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THERMAL CONDUCTIVITY AND THERMAL EXPANSION OF GRAPHITE FIBER/COPPER MATRIX COMPOSITES

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SUMMARY

The high specific conductivity of graphite fiber/copper matrix (Gr/Cu) composites offers great potential for high heat flux structures operating at elevated temperatures. To determine the feasibility of applying Gr/Cu composites to high heat flux structures, composite plates were fabricated using unidirectional and cross-plied pitch-based P100 graphite fibers in a pure copper matrix. Thermal conductivity of the composites was measured from room temperature to 1073 K, and thermal expansion was measured from room temperature to 1050 K. The longitudinal thermal conductivity, parallel to the fiber direction, was comparable to pure copper. The transverse thermal conductivity, normal to the fiber direction, was less than that of pure copper and decreased with increasing fiber content. The longitudinal thermal expansion decreased with increasing fiber content. The transverse thermal expansion was greater than pure copper and nearly independent of fiber content.

INTRODUCTION

Many aerospace applications require materials with high thermal conductivity to reduce component operating temperature, extend service life, and reduce system weight. Hypersonic vehicle applications such as high conductivity heat exchangers require service lives of hundreds to thousands of hours. They require materials with good elevated temperature tensile and creep strengths, thermal fatigue resistance and oxidation resistance. The materials for the heat exchangers must be usable at temperatures ranging from 800 K for the cooling channels up to 1200 K for the hot side in the combustor. Space power system applications include high conductivity radiator fins with service lives of 10 to 30 years. They require high stiffness and low density. The materials for the radiator fins must be usable at temperatures ranging from 300 K for solar power systems up to 1050 K for nuclear power systems.

Several materials can be used for high heat flux thermal applications. Copper offers the second best thermal conductivity of the elements (ref. 1). It is widely available in many forms and can be easily machined and formed. The primary problem with using copper for aerospace applications is its high density (8.96 g/cc) (ref. 1). Copper also suffers from a low yield strength and stiffness (ref. 2). Beryllium is often considered for aerospace applications because of its low

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Thermal conductivity and density data are required to determine the feasibility and basic design parameters of Gr/Cu components. Thermal expansion data for Gr/Cu composites is required to design joints to minimize thermal expansion mismatch with heat pipes and coolant manifolds. The effect of temperature and fiber content of unidirectionally reinforced Gr/Cu composites on thermal conductivity and thermal expansion are described in this paper.

DESIGN, FABRICATION AND TESTING OF Gr/Cu COMPOSITES

Before describing the results obtained for Gr/Cu composites, some terms used to describe metal matrix composite (MMC) lay up and the property anisotropy of MMCs and the graphite fibers will be introduced. All unidirectional MMCs have anisotropic properties. However, the highly aligned crystallinity of the ultra-high modulus (UHM) P100 fibers causes very large anisotropy in UHM graphite fiber reinforced composites. Figure 1 shows the directional axes that will be used in the discussion of the effect of longitudinal and transverse orientations on Gr/Cu composite properties. Conductivity, modulus, and strength are maximized in the longitudinal direction and minimized in the long transverse and short transverse directions. In addition, figure 1 also shows angle-plied composites can be produced by orienting the fiber direction in the individual plies. By angle-plying the properties in the longitudinal and long transverse directions can be tailored to the application requirements.

The results previously reported (refs. 8 and 9) on Gr/Cu composites were from vendor supplied panels or NASA fabricated composite panels. Some contractor supplied panels used P-120 and P-130 graphite fibers to compare the effects of different graphite fiber types. All NASA fabricated panels were made with copper-coated P100 graphite fibers supplied by American Cyanamid Co. (ref. 10). Selected results from the NASA fabricated panels in the previous studies (refs. 8 and 9) of Gr/Cu composites are reported in this paper for comparison. These Gr/Cu composites were made using a labor intensive hand layup of a copper coated graphite fiber tow. The volume percent fiber was determined from the density of the samples to be 67 volume percent (vol %). Optical microscopy of the samples revealed some porosity in the samples due to incomplete consolidation. The actual volume percent fiber is therefore somewhat lower than the calculated value.

The Gr/Cu composites containing 20 to 50 vol % graphite fibers discussed in this paper were made by a new fabrication technique developed at NASA Lewis. Yarns were supplied with different coating thicknesses to provide nominal fiber contents from 20 to 60 vol % of graphite fibers. The copper coated yarns are passed through a series of cleaning baths to remove any surface contamination and wound onto a drum at a precise spacing (fig. 2(a)). After winding, a thin coating of copper is arc sprayed onto the yarns using the arc spray process described in reference 11. This produces a well aligned unidirectional monotape such as the ones shown in figure 2(b) that can be cut, handled and positioned with ease. After arc spraying, the monotapes were laid up as 6-ply unidirectional composites and hot isostatically pressed into panels 200 mm long by 100 mm wide by 1.0 to 1.5 mm thick. An example of the panels is shown in figure 2(c). Microstructures of the as-pressed Gr/Cu composites (fig. 3) show a uniform fiber distribution and good consolidation of the monotapes with grain boundaries and twins extending through the prior particle boundaries. No porosity was observed in any of the consolidated panels. Fiber contents of the panels were determined by specific gravity measurements.

One Gr/Cu composite plate was made with eight plies cross-plied at a 90° angle. The outer two plies on each face of the plate were parallel to the longitudinal axis of the plate. The

Thermal conductivity is calculated using the equation (ref. 12)

$$\sigma_T(T) = C_P(T) \cdot \rho \cdot \lambda(T) \tag{1}$$

where

$\sigma_T(T)$	calculated thermal conductivity at temperature T
$C_P(T)$	measured specific heat at temperature T
ρ	room temperature bulk density
$\lambda(T)$	measured thermal diffusivity at temperature T

Thermal conductivity in both the longitudinal and short transverse directions is shown in figure 5 for Gr/Cu composites with a variety of fiber contents. In the longitudinal direction, the thermal conductivities of the composites fell in the narrow band shown in figure 5. Within this band there was no correlation of fiber content with the upper or lower limit of the band. This is because the longitudinal thermal conductivity of the P100 fibers used is near that of pure copper. At the lower temperatures the Gr/Cu composites have a higher thermal conductivity than copper, but at higher temperatures the thermal conductivity drops to values near copper. The observed drop in thermal conductivity is caused by the greater decrease in thermal conductivity of P100 fibers compared to Cu over the temperature range.

In the transverse directions, the thermal conductivity is less than in the longitudinal direction and decreases with increasing fiber content (fig. 6). The decrease is caused by the anisotropy of the UHM P100 graphite fibers. As graphite crystallites become more aligned, lattice orientation dependent physical properties such as elastic modulus and thermal conductivity become higher in the direction of the fiber axis. However, the anisotropy of the properties in the radial direction becomes even more pronounced (ref. 14). While the thermal conductivity of the fiber along the axis of the fiber is very high, it is extremely low in the radial direction. In the transverse directions of Gr/Cu composites, heat is conducted almost exclusively through the copper matrix. Extrapolation of the thermal conductivity versus fiber content curves indicates that the radial thermal conductivity of the P100 graphite fibers is near zero. Thus the higher the fiber content, the lower the transverse thermal conductivity of the composite. Despite the decrease in thermal conductivity, composites with fiber contents up to 60 vol % have thermal conductivities higher than titanium and niobium alloys (fig. 6), the primary competitive materials at higher temperatures where Be and Cu cannot be used.

The thermal conductivity in the long transverse direction is about 10 percent higher than the short transverse direction. This difference is caused by the sample processing. Heat must pass around all fibers in the short transverse direction. In the long transverse direction there are thin layers of copper rich material that are the result of the arc spray process. These thin layers allow for easier diffusion of the heat than in the short transverse direction. With the increase in thermal diffusivity, the thermal conductivity increases as shown in equation (1).

The density dependent specific thermal conductivity (thermal conductivity divided by density) is a more important design criterion for the reduction of mass in thermal management structures. The effect of fiber content on the specific thermal conductivity of Gr/Cu composites is shown in figure 7. In the longitudinal direction, specific thermal conductivity increases with increasing fiber content though the absolute thermal conductivity is relatively unchanged. This using available data on the thermal expansion of the PEEK matrix and the transverse thermal expansion of the Gr/PEEK composite. The results led to a calculated CTE of 23×10^{-6}/K for temperatures up to 200 °C compared to a copper CTE of 17.2×10^{-6}/K over the same temperature range (ref. 1). With the fibers expanding more rapidly than copper, the total matrix thermal strain in the long transverse direction exceeds the thermal strain for pure copper as is observed in the Gr/Cu samples.

In the longitudinal direction, thermal strain decreased with increasing fiber content. The 20 vol % Gr/Cu composite showed an almost uniformly increasing curve paralleling the copper curve. For the 30 vol % Gr/Cu composite, the curve started on an upsweeping curve parallel to the copper curve, but changed slope around 650 K and flattened out to a nearly uniform expansion up to 1050 K. The 40 vol % Gr/Cu composite shows similar behavior. At 50 vol % graphite fiber, the curve started with a negative expansion similar to the P100 fibers and reached a minimum near 650 K. Above 650 K the samples expanded slightly, but were still near zero thermal expansion.

Based on the behavior of the samples, some general ideas were developed to explain the longitudinal thermal strain data. The graphite fibers contract in the longitudinal direction but expand in the radial direction during heating. The radial expansion pushes the fibers against the copper matrix and forces a mechanical or frictional bond. The contraction of the fibers in the longitudinal direction restrains the thermal expansion of the copper matrix and therefore the Gr/Cu composite. This results in a thermal strain curve roughly parallel to the pure copper curve but at a lower absolute thermal strain value. This is the type of behavior seen during the heating of the 20 vol % Gr/Cu composite. With increasing graphite fiber content, the fibers constrain the matrix more, and the thermal strain at a given temperature is decreased. For the 50 vol % Gr/Cu composite the fibers totally dominate the thermal expansion of the Gr/Cu composite, and the composite shows a near zero thermal strain throughout heating.

The 30 and 40 vol % Gr/Cu composites show a change in slope of the thermal strain curves during heating. The observed change in slope of the thermal strain curves can be explained by yielding of the copper matrix. At elevated temperatures the yield strength of copper drops to very low values. The large CTE mismatch between the fibers and the matrix can therefore generate sufficient thermal stresses to cause compressive yielding of the matrix. Evidence of this was seen as macroscopic slip bands and cracks on the surfaces of the samples. As a result of the compressive yielding, the matrix expands less than it would otherwise. The slope of the thermal strain curve would decrease as is observed in figure 8.

Modeling is currently underway to understand better the stress states of the composites and will be presented in a later paper. For now it is sufficient to know that thermal stresses sufficient to cause compressive yielding of the copper matrix can be generated by the CTE mismatch between the fiber and matrix after heating only a few hundred degrees Celsius.

The thermal strain of P100 Gr/Cu composites in the longitudinal and long transverse directions at 800 K are plotted in figure 9. The plot shows the thermal strain in the long transverse direction increases at low fiber contents but is essentially constant above 20 vol % Gr. The longitudinal thermal strain decreases with increasing fiber content from the copper value and becomes negative around 50 vol % Gr. Above 50 vol % Gr, the thermal strains would extrapolate to a negative value equal to the thermal strain of P100 graphite fibers at 100 percent P100 graphite fibers. The longitudinal CTE values for Gr/Cu composites at 800 K is presented in figure 10. For these tests the zero point was defined as 300 K. The transverse CTE of the

For the samples with a larger volume fraction of P100 graphite fibers, the reversal of stress states occurs at higher temperatures, and the net room temperature strain is near the value for 1073 K.

Kural and Min (ref. 17) analyzed the thermal deformation of P100 graphite fiber reinforced magnesium (Gr/Mg) and P100 graphite fiber reinforced 6061 aluminum (Gr/6061) composites during low temperature thermal cycling. The Gr/Mg and Gr/6061 composites had a near zero CTE. For the Gr/6061 composite, the graphite fiber plies were angleplied at ±26° to achieve the desired longitudinal CTE. Kural and Min's analysis of the lamina in the composites showed a similar hysteresis loop. They also explained the observed behavior in terms of plastic yielding in the matrix combined with a stress state reversal during the thermal cycle. While direct comparisons between the two composites and the Gr/Cu composites studied here are complicated by several metallurgical factors, the results from the Gr/Mg and Gr/6061 composites do indicate that the basic ideas put forth for the Gr/Cu composites are probably valid. A more detailed analysis is required to confirm and quantify the results for the Gr/Cu composites.

The observed hysteresis of the thermal strain curves indicates two potential problems in using the Gr/Cu composites for applications with thermal cycling. The increase in size of the samples indicates the CTE mismatch may induce matrix stresses that are large enough to nucleate and grow voids. Voids have been observed in Gr/Cu samples following thermal exposure (ref. 18). Thermal ratcheting also could be caused by the accumulation of strains in the composite with repeated thermal cycling. For space power applications, there would be only a few thermal cycles associated with the manufacturing and testing of the composites. Once the space power system is activated in space, the radiator fins will remain at a constant temperature for the life of the system. Proper design and materials selection can be used to mitigate the problem. For hypersonic vehicles and similar applications, the composites will be required to undergo hundreds or thousands of complex thermal cycles during the life of the parts. They also will be required to maintain the integrity of the seal with manifolds and other parts of the thermal structures. As such, large thermal deformations cannot be tolerated. More work is required to address this problem.

All the data discussed thus far dealt with unidirectionally reinforced Gr/Cu composite plates. Figure 13 shows the thermal strain in the longitudinal and long transverse directions for a cross-plied 50 vol % Gr/Cu composite plate. Figure 13 also shows the heating cycle thermal strain of a unidirectionally reinforced 50 vol % Gr/Cu composite and copper for comparison. This plot shows that thermal strains of both the longitudinal and long transverse directions of the cross-plied composite fall significantly below the unidirectional longitudinal experimental data and above the unidirectional long transverse thermal experimental data. The cross-plied composites show a behavior dominated by the P100 graphite fibers.

The thermal strain results presented indicate that the thermal expansion behavior of Gr/Cu composites can be tailored by modifying the volume fraction of the fibers and orientation of the plies to meet the desired design criteria. By proper selection of fiber content and the orientation and number of plies in each direction, the thermal expansion of Gr/Cu composites can be varied from near zero to values greater than copper. This would encompass the range of most materials anticipated for use as manifolds or heat pipes to be used with Gr/Cu composites in high heat flux thermal management structures.

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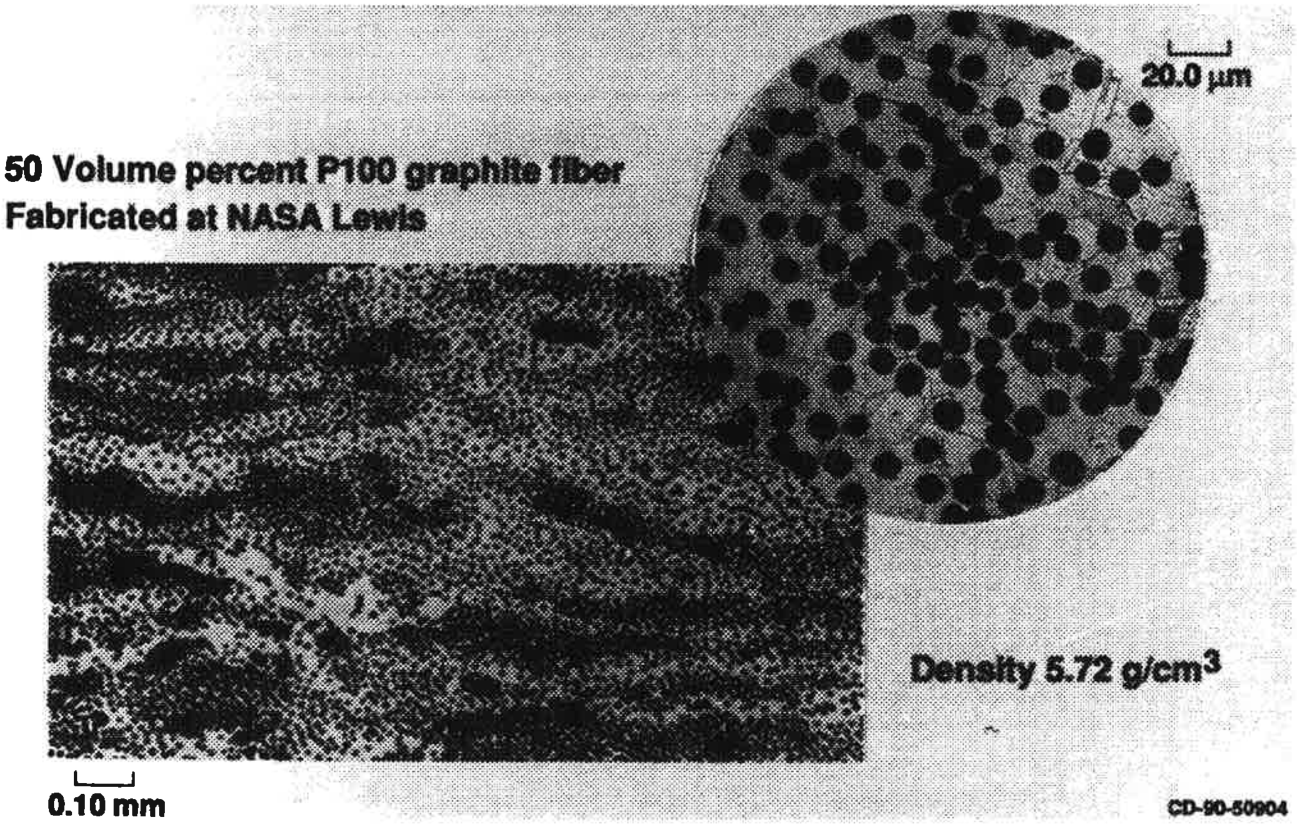

Figure 3.–Microstructure of 50 v/o P100 Gr/Cu composite. Transverse Cross-section.

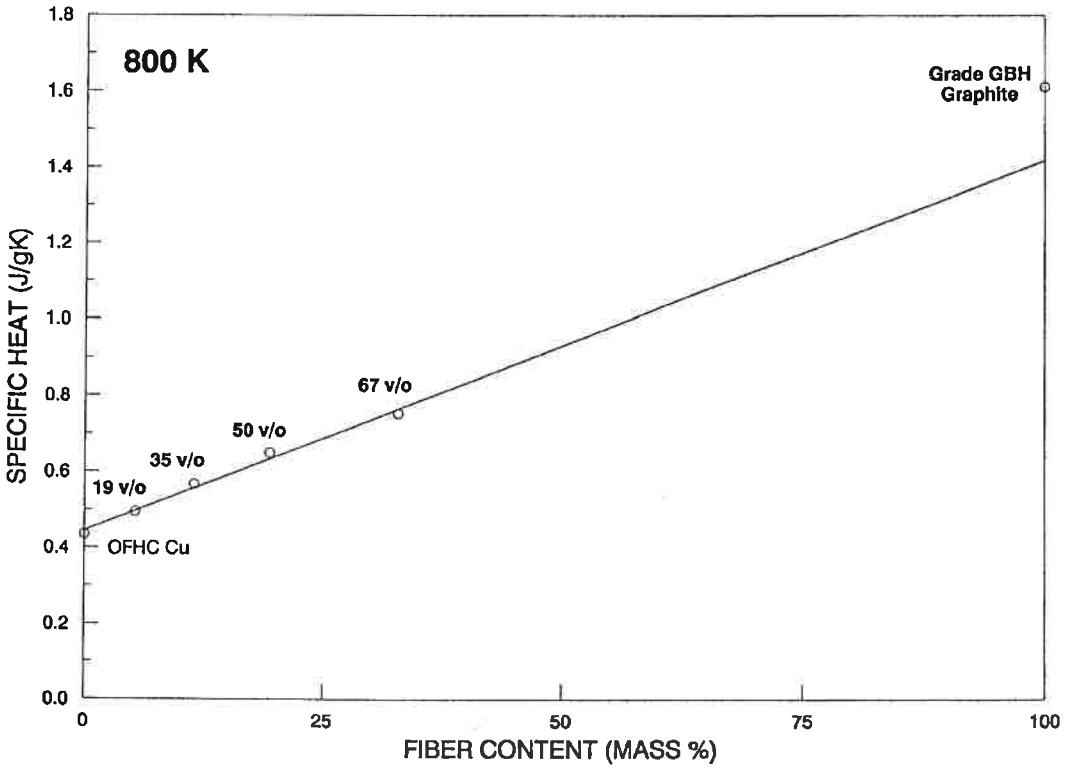

Figure 4.–Effect of fiber content on specific heat of P100 Gr/Cu composites at 800 K .

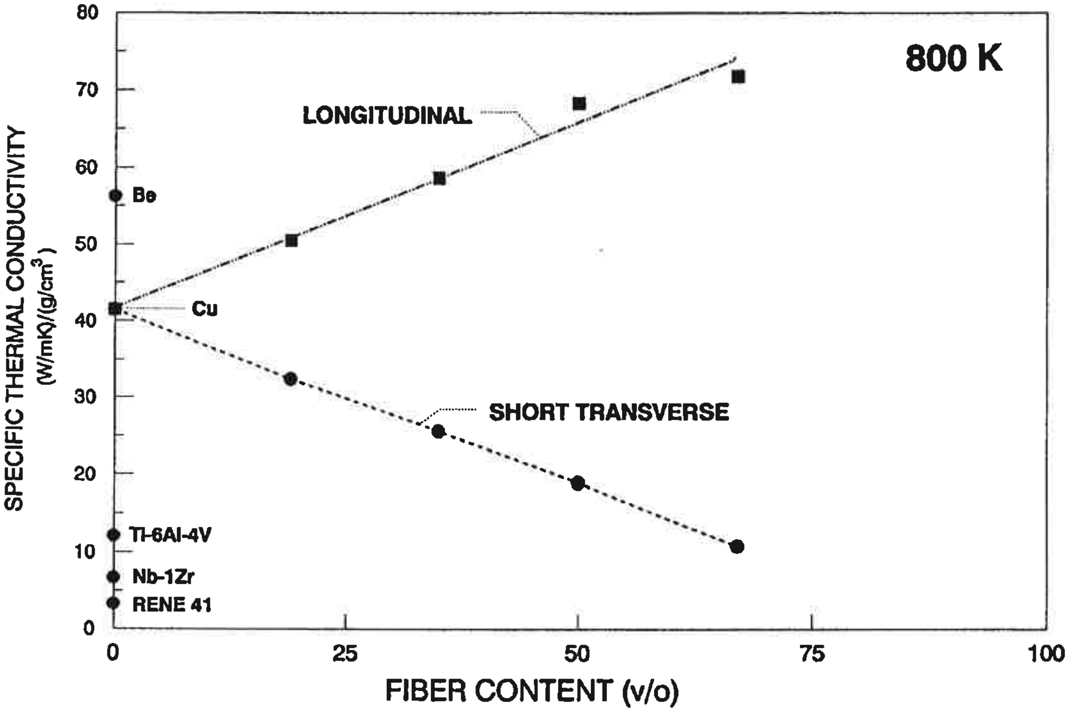

Figure 7.–Effect of fiber content on specific thermal conductivity of P100 Gr/Cu composites at 800 K.

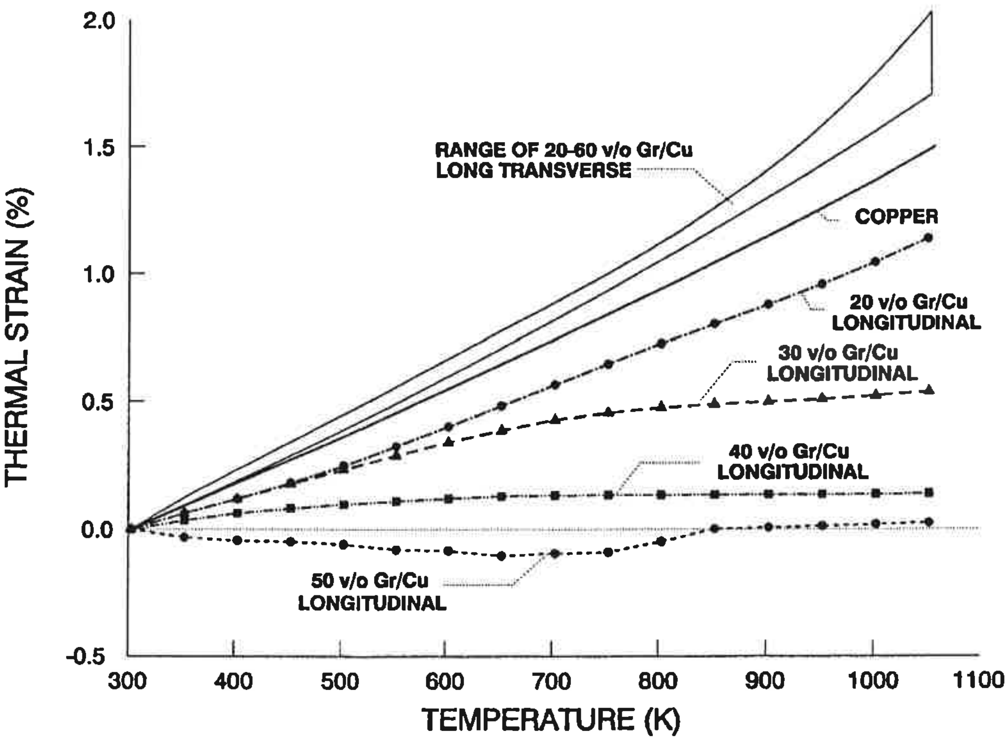

Figure 8–Effect of fiber content on thermal strain of P100 Gr/Cu composites during heating from 300 K to 1050 K.

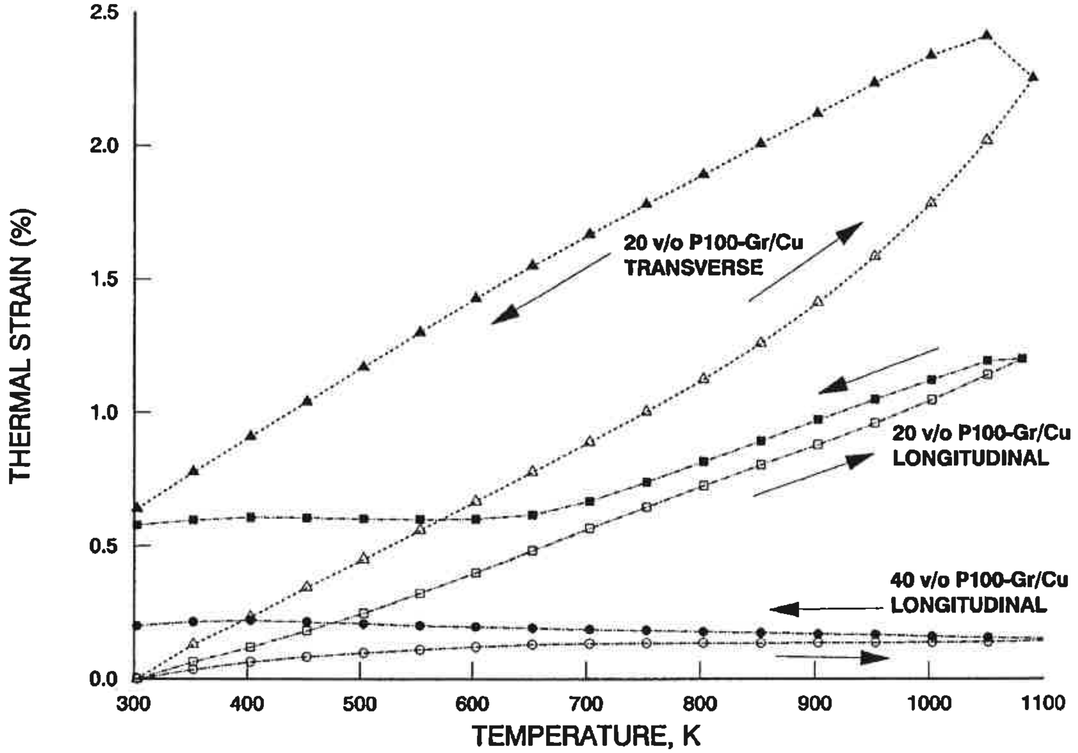

Figure 11.–Thermal strain of three typical P100 Gr/Cu composites for both heating and cooling curves.

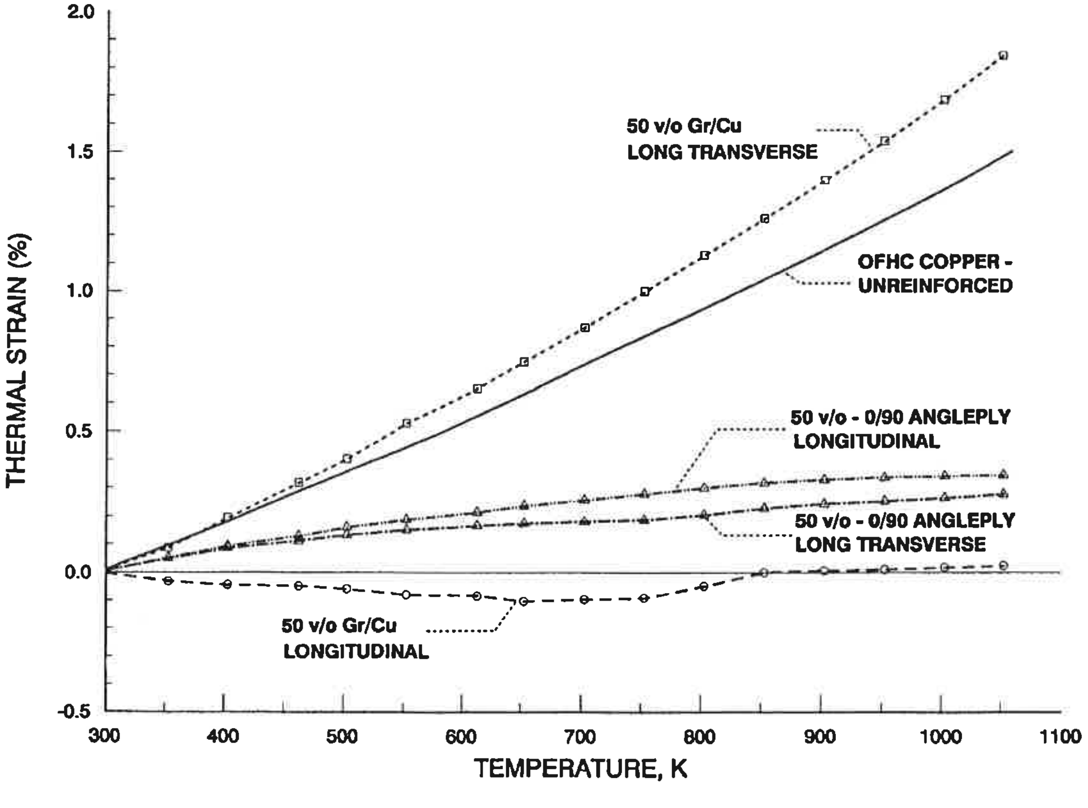

Figure 12.–Effect of fiber orientation on thermal strain of 50 v/o P100 P100 Gr/Cu composites during heating from 300 K to 1073 K..