Investigation of Enhanced Thermal Conductivity in Hexagonal Boron Nitride-Filled Polyimide Films: Effects of Size, Aggregation, and Orientation of Filler Particles

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A series of inorganic/organic composite films exhibiting high thermal conductivity and thermal stability was prepared from platelet-shaped hexagonal boron nitride (hBN) ceramic particles and polyimide (PI), a super engineering plastic widely used in electrical and aerospace industries. Thermal conductivity of hBN/PI films along the in-plane and out-of-plane directions was measured and analyzed in terms of size, shape, volume loading and orientation of hBN particles. hBN platelets of micrometer-scale diameters with different agglomeration states were dispersed in polyamic acid (PAA), a solution-processable precursor of PI. Each hBN/PI film containing 10 to 60 vol% hBN was prepared by spin-coating of the corresponding hBN/PAA slurry on a Si substrate, followed by thermal curing up to 350°C. Out-of-plane and in-plane thermal diffusivity measurements were carried out by the temperature wave analysis (TWA) method and the laser-heating angstrom method, respectively. The degree of orientation of hBN particles was estimated by cross sectional images of scanning electron microscopy (SEM) and transmission wide-angle X-ray diffraction (WAXD) patterns. All films retained high mechanical strength even at a hBN loading of 60 vol%, and the highest values of thermal conductivity achieved in this study were 5.4 W/mK for the out-of-plane direction and 17.6 W/mK for the in-plane direction. hBN/PI films filled with large and high-aspect-ratio particles exhibited large in-plane and out-of-plane anisotropy in thermal conductivity due to the strong in-plane alignment of highly thermally conductive basal (100) plane of hBN particles, while smaller anisotropy was observed for the films filled with small particles and aggregates, which are less likely to align in the in-plane direction during film processing. The anisotropic thermal conductivity observed in the hBN/PI films exhibited strong correlation with the orientation of hBN particles estimated using SEM image analyses and WAXD.