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Research Article

Replication Experiments at Tohoku University on Anomalous Heat Generation Using Nickel-based Binary Nanocomposites and Hydrogen Isotope Gas

Y. Iwamura*, T. Itoh† and J. Kasagi
Research Center for Electron Photon Science, Tohoku University, Sendai, Japan

A. Kitamura‡, A. Takahashi and K. Takahashi
Technova Inc., Tokyo, Japan

Abstract

We built a new experimental system to replicate the anomalous heat generation experiments based on the papers by A. Kitamura and A. Takahashi. The system was developed in order to measure precisely heat generation using a flow-calorimetry method with liquid hydrocarbon coolant that enables us to measure at temperatures higher than 373 K. The Ni-based binary nano-composite samples were prepared by the melt spinning method. A fabricated material at Kobe University were separated into two samples. One sample was loaded and tested at Tohoku University and the other at Kobe University, in order to compare the experimental results at the two different places. Two experiments were performed up to now. One was the PNZ4s ($Pd_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with D_2 gas experiment and the other is the CNZ5s ($Cu_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with H_2 gas experiment. For the PNZ4s with D_2 gas experiment, excess heat up to 10 W was observed. The amount of excess energy reached 2.5 MJ and it corresponded to 14.9 eV per absorbed D. CNZ5s ($Cu_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with H_2 gas experiment also showed anomalous excess heat ranging from 2 to 5 W. Coincident increase events of the pressure of reaction chamber and gas temperature, which suggested high temperature gas generation in the reactor chamber, was observed many times. The amount of excess energy amounted to 1.9 MJ and the generated energy per hydrogen atom was estimated as 67.8 eV/H. For the both samples subjected to the same fabrication process, results of the present work qualitatively agreed with those of the similar experiment performed at Kobe University. And these observations are supposed to be very difficult to explain by known chemical processes only.
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Keywords: Anomalous heat, Deuterium gas, Gas loading, Hydrogen gas, Ni-based binary nano-composites, Replication

*Corresponding author. E-mail: iwamura@lns.tohoku.ac.jp.
†Also at: CLEAN PLANET Inc., Tokyo, Japan.
‡Also at: Graduate School of Maritime Sciences, Kobe University, Kobe, Japan.

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1. Introduction

The Research Center for Electron Photon Science of Tohoku University and CLEAN PLANET Inc. agreed to the establishment of a collaborative research division – Condensed Matter Nuclear Reaction Division, in 2015 [6]. Research on anomalous heat generation started at the Condensed Matter Nuclear Reaction Division (CMNRD) although Iwamura and Itoh have been engaged in the study of transmutation reactions observed in nano-structured Pd/CaO multilayer thin film induced by D_2 gas permeation [7–15] for many years at Mitsubishi Heavy Industry and then CMNRD. Replication efforts have been made in two types of experiments as a first step at CMNRD in Tohoku University is the work using nickel-based binary nano-composites with D_2/H_2 gas by Kitamura and Takahashi [1–5], and the other is the experiment using nano Pd/Ni fabricated by glow discharge with D_2 gas by Mizuno [16,17].

In this paper, replication experimental results on the Kitamura and Takahashi works are described, which were achieved by close cooperation with Technova Inc. and Kobe University. A new experimental system using a flow-calorimetry method was built at Tohoku University, and the experimental results were compared with the results obtained at Kobe University. Similar results were obtained for the samples subjected to the same fabrication process, so it can be said that the replication efforts were successful.

2. Experiment

2.1. Experimental apparatus

A schematic of our experimental apparatus is shown in Fig. 1. It is basically similar to the apparatus described in Ref. [1], with some improvements. The main features of this system are as follows.

(1) Oil mass flow-calorimetry at high temperature.
(2) Many temperature sensors.
(3) Resistant to outside-temperature fluctuation.

The reaction chamber (RC) that contains the nickel-based binary nanocomposites and hydrogen isotope gas is located in the center of Fig. 1. Heat generation from the RC is estimated by the mass-flow calorimetry, which is widely used in this field, and is considered an accurate measurement method. Our method is standard except that we used oil coolant instead of water coolant. The coolant is input to the RC from the bottom and heated by the RC. Heat generation from the RC is calculated by the temperature difference between the coolant inlet and the outlet. The inlet and outlet oil temperatures are measured by three thermocouples each. The system is intended to accurately measure heat generation at high temperatures above 373 K. A liquid hydrocarbon coolant enables us to use the flow-calorimetry method at temperatures higher than 373 K. The coolant oil is an aromatic hydrocarbon (Barreltherm-400 (BT400); Matsumura Oil Co. Ltd.), which can work up to 600 K. The coolant is driven by a digital liquid tubing pump with a flow rate from 14.0 to 14.8 cm^3/min.

H_2 or D_2 gas is fed from a reservoir through a needle valve precision flow regulator to the nickel-based binary nanocomposite sample in the RC. Pressures in the RC and the reservoir are continuously monitored. Temperature distribution in the RC is measured by four Resistance Temperature Detectors (RTDs) and temperatures along the oil coolant pipe and the stainless-steel gas introduction pipe are monitored by thermocouples. Many temperature measurement points enable us to judge whether observed excess anomalous heat is real or not. The outer chamber is kept at constant temperature with cooling water from the thermobath $23\pm0.1^\circ$C. The entire apparatus is placed in a thermostatic chamber controlled at $23\pm0.1^\circ$C to avoid the influence of outside temperature fluctuations.

A 1 kW sheath heater (#1) is spirally wound on the outer surface of the RC and a 200 W cartridge heater (#2) is located at the central axis of the RC to heat up the sample in the RC. The power to the heater is fed from a finely

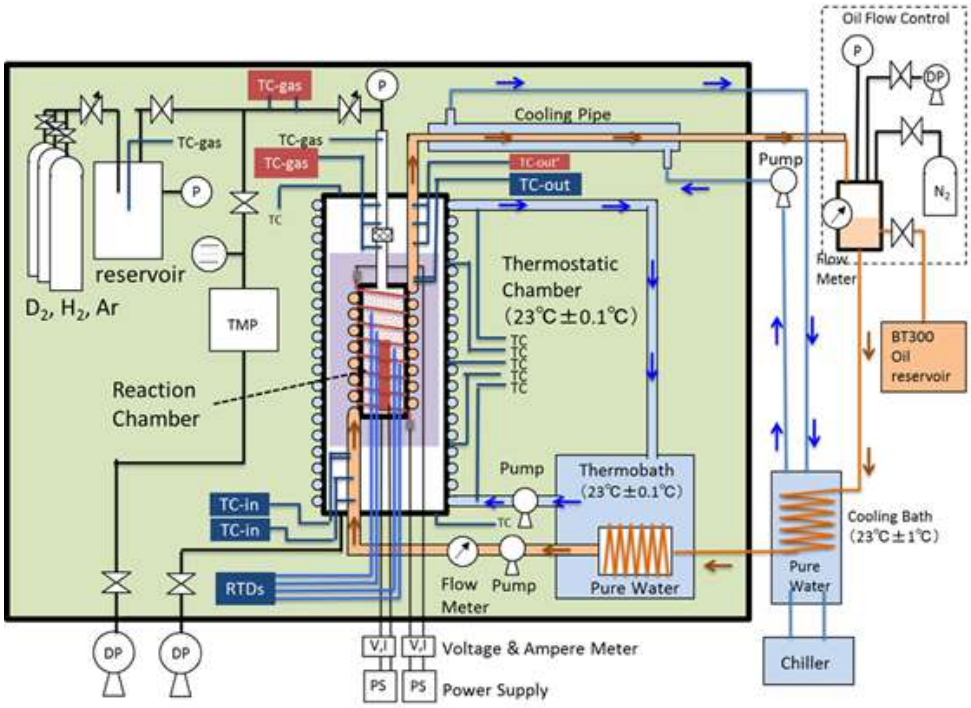

Figure 1. Experimental set-up.

regulated DC power supply. The input electric power for every heater is continuously monitored by two independent voltage and ampere meters to avoid mistakes on the input power estimation.

The coolant oil goes to the oil reservoir located outside of the thermostatic chamber and is cooled down to the ambient temperature by a cooling bath. It goes back to the tubing pump through a water bath kept at a temperature of 23±0.1°C.

2.2. Sample preparation

The procedure for sample preparation is the same as described in [1,2]. At first, an amorphous mixture of metal elements (Pd, Ni, Zr and Cu) were prepared by the melt spinning method at a Sendai company. By annealing the mixture in air at a temperature of 723 K for 60 h at Kobe University, preferential oxidation of Zr to ZrO_2 was expected with a consequent formation of binary-nanoparticles of Pd/Ni or Cu/Ni embedded in it. The sample compositions for the present paper are shown in Table 1. Atomic ratios of Pd:Ni or Cu:Ni were chosen to be 1:7 based on the Technova–Kobe work performed up until now [1–5].

The materials fabricated at Kobe University were separated into two samples. One sample was loaded and tested at Tohoku University in Sendai City and the other at Kobe University in Kobe City, in order to compare the observed experimental results at the two places. The samples CNZ5s and PNZ4s have the same physical and chemical properties as CNZ5 and PNZ4, respectively.

Table 1. Sample composition.

Sample name	Tested at	Weight (g)	Molar fraction (%)				
			Cu	Pd	Ni	Zr	O
PNZ4s	Tohoku University	109.4	–	3.6	25.2	53.4	17.8
PNZ4	Kobe University	109.4					
CNZ5s	Tohoku University	130.0	1.7	–	11.6	24.5	62.3
CNZ5	Kobe University	130.4					

2.3. Experimental procedure

Calibration or blank runs to measure the heat recovery rate of the experimental apparatus were performed using zirconia beads in the RC before and/or after foreground runs. The diameter of the beads was 1 mm and weight was 1300 g for both experiments.

At the beginning of a foreground run, PNZ4s or CNZ5s sample with the zirconia beads was put into the RC. The RC was evacuated by a turbo molecular pump and then heated up to 200–300°C to remove H_2O or other impurity gas. After the vacuum baking, the RC is cooled down to room temperature. About 1.0 MPa D_2 gas for PNZ4s and H_2 gas for CNZ5s were stored in the reservoir chamber in advance, then D_2/H_2 gas was introduced into the RC by opening the needle valve as shown in Fig. 1.

In the case of the PNZ4s sample, D_2 gas absorption and heat generation was observed at room temperature due to the presence of Pd. After observing that, we applied electric power to the heaters located at the inside and outer-surface of the RC to increase sample temperature. Data from temperatures, pressures, voltages, currents and a flow rates was logged during the experiments. Based on the data, we estimated the H/D absorption rate and excess heat generation from the samples.

3. Results and Discussion

Two experiments were performed up to now. One was the PNZ4s ($Pd_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with D_2 gas experiment and the other was the CNZ5s ($Cu_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with H_2 gas experiment.

3.1. Heat analysis and error estimation

The heat analysis of this system is based on the equation:

$$\eta Q = F_{\mathrm{R}}\rho(T_{\mathrm{ave}})C(T_{\mathrm{ave}})(T_{\mathrm{out}} - T_{\mathrm{in}}), \tag{1}$$

where η rate, Q is the heat release rate, F_{R} is oil flow rate, T_{ave} is the oil density as a function of temperature, $C(T_{\mathrm{ave}})$ is heat capacity, T_{out} and T_{in} are the outlet and inlet temperatures of the coolant oil, respectively. Physical data of $\rho(T)$ and $C(T)$ of the coolant oil are already known. As the temperature dependence of $\rho(T)$ and $C(T)$ is linear, we can postulate that T_{ave} is equal to $(T_{\mathrm{out}} + T_{\mathrm{in}})/2$. Q is expressed as

$$Q = W_1 + W_2 + H_{\mathrm{EX}}, \tag{2}$$

where W_1, W_2 and H_{EX} are the input power of heater 1, the input power of heater 2 and the excess heat power from the RC.

Based on these equations η is determined as a function of $(W_1 + W_2)$ by a blank run because Q, F_{R}, $\rho(T_{\mathrm{ave}})$, $C(T_{\mathrm{ave}})$ and $(T_{\mathrm{out}} - T_{\mathrm{in}})$ are obtained by experimental data. H_{EX} is calculated by a foreground run using the determined η.

We simplify Eqs. (1) and (2) to estimate experimental error.

$$H_{\mathrm{EX}} = \frac{F_{\mathrm{R}}\rho C}{\eta}\Delta T - W,$$

$$\Delta T = T_{\mathrm{out}} - T_{\mathrm{in}}, \qquad W = W_1 + W_2.$$

Considering that experimental variables are F_{R}, ΔT the error range of the calculated excess heat is the sum of the fluctuations of the oil flow rate, temperature difference and input electrical power.

$$\delta(H_{\mathrm{EX}}) \approx |\delta(F_{\mathrm{R}})| \frac{\rho C\,\Delta T}{\eta} + |\delta(\Delta T)| \frac{F_{\mathrm{R}}\rho C}{\eta} + |\delta(W)|\,. \tag{3}$$

Actual experimental data shows that largest contribution to the error of H_{EX} is of the F_{R} term and W is the most stable. Considering that experimental variables are F_{R}, ΔT and W, we can assume that error range of the calculated excess heat is the sum of fluctuations of oil flow rate, temperature difference and input electrical. In the case of 80 W input, for example, fluctuations are as follows.

$$|\delta(F_{\mathrm{R}})| = 0.012(\mathrm{ml/min}), \quad |\delta(\Delta T)| = 0.261(\mathrm{K}), \quad |\delta(W)| = 0.031(\mathrm{W}), \quad \delta(H_{\mathrm{EX}}) = 0.260\,(\mathrm{W}).$$

About $\pm$0.26 W for 80 W input could be the error range for excess heat estimation in our experimental set-up.

3.2. PNZ4s

The first experiment was done using the PNZ4s sample at Tohoku University after a blank run. Figure 2 shows the temperature change at four points in the RC during absorption of D into PNZ4s at room temperature. Rapid temperature rises from room temperature could be seen depending on the positions of RTDs when D_2 gas was introduced into the RC. RTD1, RTD2, RTD3 and RTD4 are located successively from the bottom as shown in Fig. 2. It could be supposed that PNZ4s sample local density was highest around the RTD2 and RTD3, considering that the weight ratio of PNZ3s to zirconia beads is less than 0.1 and sample powder would be localized somewhere. This might be the reason why peak temperatures are larger at RTD2 and RTD3. The reason for the very fast temperature rise at RTD2 is unknown.

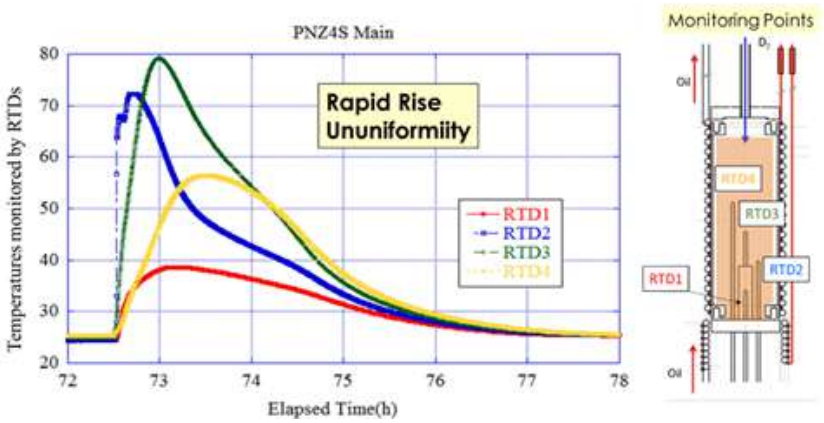

Figure 2. Temperature change at four points of the RC during absorption of D into PNZ4s at room temperature.

Figure 3 shows excess heat generation from PNZ4s with D_2 gas. The peak power induced by D_2 gas absorption at room temperature amounted to 10 W but it did not last for a long time. On the contrary, excess heat power was not so high at elevated temperatures but it continued more than 100 h. The peak ratios of excess heat to input power were about 4% and 5% for 80 and 134 W input, respectively. Integrated excess energies were 0.65 MJ for 80 W input and 1.73 MJ for 134 W input. Therefore, the amount of excess energy released from the RC was about 2.4 MJ.

Based on the pressure measurement at the RC and the reservoir for D_2 gas, the total amount of deuterium absorbed into PNZ4s was 1.73 mol. Although it is very difficult to postulate that every absorbed D atoms produced additional energy after the saturation of D-loading, average released energy at elevated temperatures per initial total D-loading is calculated as 14.9 eV/D, which is the lowest estimation of specific energy by one deuterium reaction that might happen. As the long-lasting excess heat was observed after the D-loading was saturated, the real portion of deuterons contributing to the heat generation is regarded to be very small (several orders of magnitude [1–5]) and consequently estimated specific energy might be very large). It is therefore very hard to explain that the generated excess heat was caused by some chemical reactions, as energy generation from known chemical reactions cannot exceed 10 eV/D

3.3. CNZ5s

A comparison of temperatures detected by RTDs and E1 between a blank run and an experiment with CNZ5s is shown in Fig. 4 when 134 W of heater power was input to the RC. In this experiment, no input was applied to W2 during the entire course of the test, because the W2 heater was not working due to disconnection-troubles. The thermocouple E1 is located on the top of the RC. Temperature increases of RTD3, RTD4 and E1 from the blank experiment data were observed for the CNZ5s experiment, although there was no difference between RTD1 and RTD2 temperatures compared to those of blank runs. It seems that excess energy is generated at temperatures higher than 220°C based on these observed experimental results.

Pressure of the RC (Pr) and a gas temperature (E2) at the near point of the top of RC are plotted in Fig. 5. Sudden fluctuations of Pr and E2 could be seen during the CNS5s experiment, although no fluctuation could be seen during the blank run and the baking (time-zone 0–150 h) as shown in Fig. 5.

Coincident increase events of Pr and E2 at random timing were observed as shown in Fig. 6. The pressure increase was about 0.02 or 0.03 MPa and the temperature increase ranged from 1°C. These were significant values and cannot

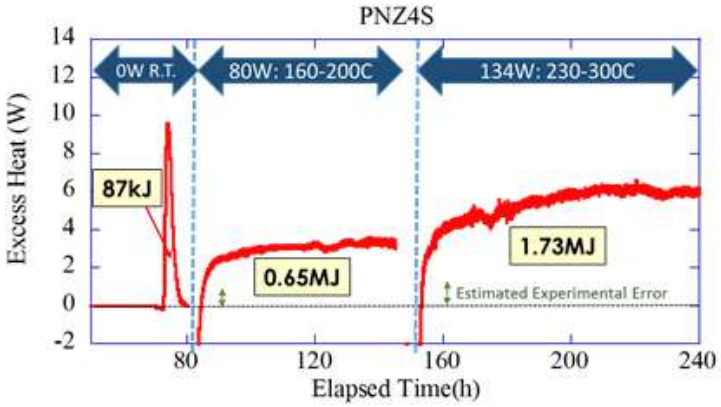

Figure 3. Excess heat generation for PNZ4s.

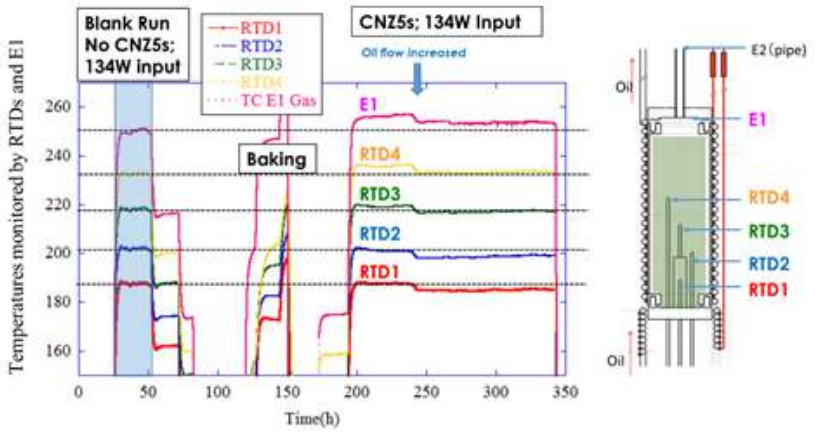

Figure 4. Comparison of temperatures detected by RTDs and E1.

be explained as instrument noise, as the other measurement data were stable and only Pr and E2 changes occurred, both at the same time.

A further close-up look at Pr and E2 confirms that Pr and E2 increased simultaneously, as shown in Fig. 7. The time interval for the plotted points for Pr and E2 was 10 s. Considering that the excess energy seemed to be generated in the upper region of the RC, as demonstrated in Fig. 4, these simultaneous increases of Pr and E2 might be due to the generation of high temperature gas. If we assume that high temperature gas was intermittently produced by certain condensed matter nuclear reactions in the upper region of the RC, this assumption will fit the experimental observations.

Excess heat for CNZ5s based on Eqs. (1) and (2) is illustrated in Fig. 8. About 4–5 W and 2–4 W were released during 80 W and 134 W heater power input, respectively. These excess powers are larger than the estimated errors based on Eq. (3). Sudden increase of excess power around 240 h seemed to be induced by the increase of the oil flow rate, but we have no accurate explanation for it.

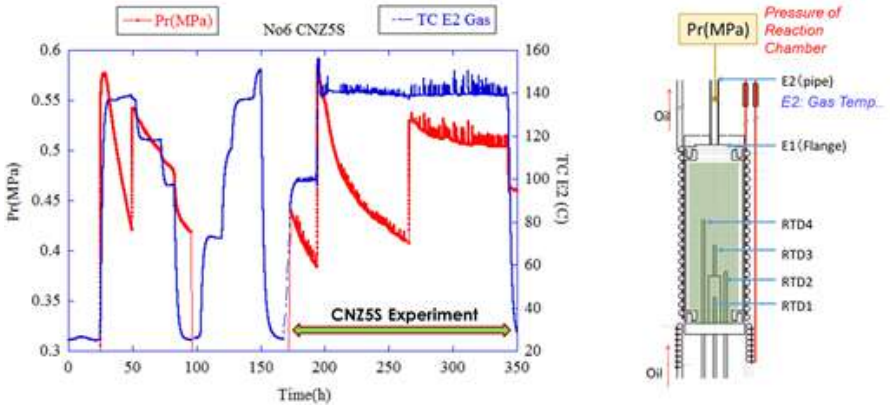

Figure 5. Fluctuations of pressure of the RC (Pr.) and a E2 Gas Temp.(E2) during a CNZ5s experiment.

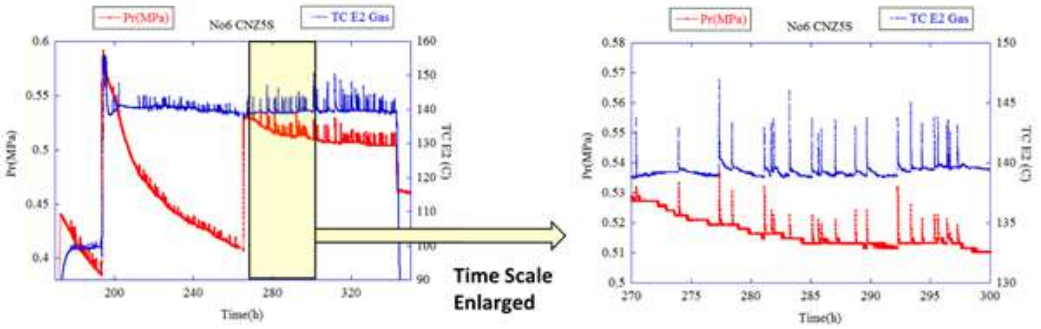

Figure 6. Coincident increase of Pr and E2.

All the integrated excess power comes to 1.86 MJ according to Fig. 8. Also estimated H atoms absorbed into CNZ5s during the experiment is 0.29 mol based on the pressure measurement at the RC and the reservoir for H_2 gas. Excess energy production is calculated as 67.8 eV/H, although it is very difficult to assume that every absorbed H atoms produced any energy same as D atoms. Also, it is very difficult to explain how the generated excess heat could have been caused by chemical reactions, as 67.8 eV/H energy generation is far larger than usual chemically released energy. This anomalous excess energy generation, in addition to the observed coincident pressure and temperature rises of the RC, suggests that some condensed matter nuclear reactions occurred in the RC.

3.4. Comparison between experimental results at Tohoku university and those at Kobe university

Table 2 summarizes experimental results obtained at Tohoku University and those at Kobe University. As previously described, the samples CNZ5s and PNZ4s have the same physical and chemical properties as CNZ5 and PNZ4, re-

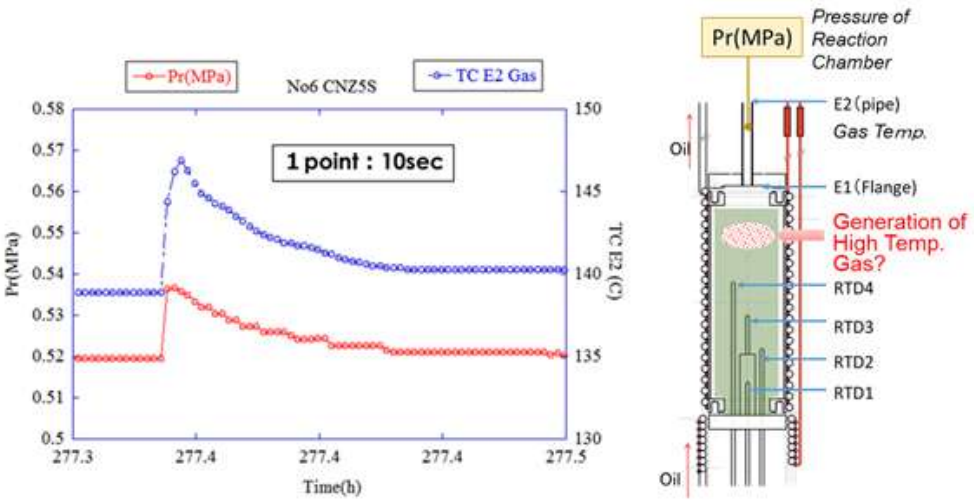

Figure 7. Simultaneous increase of Pr and E2 and the hypothesis for it.

Table 2. Comparison between experimental results at Tohoku university and those at Kobe university.

Sample	Composition	Tested at	Released energy at room temp. (eV/D)	Released excess power at elevated temp. (W)	RC temp. (°C)
PNZ4s	$Pd_{0.044}Ni_{0.31}Zr_{0.65}$	Tohoku University	0.57	2–6	160–300
PNZ4		Kobe University	0.56	Not estimated due to malfunction	
CNZ5s	$Cu_{0.044}Ni_{0.31}Zr_{0.65}$	Tohoku University		2–5	150–250
CNZ5		Kobe University		2–8	200–350

spectively.

The energy released at room temperature from PNZ4s and PNZ4 were 0.57 and 0.56 eV/D, respectively. These values are very similar. If we look at the released excess power from CNZ5s and CNZ5, they are close to each other. These results show that we successfully replicated experimental results on excess heat at least qualitatively, although there is still not enough quantitative reproducibility.

4. Conclusion

A new experimental system to replicate the anomalous heat generation using nickel-based binary nanocomposites and D_2/H_2 gas was introduced at Tohoku University. Two experiments were performed up to now. One was the experiment using PNZ4s ($Pd_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with D_2 gas, and the other was the experiment using CNZ5s ($Cu_{0\ 044}Ni_{0\ 31}Zr_{0\ 65}$) with H_2 gas. Anomalous excess heat generation were observed in both cases, and the experimental results of the present work qualitatively agreed with those of the similar experiment performed at Kobe University. In the case of CNZ5s experiment, events of coincident burst-like increases in the pressure of the reaction chamber and the gas temperature were observed many times, which suggested high temperature gas generation in the reaction chamber. These experimental observations are very difficult to explain by chemical reactions only.

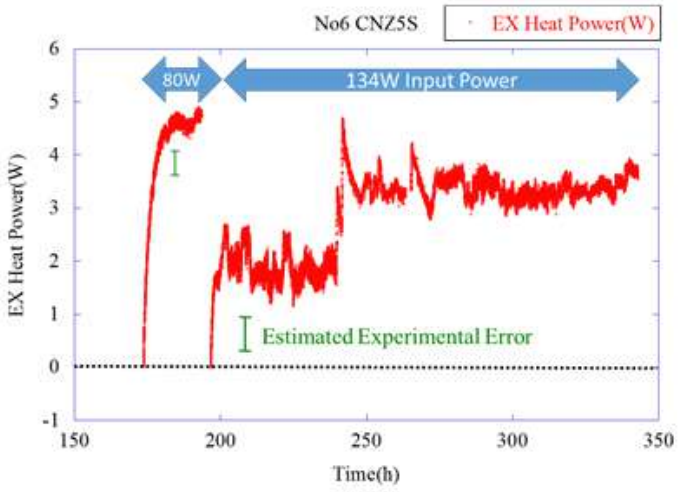

Figure 8. Excess heat generation for CNZ5s.

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