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Thermal conductivity of AlN/polystyrene interpenetrating networks

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Abstract

A new processing technique is proposed for preparing AlN/polystyrene interpenetrating network composites. This technique is based on an infiltration procedure, partly conducted in vacuo, of a liquid mixture of monomer and initiator into a porous A1N ceramic body with a percolated pore structure. Successive in situ polymerization is produced by heating up the infiltrated ceramic at ≈100°C under ambient pressure. The final morphology of the composite consists of an interpenetrating polymer network which fills in an A1N ceramic skeleton. This new infiltration procedure enabled us to prepare continuous polymeric networks whose volume fraction lies between ≈12 and 40 vol%. These fractions of polymer are consistently lower than that usually involved in traditional polymer moulding processes. It is shown both by experiments and theory that these special interpenetrating network microstructures experience relatively high thermal conductivity. Concurrently, significantly improved fracture characteristics and reliability can be achieved as compared with both that of monolithic ceramics and traditional polymeric materials containing high fractions of ceramic filler. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: AlN; Microstructure-final; Polymeric filler; Porosity; Thermal conductivity

1. Introduction

Polymers are widely employed for both their excellent thermal insulating properties and fracture resistance. For many applications, however, it is desirable to have a plastic part which can conduct heat, either to reduce hot spots, or to act as a heat sink for a thermally sensitive component. In some applications, it may be advantageous to combine light weight, chemical inertness and reliability against fracture of many plastics to uses which require high degrees of heat transfer. For example, current requirements on the thermal management of electronic packages have prompted the search of a thermally conductive material which provides a solution for high-reliability packages with a relatively long lifetime. In plastic encapsulated electronic devices, thermal problems have traditionally been moderated by the use of embedded heat sinks. However, these packages may not guarantee enough heat transfer and tend to be susceptible to thermal cracking.[1]

Currently, most package devices are moulded with a composition of 30–45 vol% polymeric resin and 55–70 vol% filler. Fused SiO_2 filler is most commonly blended into the polymer matrix in order to improve its thermal property. However, the thermal conductivity of neat polymeric resins is typically as low as ≈0.2 W/m K,[1] while fused silica experiences a value of 1.4 W/m K.[2] Thus, given the relatively low mismatch in thermal conductivity between the polymer matrix and the SiO_2 filler, the maximum thermal conductivity of polymer/SiO_2 composites usually achieve a value only ≈6–7 times higher than that of the neat polymer matrix.[3] Replacement of fused silica with higher thermal conductivity fillers has been attempted to maximize the thermal dissipation effect, while maintaining all other properties of the encapsulant material (including the fracture properties) (at least) unchanged. Being metal phases to be excluded a priori because of their high electrical conductivity, several ceramic materials such as aluminum nitride (AlN), boron nitride (BN), silicon carbide (SiC) and beryllium oxide (BeO) have been attentioned, either as potential fillers [4] or as bulk matrix materials.[5,6]

All the above mentioned ceramic phases are of the adamantine structure and possess the needed high intrinsic thermal conductivity characteristics. In particular, AlN is a non-toxic phase, readily available as high-purity powder, and with good thermal conductivity (i.e.

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150–300 W/m K).[7] On the one hand, however, bulk AlN ceramics are typically brittle and, thus, may lack the needed reliability. Reliability against fracture should be ensured by a continuous distribution of a tough polymeric phase within the composite, which it is very difficult to maintain by conventional processing procedures[8] at filler contents approaching and exceeding ≈60 vol%. In addition, the viscosity of the mixture of AlN and uncured resins rises rather rapidly above AlN volume fractions >50%, making the molding process difficult to perform. Given this compositional limitation, polymeric materials added with AlN powder, despite the high mismatch of thermal properties between matrix and filler, can experience thermal conductivity values at most ≈10 times higher than that of the neat polymeric matrix.[3]

In this paper, we report about a new processing method for preparing ceramic/polymer composites with both high thermal conductivity and improved reliability characteristics against fracture. This processing method is based on an infiltration procedure partly conducted in vacuo of both a monomer and an initiator into a porous AlN ceramic body with a percolated pore structure. After bringing the specimen back to ambient pressure, in situ polymerization occurs in the porous ceramic upon heating up, and a morphology is reached of a polymer/ceramic composite consisting of an interpenetrating polymer network completely filling the AlN ceramic skeleton. An important feature of this process is that a continuous polymeric polystyrene network can be achieved even at volume fractions as low as ≈18 vol%. Thus, the content of low thermal conductivity polymer can be minimized. achieving fractions consistently lower than that produced by traditional polymer molding processes. It is shown that this special microstructural arrangement of ceramic/polymer interpenetrating networks enables to optimize both thermal conductivity and fracture characteristics.

2. Experimental procedure

2.1. Material preparation

AlN powder (Type F, Tokuyama Corp., Ltd., Tokyo, Japan) was used, whose average grain size and oxygen content were ≈0.25 μm and 0.85 wt%, respectively. No additional sintering additives or external phases were added. The powder was preformed by uniaxial pressing (under ≈30 MPa) in the shape of small disks with diameter and thickness of 20 and 5 mm, respectively. Then, a cold isostatic pressing (CIP) cycle (under ≈200 MPa) was applied for obtaining further compaction up to a relative density ≈55%. After a pre-firing in vacuo of 2 h at 1200°C, the compacted disks were encapsulated into evacuated pyrex tubes and hot isostatically pressed (HIPed) in the interval of temperature between 1500 and 2000°C. The maximum HIPing pressure was ≈180 MPa for all the fired AlN materials reported in this study. The HIPing process enabled us to obtain pure polycrystalline AlN bodies with different densities. Density of the AlN HIPed bodies, measured by the Archimedes' method, were in the range between ≈65 and >99.5%.

Porous AlN polycrystalline bodies with relative densities in the range 65–92% were infiltrated by a mixture of monomer (styrene/divinylbenzene) and initiator (benzoyl peroxide), using the newly developed infiltration device shown in Fig. 1. As seen, the device consisted of three interconnected glass containers evacuated to 10^{-5} Pa, in which the monomer, the initiator and the porous ceramic specimen were respectively placed. Sodium sulphate powder was also introduced in the container together with the porous AlN specimen, in order to avoid large depositions of polymer on the specimen surface. According to a two-step process of "freezing and thawing" (cf. the liquid nitrogen containers in Fig. 1), a liquid mixture of monomer and initiator was obtained and a liquid state infiltration into the porous ceramic body was performed upon gradually releasing vacuum upon Ar gas addition until reaching ambient pressure. Then, in situ polymerization to polystyrene spontaneously occurred within the percolated porosity of the AlN specimen upon heating up at 100°C for ≈14 h. AlN materials with percolated porosity in the range between 8 and 35 vol% were infiltrated by polystyrene and tested with respect to both their fracture behavior and thermal conductivity. The processing parameters of the above described infiltration process were kept constant in this preliminar study, and only the HIPing temperature adopted for preparing the AlN ceramic skeleton was varied. For comparison, also an unfired AlN specimen, just preformed by CIP (density ≈56%), was infiltrated by polystyrene and tested. For simplicity, this specimen will be referred to as "green"

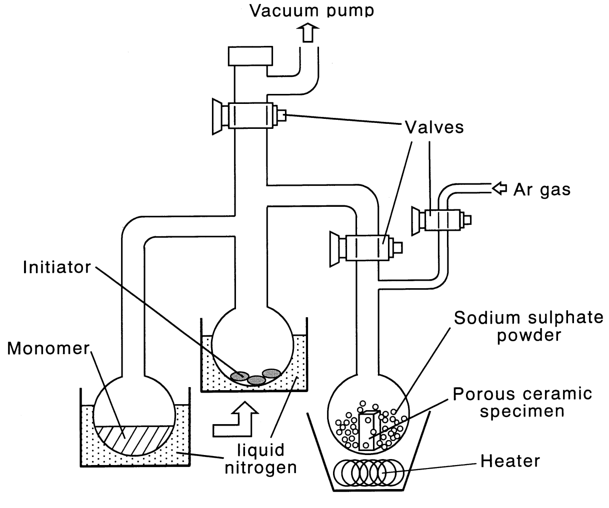

Fig. 1. Schematic of the experimental device used for polymer infiltration into the porous ceramic body.

infiltrated specimen, henceforth. It should be noted that, irrespective of the density of the AlN skeleton, in all the composites prepared in this study infiltration could be completed up to the center of the specimens without visible inhomogeneities in the network structure.

2.2. Material characterization

Microstructures of the interpenetrated network composites were analysed by a field-emission SEM (S-800, Hitachi Ltd., Tokyo, Japan) operating at 10 kV. Image analysis characterizations were performed on SEM micrographs to determine both the grain size of the AlN ceramic skeleton and the precise volume fraction of the polystyrene phase (i.e. the porosity of the AlN skeleton). AlN grain size and polystyrene volume fraction were measured according to the circle-interception and the systematic-point counting methods, respectively, as proposed by Hilliard and Cahn.[9,10] The surface area quantitatively scanned by image analysis was $\sim$0.1 mm^2 per each material investigated.

Specimens for fracture mechanics tests were parallelepipeds 3×4×20 mm cut from the respective AlN polycrystalline or composite bodies. In order to achieve a stable crack propagation, a straight-through notch with a relative length, $a/W \approx 0.5$ was introduced at the center of the composite bars by a diamond cutter (blade thickness of 0.2 mm). To reduce the influence on crack initiation of the finite notch-tip radius, the bottom part of the saw-notch was sharpened with a razor blade sprinkled with fine diamond paste. Stable fracture propagation (from the notch root) in bending flexure geometry was achieved by loading the specimens in a three-point jig (span of 16 mm), using a specially designed crack stabilizer.[11] The load relation was directly measured with semiconductor strain gauges placed on the loading bar and on the tensile surface of the specimen, respectively. Work of fracture of the composites was calculated as the ratio between the area under the load-displacement curve and twice the fractured surface area.

Thermal conductivity was measured according to the laser-flash method, using a 5J CO$_2$ laser. The measurement device was supplied with a non-contact infrared detector and a computer-controlled data acquisition system. All the thermal conductivity experiments were performed at room temperature. Data were averaged on three measurements per each investigated material.

3. Results and discussion

3.1. Microstructure and fracture behavior

The dependences of relative density and grain size of the AlN porous bodies on the HIPing temperature are shown in Figs. 2A and B, respectively. The AlN grain size experienced values in the range 0.8–9.5 μm, the higher the HIPing temperature (and relative density) the larger the grain size. The AlN ceramics HIPed at temperatures $\geqslant$1900°C were nearly fully dense. Thus, they were not infiltrated by polystyrene and were only tested for comparison.

Results of the fracture mechanics characterization are shown in Fig. 3. Load-displacement curves are shown, which were recorded during stable fracture propagation in two polystyrene infiltrated materials, in comparison with that of fully dense AlN (HIPed at 1900°C). An interesting trend is noticed in these curves. The lower limit for toughness is represented by the specimen infiltrated with a large percentage of polymer ($\approx$40 vol%). This material is extremely brittle and even liable to dimensional instability upon handling. Its consistency is similar to that of common chalks. This poor fracture behavior is certainly due to a lack of bonding both

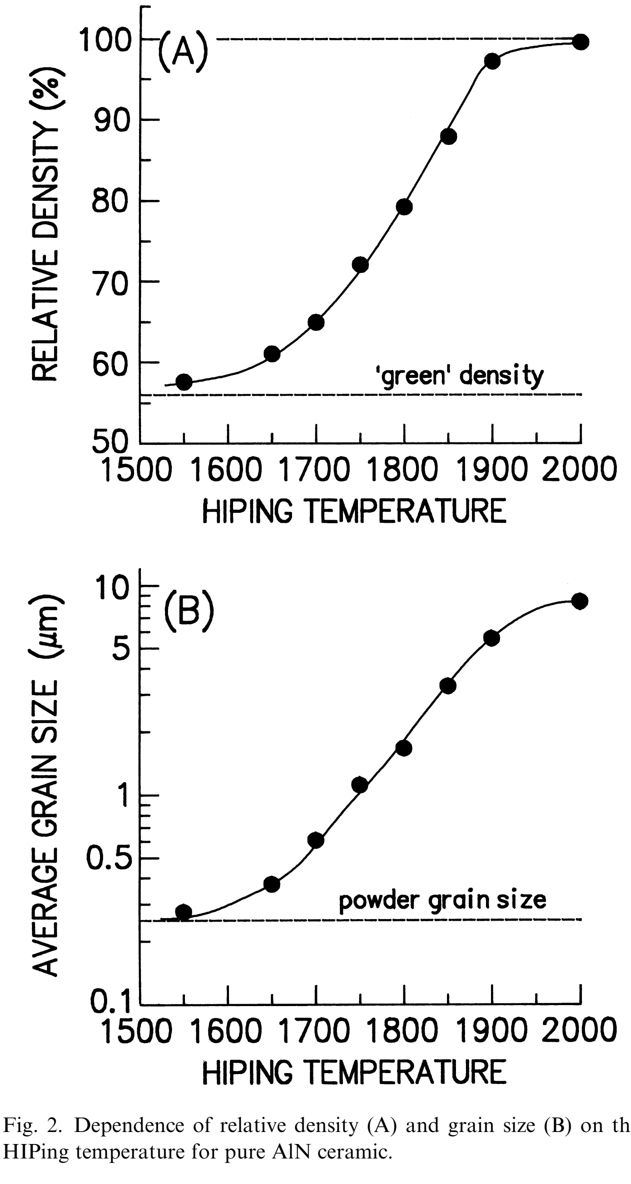

Fig. 2. Dependence of relative density (A) and grain size (B) on the HIPing temperature for pure AlN ceramic.

between the polymer and ceramic phase, and among the AlN grains constituting the polycrystalline ceramic skeleton. In the ceramic phase, large (and percolated) cracked regions are formed and the composite fails, irrespective of the presence of a large fraction of polymer. The fracture behavior of the AlN/polystyrene composite improved with increasing the HIPing temperature selected for preparing the AlN porous body, until an optimum fracture behavior was achieved upon polystyrene infiltration of the specimen HIPed at 1750°C. The load-displacement curve of this specimen is also shown in Fig. 3. It is thought that, upon a HIPing cycle which can densify the ceramic skeleton, a higher bond strength can be achieved among the AlN grains of the polycrystalline network. Porous AlN bodies before infiltration were also tested for comparison. However, none of these materials showed stable fracture. In addition, the porous AlN bodies were typically brittle, displaying toughness values, $K_{IC} < 1.6$ MPa×m$^{1/2}$. This proves that the high toughness found in the composite arises from a synergistic effect between the ceramic and polymer phases. Maximum toughness in the composite is achieved despite only a minor fraction of polystyrene polymer being infiltrated. For HIPing temperatures >1750°C (i.e. polymer fractions <28 vol%) the toughness of the composite started decreasing until approaching that of the dense monolithic AlN HIPed at 1900°C (shown in Fig. 3).

The results of the fracture mechanics characterization are summarized in the inset of Fig. 3, which shows the work of fracture of the composite as a function of the volume fraction of the polystyrene phase infiltrated into the AlN ceramic skeleton. The presence of a maximum toughness can be explained by considering the fracture data in comparison with SEM observation of the fracture surfaces. Fig. 4a, b, and c show the fracture surface of AlN/polystyrene specimens containing polymer fractions ≈12, ≈28, and ≈40 vol%, respectively. From a microstructural point of view, it is very interesting to notice how the polystyrene polymeric network could even form within the narrow ceramic porosity of an ≈88% dense AlN (Fig. 4a). The polymer network appears to be fully percolated, like as it formed a polymeric grain boundary with no visible porosity and whose average thickness experienced an average value ~350 nm. However, despite the uniqueness of this interpenetrated network microstructure, its fracture behavior was rather brittle (cf. Fig. 3). The fracture surface appears to be relatively flat and no stretched polymer ligaments are obviously found.

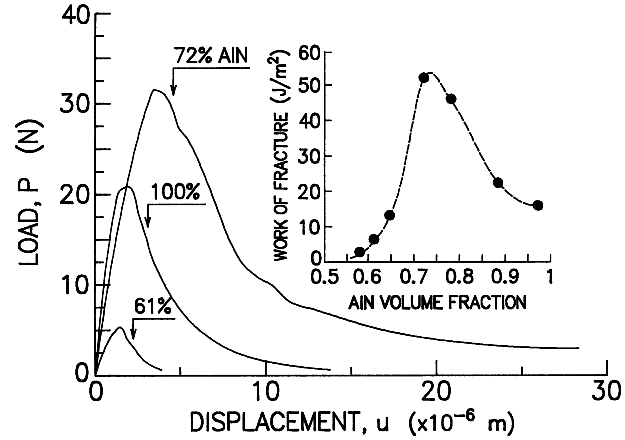

Fig. 3. Load-displacement curves for two (61 and 72 vol% AlN) polystyrene-infiltrated ceramics are compared with that of a fully dense AlN ceramic (100% AlN) HIPed at 1900°C. In the inset, a plot of the work of fracture, calculated from the respective load-displacement curves, is given as a function of the AlN content for all the compositions investigated. Work of fracture values were averaged among three point data per each material.

The composite showed in the SEM micrograph of Fig. 4b is the one which experienced optimum toughness (polystyrene fraction ≈28 vol%). Such a good fracture behavior is clearly explainable from inspection of the fracture surface. Highly stretched polymer ligaments are clearly envisaged, which should counteract brittle fracture propagation. These polymer ligaments form plastic bridging sites whose stretching may adsorb a large amount of energy during fracture propagation in the ceramic skeleton, thus explaining the relatively high work of the fracture value measured in this composite. An interesting feature in the SEM micrograph is that the polystyrene polymer appears to be strongly bonded to the ceramic AlN grains and, upon stretching, it shows ramifications and traces of local crazing (cf. arrowed regions in Fig. 4b). This implies that the strength of the polymer phase is fully exploited in the stretched bridging sites.

Fig. 4c shows the fracture surface of a composite containing a larger fraction of polystyrene (~40 vol%). Although the polymer interpenetrating network is clearly visible, no significant stretching of the network can be noticed after fracture. In other words, a clear change in the fracture mode occurred. Some agglomeration of the AlN grains are observed in which bonding among the ceramic crystallites is achieved (cf. arrowed regions in Fig. 4c). However, fracture seems rather to occur by brittle pulling out of the ceramic grains (or grain agglomerates) from the polymer network than by plastic stretching of the network itself. This fracture behavior may be similar to that observed in some polymer/ceramic composites with similar composition, but prepared by conventional methods. An important consideration lies behind the fracture mode observed in the composite containing a large fraction of polystyrene. To ensure that the grains are strongly bonded even at high fraction of porosity, porous bodies should be obtained under a higher sintering temperature, i.e. using pore formers to tailor the density. This may be a promising directive for future research using the present infiltration technique.

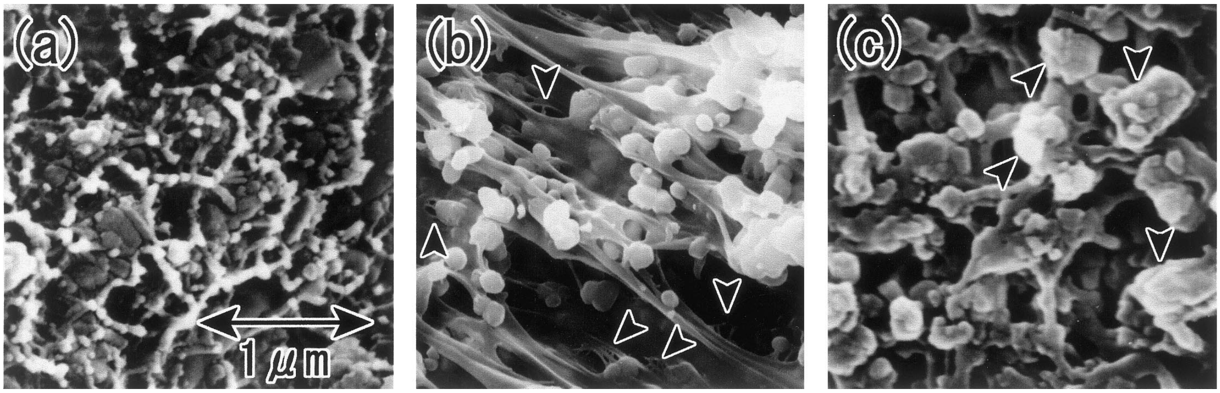

Fig. 4. Fracture surfaces of AlN/polystyrene composites containing polymer fractions ≈12 vol% (a), ≈28 vol% (b), and ≈40 vol% (c). Arrows in (b) indicate polymer ligaments heavily stretched during fracture. Arrows in (c) locates hard AlN agglomerated after partial densification by HIPing.

SEM observation may suggest that, besides the bonding strength between the ceramic grains (governed by the HIPing temperature selected for preparing the ceramic skeleton) and the polymer volume fraction, also the bonding strength between the ceramic and the polymer phase plays an important role in the fracture behavior of the composite. In other words, the present results suggest that an optimum size exists for the thickness of the percolated polystyrene network, which enhances the ceramic/polymer bond. Unfortunately, the present preliminary results do not allow to check whether this hypothesis is correct or not, and further studies are presently in progress to clarify this important point.

From the point of view of the reliability against fracture, it should be noted that, for applications mainly focused on thermal behavior, the maximum load requirements are usually not very severe. From the maxima of the load-displacement curves a fracture toughness value, K_{IC}, in the range between 3 and 5 MPa×m$^{1/2}$ can be calculated for the composites containing polystyrene fractions in the range 8–30 vol%. These K_{IC} values are similar to that of dense monolithic ceramics, however, the significant result in the present fracture characterization is that, in the composite containing ≈28 vol% polystyrene, brittle (i.e. catastrophic) fracture is suppressed. In other words, although crack initiation may occur in the material under relatively low stress level, crack propagation requires large energy dissipation and the crack eventually arrests, thus ensuring the long-term integrity of the component.

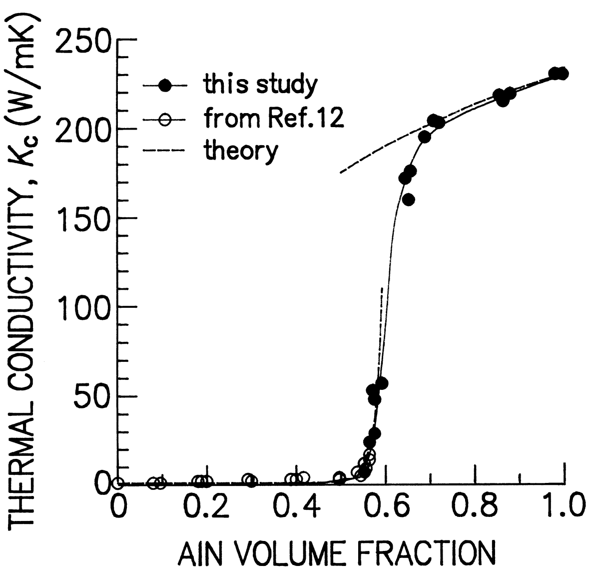

Fig. 5. Thermal conductivity of the AlN/polystyrene composite as a function of the AlN fraction. Literature data collected on materials with higher polymer contents are shown for comparison. One data point represents one tested specimen. Dotted curves correspond to theoretical predictions according to Eqs. (1)–(3).

3.2. Thermal conductivity behavior

The thermal conductivity at room temperature of the AlN/polystyrene composite samples investigated in this study is shown in Fig. 5 as a function of the composite composition. The measured thermal conductivities increased monotonically as the volume of AlN increased. However, the increase was steeper in the range of AlN volume fractions between 55 and 70%, while saturation of the thermal conductivity values could be observed at higher fractions of AlN. For comparison, data collected on polymer/AlN composites,[12,13] prepared by conventional moulding method, are also plotted in Fig. 5. As seen, the range of composite composition which can be achieved by conventional preparation method is somewhat complementary to that achievable by the present infiltration method, a narrow overlapping region being possible in the interval 50–60 vol% of AlN. Thermal conductivity data collected in this overlapping region for the composites prepared by polymer infiltration are consistent with the literature data on composites by polymer moulding method. However, according to the steep increase starting from intermediate fractions, rather high thermal conductivities can be found in composites with composition in the interval $\geqslant 70$ vol% AlN. A theoretical interpretation of the composite data over the entire compositional range is attempted hereafter.

A model by Nielsen[14] is followed which is of relatively simple formulation and applies over a wide range of materials. According to Nielsen, the thermal conductivity of a composite material, K_c, is related to that of the matrix, K_m, and the filler, K_f, according to the following equation:

$$K_c = K_m[(1 + ABV_f)/|1 - B\beta V_f|] \tag{1}$$

where the parameters B and β are given by:

$$B = [(K_f/K_m - 1)/(K_f/K_m + A)] \tag{2}$$

$$\beta + 1 = [(1 - V_M)/V_M^2]V_f \tag{3}$$

in which V_f is the volume fraction of the filler phase, V_M is the maximum packing fraction of the filler phase, and A is an adimensional shape factor which depends on the geometry of the filler particles. A is a parameter related to the generalized Einstein coefficient, published for a number of solid and liquid suspensions with various characteristics.[14] V_M has been given as ≈ 0.6 for irregular random-close particles and 0.9 for elongated unidirectional networks.[14] The A value has been given ≈ 2 and 0.5 for facetted particles and percolated networks of acicular particles, respectively.[14,15] For $A \to \infty$ and $V_M = 1$, Eq. (1) simply reduces to the rule of mixture. Predictions based on the Nielsen's model for the thermal conductivity of the AlN/polystyrene composite are also shown in Fig. 5. In the calculation, the thermal conductivity of AlN is assumed to be ≈ 230 W/m K, according to previous studies on AlN polycrystalline ceramics.[7,16] The thermal conductivity of the bulk polystyrene polymer is taken as ≈ 0.18 W/m K.[1,14] First, prediction for the polymer-rich side of the composite composition can be obtained by considering the polymer and the ceramic phases as the matrix and the filler, respectively. In this case, the appropriate value of the filler-morphology dependent parameter A is 2 and a single curve can be found from Eqs. (1)–(3), which fits both the literature and our present experimental data, when V_M is taken ≈ 0.43. Prediction can also be obtained for the ceramic-reach compositions, this time taking AlN as the matrix phase. Again according to Eqs. (1)–(3) and considering the appropriate values $A = 0.5$ and $V_M = 0.9$, a mildly decreasing thermal conductivity curve is found as the polystyrene volume fraction increases. This curve fits with good approximation the experimental data up to a polymer fraction ≈ 30 vol%.

In the present context, it is important to notice how experiments and theory agree in showing a steep increase in the thermal conductivity of the composite at intermediate polymer fractions (i.e. in the interval 30–45 vol%). A theoretical model which explains more in detail the percolation-like behavior of the thermal conductivity of AlN in presence of an intergranular secondary phase, is given in a separate paper.[17] The properties of the present hybrid composites could not be so far explored due to technological problems mainly related to composite processing. These problems can now be avoided according to the newly proposed polymer infiltration method.

4. Conclusion

A new infiltration process for producing ceramic/polymer composites with interpenetrating network microstructure is presented. The technique is applied to an AlN/polystyrene material.

Porous AlN ceramics are first prepared with relative densities in the range 55–88%. Then, infiltration in vacuo of monomer and initiator is performed in the liquid state into the porous ceramic body upon bringing back the specimen to ambient pressure. Successive in situ polymerization to polystyrene can occur upon annealing the specimen in argon gas at ≈ 100°C for 14 h. From a microstructural point of view, a well percolated polymer network could be introduced into the ceramic skeleton even at polystyrene fractions as low as ≈ 12 vol%.

The fracture behavior of the composites was investigated and an optimum composition for toughness was found at a polymer fraction ≈ 28 vol%, due to stretching of polymer ligaments in the crack wake. The thermal conductivity of the interpenetrated network composites monotonically increased first with increasing the AlN volume fraction, then it saturated at high fractions of the ceramic phase. A theoretical model by Nielsen was invoked to explain the thermal conductivity experimental data. It was shown that high thermal conductivity can be obtained (concurrently with relatively high toughness) in composites with polymer fractions in the range ≈ 20–30 vol%. The suitable combination of thermal and mechanical properties is regarded as being peculiar of the ceramic/polymer interpenetrated network structures. Although more detailed studies are required for a complete understanding of this new class of materials, the polymer infiltration of porous ceramics appear to be a promising technique to obtain microstructures which are not available by traditional moulding or sintering processes.

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