



The “Superlubricity State” of Carbonaceous Fillers on Polymer Composites

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In 2019 Ferreira et al. observed for the first time the influence of the superlubricity phenomenon of platelet-like fillers on the properties of high molecular weight polyethylene (HMWPE) in the molten state. At the time, the study was carried out in the solid state of the polymer, analyzing the influence of the superlubricity phenomenon of oxide graphene and oxide graphite (GrO) agglomerates on the mechanical properties of the HMWPE. It was observed that the agglomerates, due to their superlubricity state, toughen the HMWPE polymer matrix. Here, mechanical tests (tensile), X-ray microtomography, and scanning electron microscopy are carried out to elucidate this phenomenon. The same is observed for polystyrene containing very poorly dispersed GrO. The results presented here are a paradigm shift, as it has always been thought that the presence of agglomerates is harmful, but the opposite is proven here. In general, this paper can contribute a lot to the development of new composites and nanocomposites based on fillers that present the superlubricity phenomenon.

1. Introduction

The superlubricity phenomenon observed in platelet-like particles (graphite, molybdenum disulfide, graphene, mica, and oxide graphene) is a topic that has been widely discussed in recent decades.^[1] These particles, due to their good lubricating properties, are widely used as friction-reducing additives in various technological systems.^[1]

In 2019 Ferreira et al. may have opened up a new field of applications for these particles.^[2] For the first time, they elucidated the influence of the superlubricity phenomenon of platelet-like fillers on the viscoelastic behavior of polymers.^[2] They showed that the addition of 0.1 wt% of multilayer GO (mGO), graphite oxide (GrO), and graphite (Gr) to high molecular weight polyethylene (HMWPE), due to the superlubricity state of agglomerates, can reduce the viscosity, molecular relaxation time, and significantly increase the deformation of HMWPE in the molten state. This shows that these particles

have the potential to be used as processing aid additives for polymers.^[2] The superlubricity phenomenon of particles appears due to the high amount of out-of-register flake-flake contacts present in the agglomerates.^[2] The friction between these contacts is extremely low, giving rise to the superlubricity.^[2–5]

This paper brings a new version of a previous study. This time the study was carried out on the polymer in the solid state, analyzing the influence of the superlubricity phenomenon of graphene oxide (GO) and GrO agglomerates on the mechanical properties of HMWPE. The HMWPE-mGO and HMWPE-GrO composites showed the greatest superlubricity effect during the rheological study in the molten state.^[2] Therefore, it is also expected that the mechanical properties of HMWPE will be significantly altered

due to the strong superlubricity effect of the GrO and mGO agglomerates.

Here, the mechanical performance of HMWPE with a content of 0.1 wt% of mGO and 0.1 wt% of GrO was evaluated. GrO was studied since it is a microparticle produced only by graphite oxidation without any exfoliation process; different to GO which is mostly a nanosheet with a very low number of layers (thickness of nanometers). The composites were produced using a twin-screw extruder and molded with a hot press. The compatibility between polyethylene and mGO and GrO, given their opposite chemical nature, was achieved by thermally reducing the filler during the molding of the composites.^[6] In order to observe the influence of the agglomerates on the mechanical properties of a brittle matrix, a polystyrene-based composite with GrO (0.1% by weight) was produced. In general, this paper can contribute a lot to the development of new composites and nanocomposites that use fillers that present the superlubricity phenomenon (molybdenum disulfide, graphite, graphene oxide, graphene, carbon nanotube, boron nitride, etc.).

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2. Experimental Section

2.1. Characterization of Fillers

The mGO and GrO used here were the same used by Ferreira et al.^[2] The characterizations of the particles are meticulously detailed in their work.

2.1.1. Thermogravimetric Analysis (TGA)

The thermal stability of graphite oxide was evaluated using thermogravimetric analysis (DSC/TGA Q600, TA Instruments). This experiment was carried out to check the possibility of GrO thermal reduction during the molding of nanocomposites and composites. This experiment was conducted under an inert atmosphere over a temperature range of 30–200 °C (10 °C min⁻¹) and upon reaching the final temperature, an isotherm was kept for 120 min. All measurements were conducted under an inert atmosphere (nitrogen).

2.1.2. Fourier-Transform Infrared Spectrophotometer-Attenuated Total Reflection (FTIR-ATR)

Analysis of graphite and GrO was performed in a Shimadzu IRAffinity-1S. The spectroscopy range used was 500 to 4000 cm⁻¹. The resolution used was 4 cm⁻¹.

2.2. Processing and Characterization of Composites

Two polymers, HMWPE with a number-average molecular weight (M_n) of 1.48×10^5 g mol⁻¹ and polystyrene (PS) with M_n of 7.8×10^4 g mol⁻¹, were used.

2.2.1. Processing of Composites

The HMWPE, HMWPE-mGO (0.1 wt%), and HMWPE-GrO (0.1 wt%) used here were the same used by Ferreira et al.^[2] The HMWPE and its composites were processed in a twin-screw extruder (Process 11, ThermoScientific). The processing parameters were meticulously detailed in the work by Ferreira et al. The molding of HMWPE and its composites was carried out in a hot hydraulic press (Solab SL-1220), operating at a temperature of 200 °C on the top and bottom plate and was subjected to 6 tons of pressure for two different molding times, 10 and 60 min. The samples were cooled at room temperature. In the case of the PS/GrO composite, the mixing was only physical, and they were directly molded by injection, without going through any previous melting mixing stage. The injection machine used was the MiniJet Pro, ThermoScientific, using an ASTM Type V test specimen mold. The temperatures were 230 and 80 °C, for barrel and mold, respectively. An injection pressure of 300 bar (30 s) and post-pressure of 150 bar (20 s) were applied.

2.2.2. Tensile Test

The stress–strain test of HMWPE and its composites and PS and PS-GrO were performed at a deformation rate of 20 and 1 mm min⁻¹, respectively, at room temperature on a Zwick/Roell Z100 testing machine. Tests were performed on seven specimens for each type of sample. The sample specification follows the ASTM Type V standard.

2.2.3. X-Ray Microtomography

Pieces at least 2 mm × 2 mm × 1.2 mm from specimens were used for this characterization. Samples were analyzed in a Sky-Scanner 1272 (Bruker), at 20 kV and with a 175 μA X-ray source, with a final image resolution of 2 μm pixel⁻¹.

2.2.4. Scanning Electron Microscopy (SEM)

The fracture surface of specimens after the tensile test of the HMWPE, HMWPE-mGO (0.1 wt%), and HMWPE-GrO (0.1 wt%) molded for 60 min was analyzed on a HITACHI TM3000 tabletop microscope at 15 keV.

3. Results and Discussions

3.1. Tensile Tests of Composites of HMWPE-mGO, HMWPE-GrO, and PS-GrO

The mGO nanoparticle is obtained by exfoliating the GrO (microparticle) on ultrasound.^[2] The chemical structure and defect density of both particles are similar, allowing to estimate the physical and chemical properties of mGO through the analysis of GrO. Handling and analyzing GrO being a microparticle becomes easier. Thus, in order to evaluate the possible thermal reduction of mGO during the molding of nanocomposite on the hot press, the GrO was subjected to different times of thermal reduction at 200 °C (10, 60, and 120 min) to simulate the molding condition of the nanocomposite. The results and discussions of TGA (Figure S1, Supporting Information) and FTIR (Figure S2, Supporting Information) analysis of GrO under these thermal conditions are presented in the Supporting Information. In summary, the GrO is partially reduced after a thermal treatment of 120 min showing removal of some functional chemical group peaks (1371, 1224, and 1054 cm⁻¹) present for pristine GrO. For 10 min of isotherm, the functional chemical group peaks removed were 1371 and 1224 cm⁻¹. Between 10 and 60 min of isotherm, the 1054 cm⁻¹ peak was removed. Comparing the spectra of the GrO subjected to isotherms for 60 and 120 min, no difference is observed. Based on the FTIR spectrum (Figure S2, Supporting Information), the following decreasing order of GrO polarity is expected: nonannealed GrO > annealed GrO_{10 min} > annealed GrO_{60 and 120 min}. However, as HMWPE is an extremely nonpolar polymer, it is expected that composites molded for longer molding times at 200 °C present a better polymer-filler adhesion, due to the higher thermal reduction of GrO achieved for a longer molding time. This can be clearly seen in the results from tensile test of the HMWPE-mGO molded for 10 and 60 min, shown in **Figure 1**.

It is observed that HMWPE-mGO (0.1 wt%) molded for 10 and 60 min has a very different elastic modulus (E) (Figure 1a). The E increased drastically when the nanocomposite was molded for 60 min. For this molding condition, a cohesive interphase was formed between the two materials due to a greater thermal reduction of the GrO, as predicted in the results shown in Figures S1 and S2 (Supporting Information). However, it is observed that the HMWPE-mGO, even when molded

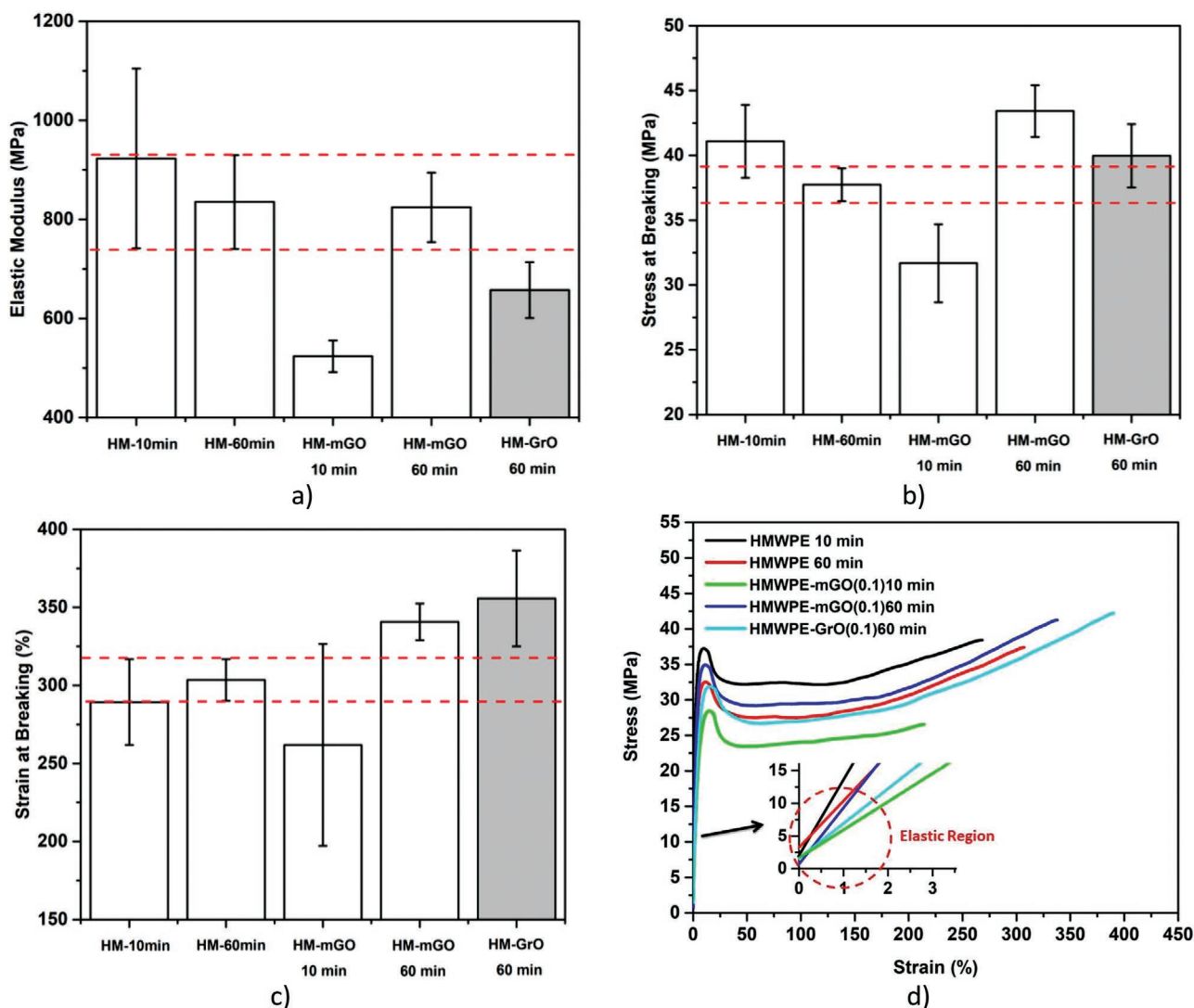


Figure 1. a) E , b) τ_b , and c) ϵ_b values of neat HMWPE and HMWPE-mGO (0.1 wt%) molded for 10 and 60 min and HMWPE-GrO (0.1 wt%) molded for 60 min. d) Stress–strain curves representative of the materials.

for 60 min, did not present larger modules than neat HMWPE. It is known that the incorporation of stiff particles, as is the case of mGO, to the polymeric matrix drastically increases its E , especially when nanofillers are added.^[7] However, it is not observed here. The nanocomposites molded for 10 and 60 min achieved an E lower than or equal to neat HMWPE. Ferreira et al.^[2] have shown that the morphology of HMWPE-mGO contains regions with mGO nanoparticles (well dispersed) and other regions with mGO agglomerates. They have reported that these regions behave the opposite way during the rheological tests of HMWPE-mGO in the molten state. The region with mGO nanoparticles acts by molecular mobility reduction (locking), leading the polymer to higher modules. Meanwhile, regions with agglomerated mGO act by facilitating molecular mobility due to the superlubricity effect, leading the polymer to smaller modules.^[2] It is believed that the E values of HMWPE-mGO (0.1 wt%), which is below expectations is a result of competition between these two regions, one rich in very well-

exfoliated mGO (increasing the modules) and the other rich in mGO agglomerates (decreasing the modules). This can be confirmed when HMWPE-GrO (0.1 wt%) molded for 60 min is evaluated (see Figure 1a). As a result of the absence of well-dispersed mGO and by the presence of a large amount of agglomerated GrO, as shown by Ferreira et al. through transmission electron microscopy and optical microscopy.^[2] Figure 1a shows that this composite displayed an E much lower than HMWPE and HMWPE-mGO (0.1 wt%) molded for the same time. These results are a strong indication that the low E values of the composites are due to the superlubricity state of the agglomerated filler. A similar conclusion to this was reported by Ferreira et al. for these composites in the molten state.^[2]

Other authors had results and conclusions similar to those in this article, but they have not clarified the issue, as is proposed here.^[8,9] Muñoz et al. reported that agglomerated mGO significantly reduced the elastic modulus of the PS matrix, while the well-dispersed mGO led the PS to the highest elastic

modulus values.^[8] Kalaitzidou et al. studied two exfoliated polypropylene/exfoliated graphite nanocomposites with different levels of filler dispersion.^[9] They observed that the nanocomposite with the well-dispersed filler presented a significant increase in the elastic modulus for low concentrations (0.01, 0.05, and 0.1 vol%) of exfoliated graphite added. In contrast, for the nanocomposites containing a greater amount of agglomerate, the elastic modulus of the nanocomposites was inferior to the neat polymer.

Figure 1b,c clearly shows that the increased molding time improved the mechanical properties of the HMWPE-mGO nanocomposite. As discussed earlier, this is a result of the higher adhesion at the HMWPE-mGO interphase. Comparing the neat HMWPE molded for different molding times, it can be observed that the neat HMWPE molded for 60 min displayed a lower stress at breaking. Ferreira and Fachine have evaluated the physical and mechanical properties of HMWPE (called HDPE high molecular weight in that previous paper) and HMWPE-UHMWPE blends molded for 10 and 60 min at 200 °C (same conditions used in this paper). They showed that the difference in the mechanical properties of HMWPE with increased molding time is not due to molecular degradation, but probably because it provides enough time for chain relaxation and disentanglement. As a result, the thickness of the lamellar crystal and the crystallinity degree of the HMWPE increased with the molding time.^[10] Brown et al. have proposed that the tie chain density is inversely proportional to the thickness of the lamellar crystal.^[11] Based on this, HMWPE molded for 60 min may have presented inferior mechanical performance due to the lower tie chain density. The superlubricity effect of agglomerates on the τ_b and ε_b of the HMWPE can be observed comparing the materials molded for 60 min. By analyzing the HMWPE-mGO molded for 60 min, it can be observed that the addition of mGO increased the τ_b and ε_b of the HMWPE.

The simultaneous increase in τ_b and ε_b is an unexpected result when adding graphene nanoparticles or graphene oxide to the polymer. Young et al. in their review article reported that the addition of graphene or graphene oxide to the polymer commonly increases its E and τ_b , to the detriment of reducing its ε_b .^[7] It is not observed here. Unlike the results reported by Young et al., the HMWPE-mGO studied here contains a large number of agglomerates, as discussed by Ferreira et al. **Figure 2** shows an overview of a large amount of agglomerate present in the HMWPE-mGO (0.1 wt%) specimen piece.

It is believed that the increase in ε_b observed for HMWPE-mGO is due to the superlubricity phenomenon of agglomerates. Ferreira et al. have shown in the creep test in the molten state that the agglomerates of mGO and GrO lead to greater polymer deformations,^[2] and the same may be occurring in the solid state. The regions with agglomerates present in HMWPE-mGO can lead the polymer to higher ε_b , while the regions with well-exfoliated mGO are leading to higher τ_b . Figure 1b,c also shows the τ_b and ε_b of the HMWPE-GrO molded for 60 min. In this case, the HMWPE-GrO presents only agglomerates of GrO flakes, as discussed by Ferreira et al.^[2] As a result of the absence of mGO nanoparticles in HMPWE-GrO, it is observed that its τ_b is considerably less when compared to HMWPE-mGO. On the other hand, due to the greater amount of flake-flake contact (agglomerates) present in HMWPE-GrO,^[2] its ε_b reaches

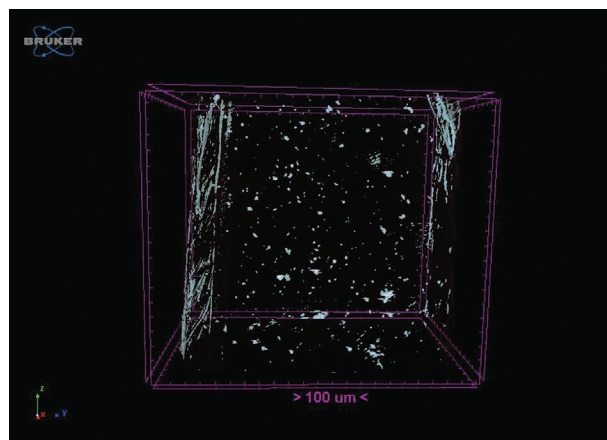


Figure 2. Microtomography image of mGO agglomerates contained in the HMWPE obtained using CTvox Software.

higher values. This leads to the conclusion that the presence of regions with agglomerated mGO and well-exfoliated mGO in the polymer matrix is ideal when it is desired to obtain a nanocomposite with high values of τ_b and ε_b . In Figure 1d shows stress–strain curves representative of the materials studied here.

Traditionally, it is accepted that agglomerates reduce the mechanical properties of polymers.^[12] However, even though HMWPE-mGO and HMWPE-GrO have a high content of agglomerates, their mechanical properties were superior to the neat HMWPE. It is believed that the agglomerates present in the HMWPE, instead of acting as a defect, are acting as a toughening mechanism, through the sliding between the flake-flake contacts, aiding the deformation of the polymer. This can be confirmed by analyzing the fracture surface (FS) of the samples after the tensile test by scanning electron microscopy. **Figure 3** shows the FS of HMWPE, HMWPE-mGO (0.1 wt%) molded for 10 and 60 min, and HMWPE-GrO (0.1 wt%) molded for 60 min. The images show three zones on the FS of the samples: river line (1), mist (2), and Hackles zone (3). The river line and Hackles zone are characteristic zones of ductile deformation, while the mist zone is characteristic of brittle fracture.^[13] In Figure 3a–c, it can be seen that the HMWPE and its composites molded for 60 min showed predominantly ductile zones along the FS; this is a result of the high tensile toughness.

Zone (1) has the characteristics of river lines that converge to a crack.^[14] This information makes it possible to locate where the crack that led to the polymer fracture arose. In Figure 3a–c, the crack nucleation that led to the fracture of the samples molded for 60 min can be seen as arising on the specimen surface. This observation leads to two conclusions. The first conclusion is that mGO and GrO agglomerates did not act as crack nucleation points; that is, they did not act as a defect. If the agglomerates had acted as a defect, multiple river line zones would be observed, or at one extreme, a smooth surface would be observed, characterizing a brittle fracture. The second conclusion is that the polymer-filler interphase debonding phenomenon did not occur, because if it had occurred, multiple river line zones would also be observed.^[14] This can be clearly understood by analyzing the fractured surface of the

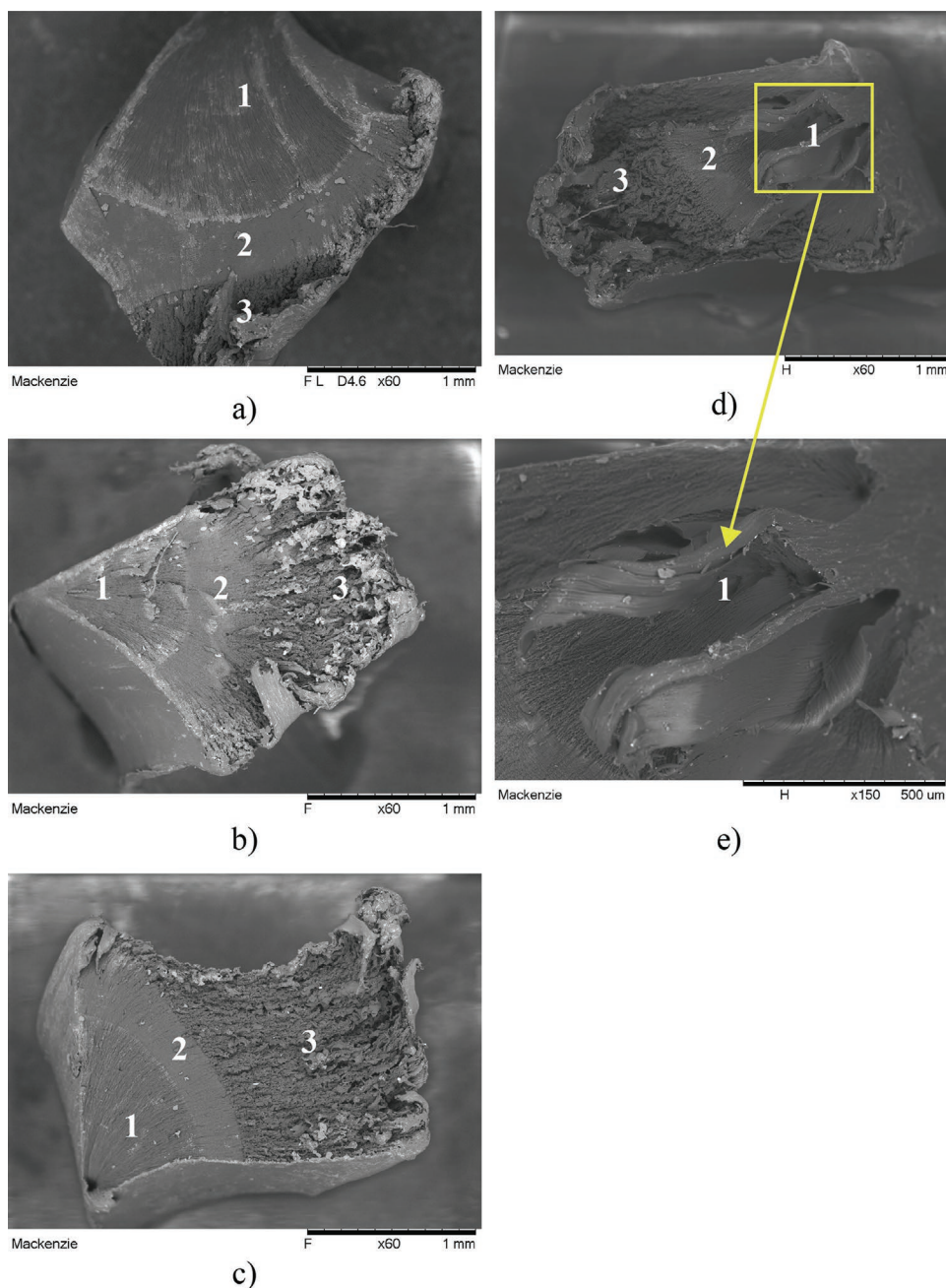


Figure 3. FS images of the specimens after the tensile test with a magnification of 60 \times . a) HMWPE, b) HMWPE-mGO (0.1 wt%), c) HMWPE-GrO (0.1 wt%) molded for 60 min, and d,e) HMWPE-mGO molded for 10 min.

HMWPE-mGO (0.1 wt%) molded for 10 min (Figure 3d,e). For this molding time, mGO acts as a defect, reducing the mechanical properties of HMWPE due to the poor adhesion achieved at the interphase, as seen earlier. These images show several river line zones converging to several points (crack nucleation), i.e., several cracks were nucleated due to the presence of the mGO particles. Differently to how it occurs for the composites molded for 60 min, crack nucleation does not occur on the specimen surface, but it occurs internally, because the mGO particles are poorly adhered to the polymer.

Images from Figure 3a–c also present mist (2) and Hakles zone (3). The mist zone (2) may be associated with crack acceleration just prior to rapid crack growth (Zone 3). A Hakles zone (3) is associated with high-energy dissipation due to localized plastic deformation on the FS.^[13] Comparing the samples molded for 60 min with each other, the Hakles zone is larger for HMWPE containing a higher content of agglomerate (HMWPE-GrO). This shows that the agglomerates do not act as a defect but acts as a toughening mechanism for HMWPE promoting a greater dissipation of energy during deformation,

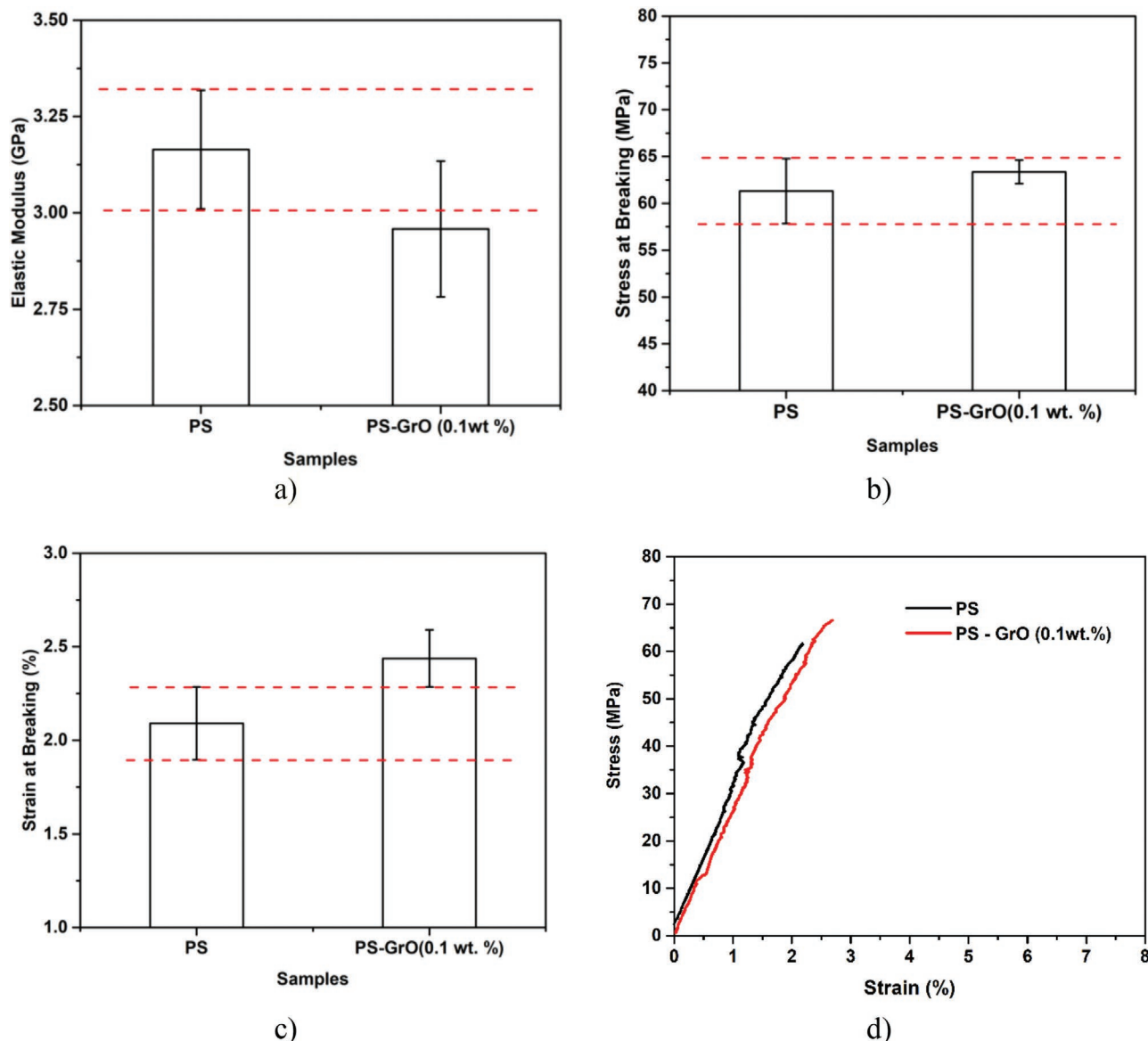


Figure 4. a) Elastic modulus, b) stress, and c) strain at breaking values and d) stress–strain curves of PS and the PS-GrO (0.1 wt%).

as earlier suggested. At this zone, very fast changes occur in the stress field, characterized by a mixture of Mode I (open crack)/ Mode III (out-plane shear) fractures.^[13] It is believed that the slipping between the flake-flakes contacts out of registry present in the agglomerates has intensified the out-plane shear component, making the Hackles zone strongly perceived for samples with a higher concentration of agglomerate.

In order to observe the behavior of the agglomerates in a brittle matrix, composites based on polystyrene were produced. Polystyrene, due to the absence of tie molecules, is extremely sensitive to defects, such as agglomerates.^[15] The filler added to the PS was GrO, i.e., the added filler has a large number of agglomerated flakes. In addition, to avoid any degree of dispersion, this material was directly molded, without going through the mixture in a twin-screw extruder. Figure 4a–d shows the E , τ_b , ε_b , and stress–strain curve, respectively, of neat

PS and PS-GrO (0.1 wt%). Similarly to HMWPE, the average elastic modulus of PS reduced with addition of GrO, while the ε_b increased significantly and the τ_b was not altered. This led to the conclusion that this type of agglomeration did not act as a defect in the polymer matrix, but acted as a toughening mechanism. However, the agglomerates behavior on mechanical properties of the amorphous polymer was quite similar to the semicrystalline polymer, leading both matrixes to higher deformations and lower modulus.

Many publications have shown that high levels of mGO or graphene added to the polymer lead to the formation of a large amount of agglomerates, causing a reduction in the ultimate mechanical properties (stress and strain at breaking) of the polymer, even with strong adhesion at the polymer-filler interface.^[16] However, note that, in this case, the filler levels added to the polymer are still very low. It is believed that the low filler

content in the polymeric matrices allows the filler to agglomerate in an “organized way.” Ferreira et al. showed through X-ray microtomography of HMWPE-mGO (0.1 wt%) that the agglomerate flakes are arranged in a stacked way, like playing cards. They reported that the agglomerates are organized in a certain way that allows easy slipping between the flakes, giving rise to the superlubricity phenomenon. Consequently, it is possible to observe a beneficial effect on the ultimate tensile properties of polymers containing agglomerates, when there is good adhesion on the interphase.

It is very well known that in semicrystalline polymers the fillers can act as a nucleating agent, changing the morphology of the polymer and consequently changing the mechanical properties.^[17] It is believed that the particle’s nucleating effect is doing little to contribute to the mechanical properties of HMWPE. Figure S3 (Supporting Information) shows that the crystallinity degree of the HMWPE is hardly affected by the insertion of mGO and GrO. Consequently, the reinforcement effect and superlubricity effect of mGO are the main causes of the analyzed composites. Proof of this is the PS-GrO (0.1 wt%) composite. The PS is not crystallizable, that is, the nucleating action of GrO is nonexistent in this case, but even so a considerable increase in the deformation of this polymer due to the presence of GrO agglomerates is observed.

4. Conclusion

In the same way that it was observed in the molten state by Ferreira et al., here the influence of the superlubricity of agglomerates on the polymer in the solid state of a polymer was demonstrated. We believe, as long as the filler content added to the polymer is low, the formed agglomerates can have a beneficial effect on the mechanical properties of the polymer. We have seen that agglomerates can lead the polymer to higher strain at breaking, due to the superlubricity phenomenon, while the well-exfoliated mGO leads the polymer to higher stress at breaking. This observation can be especially important when obtaining nanocomposites with high tensile toughness is desired. The results presented here are a paradigm shift, as it has always been thought that the presence of agglomerates is harmful, but the opposite is proven here. Well-controlled agglomerates can be extremely desirable, and the superlubricity state can also be observed in the polymer on the solid state. In general, this paper can contribute a lot to the development of new polymer composites and nanocomposites produced with fillers that present the superlubricity phenomenon (molybdenum disulfide, graphite, graphene oxide, graphene, carbon nanotube, boron nitride, etc.).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

agglomerates, composites, mechanical properties, superlubricity state, toughening mechanism

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