μA) 3 mW to (178 μA) 8 mW with polarity change. It showed a steep increase, and fluctuation reached (0.7 mA) 35 mW. These instabilities may have been caused by local changes in the sample temperature. Usually temperature rise induces an increased rate of proton flow. Average EPD input was 7 mW, and the average excess heat was estimated as 1.5 W by Eq. (1). The EPD input was stopped at 4.4 h to observe the lasting time of the excess heat; however, the temperature fell down to the value that existed before EPD was supplied. The sample no longer showed excess heat. The sample showed no excess heat in H_2 gas at the same pressure and temperature as the D_2 gas.

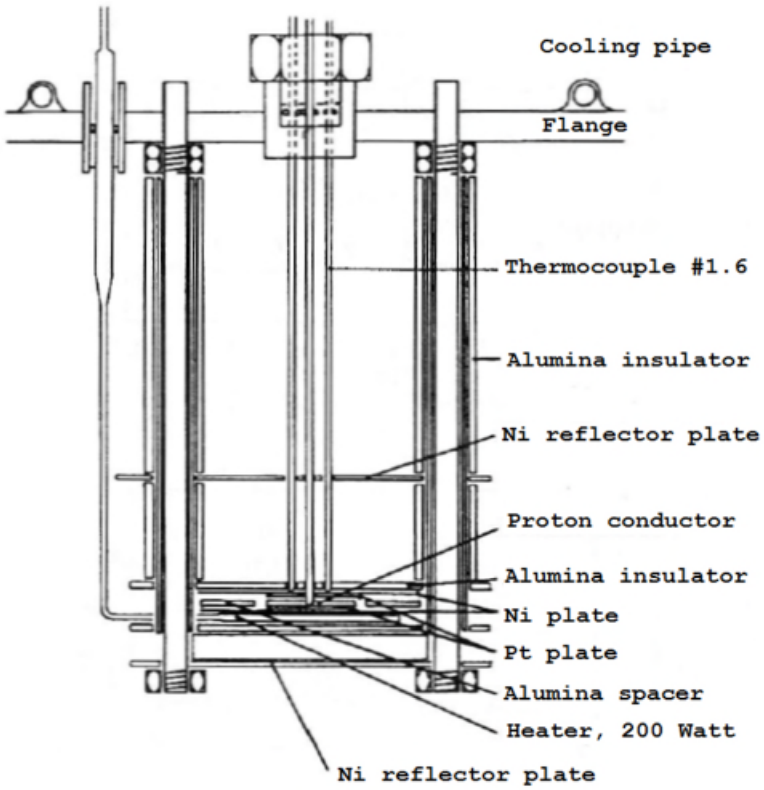

Fig. 2. Cross section of the upper part of the reactor cell.

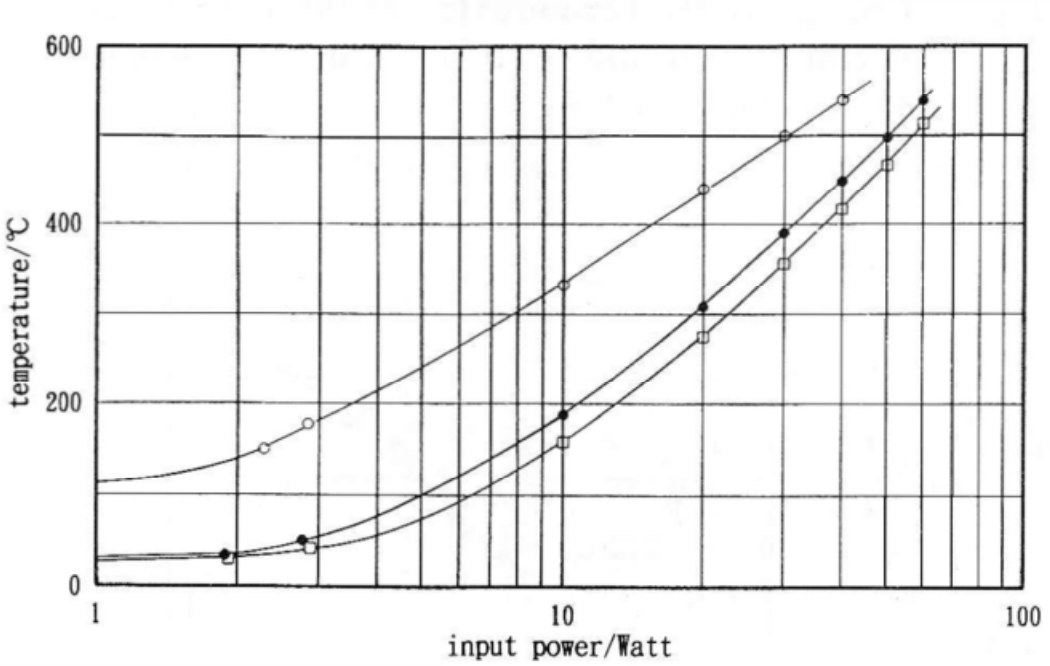

Fig. 3. Calibration curves between input power and the ceramic samples in various atmospheres: ○ = in vacuum, □ = H_2 12 Torr, ● = D_2 13.8 Torr.

Another example is shown in Fig. 5. The sample temperature was kept at 530°C in 12.4 Torr of D_2 gas. At 0.86 h after EPD was supplied, the temperature rose 5°C above the level seen with joule heating alone. This temperature elevation remained the entire time EPD was supplied. It reached a maximum of 543°C, about 13°C higher than the level with joule heating alone. Excess heat was estimated at 2 W maximum. The temperature fell immediately after EPD power was switched off, but some temperature fluctuations continued. The EPD was switched on again at 4.9 h, but the temperature increased only 5.7°C. Excess heat caused by the reintroduced EPD is difficult to estimate because of the continuing fluctuation from the earlier excess heat event. The EPD itself fluctuated by ±15% because of the temperature fluctuations. Figure 6 shows the details of temperature rise and EPD input ratio (ΔT/EPD) for Fig. 5; usually the ratio is 5 for the case of no excess heat. However, the ratio increases to 10.5 and shows a large fluctuation.

Figure 7 shows the temperature of the same sample in H_2 gas. It shows no excess heat and stable temperature change. Here EPD change caused by temperature deviation was limited to an estimated 10%. The ΔT/ EPD ratio remains stable at a value of 4.5.

Summarized results are shown in Fig. 8. The oblique straight line in this figure shows the calibration curve, the points at which input equaled output, with no excess heat. About 80% of the samples showed no excess heat, and 20% were positive. The samples that generated excess heat have large error bars because the EPD with any proton conductor becomes very unstable when the temperature changes. Samples that showed no excess heat showed stable temperatures.

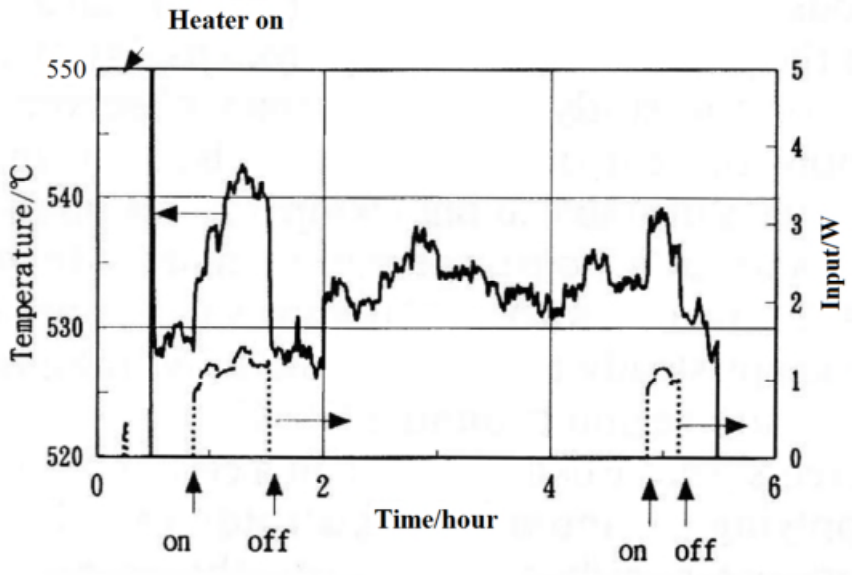

Fig. 5. Changes of temperature and EPD input in D2 gas.

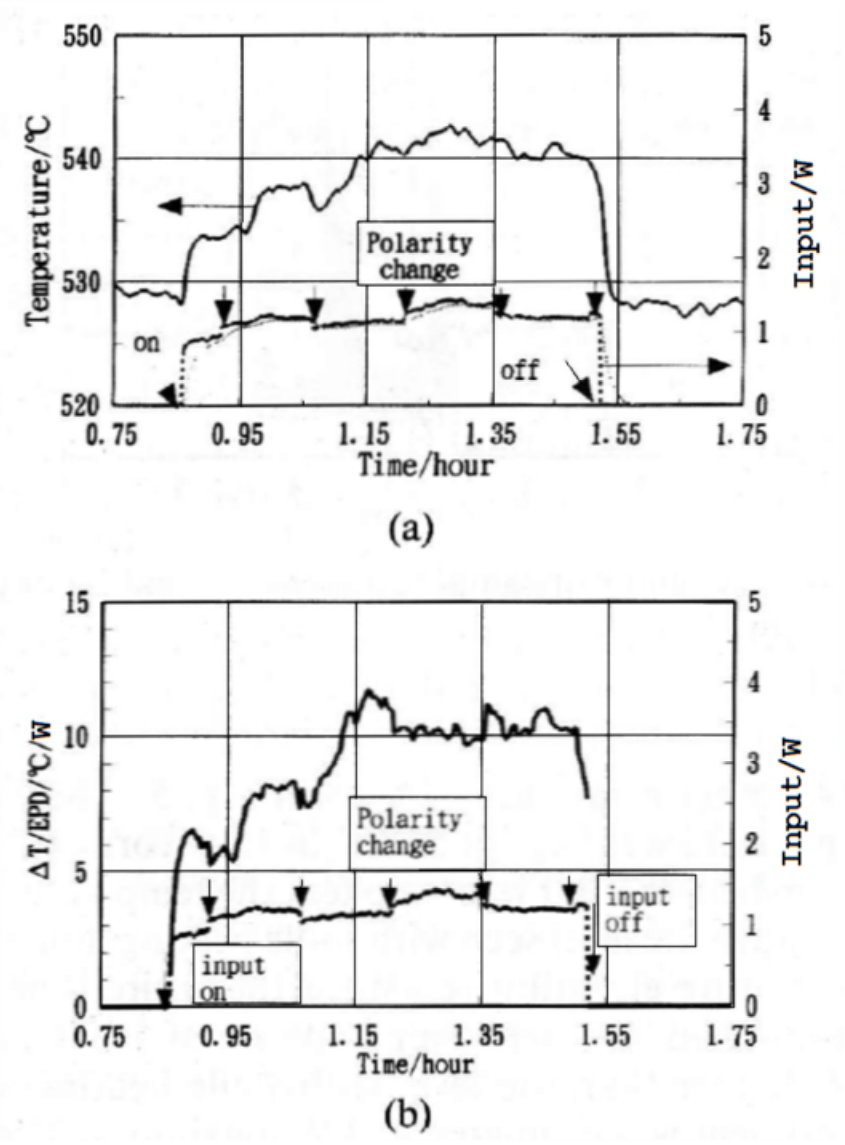

Fig. 6. Details for Fig. 5: (a) change of temperature and EPD and (b) change of the ratio of temperature rise and EPD input.

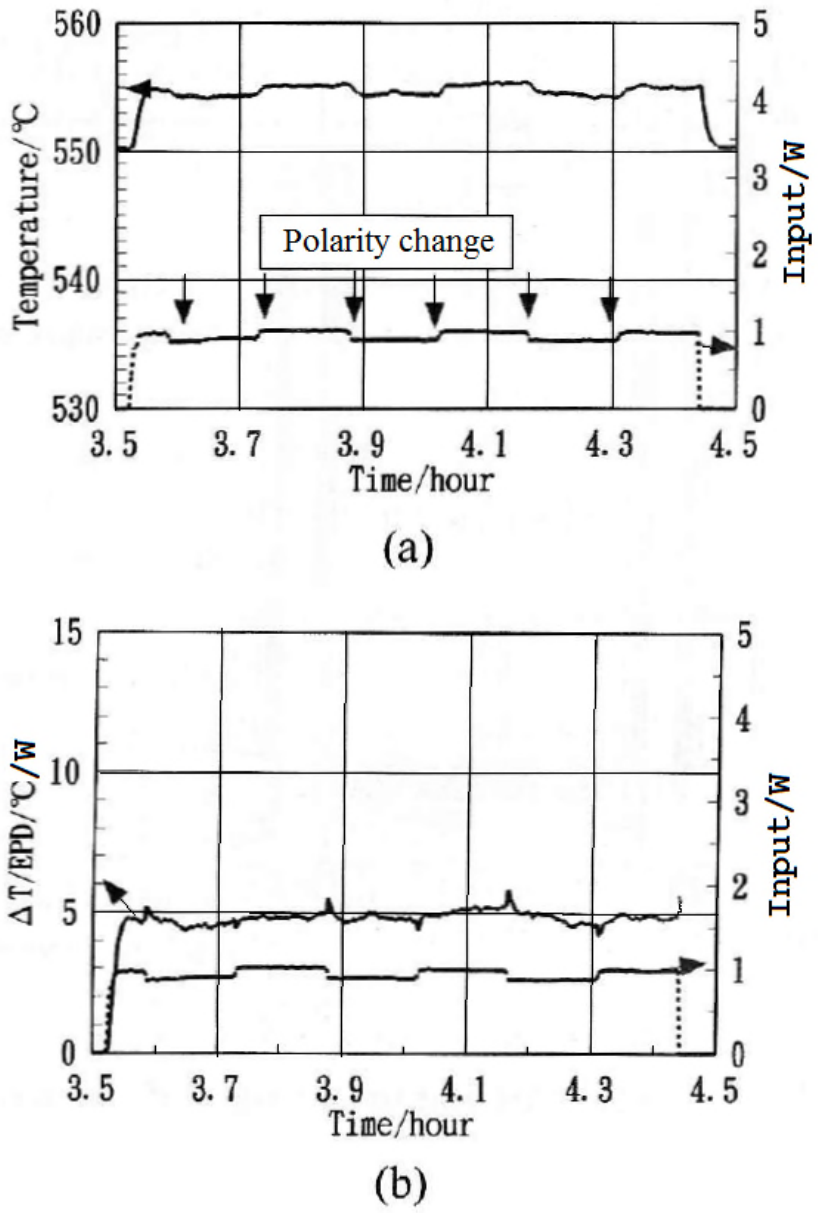

Fig. 7. Changes of temperature: (a) the ratio of temperature rise and input and (b) EPD input in H_2 gas for the same sample of Fig. 4.

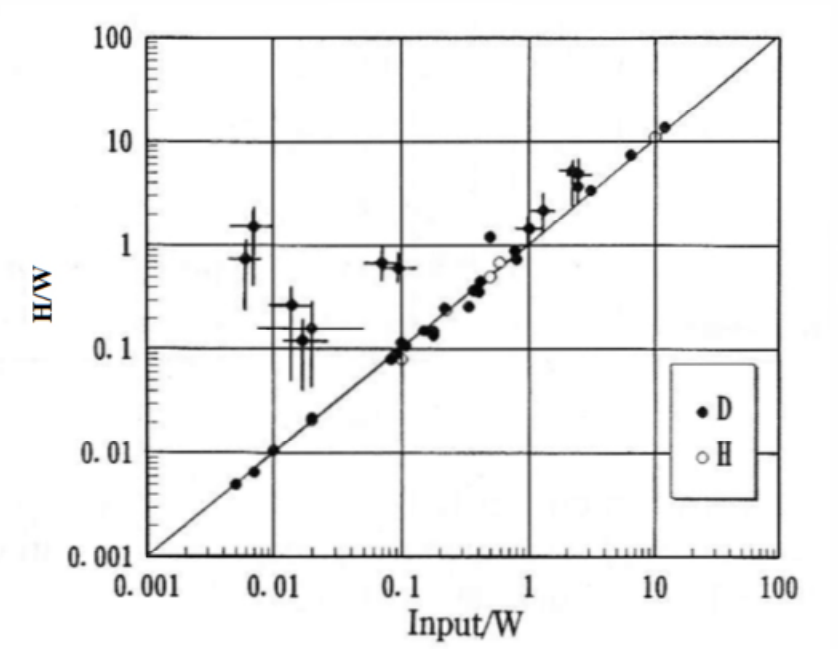

Fig. 8. Summarized results of the relationship between electrical input power and heat output for various samples.

CONCLUSION

All previous attempts at cold fusion experiments can be classified as either wet or dry loading of hydrogen isotopes. Contrary to this previous work, the initial new attempts were done by Granite and Jorne.[7] They thought it would be useful to combine the wet and dry methods to take advantage of electrochemical potential in the gas phase. They employed β-alumina specimens for the study and sometimes observed a few emissions of neutrons as low as the background. The β alumina is unstable at high temperatures up to 300°C and gets to show no proton conductivity. Meanwhile, a lattice type of proton conductor such as perovskite oxide keeps steady proton conductivity as high as the temperature region around 1000°C.

Excess heat usually evolved in a certain time lag after applying the input power. Proton mobility at the temperature is easily obtained using the Nernst-Einstein relation as follows:

$$\frac{D}{RT} - \frac{\sigma}{nZ^2e^2} \cdot \tag{2}$$

Here, Ze is the charge number as 1 for D^+, n ion density in the sample that was obtained from current density. Proton mobility 4×10^{-6} at 450°C and 1.05×10^{-5} at 540°C $cm^2/s \bullet V$ obtained by the relation are the same as Yajima's data.[8] However, the time lag between the occurrence of excess heat evolution and the supply of input power is almost ten times larger than the time of the relationship for a proton to reach from one side to another. And the excess heat clearly decreased after the input power supply ceased. This phenomenon suggests that the excess heat evolution may be induced by deuterium flow in the conductor accelerated by an electric field.

Only 5 samples in 50 showed the excess heat evolution. Also, the construction and composition of the proton conductor can be considered an important factor that decides the reaction of the excess heat evolution.

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