J. Condensed Matter Nucl. Sci. 24 (2017) 179–190

Research Article

Anomalous Excess Heat Generated by the Interaction between Nano-structured Pd/Ni Surface and D_2 Gas

Takehiko Itoh*,†, Yasuhiro Iwamura and Jirohta Kasagi
Research Center for Electron Photon Science, Tohoku University, Sendai, Japan

Hiroki Shishido
Department of Quantum Science and Energy Engineering, Graduate School of Engineering, Tohoku University, Sendai, Japan

---

Abstract

A new experimental setup based on Mizuno's work was introduced in our lab in order to investigate the anomalous heat generation phenomena. Following Mizuno's procedures, we fabricated nano-structured material composed of Pd and Ni by glow discharge on a heater located in the center of a vacuum chamber. The nano-structured Pd/Ni was prepared with D_2 before use. Then, we applied electrical power to the heater that was covered with the nano-structured Pd/Ni while evacuating the chamber, and we observed the heater temperature behavior. Next, we introduced D_2 gas at about 250 Pa to the chamber while maintaining the heater input. As a result, in three experiments, we observed that heater temperature increases compared to the reference experiment (with no nano-structured Pd/Ni). In particular, in experiments with 7 W input, we observed a123°C heater temperature increase compared to the reference experiment. It can be said that we replicated Mizuno's experiment successfully. Since the heater was covered with nano-structured material, there was a concern that a change in emissivity affected the heater temperature measurement. Numerical calculation was conducted to evaluate the effect of the change of the surface emissivity. It was concluded that even if the emissivity drop dramatically from 0.7 to 0.3 due to the coating of the nano-structured Pd/Ni, the temperature rise would be only 70°C at the most. The postulated emissivity change cannot explain the observed temperature increase of 123°C. These experimental and numerical results suggest that anomalous excess heat was generated by the interaction between nano-structured Pd/Ni surface and D_2 gas.

Keywords: Anomalous heat, D_2 gas absorption, Finite element method, Nano-structured material, Replication

---

1. Introduction

A new division devoted to Condensed Matter Nuclear Reaction (CMNR) was established at the Research Center for Electron Photon Science of Tohoku University in April 2015.

---

*Corresponding author. E-mail: itoh@lns.tohoku.ac.jp .

†Also at: Clean Planet Inc., Tokyo, Japan.

An experimental apparatus introduced in our lab was based on the experimental device and methods developed by Mizuno [1,2] for the study of anomalous heat generation. Mizuno, Yoshino and their colleagues have been developing excess heat generation methods and devices for a few years. According to their report, Ni and Pd are alternately discharged in a chamber to fabricate nano-structured Pd/Ni. After the baking process, D_2 or H_2 gas was introduced into the chamber. This process causes D (H) to absorb in the nano-structured Pd/Ni. After that, they observed anomalous heat generation. In one example, they reported as follows. When D_2 gas about 100–300 Pa was introduced at 200°C, they observed about 78 W excess power, which was almost as much as the input power. We have begun doing experiments using a similar device and procedures.

2. Experiment

2.1. Experimental apparatus

We made a device based on information from Mizuno. The chamber and components are almost identical to Mizuno's experimental apparatus. Figure 1 shows a schematic and photograph of our experimental apparatus. The vacuum chamber is made of stainless steel and its volume is 7.5 l. This chamber is evacuated with an oil-free dry scroll pump, ultimately to a vacuum of 1 Pa or less.

Pd electrodes and a ceramic heater are installed inside the chamber. Ni mesh is set around the heater and electrodes. Figure 2 shows the structure of the heater and electrodes. A ceramic heater is installed in the center of the chamber. This ceramic heater is made of alumina of size 25 mm × 25 mm, and has a built-in thermocouple inside. A 0.3 mm diameter Pd wire is wound around this heater so that high voltage can be applied to the wire. A Pd rod (φ3 mm $L = 200$ mm) is placed so as to face this heater, and this rod also makes it possible to apply high voltage. A Ni mesh (φ0.15 mm, 50 mesh) is placed surrounding the heater and Pd rod, and this is grounded to the chamber. As a result, high voltage can be applied between the Pd wire and the Ni mesh, or between the Pd rod and the Ni mesh. The

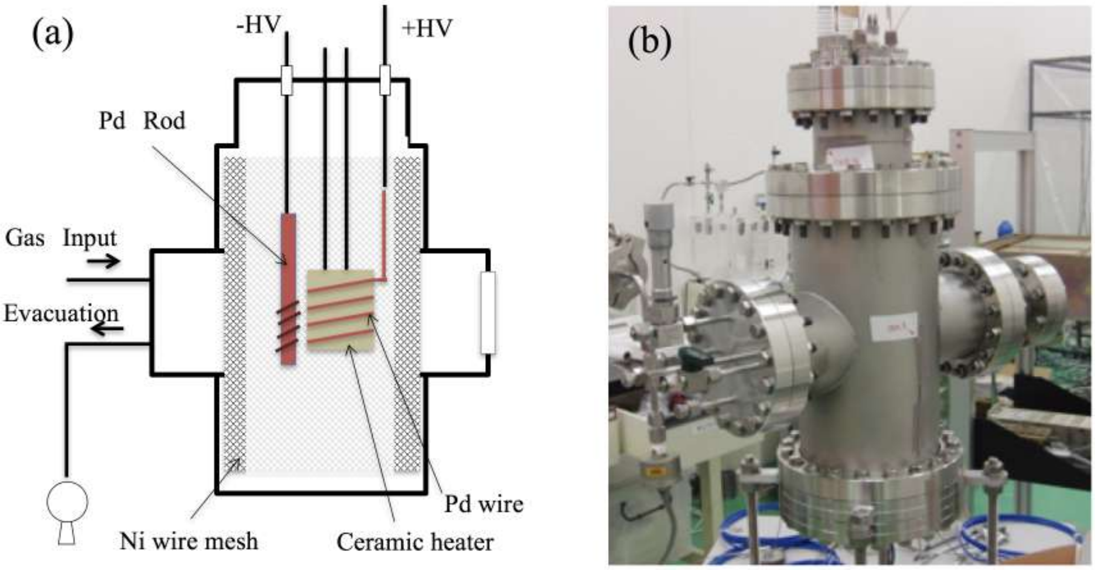

Figure 1. Experimental apparatus: (a) schematics of apparatus and (b) photograph of apparatus.

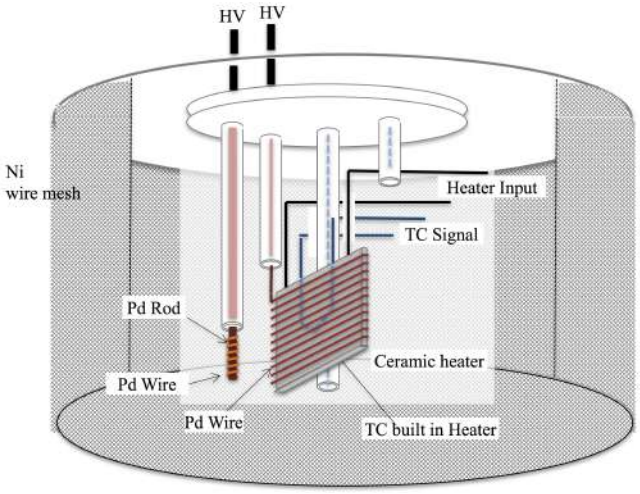

Figure 2. Schematic of components around the heater and electrodes.

chamber is covered with Al foil for thermal insulation.

2.2. Experimental procedure

Our experimental procedure followed the methods reported by Mizuno. The following three steps are taken.

(1) Reference run without activated nano-structured Pd/Ni material.
(2) Activation treatment: formation and activation of nano-structured Pd/Ni material.
  (a) Sputtering by glow discharge,
  (b) baking,
  (c) D_2 absorption.
(3) Foreground run with the activated nano-structured Pd–Ni material.

Determining whether anomalous excess heat was generated by comparing the reference run to the foreground run. Details of each process from (1) to (3) will be explained.

In the reference run, two kinds of measurements were conducted. The first was to measure a temperature of the heater by inputting power to the heater under vacuum condition of 1 Pa or less. The second was to introduce deuterium gas and measure the heater temperature while maintaining pressure of 200–300 Pa.

In activation treatment, the following three processes were carried out. First, the nano-structure was fabricated on the Pd/Ni surface in the chamber, using glow discharge. We performed two patterns of Ar glow discharge (Table 1). We introduced Ar at 10–100 Pa into the chamber. Next, we applied negative high voltage (–1 kV) to the Pd rod, so Pd is sputtered by this discharge. Figure 3 (a) is a photograph of Pd sputtering. We continued this discharge about

Table 1. Voltage of electrode for discharge.

	Pd wire winding heater	Pd rod	Ni mesh
#1	Float	–HV (1 kv)	Grand
#2	+HV (500–700 V)	Float	Grand

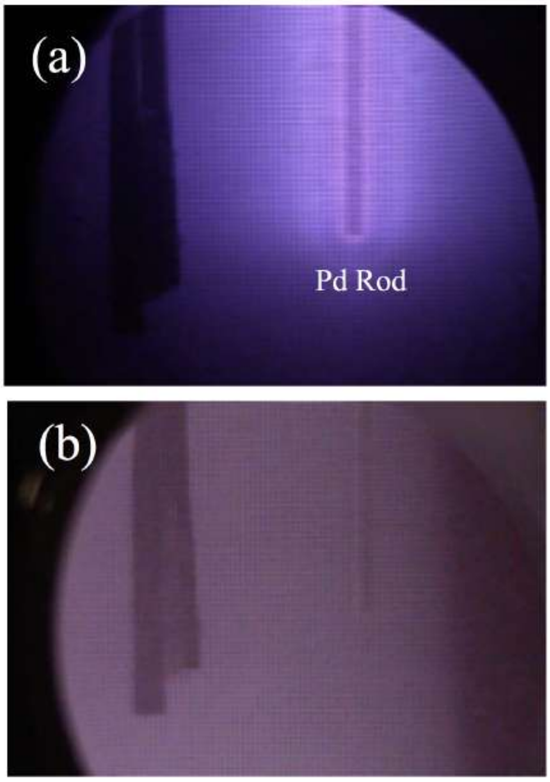

Figure 3. (a) Photograph of #1 discharge and (b) #2 discharge.

10,000 s. Next, positive high voltage (+600 to +800 V) was applied to the Pd wire. In this case, Ar ions attack the Ni mesh, so Ni is sputtered (Fig. 3 (b)). This discharge continued about 10,000 s. Table 1 shows the electrode voltage of each discharge. We repeated #1 and #2 discharge alternately three times, so Ni and Pd mixed to form nano-structured Pd/Ni.

Figure 4 (a) and (b) shows the photographs of the electrode before and after experiments. You can see there is a silver colored film on and around a heater after experiments. Figures 4(c) and (d) are the SEM images of the surface of the Pd wire. From the images of (c) and (d), it can be seen that a film covering surface of the wire, i.e., a film formed by sputtering by discharge, is composed of fine particles of several hundred nano-meters in size. According to the Energy dispersive X-ray spectrometry (EDS) analysis, most of the fine particles were Pd and a few contained Ni. We also analyzed other areas and observed similar features. As described above, with this treatment it is possible to fabricate a structure composed of Pd–Ni nano-particles on the heater and its peripheral parts. This structure is similar to the one Mizuno described.

As the second activation treatment, baking was carried out at 100–200°C for about one day to remove water molecules and hydrocarbons from the inner surface of the chamber.

The final activation process is deuterium absorption in the nano-structured Pd/Ni. After baking, D_2 gas was introduced into the chamber, which was sealed off at a pressure of 170 Pa, and this state was maintained for more than 12 h.

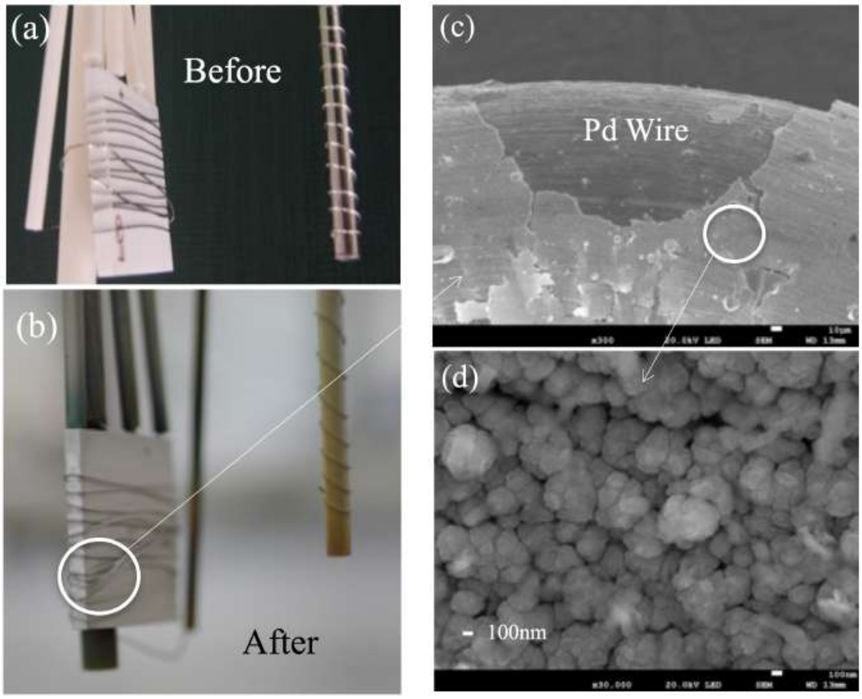

Figure 4. Photograph of electrode and SEM image. (a) Before activation process, (b) after foreground experiment, (c) SEM image of wire and (d) zoom image of (c).

After this process, a foreground run was carried out with the same procedure as the reference run. We will discuss the result in Section 3.

3. Results

Figure 5 shows the results of the foreground run of Experiment 1. In this experiment, we applied constant power of 40 W to the heater and maintained this condition. Figure 5 (a) is a reference run (before the activation process). In the reference run, under vacuum conditions (<2 Pa), the heater temperature is 714°C. When D_2 gas was introduced at 260 Pa after 1 h, the heater temperature dropped to 587°C from D_2 gas cooling. The gas pressure changes in Fig. 5(a) were caused by gas cooling of the heater temperature. As a result, it was found that in the range of 10–300 Pa, there is an effect of several degrees. Figure 5(b) is the foreground run. The chamber was sealed with D_2 gas at 170 Pa until the start of the experiment, vacuum evacuation was performed, and power of 40 W was applied to the heater. The temperature of the heater rose to 764°C, which was higher than the temperature of the reference run by 50°C. After 1.7 h, when D_2 gas was introduced at up to 260 Pa, the temperature of the heater was 611°C, which was 24°C higher than in the reference experiments. This result suggests that anomalous heat generation is occurring.

We performed Experiment 2 under the same conditions as Experiment 1. Figure 6 shows the result of Experiment 2. The chamber was filled with deuterium and left for 12 h. After that, we applied 40 W of power to the heater while simultaneously evacuating the chamber. The temperature of the heater rose to 760°C, which was 46°C higher than the reference run. This temperature rise was about the same as Experiment 1. After 2 h, when D_2 gas was introduced at up to 270 Pa, the temperature of the heater rose to 613°C, which was 26°C higher than in the reference experiments.

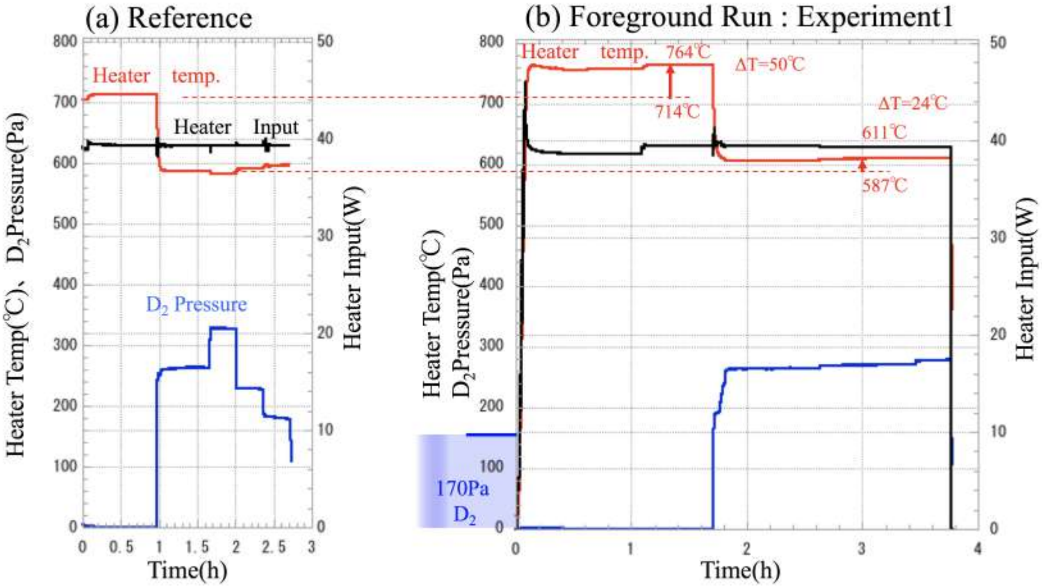

Figure 5. Heater temperature, heater input, and D_2 pressure of Experiment 1: (a) reference and (b) foreground run. Red line heater temperature, black line heater input and blue line D_2 pressure.

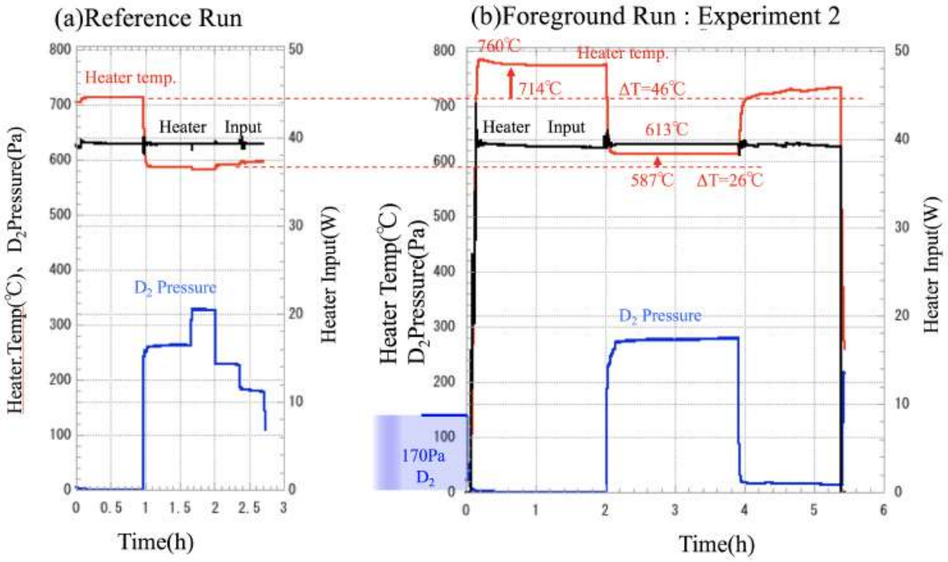

Figure 6. Heater temperature, heater input, and D_2 pressure of Experiment 2: (a) reference run and (b) foreground run. Red line heater temperature, black line heater input and blue line D_2 pressure.

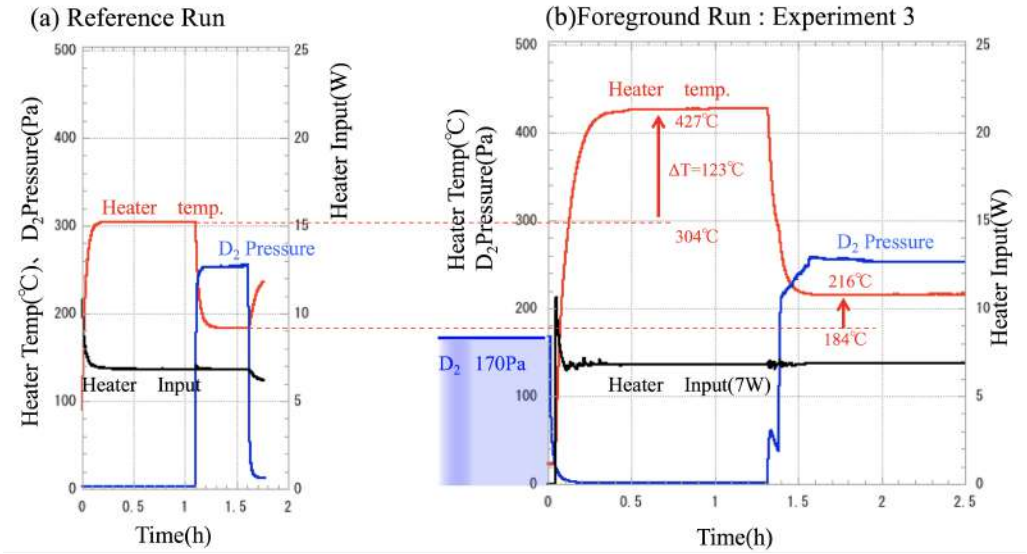

Figure 7. Heater temperature, heater input, and D_2 pressure of Experiment 3: (a) reference and (b) foreground run. Red line heater temperature, black line heater input and blue line D_2 pressure.

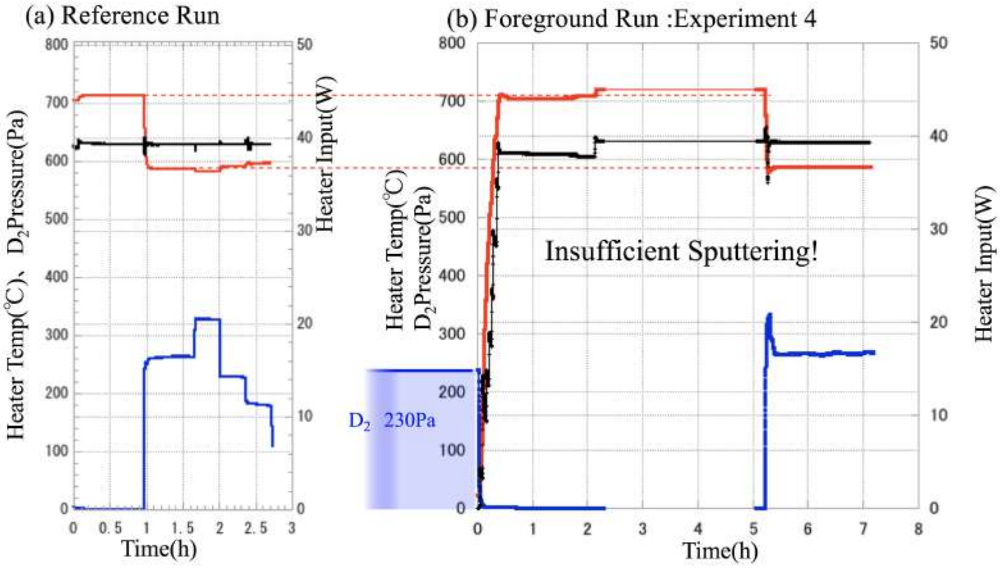

Figure 8. Heater temperature, heater input, and D_2 pressure of Experiment 4: (a) reference and (b) foreground run. Red line heater temperature, black line heater input and blue line D_2 pressure.

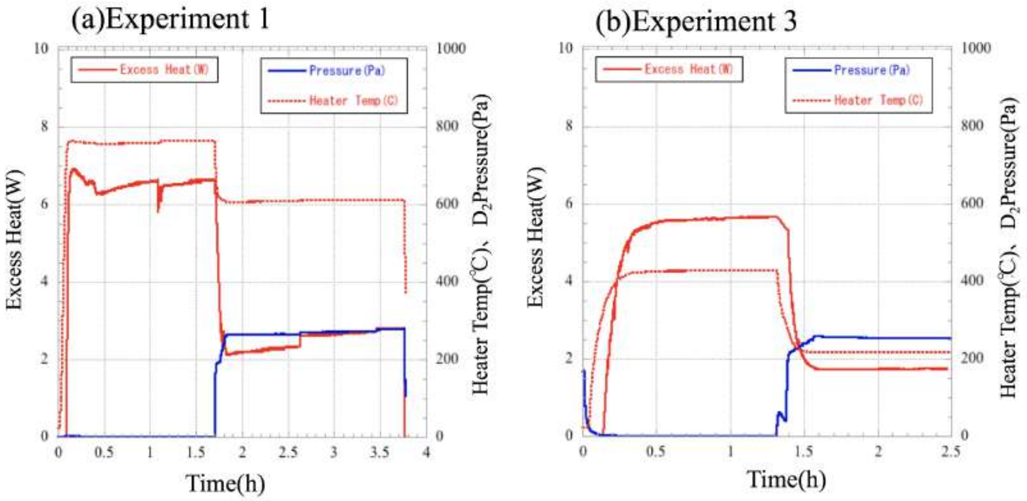

Figure 9. Thermal outputs of Experiments 1 and 3. (a) Experiment 1 (heater input 40 W) and (b) Experiment 3 (heater input 7 W). Red solid line Excess heat, red dash line heater temperature and blue line D_2 pressure.

In this way, the same extent of temperature rise was observed in the two experiments under the same conditions, so the results were reproduced.

Next, we performed an experiment with input power changed from 40 to 7 W. The results are shown in Fig. 7. In the reference run, under the vacuum condition, the heater temperature reached 304°C, and under the condition with deuterium at 260 Pa, it reached 184°C. On the other hand, in the foreground rum, the temperature in vacuum reached 427°C, which was higher than that in the reference run by 123°C. In addition, when deuterium was introduced, the heater temperature became 216°C, which was 32°C higher than in the reference run.

The next experiment failed to produce excess heat, as is shown in Fig. 8. In this experiment, in the activation process, discharge voltage was insufficient, so sufficient sputtering was not performed. In fact, after the experiment, we examined the heater and the electrode but we found the nano-structured Pd/Ni had not formed. As a result, in the reference run and Foreground run, the temperature of the heater hardly changed and no anomalous heating phenomenon was observed. This suggests that nano-structured Pd/Ni is necessary for abnormal heat generation.

The thermal output of the foreground experiment was evaluated based on the data from the control experiments.

Figure 9(a) is an evaluation of excess heat in Experiment 1. In this experiment, we input 40 W to the heater. In the first half 1.7 h, under vacuum conditions, excess heat is seen at around 7 W. In the second half, 260 Pa of deuterium was introduced, and the excess heat was 2.5 W. Figure 9(b) shows the evaluation of excess heat in Experiment 2, where input power is 7 W. In this case, under vacuum conditions, the excess heat is 5.5 W and the ratio of excess heat to input reaches 80%. When deuterium gas is introduced, the excess heat is 1.6 W.

4. Discussion

In this discussion, we will show that the amount of energy in our experiments is much larger than a normal chemical reaction, such as the combustion heat of hydrogen absorbed in nano-structured Pd/Ni.

We assume that the source of excess heat is absorbed deuterium in nano-structured Pd/Ni formed on the heater surface. The EDX analysis shows that the nano-structured film is composed of 99% Pd and 1% Ni, which is formed with a thickness of 10 μm on the surface of the heater. The amounts of Pd in nano-structured film is approximately 1.414×10^{-3} mol.

D_2 gas absorbed in this material was approximately 1.65×10^{-4} mol, which was estimated from pressure decrease of D_2 gas which was introduced into the chamber and sealed off before the experiment. This pressure decrease was about 50 Pa in each experiment. Although there are Pd rods and Pd wires in the chamber, we assume that the loading ratio of D/Pd is about 0.23, so only ~1.65×10^{-4} mol was absorbed.

If this absorbed deuterium reacts with oxygen for some reason, the total energy of combustion would be 47 J. On the other hand, in the first half of the case of Experiment 1, excess heat of 6 W lasted 1.8 h. This amount of energy is 38,880 J. In the case of Experiment 3, the total energy of excess heat is 19,800 J. These values are much larger than the heat of combustion of deuterium and cannot be explained as a chemical reaction.

Next, we will show that this cannot be apparent excess heat from a radiation change caused by the coating of the Pd / Ni nanostructure film.

In our experiments, since the heater is covered with nano-structured Pd/Ni, there is a concern that the emissivity may vary and affect the heater temperature measurement. We conducted experiments at two temperature ranges of 700°C (Experiments 1 and 2) and 300°C (Experiment 3). Comparing the two experiments, the temperature rise in the low temperature experiment (300°C) was higher than high temperature experiments (700°C). With the effects of radiation, it can be expected that heater temperature rise will increase in the high temperature region, but the experimental results are different. Consequently, we conclude that the observed temperature rise is not related to the effect of radiation.

A numerical calculation was conducted to evaluate the effect of the change of surface emissivity. We performed numerical calculation using the commercial finite element method software COMSOL Multiphysics 5.2 Heat transfer module [3].

The governing equation of heat transfer is described as follows.

$$\rho C_{\mathrm{p}}\,(\overrightarrow{\mu}\cdot\triangledown)\,T+\triangledown\cdot q=Q, \tag{1}$$

$$\overrightarrow{q}=-k\triangledown T. \tag{2}$$

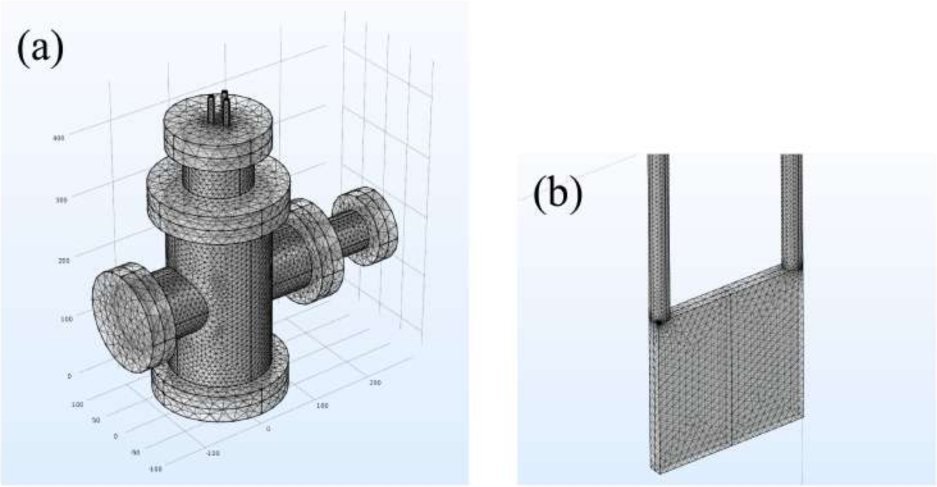

Figure 10. Chart of numerical analysis of (a) chamber and (b) heater.

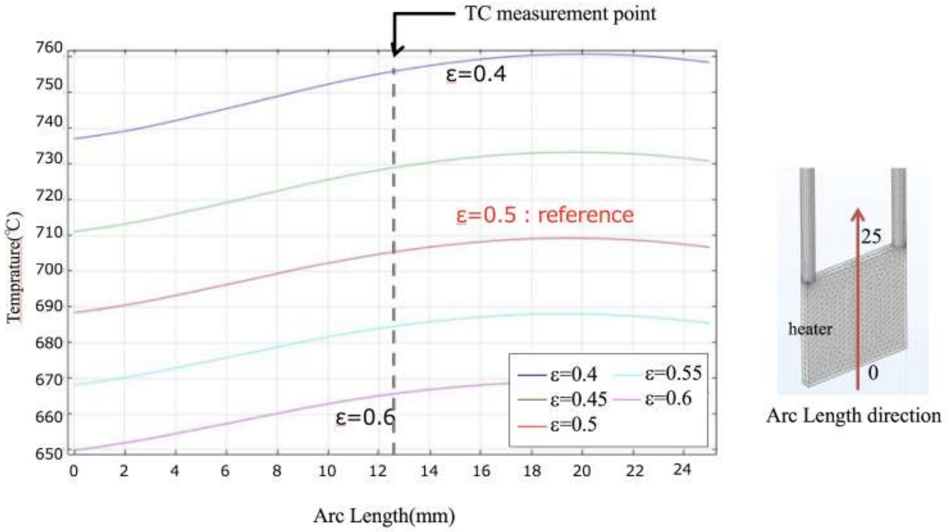

Figure 11. Calculation result of temperature distribution of heater. Case of Experiments 1 and 2. (Heater input is 40 W and heater temperature is 700°C.)

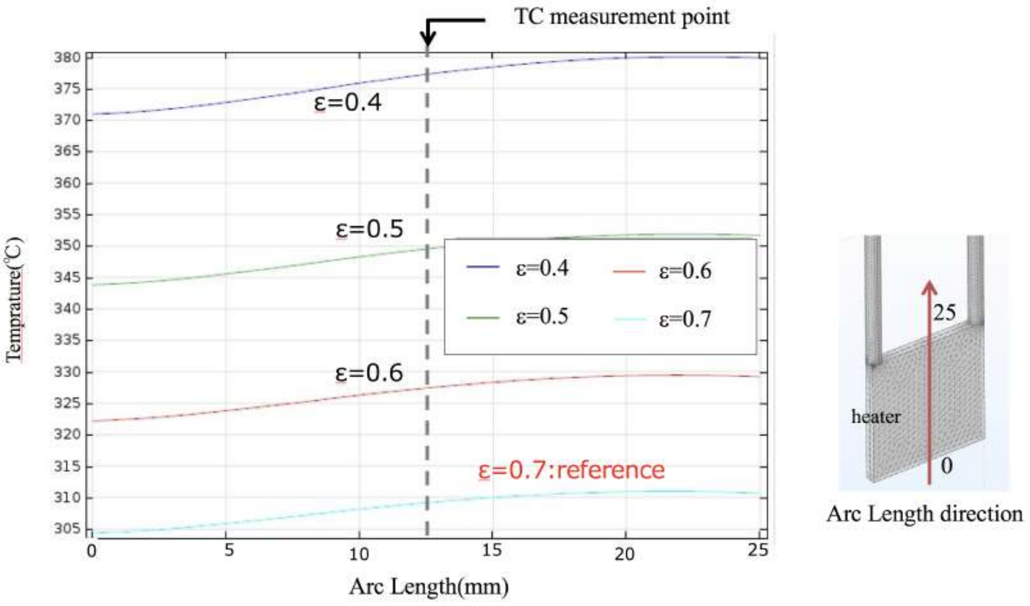

Figure 12. Calculation result of temperature distribution of heater, in the case of Experiment 3. (Heater input is 7 W and heater temperature is about 300°C.).

where

Q	heat density of heat source
q	heat flux
C_p	heat capacity of heat medium
ρ	density of heat medium
$\overrightarrow{u}$	fluid flow vector

As a first step, we simulated the experiments under vacuum conditions (the first half of experiment). In this case, since the fluid flow velocity vector is 0, the term of thermal convection in the first term of Eq. (1) can be neglected, and Eq. (1) includes only the second terms of heat conduction and radiation. The following boundary conditions are applied: adiabatic condition on the outside of the chamber, thermal radiation on the heater surface and inner surface of the chamber written as Eq. (3). A chamber, a heater, and internal members, as shown in Fig. 10 were divided into finite elements of 207,479 free tetrahedrons. We conducted numerical calculations using these procedures.

$$-\overrightarrow{n}\cdot\overrightarrow{q}=\varepsilon\{G-e_\mathrm{b}(T)\} \tag{3}$$

where

ε	total emissivity,
G	irradiation,
$e_\mathrm{b}(T)$	total emissive power of black body,
$\overrightarrow{n}$	normal vector on the boundary.

In the case of Experiments 1 and 2 with 40 W, in the reference run the heater temperature is 714°C (see Figs. 5(a) and 6(a)). On the other hand, since the catalog value is 613°C, the total emissivity ε is assumed to be 0.5. Based on the above calculation, heat density was 3.0×10^7 W/m^3 by calculation only with the ceramic heater (Fig. 10(b)). Next, a heater was placed in the chamber, and calculations were performed taking into account the effects of heat transfer and irradiation (Fig. 10(a)). As a result, the heater temperature was 706°C, which is in good agreement with the experimental result. In this state, calculations were performed when the heater surface was covered with nano-structured Pd/Ni and the emissivity changed. The results are shown in Fig. 11. According to this, if the emissivity decreases by 0.1 due to the covering of the nano-structured Pd/Ni, the heater temperature is expected to rise by 50°C. For this reason, it is difficult to completely eliminate the temperature rise due to the change in the emissivity, given the temperature rise observed in Experiments 1 and 2.

Next, we consider the case of 7 W input (Experiment 3). In this case, from the heater temperature at 7 W input, the total emissivity was 0.7 and heat density was 5.5×10^6W/m^3, as calculated by the heater alone. Based on this result, calculations were carried out when it was installed in the chamber. The heater temperature was 309°C, which is also nearly the same as the reference run (Fig. 7(a)). As with the case of 40 W, the calculated result of the temperature distribution when emissivity is changed is shown in Fig. 12. According to Fig. 12, even when the emissivity drops dramatically from 0.7 to 0.3 due to the coating of the nano-structured Pd/Ni, the temperature rise is only 70°C at the most. On the other hand, in the experimental results (Fig.7(b)), a temperature rise of 123°C has been observed, which suggests anomalous heat generation.

As described above, at least in the experiment at the 300°C region, we showed anomalous heat generation. At the present time, there are many data analyzing the effects of temperature, pressure conditions and material properties (structure and composition) of this phenomenon.

Based on the above discussion, excess heat observed in our experiments conducted by Mizuno's method cannot be explained by a normal chemical process, which suggests it is a condensed matter nuclear reaction. Since radiation measurement and element analysis were not carried out at the present time, but only thermal measurements, further experiments and analysis will now be performed, detailed data will be acquired and we hope the phenomena will be clarified.

5. Conclusion

We tried to reproduce the heat generation experiment reported by Mizuno. We followed the same procedures using same experimental apparatus. As a result, the phenomenon reported by Mizuno could be reproduced.

As a result of evaluating the total amount of excess heat observed in our experiments, we conclude that the total amount of energy was much larger than that of plausible chemical reaction. Since a heater was covered with nano-structured material, there was a concern that change in emissivity affected the heater temperature measurement. Based on numerical calculations, we concluded that the observed temperature increase rise cannot be explained even if we postulate an unrealistic emissivity drop. These experimental and numerical results suggest that anomalous excess heat was generated by the interaction between nano-structured Pd/Ni surface and D_2 gas.

Acknowledgements

The authors would like to thank Dr. Tadahiko Mizuno for the information on his device and procedures, and Prof. Hidetoshi Hashizume and Prof. Noritaka Yusa for their cooperation in the numerical analysis. The Department of Instrumental Analysis Technical Division School of Engineering, Tohoku University, supported our SEM analysis. We also thank Mr. Hideki Yoshino and Mr. Masanao Hattori for their support.

References

[1] T. Mizuno, Reactant, heating device, and heating method, Patent Application, WO2015/008859 A2.

[2] H. Yoshino, E. Igari and T. Mizuno, Presentation at 2014 CF/LANR Colloquium at MIT, March.21–23, 2014, Massachusetts Institute of Technology, Cambridge, MA, USA.

[3] COMSOL Multiphysics Heat Transfer Module User's Guide, Version 5.2.