Thermal Conductivity, Electrical Conductivity and Specific Heat of Copper-Carbon Fiber Composites*

By Keiichi Kuniya**, Hideo Arakawa***, Tsuneyuki Kanai*** and Akio Chiba***

We have developed a new copper-carbon fiber composite which possesses properties of copper, i.e., excellent thermal and electrical conductivities, in combination with a property of carbon fiber, i.e., a small thermal expansion coefficient. The composite properties can be controlled by the amount, type and orientation of the carbon fibers they contain. Therefore, this composite is considered to be useful as an electrical material.

The effects of volume and arrangement of fibers on the thermal conductivity, electrical conductivity and specific heat of the composite were studied. The results obtained were as follows:

1. The thermal and electrical conductivities of the composite became smaller as the volume of carbon fibers increased, and they were also influenced by the fiber arrangement.
2. The above conductivities could be calculated by careful application of the "rule of mixtures" for composites.
3. The specific heat of the composite was dependent not on the fiber arrangement but on the fiber volume.
4. In thermal fatigue tests, no degradation in electrical conductivity was observed.

(Received April 27, 1987)

Keywords: carbon fiber, copper-carbon composite, fiber reinforced metal, thermal conductivity, electrical conductivity, specific heat, thermal cycling test

I. Introduction

Very few studies on fiber-reinforced metal (FRM) as a structural material are available in the literature, compared with aluminum-carbon fiber composites which have been developed for improvements in specific strength, heat resistance, etc. The basic characteristics of the composites required for electric and electronic parts are high thermal and electrical conductivities. We have developed a new copper-carbon (Cu-C) fiber composite which possesses properties of copper, i.e., excellent thermal and electrical conductivities, and a property of carbon fiber, i.e., a small thermal expansion coefficient(1)-(10). Since the desirable properties of the composite can be obtained by selecting the amount, type, and orientation of the carbon fibers, it is considered to be suitable for use as electric and electronic materials.

In most of applications of the composite to electric and electronic parts, however, its properties are required to have at least two-dimensional isotropy(1)-(3)(9). In order to form a two-dimensionally isotropic composite, carbon fibers were embedded in a copper matrix in weave and spiral arrangements. In this report, the thermal conductivity, electrical conductivity and specific heat of Cu-C fiber composites and the relationship between theoretical results predicted by the rule of mixtures and the experimental results are discussed.

Further, since the Cu-C fiber composite is composed of heterogeneous materials, namely copper and carbon fiber, material degradation during a long-time use is of concern when it is applied to various electric and electronic parts. Changes in electrical conductivity of the composite were examined through high temperature storage and temperature cycling tests simulating typical service conditions of semi-

---

* This paper was originally published in Japanese in J. Japan Inst. Metals, 49 (1985), 906.
** Hitachi Research Laboratory, Hitachi Ltd., Hitachi.
** Present address: Research and Development Department, Hitachi Nuclear Engineering Co., Ltd., Hitachi 317, Japan.
*** Hitachi Research Laboratory, Hitachi Ltd., Hitachi 319-12, Japan.
conductor devices.

II. Experimental Procedure

1. Materials and specimen preparation

Copper was electrolytically plated to thicknesses of 1-3 \( \mu \text{m} \) on the surface of PAN type high strength carbon fibers (HT fibers\(^{(1)-(3)(5)} \)) having a diameter of about 7 \( \mu \text{m} \). The Cu-plated fibers were semiautomatically arranged in the forms shown in Fig. 1. They were then hot-pressed under a pressure of 24.5 MPa at 1273 K for 1.2 ks in an atmosphere of \( \text{N}_2 + 8 \text{ vol}\% \text{H}_2 \).

After hot-pressing, the specimens were cut out so that thermal and electrical conductivities could be measured in parallel, perpendicular and 45-degree directions to the fibers for the unidirectional and plain weave composites. These properties of the spiral composite were measured only in the direction perpendicular (thickness direction) to the fibers. The spiral and the plain weave, both in the perpendicular direction to the fibers, were used to measure the specific heat.

2. Measurements of thermal conductivity and specific heat

Thermal conductivity and specific heat were measured by the laser flash method. Figure 2 shows a block diagram of the experimental apparatus for the conductivity measurement. Pulsed laser beams were radiated onto specimens (diameter: 10 mm, thickness: 2 mm) having a carbon-sprayed surface. The temperature rise on the specimen backs was measured by using an InSb detector or a thermocouple. The data were sent to a data recorder, consisting of a high transient memory and a micro computer, through a high-speed DC preamplifier. At the same time, some of the laser energy was taken through a laser power monitor, consisting of a Si photo-diode.

Specific heat and thermal conductivity were calculated from the following equations\(^{(1)} \).

Specific heat \( C_p (\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}) \) was computed by using eq. (1)

\[
C_p = \frac{Q}{l \cdot \rho \cdot \Delta T_{\text{max}}}. \tag{1}
\]

Here \( Q \) represents the heat which the specimens absorbed from the pulse light, \( l \) the thickness of the specimens, \( \rho \) the density of the specimens, and \( \Delta T_{\text{max}} \) the maximum temperature on the back of the specimens after the pulse light irradiation.
Thermal conductivity $\lambda$ (W·m⁻¹·K⁻¹) was obtained from eq. (2)

$$\lambda = K \cdot C_p \cdot \rho$$  \hspace{1cm} (2)

where $K$ represents the thermal diffusivity (m²·s⁻¹) and is expressed by the equation

$$K = 1.37(l/\pi)^{1/2}/(t_{1/2})$$  \hspace{1cm} (3)

where $t_{1/2}$ is the time required for the temperature to reach half of $\Delta T_{\text{max}}$ after the pulse light irradiation.

Specific heat was measured by a drop method of measuring the temperature rise of the water absorbing the heat, in addition to the laser flash method.

3. Measurement of electrical conductivity

The size of the spirally arranged composite specimen was 15 mm in diameter and 15 mm thick. The other composite specimens were 4 mm wide, 50 mm long and 4 mm thick. A constant electric current of 1 A was applied from a high-accuracy programable DC voltage-and-current generator and the voltages generated in the specimens were measured by a precision digital voltmeter to obtain their specific resistance. Electrical conductivity was calculated from this specific resistance.

4. High temperature storage and temperature cycling tests

A plain weave composite (length: 50 mm, wide: 8 mm, thickness: 1 mm) was used as a specimen. In the high temperature storage test, the specimen was heated in vacuum at 423, 523 and 673 K for 1000 h, and the changes in electrical conductivity were measured. In the temperature cycling test, the specimen was subjected to a temperature cycle of R.T. (for 300 s) $\xrightarrow{300s} 218$ K (for 1.5 ks) $\xrightarrow{300s} \text{R.T.}$ (for 300 s) $\xrightarrow{60s} 423$ K (for 1.5 ks) using a thermal shock test apparatus, and the change in electrical conductivity was measured.

III. Results and Discussion

1. Influence of content and orientation of fibers on specific heat

Figure 3 shows the relationship between the specific heat and the carbon fiber content of the spiral and plain weave composites. The specific heat of the spiral composite was measured by the drop method, while that of the plain weave composite was measured by the laser flash method. The specific heat of the Cu-C fiber composites becomes larger as the fiber content increases, regardless of the fiber arrangements or measurement method.

The specific heat of the composites ($C_p$) is the sum of the mass ratio of specific heat of each component ($C_1, \ldots, C_n$) and is represented by the following equation\(^{(12)}:\)

$$C_p = (C_1 m_1 + C_2 m_2 + \ldots + C_n m_n) / (m_1 + m_2 + \ldots + m_n)$$  \hspace{1cm} (4)

where $m$ denotes the mass of each component. The specific heats of copper and HT fiber are 385 and 711 J·kg⁻¹·K⁻¹, respectively, and their densities are 8.94 and 1.78 Mg·m⁻³. Using these values the specific heats of Cu-35 vol% C, Cu-45 vol% C and Cu-54 vol% C are calculated from eq. (4) to be 417, 431 and 477 J·kg⁻¹·K⁻¹, respectively. These theoretical values are in reasonable agreement with the measured values shown in Fig. 3. Therefore, the specific heat data in the figure were adopted for the calculation of thermal conductivity.

2. Effect of measuring directions and fiber content on thermal conductivity

Figure 4 shows the effect of measuring directions and fiber content on thermal conductivity.
of unidirectional composites. The thermal conductivity of the composites becomes smaller as the fiber content increases. For the measuring direction, thermal conductivity decreases in the order the parallel direction, 45-degree direction, and perpendicular direction. The thermal conductivity measured in the 45-degree direction is intermediate between the other two.

Figure 5 illustrates the effects of measuring directions and fiber content on thermal conductivity in the spiral and plain weave composites. The thermal conductivity of the latter becomes smaller as the fiber content increases. As to the effect of measuring directions, the thermal conductivity measured in the parallel direction is the largest, while that in the perpendicular direction is the smallest. But the difference in the volumes between the parallel and 45-degree directions is so small that they can be regarded as being the same. Compared with the unidirectional fiber composite, the thermal conductivity in the parallel direction is smaller, but the conductivities in both perpendicular and 45-degree directions are approximately equivalent to that in the unidirectional one. The reason why the thermal conductivity in the parallel direction of the plain weave composite is smaller than that of the unidirectional composite may be attributed to the plain weave composite consisting of the conductivities parallel and perpendicular to the fibers due to its arrangement of warps and wefts.

The thermal conductivity of the spiral composite also becomes smaller in proportion to the increased fiber content. The value is larger than that of the plain weave composite and between the values of the unidirectional composite measured in the parallel direction and the 45-degree direction.

3. Effect of measuring directions and fiber content on electrical conductivity

Figure 6 shows the effects of measuring directions and fiber content on the electrical conductivity in the unidirectional, plain weave and spiral composites. The electrical conductivity is lowered in proportion to the increased fiber content. Comparing the electrical conductivity of the unidirectional composite with that of the plain weave composite, the former is larger in the parallel direction, while both of them are approximately equal in the perpendicular direc-
Thermal Conductivity, Electrical Conductivity and Specific Heat of Copper-Carbon Fiber Composites

The electrical conductivity in the perpendicular direction of the spiral composite is almost the same as that measured in the parallel direction of the plain weave composite. The spiral composite has a copper shaft at its center and the fibers are arranged in concentric circles, which means a continuous copper surface is exposed. Therefore, the spiral composite exhibits similar behavior as for the electrical conductivity measured in the parallel direction.

4. Comparison between measured and calculated values of thermal and electrical conductivities

The thermal conductivity of the unidirectional composite (Fig. 4) in the 45-degree direction was calculated. The thermal conductivities of the composite in the parallel direction, perpendicular direction and 45 degrees from the parallel direction are \( \lambda_p \), \( \lambda \perp \) and \( \lambda_{45} \), respectively, and \( \lambda_\theta \) is given by the following equation:

\[
\lambda_\theta = \lambda_p \cos^2 \theta + \lambda \perp \sin^2 \theta.
\]  

By substituting the thermal conductivities in the parallel direction and the perpendicular direction in Fig. 4 into eq. (5), the thermal conductivities for the \( \theta = 45 \) degree direction of Cu-35 vol\%C, Cu-45 vol\%C and Cu-54 vol\%C are calculated as 225, 175 and 125 W·m\(^{-1}\)·K\(^{-1}\). These agree well with the measured values.

The thermal conductivity in the plain weave composite was estimated from that of the unidirectional composite shown in Fig. 4. Generally, the composites are laminated plates of heterogeneous materials. If it is assumed that the thermal conductivities of the respective components are \( \lambda_1, \lambda_2, \ldots, \lambda_n \), and their volume ratios are \( V_1, V_2, \ldots, V_n \), as shown in Fig. 7, the thermal conductivity of the laminated plate \( \lambda_{c\perp} \) in the parallel direction and that of \( \lambda_{c\perp} \) in the perpendicular direction are expressed as follows:

\[
\lambda_{c\perp} = \lambda_1 V_1 + \lambda_2 V_2 + \ldots + \lambda_n V_n
\]

\[
1/\lambda_{c\perp} = V_1/\lambda_1 + V_2/\lambda_2 + \ldots + V_n/\lambda_n
\]

\[
V_1 + V_2 + \ldots + V_n = 1.
\]

On the other hand, in the plain weave composite, fibers are wavy as shown in Fig. 7. The plain weave composite may be modeled as two composites stacked together, one being unidirectional and the other being inclined by

\[\text{[Clad]} \quad \lambda_{c\perp} \quad \lambda_{c\parallel} \quad \lambda_1, \lambda_2, \ldots, \lambda_n \]

\[\lambda_{c\parallel} = \lambda_1 V_1 + \lambda_2 V_2 + \ldots + \lambda_n V_n \]

\[1/\lambda_{c\parallel} = V_1/\lambda_1 + V_2/\lambda_2 + \ldots + V_n/\lambda_n \]

\[V_1 + V_2 + \ldots + V_n = 1.\]

\[\text{[Plain weave]} \quad \lambda_{c\perp} \quad \lambda_{c\parallel} \quad \lambda_1, \lambda_2, \ldots, \lambda_n \]

Fig. 7 Structural analysis of thermal conductivity in clad and plain weave composites.
the angle $\theta$. Here $\theta$ is the mean angle of inclination of wave-like fibers with respect to the unidirectional fibers. Measurement of the wave angle revealed that $\theta$ was 17.5° and independent of the carbon fiber content (1)-(3). The thermal conductivity $\lambda_{p,\phi}$ in the parallel direction can be calculated as follows: In eq. (6), $\lambda_{c,\phi}$, $\lambda_1$, $\lambda_2$, $V_1$, and $V_2$ are substituted by $\lambda_{p,\phi}$, $\lambda_1$, $\lambda_2$, and $V_1 = V_2 = 1/2$ (see eq. (8)), then $\lambda_{p,\phi}$ is given by the following equation:

$$\lambda_{p,\phi} = (\lambda_1 + \lambda_2)/2.$$  

(9)

In the same way, the thermal conductivity $\lambda_{p,\perp}$ in the perpendicular direction and $\lambda_\phi$ in the $\phi$-degree direction ($\phi = 90° - \theta$) from the perpendicular direction are expressed as follows:

$$1/\lambda_{p,\perp} = 1/(2\lambda_\perp) + 1/(2\lambda_\phi)$$  

(10)

$$\lambda_\phi = \lambda_\phi \cos^2 \phi + \lambda_\perp \sin^2 \phi.$$  

(11)

Table 1 shows the calculated and measured thermal conductivities for comparison. The values agree well with each other.

The electrical conductivity of the plain weave composite was estimated from that of the unidirectional composite in the same way as in the calculation of the thermal conductivity. The electrical conductivities in the parallel and perpendicular directions are expressed as follows:

$$\delta_{p,\phi} = (\delta_1 + \delta_\phi)/2$$  

(12)

$$\delta_\phi = \delta_\phi \cos^2 \phi + \delta_\perp \sin^2 \phi.$$  

(13)

$$1/\delta_{p,\perp} = 1/(2\delta_\perp) + 1/(2\delta_\phi)$$  

(14)

$$\delta_\phi = \delta_\phi \cos^2 \phi + \delta_\perp \sin^2 \phi.$$  

(15)

where

$\delta_\phi$: electrical conductivity of the unidirectional composite in the direction parallel to fibers

$\delta_{p,\phi}$: electrical conductivity of the plain weave composite in the direction parallel to fibers

$\delta_{p,\perp}$: electrical conductivity of the plain weave composite in the direction perpendicular to fibers

From eqs. (12)-(15), the electrical conductivities are calculated and listed in Table 2. The calculated values agree well with the measured ones.

5. High temperature storage and temperature cycling tests

Figure 8 relates the electrical conductivity and heating time when plain weave composites were heated at 423, 523 and 673 K. No degradation in the electrical conductivity is observed after heating for 3.6 Ms (1000 h).

Figure 9 shows the electrical conductivity and the number of temperature cycles of the plain weave composites obtained when the specimens were tested for 1100 cycles of R.T. $\rightarrow$ 218 K $\rightarrow$ R.T. $\rightarrow$ 423 K. No degradation in the electrical conductivity is observed.

6. Estimation of thermal conductivity when applying the composite to semiconductor devices

Since the Cu-C fiber composite has an anisotropy inherent to composites, its thermal conductivity during actual use was suspected to be considerably different from that obtained from one-dimensional laboratory measurements. Overall thermal conductivity of the composite was calculated through a finite element method (FEM) by using two-dimensional heat conduction elements on a power tran-

Table 1: Comparison of experimental and calculated thermal conductivities in plain weave composites.

| Volume fraction of fiber (%) | Thermal conductivity/W·m⁻¹·K⁻¹ |  |  |
|-----------------------------|---------------------------------|---|---|
|                            | Parallel                        | Perpendicular |
|                            | Avg. meas.                     | Calc.         | Avg. meas. | Calc. |
| 35                          | 220                             | 220           | 175        | 175   |
| 45                          | 173                             | 170           | 120        | 125   |
| 54                          | 125                             | 120           | 75         | 70    |
The transistor was composed of a silicon wafer, a thermal buffer material (Cu–C) and a copper stem. Figure 10(a) illustrates the calculated temperature distribution in the transistor interior when a plain weave composite (Cu–35 vol% C) was used. In this calculation, it was assumed that the heating power of the silicon wafer was 100 W and the temperature on the back surface of the copper stem was 273 K. The thermal conductivities of the copper stem and silver solder were both set as 393 W·m⁻¹·K⁻¹. In Fig. 10(a), the solid lines indicate isothermal lines. The maximum temperature rise is 2.828 K and it appears at the center of the silicon wafer.

Then in place of the plain weave composite, maximum temperature rises were calculated on various homogeneous materials with different thermal conductivities. Figure 10(b) illustrates the relationship between the maximum temperature rise obtained from the calculated thermal conductivities of the various homogeneous materials. Thermal conductivity of a homogeneous material, which has the same maximum temperature rise as the plain weave composite, is denoted as a homogeneous material with equivalent thermal conductivity. The equivalent thermal conductivity of the above plain weave composite (Cu–35 vol% C) is 199 W·m⁻¹·K⁻¹ as seen from the figure.

In Fig. 11 are compared the homogeneous material with equivalent thermal conductivity obtained in the above-mentioned way and the thermal conductivities of the plain weave composite in the parallel and perpendicular directions. Regardless of the fiber content, the homogeneous material with equivalent thermal conductivity is larger by about 20 W·m⁻¹·K⁻¹ than that of the plain weave composite in the
perpendicular direction for all fiber volume fractions. Thus, the overall thermal conductivity of the Cu–C fiber composite applied to electric or electronic parts is between the thermal conductivity of the direction parallel to the fibers and that to the direction perpendicular to the fibers.

IV. Conclusions

The following results were obtained in this investigation of the thermal conductivity, electrical conductivity and specific heat of copper-carbon (Cu–C) fiber composites.

(1) The composite specific heat was dependent not on the fiber arrangement but on the fiber volume and it increased with volume increases. The calculated values were almost equal to the measured values.

(2) The thermal and electrical conductivities of the composite were lowered as the volume of carbon fibers increased. Expressed in terms of the fiber arrangement, those of the unidirectional composites were the largest, the spiral composites were intermediate and the plain weave composites were lowest. With respect to the effect of measuring direction, these properties were largest in the fiber parallel direction, intermediate in the 45-degree direction and the smallest in the perpendicular direction.

(3) The thermal and electrical conductivities were predictable by careful application of the "rule of mixtures" for composites.

(4) In the high temperature storage and temperature cycling tests, no degradation in electrical conductivity of the Cu–C fiber composites was observed.

(5) Although the Cu–C fiber composites have anisotropy, their actual thermal conductivity was calculated through a finite element method. This value was between that for the direction parallel to the fibers and that for the direction perpendicular to them.

REFERENCES

(1) K. Kuniya, H. Arakawa, T. Kanai, T. Yasuda, H. Minorikawa, T. Sakaue and K. Akeyama: IEEE Proceedings 33rd Electronic Components Conference, The Institute of Electrical and Electronics Engineers, Inc., New York, (1983), p. 264.

(2) K. Kuniya, H. Arakawa, T. Kanai and T. Yasuda: IEEE Trans. Components, Hybrids Manuf. Technol., CHMT-6 (1983), 467.

(3) K. Kuniya and H. Arakawa: Proceedings 3rd Japan-U.S. Conference on Composite Materials, J. Japan Society for Composite Materials, Tokyo, (1986), p. 465.

(4) K. Kuniya, H. Arakawa and T. Namekawa: Trans. Japan Inst. Metals, 28 (1987), 236.

(5) K. Kuniya, H. Arakawa and T. Kanai: J. Japan Inst. Metals, 49 (1985), 291.

(6) K. Kuniya, H. Arakawa, T. Kanai and A. Chiba: ibid., 49 (1985), 906.

(7) K. Kuniya, H. Arakawa and T. Namekawa: ibid., 49 (1985), 1137.

(8) K. Kuniya, H. Arakawa and T. Namekawa: ibid., 50 (1986), 119.

(9) K. Kuniya, H. Arakawa, T. Sakaue, H. Minorikawa, K. Akeyama and T. Sakamoto: ibid., 50 (1986), 583.

(10) K. Kuniya and H. Arakawa: J. Japan Society for Composite Materials, 10 (1984), 152.

(11) A. Maezono: Introduction to Experimental Techniques for Thermal Analysis, Sinkuriko, Tokyo, (1979), p. 189.

(12) N. Kurihara: Reinforced Plastics, 2 (1956), 164.

(13) N. W. Pilling, B. Yates, M. A. Black and P. Tattersall: J. Mater. Sci., 14 (1979), 1326.

(14) Japan Inst. Metals: Handbook of Metals, Maruzen, Tokyo (1982), p. 986.

(15) H. Maki and S. Simamura: Compilation of Composite Material Technology, Sangyo Gijutu Center, Tokyo (1976), p. 151.