Electronic Supplementary Information (ESI)

Cellulose/boron nitride core-shell microbeads providing high thermal conductivity for thermally conductive composite sheets

by Shoji Nagaoka, Takuma Jodai, Yoshihiro Kameyama, Maki Horikawa, Tomohiro Shirosaki, Naoya Ryu, Makoto Takafuji, Hideo Sakurai, and Hirotaka Ihara

1. Details of preparation of cellulose/h-BN core/shell microbeads

We investigated the preparation conditions for preparing with cellulose core and *h*-BN particles shells. We examined two phase separation methods, one in which BN was dispersed in a cellulose xanthate aqueous solution (Procedure-1), and another the BN was dispersed in a SPA aqueous solution (Procedure-2). These two procedures are described in the ESI†, and are shown in Fig. S1. The two types of sphering procedure are described in the ESI†. In Procedure-1, the cellulose xanthate aqueous solution, containing BN particles was added to the SPA aqueous solution, as reported previously.²¹⁻²⁴ In Procedure-2, the cellulose xanthate aqueous solution was added to the BN-S particles-SPA aqueous dispersion. Table S1 summarizes the loading ratios and contents of *h*-BN and cellulose.



Fig. S1 Sphering process of cellulose/BN microbeads.

Table S1 Abbreviations of cellulose/BN microbeads and their loading ratios and contents of BN

Cellulose/BN microbeads	Loading ratio (wt.%)	Content (wt.%)
BN-S/Cell-0	50	41.6
BN-S/Cell-1	50	44.9

2. Details of contents of BN, cellulose and epoxy resin

The contents of BN, Cellulose and epoxy resin in the obtained sheet were summarized in Table S2.

Sheet No.	Filler	Content of BN		Content of Cellulose		Content of Epoxy		Gravity
	Filler Type	(wt.%)	(vol.%)	(wt.%)	(vol.%)	(wt.%)	(vol.%)	g/cm3
Sheet-1		39.9	27.0	-	-	60.1	73.0	1.52
Sheet-2	BN-S	63.3	49.0	-	-	36.7	51.0	1.55
Sheet-3		80.2	69.3	-	-	19.8	30.7	1.90
Sheet-4		19.6	12.0	-	-	80.4	88.0	1.38
Sheet-5	BN-L	57.1	42.6	-	-	42.9	57.4	1.59
Sheet-6		84.4	75.0	-	-	15.6	25.0	1.97
Sheet-7		22.4	15.5	59.1	61.5	18.6	23.0	1.53
Sheet-8	BN-S/Cell-11)	24.9	17.7	65.6	70.3	9.45	12.0	1.61
Sheet-9		26.6	19.3	70.1	76.5	3.27	4.20	1.62
Sheet-10		31.6	22.1	38.8	40.8	29.6	37.1	1.51
Sheet-11	BN-S/Cell-2 ²⁾	39.3	29.3	48.2	54.1	12.5	16.7	1.65
Sheet-12		42.5	32.5	52.2	60.1	5.35	7.30	1.72
Sheet-13		28.5	18.7	13.6	13.4	57.8	67.9	1.40
Sheet-14	BN-L/Cell-33)	55.8	43.8	26.6	31.5	17.6	24.8	1.74
Sheet-15		59.9	48.5	28.6	34.8	11.5	16.7	1.85

Table S2 Thermal conductivity of *h*-BN/epoxy resin composite sheet

1) BN content: 27.5 wt.%, 2) BN content: 44.9 wt.%, 3) BN content: 67.7 wt.%.

Measure temperature: 25 °C