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Enhanced Thermal Conductivity Based on Vertical Double Percolation Morphology in Phase Separated Polyimide Blend Films

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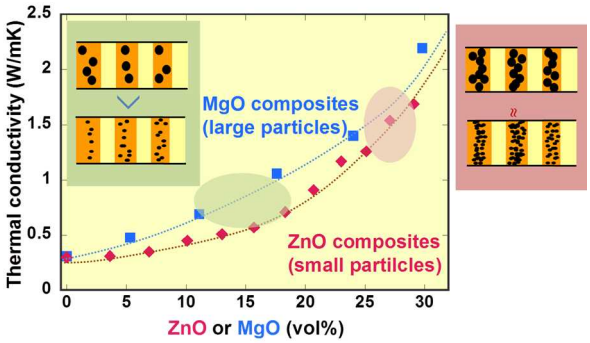
Introduction: Functional composite materials consisting of μm-sized ceramic particles, such as Al_2O_3, AlN, hexagonal boron nitride (h-BN), and thermally stable polymers have been demanded for enhanced thermal conductivity and electric resistivity in thermal interface materials and insulating layers for power-ICs. However, technological achievement is still unsatisfactory due to the large interfacial contact resistance between particles and polymer matrix. In this study, we report a significant enhancement in thermal conductivity even at a low particle loading based on a selective confinement technique of ZnO and MgO particles in micro-phase separated polyimide/polyimide (PI/PI) blend films [1–4].

Methods: Two types of PIs, sulfur- and fluorine-containing PIs, were blended with pyramidal ZnO (ϕ =0.3–0.5 μm), needle-shape ZnO (length=1.5 μm, ϕ =0.15 μm), and cubic MgO (2–3 μm) particles. Since the thermal conductivities of ZnO and MgO crystallites are nearly the same (55 W/m K), 'filler size effect' can be primarily examined. Under the optimized spin-coating and thermal curing conditions, micro-phase separated structures with "vertical double percolation (VDP)" morphology were spontaneously formed in the blend films, in which two phases are separately aligned along the out-of-plane direction, and both of ZnO and MgO particles were selectively confined in the fluorine-containing PI phase.

Results & Discussion: PI blend films with VDP morphology exhibited more than 400 % enhancement of thermal conductivity in the out-of-plane direction at 24 vol% content (Φ) of ZnO and MgO particles, whereas monophase PI films containing homogeneously dispersed particles exhibited only 90–150 % enhancement compared with a pristine PI film. These results indicate that the VDP morphology with selectively confined particles functions as an effective thermal conductive pathway which goes over the percolation threshold. Moreover, PI blend films with large-sized MgO particles exhibited higher conductivity between Φ=5 and 24 vol % than those with cubic ZnO particles, whereas similar values were observed over Φ =24 vol %. This demonstrates that “filler size effect”, which effectively reduces interfacial contact resistance, is significant at lower filler contents, but size effect is insignificant at higher filler contents because mutual contact among particles induces effective percolation.



Thermal conductivity of PI blend films containing MgO and ZnO particles with VDP morphology.

References

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