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Journal of the European Ceramic Society 23 (2003) 1773–1778

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Thermal conductivity of Al_2O_3/SiC platelet composites

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Received 5 July 2002; received in revised form 30 October 2002; accepted 8 November 2002

Abstract

The thermal conductivity of hot-pressed Al_2O_3/SiC platelet composites is determined as a function of the platelet content, from 0 to 30 vol.% of SiC. Existing heat conduction models are employed to discuss the experimental data. Data agree with the presence of an interfacial thermal resistance at the Al_2O_3/SiC grain boundaries, which precludes the effect of percolation on the thermal conductivity for the higher percentage of SiC platelets. The observed orientation effect on the thermal conductivity due to an alignment of the platelets is also modelled using the Hasselman's approach. The thermal conductivity of the SiC platelets is calculated from the effective thermal conductivity of the composites.

Keywords: Al_2O_3; Platelets; SiC platelets; Thermal conductivity

1. Introduction

Ceramic matrix composites are attractive candidates for cutting tools and structural applications due to their good thermomechanical properties and high oxidation and thermal shock resistance. The several types of SiC reinforced alumina composites, owing to the wide range in size and shape of the reinforcing inclusions (particles, whiskers and platelets), are within those candidates.[1] The risk to health that the handling of SiC whiskers involves led to the use of platelets as the reinforcing phase, in spite of the slightly worse mechanical performance of these composites when compared to that achieved by the whiskers containing composites.[1–4]

The thermal conductivity, K, is an important property in many applications of alumina ceramics such as high temperature structural components, refractories for the glass and metal manufacture industries, gas radiant burners, wear parts and cutting tools, or microelectronic packaging. Nevertheless, up to date, there are only a few papers devoted to the study of the thermal conductivity of SiC reinforced composites[5–11] and none of them analyses the thermal properties of platelet reinforced composites.

The thermal behaviour of composites containing SiC seems to be strongly dependent on the reinforcement shape and size, as well as on the matrix composition. Only moderate thermal conductivity increases, compared to those expected according to the theoretical value for SiC, have been achieved when SiC was added to alumina,[6,7,9] aluminium[8,11] or lithium aluminosilicate glass-ceramics,[10] even for whisker contents higher than 30 vol.%. Conversely, a dependence of thermal conductivity on the SiC mean particle size[8,11] and SiC whisker diameter[10] has been observed in these composites, in fact, the thermal conductivity increased with the size of the SiC dispersions. The presence of a thermal barrier at the matrix/dispersion boundaries has been proposed to explain both effects, the relatively low thermal conductivity values and its grain size dependence.

Moreover, the thermal conductivity of SiC containing composites is difficult to model because the room temperature thermal conductivity data reported for SiC materials can vary up to 2 orders of magnitude, depending on the impurity content and processing technique.[12–15] A room temperature thermal conductivity in the range of 40–100 W/mK has been estimated for SiC-whiskers using simple models.[5,7,9] Recently, higher values, in the range of 150–325 W/mK[5,8,10,11,16] have been calculated for the intrinsic

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doi:10.1016/S0955-2219(02)00449-1

thermal conductivity of SiC, applying heat conduction models that consider the presence of an interfacial thermal barrier.

In this work, the thermal conductivity of hot-pressed Al_2O_3/SiC platelet composites is evaluated as a function of the platelet content, paying special attention to two effects, the platelet orientation and the interfacial thermal barrier, on heat conduction. By using simple models, the thermal conductivity of the SiC platelets has been estimated from the composite data and compared to published values for SiC.

2. Experimental procedure

Submicronic α-Al_2O_3 powders (CS400, Lonza Martinswerk, Germany) and α-SiC platelets (grade SF, C-Axis technology, Canada) with a 17 μm mean diameter and 3 μm thickness, were used as starting materials. Homogeneous Al_2O_3/SiC mixtures, with SiC platelet contents ranging from 0 to 30 vol.%, were prepared by a flocculation route described in detail in a previous paper.[2] The homogeneous suspensions were stabilized at pH ~10, flocculated at pH =7 and then dried at 120 °C and sieved up to 100 μm.

Fully dense (⩾99% of the theoretical density) Al_2O_3/SiC platelet composites were sintered by hot pressing the homogeneous powder/platelet mixtures in an argon atmosphere. Hot pressing treatments were done using 50 MPa of uniaxial pressure, at 1500 °C/30 min for platelet contents ⩽12 vol.% and 1550 °C/60 min for contents ⩾20 vol.%.

The thermal diffusivity (α) was measured by the laser flash method on square samples of dimensions 8.8 × 8.8 × 1 mm^3, using a commercial equipment (Thermaflash 2200, Holometrix-Micromet Inc. Bedford, USA). The square surfaces of the samples were gold and graphite coated to avoid direct transmission of the laser pulse through the specimen and to improve energy absorption. Measurements were done from room temperature up to 1000 °C always in an argon atmosphere. The software implemented in the equipment is based on Koski's method[17] and considers heat losses and finite pulse corrections. The given values at each temperature are the average of three consecutive measurements.

Thermal diffusivity of each specimen was measured in the hot pressing direction, that is, with the heat flowing in a plane parallel to the hot pressing axis. As the hot pressed composites showed a preferred orientation of the platelets with their basal planes perpendicular to the hot pressing direction,[2] the thermal conductivity of the composites should depend on the testing direction, as was previously established for the wear, mechanical and electrical properties of these composites.[18,19] This orientation effect on thermal diffusivity was checked only for the 30 vol.% platelet composite. Therefore, two specimens were machined with opposite orientations, that is, with the square surface perpendicular and parallel to the hot pressing axis, respectively. As it can be seen in Fig. 1, heat flows in the hot pressing direction in

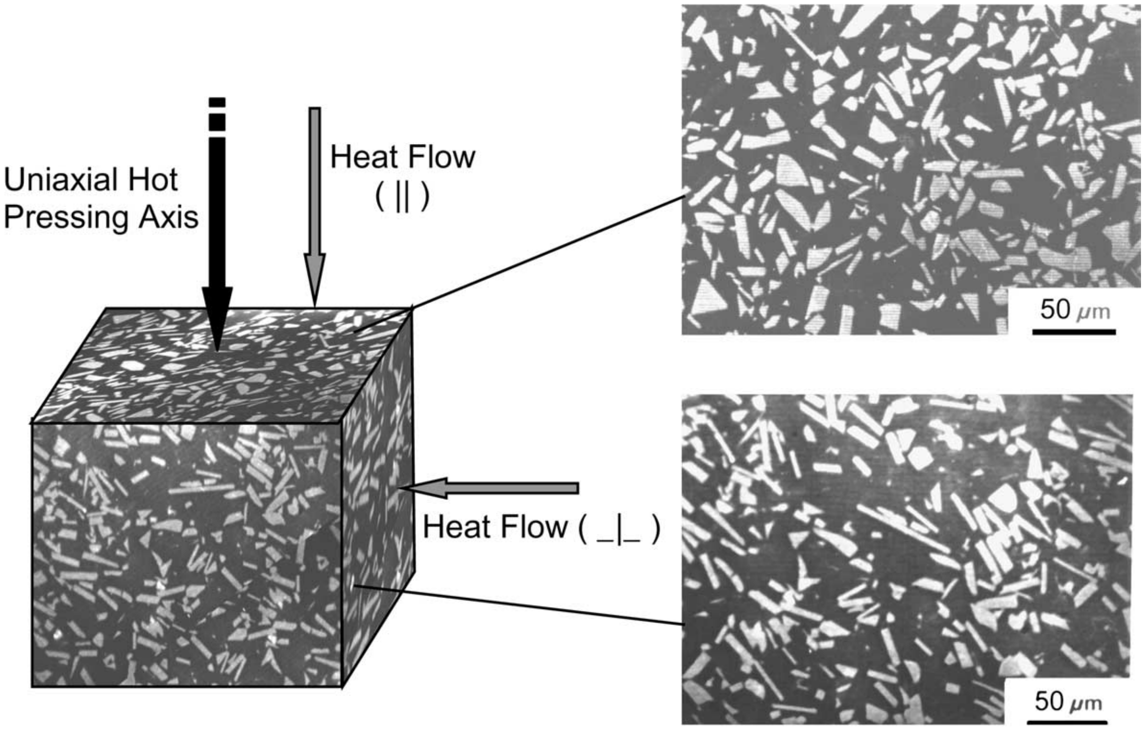

Fig. 1. Scheme of the two specimens used for the thermal diffusivity measurements, having opposite testing orientations, parallel ("‖") and perpendicular ("⊥") to the hot pressing axis.

the former (labelled "∥" specimen) whereas in the second, it flows in the plane perpendicular to the hot pressing direction ("⊥"specimen).

The specific heat of the Al_2O_3/SiC composites was calculated by the Al_2O_3 and SiC weight fractions and the corresponding specific heat data from JANAF tables[20] (see Table 1). With the density (ρ), the thermal diffusivity (α) and the calculated specific heat values (Cp), the thermal conductivity (K) was calculated for each specimen according to the following equation:

$$K = \rho \ \alpha \ Cp \tag{1}$$

3. Results

Fig. 2 depicts the thermal diffusivity versus temperature plots for the Al_2O_3/SiC platelet composites tested on the "∥" configuration. At room temperature, diffusivity increases with SiC platelet volume fraction from 0.092 cm^2/s, for the monolithic alumina, to 0.153 cm^2/s, for the 30 vol.% composite. For each composite, the thermal diffusivity decreases gradually with temperature, reaching at 1000 °C values of 0.015 cm^2/s and 0.028 cm^2/s for 0 and 30 vol.% of SiC_{pl}, respectively. Therefore, the maximum increase in diffusivity due to the platelet addition with respect to the monolithic alumina varied from 66% at room temperature up to 85% at 1000 °C.

The effect of platelet orientation on diffusivity can be seen in Fig. 3. The "⊥" configuration showed higher diffusivity than the "∥" one at all tested temperatures, reaching the maximum value of 0.178 cm^2/s at room temperature.

Fig. 4 shows thermal conductivity values derived from Eq. (1) versus temperature for each composite. These curves show similar trends to those observed for thermal diffusivity plots. A maximum K value of 42 W/mK is reached at room temperature for the 30 vol.% platelet content tested in the "∥" direction, which is 52% higher than that measured for the monolithic alumina. As with thermal diffusivity, the thermal conductivity was higher for heat flowing in the "⊥" than in the "∥" direction, with a room temperature K value of 49 W/mK.

Table 1
Specific heat values used in the specific heat calculations of the different composites. Data were collected from ref. 20

Temperature (°C)	Cp α-SiC (J/ g °K)	Cp α-Al_2O_3 (J/g °K)
25	0.6628	0.7719
100	0.8294	0.9205
200	0.9542	1.0273
300	1.0309	1.0906
400	1.0850	1.1340
500	1.1266	1.1668
600	1.1606	1.1933
700	1.1894	1.2156
800	1.2146	1.2351
900	1.2370	1.2523
1000	1.2571	1.2678

4. Discussion

The addition of SiC platelets to Al_2O_3 clearly enhances its thermal diffusivity and thermal conductivity (Figs. 2–4). Room temperature thermal diffusivity values in the "∥" direction (up to 0.153 cm^2/s for the 30 vol.% platelet composite) are similar to those reported for other SiC whisker reinforced composites,[5,9] which

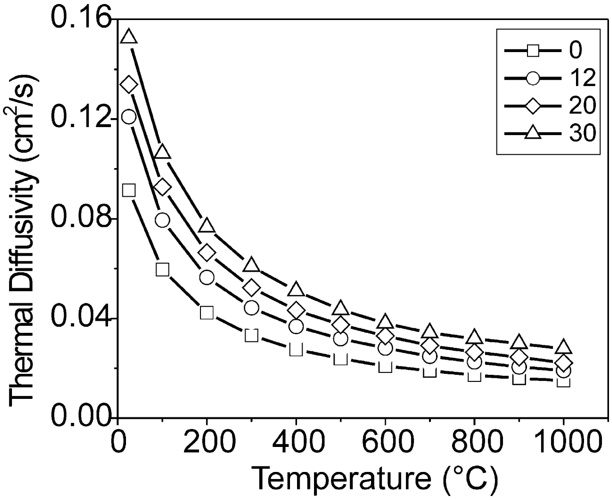

Fig. 2. Thermal diffusivity as a function of temperature for selected Al_2O_3/SiC platelet composites in the "∥" direction. Values for 5 and 8 vol.% composites are not shown as they were very closed to those measured for the 12 vol.% composite. Numbers in the legend indicate SiC platelet content (vol.%).

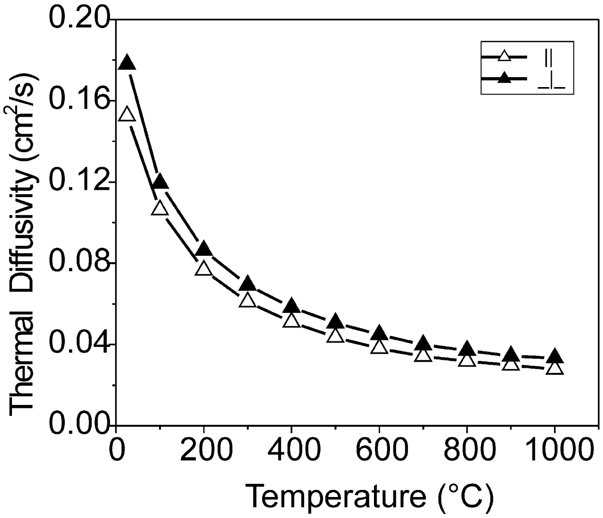

Fig. 3. Thermal diffusivity as a function of temperature for the 30 vol.% SiC platelet composite tested in the parallel ("∥") and perpendicular ("⊥") orientations.

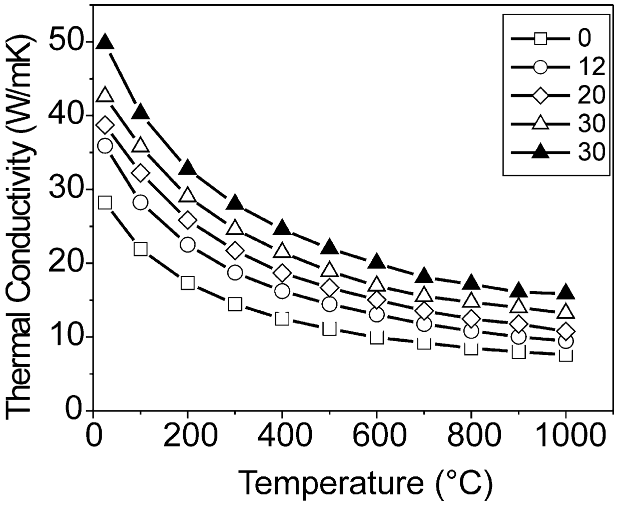

Fig. 4. Thermal conductivity as a function of temperature for the selected Al_2O_3/SiC platelet composites. Values for 5 and 8 vol.% composites are not shown as they were very closed to those measured for the 12 vol.% composite. Legend numbers indicate SiC platelet content (vol.%). Empty and full symbols correspond to the samples tested in the "∥" and "⊥" orientations, respectively.

are in the range of 0.144–0.176 cm^2/s. Room temperature thermal conductivity values are also in agreement with data found in literature for monolithic Al_2O_3 (28–35 W/mK)[5,6,9,21] and Al_2O_3/30 vol.% SiC whisker composites (33–50 W/mK).[5,9]

The maximum thermal conductivity was reached for the 30 vol.% SiC composite at room temperature, for the"⊥" direction (49 W/mK). However, a higher thermal conductivity should be expected if we consider the theoretical value for SiC (~700 W/mK),[12] or even, values measured in single-crystal SiC (490 W/mK)[13] or in pure polycrystalline SiC specimens (270 W/mK).[14] This discrepancy is probably related to the existence of an interfacial thermal barrier as it has been proposed by other authors.[5,8,10,11,16]

The effect of a grain boundary interfacial thermal resistance on the effective thermal conductivity of the SiC platelet containing composites can be analysed using the model of Hasselman and Johnson.[22] By modifying the original theory of Rayleigh and Maxwell, Hasselman and Johnson deduced expressions for the effective thermal conductivity of composites having a continuous matrix phase with dispersed particles of variable geometry (spherical, cylindrical or layered composites), introducing a thermal barrier conductance, "h", at the interface. The expression for spherical dispersions is selected as a first approximation to our composites:

$$\frac{K_{\mathrm{eff}}}{K_{\mathrm{c}}}=\frac{\left[2\left(\nu-\frac{K_{\mathrm{sic}}}{ah}-1\right)\phi+\nu+\frac{2K_{\mathrm{sic}}}{ah}+2\right]}{\left[\left(1-\nu+\frac{K_{\mathrm{sic}}}{ah}\right)\phi+\nu+\frac{2K_{\mathrm{sic}}}{ah}+2\right]} \tag{2}$$

where K_c is the thermal conductivity of the continuous phase (Al_2O_3 in the present case), ν is K_{SiC}/K_C, being K_{SiC} the unknown thermal conductivity of the dispersed phase (SiC), ϕ is the SiC volume fraction, and "a" is the inclusion radius. When $K_{SiC}/ah=0$ this equation agrees with Maxwell expression for the effective conductivity in absence of an interfacial thermal resistance.[23] Fig. 5 illustrates the relative effective thermal conductivity given by Eq. (2) as a function of the SiC volume fraction for a range of K_{SiC}/ah values and for a ν value of 8.7. Experimental thermal conductivity data at different temperatures are also included in this graph. Although the porosity of the samples was very low (<1%), its effect was corrected in this figure using Klemens' equation.[24]

The ν value of 8.7, used in Eq. (2) to get the theoretical curves of Fig. 5, is the average of the K_{SiC}/K_c ratios for all the tested temperatures, where K_c is the experimental data for the monolithic Al_2O_3 material and, as it will be described below, K_{SiC} is calculated by fitting the effective thermal conductivity of the Al_2O_3/SiC composites with 8 and 12 vol.% of SiC to the Maxwell's expression.

As it can be seen in Fig. 5, the relative effective thermal conductivity (K_{eff}/K_c) does not depend on temperature for the composites with SiC platelet contents $\leqslant$12 vol.%. For these composites, experimental data agree with the model of Maxwell ($K_{SiC}/ah=0$) with no apparent contribution of interfacial thermal barriers. Conversely, for higher SiC contents, K_{eff}/K_c shows notable lower values at room temperature than those predicted by Maxwell's model, which evidences the lack of a percolation effect. These data show a good fit to the model of Hasselman with K_{SiC}/ah values ranging from 2 to 0.5, depending on temperature and SiC contents. As K_{SiC}/ah decreases as

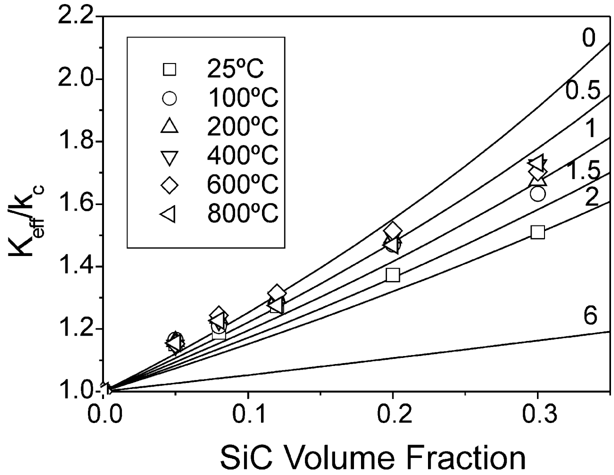

Fig. 5. Thermal conductivity data at selected temperatures as a function of the SiC volume fraction for all the Al_2O_3/SiC composites tested in the "∥" orientation. Continuous lines correspond to the relative effective thermal conductivity calculated from the Hasselman's equation for a range of K_{SiC}/ah values.

the temperature increases, it would agree with a thermal barrier at the Al_2O_3/SiC grain boundaries associated with the development of high thermal stresses in the composites with high SiC contents.

As was mentioned above, the intrinsic thermal conductivity of SiC platelets was estimated from the effective thermal conductivity of the Al_2O_3/SiC composites applying Maxwell's model,[23] which is one of the simplest that often shows good results for dilute media. Therefore, it can be considered to be valid for SiC contents below 15 vol.%.[25] We have tried to apply it for the 5, 8 and 12 vol.% containing composites, in the range of tested temperatures. However, the application of Maxwell's expression to the 5 vol.% composite led to large scattering in the values of SiC thermal conductivity because of the small conductivity increases compared to the Al_2O_3 matrix. Accordingly, we have applied it for the 8 and 12 vol.% composites, obtaining a room temperature value of 244 W/mK, that is similar to that calculated for SiC particles by other authors.[8,11] Averaging the thermal conductivity values deduced from the model of Maxwell at each temperature, the following fit for the intrinsic thermal conductivity of SiC platelet as a function of temperature [K] was deduced:

$$K_{\mathrm{SiC}} = \frac{1}{0.0011 + 10^{-5}T} \quad (3)$$

which is very close to the temperature dependence reported for SiC.[7]

The presence of a grain boundary interfacial thermal resistance can also explain the increase in K observed for the 30 vol.% sample tested on the *"⊥"* orientation, although the anisotropy of the crystalline structure of SiC might also influence it. The SiC platelets are hexagonal single-crystals, with C and Si atoms tetrahedrically coordinated, the interatomic distance being three times longer along the C-axis than in the basal plane, which is formed by three similar atoms. Therefore, it might show anisotropic properties as it has been observed in other properties related to phonon vibration, such as the elastic constants and the thermal expansion coefficient.[26,27] Accordingly, an anisotropy in the thermal conductivity of hot-pressed Al_2O_3/SiC_{pl} materials could also be expected as the phonon mean free path will be higher in the basal plane, which corresponds to the platelet facet, improving heat conduction in the *"⊥"* direction.

Observing the microstructures for both specimen orientations (Fig. 1), it can be inferred that the value of "a" in Eq. (2) will depend on the orientation. For the parallel orientation, "a" should be the platelet half-thickness, whereas for the perpendicular direction it should correspond to the half-diagonal of the platelet, 1.5 and 8.5 μm, respectively.[28] Considering the K_{eff} as the experimental data for the 20 and the two 30 vol.% composites (in both orientations), the K_c as the value measured for the monolithic Al_2O_3 sample and the intrinsic K_{SiC} as that previously estimated [Eq. (3)], the interfacial thermal conductance was deduced for each temperature. Values for *"h"* were almost constant with temperature and the room temperature value, averaged from the data obtained for 20 and 30 vol.% (parallel and perpendicular), was 1.8×10^8 W/m^{2}K which is within the range given by Collin and Rowcliffe[5] for Al_2O_3/SiC composites (0.8×10^8–9.5×10^8 W/m^{2}K).

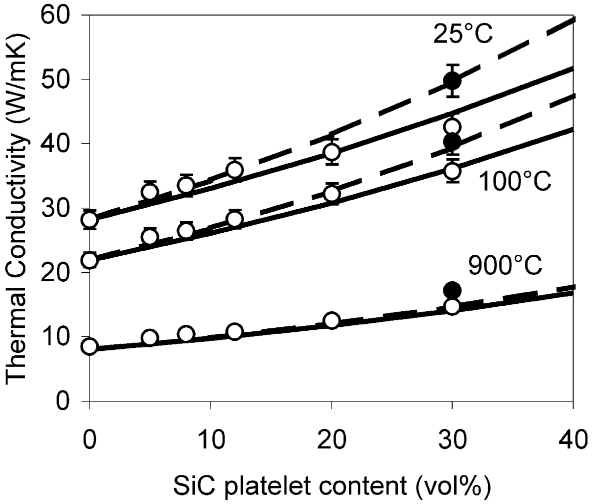

Fig. 6. Thermal conductivity at three selected temperatures, as a function of the SiC volume content, for all the Al_2O_3/SiC composites. Points are the experimental data, empty for parallel and full for perpendicular orientation. Lines correspond to the effective conductivity obtained from the Hasselman's equation, continuos for the "‖" direction (a = 1.5 μm), and dashed for the "⊥" orientation (a=8.5 μm).

The plot of Fig. 6 has been done introducing the proposed values for K_{SiC}, h and a in the equation of Hasselman [Eq. (2)]. As it can be seen, experimental data, which are also plotted in the graph, agree well with the model proving that the presence of an interfacial thermal barrier explains conveniently heat conduction in Al_2O_3/SiC materials. Data for the 30 vol.% specimen tested in the perpendicular orientation are also explained by this model because the contribution of the thermal barrier is lower due to the higher value of "a" for this orientation. The effect of the thermal resistance at the Al_2O_3/SiC_{pl} interface will diminish with temperature as thermal stresses disappear and the phonon-phonon scattering becomes the limiting factor for the phonon mean free path. This agrees with the experimental evidence that the difference in thermal conductivity for the perpendicular and parallel orientations decreases as temperature raises.

5. Conclusions

The addition of SiC platelets enhanced the thermal diffusivity and thermal conductivity of Al_2O_3. The intrinsic thermal conductivity of SiC platelets as a function of temperature was calculated from the effective thermal conductivity of the Al_2O_3/SiC composites having low SiC contents, using the Maxwell's model. The experimental data agreed with the presence of an interfacial thermal conductance at the Al_2O_3/SiC grain boundaries, which was estimated using the Hasselman's model as 1.8 10^8 W/m^2K at room temperature. This interfacial thermal barrier precluded the effect of percolation on the thermal conductivity for the higher percentage of SiC platelets. An orientation effect on the conductivity due to an alignment of the platelets was experimentally observed and also modelled with the Hasselman's approach.

Acknowledgements

This work was supported by Mcyt (ES), project 2FD97-0345-C02-01, and CAM (ES), Project 07N/0030/2001.

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