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Supporting information

A Novel Environmentally Friendly Material of Boron Nitride/Lignosulfonate/Nature Rubber

Composite with improved thermal conductivity

Dong An^{1,3}, Shuaishuai Cheng¹, Can Jiang², Xiaoyuan Duan¹, Bo Yang¹, Zhiyi Zhang^{1*}, Jiaxiong Li⁴,

Yaqing Liu^{1*}, Ching-Ping Wong³

1. Shanxi Key Laboratory of Nano-functional Composite Materials, North University of China, Taiyuan

030051, China.

2. School of Materials Science and Engineering, Wuhan Institute of Technology, Wuhan 430205, China.

3. School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, 30332, USA.

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Experimental Section

Materials

The raw h-BN powders were obtained from Sigma-Aldrich. Nature rubber latex (NRL, 60%) was supplied by the Chengyi Rubber Company. Lignosulfonate were purchased from Shangdong Gaotang Duoyuan Lignin Co. Ltd. The sucrose, gelatin, Zinc oxide (ZnO), Zinc diethyl dithiocarbonate (ZDC), Potassium hydroxide (KOH) and Sulfur (S) were obtained from the Aladdin without further processing. The ZnO, ZDC, KOH and S were used as activator, accelerator, stabilizers and vulcanizing agents for the NRL, respectively. *Preparation of carbonized BN/Lignosulfonate hybrids*.

Typically, a certain ratio of h-BN and sucrose were mixed together, then the hydroxyl-functionalized BN could be obtained after the ball milling treatment. Then, the modified BN were added the lignosulfonate solution with agitation and sonicated until forming a homogeneous mixture. Next, the mixed solution was filtrated and dried with the temperature (60 °C). Finally, the hybrids were carbonized from room temperature to 800 °C for 4 h under the Ar atmosphere.

Preparation of carbonized BN/Lignosulfonate/NR composites.

The BN/Lignosulfonate/NR composites were fabricated by the vacuum-assisted infiltration method. First, certain mass ratio of NRL, ZnO, ZDC, KOH and S were well mixed at the room temperature, where ZnO, ZDC, KOH and S accounted for 0.4 wt%, 0.5 wt%, 0.1 wt% and 1 wt% of the natural rubber, respectively. hen, the carbonized BN/Lignosulfonate were added into the NRL mixture with agitation for 1 h until get a

uniform mixture. After that, the mixture were filtered with a membrane through vacuum system equipment and then peeled of it. Finally, the samples were degassed the air under vacuum oven at 50 °C for 2 h and vulcanized at the temperature of 80 °C for another 2 hours to obtain the composites. Different mass ratios of BN/Lignosulfonate hybrids (1:1; 2:1; 4:1; 8:1) were prepared and the composites with different filler contents were also fabricated. For comparison, the pure NR and BN/lignosulfonate/NR composites without carbonization were prepared with an identical vulcanized process.

Characterization

The lateral size of raw h-BN, modified BN and the hybrids was tested through a ZetaPALS particle size analyzer (Brookhaven Instruments Corp) and all materials were dissolved in the deionized water and sonicated for 30 min with a concentration of 0.1mg/mL before testing. The morphologies of the fillers and composites were characterized by the field-emission scanning electron microscopy (FESEM, Hitachi SU8010) and transmission electron microscopy (TEM, JEM-2100F). The X-ray diffraction (XRD, RU-300, Rigaku, Japan) with Cu Ka radiation (k = 0.1546 nm) was carried out to know the phase structures of fillers. Raman spectra was characterized through a DXR Raman microscope with a 514.5 nm excitation source from an Ar⁺ laser at the room temperature. Fourier transform infrared spectroscopy (FTIR, Thermo Nicolet 360) and the surface elemental state (XPS, ESCALAB210) were also measured. Thermogravimetric analysis (TGA) was characterized by a Q50 TGA (TA instruments) from 40 to 800 °C under N₂ with a heating rate of 10 °C/min. Dynamic mechanical analysis (DMA/SDTA861°) were used to confirm the modulus of the composites. As for the performance characterization, the thermal conductivity was tested by steady-state heat flow method (DRL-III, China), which the value could be obtained from the following equation:

$$k = -\frac{Q\Delta X}{A\Delta T} \tag{1}$$

Where K is the thermal conductivity (W m⁻¹ K⁻¹), Q is the heat flux (W), and ΔX and A are the thickness

(m) and area (m²) of the sample, respectively. ΔT is the temperature difference between the two hot meter bars. In addition, the size of test samples were typically in the form of discs 20 mm in diameter and about 2 mm thick. For the thermal dissipation test, which performed by an IR thermal camera with the spatial resolution of 1.31 mRad (Fluke Ti400 IR Fusion Camera, USA). Before testing the heat absorption capability, three different samples should place on a stainless steel plate and then put into an 80 °C oven to heat the sample. Similarly, the heat dissipation test were needed to put all samples in an oven at 80 °C for 30 min to guarantee the same temperature and then transferred to a thermally insulating stage at a room temperature environment. The tensile of the samples were tested by a testing machine (Instron 5567, US). Meanwhile, the measurement of volume resistance was tested with a resistivity meter (TH2683, China), which the AC voltage were 1000 V and the samples were cut into a rectangle from the thermal conductive samples. Then, the resistivity could be calculated through the following formula:

$$R = \rho \frac{l}{A} = \rho \frac{l}{ab}$$

where ρ , *l*, A, a, and b represented the resistivity, length, cross-sectional area, thickness and width of the rectangular sample, respectively.



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Figure S2. Particle size distribution of raw h-BN and h-BN after ball milling method with sucrose according to the

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Figure S3. The FTIR curves of pure BN, sucrose and hydroxyl-surface modified BN with mixing the sucrose by the ball-

milling method.



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Figure S5. The SEM image of lignosulfonate with different magnification.



Figure S6. The FTIR curves of lignosulfonate.



Figure S7. The TGA curve of the pure lignosulfonate.



Figure S8. The dynamic mechanical analysis of pure NR and BN/lignosulfonate/NR composites (before and after

carbonization, BN/lignosulfonate=4:1, 20 wt %).

BN/lignosulfonate	V _c (%)	β	\mathbf{K}_{0}
Before carbonization	10.4	0.14	0.44
After carbonization	8.2	0.032	1.10

 Table S1. The value of different parameter.



Figure S9. The fitting process of Foygel model for BN/lignosulfonate/NR composites after carbonization.

Firstly, do the tangent for the original experimental data of BN/rGO/NR composites and the abscissa of intersection from two tangent lines was the critical percolation. Then transformed into a linear form: $y=A + B \times x$, where A equals to lg K₀, B equals to β , x equals to lg [(V_f- V_c) / (1 - V_c)], y equals to lg (K - K_m), respectively. After fitted, the values of A and B were

obtained. Based on this, the values of K_0 and β were calculated.



Figure S10. The lateral size distribution of BN/lignosulfonate hybrids after carbonization.



Figure S11. The fitting process of Foygel model for BN/lignosulfonate/NR composites before carbonization.