Effect of Interfacial Thermal Resistance on Effective Thermal Conductivity of MoSi$_2$/SiC Composites

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The thermal conductivity of molybdenum disilicide composites reinforced with 100 nm and 0.5 μm silicon carbide particles was determined as the function of volume fraction of silicon carbide. Due to interface effect, the thermal conductivity of composites with same volume fraction of silicon carbide decreased with decreasing inclusion size. It has been found that at lower inclusion content, interfacial thermal resistance dominates in determining composite thermal conductivity, while with increasing content of dispersions, percolative thermal transport paths diminishes the effect of interfacial thermal resistance.

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1. Introduction and Background

The thermal conductivity of composites has intrigued great interest for about a century. It is well known that due to the mathematical analogy between thermal conduction, electrical conduction and electrostatics, results obtained in one area can be applicable to the other. Effective thermal conductivity of composite materials is usually determined not only by the constituent conductivities, volume fraction, shape and microscale arrangement but also by interfacial thermal resistance between the constituent phases. The interfacial thermal resistance (the reciprocal of interfacial thermal conductance) is possibly attributable to two factors: (a) thermal contact resistance due to poor mechanical and chemical bonding between constituent phases and (b) thermal boundary resistance due to differences in the physical properties of the composite’s constituents, also known as Kapitza resistance due to the diffuse scattering of phonons at interfaces.

The interfacial thermal resistance in a composite refers to the combined effect of the thermal contact resistance and the thermal boundary resistance.

Many experiments have shown that the interfacial thermal resistance has a dramatic effect on the effective thermal conductivity of composites. Based on Effective Medium Theory, Hasselman and Johnson, and recently Nan obtained the same expression of the effective thermal conductivity of composites with spherically dispersed dispersions:

$$K_{\text{eff}} = K_m + \left(1 - \frac{K_d}{K_m} \frac{V_d}{V_m} + \frac{K_d}{K_m} \frac{2K_d}{ah_c} + 2\right)$$

where $K_{\text{eff}}$ is the effective thermal conductivity of a composite; $K_d$ and $K_m$ are thermal conductivity of the dispersions and the matrix material; respectively; $V_d$ refers to the volume fraction of dispersion; $h_c$ is the interfacial thermal conductance, and $a$ the radius of dispersion.

In this paper, we reported the experimental results for the effect of interfacial thermal resistance and volume fraction on the effective thermal conductivity of Molybdenum disilicide matrix composites reinforced with SiC particulate.

2. Experimental Procedures

High purity MoSi$_2$ (Riken, Japan, 99.5%, 3.6 μm) and two kinds of SiC with particle sizes (Siccs, China, 100 nm and 0.5 μm) were used as starting materials. Specimens of MoSi$_2$ reinforced with 0%, 10%, 20%, and 30% volume of two kinds of SiC particles were prepared via Spark Plasma Sintering at the temperature range from 1720 to 1820 K, depending on the content of SiC, under 50 MPa of applied pressure in vacuum with dwelling time of 5 min. Slices with thickness of about 2 mm and diameter of 10 mm were machined from as-prepared samples. All composites were fully dense. The distribution of SiC reinforcements was observed by optical microscopy (Olympus BX51).

Thermal diffusivity was measured by means of laser flash technique at temperatures ranging from room temperature to 1000 K. The specimens were in the form diameter of 10-mm-diameter circular disks with a thickness of about 2 mm. Specific heat of composites was obtained from specific heat of MoSi$_2$ and SiC using the rule of mixtures. The thermal conductivity was calculated by multiplying the experimental data for the thermal diffusivity, specific heat and density. In the thermal conductivity determination, the changes in specimen thickness and density were found to be negligible.

3. Results and Discussion

3.1 Microstructures

Figure 1 shows the optical micrographs of MoSi$_2$/SiC composites with various SiC content and different SiC particle size. It can be seen that SiC is homogeneously distributed in MoSi$_2$ matrix. For composites reinforced with 100 nm and 0.5 μm SiC, the aggregate size of SiC increases with increasing SiC volume fraction. For composites with same volume fraction of SiC, the aggregates in composite with 0.5 μm SiC exhibit a larger averaged size than that in the composite with 100 nm SiC.
3.2 Effective Thermal Conductivity of composites

Figures 2(a) and (b) show the temperature dependency of the thermal conductivity of the composites with 0.5 μm SiC and 100 nm SiC, respectively. In both figures, the thermal conductivity of all composites shows a negative temperature dependency. In Fig. 3, it can be found that, with increasing volume fraction of SiC, the thermal conductivity of the composite with 0.5 μm SiC at room temperatures decreases until 20 Vol% SiC, then increases. The thermal conductivity of both composites bears a similar variation with temperature.

3.3 Determination of interfacial thermal resistance and inclusion thermal conductivity

Figure 3 gives the thermal conductivity variation of the composites reinforced with both 100 nm SiC particles. From Fig. 3, for the composites with 0.5 μm SiC, \( K_m = 55.3 \text{ W/mK} \), \( a = 0.5 \times 10^{-6} \text{ m} \). Substitution of thermal conductivities of the composites with 10 and 20 vol% SiC into eq. (1) gives rise to two equalities with two unknown parameters, \( K_d \) and \( h_c \). The set of two equalities yields \( K_d = 325 \text{ W/mK} \) and \( h_c = 2.11 \times 10^8 \text{ Wm}^{-2} \text{ K}^{-1} \). Similar calculation for composites with 100 nm SiC gives \( 310 \text{ W/mK} \) and \( 1.32 \times 10^8 \text{ Wm}^{-2} \text{ K}^{-1} \) for \( K_d \) and \( h_c \). It is plausible that the thermal conductivity of SiC dispersions is larger than that of polycrystalline SiC materials and lower than that of single crystal SiC. Since thermal conductivity of materials is proportional to the size of materials, \( K_d \) for 100 nm SiC is reasonably smaller than that for 0.5 μm. The values of \( h_c \) is of the same order of magnitude with those obtained by Xu et al. \( h_c \) obtained for the composites with 100 nm SiC is a little lower than that for the composites with 0.5 μm SiC, which is due to dislocation and high interface density induced by SiC particle loading. In the above calculation, the data for composites with 30 vol% SiC was not used since possible percolation occurs for SiC phase, which is sure to result in a higher \( h_c \).

3.4 Effect of interfacial thermal resistance and SiC content

With increasing volume fraction of SiC, the thermal conductivity variation of the composites reinforced with both...
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100 nm and 0.5 μm SiC shows a decreasing and then increasing trend. As a matter of fact, interfacial thermal resistance in eq. (1) results a monotonous decrease in composite thermal conductivity, which is in contrast with the experimental results. If we further increase the content of SiC, the thermal conductivity of the composites will increase further since the thermal conductivity of SiC is larger than that of MoSi$_2$ and larger aggregates of SiC result in the less pronounced effect of interfacial thermal resistance. Interestingly, there must be certain volume fraction ($V_{cri}$) at which the addition of SiC has no enhancement or decrease effect on the thermal conductivity of composites, compared to that of MoSi$_2$. We call this volume fraction “critical volume fraction”. For a given composite system and inclusion size, $K_{eff} = K_0$ should yield two results: $V_d = 0$ and $V_{cri}$. However, equation (1) only gives $V_d = 0$. With increasing volume fraction of SiC, the aggregates become bigger and bigger, forming percolative heat conduction paths, which results in an additional enhancement in thermal conductivity. However, the ratio of conductivities $K_d/K_m \sim 10$ is not high enough to induce a strong percolation threshold effect on thermal transport. From theoretical viewpoint, the $V_{cri}$ does not belong to the dilute case, on which eq. (1) works. Obviously, at lower content of SiC, interfacial thermal resistance tends to decrease the thermal conductivity of composites, while at higher content of SiC, the enhancement due to percolative heat conduction paths of the dispersions counteracts the decreasing effect of interfacial thermal resistance on the composite thermal conductivity. And hence, the thermal conductivity of composites decreases and then increases with increasing volume fraction of SiC, which is interesting and not mentioned in previous works$^{1-6,8}$)

4. Conclusion

The effect of interfacial thermal resistance on effective thermal conductivity of MoSi$_2$/SiC composites was investigated. Interfacial thermal resistance between MoSi$_2$ and SiC tends to decrease composite thermal conductivity, but with increasing volume fraction of SiC, the decreasing effect becomes less pronounced due to the enhancement resulted from percolative heat conduction paths.

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