Effects of Vertical Double Percolation Morphology and Filler Size on the Thermal Diffusivity of Polyimide Blend/Metal Oxides Hybrid Films

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I. INTRODUCTION

Thermally stable polymers are quite important in industrial applications such as low-dielectric insulators and heat spreaders. 'Polyimides (PIs)', one of the representative super engineering plastics, have high thermal stability with good mechanical and insulation properties. Thereby, PIs have been widely used as interlayer dielectrics (Fig.1) and substrates for flexible printed circuit boards. Thermal interface materials (TIM) are used to transfer heat across jointed solid surfaces such as between CPUs and heat sinks or heat spreader. However, the thermal diffusivity ($D_{\perp}$) of PIs is much lower than those of ceramics and metals. Recently, hybrid materials consisting of polymer matrix and homogeneously dispersed inorganic fillers (Fig.2 (a)) have been investigated to enhance thermal diffusivity. We have recently reported that immiscible PI blends with selectively dispersed zinc oxide (ZnO) particles (~0.3 μm) show much higher thermal conductivity than PIs containing homogeneously dispersed ZnO (Fig2 (b)) [1].

II. EXPERIMENT

A PI blend prepared from TF and SD PIs (Fig.3) was used as matrix. Hybrid films were obtained by thermal curing of PI precursors containing metal oxide particles at 350°C for 1 h. The volume ratios of SD, TF and metal oxide particles were set to $100-x$: $100-x$: $2x$ (x=0~29.8).

III. RESULTS AND DISCUSSION

Magnesium oxide (MgO) particles (~1.2 μm) were chosen as new conductive filler in this study, and the degree of particle segregation was investigated to examine relationship between the phase separation morphology and thermal diffusivity. Under well-controlled conditions, thermal conductive pathways were spontaneously formed by selectively incorporation of particles in one phase, which is called '*vertical double percolation*' morphology. Owing to the pathways, $D_{\perp}$ is significantly enhanced. In addition, PI films containing large MgO fillers exhibit larger $D_{\perp}$s than those with small ZnO fillers due to reduced interfacial thermal resistance between PI matrix and particles. The films with MgO exhibited higher $D_{\perp}$s than those with ZnO between 0 and 24 vol % for blend films, whereas similar $D_{\perp}$s were observed over 24 vol % (Fig.2 (c)), which demonstrates that 'filler size effect' is significant at lower filler contents.

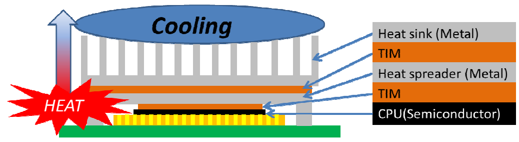

Figure 1. Schematic illustration of CPU unit.

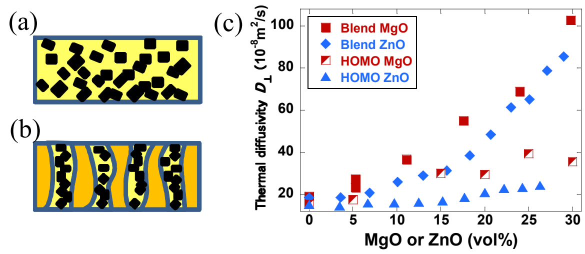

Figure 2. Schematic cross-sectional images of a polymer material with (a)homogeneous (HOMO).(b) selective dispersion of fillers (Blend) (c) Thermal diffusivity of PIs hybrid films with different volume ratios of MgO or ZnO

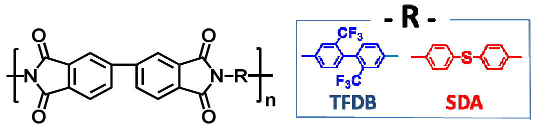

Figure 3. Molecular structure of the PIs blend:BPDA-TFDB/BPDA-SDA (TF/SD).

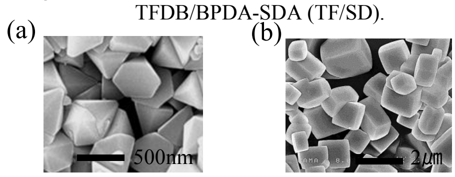

Figure 4. (a) ZnO nano-particles (b) MgO micro-particles

REFERENCE

[1] D. Yorifuji, S. Ando, "Enhanced Thermal Conductivity over Percolation Threshold in Polyimide Blend Films Containing ZnO Nano-pyramidal Particles: Advantage of Vertical Double Percolation Structure," *J. Mater. Chem*, **21**, pp. 4402-4407, Feb 2011.