# Supporting Information

## Jelly-inspired Construction of 3D Interconnected BN Network for Lightweight, Thermally Conductive, and Electrically Insulating Rubber Composites

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## Characterization

S-4700 scanning electron microscope (Hitachi Co., Japan) was utilized in the observation of the fracture surfaces of 3D porous BN aerogel and 3D BN-PDMS composites. A thermogravimetric analyzer (Mettler-Toledo, Switzerland) was employed to estimate the thermal property of the 3D BN-PDMS composites. The volume resistivity of 3D BN-PDMS composites were measured by a digital insulated resistor tester (PC68, China). The thermal conductivity of the composites were calculated from the following formula:

$$\lambda = \rho C_{\mathrm{p}} \alpha \quad (1)$$

where $\rho$, $C_p$ and $\alpha$ are the density, specific heat capacity and thermal diffusivity of the composites respectively. To obtain the more accurate values, $C_p$ was obtained by differential scanning calorimetry (DSC, TA Q2000) with the sapphire method. $\alpha$ was determined by a laser flash apparatus (Netzsch LFA-467). It should be pointed out that there would be a layer of rubber-enriched edges on the surface of the sample. This transparent rubber film can be easily peeled off. In order to avoid its influence on the test result, we would use a knife to polish the surface of the sample. The temperature distribution images of the samples were taken by an infrared imaging camera (FLIR E30, FLIR Systems, Inc., USA). The spatial distribution of BN was observed by X-ray tomography (XRT, nanoVoxel 4000, Sanying Precision Instruments Co., Ltd, China).

**Figures**

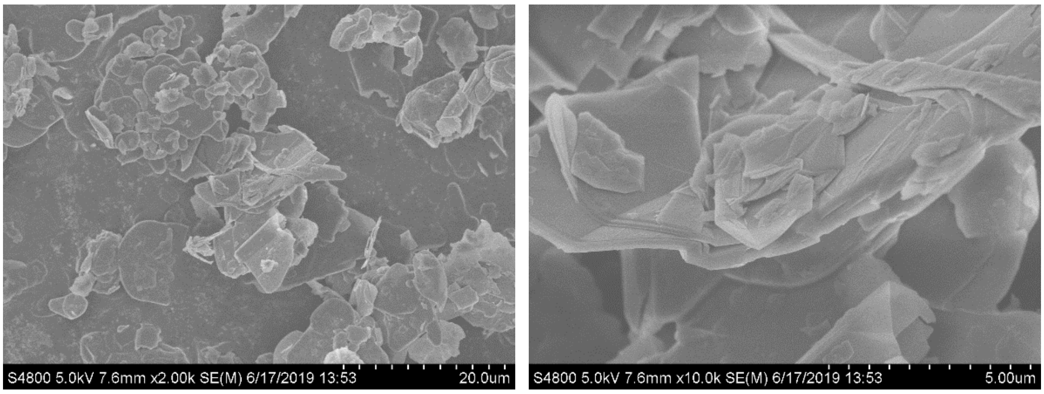


**Figure S1.** SEM images of the as-received BN powder.

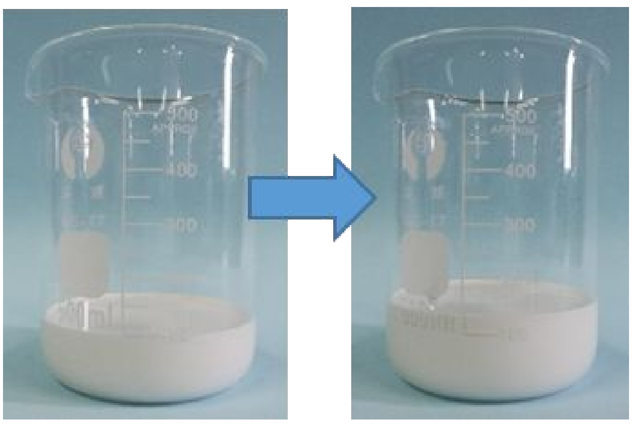

**Figure S2.** Volume changes before and after foaming.

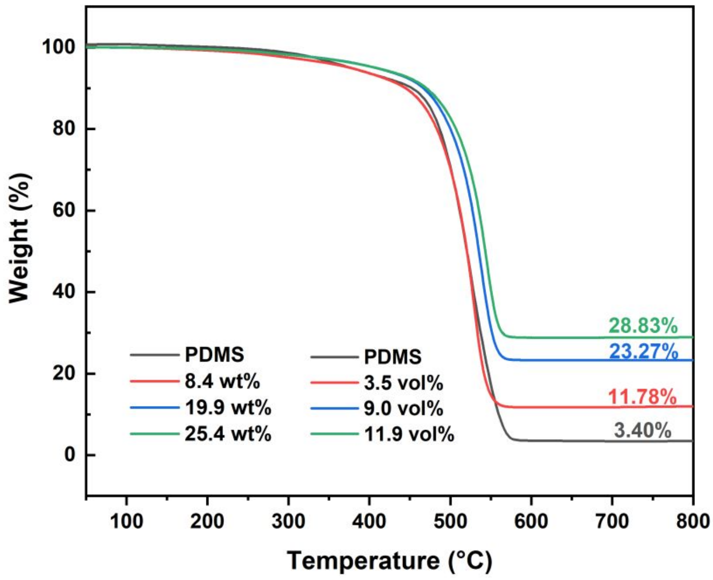


**Figure S3.** TGA curves of the 3D BN-PDMS composites with different BN contents.

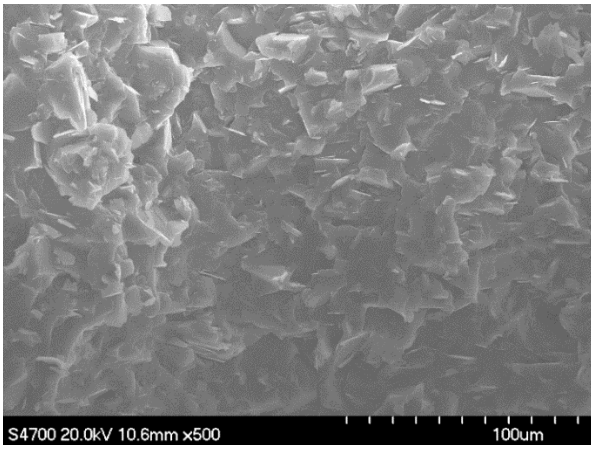


**Figure S4.** Fracture surface SEM microstructure image of the random BN-PDMS composite.

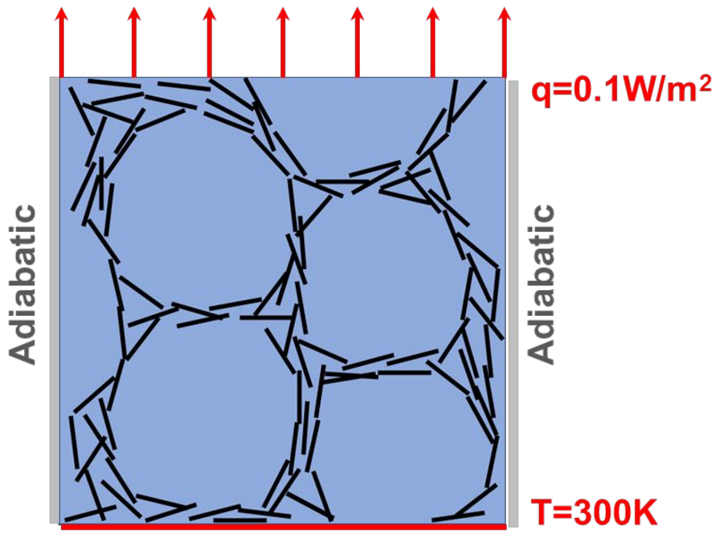


**Figure S5.** Boundary conditions of the Finite Element Simulation.

**Table S1** The performance comparison between our silicone rubber composites and the commercially-available thermally conductive silicone rubber gap pad.

| Company Referance | Product Code | Volume Resistance (Ohm/cm) | Thermal conductivity (W/(m·K)) | Density (g·cm$^{-3}$) |
|---|---|---|---|---|
| ShinEtsu | TC-80S2 | $10^{13}$ | 2.0 | 2.9 |
| | TC-80C | $10^{12}$ | **1.5** | **2.6** |
| | HC-20AS | $10^{13}$ | 0.7 | 2.23 |

| | | | | |
|---|---|---|---|---|
| Parker | THERM-A-GAP HCS10 | $10^{14}$ | 1.0 | 2 |
| Chomerics | THERM-A-GAP 569 | $10^{14}$ | **1.5** | **2.2** |
| Bergquist | Gap Pad A2000 | $10^{9}$ | 2.0 | 2.9 |
| | Gap Pad 1500 | $10^{9}$ | **1.5** | **2.1** |
| | Gap Pad EMI1.0 | $10^{8}$ | 1.0 | 2.4 |
| Laird | Tflex 300TG | - | 1.2 | 1.79 |
| **Our work** | **3D BN-PDMS (25.4 wt%)** | $\mathbf{10^{15}}$ | **1.58** | **1.27** |

**Supplementary method S1. Calculation of contact resistances**

It is known that effective medium theory (EMT) and Foygel theory were usually used to calculate the interfacial thermal resistance at filler/filler and filler/matrix interface. According to the previous report[1-2], the Foygel theory was much more precise than EMT models when the well-ordered structures in the polymer matrix, which expressed as follows:

$$\lambda - \lambda_m = K_0\left(\frac{V_f - V_c}{1 - V_c}\right)^{\beta}$$

$$R_c = \frac{1}{K_0 \mathrm{L}(V_c)^{\beta}}$$

where $\lambda$ and $\lambda_m$ were represented the thermal conductivity of BN-PDMS composites and pure PDMS matrix, respectively. $K_0$ was a pre-exponential factor ratio related to the contact between BN as well as to the topology of the percolation cluster. $\beta$ were the conductivity exponent. $V_c$ was the critical percolation of fillers. $R_c$ is the contact resistance between BN; L is the lateral size of BN (10μm). For the randomly distributed and 3D BN-PDMS composites (11.9 vol%), by fitting the thermal conductivity, the values of $K_0$ are obtained as 0.32 and 0.49, respectively. Then, the calculated $R_c$ values are $2.1\times10^6$ K/W and $5.3\times10^5$ K/W.

**Supplementary method S2. Finite Element Model**

The heat transfer processes of the 3D BN-PDMS composite and randomly dispersed BN-PDMS composite were modeled in Abaqus, which included square composites (150μm*150μm), linear heat sources at the bottom with a heat flux density of 0.1 $W/m^2$ and a temperature of 300 K. The length and aspect ratio of BN were set to about 10μm and 10 in the model, which is basically the same as the actual size of BN. The thermal conductivity of BN and pure PDMS were 300 W/(m·K) and 0.186 W/(m·K) respectively, and the BN loading was 25.4 wt% (11.9 vol%).

## Supplementary References


1. Chen, J.; Wei, H.; Bao, H.; Jiang, P.; Huang, X., Millefeuille-Inspired Thermally Conductive Polymer Nanocomposites with Overlapping BN Nanosheets for Thermal Management Applications. ACS Appl Mater Interfaces 2019, 11 (34), 31402-31410.
2. An, D.; Cheng, S.; Xi, S.; Zhang, Z.; Duan, X.; Ren, Y.; Li, J.; Sun, Z.; Liu, Y.; Wong, C.-P., Flexible thermal interfacial materials with covalent bond connections for improving high thermal conductivity. Chemical Engineering Journal 2020, 383, 123151.