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Effect of two types of graphene nanoplatelets on the physico–mechanical properties of linear low–density polyethylene composites

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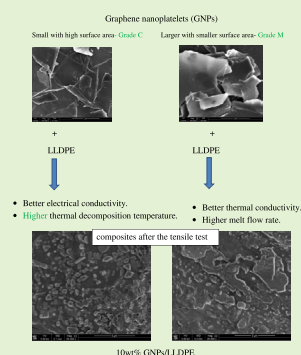[3]Innovation & Product Development Manager, Qatar Petrochemical Company (QAPCO)
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Abstract The influence of two types of graphene nanoplatelets (GNPs) on the physico-mechanical properties of linear low-density polyethylene (LLDPE) was investigated. The addition of these two types of GNPs – designated as grades C and M – enhanced the thermal conductivity of the LLDPE, with a more pronounced improvement resulting from the M-GNPs compared to C-GNPs. Improvement in electrical conductivity and decomposition temperature was also noticed with the addition of GNPs. In contrast to the thermal conductivity, C-GNPs resulted in greater improvements in the electrical conductivity and thermal decomposition temperature. These differences can be attributed to differences in the surface area and dispersion of the two types of GNPs.

Keywords Graphene nanoplatelets, Polyethylene, Physico-mechanical properties

Graphene nanoplatelets (GNPs)

Small with high surface area- Grade C   Larger with smaller surface area- Grade M

+ LLDPE   + LLDPE

• Better electrical conductivity.   • Better thermal conductivity.
• Higher thermal decomposition temperature.   • Higher melt flow rate.

composites after the tensile test

10wt% GNPs/LLDPE

Introduction

Graphene and graphene nanoplatelets (GNPs) have been used extensively in various applications due to their exceptional properties, including high electrical and thermal conductivities, and excellent mechanical strength and stiffness.[1–4] Compared to single-layer graphene, GNPs can be produced more easily and starting from natural graphite precursors, yielding large quantities of GNPs at a lower cost.[5–7] Graphene and GNP polymer nanocomposites have attracted much scientific and industrial attention because their physico-mechanical properties can be significantly enhanced with the addition of relatively small quantities of graphene.[8–11] In particular, the thermal conductivity of polymers can be greatly increased by the addition of GNPs because the two-dimensional planar structure of GNPs can reduce the phonon scattering at the polymer–nanofiller interface compared to one-dimensional additives (e.g. carbon nanotubes).[12] Other properties of GNP-based nanocomposites include ease of processing and fabrication, high thermal decomposition temperature, enhanced electrical conductivity and tensile strength, which are also crucial for mass market production and specific commercial applications. These properties depend on filler agglomeration, dispersion, and the adhesion/contact between the filler materials and the matrix.

Several processing methods for incorporating and dispersing graphene and GNPs into polymer matrices have been studied,[13–15] among which, melt mixing is one of the most common due to its low cost for mass production of thermoplastics and thermoplastic composites.[16] Under melt mixing, the constituent materials experience high levels of mechanical shear, which plays an important role in the materials produced.[17] Linear low-density polyethylene (LLDPE) is routinely processed via melt mixing and has significant commercial importance due to its good balance of mechanical properties and processability compared to other types of polyethylene (PE).[18] Addition of fillers can enhance the physical and mechanical properties of PE for various applications, such as electromagnetic reflective materials and high voltage cables. The improvements are linked mostly to the change in crystallinity, tensile strength, thermal conductivity, electrical properties and thermal stability.[17–19] In this paper, we investigate the effect of adding two grades of GNPs in contents ranging from 1 to 10 wt% in an LLDPE matrix. We report the detailed characterization of the physical and mechanical properties of the resulting LLDPE nanocomposites, and the effect of the GNP size and morphology on these properties. The paper

presents the way to tailor important properties such as thermal and electrical conductivities by changing the percentage and size of the GNPs. We use common industrial processing technique (melt processing) for powder polymer (without additives) which is essential for mass production.

Experimental

Materials

Polymer matrix

LLDPE in powder form (density 0.918 g/cm³) was supplied by Qatar Petrochemical Company (QAPCO), Qatar. Prior to the melt processing, 0.4 g of phenolic stabilizer was added to each 1 kg of LLDPE to prevent degradation during the high temperature processing.

Fillers

Two grades of GNPs were used: C grade (C-GNPs), and M grade (M-GNPs), each supplied by XG Sciences, USA. The properties specified by the supplier for each type of GNP are given in Table 1.

Preparation of C-GNPs/LLDPE and M-GNPs/LLDPE

LLDPE composites reinforced with 1, 2, 4, 6, 8, and 10 wt% of C-GNPs or M-GNPs were processed using a five-stage Brabender twin screw extruder at a screw speed of 100 rpm and a feeder speed of 100 rpm. The temperatures of the

Table 1   Technical specifications of C and M grade GNPs

	Grade C (C-GNPs)	Grade M (M-GNPs)
Number of layers	18–24	2–15
Thickness (nm)	Few nanometers	6–8
Diameter (µm)	1–2	5
Surface area (m²/g)	300–500	120–150

processing zones were in the range 190-230 °C. Mixtures of the C-GNPs/LLDPE and M-GNPs/LLDPE were extruded into filaments, which were then cooled in water, and granulated into pellets. Figure 1 shows a schematic diagram of the nanocomposite melt-mixing process. The extruded pellets were subsequently hot pressed into plaques 5 cm × 5 cm × 0.5 cm via compression molding at a temperature of 170° and a pressure of 1654.7 Bar for a period of 20 min, after which the plaques were cooled to room temperature.

Characterization

Environmental scanning electron microscopy

A Quanta 200 (FEI, USA) environmental scanning electron microscope (ESEM) was used to observe the size, shape, and morphology of the GNPs before and after incorporation into GNPs/LLDPE nanocomposites. Ten milligram of each type of GNPs was dispersed in 10 ml of acetone, the solution was sonicated for 30 min then analyzed by the ESEM for imaging. Sonication was done for 30 min after dispersion of 10 mg of GNPs in 10 ml of acetone. GNPs/LLDPE nanocomposites were also inspected using the ESEM to image the failure surfaces of tensile specimens after mechanical testing.

Melt flow rate

The melt flow rates of C-GNPs/LLDPE and M-GNPs/LLDPE composites were measured using a LMI 4004 Melt Flow Indexer (Dynisco, USA) according to ASTM D1238-10 at a temperature of 190 °C and load of 21.6 kg. Five replicate samples were tested and properties are reported as the mean values.

Thermal conductivity

The thermal conductivities of the C-GNPs/LLDPE and M-GNPs/LLDPE composites were measured at room temperature using a TPS 2500S instrument (Hot Disk, Sweden). Four tests were conducted and the mean values reported for each material.
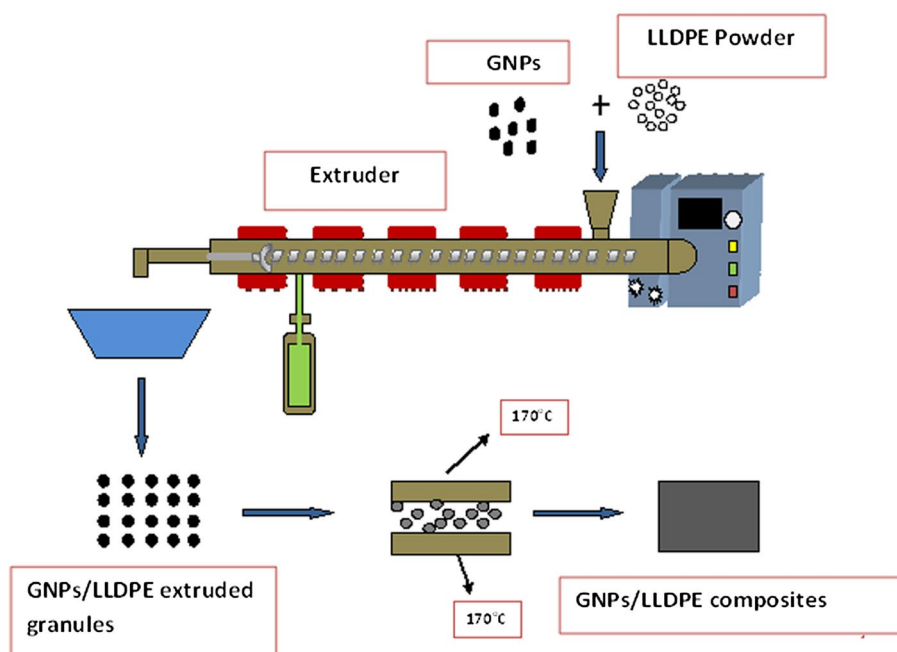

Figure 1   Schematic diagram of melt mix processing of GNPs/LLDPE nanocomposites

Electrical conductivity

A Keithley Model 2400 electrometer was used to measure the electrical resistance of C-GNPs/LLDPE and M-GNPs/LLDPE composites at room temperature using the four-point probe method. Silver paint was coated on the upper and lower surfaces of the prepared plaques to make good electrical contact. Five tests were conducted and the mean values reported for each material. The electrical conductivity (σ) of the composite was calculated using the average resistance, the sample thickness and the area of the electrodes.[20]

Thermogravimetric analysis

Thermogravimetric analysis (TGA) of the C-GNPs/LLDPE and M-GNPs/LLDPE composites was conducted using a PerkinElmer Pyris 6 under a nitrogen atmosphere from room temperature to 700 °C at a heating rate of 10 °C/min. Two tests were conducted and the mean values were reported.

Mechanical testing

Tensile testing of C-GNPs/LLDPE and M-GNPs/LLDPE composites was performed at room temperature according to the ASTM D638-10 using a Lloyd universal tensile testing machine. Five replicate samples were tested and properties are reported as the mean values.

Results and discussion

Morphology of C-GNPs, M-GNPs, and LLDPE nanocomposites

The ESEM images of the C-GNPs and M-GNPs are shown in Figure 2(a) and (b), respectively, after dispersion in acetone and solution casting. As expected (based on the specifications in Table 1), C-GNPs exhibited smaller in-plane dimensions compared to M-GNPs. ESEM images of the failure surfaces of pure LLDPE, 10 wt% C-GNPs/LLDPE, and 10 wt% M-GNPs/LLDPE composites after tensile testing are shown in Figure 3(a)–(c), respectively. The ductile behavior of pure LLDPE is reflected in the more tortuous failure surface, as compared with the flat failure surfaces associated with the more brittle behavior of the nanocomposites. The C-GNPs appear to be better dispersed in the LLDPE matrix, as shown in Figure 3(b), when compared with the M-GNPs, which appear more agglomerated in Figure 3(c); as confirmed by further characterization reported herein).

Melt flow rate

Melt flow rate (MFR) was used to assess the suitability of potential post-processing and fabrication techniques for the various LLDPE nanocomposites. A pronounced reduction in the MFR was observed with the addition of the GNPs, as shown in Figure 4, which may be attributed to the hindrance of flow by the additives and the internal friction between the additive and the matrix. C-GNPs had a stronger effect on MFR, with a 43% reduction in MFR at the highest loading of 10 wt%, compared to the 35% reduction at 10 wt% M-GNPs. The stronger effect of C-GNPs can be attributed to their higher surface area, which causes more mechanical interaction between the GNPs and the LLDPE, leading to the reduction in the movement between the layers.[21] The higher MFR of M-GNPs
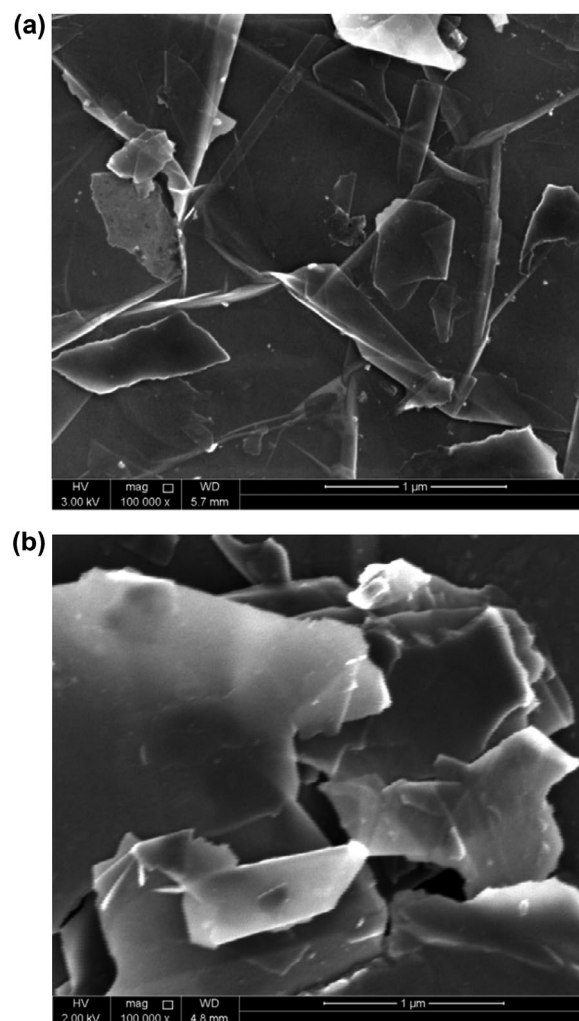

(a)

(b)

Figure 2 Typical ESEM images of *a* C-GNPs and *b* M-GNPs

nanocomposites indicates their better processability, which is favorable for large-scale production and industrial applications (noting that both grades of GNPs have the same price).

Thermal conductivity

The thermal conductivities of the C-GNPs/LLDPE and M-GNPs/LLDPE composites are plotted in Figure 5 as functions of the GNP content. The thermal conductivity continuously increased as the content of GNPs increased, with the maximum of 10 wt% resulting in an increase of 31% for C-GNPs and 92% for M-GNPs. These results are consistent with the literature, where the thermal conductivity of composites can reportedly be affected by the constituent materials, the filler content, and the size and shape of the filler.[22, 23]

GNP fillers provide a large surface area for contact with the polymer matrix and form a 2D path for phonon transport, which results in increased thermal properties of the composites. The increased thermal conductivity of the GNPs/LLDPE nanocomposites can be attributed to the formation of conductive networks by C-GNPs or M-GNPs in the LLDPE matrix. At low loadings of nanofiller (i.e. <2 wt%), the increase in thermal conductivity is small because isolated GNPs do not form a continuous network and a conductive pathway
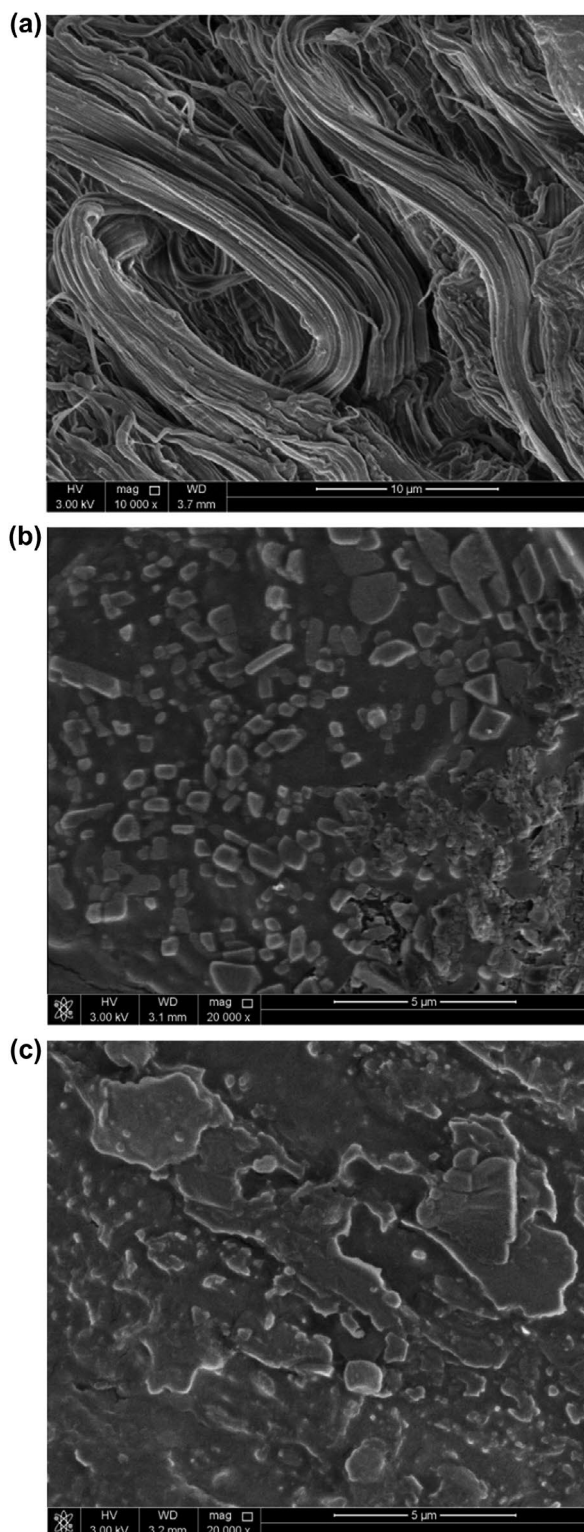
(a)

(b)

(c)

Figure 3 Typical ESEM images of *a* pure LLDPE, *b* 10 wt% C-GNPs/LLDPE composite, and *c* 10 wt% M-GNPs/LLDPE composite after tensile testing

Figure 4 Effect of C-GNPs and M-GNPs content on the melt flow rate of LLDPE composites

Figure 5 Effect of C-GNPs and M-GNPs on the thermal conductivity of LLDPE composites

The apparent formation of conductive pathways at lower weight fractions of M-GNPs (at approx. 2 wt% M-GNPs) composites is consistent with the larger diameter, and more agglomeration of M-GNPs. The improvement in thermal conductivity due to agglomeration of graphite platelets was reported by Tua and Ye,[24] and was similarly attributed to the formation of conductive pathways. The lower thermal conductivity of the C-GNPs is also consistent with the stronger phonon scattering that can be expected for a given weight fraction of C-GNPs, which will result in more interfacial surface area compared to M-GNPs. Phonon scattering, which reduces thermal conductivity, can result from poor interactions and defects between the matrix and fillers.[25]

Electrical conductivity

The electrical conductivity of C-GNPs/LLDPE and M-GNPs/LLDPE composites with different GNP contents is shown in Table 2. There was a considerable increase in the electrical conductivity as the C-GNP and M-GNP contents increased, which is consistent with reports[26, 27] for other nanocarbon–polymer nanocomposites. The electrical conductivity of the composites

through the polymer matrix. At these low GNP loadings, high thermal resistance between the GNPs and LLDPE interface are expected. As the content of GNPs in the polymer matrix increases, the GNPs are more likely to be in closer contact and the thermal conductivity increases more rapidly.
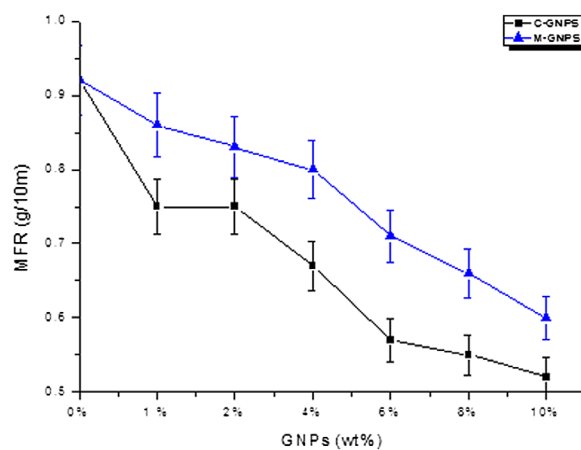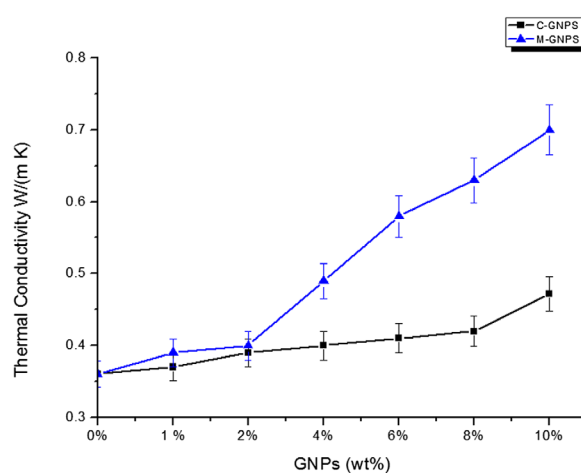
Table 2   Electrical conductivity of LLDPE composites with C-GNPs and M-GNPs

| Samples	Electrical conductivity (S/cm) (C-GNPs)	Electrical conductivity (S/cm) (M-GNPs)
LLDPE	$2.814 \times 10^{-12} \pm 8.36 \times 10^{-14}$	$2.814 \times 10^{-12} \pm 8.36 \times 10^{-14}$
LLDPE/1% Graphene	$7.98 \times 10^{-10} \pm 7.071 \times 10^{-12}$	$6.3 \times 10^{-10} \pm 5.00 \times 10^{-12}$
LLDPE/2% Graphene	$1.5 \times 10^{-8} \pm 5.77 \times 10^{-10}$	$2.4 \times 10^{-9} \pm 5.77 \times 10^{-10}$
LLDPE/4% Graphene	$2.9 \times 10^{-8} \pm 7.07 \times 10^{-10}$	$8.4 \times 10^{-9} \pm 7.07 \times 10^{-10}$
LLDPE/6% Graphene	$2.8 \times 10^{-7} \pm 5.48 \times 10^{-9}$	$2.5 \times 10^{-8} \pm 4.85 \times 10^{-10}$
LLDPE/8% Graphene	$9.75 \times 10^{-7} \pm 8.37 \times 10^{-9}$	$7.5 \times 10^{-8} \pm 4.86 \times 10^{-10}$
LLDPE/10% Graphene	$4.28 \times 10^{-5} \pm 7.07 \times 10^{-7}$	$1.1 \times 10^{-7} \pm 5 \times 10^{-9}$

increases due to the GNP addition, which impart electrical conductivity via the formation of a conductive network of GNPs.[28] The electrical conductivity of pure LLDPE was 2.81×10^{-11} S/cm, which was increased to a value as high as 4.28×10^{-5} S/cm by the addition of 10 wt% C-GNPs. Incorporation of M-GNPs into the pure LLDPE also improved the electrical conductivity but to a lesser extent (addition of 10 wt% M-GNPs/ increased the conductivity to 1.1×10^{-7} S/cm). It is known that filler geometry plays a role in the electrical properties of composite materials. Fillers with large surface area can have a high electrical conductivity properties compared to low surface area as they can easily form the conductive networks in polymer matrix.[2] Because of the higher surface area and smaller size, C-GNPs/LLDPE composites have higher electrical conductivity compared to M-GNPs/LLDPE composites. The electrical conductivity of both C-GNPs and M-GNPs composites was lower at lower filler contents, which can be attributed to a lack of conductive GNP networks formed in the LLDPE matrix.[29]

Thermal degradation

TGA was used to investigate the thermal stability of C-GNPs/LLDPE and M-GNPs/LLDPE composite granules. The decomposition temperature (T_d) of the composites is plotted as a function of filler content for each type of GNP as shown in Figure 6. TGA revealed that the decomposition temperature increased with increasing filler content in both cases. These results suggest that GNPs can act as effective thermal barriers which help to hinder the degradation of LLDPE[30] by preventing the emission of gaseous molecules, disrupting the oxygen supply during thermal degradation, and the formation of char layers on the surface of the nanocomposite.[30–32] The thermal decomposition temperature increased from 477 °C to a maximum of 489.28 °C with addition of 10 wt% C-GNPs, and to 483.17 °C with 10 wt% M-GNPs. The higher decomposition temperature of C-GNPs composites can be attributed to the higher aspect ratio and more homogeneous distribution of C-GNPs (as shown in Figure 3), which is expected to result in stronger interphase bonding between the matrix and the C-GNPs, in comparison with M-GNPs composites. Similar behavior for montmorillonite ethyl vinyl acetate nanocomposites has been reported by Osman *et al.*[33].

Tensile strength

The tensile strengths of C-GNPs/LLDPE and M-GNPs/LLDPE composites are shown in Figure 7. Modest improvements in tensile strengths (6% at 2 wt% C-GNPs, and 5% at 1 wt% M-GNPs) were observed with the addition of low fractions of GNPs. This improvement with small amounts of additives was also reported by Panaitescu *et al.*[34] who studied ~2 wt%
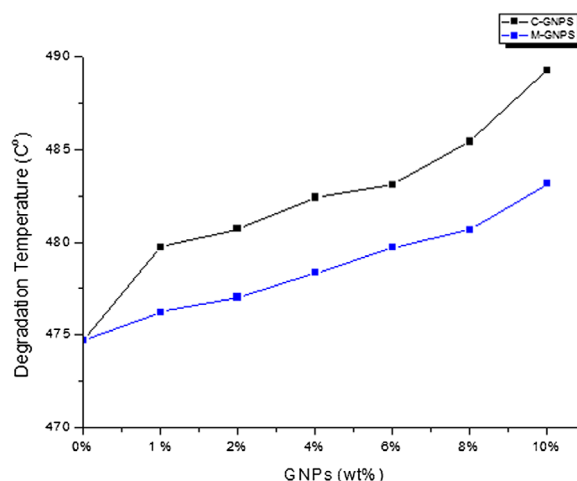
Figure 6   Effect of C-GNPs and M-GNPs addition on thermal decomposition temperature of LLDPE composites
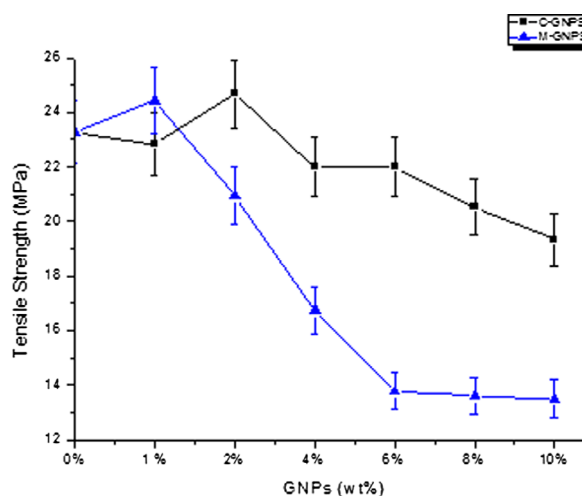
Figure 7   Effect of C-GNPs and M-GNPs loading on tensile strength of LLDPE composites

nano-oxide additives in PE nanocomposites. The improvement in tensile strength has been attributed to good, uniform interfacial adhesion,[18] which avoids stress propagation and indicate that the load is effectively transferred from the matrix to the GNPs.

Higher wt% addition (>2 wt%) led to reductions in tensile strength, and brittle behavior of the materials (see Figure 3). This reduction can be attributed to the agglomeration and lower interfacial bonding between GNPs and the LLDPE, as reported elsewhere.[35] The reduction in tensile strength is more pronounced in M-GNPs composites, which again correlates to

the higher degree of agglomeration[36] of M-GNPs and which contributed to the better thermal conductivity.

Conclusions

A comparative study has been carried out to investigate the effects of two types of GNPs (with different size and surface areas) on the physico-mechanical properties of LLDPE. From the results obtained, the following conclusions can be drawn:

- The addition of GNPs leads to improvement in the thermal conductivity of the composites, with the highest improvement of 31% achieved for C-GNPs composites, and 92% for M-GNPs composites, both at a filler content of 10 wt%. The higher conductivity of M-GNPs was attributed to two factors: (1) agglomeration contributed to the formation of conductive pathways, and (2) lower GNP surface area reduced the scattering of phonons at interfacial defects.
- Both the electrical conductivity and the decomposition temperature increased with increasing GNP content. C-GNPs give superior property enhancement due to better dispersion and the good interfacial contact between the LLDPE and the GNPs.
- The tensile strength of the nanocomposites is slightly increased with the addition of 1 to 2 wt% GNPs, but is subsequently reduced with higher loading of nanofillers. The deterioration in tensile strength is more pronounced in the M-GNPs compared to C-GNPs due to their propensity to agglomerate and their reduced interfacial adhesion with matrix.
- M-GNPs composites demonstrated higher MFR compared to C-GNPs composites, indicating a better ability to flow and hence better processability using common manufacturing techniques such as extrusion and injection molding.
- Depending on the product requirements and market needs, careful consideration needs to be given when selecting among different types of GNPs nanofillers, as their specific properties and morphology (e.g. size, shape, surface area) can have a strong influence on the resulting structure, properties, and the processability of the resulting composites.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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