Optical Observation on Anomalous Heat Generation from Nano-sized Metal Composite

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Abstract

We describe the first measurement of optical spectrum correlated with the anomalous excess heat generation from the NiCu multilayer foil with H_2 gas. The method to induce the anomalous excess heat is the one we have been developing: the anomalous excess heat occurs when a nano-sized metal composite film that has absorbed H_2 is forcibly evacuated and heated up for the gas to discharge. Spectra of the visible light of photon energy between 1.3 and 1.9 eV were measured, in addition to the temperature measurement near the heater at the sample core, which has been performed so far. Two types of samples, Ni plate and NiCu multilayer thin film, were examined under the conditions in which the sample was in vacuum (without H_2) and during desorption of H_2 (with H_2). It turns out that the measured spectrum is understood as that of gray-body radiation. Surface temperature and radiant intensity of the sample are deduced from the spectrum. For the NiCu multilayer sample which shows anomalous excess heat that is evaluated only from Tc, corresponding excessive radiant emission is clearly shown. Although the deduced quantities from the optical radiation qualitatively follow the evaluated values of excess heat, it is indicated that more sophisticated analysis that takes into account the radiation information is highly required for more reliable evaluation of the excess heat. Furthermore, we discuss that the optical measurement plays an important role in the analysis of the heat burst event observed during long-term observation. Our future experimental plans are also discussed.

1. Introduction

Recently, anomalous heat generation, producing much larger energy than ordinary chemical reactions produce, has been reported in a system consisting of nanostructured metals (Ni, Cu, etc.) and hydrogen (or deuterium) gas [1]-[3]. Such a system that produces a large amount of energy without CO_2 emissions is very promising as a future energy source; the realization of clean, powerful and inexpensive energy will have a great social and economic impact.

We have been working on development of a new experimental method that emphasizes the role of hydrogen diffusion in nano-sized metal multilayer composite [4]-[6]. The method currently under development is to induce a reaction of H_2 (or D_2) gas with the thin film layer structure of Ni and Cu metal [4]-[6] instead of the composite amorphous metal powder developed in the joint project of NEDO. We have already published some results using multi-layer foil with good reproducibility suggesting anomalous excess heat generation [3]-[6], it is still necessary to develop experimental equipment for new measurement which may provide more accurate and quantitative discussions. In the previous measurements, the quantity of generated heat was evaluated only by the temperature of the sample core although together with the radiation temperature of the surface was also measured preliminarily.

Accurate excess heat measurement is highly required to discuss the phenomena more quantitatively, to clarify quantities that correlate with the energy generated, and, then, to explore the mechanism. In this work, we describe the first experiment performed with an optical spectroscopic measurement as the next step towards a more complete measurement.

2. Importance of optical measurement

Fig.1 shows a basic structure of a sample installed in a vacuum chamber; two sheets of Ni plate coated with NiCu multilayer are placed on both sides of a ceramic heater (25mm square ; MS-1000R; Sakaguchi E. H Voc Corp.) which has an R-type thermocouple (Tc) in the center. In the previous reports, values of the excess heat were evaluated only by temperature measured Tc. When the sample is heated up (typically between 700 and 900 ℃), the heat generated by the heater (and the excess one) becomes a heat flow and is transferred to the low temperature region by the three processes as shown in Fig. 2; (1) conduction heat transfer through supporting rods (connected with the chamber wall) and wires of the heater and thermocouple, (2) radiation heat transfer from surfaces of heated element to the wall, and (3) convection heat transfer by gas. Since the pressure in the chamber is 1×10^{-4} Pa or less during measurements, the process (3) can be ignored. In processes (1) and (2), the latter contributes most of the heat flow, because a large heat-insulating substance (photveel) is used to support the sample. Thus, the measurement of radiant heat plays an essential role to improve the accuracy of excess heat measurement by measuring radiation.

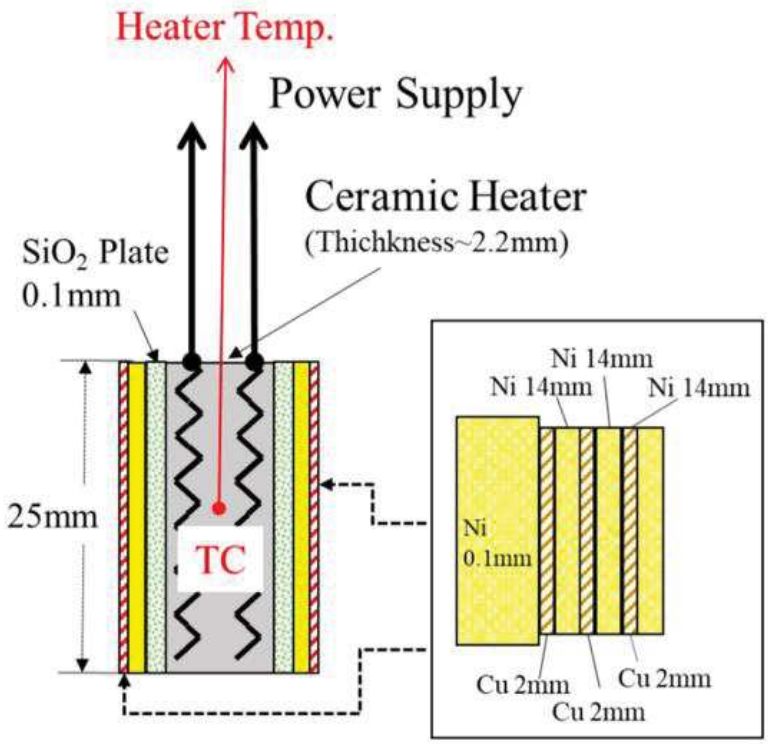

Fig.1 Schematic of components around Nano-sized Multilayer Metal Composite.

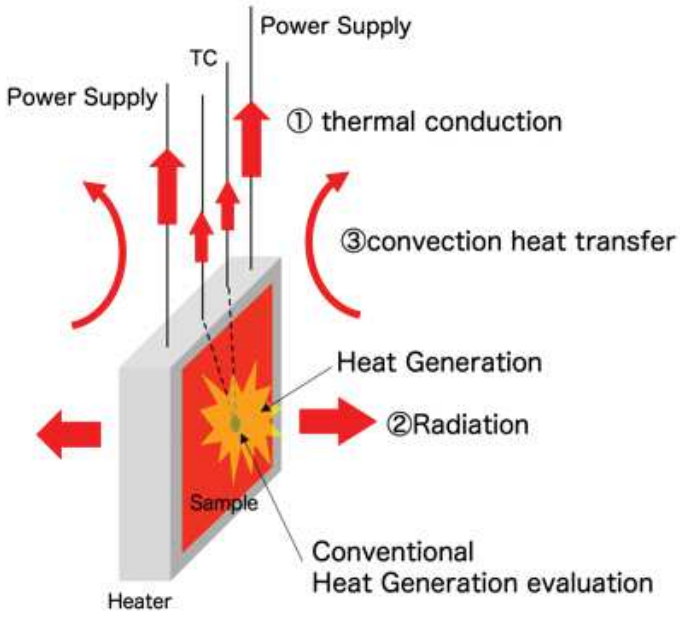

Fig.2 Heat transfer path when a heat generation phenomenon occurs.

Spectroscopic measurements contribute to search for any transitions of special electronic states that accompany the generation of excess heat [7]. In addition, it provides important information on temperature and emissivity, even if it is a continuous thermal spectrum. As an example of the latter, hotspot (local high-temperature region) observation is one of the challenges we are pursuing as discussed at JCF20. Consider the case where a reaction that produces high energy, such as a nuclear reaction, occurs in the sample as shown in Fig. 3(a). If the energy is deposited to several to many electrons immediately after the reaction occurs, those electrons may move in a random direction in the sample and lose their kinetic energies as shown in Fig. 3(b). This region may locally form a hot thermal equilibrium, temperature much higher than that the region heated up only by the heater. One can expect a radiation spectrum to have two temperature components (Fig.3(c)).

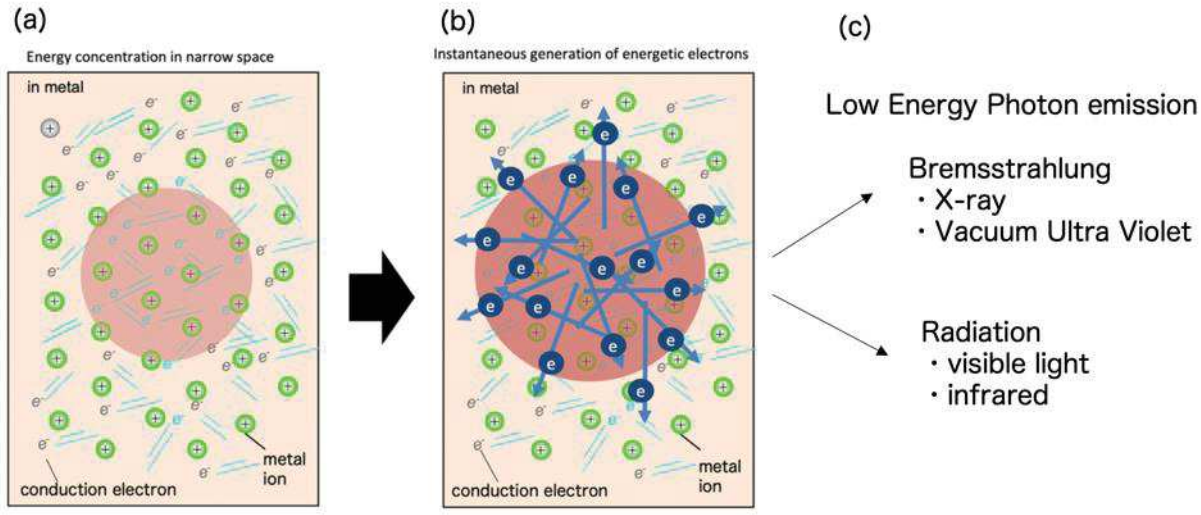

Fig.3 Schematic of energy distribution to electrons when energy is generated locally. (a)Energy concentration in narrow space, such as a nuclear reaction, (b)Instantaneous generation of energetic electrons, (c)Low Energy Photon Emission due to energetic electrons

3. Experimental procedure

The experimental apparatus is shown in Fig 4. A vacuum chamber is made of stainless steel and the volume inside the gate valve is 4.6 litters. It is evacuated with a turbo molecular pump down to 3×10^{-6} Pa. The sample with the structure shown in Fig. 1 is set in the chamber. H_2 gas was introduced from the lower port (gas inlet) and its pressure were monitored by a Pirani gauge. Heater input power is supplied by a DC power supplier with constant voltage mode. The voltage and current values indicated by the device are monitored and those of independently measured are recorded during the measurement.

The sample is placed at an angle of 45 degrees with respect to the direction of the center of the quartz window as shown in Fig. 4; this geometry enables us to put an X-ray detector behind the Be window. The light emitted from the sample goes out through the quartz window, is focused on the optical fiber by a focus lens, and is guided to the spectroscope (C10027-01; Hamamatsu Photonics K.K.) that has a measurement range of 200-950 nm and a wavelength resolution of 2 nm. In order to make the signal-to-noise ratio improve, we decided to obtain a spectrum that averaged 50 measurements with an exposure time of 1 second.

In the present work, we measured two types of samples: (1) nanostructured Ni-Cu multilayer film deposited on a Ni plate and (2) Ni plate (each side of 25 mm and a thickness of 0.1 mm) which is used as a substrate. For the Ni-Cu multilayer film, Cu and Ni are alternately deposited 6 times each on the substrate by the sputtering method. By this way, the sample with a laminated 6-layer structure of 2nm-Cu and 14nm-Ni is prepared. The Scanning Transmission Electron Microscope (STEM) image of the sample structure is shown in Fig. 5.

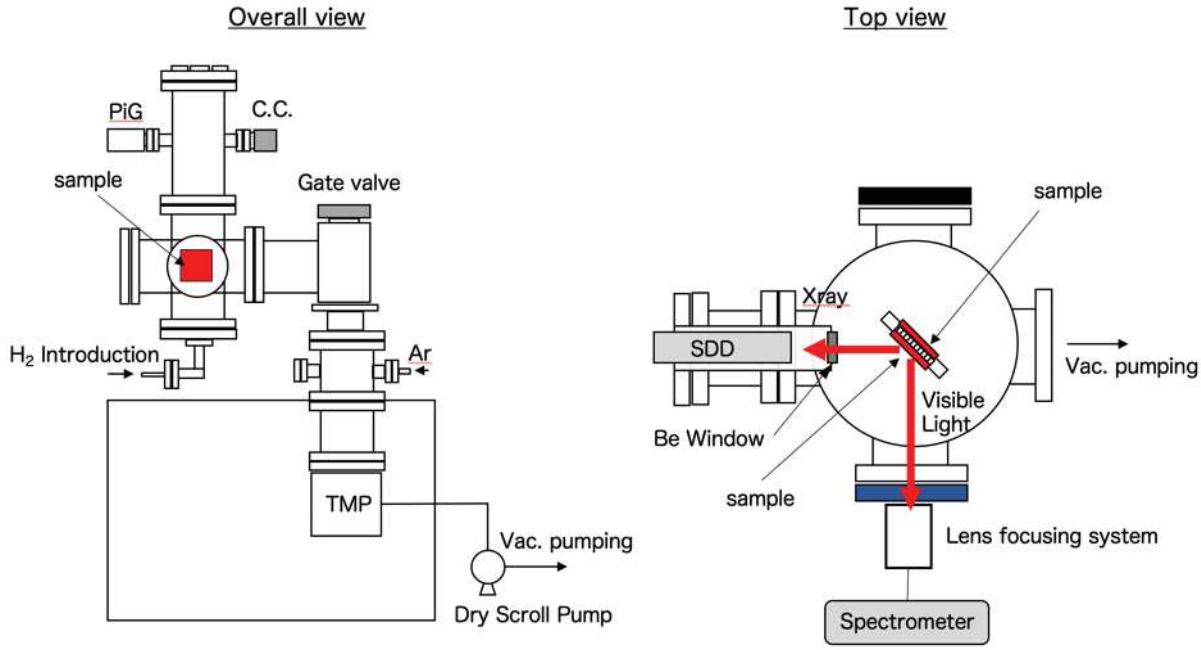

Fig.4 Schematic of Experimental apparatus.

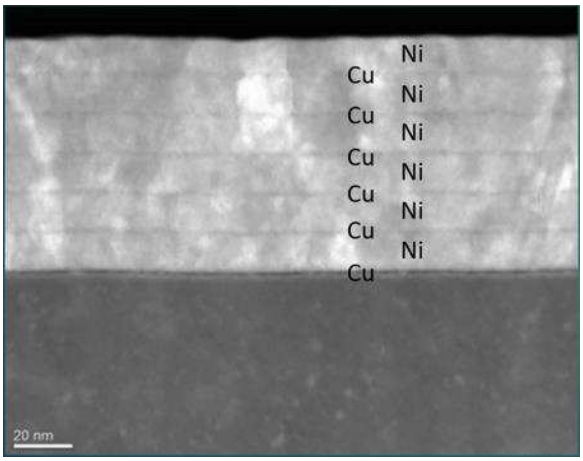

Fig. 5 Scanning Transmission Electron Microscope (STEM) Image
of Cu-Ni Multilayer Thin Film.

The present experiment has been carried out as follows.
(1) Setting sample: Two sheets of sample are fixed on both sides of a ceramic heater and placed at the right position in the vacuum chamber, which is evacuated for a vacuum bake-out. After about 3 days baking out, the heater is turned off.
(2) Measurement without H_2 gas: In a vacuum condition (i.e., without H_2 gas introduction), the voltage of the power supply of the heater is set at various values (between 20 and 48 V) and measured are the temperature inside the heater (Tc) and the optical spectrum. These are regarded as the reference data corresponding to no excess heat.
(3) Measurement during desorption of H_2 gas: The sample absorbs H_2 gas by filling the chamber with H_2 gas to 200-300 Pa and keeping the temperature at about 250 ℃ for 12 -15 hours. At the same time as setting the heater input voltage to the value of the measurement condition, the evacuation of the vacuum chamber is started. The sample temperature (Tc), chamber wall temperature, heater input voltage and current, and vacuum chamber pressure are read and recorded every second. In addition to this, optical spectral measurements in the visible light region are taken every 5 to 30 minutes and saved as data. Data collection continues for at least 6 hours after the voltage once set, since H_2 desorption is expected to continue sufficiently. For measurement under different conditions by changing the set voltage, return to the beginning of (3) and start from H_2 absorption.

4. Results and Discussion

As discussed in Section 2, the heat, including the one from heater and the other generated by reactions between the sample and the H_2 gas, is dissipated as a heat flow from the sample: a radiant heat transfer and a conduction heat transfer. However, the present experiment cannot be a complete measurement of above quantities because of a narrow and limited wavelength range of optical radiation. Thus, discussing more accurate values of excess heat must wait for future experiments. Here, we show the optical spectrum newly introduced, and discuss its change as the excess heat was produced.

Fig.6 shows simple estimates of excess heat from (a) the NiCu multilayer sample and (b) the Ni substrate sample. Each dot against the elapsed time represents the input power of the heater (black dot) and the excess power (orange dot). The time regions without dots correspond to the time for the sample to absorb H_2 gas. Note that the evaluation of the excess heat is based only on the temperature Tc as reported in our previous reports [3-6], and is not a quantitative evaluation with

the addition of radiant heat flow. As seen in Fig.6, excess heat of about 2 ±1 W is observed for the NiCu multilayer sample, while for the Ni plate, almost no excess heat is detected.

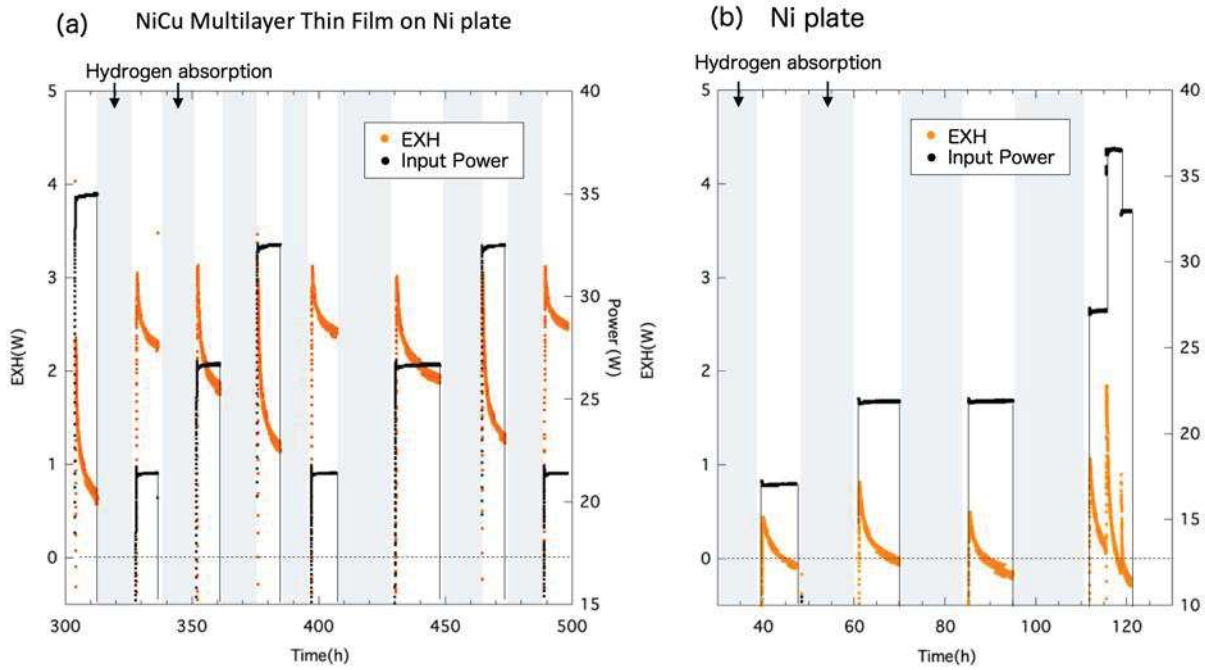

Fig. 6 Excess heat and Input power.
(a) Experiment with NiCu Multilayer Thin Film on Ni plate,
(b) Experiment with Ni plate.

Fig.7 shows intensity spectra of emitted light photon with energy above 1.2 eV from the NiCu multilayer sample: the sample clearly shows the excess heat generation as indicated in Fig.6. The vertical axis corresponds to the radiation energy observed per second (i.e., photon energy times number of photons /sec). In the figure, the spectrum measured during H_2 desorption (red dot) and that without H_2 (blue dot) for heater input power of 21, 27 and 35 W are compared.

The first point to be emphasized is the fact that the radiation intensity is clearly increased by desorption of H_2 as compared without H_2. This is a clear indication of the excess heat: the greater the amount of heat, the greater the brilliance. That is, it is shown that the radiant energy is increased by increasing the temperature of the sample surface due to heat generation. Moreover, the amount of yield increases as the input power increases. This is different from the behavior of excess heat in Fig.6. This indicates that more sophisticated analysis that takes into account the radiation information is highly required in order to obtain accurate value of the excess heat.

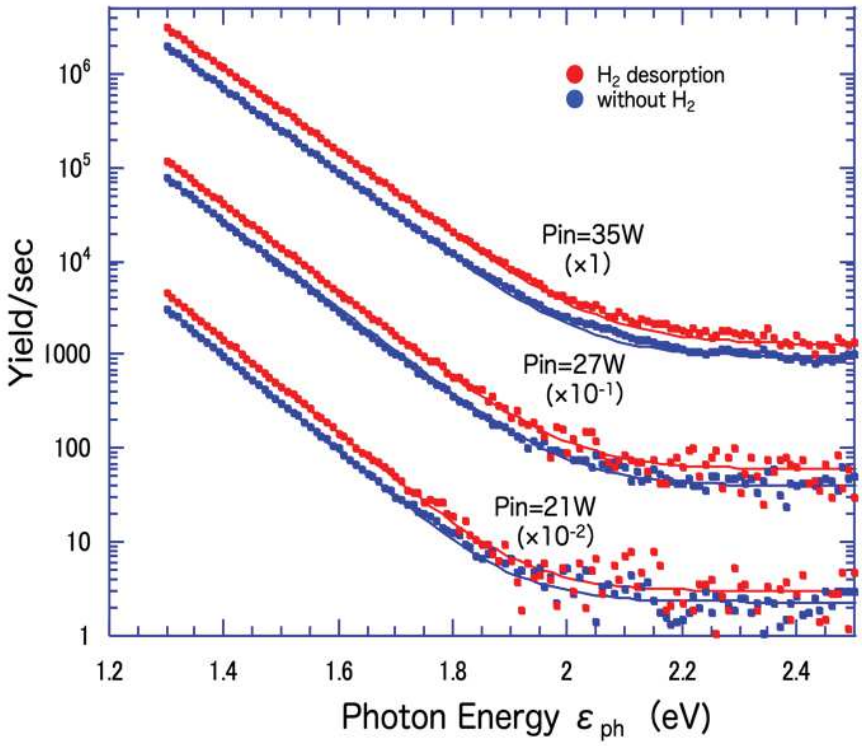

Fig. 7 Photon emittion from NiCu Multilayer Thin Film.

Second, the measured spectrum is well understood as the gray-body radiation, at least in this energy region. Since the photon energy ε we measure is much larger than the radiation temperature (ε >> kTs), Planck's radiation equation can be approximated as follows.

$$Y(\varepsilon) = A\varepsilon^3 \frac{1}{\exp\left(\frac{\varepsilon}{kT}\right)-1} = \sim A\varepsilon^3 \exp(-\varepsilon/k_B T)$$

The solid curves in Fig.7 are the best fitting curves calculated to explain the experimental data with 3 parameters under the assumption that the emissivity of the sample does not depend on energy. We used the following approximate function for the intensity Y(ε):

$$Y(\varepsilon) = A\varepsilon^3 \exp\left(-\frac{\varepsilon}{kT_s}\right) + C \quad , \qquad (1)$$

where A is the normalization factor, ε is photon energy, k is the Boltzmann constant, Ts is the surface temperature of the sample and C represents the background yield due to the dark current of the device: the 3 parameters are A, T_s and C. In this way, the important values A and Ts can be obtained: the total (integral) radiation power from the sample is proportional to AT_s^4, if the emissivity is a constant. The radiant power mentioned below refers to the AT_s^4.

Figure 8 shows a comparison of the emitted radiation from the two samples. Fig.8 (a) and (b) show the radiant power and the surface temperature, respectively, obtained for the NiCu multilayer film, and Fig.8 (c) and (d) show those for the Ni plate sample. Red circles correspond to the foreground experiment with H_2 storage, and open circle to the reference experiment without H_2.

The comparison of (a) and (c) in Fig.8 shows more clearly that the NiCu multilayer sample generates a large amount of excess heat as compared with the Ni plate sample. In the NiCu multilayer sample, the radiant power during H_2 desorption is always higher than without H_2, demonstrating the presence of the excess radiation which increases as the input power increases. In the Ni plate sample, on the other hand, no significant difference is observed; i.e., no excess radiation in the Ni plate. The same behavior can be clearly seen in the comparison of (b) and (d) in

Fig.8. Observed temperatures, both Tc and Ts, are always larger during desorption of H_2 for the NiCu multilayer sample, while no change in temperature for the Ni plate sample.

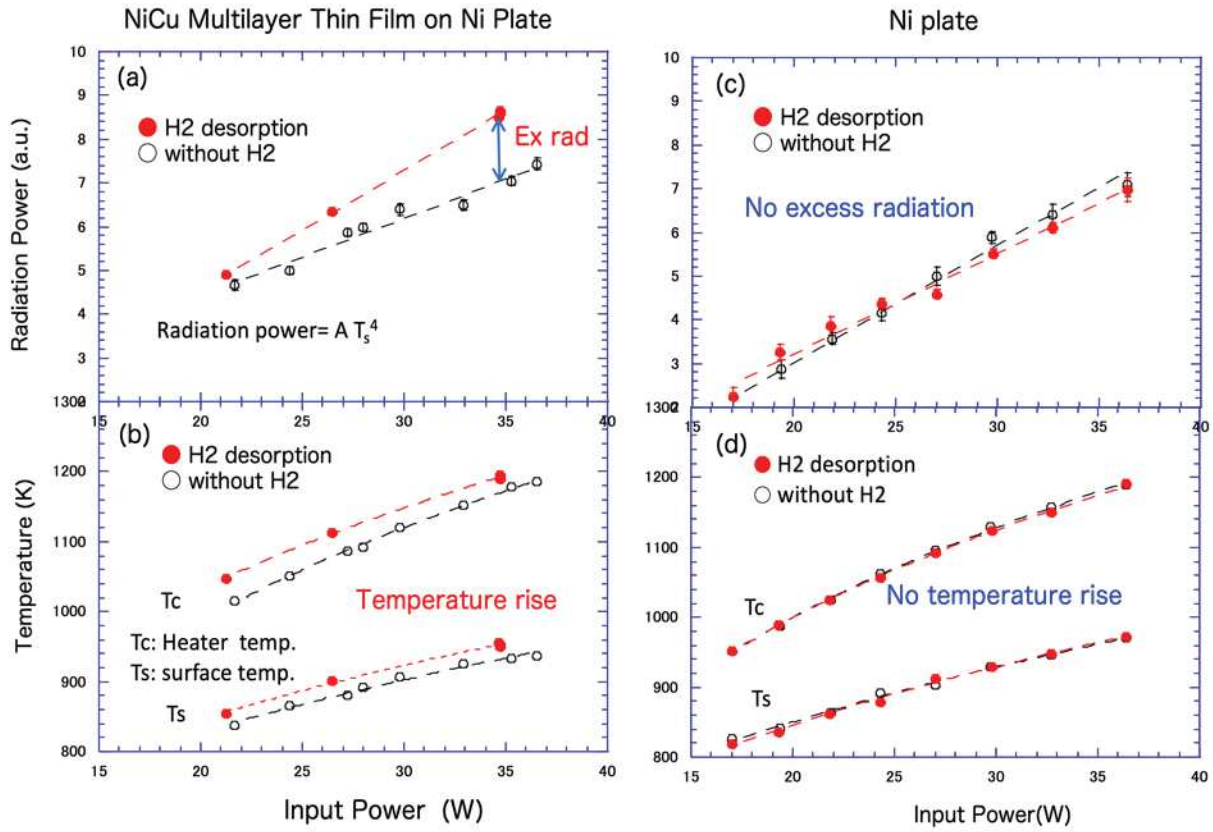

Fig. 8. Comparison of emitted radiations between NiCu Multilayer Thin Film on Ni Plate and Ni Plate. (a) Radiation Power and (b) Temperature of experiment using NiCu Multilayer Thin Film (c) Radiation Power and (d) Temperature of experiment using Ni Plate.

When the H_2 desorption experiment with the NiCu multilayer sample was conducted for several hours continuously, a phenomenon in which Tc rises suddenly did occur as reported in [4,5,8]. We call it heat burst. Fig.9 shows two such events observed simultaneously with measurements of optical spectra, for the first time. As seen in Fig.9 (a) where heater temperature (Tc) is plotted against elapsed time, the heat burst events are recorded at 8.5 and 22 hours after the start of the experiment. Fig. 9 (b) shows corresponding excess heat evaluated based solely on Tc. The surface temperature (Ts) and integrated yield between 1.2 and 1.5 eV (proportional to radiant intensity) deduced from light spectrum are shown in Fig. 9 (c). The bump structures corresponding to those of Tc are clearly confirmed in both Ts and the radiant intensity. However, the data were not obitained immediately before and after the bump, since the number of sampling of the optical measurement is small for the elapsed time after 15.5 h. Fig.9 (d) and 9 (e) show the spectra before and after the first burst and the second burst, respectively.

One may interpret the time sequence of the bump events observed in Fig.9 as follows: A heat burst occurs locally on the surface of the NiCu multilayer sample, part of the power is dissipated immediately from the surface as the radiant power, and at the same time, the generated heat is transferred to the core of the sample, then, temperature Tc raises. It is considered that investigating the heat burst in more detail, for example, to obtain precise time correlation between

the busts in Tc and in the radiant power, might lead to the elucidation of the mechanism of the excess heat generation.

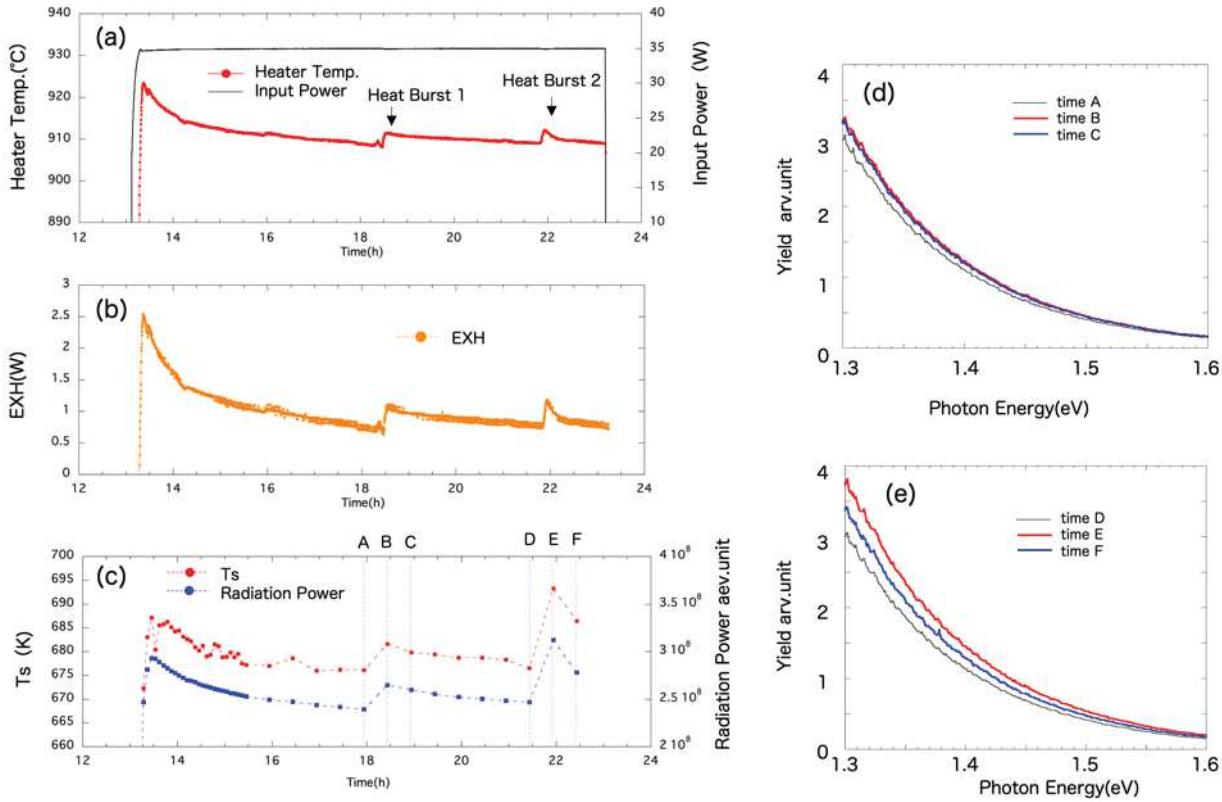

Fig. 9 Simultaneously detection of Heat Burst by Radiation of spectrometer Looking at the Surface of the Multilayer Thin Film and a Thermocouple Located in the center of Heater. (a)Heater Temperature and Input Power, (b)Excess Heat, (c) Surface temperature and Radiation Power by Spectrometer, (d)Radiation Spectrum of 1st Heat Burst, (e) Radiation Spectrum of 2nd Heat Bust.

5. Future Plan

In the present experiment, we were able to observe the anomalous heat generation phenomenon from the observation of radiation, although the observed region is very narrow as 1.3-1.9 eV. Also, we discussed, in Section 2, the importance of the radiation measurement to explore the mechanism of the anomalous excess heat. Thus, it is natural to extend the region of the wavelength to be measured. This is our future plan as a next step.

In Fig.10, calculated spectra of the black-body radiation are shown for temperatures from 800 to 1100 K, expected temperatures in our experiments. Shown in the lower part of Fig.10 are the detectable energy regions of the detectors under consideration. Since region 1 is already introduced in this work, we will extend our measurements to low energy regions using a near-infrared spectrometer (1.1-2.5 μm) and a mid-infrared detector (3-5.6 μm). It is expected, then, to grasp the whole picture of the radiation: This is equivalent to a sophisticated calorimeter measurement, and the reliability of the excess heat is dramatically increased.

In addition, in order to elucidate the mechanism related to the existence of hot spots, it is necessary to measure the visible light region on the high energy side more carefully. We are also considering ways to reduce the effects of radiation and improve S/N for measurement in higher energy side of region ①

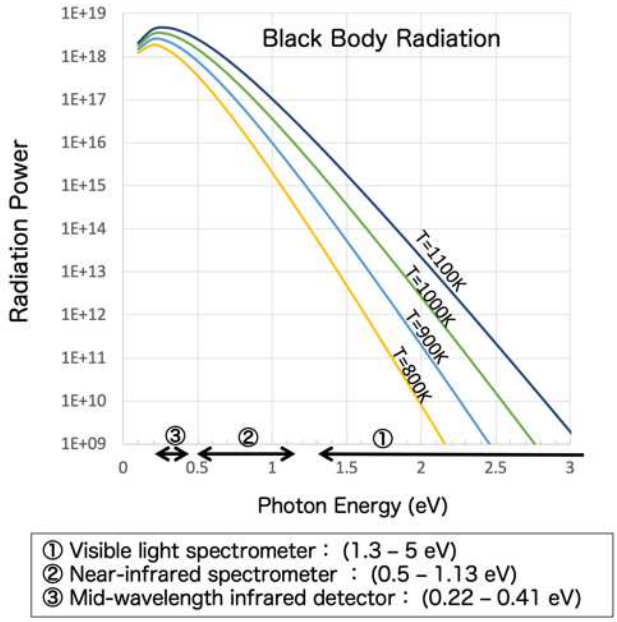

Fig.10 Extended spectra; from Visible light to infrared.

6. Summary

In the present work, we aimed to improve the accuracy of measurement of the anomalous excess heat generation observed in Ni-hydrogen system by adding optical measurement to the conventional measurement system. This is the first measurement of optical spectrum correlated with the anomalous excess heat generation from the NiCu multilayer sample with H_2 gas.

In the experiment with the Ni substrate sample, the temperature and radiation intensity deduced from the light spectrum hardly changed before and after hydrogen absorption, but with the NiCu multilayer sample, they showed clearly excessive radiations.

The heat burst phenomenon was observed multiple times in the measurement with the NiCu multilayer sample. When heat burst phenomena occurs, sudden increase in the radiation intensity together with the surface temperature was observed at the same time as the heater temperature increases.

In conclusion, we have constructed the measurement system for the optical spectroscopy by adding the previously used one. The optical measurement is found out to be an important one for the study of anomalous excess heat generation. Although the deduced quantities from the optical radiation qualitatively follow the evaluated values of excess heat, it is indicated that more sophisticated analysis that takes into account the radiation information is highly required for more reliable evaluation of the excess heat.

Acknowledgements

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