



Conductive nanocomposites based on polymer with high concentrations of graphene nanoplatelets

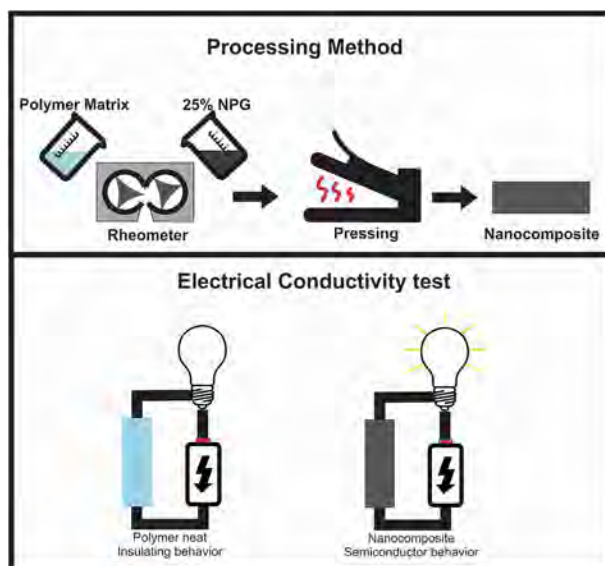
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Abstract

The power density of electronic devices has been progressively increased in the last years, thus raising the urgent demand for the efficient systems of electrical conductivity. In a sense a promising strategy to increase the electrical conductivity of polymer composites is to construct interconnected three-dimensional graphene nanoplatelets networks. Due to the variety commercialized graphene nanoplatelets, some researchers have reported the need to incorporate higher concentrations. This research aims to develop nanocomposites with electrical conductivity potential, based on high concentrations of graphene nanoplatelets (i.e., 12.5 and 25 wt%) and conventional polymers (i.e., polystyrene (PS) and acrylonitrile butadiene styrene (ABS)). Moreover, it will investigate the effects of the high concentrations of graphene nanoplatelets on the mechanical, rheological and morphological properties of the nanocomposites. The results showed that the graphene nanoplatelets directly interfere in the complex viscosity and in the dynamic–mechanical properties of the polymers matrices. A significant increase in volume electrical conductivity was verified in both polymeric matrices when graphene nanoplatelets were added. While polymeric matrices acted as insulating materials, the nanocomposites containing 25 wt% of graphene nanoplatelets acted as semiconductors, for both matrices (PS and ABS). However, the mechanical properties of the tensile strength and impact were strongly reduced, due to the increased stiffness of the nanocomposites. These results indicated a potential application of these nanocomposites with high contents of graphene nanoplatelets in the electronics field, possibly as an alternative to conventional semiconductor materials, provided that the required mechanical properties are of low performance.

Graphical abstract



Extended author information available on the last page of the article

Keywords Graphene · Polymer · Composite · Rheometer · Dynamic–mechanical analysis · Conductivity

Introduction

Along with the technological development in 5G, big data, artificial intelligence, etc., the power density of electronic devices has been progressively increased, thus raising the urgent demand for efficient systems with electrical and thermal conductivity to ensure the efficiency, reliability, safety, durability, and stability [1, 2]. In this sense, recent research works emphasized the use of metallic ions as candidate materials to improve the performance of supercapacitors and batteries. [3–5].

At the same time, searching for non-metallic materials that conduct electricity well has become essential for different applications [6]. In this scenario, polymers have attracted great attentions owing to their impressive properties, such as light weight, low cost, high flexibility and excellent processibility [2]. Although, traditional polymers are generally considered insulating materials, since they promote resistance to the flow of electrons because of the low number of subatomic particles, making electrical charges difficult to transit.

Conventional conductive polymers have been investigated for applications in supercapacitors due the high electrical conductivity, rapid charge and discharge. However, in the process of long-term charge and discharge, its electrical conductivity gradually deteriorates [7]. Thus, it is evident the need to develop new polymeric materials with specific properties and with great added value and lower cost [8]. A promising strategy to increase the electrical conductivity of polymer is to construct interconnected three-dimensional carbon nanofibers networks [9–11].

In their review research, Alemour et al. [6] stated that recently, graphene has become the favorite candidate for scientist to be utilized as nanofiller for the polymer matrices. High electrical conductivity is the most common and important character of graphene [7, 12]. However, the electrical properties of graphene and its derivatives strongly depend on several crucial factors, including the type of filler and matrix, graphene geometry, properties of the interphase region, quantum tunneling effect, and the volume fraction of the filler in the matrix [13].

Single-layer graphene has properties that approximate single-walled carbon nanotubes (SWCNT). However, the high manufacturing costs of SWCNT and graphene (i.e., US\$700 per gram and US\$1,000 per gram, respectively) make them both unideal for polymer processing and composite industry. Instead, multi-walled carbon nanotubes (MWCNT) and graphene nanoplatelets (GNP), having far

lower manufacturing costs (i.e., US\$30 per kilogram and potentially US\$10–20 per kilogram, respectively) yet satisfactory mechanical and functional properties, are suitable to engineering applications [14].

NPG have different characteristics and properties depending on their size and number of layers, when compared to graphene. These characteristics are related to their aspect ratio and specific surface area. For example, the excellent electrical conductivity of few-layer graphene (layer number of 2–9) is decreased rapidly with increasing the layer numbers, remained constant for layer numbers up to 13–165 [15]. Thus, it is clear that the graphene electrical conductivity is closely related to the number of graphene layers [15, 16].

Raza et al. [17] studied the properties of nanocomposites of polystyrene matrix at different concentrations (0.1–0.7 wt%) of few-layer graphene. Thus, due to the dimensions of the graphene used, the authors got a noticeable increase of tensile strength by varying the value of few-layer graphene [17]. However, due to the distinct properties of graphene nanoplatelets, higher concentrations need to be tested to achieve changes in the properties of the composites.

Ding et al. [18] also studied PS/NPG nanocomposite. Interactions between PS and NPG led to anisotropic thermal conductive properties. Incorporation of 10 wt% NPG caused 66% enhancement in thermal conductivity [18]. Wei et al. [19] developed a 3D printable graphene/ABS composite filament. The authors concluded that adding 5.6 wt% graphene to ABS increased the electrical conductivity of polymer nanocomposite by four orders of magnitude [19].

Dul et al. [20] studied the effect of the incorporated NPG at 4 wt% in acrylonitrile–butadiene–styrene (ABS) filaments obtained by a solvent-free process consisting of melt compounding and extrusion. Nanocomposite filaments were then used to feed a fused deposition modelling (FDM) machine for 3D printed. According to the authors, the presence of graphene nanoplatelets improved the tensile modulus of ABS and the positive effect was also verified along several different orientations in FDM samples [20]. Li et al. [21] searched the production of polystyrene- (PS-) graphene nanoplatelets (GNP) (0.1, 1, and 10 wt%) nanofibers via electrospinning of dimethylformamide stabilized GNP and PS solutions. GNP modified PS nanofibers showed an increase of 7–8 orders of magnitude in electrical conductivity of the nanofibers at 10 wt% GNP loading [21].

Alauddin et al. [22] incorporated high concentrations (10, 20 and 30 wt%) of graphene nanoplatelets in ABS, through ball mill mixing, extrusion and injection. The electrical conductivity measurements were performed using the electrical

impedance spectroscopy (EIS) technique. According to the authors, the conductivity of pure ABS polymer was enhanced when NPG was incorporated into the composite as the electrical conductivity of ABS/graphene nanocomposite significantly increased to 10^{-9} S/cm (at 30 wt%). However, polymer matrix thus considerably reduced its impact strength [22].

Huang et al. [23] studied the effects of increased NPG concentration on variations in the structure and properties of electrospun GNP-filled poly(trimethylene terephthalate) (PTT/GNP) composite fiber. For the PTT/GNP composite fibers exhibited an improvement in conductivity, and its gradual transition was observed from 1.77 to 9.98 vol% of GNP [23].

Mergen et al. [16] developed the nanocomposites of polystyrene/graphene nanoplatelets (PS/NPG) and polystyrene/multi-walled carbon nanotube (PS/MWCNT) prepared through solution mixing processing. The observed behavior surface conductivity for nanocomposites was explained according to the classical percolation theory, where the percolation thresholds for PS/NPG and PS/MWCNT nanocomposites were determined as 12.0 and 3.81 vol%, respectively. According to the authors, the results indicate that both carbon fillers created a three-dimensional network in PS matrix [16].

Vidakis et al. [24] studied the influence of various concentrations of graphene nanoplatelets in the acrylonitrile–butadiene–styrene (ABS) nanocomposite filaments for 3D-printing, produced by melt compounding and extrusion. Regarding the electrical properties of the composites, the authors reported that the dielectric constant increased by increasing NPG content. The electrical conductivity of the nanocomposites was increased at the concentration of 10 wt% in NPG. The nanocomposites started to present characteristics of semiconductor materials [24].

Given the above, this research aimed to develop nanocomposites with electrical conductivity potential, based on high concentrations of graphene nanoplatelets and conventional polymers (polystyrene (PS) and acrylonitrile butadiene styrene (ABS)) and studied the effects of the high concentrations of graphene nanoplatelets on the properties (i.e., mechanical, rheological and morphological) of the nanocomposites.

Experimental

Materials

In this work, the polymers, i.e., PS (polystyrene, grade N1921, Innova, Brazil) and ABS (acrylonitrile butadiene styrene, grade GP-22, Ineos Styrolution, Germany) were used. Graphene nanoplatelets (NPG) were supplied from

Strem Chemicals (USA) with a thickness 6–8 nm, width of 5 μm , surface area of $120 \text{ m}^2 \text{ g}^{-1}$ and number of layers between 20 and 40. The carbon, oxygen and residual acid content supplied by the manufacturer was 99.5%, 1% and 0.5%, respectively (CAS 1034343-98-0).

Methods

For the development of composites, 40 g of polymer (PS or ABS) were added to a torque rheometer at 180 °C and 100 rpm until complete melting. With the polymer melted, the NPGs were added at specific concentrations at 180 °C and 100 rpm until complete homogenization was achieved (average time of mixing 30 min). The composites were ground in a knife mill (Marconi Equipment, Algodoal, Brazil). The ground composites were placed in metallic molds ($79 \times 165 \times 3 \text{ mm}^3$) and formed in a thermopress (Schulz, Brazil) for 2 min at 180 °C with 10 tons of force.

Subsequently, the metal molds were transferred to a cooling press (Bovenau, Brazil) and held for 2 min with 8 tons of force. The plates formed were then cut into the dimensions of the specimens suitable for the analyses. The pure polymer samples underwent the same processing for comparison purposes.

The nomenclature adopted for the identification of composite samples corresponded to the used polymers and content of NPG. Example: ABS/NPG25, corresponded to the composite containing 75 wt% (m/m) of ABS and 25 wt% (m/m) of NPG in its composition.

Characterization

Field emission scanning electron microscopy with gun (FESEM) (Tescan, MIRA 3, Czech) was used to evaluate the composite morphology. The viscoelastic properties were evaluated by a dynamic rheometer (Anton Paar, Physica MCR 502, Austria) with parallel plate geometry at a temperature of 190 °C, in an angular frequency range from 100 to 0.1 rad/s, with an amplitude of 0.1 rad, under ambient atmosphere.

The hardness determination test was carried out following the ASTM D2240-15 standard on a Shore D durometer, model M-702 from the Minard brand. Dynamic–mechanical analysis (DMA) of the post-cured composites was done by a Q800 TA Instruments (USA) dynamic mechanical analyzer, using a single cantilever clamp, from 30 to 150 °C for PS and from – 100 to 150 °C for ABS, at 3 °C min^{-1} , frequency of 1 Hz and deformation amplitude of 0.10%.

Tensile tests were performed in a EMIC DL 2000 universal testing machine equipped following ASTM D638-10, 100 kgf load cell and 5 mm/min speed. It was used 10 specimens of each sample, with specific type I dimensions ($165 \times 24 \times 2.9 \text{ mm}^3$).

Izod impact test was carried out by a Resil 25, CEAST equipment with a 2.75 J pendulum, according to ASTM D256-10 using unnotched specimens. It was used 10 specimens of each sample, with $127 \times 12.7 \times 2.9 \text{ mm}^3$ dimensions.

Volume electrical conductivity of the samples was measured according to ASTM D257-07. In this technique, the direct current conducted through the samples of $50 \times 1 \text{ mm}^2$ (diameter \times thickness) was measured using a benchtop multimeter (Fluke, 8846A) with a voltage of 100 V, inside a grounded Faraday cage. The volume electrical conductivity was calculated from the current measurements obtained, combined with the dimensions of the circular samples according to Eq. (1):

$$\gamma_v = \frac{t}{R_v \times A} \quad (1)$$

where γ_v is the volume electrical conductivity (S/cm), t is the average thickness of the specimen (cm), R_v is the measured

volume resistance (ohms), and A is the effective area of the measuring circular electrode (cm^2) (ASTM D257-07).

Results and discussion

Figure 1 shows the FESEM micrographs of the cross section of the studied samples. It is observed that the samples of pure polymers (PS and ABS) present surfaces with signs of fragile fractures, typical of these materials. Samples containing graphene nanoplatelets (i.e., PS/NPG12.5; PS/NPG25; ABS/NPG12.5; ABS/NPG25) show lamellar aggregates characteristic of NPGs, confirming the morphological tendency of NPG to present itself as very thin graphene sheets, randomly scattered. Although a good dispersion was observed, it is possible to perceive voids between the interfaces of the graphene nanoplatelets and the polymer, evidencing the low mechanical bond between the dispersed phase and the matrix.

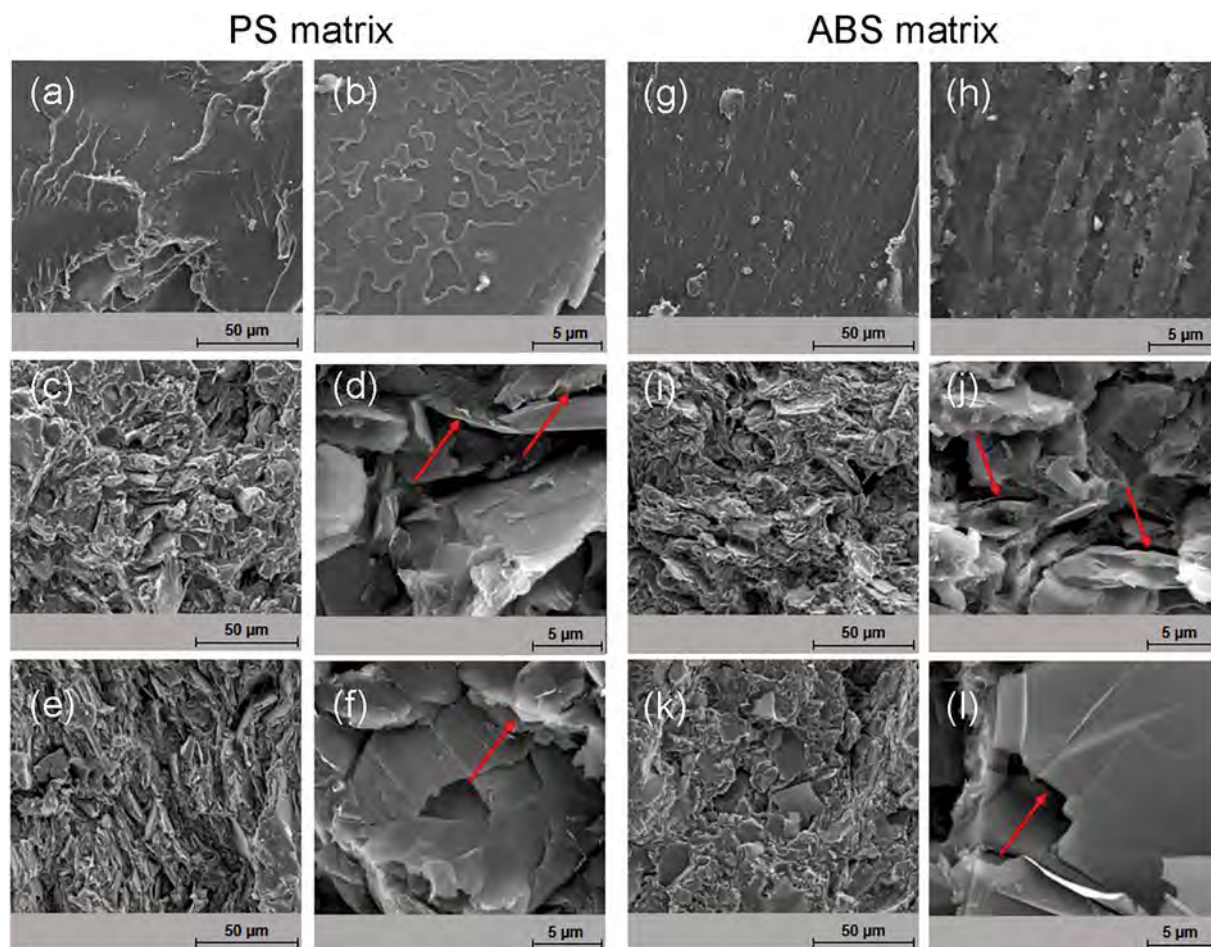


Fig. 1 FESEM micrographs of the cross sections of: **a, b** PS, **c, d** PS/NPG12.5, **e, f** PS/NPG25, **g, h** ABS, **i, j** ABS/NPG12.5, and **k, l** ABS/NPG25 samples

The results of complex viscosity, obtained by rheometry, is presented in (Fig. 2). It is well-known that rheological measurements not only provide the information about the flowability of the polymer composites but also reflect the dispersion state and microstructure of fillers in the composites [25]. For the samples with PS matrix (Fig. 2a), it is possible to observe that the graphene nanoplatelets contributed to the reduction of viscosity. This result indicates that, in this polymeric matrix, the NPG act as a lubricant in the alignment of polymer chains [26]. However, at high angular frequency values, the complex viscosity of the samples with NPGs approaches the value of the pure polymer sample. This result may be associated with changes in the interfacial interactions of the matrix with the nanofiller, altering the viscosity of the system.

In general, the viscosity increases gradually with increasing content of fillers in the composites at low frequencies, once fillers form the percolated network-like structure [25]. For the samples containing ABS matrix (Fig. 2b) it was observed that the graphene nanoplatelets contributed to the increase in viscosity, when compared to the pure ABS

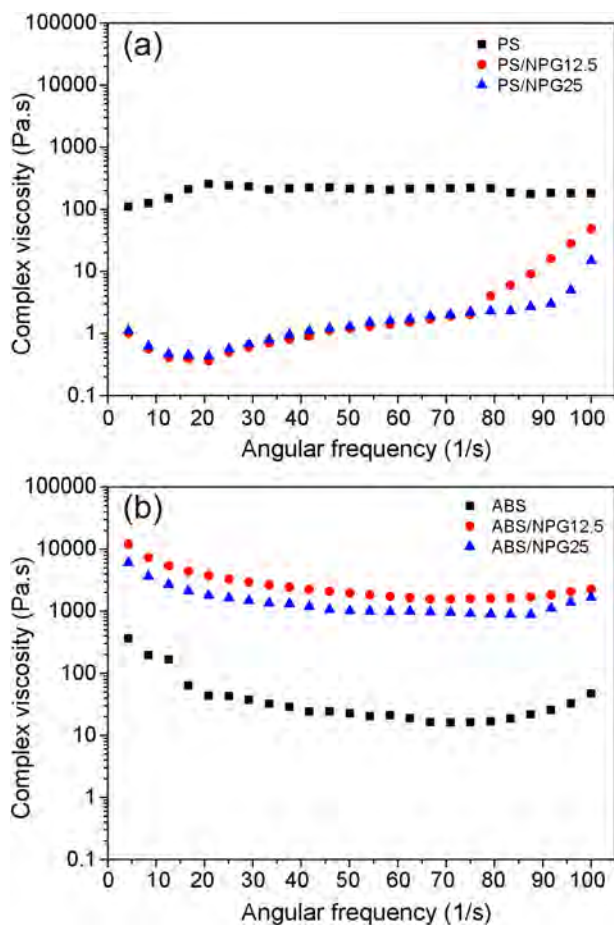


Fig. 2 Complex viscosity by rheometry for: **a** PS/NPG and **b** ABS/NPG samples

sample. In this case, the graphene nanoplatelets did not act as a lubricant, probably due to the molecular configuration of ABS, the lateral presence of acrylonitrile and longer chains, hindering the flow of NPG between the polymer chains.

In their research, Alvaredo et al. [27] found similar results to those observed for ABS/NPG composites in the present work. The researchers studied the effect of the graphene nanoplatelets (GNP) in polyetheretherketone (PEEK) composites. According to these authors, the increase of GNP content to 5 and 10 wt% triggered severe falls in the rheological behavior of the composites in the whole frequency range analyzed. The viscosity of the composites has increased considerably, due to the increase of the polymer chain mobility restrictions, limits the large-scale motions of the PEEK chains [27].

Figure 3 presents the Shore D hardness results for the studied samples. It was compared the hardness of unfilled polymer matrices. It is possible to observe that the values obtained for PS were superior to ABS, which is in agreement with the results presented in the study of hardness of several polymers comparing different hardness methods, with mean values of 79.2 and 80.8 Shore D points for ABS and PS, respectively [28].

For the behavior of the composites, the addition of NPG promoted the hardness decrease for both PS and ABS. This can be attributed to the NPG acting as a lubricant, facilitating the mobility of the polymer chains, and thus impacting on the decrease of the hardness of these materials. In addition, the smaller decrease in hardness according to the addition of NPG to ABS (1 point) in relation to PS (2 points) may be related to the complexity of ABS polymer chains which is greater in relation to PS chains, reducing the lubricating effect of NPG and thus having a greater restriction between its chains [26]. This result corroborates the result

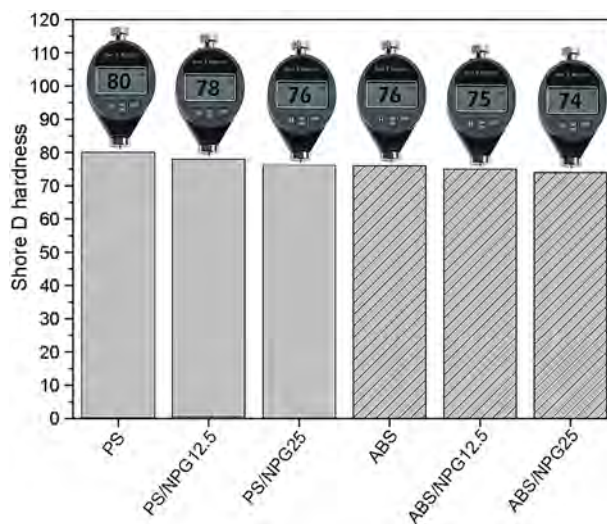


Fig. 3 Shore D hardness for the samples studied

of complex viscosity of the samples (Fig. 2) where depending on the complexity of the polymer matrix, the presence of nanoplatelets acts differently.

Figure 4 presents the storage modulus, loss modulus and tan delta by DMA analysis for the samples studied. Evaluating the storage modulus results it is observed that the addition of NPG promoted an increase in the modulus in

the vitreous region, when compared to the pure polymer samples (PS and ABS). This behavior can be attributed to the increase in the stiffness of the composite, conferred by the high levels of NPG. This behavior extended over the entire temperature range (glassy and elastomeric regions), where a higher storage modulus was observed for composites containing NPG. This result may be associated with

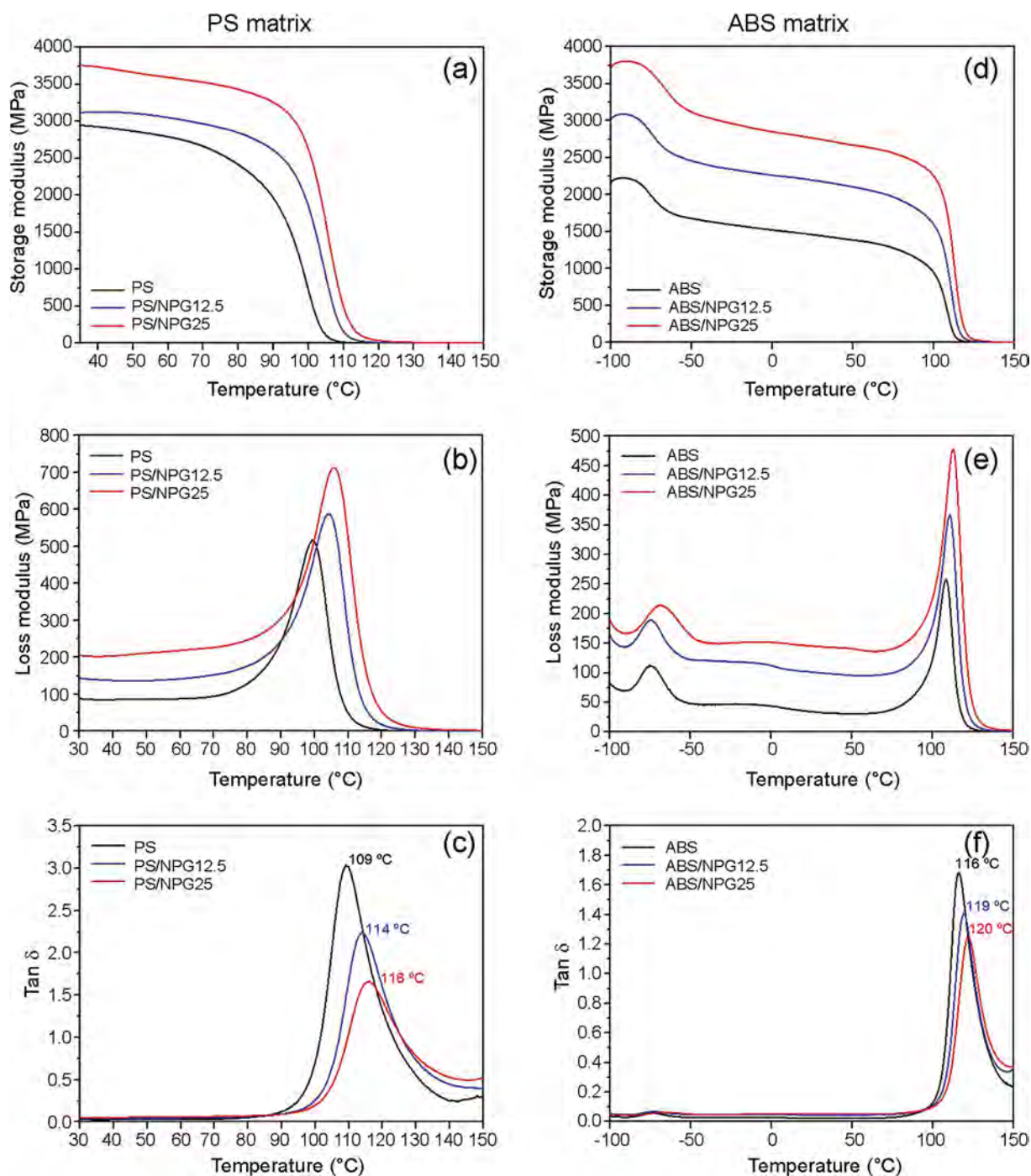


Fig. 4 Variations of **a** storage modulus, **b** loss modulus and **c** tan delta for PS/NPG and **d** storage modulus, **e** loss modulus and **f** tan delta for ABS/NPG samples by DMA analysis

the formation of cross-links in the polymeric chain of the matrices, forming a more rigid composite. The results are supported by the loss modulus, where the composite samples containing NPG showed higher values than the pure polymer samples.

Dul et al. [20] in their study about the effect of incorporated of the graphene nanoplatelets at (4 wt%) in acrylonitrile–butadiene–styrene (ABS), reported that the storage modulus of composite materials was more than twice that of the neat ABS for all investigated samples, thus revealing a positive stiffening effect of graphene nanoplatelets in the molten state.

From the results for the damping factor (tan delta) it is possible to obtain the glass transition temperature (T_g) of the samples studied. It is observed that there is a gradual increase in T_g and a decrease in peak height as the concentration of NPG in the composites was increased, for both polymer matrices (PS and ABS). This increase may be associated with the increase in the stiffness of the composites, due to the cross-links formed by the high concentrations of NPG. Furthermore, the decrease in the height of the peaks in the tan delta curves may be associated with the reduction in the mobility of the polymeric chains imposed by the nanofillers. Other researchers also reported the presence of the NPG in ABS composites caused an increase of T_g values by about 1 °C due to the restriction of the motion of macromolecules [19, 20].

The results of the mechanical property of tensile strength are shown in Fig. 5 and Table 1. The stress–strain curves (Fig. 5a) showed that the studied samples presented brittle behavior, since the samples broke easily, even in the elastic phase. For these materials, the plastic domain was practically non-existent, with no flow phase, indicating its low capacity to absorb permanent deformations.

It is observed that the addition of graphene nanoplatelets promoted a decrease in the tensile strength of the composites, in both polymeric matrices (PS and ABS) (Fig. 5b and Table 1). This result is associated with the possible formation of agglomerates that generate structural defects in the matrix, forming micro voids and flaws in the structure, contributing to the reduction of the mechanical performance of the composites.

Furthermore, it is observed that the addition of graphene nanoplatelets promoted a significant increase in the elastic modulus of the composites in both polymeric matrix (PS and ABS). This result indicated that the increase of the NPG concentration was directly associated with the increase in the stiffness of the composite (Fig. 5c and Table 1). This result was in agreement with what was previously observed in the shore D hardness test and DMA tests, where it was evident that the incorporation of NPG in the polymeric matrices restricts the mobility of the polymer chains, thus increasing the stiffness of the composites.

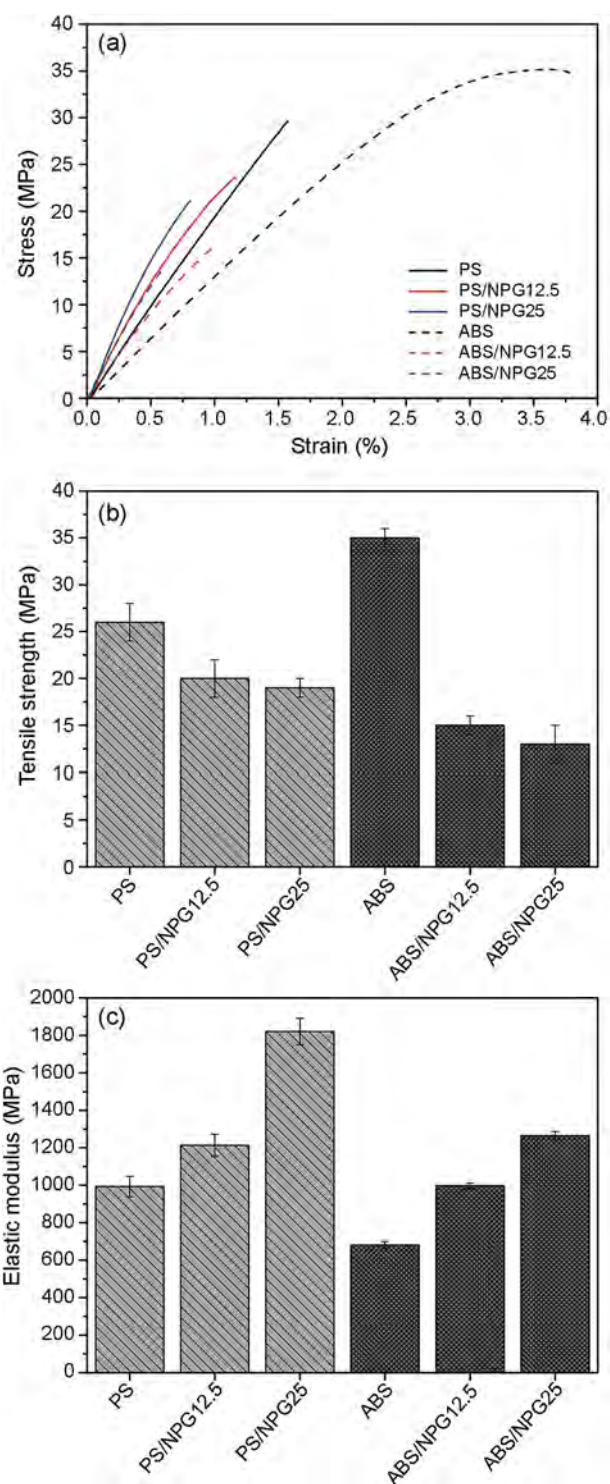


Fig. 5 Tensile test for the samples studied: **a** Stress–strain curves, **b** tensile strength and **c** elastic modulus

Similar results were observed by Dul et al. [20], when studying the effect of graphene nanoplatelets (4 wt%) on the mechanical properties of ABS nanocomposites. The authors observed that presence of graphene nanoplatelets

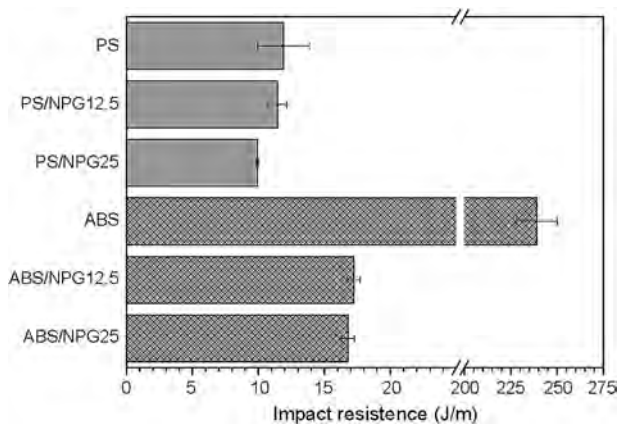
Table 1 Tensile strength and elastic modulus results of the samples studied

Sample code	Tensile strength (MPa)	Elastic modulus (MPa)
PS	26 ± 2	993 ± 54
PS/NPG12.5	20 ± 2	1213 ± 60
PS/NPG25	19 ± 1	1821 ± 70
ABS	35 ± 1	681 ± 19
ABS/NPG12.5	15 ± 1	998 ± 14
ABS/NPG25	13 ± 2	1264 ± 23

promoted an increase in the elastic modulus of the ABS matrix, but decreased its ultimate tensile strength. According to the authors, the reduction of ultimate properties could be attributed to a poor adhesion level between the nanofiller and ABS matrix.

Vidakis et al. [24] also reported similar results in their research work on ABS/NPG nanocomposites. The authors observed that increasing the concentration of NPG promoted a decrease in the tensile strength and an increase in the elastic modulus of the nanocomposites, in comparison with pure ABS. The authors related the results to the weak bonding connection of the graphene nanoplatelets to the polymer matrix due to the incorporation of a non-functionalized carbon-based filler.

The results of the impact resistance test are shown in Fig. 6. It is observed that the addition of NPG contributes directly to the reduction of the impact resistance of the composites, when compared to the pure polymer sample (PS and ABS). For PS matrix composites, a small magnitude of 15% decrease in the impact strength value was observed for the sample with the highest NPG content (PS/NPG25) when comparing with pure polymer sample (PS). This brief decrease in the toughness of the material might be associated

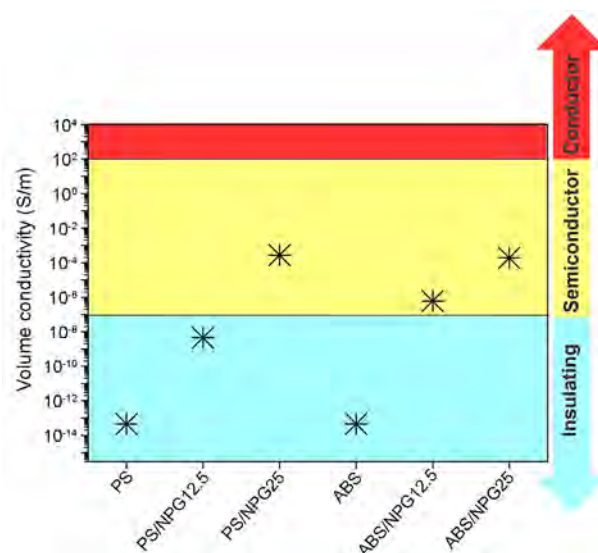
**Fig. 6** Variations of impact resistance for the samples studied

with the high concentration of nanofiller that increased the stiffness and brittleness of the nanocomposites, due to the probable formation of agglomerates and restriction of the mobility of the polymer chains.

Significantly, ABS matrix composites containing NPG (ABS/NPG12.5 and ABS/NPG25) showed a 95% decrease in the impact strength when compared to pure polymer sample (ABS). It is known that the butadiene (elastomeric component) is responsible for flexibility and impact resistance, characterizing the rubber phase of the polymer. It was believed that the addition of high concentrations of NPG promoted changes in the structure of the ABS polymer chains, making it more stiff and brittle. Moreover, other possible reasons for the observed reduction in interlayer bonding could be related to the local stress concentration induced by graphene nanoplatelets and to the intrinsically brittle nature of the graphene nanoplatelets [20].

Similar results were reported by Alauddin et al. [22] in their research. The authors also observed that the addition of graphene platelets to the ABS polymer tremendously reduced the impact strength of the nanocomposite. Apparently, presence of graphene weekend the structure of the polymer matrix that was built based on the cross-linkage of poly-butadiene with styrene–acrylonitrile.

The volume electrical conductivity of the composites as a function of the NPG content and polymer matrix is shown in Fig. 7. As previously mentioned, graphene nanoplatelets have electrical conductivity properties different from those of the few-layer graphene. The electronic homogeneity of graphene is violated by the introduction of disorders into the graphene structure. These disorders are able to alter the bond length of the interatomic bonds and lead to the re-hybridization of the σ and π orbitals. Thus, the defects

**Fig. 7** Volume electrical conductivity for the samples studied

might cause the scattering of electron waves and change the electron trajectories [15].

The addition of 12.5% of NPG in the composition induced significant changes in the resistivity of the polymeric matrix, both for PS and ABS, of the 5 and 7 orders of magnitude, respectively, when compared to the pure polymers. These results indicated that the presence of greater dimensions and/or larger clusters was important for the formation of an interconnected NPG conductive network that allowed transport of electric current.

A significant enhancement in the volume electrical conductivity was obtained for both PS and ABS, up to 10 orders of magnitude for the composites with 25 wt% of NPG, showing a sharp transition from insulating to semiconducting behavior. PS and ABS have a conductivity of 4.5×10^{-14} S/m, therefore, are classified as insulating materials. Thus, the addition of conductive fillers, such as NPG, had the main goal of increasing the volume electrical conductivity of the final material, besides improving its mechanical properties.

It is known that the level of dispersion and morphology of the agglomerates affect the final properties of the nanocomposite, mainly because they are insulating matrices, making it essential that there is a path for the current to travel through the length of the composite. Thus, the increase in the volume electrical conductivity of the composites indicated that the addition of high concentrations of NPG promoted the formation of a three-dimensional network that favored the passage of electrical current through the composites. Considering the SEM micrographs presented in Fig. 1, it is possible to verify any prevailing contact between the fillers allowing the electrical current to pass through the entire sample.

The fact that composite ABS/NPG12.5 (6.0×10^{-7} S/m) has a higher conductivity than PS/NPG12.5 (4.5×10^{-9} S/m) might be related to the lubricating action of the filler during processing stage, which aided in a more homogeneous dispersion of the filler in the ABS matrix compared to its dispersion in the PS matrix. As for the ABS/NPG25 and PS/NPG25 composites, the results were very similar, which was possibly related to the formation of the NPG–NPG network due to the high amount of filler and due the compression method chosen to produce the specimens, forcing the contacts among the dispersed NPG content.

Alauddin et al. [22] managed to increase the volume electrical conductivity from 10^{-17} S/cm for pure ABS polymer to 10^{-9} S/cm for nanocomposite ABS/graphene(30 wt%). As mentioned by the authors, the intermolecular interactions between NPG and ABS increased the electron mobility inside the polymer matrix, thus, enhanced the conductivity. Nevertheless, the value was still low to consider this nanocomposite as a semiconductor.

Vidakis et al. [24], in their research on ABS and NPG nanocomposites, reported a conductivity of 10^{-6} S/m, for

the sample containing 10 wt% of NPG, concluding that NPG might increase the conductivity of pure ABS.

Conclusions

The primary methods for preparing graphene/polymer composites is based on simple physical mixing of very low concentration of graphene with polymer. An outstanding electrically conductive graphene/polymer composite is expected to have higher electrical conductivity at a lower graphene loading. However, due to the distinct properties of the graphene nanoplatelets, when compared to graphene, it is necessary to add higher concentrations of this nanofiller. Wherefore, in this research work, nanocomposites were produced from different polymer matrices (PS and ABS) and high concentrations of graphene nanoplatelets. By DMA results, it was observed that, for both polymer matrices, the addition of graphene nanoplatelets gradually increased the storage modulus, loss modulus and T_g values associated with the formation of cross-links in the polymeric chain of the matrices, forming stiffer composites. Due to the addition of NPGs, the complex viscosity decreased in the PS nanocomposites, due to the lubricating effect of the graphene nanoplatelets. On the other hand, in the ABS nanocomposites, the complex viscosity increased, probably due to the molecular configuration of ABS, the lateral presence of acrylonitrile and longer chains, hindering the flow of graphene sheets between the polymer chains. The mechanical properties of tensile strength and impact strength were strongly reduced with the addition of NPGs, due to the increased stiffness of the nanocomposites. Fortunately, an expressive enhancement in volume electrical conductivity was obtained for the nanocomposites with 25 wt% of NPG, for both matrices (PS and ABS). It was found a surprising increase up to 10 orders of magnitude, increasing from 10^{-14} S/m for pure polymer matrices to 10^{-4} S/m for nanocomposites containing 25 wt% NPG. Therefore, the sharp transition from insulating materials to semiconducting materials was confirmed. The results presented shed light on the potential applications of the developed nanocomposites, since they exhibited good electrical performance (i.e., semiconductor behavior) becoming a promising candidate in future electronics applications. However, it is worth mentioning that the addition of high concentrations of NPG in these polymeric matrices decreased their mechanical properties, limiting their applications in systems with high mechanical performance.

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References

- Shanmugasundram HPPV, Jayamani E, Soon KH (2022) A comprehensive review on dielectric composites: classification of dielectric composites. *Renew Sustain Energy Rev* 157:112075
- Wu N, Che S, Li H, Wang C, Tian X, Li Y (2021) A review of three-dimensional graphene networks for use in thermally conductive polymer composites: construction and applications. *New Carbon Mater* 36:911–929
- Bai Y, Liu C, Chen T, Li W, Zheng S, Pi Y, Luo Y, Pang H (2021) MXene-copper/cobalt hybrids via lewis acidic molten salts etching for high performance symmetric supercapacitors. *Angew Chem* 60:25318–25322
- Li W, Guo X, Geng P, Du M, Jing Q, Chen X, Zhang G, Li H, Xu Q, Braunstein P, Pang H (2021) Rational design and general synthesis of multimetallic metal–organic framework nano-octahedra for enhanced Li–S battery. *Adv Mater* 33:2105163
- Zheng Y, Zheng S, Xue H, Pang H (2018) Metal-organic frameworks/graphene-based materials: preparations and applications. *Adv Funct Mater* 28:1804950
- Alemour B, Yaacob MH, Lim HN, Hassan MR (2018) Review of electrical properties of graphene conductive composites. *Int J Nanoelectron Mater* 11:371–398
- Zhang R, Pang H (2021) Application of graphene-metal/conductive polymer based composites in supercapacitors. *J Energy Storage* 33:102037
- Zamiri G, Haseeb ASMA (2020) Recent trends and developments in graphene/conducting polymer nanocomposites chemiresistive sensors. *Materials* 13:3311
- Gaikwad SD, Goyal RK (2018) Effect of manufacturing processes on percolation threshold and electrical conductivity of polymer/multi layers graphene nanocomposites. *Diam Relat Mater* 85:13–17
- Li J, Zhang W, Zhang X, Huo L, Liang J, Wu L, Liu Y, Gao J, Pang H, Xue H (2020) Copolymer derived micro/meso-porous carbon nanofibers with vacancy-type defects for high-performance supercapacitors. *J Mater Chem A* 8:2463–2471
- Kausar A (2020) Technical viewpoint on polystyrene/graphene nanocomposite. *J Thermoplast Compos Mater* 1:1–15
- Itapu BM, Jayatissa AH (2018) A review in graphene/polymer composites. *Chem Sci Int J* 23:1–16
- Mazaheri M, Payandehpeyman J, Khomehchi M (2020) A developed theoretical model for effective electrical conductivity and percolation behavior of polymer-graphene nanocomposites with various exfoliated filleted nanoplatelets. *Carbon* 169:264–275
- Su X, Wang R, Li X, Araby S, Kuan H, Naeem M, Ma J (2021) A comparative study of polymer nanocomposites containing multi-walled carbon nanotubes and graphene nanoplatelets. *Nano Mater Sci*. <https://doi.org/10.1016/j.nanoms.2021.08.003>
- Yang G, Li L, Lee WB, Ng MC (2018) Structure of graphene and its disorders: a review. *Sci Technol Adv Mater* 19:613–648
- Mergen OB, Umut E, Arda E, Kara S (2020) A comparative study on the AC/DC conductivity, dielectric and optical properties of polystyrene/graphene nanoplatelets (PS/GNP) and multi-walled carbon nanotube (PS/MWCNT) nanocomposites. *Polym Test* 90:106682
- Raza H, Aized T, Khan MB, Imran M (2018) Tensile testing of polystyrene graphene 2D nano composite membrane. *Int J Adv Manuf Technol* 94:4343–4349
- Ding P, Zhang J, Song N, Tang S, Liu Y, Shi L (2015) Anisotropic thermal conductive properties of hot-pressed polystyrene/graphene composites in the through-plane and in-plane directions. *Compos Sci Technol* 109:25–31
- Wei X, Li D, Jiang W, Gu Z, Wang X, Zhang Z, Sun Z (2015) 3D printable graphene composite. *Sci Rep* 5:11181
- Dul S, Fambri L, Pegoretti A (2016) Fused deposition modeling with ABS-graphene nanocomposites. *Compos Part A Appl Sci Manuf* 85:181–191. <https://doi.org/10.1016/j.compositesa.2016.03.013>
- Li Y, Porwal H, Huang Z, Zhang H, Bilotti E, Peijs T (2016) Enhanced thermal and electrical properties of polystyrene-graphene nanofibers via electrospinning. *J Nanomater* 2016:4624976
- Alauddin SM, Ismail I, Zaili FS, Ilias NF, Aripin NFK (2018) Electrical and mechanical properties of acrylonitrile butadiene styrene/graphene platelet nanocomposite. *Mater Today Proc* 5:S125–S129
- Huang C, Wu H, Jeng Y, Liang W (2019) Electrospun graphene nanosheet-filled poly(trimethylene terephthalate) composite fibers: effects of the graphene nanosheet content on morphologies, electrical conductivity, crystallization behavior, and mechanical properties. *Polymers* 11:164–187
- Vidakis N, Maniadi A, Petousis M, Vamvakaki M, Kenanakis G, Koudoumas E (2020) Mechanical and electrical properties investigation of 3D-printed acrylonitrile–butadiene–styrene graphene and carbon nanocomposites. *J Mater Eng Perform* 29:1909–1918
- Bai Q, Jin X, Yang J, Qi X, Wang Y (2019) Constructing network structure of graphene nanoplatelets/carbon nanofibers in polystyrene and the resultant heat resistance, thermal and conductive properties. *Compos Part A Appl Sci Manuf* 117:299–307
- Omran E, Moghadam AD, Algazzar M, Menezes PL, Rohatgi PK (2016) Effect of graphite particles on improving tribological properties Al-16Si-5Ni-5graphite self-lubricating composite under fully flooded and starved lubrication conditions for transportation applications. *Int J Adv Manuf Technol* 87:929–939
- Alvaredo A, Martín MI, Castell P, Guzmán de Villoria R, Fernández-Blázquez JP (2019) Non-isothermal crystallization behavior of PEEK/graphene nanoplatelets composites from melt and glass states. *Polymers* 11:124
- Vian WD, Denton NL (2018) Hardness comparison of polymer specimens produced with different processes. ASEE IL-IN Section Conference. 3, <https://docs.lib.purdue.edu/aseeil-insectionconference/2018/tech/3>. Accessed 25 July 2022

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