Supporting Information

Impact of Simultaneous Silane and PCPA Grafting on the Thermal and Mechanical Properties of BN and GO Hybrid Composites

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Table S1. Density, heat capacity, thermal diffusivity, and thermal conductivity of the composites.

Composite	**EP**	**BP10**	**BP30**	**BP50**	**BN50**	**BP50 /GP1**	**BP50 /GP3**	**BP50 /GP5**	**BN50 /GO5**
Density (g/cm^3)	1.110	1.129	1.289	1.368	1.450	1.450	1.454	1.459	1.522
Heat capacity (J/g·K)	1.401	1.317	1.306	1.284	1.035	1.271	1.267	1.22	1.076
Thermal diffusivity (m^2/s)	0.132	0.297	0.564	0.954	0.794	1.099	1.679	2.713	2.059
Thermal conductivity (W/m·K)	**0.205**	**0.442**	**0.949**	**1.677**	**1.191**	**2.026**	**3.093**	**4.829**	**3.374**

Extended discussion: Theoretical calculation of the thermal conductivity of composites.

To demonstrate the impact of fillers on the thermal conductivity of composites when the filler content surpassed the percolation threshold, we utilized Agari-Uno models to assess the role of fillers in establishing thermal conductive networks. The detailed description is provided below.

To ensure precise predictions, we achieved a remarkable agreement with thermal conductivities by incorporating a specific probability factor, denoted as P, which takes into account the percolated conductive paths formed by the fillers within the composite. The Agari-Uno model is a modification of the Maxwell model, which considers the influence of fillers in creating a thermally conductive network within the polymer matrix. In this context, k_c represents the thermal conductivity of the composite, k_m stands for the thermal conductivity of the matrix, k_p corresponds to the thermal conductivity of the particles, and V_f signifies the volume content of particles. According to this theory, the thermal conductivity can be expressed by the following equation:

$$k_c = k_m \frac{k_p + 2k_m + 2(1-P)(k_p - k_m)}{k_p + 2k_m - (1-P)(k_p - k_m)} + V_f P c^2 k_p$$

where c^2 is the cross-sectional area of the thermal conducting path, $P = (V_f)^{(V_f)^{-2/3}}$ and $V_f = 3c^2 - 2c^3$. In the study, an alternative model was introduced to estimate the thermal conductivity of a two-phase blended composite using the following equation:

$$log(k_p) = V_f[P^* - log(C_1 k_m)] + log(C_1 k_m)$$

where $P^* = X_2 C_2 \log k_p + X_3 C_3 \log k_3 + \cdots$; C_i is the factor, indicating the formation of a thermal conducting path within the matrix, and X_i represents the ratio of fillers in the blended composite. k_i represents the thermal conductivity of a polymer in a multiphase composite. In our composite system, where X_1 is assumed to be 1, the other coefficients can be disregarded, simplifying the equation to:

$$\log k_c = V_f C_2 \log k_f + (1 - V_f) \log(C_1 k_p) = (C_2 \log k_p - \log C_1 k_m) V_f + \log(C_1 K_m)$$

The coefficients, C1 and C2, were determined by fitting the plot of K_c versus V_f using experimental measurements. The slopes which were calculated as 1.21 for BPx and 0.96 for BP50/GPx composites, were used to predict the thermal conductivity of these composites. This prediction was achieved by substituting the slope and intercept values into the resulting equation, which is presented in Table S2.

Table S2. C1 and C2 value of cellulose/GO and cellulose/APTES-GO composites.

	BPx	**BP50/GPx**
Equation of predicted k_c	$k_c = 10^{(0.03Vf - 0.61)}$	$k_c = 10^{(0.11Vf + 0.21)}$
C_1	**1.21**	**0.96**
C_2	**0.07**	**0.14**

Following the detailed deduction procedure, the thermal conductivities (k_c) of the composites were simplified using the following equation:

$$\log k_c = V_f \cdot C_2 \cdot \log k_p + (1 - V_f) \cdot \log(C_1 \cdot k_m)$$

where C_1 quantifies the impact of heat transfer associated with the secondary structure of the matrix, while C2 signifies a factor linked to the ease with which the thermal conductive path is established by the particles within the composite. Consequently, the thermal conductivities of BP50 and BP50/GP5 were effectively engineered, demonstrating that PCPA and GPTMS facilitated the formation of a heat transport pathway more efficiently, thereby enhancing the matrix-filler network.