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Supporting information

Structural design of multilayer thermally conductive nanofibrillated cellulose hybrid film with electrical insulating and antistatic property

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Preparation of G2s: The G2s dispersion were prepared using a classical reported method.[1] NaC was dissolved in deionized (DI) water at the concentration of 0.1 mg/mL. G2s dispersion was prepared by adding 2 g of G2 powders (the pristine graphene) to 400 mL of NaC solution in a 500 mL capped beaker, and then the beaker was placed in a low-power (80 W) sonic bath for 24 h. Samples were left overnight and then centrifuged for 90 min at 1500 rpm. Then, the top two-thirds of the dispersion was extracted and retained for use. **Preparation of BNs:** The preparation of BNs was consistent with the method reported by Zhou,[2] 1.5 g of pristine BN powder was added into 500 mL of ethanol/H_2O mixed solvent (v/v = 9:1vol%). The mixed solution was sonicated for 48 h in a low-power (80 W) sonic bath, and then settled at room temperature for 24 h. In the end, the top 3/4 of the supernatant was collected for use. **Preparation of Nano fibrillated cellulose (NFC).** The NFC was prepared according to the method reported by Saito.[3, 4] Cellulose fibers (cellulose

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content of 2 g) were suspended in DI water (200 mL) that contained TEMPO (0.025 g) and NaBr (0.25 g). The oxidation reaction of the cellulose slurry was initiated by adding the desired amount of the NaClO solution (10 mmol•g^{-1} cellulose), and the reaction continued at room temperature with stirring for 6 h. The pH of the reaction solution was maintained at 10 by adding 0.5mol of NaOH. The oxidized cellulose was thoroughly washed with DI water by filtration on a filter membrane (PTFE, 0.45 μm). Subsequently, 2 mg • mL^{-1} of oxidized cellulose/water slurries were sonicated for 15 min at a power of 300 W in an ice bath using an ultrasonic generator with a probe tip that had a diameter of 15 cm. Then, the slurry was centrifuged at 8000 rpm for10 min to remove the un-nanofibrillated cellulose. The transparent NFC suspension was stored at 4 °C before use.

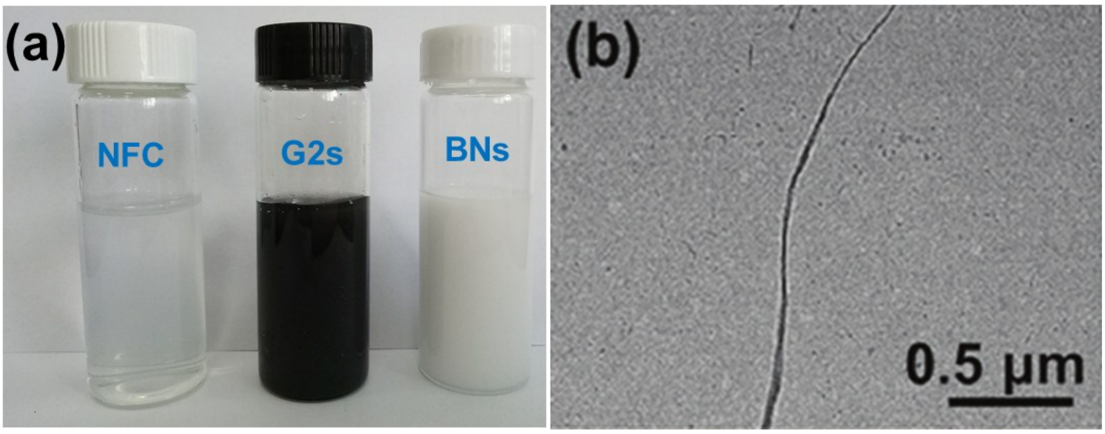

Fig. S1 (a) The photograph of the NFC suspension and the filler of G2s and BNs dispersion (b) the TEM of the NFC whisker

The particle size of the fillers. From the Fig.S2a and Fig.S3b, most of the G2s was distributed about 10μm, and the BNs was mainly distributed about 200nm (Fig.S2b and Fig.S3d).

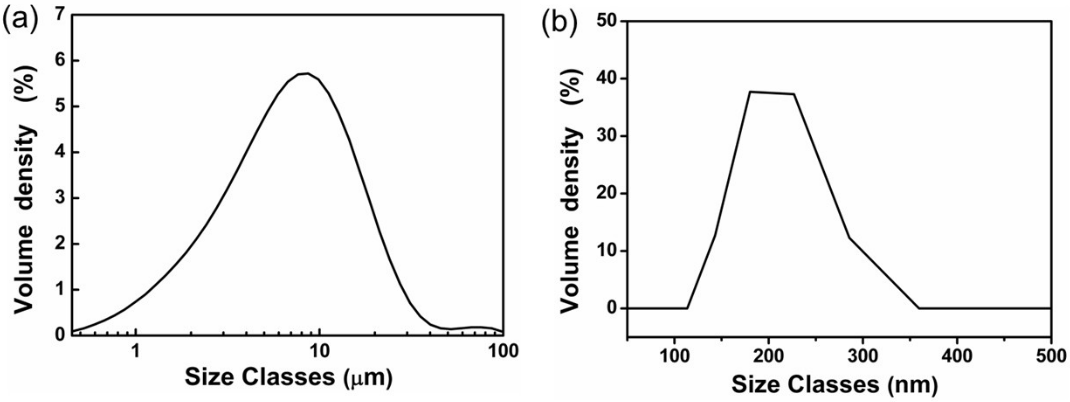

Fig. S2 The particle size of the (a) G2s (b) about BNs

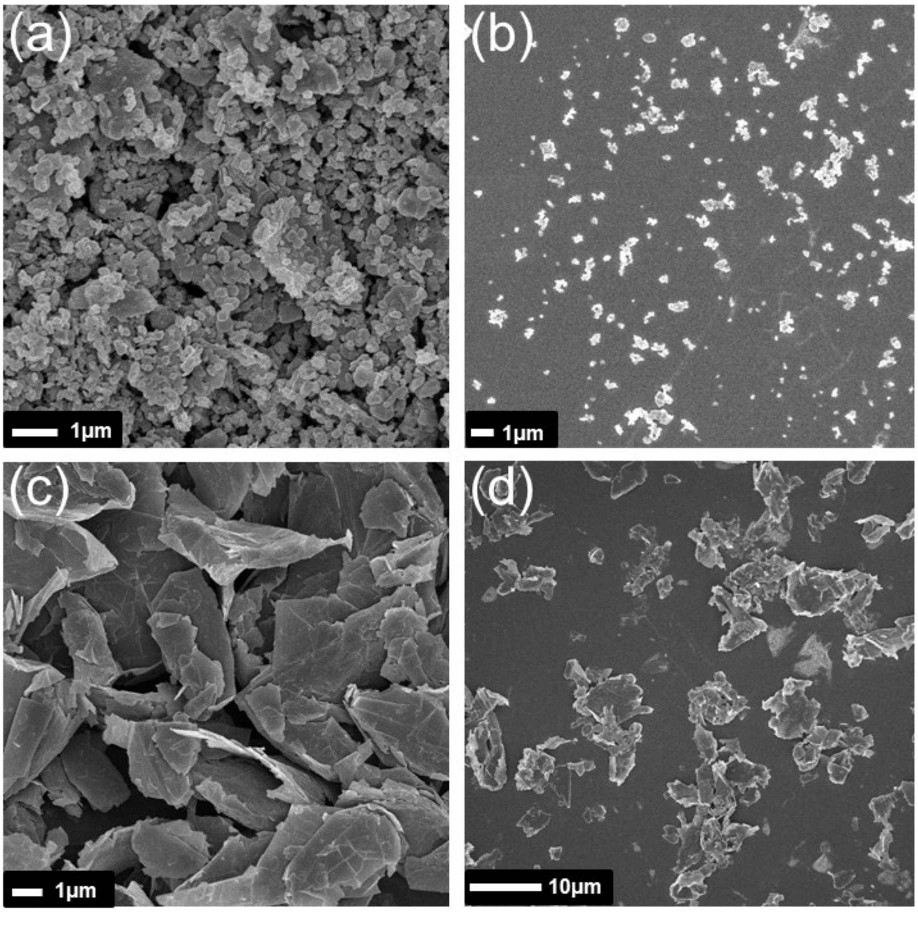

Fig. S3 The SEM pattern about the fillers before and after ultrasonication (a) BN (b) BNs (c) G2 (d) G2s

Characterization of Raman. The Raman peak of the BNs was red-shift from 1365.2 cm^{-1} to 1367.2 cm^{-1} (Fig.S4a), which was caused by a hardening of the corresponding

E_{2g} phonon mode in monolayers and indicated that the BNs sample might be exfoliated into monolayers.[5, 6] Gorbachev and co-workers also found the 2-4 cm^{-1} red-shift for BN monolayer.[7] The full width at half-maximum of Raman characteristic peak of BNs was narrower than 30 cm^{-1}, suggesting that BNs sheets had good (002) crystallinity.[8] And the situation of graphene is similar to above, which red-shift from 1575.2 cm^{-1} to 1578.7 cm^{-1} (Fig.S4b).

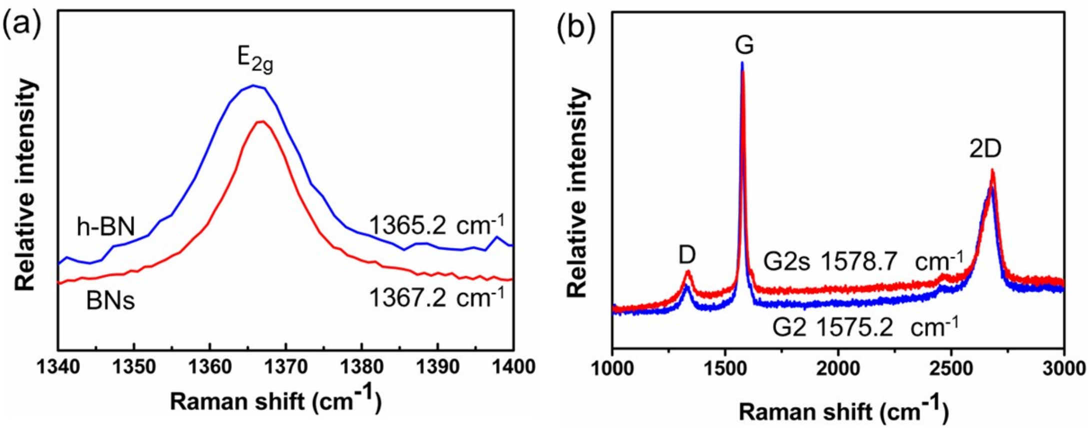

Fig.S4 The characterization of Raman before and after ultrasonication (a) the filler of BN (b) the filler of graphene

The existence of intermolecular hydrogen. The primary contributors to the interactions between the NFC and G2s-Sodium cholate were considered to be the hydrogen bonds between plentiful −OH groups on NFC and G2s-Sodium cholate. The intermolecular hydrogen bonds in the hybrid film can cause a downshift of the broad vibration peak position relating to the −OH group.[9, 10] In the present study, the −OH peaks at approximately 3100-3500 cm^{-1}, shown in Figure S5, clearly illustrate a downshift in the NFC/G2s layer, confirming the hydrogen bonding between the NFC and the G2s.

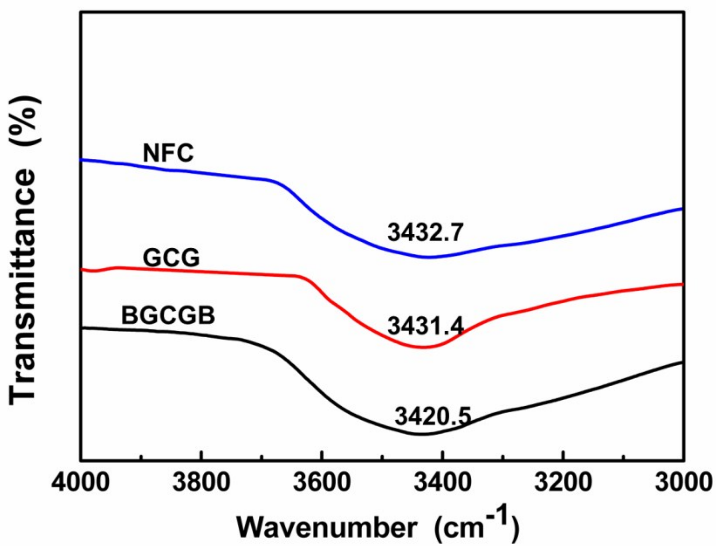

Fig.S5 FTIR spectra of the film about NFC, GCG and BGCGB

Characterization of EDS. To further demonstrate the multilayer structure of film, EDS pattern was employed to evaluate it. The element of boron mainly distributes on the top and bottom layer, it may be generated by the layer of BNs/NFC, indicating that the multilayer structure was formed (Fig. S6). We want to manifest the location of the layer of G2s/NFC simultaneously, however, the characteristic element of carbon was contained everywhere in the G2s and the substrate of NFC, therefore, it is hard to prove it using this equipment merely. Cooperation with the characterization of SEM, the multilayer structure of the NFC hybrid film got confirmed effectively. The gap between layers may be caused by the unequal breaking power when cracking due to different materials among the layer and layer, and the uneven thickness may be created by the force of vacuum filtration.

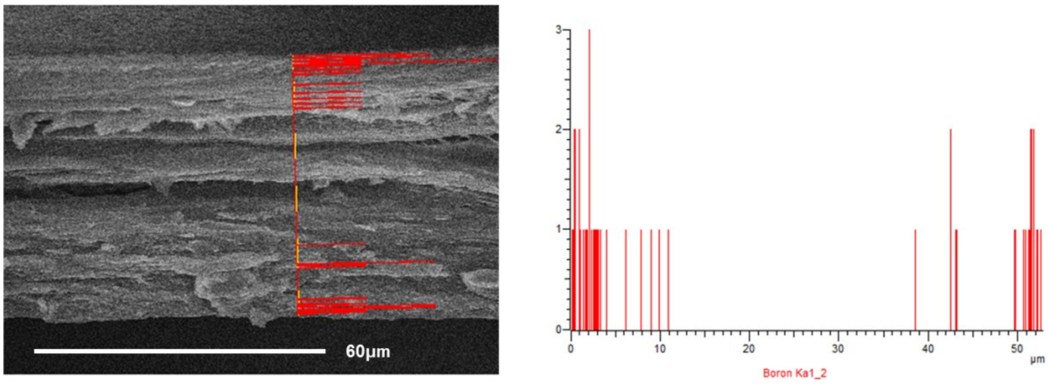

Fig.S6 The EDS pattern about the element of boron of the BGCGB film

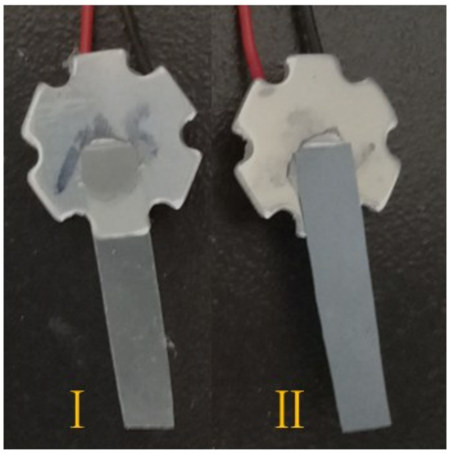

Fig.S7 The photograph of the IR thermal imaging test (Ⅰ is NFC film and Ⅱ is BGCGB film).

The transparency of the BCB film

The tri-layer BCB hybrid film with 8wt% loading of BNs is relatively optical transparent. Compared with pure NFC film (84.1%), it exhibits 33.1% transmittance at 550 nm wavelength, which can be applied as substrate in thermal management of optoelectronics (Fig.S8).[11]

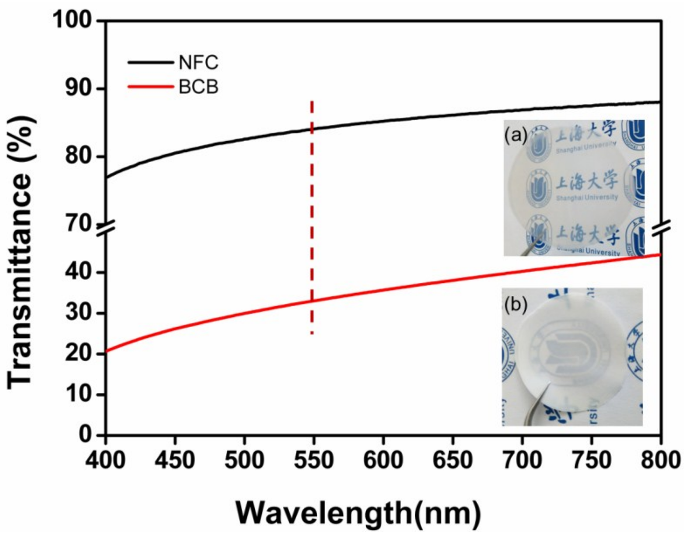

Fig.S8 Transmittance of pure NFC film and BCB film, and (inset) corresponding the photograph of the films

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