Supporting Information

Crafting "Brick-Mud" Segregated Nanocomposites: A Novel Approach to Superior Electromagnetic Interference Shielding, Electrical Insulation, and Thermal Conductivity in Biopolymers

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Experimental section

Differential scanning calorimetry (DSC) analysis.

The melting points of PCL/BN and PLA/CNTs composites were characterized by DSC analysis. DSC analyses were conducted with a Q2000TA instrument (Delaware, USA) in a nitrogen atmosphere at a flow rate of 50 mL/min. The analysis of PLA/CNTs composites was carried out in three steps: initial heating from 25 °C to 200 °C, cooling back to 25 °C after a 2 min hold, followed by a second heating from 25 \degree C to 200 \degree C. A sample size of 5 mg was used, with both heating and cooling rates set at $10\degree$ C/min. The analysis of PCL/BN composites is the same as above, but the maximum temperature is set to 100°C.

Morphological characterization.

The morphology of the fracture surface for s-*x*B*y*C and r-*x*B*y*C composites was observed using a Digital Microscope (DM, DM4, China). The fracture morphology of PLA/CNTs composites was observed using a scanning electron microscope (SEM, Regulus 8100, HITACHI). Before SEM analysis, the fracture surfaces were gold-plated for 90 s in vacuum conditions. The operating voltage for SEM was set at 10 kV. The brittle fracture of the sample was obtained by cooling the sample with liquid nitrogen for 2 h and then rapidly cracking it with force.

Rheological properties measurement.

Rheological properties of the PLA/CNTs composites, with dimensions of 25 mm in diameter and 1 mm in thickness, were assessed using a MCR302 rheometer (Anton Paar, Austria). Analysis was conducted at 180 °C, employing a frequency sweep from 100 to 0.1 rad/s at a constant amplitude of 1 % to guarantee the identification of the linear viscoelastic (LVE) region.

Electrical conductivity measurements.

Electrical conductivity measurements for PLA/CNTs composites were conducted in dry air at ambient temperature using a four-point probe resistivity system (RTS-9, Guangzhou Four Probes Technology Company Ltd., China) and a high-resistance meter (ZC36, Shanghai Anbiao Electronic Co., Ltd., China).

Electromagnetic interference (EMI) shielding performance measurements.

The EMI performance of the 3mm PLA/CNTs composites was evaluated within the Xband frequency range (8.2-12.4 GHz) via a vector network analyzer (Ceyear 3672C-S, Ceyear Technologies Co., Ltd., China). The EMI shielding performance of the material is quantified by the shielding efficiency (SE), which consists of absorption (SE_A), reflection (SE_R), and multiple internal reflections (SE_M). When the total SE (SE_T) exceeds 15 dB, the effect of SE_M becomes negligible. The relevant calculation equations are provided in formulas (1)-(6)[1].

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T = |S_{12}|^2 = |S_{21}|^2 \#(1)
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$$
R = |S_{11}|^2 = |S_{22}|^2 \#(2)
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$$
A = 1 - T - R \#(3)
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$$
SE_A = -10log(\frac{T}{1-R}) \#(4)
$$

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$$
SE_A = -10log(1-R) \#(5)
$$

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$$
SE_T = SE_R + SE_A \#(6)
$$

Here, T, R, and A represent the transmission, reflection, and absorption coefficients, respectively. The S-parameters $(S_{11}, S_{12}, S_{22},$ and $S_{21})$ were measured using a vector network analyzer.

Supplementary Figures and Tables

Fig. S1. The second-heating curves of (a) PCL, PCL/20B and PCL/40B and (b)PLA, PLA/5C and PLA/10C.

Fig. S2. Digital Microscope images of cryo-fractured surfaces: (a) s-4B4C, (b) s-4B8C, (c) s-8B8C, (a') r-4B4C, (b') r-4B8C, (c') r-8B8C.

Fig. S3. SEM images of cryo-fractured surfaces: (a)PLA, (b) PLA/5C, (c) PLA/10C.

Fig. S4. Rheological properties of PLA and PLA/CNTs composites: (a) storage modulus, (b) loss modulus, (c) tan δ, (d) complex viscosity.

Fig. S5. (a) Electrical conductivity and (b, c) EMI shielding performance of PLA and PLA/CNTs composites.

Fig. S6. SEM and EDS images of (a) r-8B8C and (b) s-8B8C.

Fig. S7. Reflection (R), absorption (A), and transmission (T) coefficients of various materials.

Fig. S8. Thermal performance test of the CPU-J1800.

Table S1. Electrical conductivity and volume resistivity of various samples.

Table S2. Thermal conductivity (TC) of various samples.

References

[1] Wang J, Song T, Ming W, Yele M, Chen L, Zhang H, et al. High MXene loading, nacre-inspired MXene/ANF electromagnetic interference shielding composite films with ultralong strain-to-failure and excellent Joule heating performance [J]. Nano Research. $2023;17(3):2061-2069.$