High thermal conductivity through simultaneously aligned polyethylene lamellae and graphene nanoplatelets†

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The effect of simultaneous alignment of polyethylene (PE) lamellae and graphene nanoplatelets (GnP) on the thermal conductivity \( k \) of PE–GnP composites is investigated. Measurements reveal a large increase of 1100% in \( k \) of the aligned PE–GnP composite using 10 wt% GnP relative to unoriented pure PE. The rate of increase of \( k \) with applied strain for the pure PE–GnP composite with 10 wt% GnP is found to be almost a factor of two higher than the pure PE sample, pointing to the beneficial effect of GnP alignment on \( k \) enhancement. Aligned GnP are further found to be 3 times as effective in enhancing \( k \) as in the randomly oriented configuration. Enhancement in \( k \) is correlated with the alignment of PE lamellae and GnP through wide-angle X-ray scattering and polarized Raman spectroscopy. At the maximum applied strain of 400% and using 10 wt% GnP, a composite \( k \) of 5.9 W mK\(^{-1}\) is achieved. These results demonstrate the great potential of simultaneous alignment effects in achieving high \( k \) polymer composites.

Polymer heat exchangers1,2 based on polymers such as polyethylene and polypropylene are widely used in applications including water desalination,3,4 solar energy harvesting,5 automotive control units6 and micro-electronics cooling.7–10 Polymers offer several advantages such as lower cost and weight which make them more economically competitive compared with metallic heat exchangers.7 Polymeric materials, however, have much lower intrinsic thermal conductivity,11 \( k \) (<0.5 W mK\(^{-1}\)) compared to metals (>20 W mK\(^{-1}\)) which limits their more widespread applicability in thermal management technologies. As means to achieve high \( k \) polymers, the alignment of polymer chains has emerged as a new and growing field of research. Recently, \( k \) of a single PE fiber with ultra-aligned PE chains was measured to be 104 W mK\(^{-1}\) almost 200 times12 larger than \( k \) of bulk PE (<0.5 W mK\(^{-1}\)). In another report, the effect of the alignment of polymer chains was used to achieve a thermal conductivity of \(~16\) W mK\(^{-1}\) in polyethylene films at draw ratios approaching \(~100\).13 Similarly, the alignment of filler material has been explored in the past to enhance \( k \). In recent work, magnetic GnP–Fe\(_3\)O\(_4\) hybrids at volume fractions of less than 1% were aligned through a magnetic field in epoxy composites.14 Aligned GnP–Fe\(_3\)O\(_4\) hybrids were found to enhance epoxy \( k \) by almost 40% relative to epoxy with randomly oriented GnP. In this work, only the GnP were aligned. Other studies have also explored the alignment of GnP through an electric field,15,16 flow-assisted alignment17 as well as self-alignment.18–20 Large enhancements in \( k \) through the alignment of either the PE chains or filler material suggest that by combining the \( k \)-enhancements through the alignment of both the polymer chains and the GnP, even larger \( k \) values can be achieved.

Graphitic nanoplatelets (GnP) with the thickness in the range of \(~10–100\) nm are excellent heat conductors with in-plane \( k \) \sim 1500 W mK\(^{-1}\) at room temperature.21–23 GnP also show lower thermal boundary resistance (TBR)24 with polymers relative to carbon nanotubes (CNTs). TBR between CNTs and the polymer matrix is estimated to be in the range of \(10^{-8}–10^{-7}\) m\(^2\) K W\(^{-1}\),25 the corresponding value for GnP is in the range of \(10^{-9}\) m\(^2\) K W\(^{-1}\).24 This lower TBR has led to a significant growth in the use of GnP as the filler material to increase the thermal conductivity of a polymer. While in-plane \( k \) of GnP is very high (\(~1500\) W mK\(^{-1}\)), it should be noted that the through thickness value is much lower of the order of \(~10–20\) W mK\(^{-1}\).26 Alignment achieves full advantage of the high in-plane \( k \) of GnP. Similarly, by aligning polymer chains, highly efficient heat transfer along their chain axis can be achieved.

In this work, the effectiveness of such simultaneous alignment in enhancing the thermal conductivity is explored. The alignment of both the polymer chains and embedded GnP is achieved through the application of mechanical strain to the PE–GnP composites. Mechanical strain is known to align the polymer chains in crystallites along the direction of applied

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strain enhancing $k$ along it (Fig. 1a and b). Similarly, strain has also been shown to align the embedded filler material (Fig. 1c) such as carbon nanotubes. The alignment of GnPs in this work is characterized using laser scanning confocal microscopy and polarized Raman spectroscopy. The alignment of PE crystallites is measured using wide-angle X-ray scattering (WAXS) analysis.

While GnPs are chosen both because of their high intrinsic $k$ and better thermal interaction with the polymer matrix, PE is chosen as the base polymer because it exhibits a large change in $k$ upon alignment. The GnPs used in this work are about $\sim 5 \mu m$ (Fig. 2a) in lateral size and $\sim 60$ nm in average thickness. They were produced through mechanical cleavage of raw graphite and have relatively low defect density. This is seen in the Raman analysis as a low intensity of the D peak (Fig. 2b). The oxygen content of the used GnPs was measured to be less than 2.9% through X-ray photoelectron spectroscopy (Fig. 2c). The polymer matrix used in this work was high density polyethylene with a melt index of 2.2 g per 10 min and with a molecular weight of $M_n \sim 10,600$. PE–GnP composites were prepared by melt-compounding PE pellets and GnP nanopowder together in a DSM-5 micro-compounder. A mixing time of 40 min and a mixing temperature of 200 °C were used while preparing the composite through melt-compounding. The resulting nanocomposite was then compression-molded to achieve the composite samples with a thickness of $\sim 1$ mm.

GnP used in this work are not functionalized; the interaction between GnPs and the polymer matrix is therefore through van der Waals forces.

Mechanical strain was applied to the composite samples using a motorized slide to induce the alignment of the graphene nanoflakes and polymer chains. The use of low draw rates ($\sim 40 \mu m \ min^{-1}$) coupled with heating the sample to 60–70 °C during the drawing process prevented brittle failure and allowed total strains of up to 400% to be achieved. Fig. 2d and e show an unstrained and strained PE–GnP composite, respectively.

The thermal conductivity of both the unstrained and strained composites was measured through the measurement of the thermal diffusivity ($\alpha$), specific heat capacity ($c_p$) and density ($\rho$), using the relationship $k = \rho c_p \alpha$. The specific heat capacity of composites was measured using a TA instruments Q-2000 differential scanning calorimeter. The density of samples was measured using Archimedes’ principle. Thermal diffusivity was measured using the Angstrom method by applying a sinusoidal heat signal at one end of the sample and measuring the temperature ($T$) response at two different locations along the sample. The accuracy of the measurement set-up was established through the good agreement between the measured $k$ of the pristine PE ($0.5 \ W \ mK^{-1}$, Fig. 3) and that reported in the literature.

The measured thermal conductivities of pure PE and PE–GnP composites as a function of applied strain ($\varepsilon = \Delta l / l_o$, where $\Delta l$ is the change in sample length and $l_o$ is the original length) are shown in Fig. 3. The thermal conductivities of both the pure PE sample and the composite samples increase with applied strain due to the alignment of PE chains and embedded GnPs. An extraordinary increase in $k$ for aligned PE–GnP composites can be observed. At the highest strain of $\varepsilon = 4$, a $k$ value of 5.9 W mK$^{-1}$ is achieved for the aligned PE–GnP composite with 10 wt% GnP. This represents a $k$-enhancement of almost 1100% relative to the unoriented pure PE matrix ($k \sim 0.5 \ W \ mK^{-1}$). In addition, $k$ of the 7 wt% composite at a strain of $\varepsilon = 4$ is measured to be 5.1 W mK$^{-1}$, almost 5 times higher compared to the value for the unstrained composite ($\varepsilon = 0$) $k = 1.1 \ W \ mK^{-1}$. These results show significant enhancements in $k$ of the aligned versus the unoriented composites and point to the great potential of alignment effects in yielding high $k$ polymeric materials.
results provide the first evidence that simultaneous alignment effects can provide large enhancements in composite $k$.

While the above values provide an estimate of the absolute $k$-enhancement in aligned systems, direct evidence of the beneficial effect of aligned GnP's in enhancing $k$ is gained by comparing the slopes of the best-fit lines for the three samples. For the pure PE sample, a slope of $k$-increase with respect to applied strain is found to be $(d k / d \varepsilon)_{\text{pure-PE}} \approx 0.67$. In the pure PE sample, this $k$ enhancement with respect to the applied strain is solely due to the alignment of PE chains and has been well studied before. As 7 wt% GnP's are added to the sample, the slope increases to $(d k / d \varepsilon)_{\text{composite}} \approx 1.03$. In the composite sample, $k$-enhancement with applied strain is through the alignment of both PE chains and GnP's. The larger slope is a direct manifestation of the beneficial effect of aligned GnP's in enhancing composite $k$ (if the aligned GnP's did not cause any additional increase in $k$ relative to randomly oriented GnP's, the only effect responsible for $k$-increase with respect to applied strain in the composite would be the alignment of PE chains, this would have resulted in the same slope as that of the pure PE sample assuming that the alignment of the PE was the same in the pure and composite samples). As the weight fraction of GnP's in the composite is increased to 10 wt%, the slope increases to $(d k / d \varepsilon)_{\text{composite}} \approx 1.24$. This points to the enhanced effect of alignment of larger quantities of the GnP's within the composite. This almost a factor of two higher rate of $k$-increase with applied strain in the PE-GnP (10 wt%) composite relative to the pure PE sample is due to the cumulative contributions of the alignment of PE chains and GnP's to $k$ enhancement and clearly points to the promise of such simultaneous alignment in enhancing composite $k$.

Quantitative estimate of the effectiveness of aligned versus randomly oriented GnP's can also be achieved by comparing the difference between the thermal conductivity values of the pure PE sample and composite at the same strain. At zero strain, the difference between the two is due to randomly oriented GnP's. At higher strains, the difference between the two $k$ values is through GnP's which are now aligned providing a direct comparison of the $k$ enhancement between aligned GnP's and randomly oriented GnP's. For the 7 wt% sample, the enhancement at zero strain is $k_{\varepsilon=0}^\text{composite} - k_{\varepsilon=0}^\text{PE} = 0.5$ W m K$^{-1}$. As the strain is increased to $\varepsilon = 3$, the difference becomes $k_{\varepsilon=3}^\text{composite} - k_{\varepsilon=3}^\text{PE} = 1.7$ W m K$^{-1}$. Comparing these two enhancement values, aligned GnP's are found to be more than 3 times as effective as randomly oriented GnP's in enhancing the thermal conductivity. Similar results are found comparing enhancements for the 10 wt% sample. This analysis clearly demonstrates the increased effectiveness of GnP's in enhancing $k$ in the aligned form versus randomly oriented configuration.

We further compare the effectiveness of mechanical strain mediated alignment in enhancing $k$ with that through other means such as electric field. In Table 1, we compare the results of $k$-enhancement through GnP's aligned by different techniques. In the first study in Table 1, GnP's with volume fractions of up to $\sim 0.08\%$ were aligned through an electric field in epoxy composites. At low volume fractions $\sim 0.6\%$, $k$-enhancement through aligned GnP's was almost twice that due to the randomly oriented case. At higher volume fractions $\sim 1\%$, however, the effectiveness of the electric field in aligning GnP's diminished due to the presence of.

![Image](https://example.com/image.png)

**Fig. 3** Measured thermal conductivities of pure PE and PE–GnP composites as a function of applied strain. A highest $k$ of 5.9 W m K$^{-1}$ is achieved representing a 12-fold improvement over pristine PE. Increasing slopes of linear fits point to the role of aligned GnP's in enhancing $k$.

### Table 1: Thermal conductivity enhancement through alignment effects

<table>
<thead>
<tr>
<th>Sample composition</th>
<th>$k$ (W m K$^{-1}$)</th>
<th>Fraction</th>
<th>Alignment method</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aligned GnP/epoxy</td>
<td>0.45</td>
<td>1.05 vol%</td>
<td>Electric field</td>
<td>15</td>
</tr>
<tr>
<td>Aligned-interconnected GnP/PDMS</td>
<td>2.13</td>
<td>0.92 vol%</td>
<td>Freeze casting of self-aligned GO' liquid crystal</td>
<td>18</td>
</tr>
<tr>
<td>Aligned GnP/epoxy</td>
<td>0.6</td>
<td>1 vol%</td>
<td>Magnetic field</td>
<td>14</td>
</tr>
<tr>
<td>Aligned GnP/epoxy</td>
<td>0.37</td>
<td>0.52 vol%</td>
<td>Magnetic field</td>
<td>34</td>
</tr>
<tr>
<td>Aligned GO’/cellulose</td>
<td>6.17 (3.56)</td>
<td>30 wt% (10 wt%)</td>
<td>Self-alignment</td>
<td>19</td>
</tr>
<tr>
<td>Aligned GnP/PDMS</td>
<td>6.05 (1.04)</td>
<td>20 vol% (7.5 vol%)</td>
<td>Electric field</td>
<td>16</td>
</tr>
<tr>
<td>Aligned GnP/PVDF</td>
<td>10 (1.06)</td>
<td>25 vol% (5 vol%)</td>
<td>Flow induced</td>
<td>17</td>
</tr>
<tr>
<td>Aligned GnP/aligned PE</td>
<td>3.9</td>
<td>10 wt% (6.2 vol%)</td>
<td>Mechanical strain</td>
<td>This work</td>
</tr>
</tbody>
</table>

GnP agglomerates, leading to smaller $k$-enhancements. The alignment of only low volume fractions (<1 vol%) limited the highest $k$ value achieved to 0.45 W mK$^{-1}$. In this work, we find that $k$-enhancements for 7 wt% and 10 wt% GnP content are proportional to the volume fractions, indicating no decrease in the ability of mechanical strain to align greater quantities of GnP. This points to the advantages of mechanical strain relative to an electric field in achieving higher thermal conductivity values. The results suggest that even higher $k$ values can be achieved through alignment of larger quantities of GnP using mechanical strain induced alignment.

Aligned GnP are also found to be more effective in enhancing $k$ relative to aligned CNTs through comparison with previously reported results on aligned PVA (polyvinyl alcohol)–CNT composites. For the 10 wt% CNT composite and at a strain of 4.5, aligned CNTs were found to enhance thermal diffusivity by $\alpha_{\text{PVA-CNT}}^{4.5} - \alpha_{\text{PVA}}^{4.5} = 0.55$ mm$^2$ s$^{-1}$ relative to pure PVA (at the same strain of 4.5) in the PVA–CNT composites. In this work, the same comparison for 10 wt% GnP composite yields an enhancement of $\alpha_{\text{PE-GnP}}^{4.0} - \alpha_{\text{PE}}^{4.0} = 1.13$ mm$^2$ s$^{-1}$ through the use of aligned GnP. Aligned GnP are thus found to be twice as effective in enhancing $k$ as aligned CNTs.

The above comparisons shed light on the advantages of mechanical strain and the use of GnP for alignment mediated thermal conductivity enhancement. We next show that simultaneous alignment of polymer lamellae and GnP provides higher $k$ enhancement than achievable through alignment of GnP alone. In Table 1 we further compare $k$-enhancement through the alignment of GnP induced by techniques such as magnetic field, flow processes and self-alignment with the present work. For each study, we present the highest $k$ achieved, and also the $k$ values at volume fractions close to the 6.2 vol% used in this study (volume fraction for the 10 wt% composition is ~6.2 vol%, determined through density measurements). Comparison of these studies clearly shows that the $k$ value achieved in this work (5.9 W mK$^{-1}$) through simultaneous alignment of PE chains and GnP (10 wt%) is significantly higher than achieved through the alignment of GnP alone at similar compositions. These comparisons shed light on the great potential of simultaneous alignment effects in developing high $k$ polymeric composites.

To analyze the influence of GnP alignment on the composites thermal properties, the degree of its alignment must be determined. The alignment of GnP within the nanocomposite was analyzed in this work through the use of laser scanning confocal microscopy (LSCM) and polarized Raman spectroscopy. While LSCM provides a visual understanding of GnP alignment, polarized Raman allows a quantitative estimate of the GnP orientation. LSCM generates optical sections as thin as 300 nm axially by using reflected light to create images while blocking out any out-of-focus light. By collecting a series of these optical sections along the optical axis, one can generate a 3D reconstruction of a volume within an intact specimen. In this work, we use a Leica SP8 laser scanning confocal microscope with a 561 nm diode pumped solid state (DPSS) laser. The samples are imaged with a 63 × 1.4 oil immersion objective with a pinhole aperture at 0.2 airy units (AU) and voxel dimensions of 120 nm × 120 nm × 120 nm. The image volume for each sample was 246 μm × 246 μm × 10 μm with three images per sample. Fig. 4 shows the LSCM maximum intensity projections of GnP in the unstrained nanocomposite sample (7 wt% GnP) and the same composition sample with strains ($\varepsilon$) of 1, 2, 3 and 4. While GnP are seen to be randomly oriented in Fig. 4a for the unstrained sample, increasing alignment of GnP with increasing applied strain is clearly visible in Fig. 4b, c, d and e. These LSCM images visually demonstrate the alignment of GnP upon application of strain.

The absolute alignment of GnP in terms of their angle of orientation is also measured using polarized Raman spectroscopy. Raman spectra were obtained using a Renishaw inVia Reflex Spectrometer System, equipped with a laser with wavelength $\lambda = 532$ nm. To measure orientation using polarized Raman spectroscopy, we used the 7 wt% graphene/PE sample with a strain of $\varepsilon = 4$. The sample was set up with the laser illuminating along the $X$ direction, as shown in Fig. 5(a). The orientation of GnP is described by the angle $\phi_i$ between the surface normal of the composite sample and the surface normal of an arbitrary GnP flake (Fig. 5(a)). The angle between the incident laser polarization and the $Y$ axis when viewed along the $X$ axis is defined as $\Phi$ (Fig. 5(b)). The scattered radiation was not polarized in this work. During the measurement, the incident laser polarization was rotated, while the sample was kept fixed. The intensity of the Raman 2D band ($I_{2D}$) as a function of $\Phi$ measured at five areas is shown in Fig. 5(c). For each area, the intensity values at different $\Phi$ were normalized to the maximum. To quantify the orientation of GnP, the model in ref. 38 is modified to account for the absence of polarization of scattered radiation in this work. The modified

![Fig. 4 Confocal microscopy images of GnP in the 7 wt% PE–GnP composite sample for different applied strains ($\varepsilon$) of (a) 0, (b) 1, (c) 2, (d) 3 and (e) 4. Increasing alignment of GnP with increasing strain is clearly visible in the above maximum intensity projections. Scale bar = 20 μm.](image-url)
model is used to express the Raman intensity as a function of angle $\Phi$ and the parameter $\langle P_2(\cos \phi_i) \rangle$ as below,

$$I_{2D}(\Phi) = I_0 \left\{ \frac{2}{3} + \langle P_2(\cos \phi_i) \rangle \left( -\frac{2}{3} + \cos^2 \Phi \right) \right\}$$

where $I_0$ is the amplitude. $\langle P_2(\cos \phi_i) \rangle$ describes the average orientation of GnPs through the relation $\langle P_2(\cos \phi_i) \rangle = \{3\langle \cos^2 \phi_i \rangle - 1\}/2$, where $\langle \cos^2 \phi_i \rangle$ is the average value of $\cos^2 \phi_i$. By fitting the measured result to eqn (1), $\langle P_2(\cos \phi_i) \rangle$ can be obtained, as an indication of the spatial orientation of GnPs as shown in Fig. 5c. The values of $\langle P_2(\cos \phi_i) \rangle$ are 0 and 1 for random and completely aligned orientations, respectively (corresponding values of $\langle \cos^2 \phi_i \rangle$ are 0.33 and 1.0). The value of $\langle \cos^2 \phi_i \rangle$ obtained by fitting eqn (1) to the measured intensity data is achieved to be 0.47 (Fig. 5c). This large value ($\langle \cos^2 \phi_i \rangle \sim 0.47$) indicates a high level of GnP alignment in the strained composite sample. We find that this orientation level of GnPs approaches the alignment level of PE crystallites discussed next.

While the above measurements shed light on the alignment of GnPs, the alignment of crystalline PE chains is measured through wide-angle X-ray diffraction measurements (WAXS). Through WAXS, the average relative orientation of the PE chains in crystallites with respect to the stretch direction is measured in terms of $\langle \cos^2 \gamma \rangle$ (the average value of $\cos^2 \gamma$) where $\gamma$ is the angle between the PE chain axis and draw direction. The WAXS patterns in the pure PE sample obtained for different strains are shown in Fig. 6a–c. The 2D patterns clearly show alignment effects; upon stretching, the rings become arcs in WAXS spectra. The obtained intensities from these plots are integrated to compute the average orientation $\langle \cos^2 \gamma \rangle$, giving the alignment of PE chains in crystallites with respect to the stretch direction. The value of $\langle \cos^2 \gamma \rangle$ is presented in Fig. 6(d) where the increasing value of $\langle \cos^2 \gamma \rangle$ with increasing strain indicates increasing alignment. A high degree of PE alignment at the largest strain ($\varepsilon = 4$) is indicated by a large value of $\langle \cos^2 \gamma \rangle = 0.87$.

WAXS measurements were also performed on the 7 wt% composite sample for strain $\varepsilon = 4$ to compare PE alignment levels with the pure PE samples. The value of $\langle \cos^2 \gamma \rangle$ was identical to the one obtained for the pure PE sample, indicating that PE chain orientation is identical in the composite and the pure PE sample for the same applied strain. This also implies that the small GnP content of 4.4 vol% (for the 7 wt% sample) does not impact the alignment of polymer lamellae.

We further use Raman and WAXS alignment measurements to extract the relative contributions of aligned PE chains and GnPs to total $k$-enhancement. For 10 wt% filler content, increasing the strain from $\varepsilon = 0$ to $\varepsilon = 2$ leads to a total $k$-enhancement of 2.9 W mK$^{-1}$ (Fig. 3). The contribution of alignment of PE chains to this is considered to be the same as in the pure PE sample, 1.1 W mK$^{-1}$ from the difference between the $k$ values of the pure PE sample for $\varepsilon = 2$ and $\varepsilon = 0$. This is due to the same PE alignment in both the composite and the pure PE sample (as shown by the above WAXS measurements). The further enhancement induced in composite $k$ due to the alignment of GnPs is thus 1.8 W mK$^{-1}$ (obtained by subtracting the PE contribution from total enhancement). For a strain of $\varepsilon = 3$, the total $k$-enhancement reaches 3.9 W mK$^{-1}$. The contributions of the aligned PE chains and aligned GnPs to this are estimated to be 1.9 and 2.0 W mK$^{-1}$, respectively. These near equal contributions of the alignment of PE and GnPs to total $k$-enhancement lead to an almost doubling of the slope of $k$-increase with the strain for the 10 wt% composite relative to the pure PE sample and
yield overall $k$-value of 5.9 W mK$^{-1}$. These results provide the first proof that strain induced higher $k$-enhancement via mechanical stretching can occur through combined contributions of the alignment of the polymer matrix and the embedded graphitic nanoplatelets and may provide new pathways for achieving high $k$ polymeric materials.

It should be noted that alignment levels presented in this work can be realized on a bulk scale through widely used manufacturing processes such as polymer extrusion. In the past, extrusion has been found to align both polyethylene$^{40}$ and polypropylene$^{41,42}$ chains. The alignment of filler material such as CNTs$^{43,44}$ and GnPs$^{45}$ upon extrusion has also been reported. The simultaneous alignment of polymer chains and high $k$ filler material such as GnPs through composite extrusion may lead to inexpensive and scalable methods for processing of high thermal conductivity polymer composites.

In summary, we have demonstrated the effect of the strain induced simultaneous alignment of PE chains and GnPs on the $k$-enhancement of PE-GnP composites. These composites were prepared by melt-compounding PE pellets and GnPs. GnP weight fractions of up to 10% were included in the study. Both laser scanning confocal microscopy and polarized Raman spectroscopy demonstrated a high level of GnP alignment in strained samples. Similarly, wide-angle X-ray scattering measurements revealed the alignment of PE chains. At the highest strain of 400% and for 10 wt% GnP composition, an overall thermal conductivity of 5.9 W mK$^{-1}$ was achieved representing an 1100% enhancement over the $k$ of unoriented pristine PE (0.5 W mK$^{-1}$). Aligned GnPs were found to be 3 times as effective in enhancing $k$ as randomly oriented GnPs. A similar comparison with aligned CNTs showed the aligned GnPs to be twice as effective as aligned CNTs in enhancing composite $k$. These results shed light on the great potential of simultaneous alignment effects in enhancing $k$ and provide new avenues for developing high $k$ polymeric composites.

Conflicts of interest

There are no conflicts of interest.

References


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