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Specific Heat Capacity and Emissivity Measurements of Ribbon-Shaped Graphite Using Pulse Current Heating[1]

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A measurement method for specific heat capacity and hemispherical total emissivity of electrically conductive materials with pulse current heating is investigated, in which a ribbon-shaped sample is heated up to 3000 K in a subsecond-duration experiment. Specific heat capacity and hemispherical total emissivity of the sample are calculated from the time variations of heat generation and surface temperature of the sample measured during heating and cooling phases. The true surface temperature of the ribbon-shaped sample is obtained with a radiation thermometer; the directional spectral emissivity of the sample surface is measured using a hemispherical mirror centered at the sample surface. Measurements are performed for POCO AXM-5Q1 graphite in the temperature range from 1500 to 3000 K.

KEY WORDS: emissivity; graphite; high temperatures; pulse heating; specific heat capacity.

1. INTRODUCTION

Graphitic materials, including C/C (carbon/carbon) composites, which can be used at extremely high temperatures have become important. As the conditions of temperature and heat flux for use become critical, thermophysical properties of graphitic materials (such as, specific heat capacity, thermal conductivity, and emissivity) at high temperatures are becoming increasingly important in the thermal design of components, such as the first wall of fusion reactors and the nose cone of space planes. On the other

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hand, the thermophysical property data of graphitic materials are not well evaluated compared to those of metals, because they depend strongly on sample characteristics. Data on the specific heat capacity of graphite are also needed to derive the thermal conductivity at high temperatures from the thermal diffusivity data measured by the laser flash method.

In the pulse current heating method, temperature measurement of the sample with a radiation thermometer is a key technique to attain a high degree of accuracy for the heat capacity and emissivity. Making a small hole in the wall of a tubular sample has been used to attain blackbody conditions [1]. For graphitic materials, it may not be always possible to machine a small precision hole in a fabricated tubular sample because graphitic materials are often brittle and heterogeneous. Thus, a measurement method using a simple ribbon-shaped sample and a radiation thermometer technique combined with a hemispherical mirror has been investigated. This paper describes extended work of the investigation by the authors [2] for a feasibility study of the measurement method.

2. PRINCIPLE OF MEASUREMENT

Assuming that the temperature distribution in the ribbon-shaped sample is uniform and heat loss from the sample is only radiative, the heat balance of the sample between a pair of voltage probes on the sample is given by the following equation:

$$C_p(T_s)\,\rho\,\frac{dT_s}{dt}=\frac{\Delta VI}{WdL}-2\,\frac{W+d}{Wd}\,\varepsilon_{ht}(T_s)\,\sigma(T_s^4-T_c^4) \tag{1}$$

where, C_p, ρ, and ε_{ht} are the specific heat capacity, density, and hemispherical total emissivity of the sample, respectively, σ is the Stefan–Boltzmann constant, t is time, T_s and T_c are the temperatures of the sample surface and the environment, respectively, ΔV is the voltage drop across the voltage probes, I is the current through the sample, and d, L, and W are the thickness, length, and width of the sample between the voltage probes, respectively.

In the heating and cooling phases, the surface temperature, current, and voltage drop are measured. From these data, a pair of time derivatives of the surface temperature in the heating and cooling phases at a given temperature is calculated. Then the specific heat capacity and hemispherical total emissivity of the sample at that temperature are obtained using Eq. (1).

The surface temperature of the sample was measured by a single-band radiation thermometer. The radiation thermometer was calibrated by

determining the coefficients of the calibration equation given by the equation

$$V_b(T) = C \exp\{-c_2/(AT + B)\} \tag{2}$$

where V_b is the output voltage of the radiation thermometer when viewing a blackbody, c_2 is the second radiation constant, T is the temperature of the blackbody, and A, B, and C are the constants characteristic of the radiation thermometer. The output voltage of the radiation thermometer viewing the sample surface through an optical window, V_s, is given by the following equation assuming that the temperature of the sample surface is much higher than that of the environment:

$$V_s = \varepsilon_\lambda \tau V_b(T_s) \tag{3}$$

where ε_λ is the directional spectral emissivity of the sample, and τ is the transmissivity of the optical window. Since the directional spectral emissivity is generally a function of the surface temperature, it was approximated by

$$\varepsilon_\lambda(T_r) = \varepsilon_0 + \varepsilon_1 T_r \tag{4}$$

where ε_0 and ε_1 are constants, and T_r is the radiance temperature of sample surface obtained from Eq. (2) by setting $V_b = V_s$.

The directional spectral emissivity of the sample surface was evaluated before measurements of the surface temperature of samples with a hemispherical mirror having an aperture. The hemispherical mirror is centered at the middle of the sample surface where the surface temperature is to be measured. Assuming that the sample surface is a diffuse reflector and the output voltage of the radiation thermometer is proportional to the radiance of the surface, the directional spectral emissivity of the sample surface, ε_λ, is obtained approximately by the equation [3]

$$\varepsilon_\lambda(T_s) = 1 - \{1 - V_s(T_s)/V_m(T_s)\}/\rho_{me} \tag{5}$$

where V_m and V_s are the output voltages of the radiation thermometer viewing the sample surface with and without the hemispherical mirror, respectively, and ρ_{me} is the effective reflectivity of the hemispherical mirror.

To obtain the ratio of V_s to V_m exactly at the same surface temperature, the temperature dependence of the electrical resistivity of the sample was used. From two data sets measured during heating of the sample with and without the hemispherical mirror, a pair of radiance data where the electrical resistivities are equal was selected to calculate the directional spectral emissivity.

3. APPARATUS

Figure 1 shows a closeup view near the sample in a cylindrical vacuum chamber with an optical window. The inside of the vacuum chamber was evacuated to about 0.2 Pa in the measurements of the specific heat capacity and hemispherical total emissivity, while filled with argon gas at atmospheric pressure to prevent contamination of the hemispherical mirror in the directional spectral emissivity measurement.

The sample was held at both ends by a pair of electrodes. In order to accommodate thermal expansion of the sample, one electrode was allowed to move horizontally. The voltage probes made of graphite having sharp ends were pressed on the sample surface at small cavities machined on the surface (about 0.3 mm in depth). The distance between the small cavities was about 30 mm, and the voltage probes were allowed to be deflected horizontally according to the thermal expansion of the sample.

When the directional spectral emissivity was measured, a hemispherical mirror having an aperture in the oblique direction 15° off normal to the sample surface was placed in front of the sample, as shown in Fig. 1, in such a manner that the center of the hemisphere is positioned exactly on the sample surface to be measured. The inner surface of the hemispherical mirror was coated with gold and the effective reflectivity was estimated to be 0.976 taking into account the aperture.

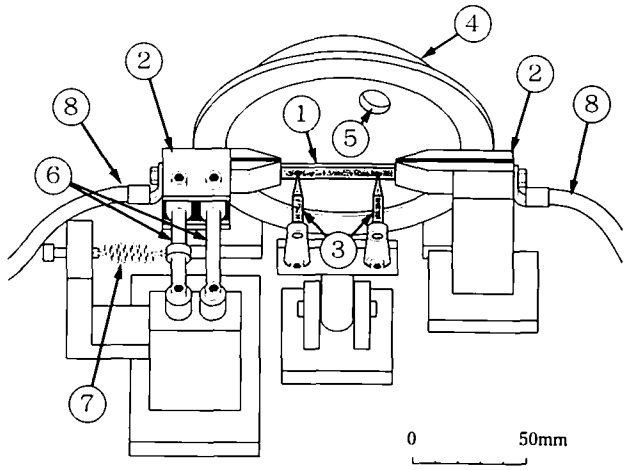

Fig. 1. A closeup view of the sample, electrodes, etc., inside the vacuum chamber. 1, Sample; 2, electrodes; 3, voltage probes; 4, hemispherical mirror (only in the directional spectral emissivity measurement); 5, aperture; 6, links; 7, spring; 8, current wires.

The current was supplied from a condenser bank for a preset duration using FET current switches. The maximum current and voltage available by this current heating system were 3000 A and 50 V, respectively. Current was measured from the voltage drop across a standard resistor inserted in the circuit.

A radiation thermometer using a silicon photodiode (Model IR-RST-90SP, Chino Co. Ltd.) was used for the temperature measurement. The measuring wavelength and bandwidth of the radiation thermometer were 0.9 μm and 25 nm, respectively, and the target size was 3 mm in diameter at a distance of 450 mm. The radiation thermometer had two gains whose ratio was 101.2. The low gain was used for the measurement itself, while the high gain was used to calibrate the radiation thermometer at lower temperatures.

The radiation thermometer was calibrated against a standard radiation thermometer of the same measuring wavelength at seven temperatures in the range from 950 to 1600 K. The standard radiation thermometer had been calibrated against fixed-point blackbody furnaces at the zinc (692.7 K), aluminum (933.5 K), and copper (1357.8 K) points.

The output voltage of the radiation thermometer was recorded in a 16-bit transient memory, while the current and voltage drop were recorded in a 12-bit transient memory. These data were sampled simultaneously every 20 μs and integrated into a data set separated at a time interval of 1 ms by a personal computer.

4. MEASUREMENTS

The measurements were performed on POCO AXM-5Q1 graphite. This material was supplied by the U.S. National Institute of Standards and Technology (RM No. 8426, rod 43, piece 3). The room temperature electrical resistivity is 13.95 $\mu\Omega \cdot$ m. The material, supplied in a cylindrical shape, was fabricated into ribbon-shaped samples, the measurements were made on three such samples. The sample size and density are listed in Table I.

Table I. Size and Density at Room Temperature and Coefficients of the Directional Spectral Emissivity, According to Eq. (4), of the Three Samples of POCO AXM-5Q1

Sample No.	Thickness (mm)	Width (mm)	Length (mm)	Density ($g \cdot cm^{-3}$)	ε_0	ε_1 (K^{-1})
1	0.491	10.05	51.20	1.696	0.92	-3.4×10^{-5}
2	0.498	10.02	51.10	1.701	0.95	-3.5×10^{-5}
3	1.000	10.03	51.19	1.703	0.92	-3.8×10^{-5}

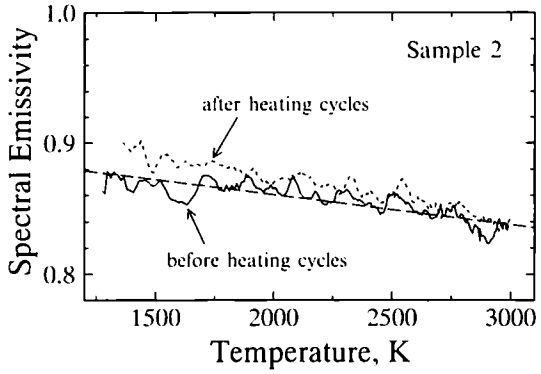

Fig. 2. Directional spectral emissivity of POCO AXM-5Q1 graphite at 0.9 μm measured with a hemispherical mirror in the direction 15° off-normal to the surface.

Figure 2 shows the results of the directional spectral emissivity of Sample 2 measured using the hemispherical mirror. The abscissa shows the true surface temperature of the sample after correcting for the directional spectral emissivity. The fitted broken line was used as the directional spectral emissivity of the sample in the following true temperature determination. The dispersion of the data from the fitted line was 0.8% in standard deviation. The directional spectral emissivity of the same sample measured after more than 10 heating cycles up to 3000 K in a vacuum is also shown by the dotted line. The results show that spectral emissivity increased by 0.02 or less than that before the heating cycles. The coefficients of directional spectral emissivity in Eq. (4) are listed in Table I for the three samples.

Figure 3 shows the results of the apparent electrical resistivity of Sample 3. Two data sets measured under different settings of the voltage probes are presented. One of the settings is the normal one (fixed probe), where the distance between the probes changes according to the thermal expansion of the sample. The other is to allow the probes to slide on the sample surface upon expansion of the sample (sliding probe), where the distance between the probes is kept constant. Assuming that the difference between the two data sets is caused by the thermal expansion of the sample, the average coefficient of linear thermal expansion of the sample is estimated to be about $8 \times 10^{-6}\ K^{-1}$.

Figure 4 shows the results of the specific heat capacity. Sample 1 was measured three times varying the pulse duration (i.e., heating rate). Literature data on the POCO AXM-5Q1 graphite [4] are also shown by the broken line. The differences between the measured data and the literature data are almost within 2%, and the dispersions of the measured

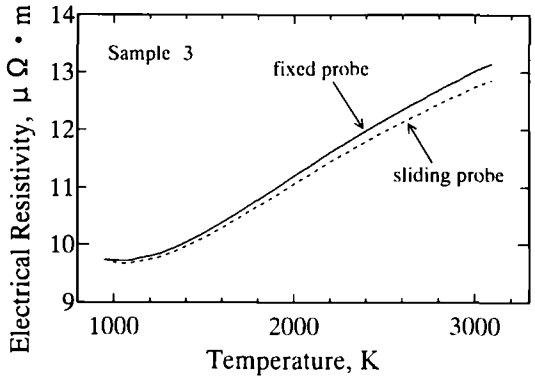

Fig. 3. Apparent electrical resistivities of POCO AXM-5Q1 graphite measured using fixed probes and sliding probes.

data from a fitted curve are about 0.05% in standard deviation above 1700 K. Among the three data sets of Sample 1, a significant systematic deviation related to the heating rate could not be observed. Comparing the data of the three samples, the reproducibility of data measured independently using different samples is considered to be almost within $\pm 2\%$.

Figure 5 shows the results of the hemispherical total emissivity. The dispersions of these data from a fitted curve are about 0.1% in standard deviation above 2000 K. The increase in the dispersions of these data at lower temperatures is due to the increased temperature imprecision of the

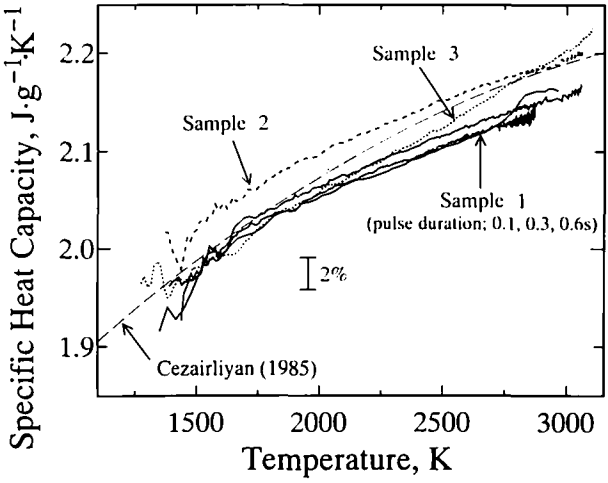

Fig. 4. Specific heat capacity of POCO AXM-5Q1 graphite.

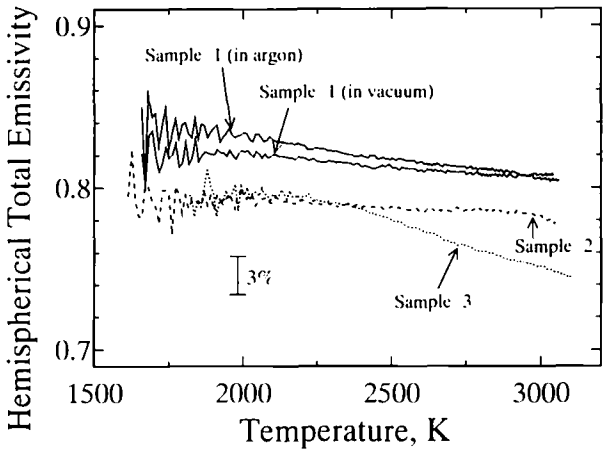

Fig. 5. Hemispherical total emissivity of POCO AXM-5Q1 graphite.

radiation thermometer at lower temperatures. Sample 1 was measured both in vacuum and in argon gas at an atmospheric pressure. The data corresponding to measurements in argon gas are higher than those in vacuum by 2% or less, and the difference becomes larger at lower temperatures. This difference is due to heat conduction from the sample to the gas surrounding it. It should be noted that the data on Sample 3 shows a decrease at high temperatures; the reason for this decrease is not fully understood.

5. SUMMARY

The specific heat capacity, hemispherical total and directional spectral emissivities, and electrical resistivity of POCO AXM-5Q1 graphite were measured in the temperature range from 1500 to 3000 K in order to develop a measurement method for ribbon-shaped samples. The measured specific heat capacity agreed with the reference data almost within $\pm 2\%$. As a result of measurements of three samples cut from the same material, the reproducibility of the measured data of the specific heat capacity was estimated as $\pm 2\%$ standard deviation.

In addition, the difference between two apparent electrical resistivities of the sample measured with different settings of the voltage probes (see Fig. 3) suggests the possibility of a simplified measurement of thermal expansion by measuring the apparent electrical resistivity of the sample.

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