

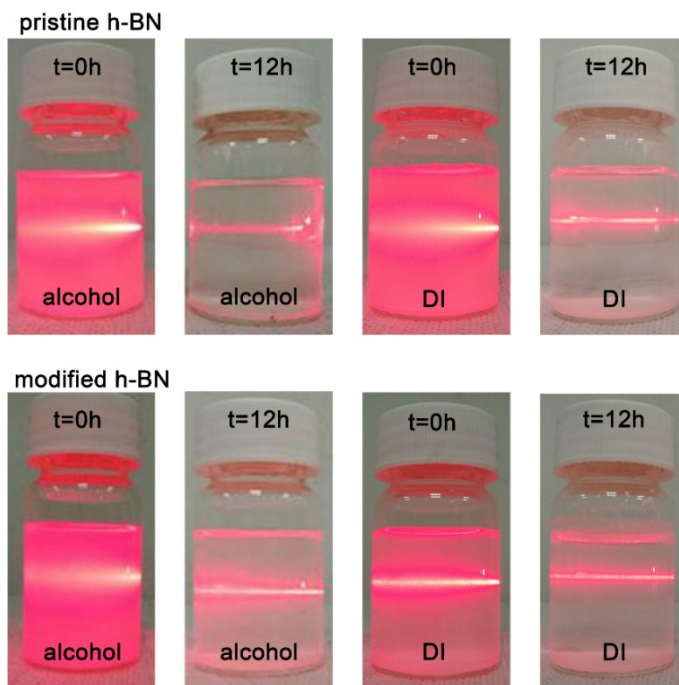
## SUPPORTING INFORMATION

### **Hybrid fillers of hexagonal and cubic boron nitride in epoxy composites for thermal management applications**

Yuyuan Zhang,<sup>a</sup> Wei Gao,<sup>a</sup> Yujing Li,<sup>a</sup> Dehe Zhao<sup>a</sup> and Hong Yin<sup>\*a</sup>

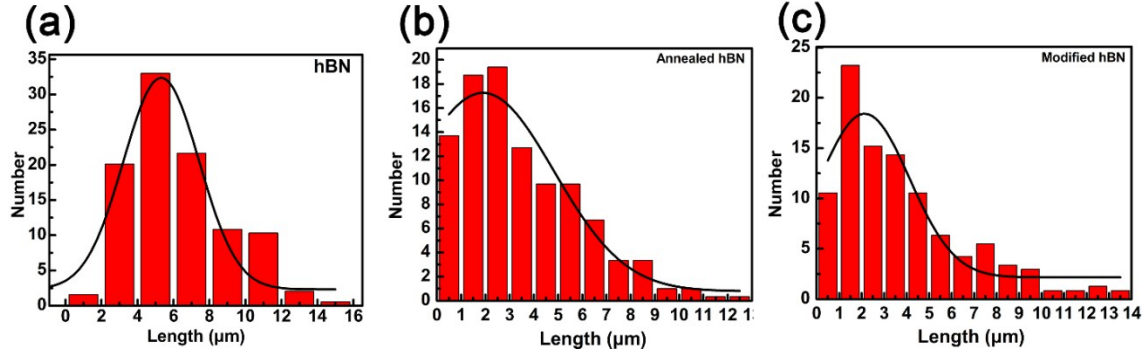
<sup>a</sup>State Key Lab of Superhard Materials, College of Physics, Jilin University, Changchun 130012, P.R. China.

<sup>\*</sup>Corresponding authors. E-mail: hyin@jlu.edu.cn



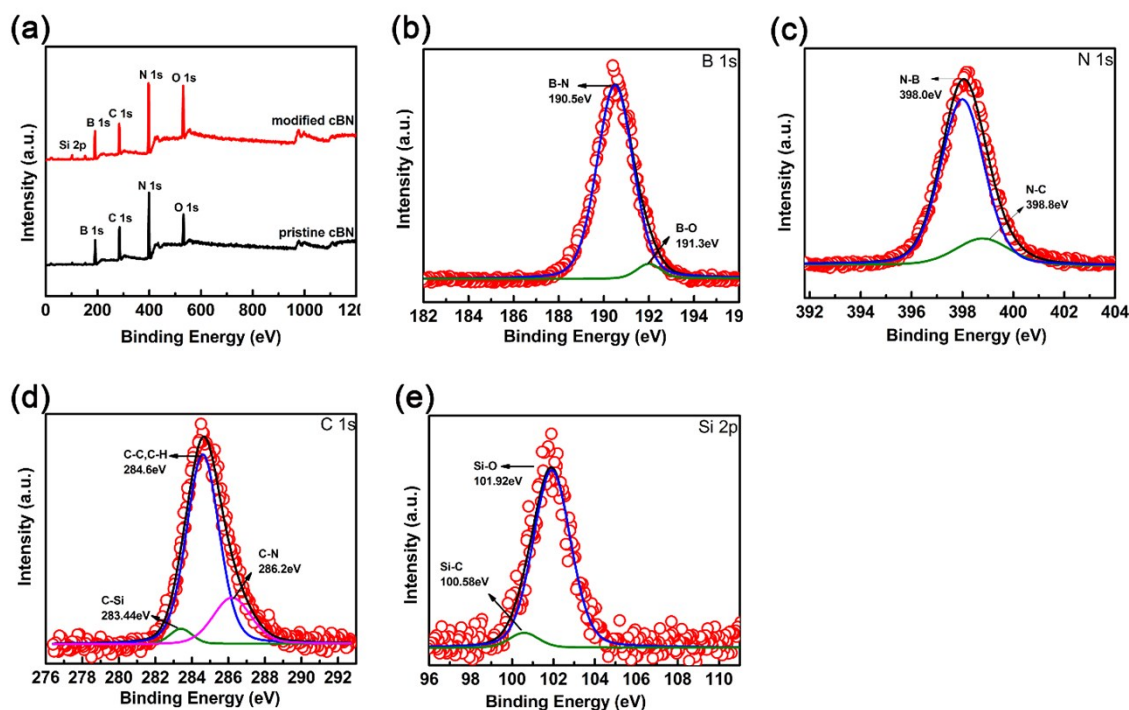
**Figure S1.** Dispersed solution of pristine and modified *h*-BN in alcohol and DI water with varied time of  $t = 0$  and 12 h, respectively.

The pristine *h*-BN powder and modified *h*-BN powder (5 mg each) were added to 15ml alcohol and deionized water (DI water) to disperse them evenly. As shown in Figure S1, all the mixed solutions were precipitated to some extent, but the concentration of modified *h*-BN dispersion was obviously higher than that of supernatant in pristine *h*-BN solution, especially in ethanol solution. When irradiated with infrared ray, the effect of Dindall is obvious.<sup>1</sup> This strongly suggests that is the modified *h*-BN has better hydrophilicity than the pristine *h*-BN, which results in better dispersion. The following measured thermal conductivity of the polymeric EP composites containing modified *h*-BN is much improved than that containing pristine ones, which can be ascribed to the reduced thermal resistance of the interface.<sup>2</sup>



**Figure S2.** Histogram of particle size analysis of pristine (a), annealed (b) and modified *h*-BN (c).

Figure. S2 illustrate the histogram of particle size of pristine *h*-BN, annealed *h*-BN and modified *h*-BN. As shown in figure S2(a), S2(b), S2(c), the particle sizes of the pristine *h*-BN, annealed *h*-BN and modified *h*-BN are about 5.3  $\mu$  m, 1.9  $\mu$  m and 2.1  $\mu$  m, respectively. This indicates that the transverse size of *h*-BN after annealing decreases due to thermal effect and suggests that after further KH-550 modification, *h*-BN platelets remains the similar geometric sizes from thermal treatment.<sup>3</sup>



**Figure S3.** (a) XPS survey spectra of pristine and modified *c*-BN. Core level are peaks of B 1s (b), N 1s (c), C 1s (d), and Si 2p (e) of modified *c*-BN, respectively.

XPS investigation has been validating the surface composition of *c*-BN after modification, as shown in Figure S3a-e. The binding energy was calibrated with reference to the C 1s energy as 284.5 eV. When compared to the pristine powder, the XPS survey spectra (Figure S3a) of *c*-BN after surface modification displays four similar peaks of B 1s, N 1s, C 1s, and O 1s at approximately 190.5, 398, 284.6, and 531.4 eV, but with different percentages. The enhancement of C 1s and O 1s implies the successful attachment of the silane coupling agent on the surfaces of *c*-BN, which is further evident by the emerge of Si 2p at 101.92 eV for the modified sample.

The chemical bonding of the modified *c*-BN is further confirmed by B 1s, N 1s, C 1s and Si 2p core level spectra together with their corresponding de-convolutions in a more detailed description as demonstrated in

Figure S3b-e. In addition to the strong B-N bond in B 1s and N 1s core level spectra clearly attributed to *c*-BN, the extra weak C-N component in N 1s and B-O component in B 1s are fitted at 398.8 eV and 191.3 eV, respectively. The former is from the contribution of surface agents, however the latter is evidencing that B is the only reactive site for the silane coupling agents through hydroxyl groups on the BN surfaces. Furthermore, the strong C-C, C-H peaks and the weak C-N, and C-Si in C 1s core level spectrum as well as Si-O, Si-C bonds are present in the structure of the silane coupling agent. This indicates that the surfaces of *c*-BN are successfully grafted by the silane coupler. Therefore, the above results clearly suggest that the silane coupling agent has been chemically grafted on the surface of *c*-BN.

**Table S1** The concentration of each element for pristine *h*-BN and modified *h*-BN.

Sample	Chemical composition (at. %)				
	<b>B 1s</b>	<b>N 1s</b>	<b>C 1s</b>	<b>O 1s</b>	<b>Si 2p</b>
pristine <i>h</i> -BN	31.5	37.5	24.1	6.7	0.2
modified <i>h</i> -BN	19.8	20.2	45.8	12.3	1.9

**Table S2** The concentration of each element for pristine *c*-BN and modified *c*-BN.

Sample	Chemical composition (at. %)				
	<b>B 1s</b>	<b>N 1s</b>	<b>C 1s</b>	<b>O 1s</b>	<b>Si 2p</b>
pristine <i>c</i> -BN	29.0	29.2	30.5	10.5	0.8
modified <i>c</i> -BN	29.3	29.4	20.9	16.0	4.4

The elemental compositions of B, N, C, O and Si have been evaluated based on the XPS results of *h*-BN (Fig. 2d) and *c*-BN (Fig. S3a). The results are listed in Table S1 and Table S2, respectively. It can be seen that after modification of *h*-BN and *c*-BN, the content of Si increases significantly reaching 1.9 at.% for the modified *h*-BN surface and , and 4.4 at.% for the modified *c*-BN. This indicates that the both surfaces of *h*-BN and *c*-BN are successfully decorated by the silane coupler.

## References

- 1 Y. Lin, T. V. Williams, T. B. Xu, W. Cao, H. E. Elsayed-Ali and J. W. Connell, *J. Phys. Chem. C*, 2011, **115**, 2679-2685.
- 2 I. Jang, K.-H. Shin, I. Yang, H. Kim, J. Kim, W.-H. Kim, S.-W. Jeon and J.-P. Kim, *Colloid. Surf., A*, 2017, **518**, 64-72.
- 3 B. Yu, W. Y. Xing, W. W. Guo, S. L. Qiu, X. Wang, S. Lo and Y. Hu, *J. Mater. Chem. A*, 2016, **4**, 7330-7340.