Cellulose-based BN core-shell spherical microbeads provide continuous pathways composed of BN in polymer composites leading to high thermal conductivity

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We fabricated sheets with high thermal conductivity in a way that required less filler. Our approach is as follows: (1) We used core-shell spherical microbeads as the thermally conductive filler; (2) we developed cellulose/*h*-boron nitride (BN) core-shell spherical microbeads using phase separation of a cellulose xanthate aqueous solution (viscose); (3) we hybridized the cellulose/*h*-BN core-shell microbeads with epoxy resin by using compression molding. This process reduced the amount of *h*-BN required because the microbeads efficiently formed thermally conductive pathways among the shells in the insulating resin. The thermal conductivities of our thermally conductive sheet in the thickness and in-plane directions were 10.6 and 15.6 $Wm^{-1}K^{-1}$, using only 48.5 vol.% *h*-BN. In contrast, the thermal conductivities of the composite sheet with 75 vol.% of naked *h*-BN particles were 6.31 and 22.9 $Wm^{-1}K^{-1}$ in the thickness and in-plane directions, respectively. This large difference came from the anisotropic structure of *h*-BN. The changes in thermal conductivity with *h*-BN content did not agree with percolation theory, using the cellulose/*h*-BN core-shell microbeads as filler. The thermally conductive sheets fabricated with microbeads showed thermal conductivities several times greater than that of sheets fabricated with naked *h*-BN, indicating that thermally conductive pathways had formed in the insulating resin.

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