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Research Article

Anomalous Heat Effects Induced by Metal Nano-composites and Hydrogen Gas

Yasuhiro Iwamura*, Takehiko Itoh† and Jirohta Kasagi

Research Center for Electron Photon Science, Tohoku University, Sendai, Miyagi 982-0826, Japan

Akira Kitamura‡, Akito Takahashi, Koh Takahashi, Reiko Seto and Takeshi Hatano

Technova Inc., Tokyo 100-0011, Japan

Tatsumi Hioki and Tomoyoshi Motohiro

Green Mobility Research Institute, Institutes of Innovation for Future Society, Nagoya University, Nagoya, Aichi 464-8603, Japan

Masanori Nakamura, Masanobu Uchimura, Hidekazu Takahashi and Shunsuke Sumitomo

Research Division, Nissan Motor Co. Ltd., Yokosuka, Kanagawa 237-8523, Japan

Yuichi Furuyama

Graduate School of Maritime Sciences, Kobe University, Kobe, Hyogo 658-0022, Japan

Masahiro Kishida and Hideki Matsune

Graduate School of Engineering, Kyushu University, Fukuoka 819-0395, Japan

Abstract

Collaborative research among Technova Inc., Nissan Motor Co. Ltd., Kobe Univ., Kyushu Univ., Nagoya Univ. and Tohoku Univ. was done from Oct. 2015 to Oct. 2017. For this collaborative work, a new accurate oil mass-flow. . . (continued in the next page)

Keywords: Anomalous heat, Deuterium gas, Excess heat, Gas loading, Heat burst, Hydrogen gas, Metal nano-composite, Nano-material, Replication

*Corresponding author. E-mail: iwamura@lns.tohoku.ac.jp.
†Also at: CLEAN PLANET Inc., 105-0022 Japan.
‡Also at: Graduate School of Maritime Sciences, Kobe University, Kobe 658-0022 Japan.

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(continued from the title page)
calorimetry system was developed at Tohoku University to replicate anomalous heat generation experiments reported by Technova and the Kobe Univ. Group. In this paper, we present evidence of anomalous excess heat effects obtained from experiments at our laboratory at Tohoku University. Excess energy experiments were done using nano-sized metal composites with H_2 or D_2 gas. Anomalous excess heat generation were observed for all the samples at elevated temperature (150–350°C) except for the palladium nanoparticles embedded in mesoporous SiO_2 (PSn1). The amount of anomalous heat generation per hydrogen atom ranged from 15 eV/H or D to 2.1 keV/H or D, which is too much to be explained by any known chemical process. Coincident burst events of pressure and gas temperature were observed for all the experiments using the $CuNi_7Zr_{15}$–O_x with H_2 gas, which suggested sudden energy releases in the reaction chamber. These observations suggest large local energy bursts. Excess heat experiments using the same material at Kobe and Tohoku Universities showed similar experimental results. Qualitative reproducibility between the Kobe and Tohoku experiments was good.

1. Introduction

Akira Kitamura and Akito Takahashi team of Technova Inc. and Kobe University have been studying anomalous heat effects by the interaction of metal nanoparticles and hydrogen isotope gas for several years [1–3]. Based on their results, a new research project started on October 2015 through the collaboration of six Japanese organizations: Technova Inc., Nissan Motor Co. Ltd., Kyushu University, Kobe University, Nagoya University and Tohoku University.

The objective of the collaborative research is to clarify the existence of the anomalous heat generation phenomena and to confirm the reproducibility of the phenomena. For these purposes, anomalous heat experiments at Kobe and Tohoku Universities and sample preparation and analyses at Nissan, Kyushu and Kobe Universities have been performed. Replication experiments were performed at Tohoku University using a high-quality heat measurement system similar to the apparatus at Kobe University.

The Research Center for Electron Photon Science of Tohoku University and CLEAN PLANET Inc. established a collaborative research division in 2015 – the Condensed Matter Nuclear Reaction Division [4] – and research on anomalous heat generation was started. Replication efforts have been made on two types of experiments as a first step at our division in Tohoku University; one is the present collaborative work [1–3], and the other is the experiment using nano-Pd/Ni fabricated by glow discharge with D_2 gas [5].

A summary of the collaborative research is shown in Table 1 and in Fig. 1. Excess energy experiments were done using nano-sized metal composites with H_2 or D_2 gas. The nano-sized metal composite samples are composed of nickel, palladium or copper nanoparticles embedded in ZrO_2 or SiO_2 particles with diameter of several microns to tens of micronsμ [1–3]. Experiments using CNZ($Cu_1Ni_7Zr_{15}$–O_x) with H_2, PNZ($Pd_1Ni_7Zr_{15}$–O_x) with D_2, CNS(Cu_1Ni_{10}/SiO_2) with H_2 and PSn1(Pd/SiO_2) with D_2 were performed. Anomalous excess heat generation was observed for all the samples at elevated temperatures (150–350°C), except for the palladium nanoparticles embedded in mesoporous SiO_2 (PSn1). The amount of anomalous heat generation per hydrogen atom ranged from 10 eV/H or D to 100 eV/H or D, which could not be explained by any known chemical process (Fig. 1). Experiments, Nos. 15 and 16, were performed to demonstrate the reproducibility of this excess heat effect.

2. Experimental

A schematic of the experimental apparatus is shown in Fig. 2; it is based on the instrument described previously in [1] with some improvements. The experimental set-up was described previously in [6,7].

The reaction chamber (RC) that contains nickel-based binary nanocomposites and hydrogen isotope gas is located at the center of Fig. 2. Heat generation from the RC is estimated by mass flow calorimetry. The inlet oil temperature

Table 1. Summary of collaborative research

No.	Place	Sample name	Composition	Gas	Temp. (°C)	Max. power (W)	Released enthalpy
1	Kobe	PS3	Pd/SiO_2	D	200–300	~ 0	~ 0
2	Kobe	PNZ3	$PdNi_7Zr_{15}$–O_x	D	200–300	10	7.7 MJ/mol-D, 80 eV/D
3	Kobe	PNZ3r	PNZ3- re oxidized	H	200–300	8.0	2.0 MJ/mol-H, 21 eV/H
4	Kobe	CNZ5	$CuNi_7Zr_{15}$–O_x	H	200–300	3.3	3.6 MJ/mol-H, 37 eV/H
5	Tohoku	PNZ4s	$PdNi_7Zr_{15}$–O_x	D	160–300	3.3	1.4 MJ/mol-D, 15 eV/D
6	Tohoku	CNZ5s	$CuNi_7Zr_{15}$–O_x	H	160–250	5.0	6.5 MJ/mol-H, 68 eV/D
7	Kobe	PSf1	$Pd/SiO_{2-\text{covered}}$	D	200–300	~ 0	~ 0
8	Tohoku	PSn1	Pd/meso–SiO_2	D	200–300	~ 0	~ 0
9	Kobe	CNS3	$CuNi_{10}/SiO_2$	H	200–400	4.4	67 MJ/mol-H, 700 eV/H
10	Tohoku	CNS3s	$CuNi_{10}/SiO_2$	H	15–300	4.2	11 MJ/mol-H, 120 eV/H
11	Kobe	PNZ5	$PdN_{i7}Zr_{15}$–O_x	D	250–350	4.2	7.6 MJ/mol-D, 70 eV/D
12	Tohoku	CNZ6s	$CuNi_7Zr_{15}$–O_x	H	150–300	2.5	5.3 MJ/mol-H, 55 eV/H
13	Kyushu	PNZ	$PdNi_7Zr_{15}$–O_x	H	23-450	–	–
14	Kobe	PNZ6	$PdNi_{10}Zr_{20}$–O_x	D	250–350	25	200 MJ/mol-D, 2.1 keV/D
15	Kobe	PNZ7k	$PdNi_7Zr_{15}$–O_x	D	250–350	5.0	3.4 MJ/mol-D, 35 eV/D
16	Tohoku	PNZ7s	$PdNi_7Zr_{15}$–O_x	D	250–350	4.0	3.0 MJ/mol-D, 31 eV/D

(T_{in}) is measured by three independent thermocouples and the outlet oil temperature (T_{out}) is also measured by three thermocouples. A oil coolant (BT400; Matsumura Oil Co. Ltd.) enabled the use of the flow-calorimetry method at elevated temperatures. The coolant is driven by a digital liquid tubing pump. A 1 kW sheath heater (W_1) is spirally wound on the outer surface of the RC and a 200 W cartridge heater (W_2) is located at the central axis of the RC to

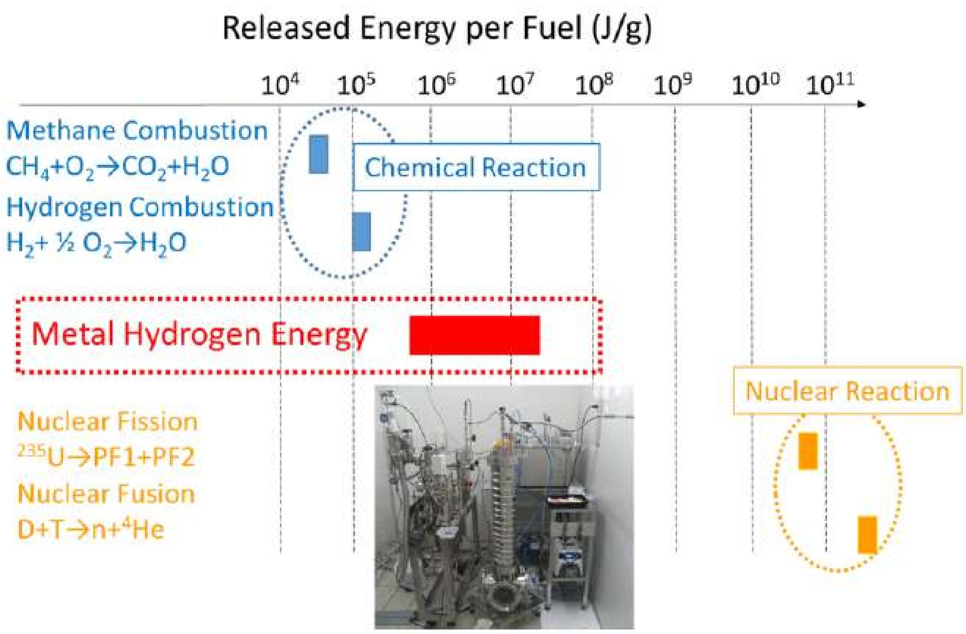


Figure 1. Released energy per gram of fuel; summary of collaborative research.

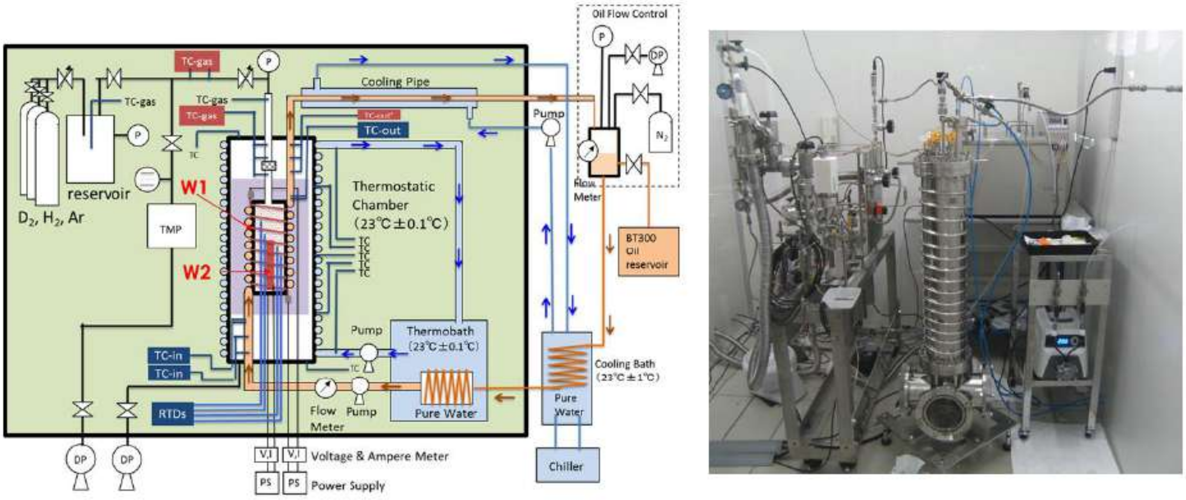


Figure 2. Experimental set-up.

heat the sample in the RC. The power to the heater is supplied by a precision regulated DC power supply. The input electrical power for each heater is monitored by two independent voltage and current meters. H_2 or D_2 gas is fed from a reservoir through a super needle flow regulator to the sample in the RC. Pressures in the RC and the reservoir are monitored continuously. Temperature distribution in the RC is measured by four Resistant Temperature Detectors (RTDs) and temperatures along the oil coolant pipe and the stainless-steel pipe for gas introduction are monitored by thermocouples. Having several temperature measurement points enables us to judge the accuracy of an observed excess heat measurement, although the heat recovery rate of this system rate is low. All the components in this thermostatic chamber are controlled at 23 ± 0.1°C to avoid the influence of outside temperature fluctuations.

Sample preparation and experimental procedures were given in [6,7]. At first, the RC was evacuated by a turbo molecular pump and then heated to 200–300°C remove H_2O or other impurity gases such as hydrocarbons. After baking, the RC is cooled to room temperature. H_2 or D_2 gas were stored in reservoir chambers (1 and 2 L, respectively) at a pressure of about 1 MPa. H_2 or D_2 gas was introduced into the RC by opening the super needle valve.

Initially, we observed H_2 or D_2 gas absorption and heat generation at room temperature due to the presence of palladium. Subsequently, we applied electric power to the heaters located at the inside and outer-surface of the RC to increase the sample temperature. Values of temperature, pressure, voltage, current and a flow rate were logged during experiments. From these measurements, we estimated H or D absorption rates and excess heat generation from the samples. Blank runs to obtain the heat recovery rate of the experimental apparatus were performed using 1 mm-diameter ZrO_2 beads of total mass 1300 g in the RC before or/and after foreground runs. Typical masses of the PNZ and CNZ samples were 100 g.

3. Results and Discussion

3.1. Heat analysis and its error estimation

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 is the heat recovery rate, Q is the heat release rate, $F_{\rm R}$ is oil flow rate, $\rho(T_{\rm ave})$ is the oil density as a function of temperature, $C(T_{\rm ave})$ is heat capacity, $T_{\rm out}$ and $T_{\rm in}$ is 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 temperature dependence of $\rho(T)$ and $C(T)$ is linear, we can postulate that $T_{\rm ave}$ is equal to $(T_{\rm out} + T_{\rm in})$/2.

The value of Q is expressed as

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

where W_1, W_2 and $H_{\rm EX}$ 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, $F_{\rm R}$, $\rho(T_{\rm ave})$, $C(T_{\rm ave})$ and $(T_{\rm out}$–$T_{\rm in})$ are obtained by experimental data. $H_{\rm EX}$ is calculated by a foreground run using the determined η. We simplify Eqs. (1) and (2) to estimate experimental error.

$$H_{\rm EX} = \frac{F_{\rm R}\rho C}{\eta}\Delta T - W, \qquad \Delta T = T_{\rm out} - T_{\rm in}, \qquad W = W_1 + W_2.$$

Considering that experimental variables are $F_{\rm R}$, 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 power.

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

Experimental data show that the largest contribution to the error of $H_{\rm EX}$ is the $F_{\rm R}$ term; W is the most stable parameter.

3.2. Burst-like coincident increase events of chamber pressure and gas temperature

During the collaborative work, we observed interesting coincident burst events of the pressure in the reaction chamber and gas temperature for CNZ5s and CNZ6s experiments in Table 1, which suggested sudden energy releases in the reaction chamber. After the period of collaborative research, we set out to replicate the phenomena using a re-oxidized CNZ5s sample. As shown in the last column of Table 2, we were able to replicate this. A summary of these experiments is shown in Table 2. These samples contain the same compositions although sample treatment was different for the No. 3 sample. The CNZ5sR sample (No. 3) was prepared by oxidizing the used CNZ5s sample (No. 1) in air for 180 h at 450°C.

Released excess enthalpy for these CNZ samples was 3.3– 6.5 MJ/mol-H, which is too much to be explained by any known chemical reactions. Furthermore, coincident burst events of chamber pressure and gas temperature were

Table 2. Summary of CNZ experiments at Tohoku university.

No.	Samples	Date	Temp. (°C)	Max. power (W)	Released excess enthalpy	Coincident increase of Pr and E2
1	CNZ5s ($CuNi_7Zr_{15}$–O_x)	05–19 Aug. 2016	160–250	5.0	6.5 MJ/mol-H 68 eV/H	Yes; $W_1 = 134$ W, $W_2 = 0$
2	CNZ6s ($CuNi_7Zr_{15}$–O_x)	1 Mar. 1 to 18 April, 2017	150–350	2.5	5.3 MJ/mol-H 55 eV/H	Yes; $W_1 = 80$ W, $W_2 = 0$
3	CNZ5sR ($CuNi_7Zr_{15}$–O_x (Re-oxidized)	11–30 May, 2018	140–260	2.6	3.3 MJ/mol-H 34 eV/H	Yes; $W_1 = 134$ W, $W_2 = 0$

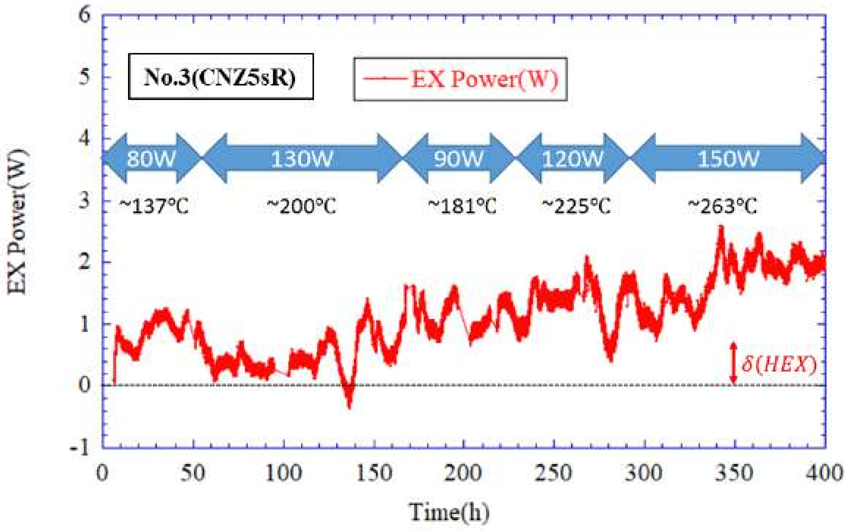


Figure 3. An example of excess heat generation; CNZ5sR with H_2.

observed in all cases. Excess heat data for CNZ5sR is shown as an example in Fig. 3. Total input power $(W_1 + W_2)$ and rough temperature in the RC is shown. Although the measured excess power during 130 W input power was not obvious, excess power continued for more than 200 h. The error range of the excess power for 150 W input based on Eq. (3) is shown in Fig. 3. The error ranges for other input power levels are smaller than the range for 150 W input.

Figure 4 shows blank run data for the pressure of RC (Pr) and upper pipe temperature (E2). The pipe is connected

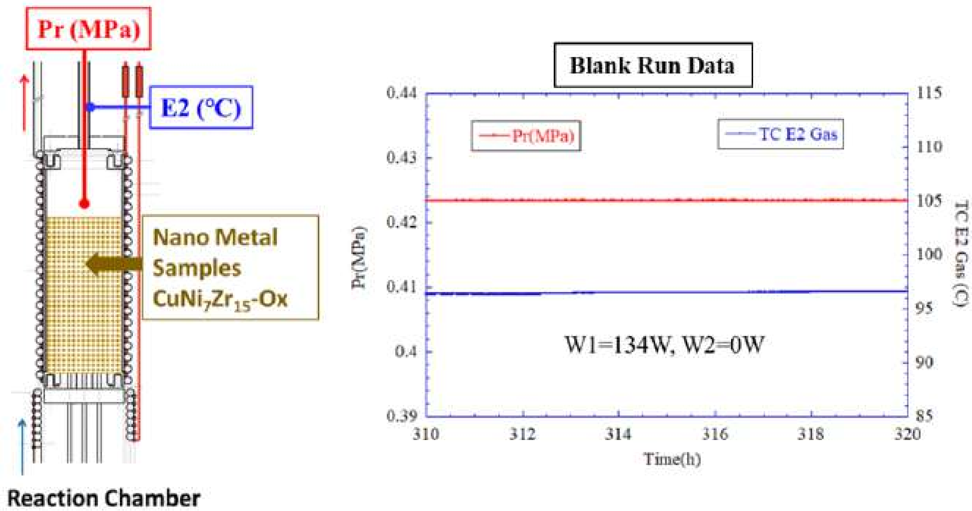


Figure 4. Blank run data for reaction chamber pressure (Pr) and upper pipe temperature (E2).

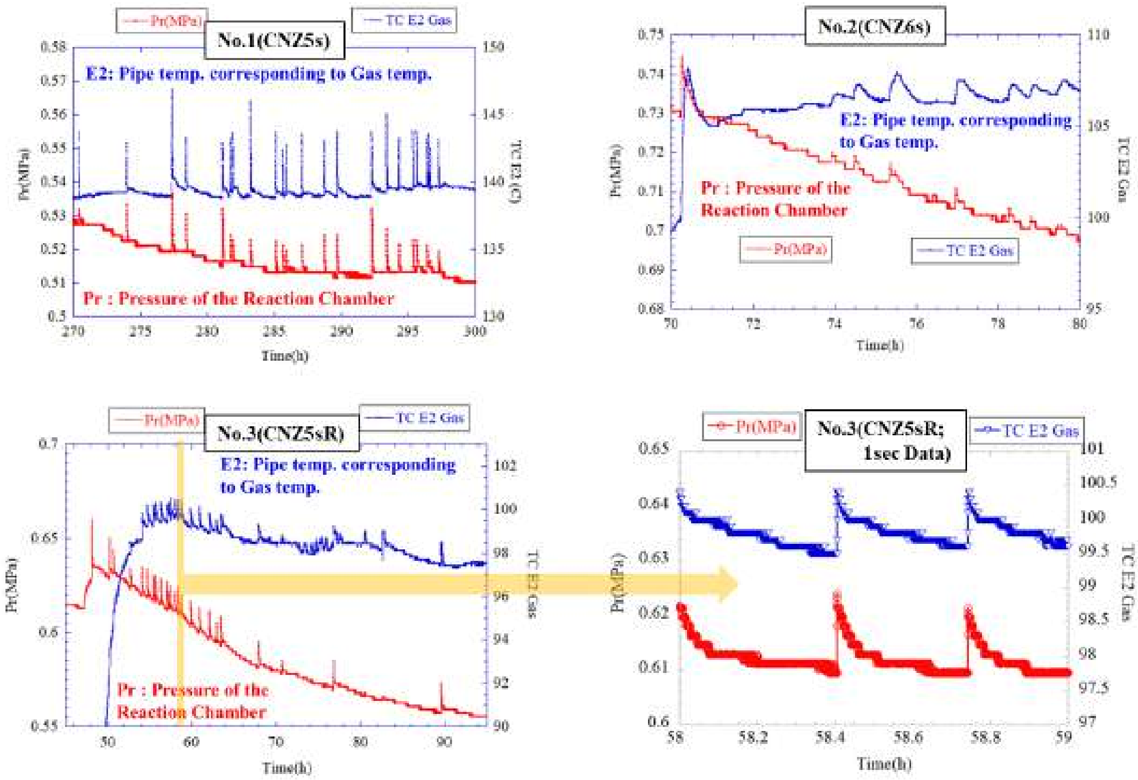


Figure 5. Burst-like coincident increase events of chamber pressure (Pr) and upper side temperature (E2).

to the hydrogen gas reservoir through a super needle flow regulator as shown in Fig. 2. A pressure gauge is connected to the pipe to measure the pressure of RC (Pr) and the surface temperature is monitored by thermocouple (E2). Pr and E2 are kept nearly constant under constant input power and oil flow rate conditions as shown in Fig. 4.

During certain test runs, coincident burst events of Pr and E2 were observed. These events were observed when we used the $CuNi_7Zr_{15}–O_x$ sample with H_2 gas when the cartridge heater (W_2) power was set to zero. In other words, the coincident events occurred when the input power was supplied by the sheath heater (W_1), which was wound on the outer surface of the RC when it contained $CuNi_7Zr_{15}–O_x$ nano-particles.

These coincident burst events occurred at random for the three experiments in Table 2; the data for these events are given in Fig. 5. The pressure and temperature increases were significant and cannot be explained by noise. For experiment No. 3, an expanded time scale is provided. For Pr and E2 the data acquisition interval was 1 s. Examining these two coincident events closely, we see that the pressure in the RC increased first, followed by a temperature increase of the pipe surface few seconds later. Noting that the time response of the pressure gauge is faster than the thermocouple attached to the pipe, it is reasonable to assume that high temperature H_2 gas was emitted intermittently.

Although it is difficult to interpret the data at the upper end of the RC, coincident temperature and pressure rises suggest burst energy releases from the RC. The temperature distribution in the RC and the temperature gradient in the metal nanocomposites must play some role in inducing these events, as these events were observed only for $W_2 = 0$. An important observation is that these coincident burst events were replicated qualitatively.

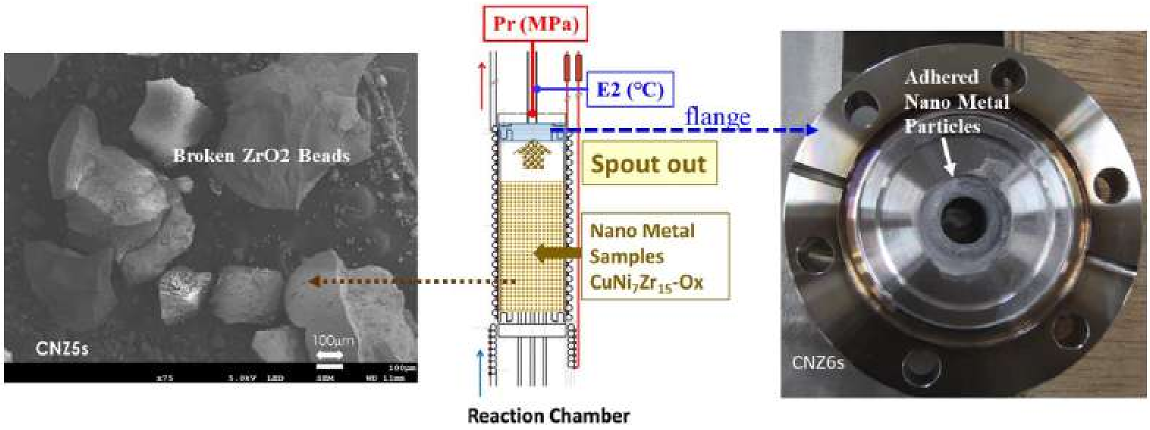


Figure 6. Examples of broken ZrO_2 beads and adhered nano-material on the upper flange after excess heat generation.

Other experimental observations supporting the assumption of burst energy releases are shown in Figs. 6 and 7. After completing these CNZ experiments, the metal nanocomposite samples were sieved out to separate them from the 1-mm diameter ZrO_2 beads. We found that broken parts of ZrO_2 beads were mixed with the sample as shown in the left photo of Fig. 6. ZrO_2 beads are very hard and difficult to break; they are used to grind materials into a powder. This suggests that large local stresses, presumably due to heat bursts, shattered the ZrO_2 beads.

Other evidence that supports the assumption of burst energy release is shown in the right photo of Fig. 6. This photograph shows the lower side surface of the upper flange at the conclusion of the CNZ6s experiment. Material adhered to the surface around the pipe of the upper flange. This white substance was dissolved by HNO_3 and subsequently analyzed by ICP-MS. Figure 7 shows the results of ICP-MS analysis of this material. Reference data for HNO_3 solutions are plotted in black and blue lines and test material data are plotted in red. This analysis shows large clear peaks for Cu, Ni and Zr. As the flange is made of stainless steel, this analysis suggests that $CuNi_7Zr_{15}$–O_x particles were embedded in the flange. This observation is consistent with the assumption of burst energy release and suggests that

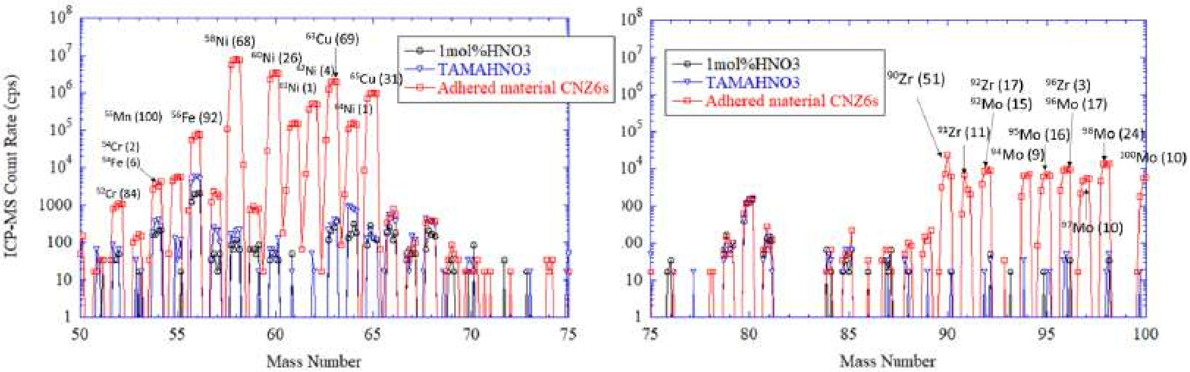
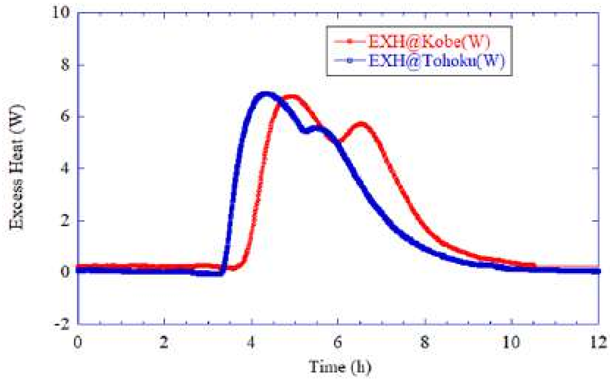


Figure 7. ICP-mass analysis for the adhered nano-metal particles on the upper flange.



	Kobe	Tohoku
Released Enthalpy (KJ/mol-D)	53.7	50.9
Released Enthalpy (eV/D)	0.56	0.53

Figure 8. Comparison of released heat power between Kobe and Tohoku at room temperature..

certain condensed matter nuclear reactions occur intermittently in or around the metal nanocomposites with hydrogen.

3.3. Experimental reproducibility at different site using same metal nano-composites

As shown in Table 1, PNZ, CNZ and CNS metal nano-composite samples showed anomalous energy generation that cannot be explained by known chemical reactions. However, palladium nano-particles expressed as PSf1 and PSn1 did not show any anomalous phenomena. These experimental results indicate rough experimental reproducibility. To better test reproducibility, we performed experiments at Kobe and Tohoku University using the same $PdNi_7Zr_{15}$–O_x with D_2 gas.

Figure 8 shows a comparison of released heat between Kobe and Tohoku at room temperature. The released heat power was calculated based on the difference between inlet and outlet coolant temperatures as was done for the

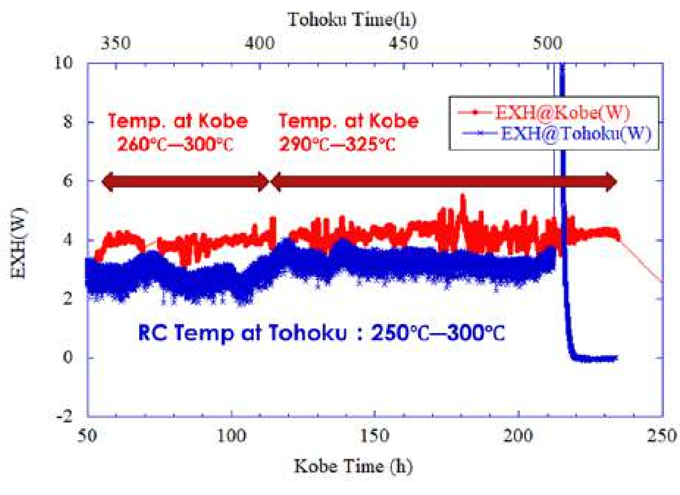


Figure 9. Comparison of excess heat power between Kobe and Tohoku at elevated temperature.

experiments at elevated temperatures. The heat power has two peaks, because D_2 gas was re-pressurized to the RC after the initial absorption of D_2 gas. After the resupply, the pressure of D_2 the absorption rate of D_2 into the $PdNi_7Zr_{15}$–O_x sample increased and enhanced the rate of heat release associated with absorption of D_2. Released excess enthalpies obtained at Kobe and Tohoku Universities agree.

Figure 9 shows a comparison of excess heat power between the experiments at elevated temperatures performed at Kobe and Tohoku Universities. As the excess power obtained by the present method has strong correlation with the temperature in the RC, we compared excess heat at similar RC temperatures. The same level of excess power were observed for similar RC temperatures. These experimental results demonstrate that a similar level of excess power can be obtained if we use the same metal nanocomposites and similar experimental setups.

4. Concluding Remarks

The Collaborative Research Project between six parties on anomalous heat effects was done from Oct. 2015 to Oct. 2017. Anomalous heat generation was observed for all the samples at elevated temperature, except for the palladium nanoparticles. Integrated excess heat reached 1.4–200 MJ/mol-H(D), which is too much to be explained by any known chemical process. Coincident burst events of the reaction chamber pressure and gas temperature were observed many times using $CuNi_7Zr_{15}$–O_x with H_2 gas. This suggested burst energy releases in the reaction chamber. These burst-like events were observed three times and suggest large local heat releases. Qualitative reproducibility was good between the Kobe and Tohoku experiments.

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