J. Condensed Matter Nucl. Sci. 36 (2022) 274–284

Research Article

Optical Observation of Spontaneous Heat Burst Phenomena during Hydrogen Desorption from Nano-Sized Metal Composite

Takehiko Itoh*

Research Center for Electron Photon Science, Tohoku University, 982-0826 Japan
CLEAN PLANET Inc., 105-0022 Japan

Yoshinobu Shibasaki, Tomonori Takahashi, and Mari Saito

CLEAN PLANET Inc., 105-0022 Japan

Jirohta Kasagi and Yasuhiro Iwamura

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

Abstract

We have constructed a photon measurement system to observe the radiant power emitted from an NiCu multilayer film for a wide energy range, from visible light to mid-infrared light. Using the new system, we attempted to measure the radiation from the sample when a heat burst occurred while anomalous excess heat was being continuously generated. About 10 big burst events were observed during the 5-day continuous measurement: it was found that the radiation intensity increased simultaneously as the heater temperature suddenly increased. We have made a comparison of the radiant spectrum of the burst event with the spectrum in normal times. There is no particular difference due to the burst; both spectra can be simply interpreted as grey body radiation emitted from a thermally equilibrated object. The correlation of radiant intensity between near-IR and visible light has been deduced; although it is expected there is a linear correlation between them, two correlation lines can be drawn instead of one. This indicates that the contribution of photon emission from the non-equilibrium stage may have an effect.

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

1. Introduction

In recent years, in a system consisting of nanostructured metals (Ni, Cu, etc.) and hydrogen (deuterium or hydrogen), anomalous heat generation that produces huge thermal energy (at least one order of magnitude larger than ordinary

*Corresponding author: itoh@lns.tohoku.ac.jp

chemical reactions) without CO_2 emission has been reported [1]–[6]. If power generation technology utilizing this phenomenon can be put into practical use, clean, powerful and inexpensive energy will be obtained, and the social and economic impact will be immeasurable.

We have been studying on this subject since 2015. The following has become clear from our measurements, although systematic basic data are still required.

(1) This heat generation phenomenon largely depends on the substance and structure of the nanostructured metal material [3]–[6].

(2) Surface analysis of nanostructured metal materials suggests that the reaction occurs locally on the surface of the material [3], [7], [8].

In addition to the material issues, we often observe sudden heat generation in the thin film samples during the measurements. We call this a 'heat burst.' The heat burst consists of a simultaneous sudden rise of the temperature of both the heater and the film surface measured by the radiation thermometer. We have interpreted it as follows: there is sudden energy generation in the sample, part of which is dissipated quickly by radiation from the surface, and part of which is propagated to the inside, raising the heater temperature [6]–[8]. In the subsequent measurement of visible light, we observe a sudden increase in light intensity associated with the heat burst [9].

In the experiments performed so far, only the temperature and a small fraction of the heat radiation were measured (with a radiation thermometer), despite the rather high temperature of the sample. The entire radiation spectrum was not observed. We have now improved the experimental equipment to allow for a wide range of radiation spectrum measurements in addition to temperature measurements, as reported in another presentation at ICCF23 [Kasagi et al.]. The measurement has been extended to the visible light, near infrared, and mid-infrared regions so that the whole picture of the radiation can be drawn almost perfectly. Observation of the radiant spectrum as a whole is indispensable for explaining the heat burst phenomenon. For example, when a reaction that locally generates high energy (such as a nuclear reaction) occurs suddenly and the generated energy is transferred to several electrons, the electrons may move in a random direction in the sample and lose their kinetic energy. The small region may locally form a hot thermal equilibrium, the temperature much higher than that heated up only by the heater and, then, one may expect a radiation spectrum with two temperature components [10].

The aim of the present work is to (1) try to observe the heat burst phenomenon with a photon detection system newly developed for wide-region measurement of the radiation, and (2) to explore the mechanism of the burst generation by measuring the time correlation between temperature and radiation, as well as examining the radiant spectrum in detail.

2. Experimental Equipment and Procedure

The experiment was performed by improving the reaction chamber used in the optical measurement reported in [9]. The improvements allowed multiple radiation detectors to be attached. The experimental procedure and the sample tested in the present work are basically the same as previously reported in [6]–[9]. We describe only procedures that are important in the present experiment.

2.1. Sample - Heater Structure

The metal thin film sample, which is the key to the success or failure of AEH (anomalous excess heat) generation, is installed in a vacuum chamber into which hydrogen gas is introduced. Two sheets of nanostructure samples are set on both sides of the ceramic heater (SAKAGUCHI E.H VOC Corp.), as schematically shown in Fig. 1. The ceramic heater is made of alumina, the size is 25 mm $\times$ 25 mm $\times$ 0.25 mm, and an R-type thermocouple is placed in the center. As described in [3], [4], [6], [8], the nanostructure metal is formed by alternately depositing Cu and Ni on a Ni plate. The metal is covered and fixed by a holder with a 20 mmϕ hole in the middle and facing the film-forming surface

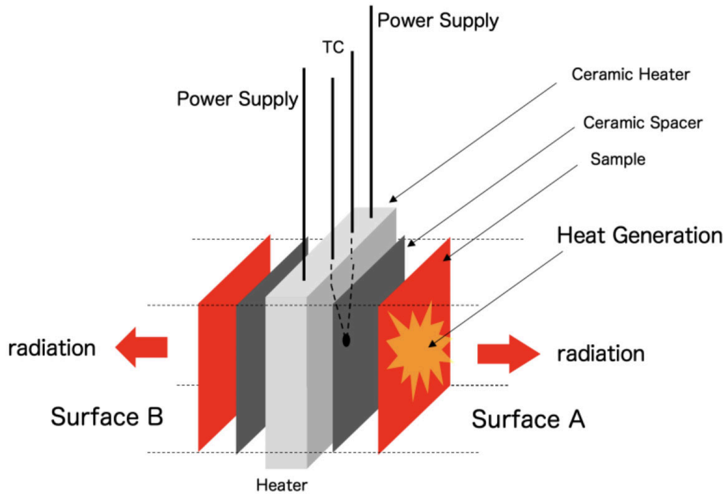

Figure 1. Schematic of components around nano-sized multilayer metal composite.

outward. The holder (not shown) is made of ceramic (Photoveel). A 1-mm-thick ceramic (Photoveel II k70) plate is sandwiched between the heater and the sample. In addition to the heater thermocouple for temperature measurement, we set K-type thermocouples on the ceramic holder and the chamber wall.

The heater input was supplied from a regulated DC power supply (PSW-360L80: TXIO TECHNOLOGY Corp.). The current and voltage values are measured with a digital multimeter (GDM-8342: TXIO TECHNOLOGY Corp.) in parallel with capturing the monitor value of the power supply into the data logger, and the input power is cross-checked. The temperature values measured by the thermometers were recorded with a data logger (GL820: Graphtec Corp.) every second.

2.2. Sample Preparation

The sample is a NiCu multilayer film which is formed on a Ni substrate plate (size: 25 mm $\times$ 25 mm $\times$ 0.1 mm, purity: 99% up, Furuuchi Chemical Co.), as follows.

(1) The Ni plate was washed with acetone, alcohol, and ultrapure water in order to remove the oil component.

(2) After that, in order to remove the processing history, annealing was performed at 900°C for 72 hours under a high vacuum ($<10^{-4}$ Pa) condition.

(3) After that, chemical etching was performed using nitric acid to remove impurities on the surface.

(3) Using a magnetron sputtering device, a nano-multilayer film in which 6 layers of Cu: 2 nm and Ni: 7 nm are alternately formed.

Figure 2 shows a STEM image of the cross section of the multilayer film.

2.3. Optical Measurement Apparatus

Figure 3 shows a top view of the arrangement of photon detection devices employed in the present work. Two sheets of samples put on both sides of the heater are placed in the center of the vacuum chamber. Photons emitted from the surfaces of the samples are detected outside the vacuum windows. A photodetector TMHK-CLE1350 (wavelength

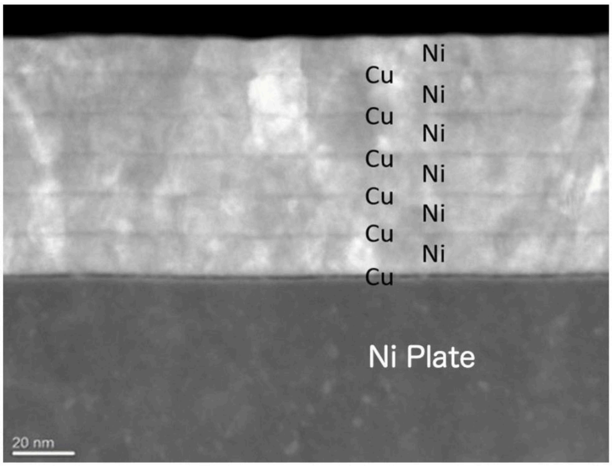

Figure 2. STEM (Scanning transmission electron microscope) image of Cu-Ni multilayer thin film.

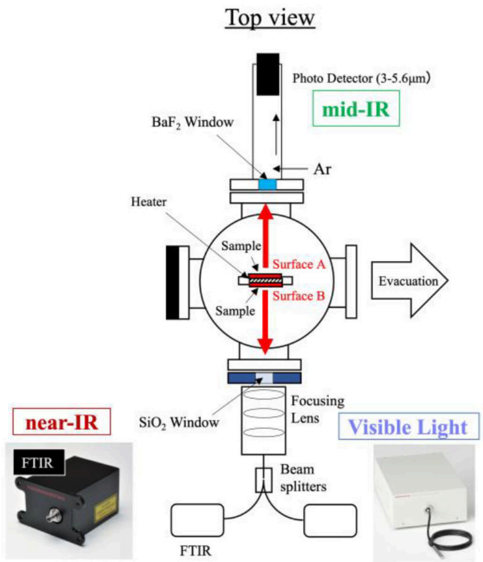

Figure 3. Schematic of experimental apparatus. Top view of apparatus and optical measurement device.

3-5.5 μm photon energy: 0.23-0.41 eV) for mid-infrared measurement is placed on one side of the chamber via a BaF2 window. On the other side through a quartz window two spectrometers are placed, one for near-infrared and the other for visible light; the light emitted from the sample is focused on a fiber by a focus lens and is split by a splitter. Employed are FTIR (Fourier Transform Infrared Spectrometer C15511-01: Hamamatsu Photonics K.K.) to measure 1.5-2.5 μm light (photon energy: 0.5-0.83 eV) and spectroscope (PMA-012 C10027: Hamamatsu Photonics

K.K.) to measure 0.3-0.9 μm light (photon energy: 1.4-4.1 eV). Each detector faces two samples by rotating the sample 180 degrees. In the present work, we report the results of long-term observation with the sample fixed without rotation. In the following description, the surface which faces the mid-infrared detector (MIR) will be referred to as Surface A, and the other surface facing to the near-infrared (FTIR) and visible light (Light) spectroscope will be referred to as Surface B.

2.4. Experimental Procedure

The NiCu multilayer film was used for the long-term observation. Before the measurement run, the sample was baked, and a measurement without hydrogen was performed as follows:

(1) Mount the two samples in the sample holder and place it in the chamber, which is evacuated to $1\text{x}10^{-7}$Pa or less.

(2) Turn on the heater and keep the heater temperature at 900°C. After about 3 days of baking, turn off the heater.

(3) Reference experiment without hydrogen: Power is applied to the heater without hydrogen absorption, and the heater temperature and radiation are measured.

The observation run with hydrogen was started by letting the sample absorb hydrogen gas. We set the sample temperature to 270-300°C, maintain it under the pressure of hydrogen gas 200 Pa for 15 hours. After that, the evacuation of the vacuum chamber is started at the same time as setting the heater input voltage to the value of the measurement condition.

The heater temperature Tc, radiation thermometer MIR, chamber wall temperature Tw, holder temperature Tpr, heater input voltage and current, and vacuum chamber pressure are constantly read every second and recorded by the logger. In addition to this, spectral measurements in the infrared and visible light region are recorded every 30 minutes. Data collection continues for at least a few days at the voltage once set, since H_2 desorption is expected to continue measurably.

3. Results & Discussion

Figure 4 shows an example of heat bursts observed through temperature Tc; the CuNi sample was heated to about 900°C, and the heater power is kept constant at about 33 W for 130 hours. In Fig. 4(a), the red line indicates Tc and the black line is the heater input (Pin). As shown, heat bursts with a sharp rise in Tc were frequently without any change in Pin. Figure 4(b) shows the excess power evaluated by a simple method that has been used so far [3], [4]. Since the evaluation is based only on the Tc values, errors of about 30% are expected, but it can be said that bumping on the order of 1 W occurs momentarily at intervals of 10 to 20 hours.

Results of photon observation are shown in Fig. 5. Figure 5 (a) shows the temperature of the heater (red line) and the radiation intensity measured by MIR (green line), for the A side of the sample. Vertical dotted lines with the alphabet above the graph indicate when large bursts occur (heater temperature rises by about 10°C). As expected, the mid-infrared radiation also increases in response at each time.

Figure 5 (b) shows the radiation intensity detected by FTIR and Light spectrometer for the B surface of the sample. The brown square in Fig. 5 (b) shows the near-infrared intensity (integrated intensity from 0.5 to 0.75 eV in the measured 0.3-0.9 eV spectrum) and the blue circle indicates the visible light intensity (integrated intensity from 1.5 to 1.8 eV in the measured 1.5-3 eV spectrum). It can be concluded that heat bursts are observed as a wide range of simultaneous radiation increases, from mid-infrared rays, which mainly contribute to heat radiation, to visible light, which is a high-energy component of heat radiation.

Figure 6 is an enlarged view of Fig. 5 between 60 to 80 h. Figure 6 (a) is for Tc (red line) and MIR (green line) and Fig. 6 (b) for FTIR (brown square) and Light (blue circle). It is shown that not only large bursts of Tc but also small

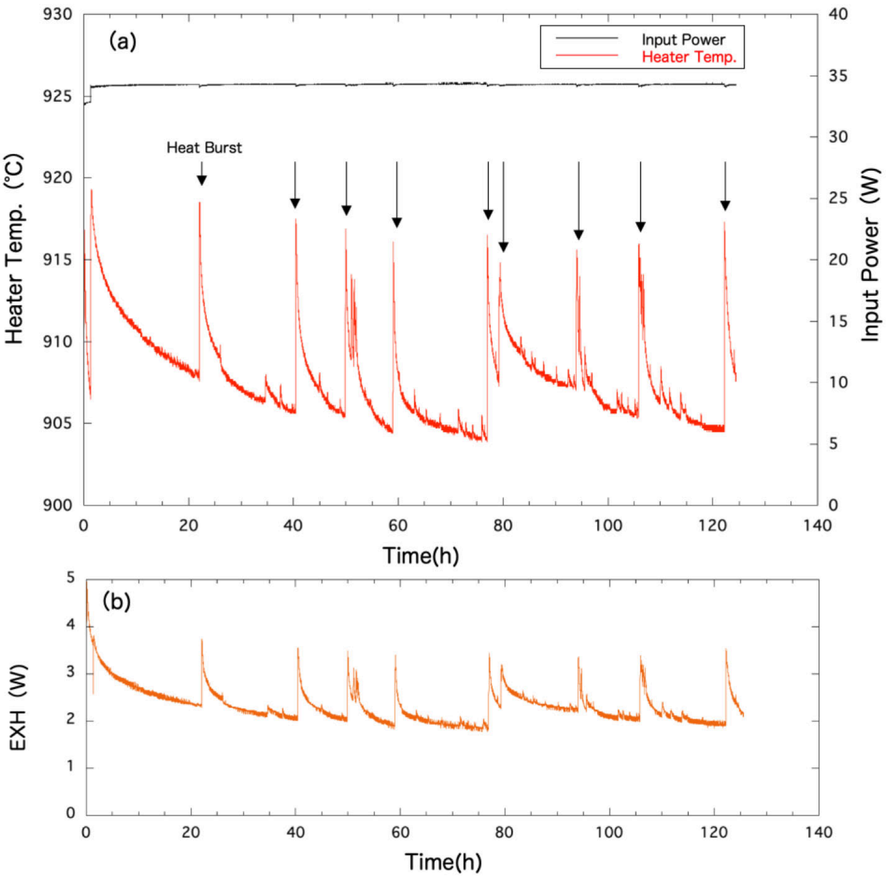

Figure 4. Observation of heat bursts: (a) time evolution of heater temperature and input power. (b) Excess heat evaluated from heater temperature.

ones cause an increase in radiant intensity of high-energy photons. The largest burst in this region is the one at 77.6 h. This burst is used to compare the spectrum of burst events with the spectrum in normal times. Such a comparison is made in Fig. 7. Figure 7 (a) shows the FTIR spectrum and Light at time 77.6 h and 77.1 h. The blue dots correspond to data at time 77.1 h, and the red dot 77.6 h. Figure 7 (b) and Fig. 7 (c) are enlarged views of the low energy side (0.55-0.75 eV) and the high energy side (1.6-1.8 eV), respectively. Both spectra can be simply interpreted as grey body radiation emitted from a thermally equilibrated object. We can deduce values of temperature to be 923.1 K for the burst and 915.5 K for the normal time.

Figure 8 shows the correlation between the near-infrared radiant intensity and the visible light intensity. Light intensity is integrated yield between 1.6 eV and 1.8 eV and near IR intensity between 0.5 eV and 0.75 eV. In the case of gray-body radiation, if the emissivity of the surface does not change, the radiant intensities in near-infrared and in visible light are linearly connected in a narrow temperature range as in this measurement. The correlation of radiant intensity shown in Fig. 8 roughly indicates that, but it is better to say that two correlation lines can be drawn instead of one, as shown by the red dotted lines in Fig. 8. This indicates that the contribution of photon emission from the non-equilibrium stage may have an effect.

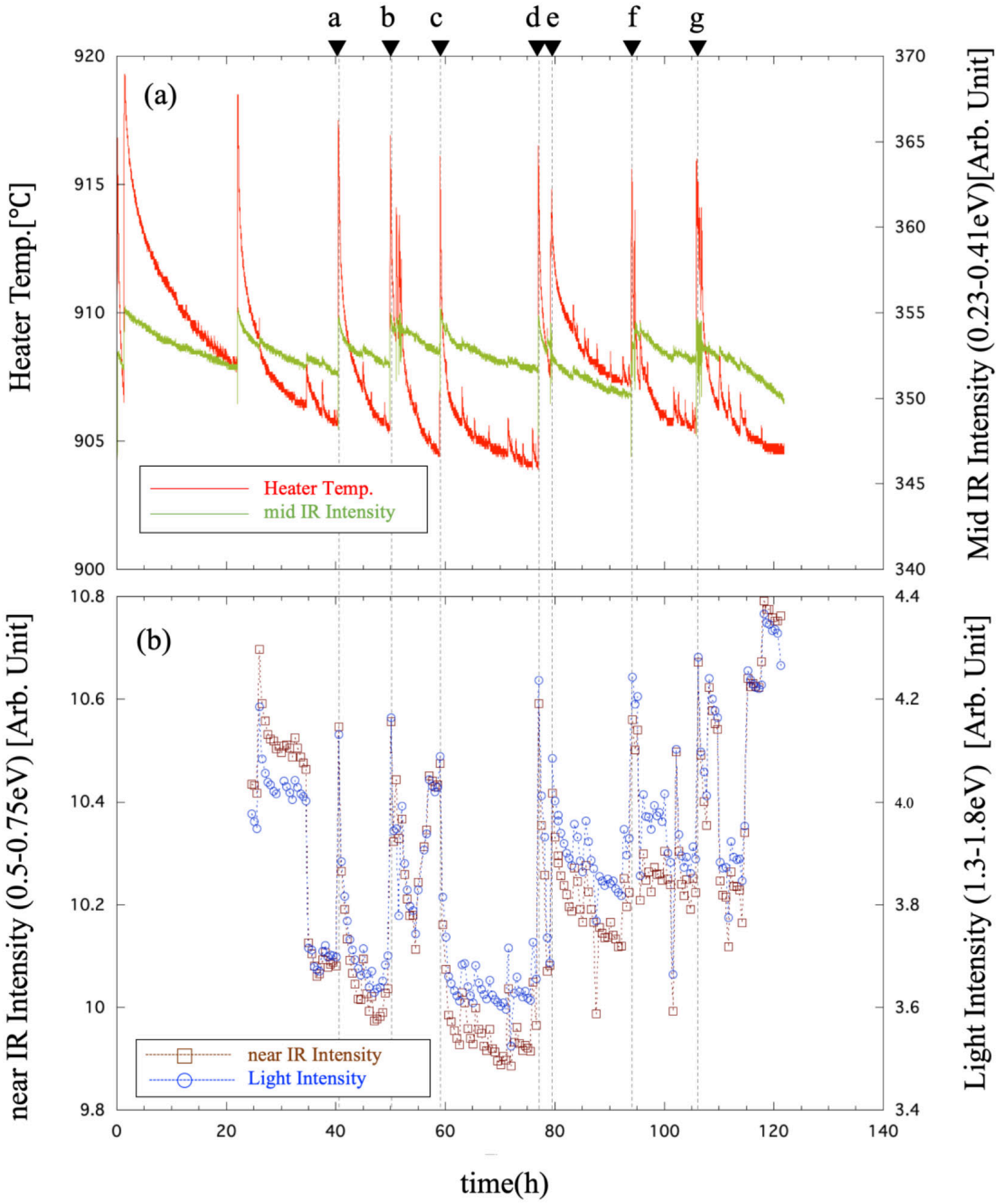

Figure 5. Simultaneously detection of heat burst by radiation of spectrometer and a thermocouple located in the center of heater. (a) Heater temperature and Mid-IR intensity, (b) Near IR intensity (integrated yield between 0.25-0.75 eV) and light intensity (integrated yield between 1.3-1.8 eV).

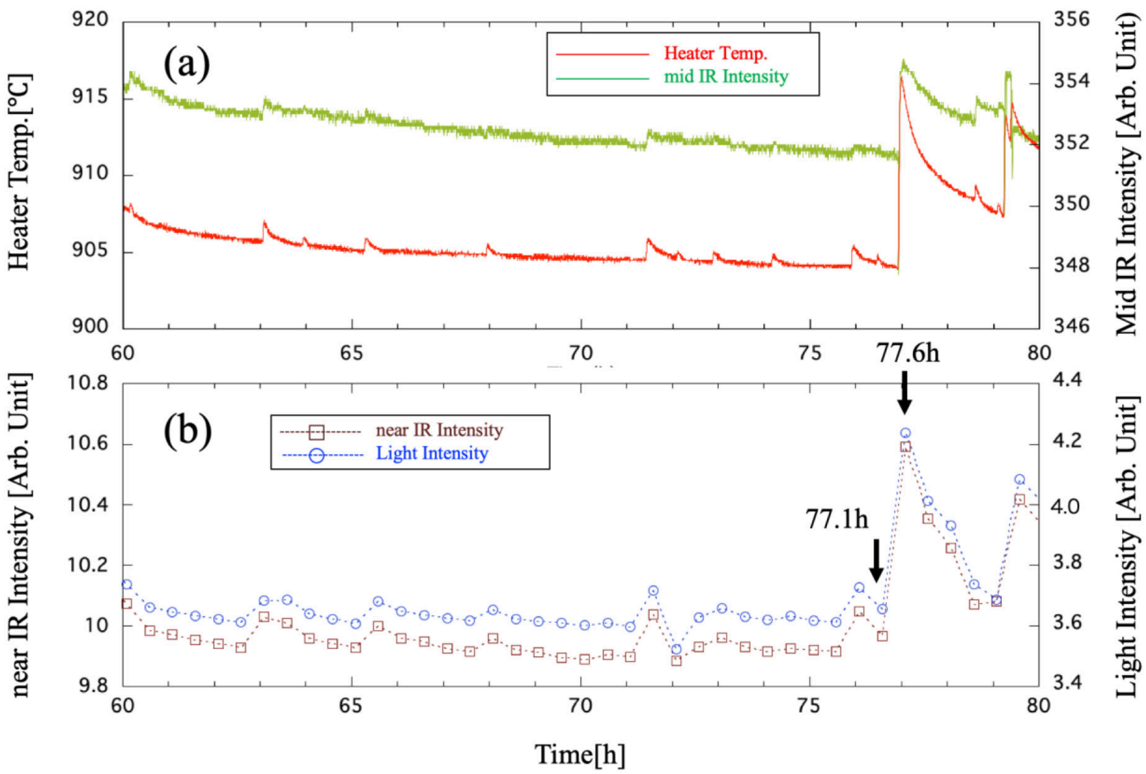

Figure 6. Simultaneous measurement of heat bursts from time 60 h to 80 h (a) Heater temperature and Mid-IR intensity, (b) Near IR intensity and light intensity.

The present observation shows that when a heat burst occurs on the sample surface, the radiation in the entire region from the mid-infrared to visible light increases immediately. It is considered that when the heat burst occurs locally on the surface of the 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 to raise the heater temperature. The existence of pre-equilibrium photon emissions is one of the issues to be pursued in the future. Thus, it is highly desirable to investigate the heat burst phenomena in more detail, for example, to obtain precise time correlation between the bursts in heater temperature and in the radiant power, to analyze the shape of the spectrum, and so on. A detailed analysis is in progress.

4. Summary

We have constructed a photon measurement system to observe the radiant power emitted from NiCu multilayer film for a wide energy range, from visible light to mid-infrared light.

In the present work, we attempted to measure the radiation from the sample when a heat burst occurred while AEH is being continuously generated. About 10 big burst events were observed during the 5-day continuous measurement. It was found that the radiation intensity increased simultaneously for all detectors as the heater temperature suddenly increased. We have made a comparison of the radiant spectrum of the burst event with the one in normal times. There is

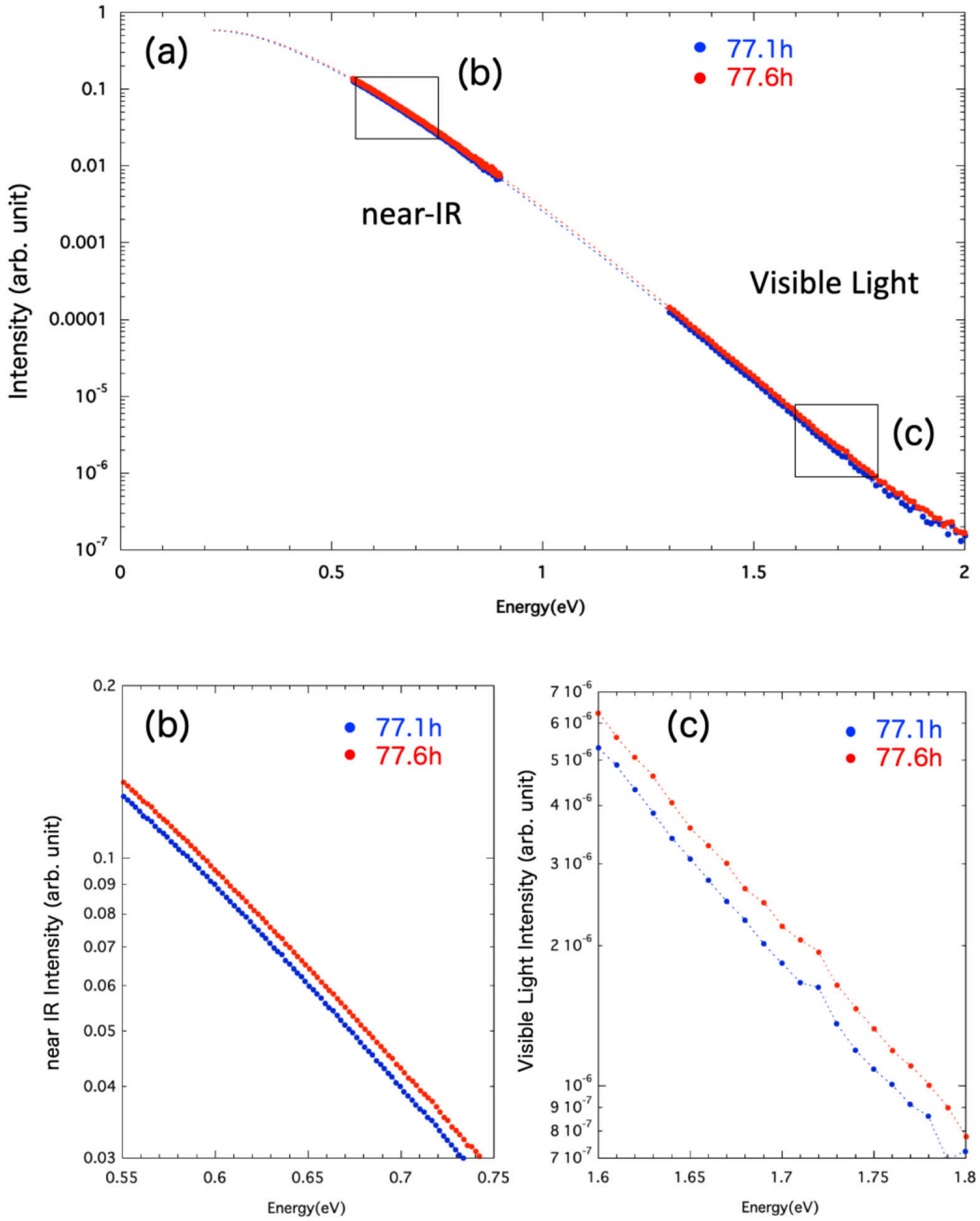

Figure 7. Visible light and near-infrared spectra at time 77.1 h (immediately before heat bursts) and 77.6 h (when heat bursts occur).

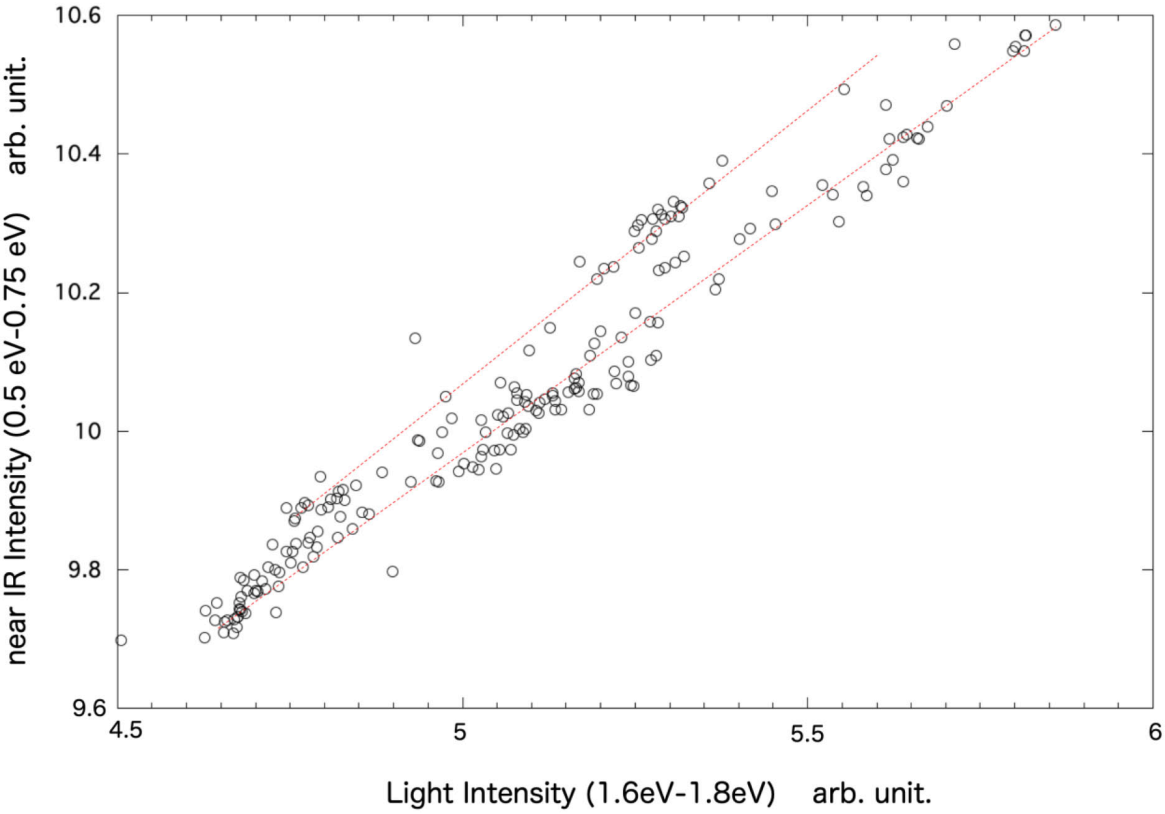

Figure 8. Correlation between light intensity and near-infrared intensity.

no particular difference due to the burst; both spectra can be simply interpreted as grey body radiation emitted from a thermally equilibrated object.

The correlation of radiant intensity between near-IR and visible light has been deduced; although it is expected a linear correlation between them, two correlation lines can be drawn instead of one. This indicates that the contribution of photon emission from the non-equilibrium stage may have an effect.

It is highly desirable to investigate the heat burst phenomena in more detail, for example, to obtain precise time correlation between the bursts in heater temperature and in the radiant power, to analyze the shape of the spectrum, and so on. A detailed analysis is in progress.

5. Acknowledgements

This work is partially supported by CLEAN PLANET Inc. The authors acknowledge also the members of CLEAN PLANET Inc., Mr. H. Yoshino, Mr. S. Hirano, and Mr. H. Shimokawa, for their significant assistance.

References

[1] A. Kitamura, A. Takahashi, R. Seto, Y. Fujita, A. Taniike and Y. Furuyama, "Effect of Minority Atoms of Binary Ni-Based Nano-Composites on Anomalous Heat Evolution under Hydrogen Absorption", *J. Condensed Matter Nucl. Sci.*, **19** (2016) 1-10.

[2] A. Kitamura, A. Takahashi, K. Takahashi, R. Seto, T. Hatano, Y. Iwamura, T. Itoh, J. Kasagi, M. Nakamura, M. Uchimura, H. Takahashi, S. Sumitomo, T. Hioki, T. Motohiro, Y. Furuyama, M. Kishida, H. Matsune, "Excess heat evolution from nanocomposite samples under exposure to hydrogen isotope gases", *International Journal of Hydrogen Energy* **43** (2018) 16187-16200.

[3] Y. Iwamura, T. Itoh, J. Kasagi, A. Kitamura, A. Takahashi, K. Takahashi, R. Seto, T. Hatano, T. Hioki, T. Motohiro, M. Nakamura, M. Uchimura, H. Takahashi, S. Sumitomo, Y. Furuyama. M. Kishida and H. Matsune, Anomalous Heat Effects Induced by Metal Nano-composites and Hydrogen Gas, *J. Condensed Matter Nucl. Sci*. **29** (2019) 119-28.

[4] S. Murakami, T. Itoh, Y. Iwamura, M. Saito and J. Kasagi, "Excess Energy Generation Experiments using a Nano-sized Multilayer Metal Composite and Hydrogen Gas", *Proceedings of the 20th Meeting of Japan CF Research Society*, JCF20, December 13-14, 2019, Reference Hakata Eki-Higashi Rental Room, Fukuoka, Japan p.86-96.

[5] M. Saito, T. Itoh, Y. Iwamura, S. Murakami and J. Kasagi, "Elemental Analysis for Elucidation of the Anomalous Heat Generation Phenomena", *Proceedings of the 20th Meeting of Japan CF Research Society*, JCF20, December 13-14, 2019, Reference Hakata Eki-Higashi Rental Room, Fukuoka, Japan p.74-85.

[6] Y. Iwamura, T. Itoh, J. Kasagi, S. Murakami and M. Saito, "Excess Energy Generation using a Nano-sized Multilayer Metal Composite and Hydrogen Gas", *J. Condensed Matter Nucl. Sci*. **33** (2020) 1-13.

[7] Y. Iwamura, T. Itoh, M. Saito, S. Murakami and J. Kasagi, "Evidence for Surface Heat Release Reaction over Nano-sized Multilayer Metal Composite with Hydrogen Gas", *Proceedings of the 21st Meeting of Japan CF Research Society*, JCF21, December 11-12, 2020, Virtual Meeting, p.1-p14.

[8] Y. Iwamura, T. Itoh, J. Kasagi, M. Saito, T. Takahashi, Y. Shibasaki, and M. Murakami, "Progress in Energy Generation Research using Nano-Metal with Hydrogen/Deuterium Gas", *Proceedings of the 23rd Meeting of International Conference on Condensed Matter Nuclear Science, Xiamen, China*, to be published.

[9] Takehiko Itoh, Yoshinobu Shibasaki, Jirohta Kasagi, Shouichi Murakami, Mari Saito and Yasuhiro Iwamura, "Optical Observation on Anomalous Heat Generation from Nano-sized Metal Composite", *Proceedings of the 21st Meeting of Japan CF Research Society*, JCF21, December 11-12, 2020, Virtual Meeting, p.15-25.

[10] J. Kasagi, "Possible radiation from thin metallic multilayers with hydrogen-permeated anomalous excess heat: Can we observe hot spots or Bremsstrahlung?", *Abstract of ICCF22* 8-13 Sept 2019 Assisi Italy p.10