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Characteristics of Highly Thermal Conductive Carbon-Metal Composites and Influence of Interface

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Abstract

The graphite flakes and metals such as Copper (Cu) or Alminium (Al) composites were prepared by pulsed electric current sintering process.Thermal conductivity of the composites was found to depend on the shape and volume ratio of graphite in composites and the influence of interfaces has been clarified. Thecharacteristics were discussed in terms of a simple phenomenological model by taking theinfluence of resistance at the interface of graphite -flakes and metallic (Cu or Al) matrix for the thermal flow into consideration.

1. Introduction

In electronic and optoelectronic devices and apparatuses, dissipation of the heat evolved in them under operation is crucial for realizing excellent characteristics, high reliability and long life. We have developed highly thermal conductive composites of metals such as Copper (Cu) and Alminium (Al) and carbons such as carbon fibers, carbon nanotubes[1)2)].To establish lower costs of composites of high thermal conductivity, in the next step we used graphite flakes as carbons in metal-carbon composites. In this paper, the characteristics of graphite flakes-Cu composites and the influence of the interface thermal resistance are discussed.

2.Experimental

The starting materials were the mixtures of copper powder (mean grain size of 5mm) and graphite flake powder. Three types of graphite flakes with the mean grain size of 500, 200 and 100mm were used. The aspect ratio of length and thickness of the graphite flake was around 10.

Composites were fabricated by the spark plasma sintering (SPS) method. The SPS is a sintering process applying pulsed high current and uniaxial pressure to raw powder materials. The composites were sintered at around 1273K under 50MPa uniaxial pressure with evacuating around 10Pa.

3. Results and discussion

The thermal conductivity of the copper-graphite composites increases with increasing the concent-ration of graphite flakes, which is due to the higher thermal conductivity in graphite than in copper.

The evaluated thermal conductivity of copper-graphite composites with 50% volume fraction of graphiteis shown in Table 1 for various sizes of graphite flakes.

The plane of the graphite flake in sintered

Table 1 properties of copper- graphite composites

No	mean grain size of graphite particle (μm)	Volume fraction of graphite (%)	density (g/cm^3)	relativ density (%)	Thermal conductivity XY (W/m/K)	Thermal conductivity Z (W/m/K)
1	500	50	5.38	96.3	567.4	52.7
2	200	50	5.42	97	451.7	57.8
3	100	50	5.4	96.7	342.3	70.5

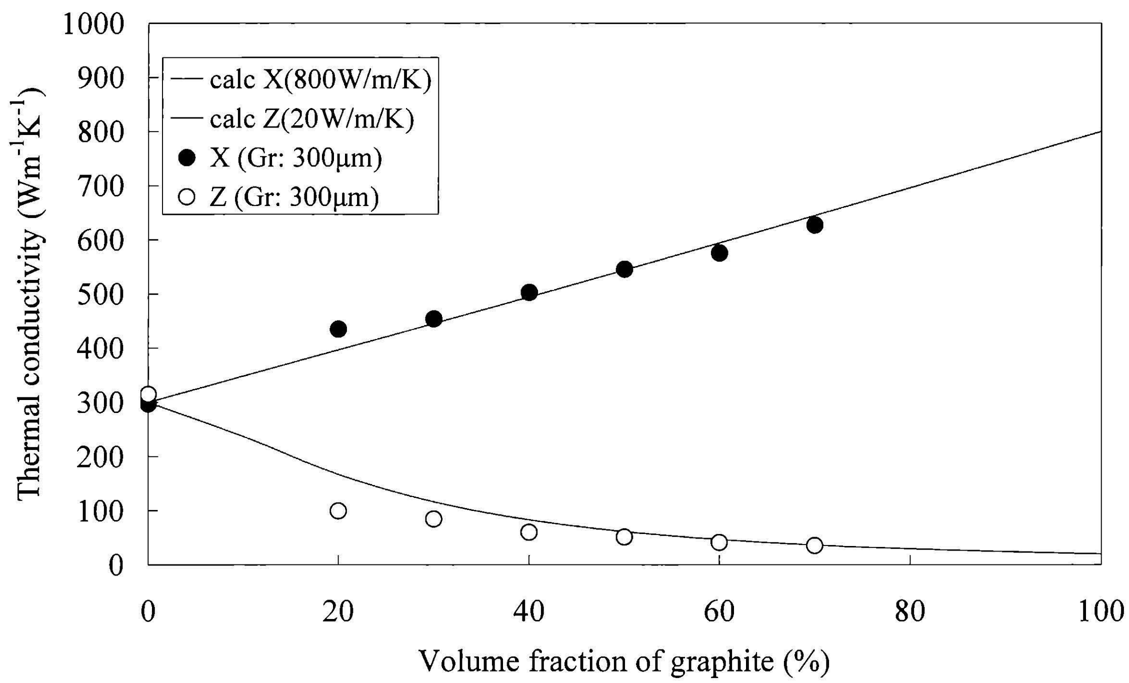

Fig.1 Dependence of thermal conductivity of composites on volume fraction of graphite

samples was aligned perpendicular to pressure axis. In this table, Z-axis is the direction of uniaxial pressure in SPS and X-axis and Y-axis are perpendicular to the direction of pressure. It should be noted that the thermal conductivity along directions in X-Y plane, that is, the thermal conductivity along the direction of mean graphite flake planes, is much higher than the value in the Z-axis direction. This is reasonable, because in graphite flakes themselves the thermal conductivity parallel to the direction of graphite plane is much higher compared to that perpendicular direction. With decreasing the size of graphite flakes, the thermal conductivity in X-Y planes decrease. On the other hand, the thermal conductivity in the Z-direction increases slightly.

For fixed volume fraction of graphite flakes, the number of the interface between copper matrix and graphite flakes in the same volume of the compositeis larger in the smaller graphite flakes. The observed results suggest that the interface thermal resistance of copper and graphite flakes is higher than the thermal resistance of graphite flakes in the direction parallel to the plane. On the

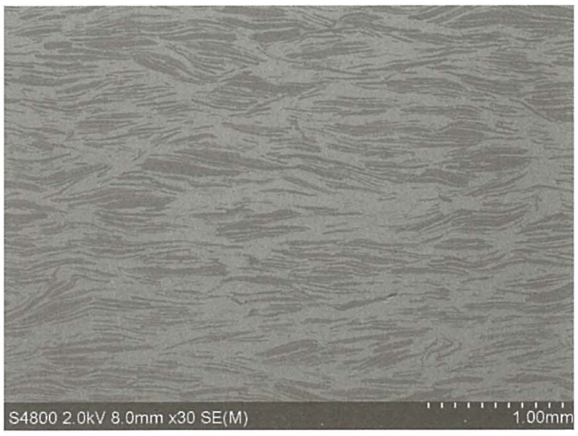

Fig.2(a) SEM image of the sample No. 1 in Table 1

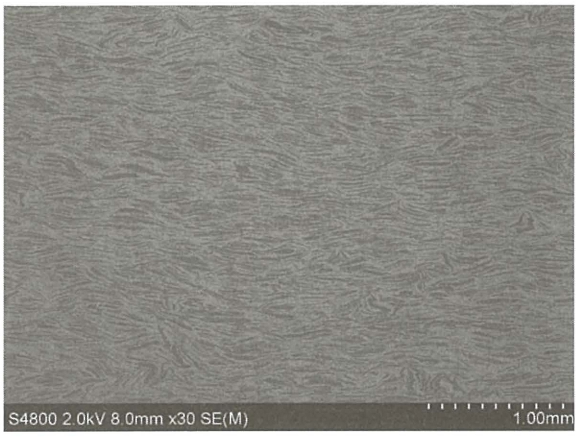

Fig.2(b) SEM image of the sample No. 2 in Table 1

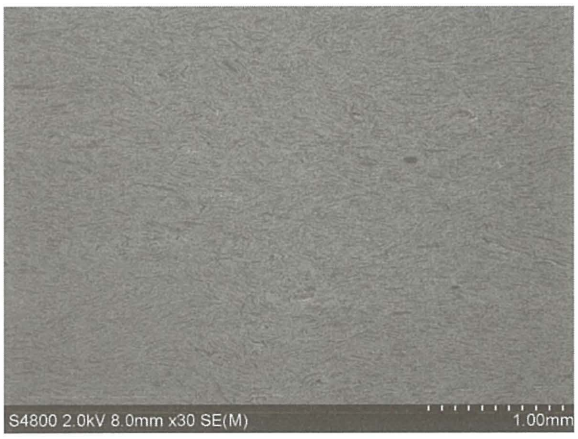

Fig.2(c) SEM image of the sample No. 3 in Table 1

other hand the interface thermal resistance in the Z-axis direction may be even less than the thermal resistance of flakes perpendicular to the flake.
Dependence of the thermal conductivity on volume fraction of graphite flake is indicated in Fig.1.

With increasing volume fraction of graphite, thermal conductivity in the parallel direction to graphite plane increases as indicated by black points and on the contrary that in the perpendicular direction decreases as indicated by white points. This is also reasonable, because with increasing volume fraction of graphite with high thermal conductivity of 800W/m/K, the thermal conductivity of the composite without graphite (thermal conductivity of copper of 300 W/m/K) comes to close to the value of pure graphite of 800W/m/K. On the other hand, the thermal conductivity perpendicular to flakes plane decreases with increasing volume fraction approaching to the value of pure graphite of 20W/m/K. However they are not linear function of the graphite fraction.

In this figure solid lines are theoretical dependence obtained by relatively simple phenomenological calculation, which will be reported in the separate paper. For precise fitting of theoretical curves we must take not only the mean orientation of graphite flakes but also fluctuation of the orientation around the mean orientation into consideration.

Indeed as shown in Fig.2, fluctuation of the orientation was confirmed by electron microscopic observation. Figure2 shows the SEM images of copper- graphite composites. The SEM images are the cross-sectional view along pressure axis in sintering. By the introduction of this fluctuation of orientation in the calculation still better theoretical fitting curve to the experimental data was obatained.

Detailed discussion on thermal conductivity in carbon-metal composites by taking interface resistance so called Kapitzaresistance into consideration will be reported in separated paper[3].

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