J. Condensed Matter Nucl. Sci. 33 (2020) 1–13

Research Article

Excess Energy Generation using a Nano-sized Multilayer Metal Composite and Hydrogen Gas

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Abstract

New type of excess heat experiments using a nano-sized metal multilayer composite and hydrogen gas have been performed based on the permeation-induced transmutation experiments with multilayer thin film and excess heat experiments with nano-particles. Two nano-sized metal multilayer composite samples, which were composed of Ni, Cu, CaO, Y_2O_3 thin films on bulk Ni (25 mm $\times$ 25 mm $\times$ 0.1 mm), were placed in a vacuum chamber. These samples were fabricated by Ar ion beam sputtering method. After baking of the samples, H_2 gas was introduced into the chamber up to about 230 Pa at 250°C. Then, the Ni based multilayer thin films started to absorb H_2 gas. Amount of absorbed H_2 gas can be evaluated by the pressure measurement of the chamber. Typically, after about 50,000 s, H_2 gas was evacuated and simultaneously the samples were heated up by the ceramic heater up to 500–900°C. The evacuation and heating process seem to trigger heat generation reactions. Heat burst phenomena were simultaneously detected by a radiation thermometer looking at the surface of the multilayer thin film and a thermocouple located near the metal composite. It shows that heat measurement by the thermocouple embedded in the ceramic heater correctly reflects surface temperature detected by the radiation thermometer. Excess energy generation using nano-sized multilayer Cu/Ni metal composite and Cu/Ni metal with third material (CaO, Y_2O_3) composite were presented. Maximum released excess energy reached 1.1 MJ and average released energy per absorbed total hydrogen was 16 keV/H or 1.5 GJ/H-mol. It cannot be explained by any known chemical process and suggests that the observed heat generation must be of nuclear origin. Various analysis methods, such as SEM-EDX or TOF-SIMS, had been applied to obtain information about what kind of reactions occur by the interaction of the nano-sized multilayer metal composite with hydrogen gas.
 ISSN 2227-3123

Keywords: Anomalous heat, Excess heat, Gas loading, Heat burst, Hydrogen gas, Multilayer thin film, Nano material, Nano-sized metal composite

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1. Introduction

Permeation-induced transmutation phenomenon, which is completely different from conventional transmutation by nuclear reactors or accelerators, was first reported in 2002 [1]. D_2 gas permeation through a nano-structured multilayer thin film composed of Pd and CaO thin film and Pd substrate with a target element induces nuclear transmutation reactions [2–4]. The temperature of the Pd complex is typically 70°C and the pressure of D_2 gas is 1 atm. Permeation-induced nuclear transmutation reactions were firstly observed at Mitsubishi Heavy Industries and were successfully replicated by other institutes such as Toyota R&D center [5]. Typical target element is Cs and produced element is Pr. Transmutation reactions of Sr, Ba, W into Mo, Sm, Pt were also observed by this method. It seemed that 2, 4 or 6 deuterons make fusion with the target materials. In this research, deuterium diffusion through nano-sized multilayer thin film was a key factor and the elemental analysis was important technique.

The Collaborative Research Project between six Japanese organizations funded by New Energy and Industrial Technology Development Organization (NEDO) on anomalous heat effects was done from Oct. 2015 to Oct. 2017 using Ni, Pd, Cu, and Zr nano-particles. Anomalous heat generation, which is too much to be explained by any known chemical process, was observed. Qualitative reproducibility was confirmed between the Kobe University and Tohoku University [6–9]. The authors replicated the experiments using nano-Pd/Ni fabricated by glow discharge with D_2 gas developed by Mizuno [10]. In these experiments, nano-sized particles and diffusion of hydrogen and deuterium were one of key factors to observe the heat effects and precise heat estimation was crucial.

Combining above factors and methods, we developed a new type of excess heat experiments using a nano-sized metal multilayer composite and hydrogen gas. Larger excess energy per H was obtained using the present method.

2. Experimental

A schematic of the experimental set-up is shown in Fig. 1(a). Two nano-sized metal multilayer composites were placed in the center of the chamber. The chamber, made of stainless steel (type 304), had two ports for introducing and evacuating gas, respectively. H_2 gas and its pressure were monitored by a Pirani gauge. The chamber could be evacuated by a turbo molecular pump. The multilayer samples could be heated up by a ceramic heater (MS-1000R; Sakaguchi E. H Voc Corp.) in which a thermocouple (Pt-PtRh13%) was embedded. Heater temperature was measured by the thermocouple. The surface temperature of a sample was evaluated by an infrared radiation thermometer (IR-CAQ3CS; Chino Corp.). The detector was made of InGaAs and two wavelengths, 1.55 and 1.35 μm, were used in this work. Gamma-rays were monitored by a NaI (Tl) scintillation counter (TCS-1172; Hitachi, ltd.) during all experiments, for safety. Heater input power was supplied by a DC power source with constant voltage mode. The input voltage and current were measured both by voltage and current monitors provided by the power supply and an independent voltmeter and amperemeter, respectively.

A detailed drawing of the Ni based nano-sized metal multilayer composite is shown in Fig.1(b). It was composed of a Ni Plate (25 mm square and 0.1 mm thickness) and Cu–Ni multilayer thin film (25 mm diameter circle and few 10 nm thickness). Two nano-sized metal multilayer composite samples were heated by the ceramic heater (25 mm square and 2.2 mm thickness) through SiO_2 plates (0.1 mm thickness). If certain energy generation reactions would happen on the surface of samples, the temperature of the embedded thermocouple (heater temperature) would rise. Simultaneously, infrared emission detected by the radiation thermometer, which corresponds to surface temperature of the sample, would increase. Photos of the experimental set-ups and STEM image of Cu-Ni multilayer thin film are shown in Fig. 2.

A Ni plate (25 mm square and 0.1 mm thickness, purity up to 99.9%, Furuuchi Chemical Co.) was washed with acetone and annealed in vacuum ($<10^{-4}$Pa) at 900°C for 72 h. It was then cooled to room temperature in a furnace and washed with HNO_3/H_2O to remove impurities on the surface of the Ni plate. The surface of the plate was covered by layers of Cu (2 nm) and Ni (14 nm) which were obtained by alternatingly sputtering 2 nm thick Cu and 14 nm

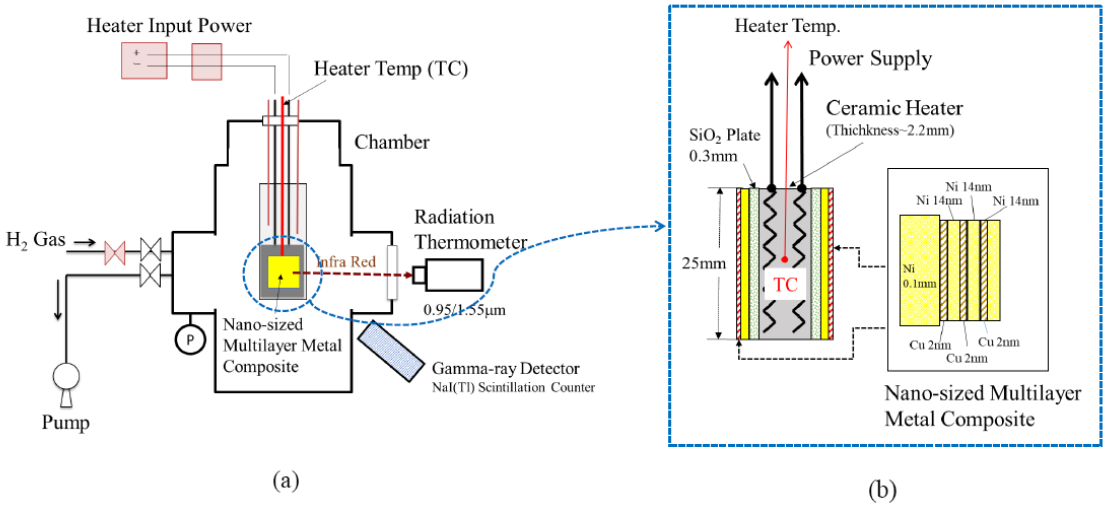


Figure 1. Experimental set-up. (a) Schematic of experimental apparatus, (b) detail drawing around nano-sized multilayer metal composite.

thick Ni layers. 2 nm thick CaO or Y_2O_3 thin films were inserted into the Ni layers in some cases. The thicknesses of the layers were measured by a crystal thickness monitor. Fabrication process is performed by Ar ion beam sputtering.

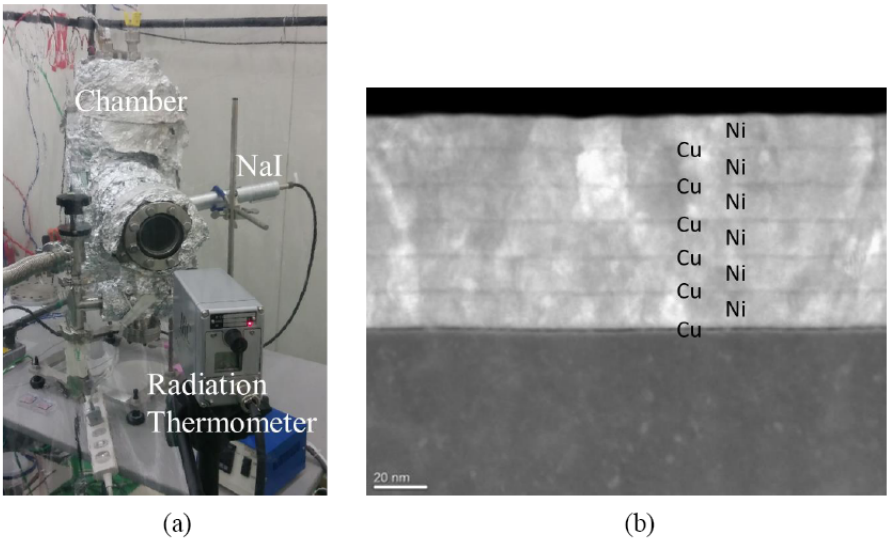


Figure 2. Photos of experimental apparatus and cross-sectional view of nano-sized metal multilayer composites; (a) outer view of the present experimental set-ups, (b) STEM (scanning transmission electron microscope) image of Cu–Ni multilayer thin film.

The samples are sputtered at room temperature and the voltage and current of the Ar beam are 1 kV and 10 mA, respectively.

The experimental procedure is as follows. Two nano-sized metal multilayer composites were placed in the chamber and baked for 1–2 days at temperature more than 200°C to remove H_2O on the surface *under vacuum condition*. After baking, H_2 gas was introduced into the chamber up to about 230 Pa at 250°C. Usually H_2 gas was loaded for about 16 h. Then, H_2 gas was evacuate by the turbo molecular pump and simultaneously the samples were heated up by the ceramic heater up to 500–900°C. These process triggers heat generation reactions and observed excess heat. Typically, after 8 h, the heater input was turned down and the samples were made cool down to 250°C. These processes (H_2 loading, heating up and cooling down samples) were repeated several times changing heating temperature.

During the above experimental procedure, hydrogen atoms are supposed to diffuse from the Ni plate through the nano-sized metal multilayer to the surface. The diffusion mechanism of hydrogen atoms is well known as "quantum diffusion" [11]. Hydrogen atoms are hopping from a site to another site in metal. We assume that hydrogen flux is one of the key factors to induce condensed matter nuclear reactions and the hydrogen flux is intentionally arranged by the present experimental method. Hydrogen flux **J** from the nano-sized metal multilayer composite to the chamber is caused by gradient of hydrogen concentration and gradient of temperature as shown in Eq. 1 [12].

$$\mathbf{J} = -nD\left(\nabla c + \frac{cQ^*\nabla T}{k_{\mathrm{B}}T^2}\right), \tag{1}$$

where n is the number of lattice atoms per unit volume, c is hydrogen concentration defined as the hydrogen/host-metal atom ratio, D is diffusion coefficient and Q^* is the heat of transport.

3. Results and Discussion

3.1. Excess heat estimation

Heat analysis of this system is based Eq. (2) as explained in Fig. 3(a).

$$k\frac{T_{\mathrm{H}}-T_{\mathrm{w}}}{L}A_{\mathrm{H}}+2A_{\mathrm{S}}\varepsilon\sigma\left(T_{\mathrm{S}}^4-T_{\mathrm{w}}^4\right)=P_{\mathrm{in}}+H_{\mathrm{ex}}, \tag{2}$$

where k is thermal conductivity, T_{H} the heater temperature, T_{S} the surface temperature, T_{W} wall temperature of the chamber, L the length between the heater and wall, A_{S} surface area of the sample, ε the emissivity of the sample, σ the Stefan–Boltzmann constant, P_{in} the electrical heater input and H_{ex} is excess power that is generated heat power by the condensed matter nuclear reactions. This equation is obtained under the following assumptions.

(1) Thermal conduction via H_2 gas is negligible as H_2 pressure is low enough.
(2) Radiation from chamber wall is negligible because T_{W} is room temperature.
(3) The electrical input power is constant. blank run, in which same sized Ni bulk.

A blank run, in which same sized Ni bulk samples without multilayer thin films were used, was performed with the same procedure described above. Figure 3(b) shows the relationship between input power given to the ceramic heater and heater temperature detected by the thermocouple. Generated excess heat power can be evaluated based on the blank run result. Generated heat can be evaluated by Eq. (1).

The radiation thermometer was introduced recently, so we now have many experimental results with the heater temperature only. As a first step of data analysis, experimental data analysis was done based on the assumption that ε is constant for Ni based nano-sized metal multilayer composite and Ni bulk. In the next step, excess heat will be evaluated more preciously by measuring ε for each sample.

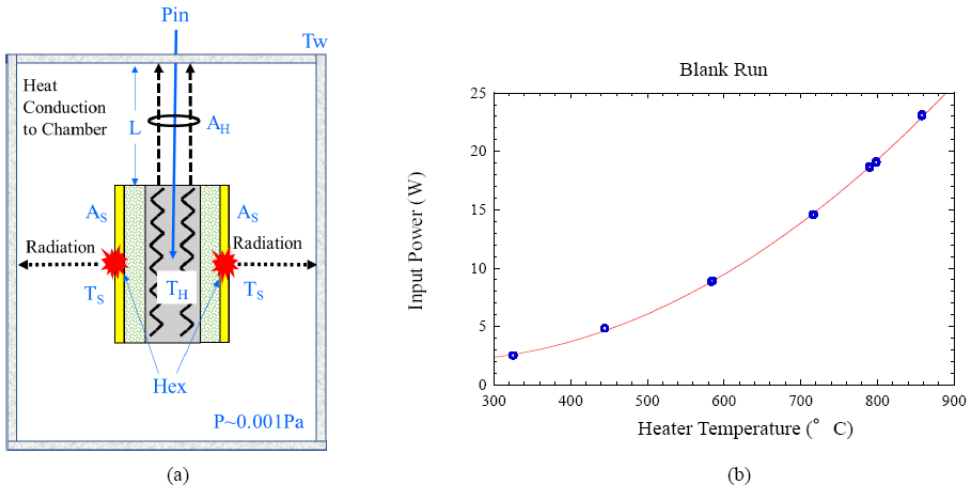


Figure 3. Excess heat evaluation; (a) Model of excess heat evaluation, (b) Relationship between input power (W) and heater temperature for blank run.

3.2. Excess heat generation and its evaluation

Examples of experimental results are shown in Figs.4 and 5. The structures for the samples were shown in Fig. 4. The ratio of Ni and Cu was the same for the four samples; Cu/Ni=1/7 in these examples. It was selected based on the past NEDO project results [7–9]. Also CaO and Y_2O_3 were selected based on the permeation-induced transmutation results [1–4].

Figure 5 shows pressure of the chamber and excess heat for each sample. Red and blue lines mean excess heat

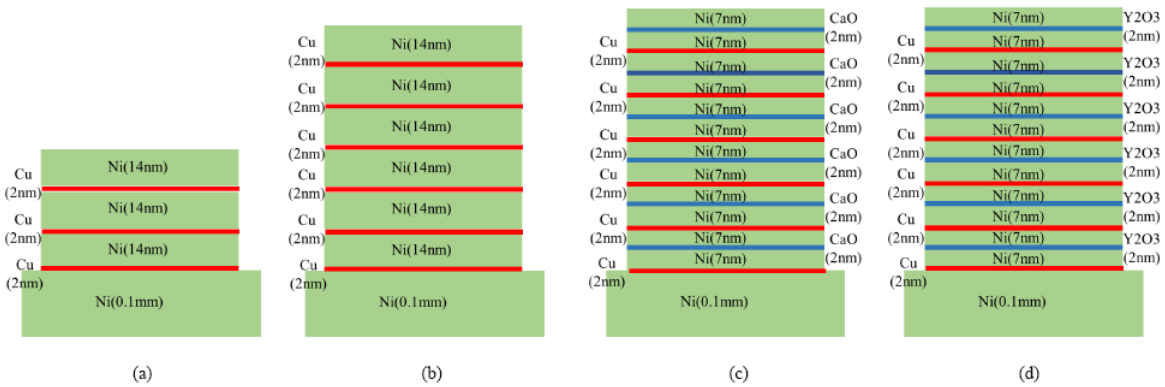


Figure 4. Structures of nano-sized multilayer metal composite samples; (a) example #1: 3 layers of Cu 2 nm and Ni 14 nm, (b) Example #2: 6 layers of Cu 2 nm and Ni 14 nm, (c) Example #3: 6 layers of CaO 2 nm, Cu 2 nm and Ni 14 nm, (d) Example #4: 6 layers of Y_2O_3 2 nm, Cu 2 nm and Ni 14 nm.

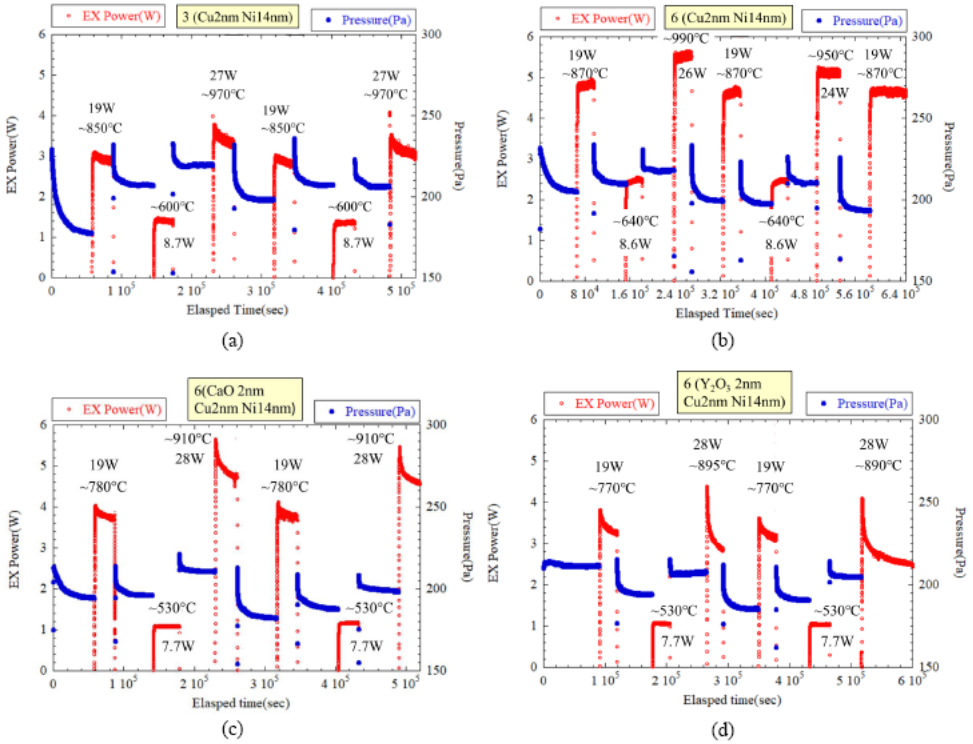


Figure 5. Examples on excess heat generation; (a) example #1: 3 layers of Cu 2 nm and Ni 14 nm, (b) example #2: 6 layers of Cu 2 nm and Ni 14 nm, (c) example #3: 6 layers of CaO 2 nm, Cu 2 nm and Ni 14 nm, (d) example #4: 6 layers of Y_2O_3 2 nm, Cu 2 nm and Ni 14 nm.

and pressure of the chamber, respectively. At the beginning of each experiment, hydrogen gas was introduced to the chamber and absorbed into the Ni based nano-sized multilayer metal composite at 250°C. The pressure for each experiment gradually decreased as shown in the Fig. 5. The amount of hydrogen absorbed by each sample was estimated based on the pressure change and temperature *of* the chamber. After about 16 h, H_2 gas was evacuated and simultaneously each sample was heated up by the ceramic heater. After that, excess heat more than input power was observed for each experiment as shown in Fig. 5. The input power for each experiment was stable during a one cycle; for example, 19 W was applied to the ceramic heater from about 6×10^4 s to about 9×10^4 s in Fig. 5(a).

Comparing experiments (a)–(d), excess heat for experiment (b) was largest. Although input power for the first excess heat event (from 6×10^4 to 9×10^4 s) in Fig. 5(a) and (b) was the same (19 W), heater temperatures were about 850°C and 880°C, respectively. A significant temperature difference was observed between the two cases. For each experiment (c) and (d), which CaO and Y_2O_3 was inserted to each Ni film, excess heat was different. Although input power for the third excess heat event (CaO: from 2.3×10^6 to 2.6×10^6 s, Y_2O_3: from 2.6×10^6 to 2.9×10^6 s) in Fig. 5(c) and (d) was the same (28 W), heater temperatures were about 910°C and 895°C, respectively. A significant temperature difference was also observed between the experiments (c) and (d). It should be noted that the properties of heat dissipation were different for experiments (a) and (b) and experiments (c) and (d) because experimental set-ups were different for the two cases.

Time dependence of excess heat for the four experiments was different; excess heat tended to increase for the 6 Cu

Table 1. Released excess energy per hydrogen for the four examples.

	#1 (three layers of Cu–Ni)	#2 (six layers of Cu-Ni)	#3 (six layers of CaO–Cu–Ni)	#4 (six layers of Y_2O_3–Cu–Ni)
Absorbed H (mol)	7.6×10^{-4}	7.3×10^{4}	5.1×10^{4}	3.7×10^{4}
Absorbed number of H	4.6×10^{20}	4.4×10^{20}	3.0×10^{20}	2.3×10^{20}
Total excess energy (J)	4.6×10^{5}	1.1×10^{6}	5.9×10^{5}	2.6×10^{5}
Excess energy per H (J/H-mol)	6.0×10^{8}	1.5×10^{9}	1.2×10^{9}	6.8×10^{8}
Excess energy per H (eV/H)	6.3×10^{3}	1.6×10^{4}	1.2×10^{4}	7.1×10^{3}

and Ni-layer experiment (b), however, excess heat decreased for the other experiments. We can see that excess heat power and its time dependence changed depending on the structure the multilayer thin film region.

Figure 6 illustrates the temperature dependence on the excess heat power for the four examples. Excess heat tended to increase as temperature increase, however, Y_2O_3 sample (Example #4) take a peak around 750°C. Temperature dependencies for the four samples looks like convex shapes. They did not depend on fourth power of temperature and it suggests that these observed excess heat powers were not attributed to the artifacts caused by thermal radiation.

Released excess energies per hydrogen for the four examples were evaluated based on the experimental results. Table 1 summarizes the amount of absorbed hydrogen, total excess energy and excess energy per absorbed hydrogen. The amount of excess energy was calculated by the time integration of excess powers for each experiment. They ranged from 0.26 to 1.1 MJ. Although it seems highly unlikely that all the absorbed hydrogen atoms reacted, we can still estimate that average released energies per absorbed total hydrogen for examples #1–#4 were 6.3 keV/H, 16 keV/H, 12 keV/H and 7.1 keV/H, respectively. Obviously, the released excess energy per hydrogen atom for all the experiments demonstrated here are too large to be explained by any known chemical reactions. This strongly suggests

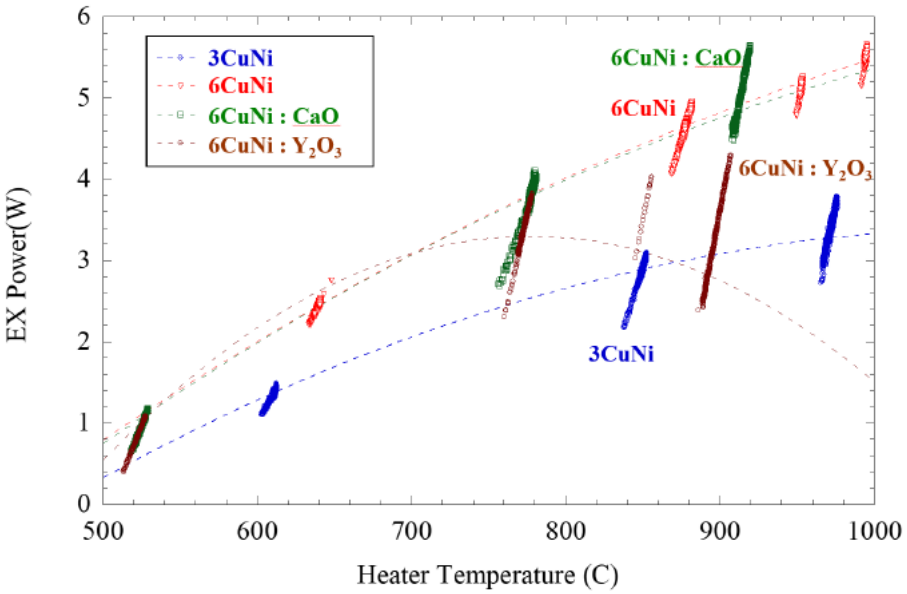


Figure 6. Temperature dependence on excess power for four examples.

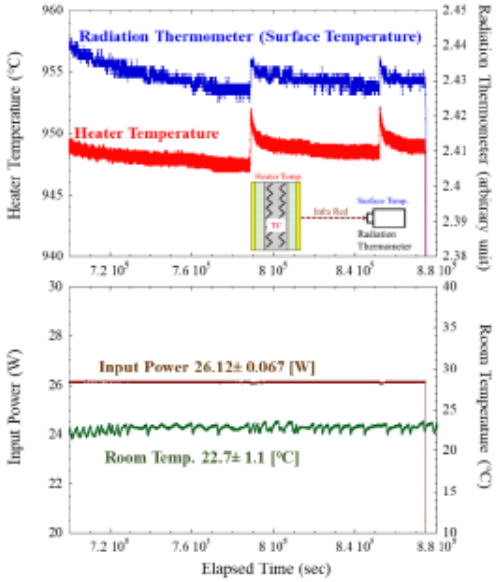


Figure 7. Simultaneously detected by a radiation thermometer looking at the surface of the multilayer thin film and a thermocouple located in the center of heater.

that some condensed matter nuclear reactions were induced in our experiments, although it is not clear at present what kind of reactions occurred. According to the results, about 10^{-3} of the absorbed hydrogen might react if the condensed matter nuclear reactions would release energy order from 1 to 10 MeV. Identification of reaction products is very important future work, in addition to more preciously excess heat evaluation.

Heat burst phenomena were observed by the radiation thermometer and the thermocouple in the heater simultaneously as shown in Fig.7. Output from the radiation thermometer, which corresponds to the surface temperature of samples, is plotted as the upper blue line, and heater temperature is plotted as the lower red line in the upper graph of Fig.7. Input electrical power and room temperature around the chamber are drawn in the lower graph. A significant simultaneous increase of surface and heater temperature were observed twice, while no significant changes for input electrical power and room temperature were detected during these burst events. This means that heat measurement by the thermocouple embedded in the ceramic heater correctly reflected surface temperature detected by the radiation thermometer.

3.3. Analysis of nano-sized multilayer metal composite

It is very important to identify what kind of reactions occur by the interaction of the nano-sized multilayer metal composite with hydrogen gas. Scanning Electron Microscope–Energy Dispersive X-ray Spectroscopy (SEM–EDX), Inductively Coupled Plasma–Mass Spectrometry (ICP–MS), Time-of-Flight Secondary Ion Mass Spectrometry (TOF-SIMS), Quadrupol Mass Spectrometer (Q-Mass), X-ray Photoelectron Spectroscopy (XPS), Transmission Electron Microscope (TEM), Scanning Transmission Electron Microscope (STEM) and the other analysis methods have been applied to the nano-sized multilayer metal composite before and after experiments and released hydrogen gas as shown in Table 2. We would like to search for reaction products and investigate what is happening in the nano-sized multilayer metal composite. It is considered that the material analysis leads to obtain key factors to control condensed matter

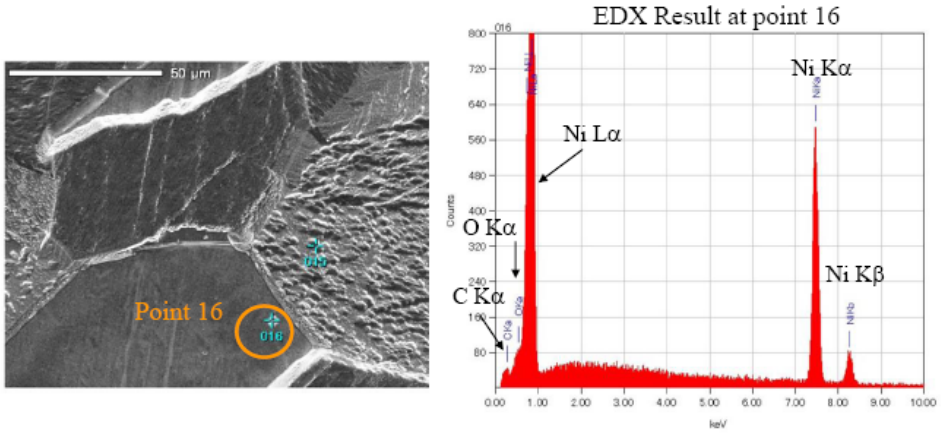
Table 2. Analysis methods for understanding reaction mechanism.

Method	Outline
SEM-EDX	Surface shape and atomic electron structure
ICP-MS	High sensitivity mass spectrometry
TOF-SIMS	High resolution mass spectrometry
Q-Mass	Mass spectrometry for released gas
XPS	Surface sensitive photo electron spectrometry
TEM, STEM	Cross sectional view of nano scale material

nuclear reactions.

The SEM and EDX results for a blank run Ni plate sample are shown in Fig. 8. Model JSM-6500F by JEOL was used for the analysis and acceleration voltage was 15 kV. The analysis result for the point 16 in the SEM image is shown. Ni, O and C peaks are detected by EDX and we understand Ni is the main element. As C is the common contamination element and the sample after blank run was exposed to the air, this is a reasonable result.

Figure 9 shows SEM and EDX results of the sample with six layers of Cu 2 nm and Ni 14 nm (Example #2) before and after the excess energy generation experiment. Before the experiment, Ni, Cu, O, C and W were observed as shown in Fig. 9(a). As W (tungsten) was used as a filament of ion beam sputtering device, it was possible to consider that W was detected by EDX. In contrast, many elements after heat generation experiment were detected by EDX. Ni, Cu, O, C, Si, Na, Mg, Al and K were detected, although each element should be confirmed by other method. Ni, Cu, O and C are reasonable to be detected, however, origins of Na, Mg, Al and K are difficult to identify. Although Si is one of common elements and often detected as a contaminant, amount of Si in this case seems too much. Ni, Cu, O and C were detected on almost all the analysis points for the sample with six layers of Cu 2 nm and Ni 14 nm, however, Na,



Element	Energy(keV)	Mass(%)	Error(%)	Atomic(%)
C (K)	0.277	0.80	0.05	3.80
O (K)	0.525	0.06	0.05	0.22
Ni (K)	7.471	99.13	0.56	95.98

Figure 8. SEM and EDX results of blank run Ni plate.

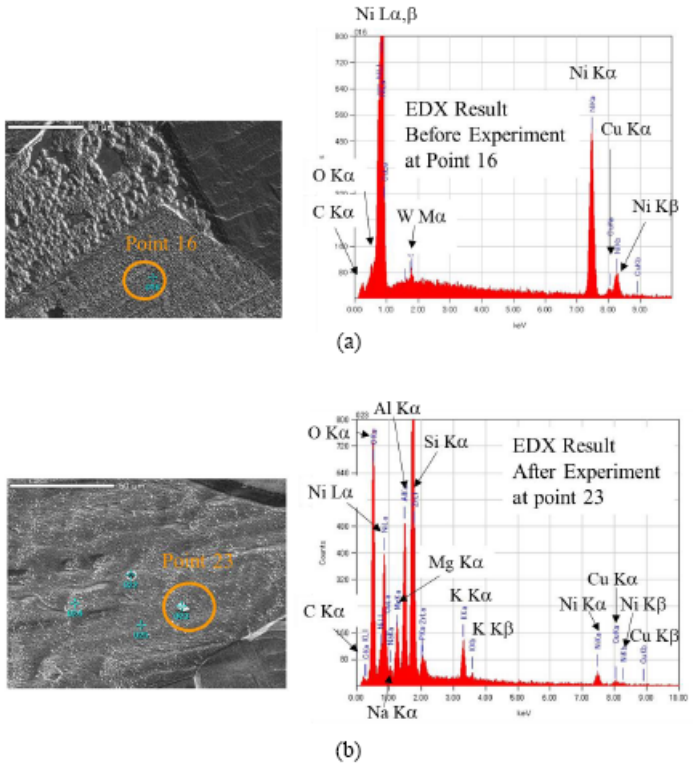


Figure 9. SEM and EDX results of the sample with six layers of Cu 2 nm and Ni 14 nm; (a) before experiment, (b) after experiment..

Mg, Al and K were detected at a few points. Of course, they are common elements and we should take consideration of many contamination chances and careful analysis is necessary.

TOF–SIM was applied for the sample with six layers of Cu 2 nm and Ni 14 nm (Example #2). Elemental surface distribution analyzed by TOF-SIMS is shown in Fig. 10. SIMS 5 by ION-TOF GmbH was used for the analysis and primary ion was Bi+. Ion energy was 25 kV, ion current was 1.0 pA and the field of view was 101.6$\times$ 101.6 μm^2.

Non-uniform distribution for each element was observed. Lighter place means larger signal strength. Na and K were detected at similar places but not corresponded exactly. A hot spot for Ca overlapped with higher places for Na and K. A hot spot for Ti overlapped with K. At present, we do not have an explanation for the distributions for Na, Ca, K and Ti.

We can see that Mn TOF–SIMS signal is strong while Fe signal is weak at the same region. Fe is a common element and Fe can be contaminated easily, because Fe is main element of stainless steel. Mn is also contained in stainless steel. However, inverse surface distributions for Fe and Mn does not give us reasonable explanation if we would postulate that Mn and Fe were derived from stainless steel.

Example of depth profiles of the sample with six layers of Cu 2 nm and Ni 14 nm with Y_2O_3 is shown in Fig. 11. TOF-SIMS device is the same as described in Fig.10 and the sputter Ion was O^{2+}, energy and current were 1 kV and 120 nA, respectively. According to the depth profile of Y (yttrium), we can see that multilayer structure remained partially. Fe, Cr and Mn, which could be supposed to be derived from stainless steel during sputtering processes,

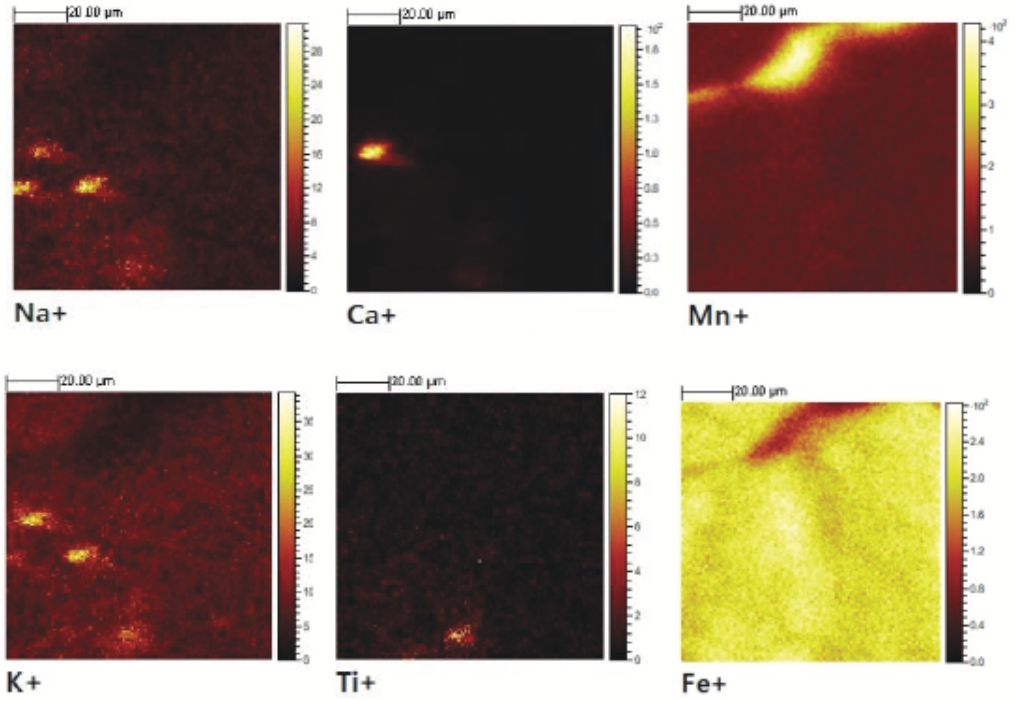


Figure 10. Elemental surface distributions the sample with six layers of Cu 2 nm and Ni 14 nm after excess heat generation analyzed by TOF–SIMS.

differently distributed. This fact might be in contradiction with stainless steel contamination process. Also we notice that the ratio of Cu/Ni got larger near the surface. The reason is unclear at present. Si, major surface impurity, seems to be reasonably distributed if it diffused from the surface.

It is important to continue to investigate further by these analysis methods paying attention to the discrimination of contamination, in order to identify what is happening in the nano-sized multilayer metal composite with hydrogen gas.

4. Concluding Remarks

New type of excess heat experiments using a nano-sized metal multilayer composite and hydrogen gas have been performed based on the permeation-induced transmutation experiments with multilayer thin film and excess heat experiments with nano-particles. Anomalous Excess energy generation using nano-sized multilayer Cu/Ni metal composite and Cu/Ni metal with third material (CaO, Y_2O_3) composite were observed. Maximum released excess energy reached 1.1 MJ and average released energy per absorbed total hydrogen was 16 keV/H or 1.5 GJ/H-mol. It cannot be explained by any known chemical process and suggests that the observed heat generation must be of nuclear origin. Many analysis methods have been applied to identify what kind of reactions occur by the interaction of the nano-sized multilayer metal composite with hydrogen gas.

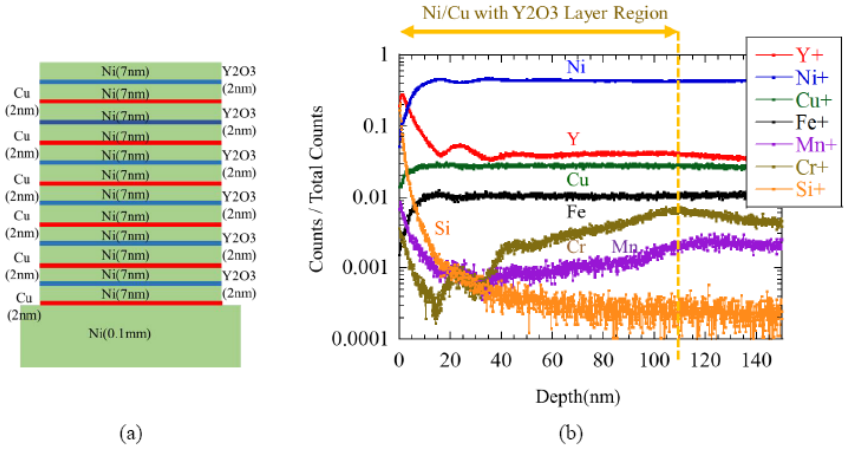


Figure 11. An example of TOF–SIMS analysis; (a) structure of the sample before experiment, (b) depth profiles of the after excess heat generation.

Acknowledgements

The authors acknowledge Mr. H. Yoshino, Mr. S. Hirano, Mr. M. Ise, Dr.T. Hioki and Mr. M. Hattori, who are the members of CLEAN PLANET Inc., for their significant assistance. The authors also thank Mr. M. Takeya, Prof. H. Kikunaga and Mr.Y. Shibasaki of Tohoku University for their support. This work is supported by CLEAN PLANET Inc., Research Center for Electron Photon Science of Tohoku University Electron Photon, Tanaka Kikinzoku Memorial Foundation and The Thermal and Electric Energy Technology Foundation.

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