

Recycling of a multilayer barrier food packaging through the use of a nanofiller: Effect of post-consumer plastic bag conditions

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ABSTRACT

Barrier food packaging films result very difficult to recycle since they are multilayer films composed by at least two different materials. In this context, aim of this study was to assess the use of a nanofiller for the mechanical recycling of multilayer barrier plastic bags used for food vacuum packaging. In particular, graphene nanoplatelets (GnPs) were used as nanofiller studying the effect of their concentration on the properties of the nanocomposites obtained by using polyamide (PA)/polyethylene (PE) bags as polymeric matrix. Moreover, the effect of plastic bags life conditions on the nanocomposite properties were evaluated by exposing the multilayer films to accelerated weathering or to contact with a fatty food simulant (olive oil). The results indicated that the incorporation of GnPs led to an enhancement of the morphology of the polymer blends, consequently resulting in nanocomposites with increased stiffness without a drastic reduction in ductility. The nanocomposites produced using post-consumer bags exhibited lower properties compared to materials produced using pre-consumer bags. However, even in this case, the addition of GnPs contributed to an improvement in the properties demonstrating that the use of this nanofiller can be a viable method to recycle multilayer packaging.

1. Introduction

Today, plastic production has increased globally and almost half of it is used in the packaging sector, which ultimately becomes waste due to its brevity of use [1]. Recycling or reusing plastic is therefore essential to prevent the unstoppable increase of polymeric materials being released into the environment and to minimize environmental pollution [2,3].

Mechanical recycling is a possible way to valorize the polymeric materials in circulation, as it is economical, large-scale, solvent-free and applicable to many polymers [4–7]. However, the quality of the final products largely depends on the quality of the selection made on the product to be recycled. In fact, chemical changes in the structure of the polymer during the previous treatment and its lifetime can affect the deterioration of the physical properties and mechanical performance of the recycled product [8,9]. In addition, it is not advisable to recycle polymeric materials of different chemical nature in a single phase, as the strong diversity between the chemical and molecular structures of the constituent polymers will create a brittle polymer system with poor mechanical properties and unsightly appearance [10]. This is one of the

reasons why so far, little attention has been paid to the mechanical recycling of multilayer packaging, compared to other methods that are more resource- and cost-intensive, such as those in which the target polymers are recycled through dissolution and precipitation [11–13], or through delamination processes by separating the different layers [14, 15]. However, recently, some studies have focused on the recycling of multilayer food packaging in a single flow without separation. These studies involve the use of compatibilizers to enhance the stability and mechanical properties of these systems [16–18]. Nevertheless, while the use of compatibilizer additives results in recyclable multilayer material, the possibility of their migration can be a limiting factor for its broad commercial use.

The use of a nanofiller could represent a valid alternative to the use of compatibilizers, given that they have been employed to improve the morphology and consequently, the properties of many polymer blends. Over the years, graphene nanoplatelets (GnPs) have attracted much attention for their ability to improve properties such as thermal, mechanical and electrical properties of polymer nanocomposites as well as their behaviour thermomechanical and photo oxidative degradation

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Table 1

Relevant properties of the plastic bags used in this work.

	Thickness, μm	Yield Strength, N/15 mm	Elongation at Break, %
Orved type	90	MD ^a ≥ 45	MD ^a ≥ 200
95		TD ^b ≥ 35	TD ^b ≥ 250

^a MD = machine direction.^b TD = transversal direction.

[19–24]. Today, their effectiveness as compatibilizers is being discussed along with other nanoparticles. In reality, rather than a true compatibilizing action at the polymer-polymer interface, nanoparticles are believed to be able to extensively alter the microstructure of the blend, which is attracting particular interest in the field of recycled plastics [25].

In this context, aim of this study was to assess the use of graphene nanoplatelets for the mechanical recycling of multilayer barrier plastic bags used for food vacuum packaging. Specifically, pre-consumer polyamide (PA)/polyethylene (PE) bilayer commercial packaging was melt-mixed with different amounts of nanofiller in order to evaluate the concentration's effect on the resulting materials' properties. Furthermore, nanocomposites were produced using plastic bags subjected to accelerated weathering or in contact with a fatty food simulant (olive oil). The materials underwent comprehensive characterization, including morphological analysis, rheological characterization and tensile tests.

2. Materials and methods

2.1. Materials

The multilayer barrier food packaging used in this work was a polyamide (PA)/polyethylene (PE) plastic bags supplied by Orved (Orved S.P.A., Venezia, Italy) under name commercial Orved type 95. Table 1 shows some main properties of the used plastic bags [26].

The nanofiller used in this study was a sample of graphene nanoplatelets (GnPs) supplied by XG Sciences Inc (Lansing, MI, USA). The specifications provided by the manufacturer are as follows: surface area of approximately 750 m²/g and average diameter between 1 and 2 μm .

2.2. Accelerated weathering

In order to examine the impact of environmental conditions on plastic bags, they underwent accelerated weathering tests in a QUV chamber (Q-Labs Corp., Westlake, OH, USA). The chamber was equipped with UVB-313 fluorescent lamps, and the bags were exposed to these lamps for approximately 72 h. The exposure involved a cyclic pattern of 8 h of light at 55 °C followed by 4 h of condensation at 45 °C, following a modified ISO 4892-3 method [27]. In this case, the layer exposed to fluorescent lamp was that of polyamide (PA).

2.3. Contact with fatty food simulant

To simulate the wetting and lubrication of solid food on the plastic bags, they were exposed to fatty food simulant (olive oil) for a period of 30 days at temperature of 25 °C and relative humidity (RH) of 50%. In this case, the layer exposed to contact with the food simulant was that of polyethylene (PE).

Fig. 1 shows a schematic of the arrangement of the plastic bags and photos during exposure to food simulant.

2.4. Melt compounding

Plastic bags were ground and compounded in a batch mixer Brabender mod. PLE330 (Brabender, Duisburg, Germany) set to operate at 240 °C and 60 rpm for 5 min. Before blending, both plastic bags and GnPs were dried in a vacuum oven overnight at 90 °C and 120 °C, respectively.

Tables 2 and 3 shows of the compositions of all investigated systems.

All the samples for further characterizations were obtained by hot compression molding using a Carver laboratory hydraulic press (Carver, Wabash, IN, USA). The hot pressing process was conducted at a temperature of 240 °C with a mold pressure of 300 psi for approximately 3 min.

Fig. 2 illustrates the methodology used in this work.

2.5. Characterization

2.5.1. Rheological characterization

Rheological tests were performed through a rotational parallel-plate

Table 2

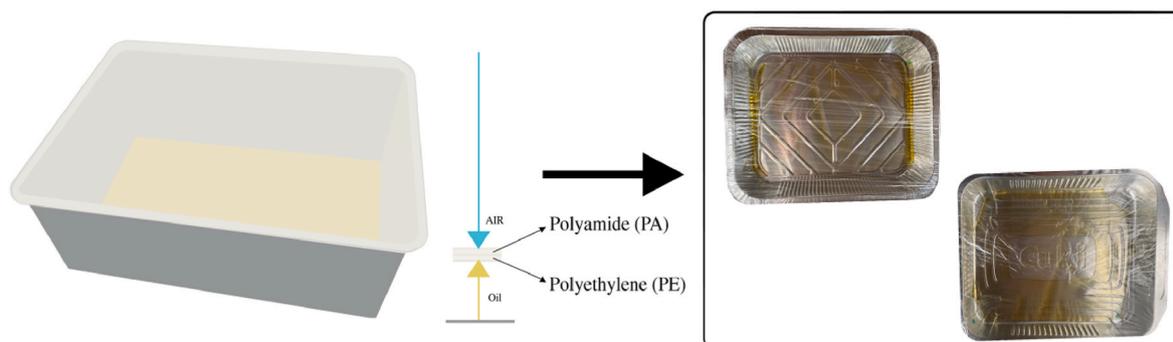
Composition of all investigated systems using PA/PE Bags.

	PA/PE Bags, %	GnPs, %
PA/PE Bags	100	–
PA/PE Bags +1% GnPs	99	1
PA/PE Bags +3% GnPs	97	3
PA/PE Bags +5% GnPs	95	5

Table 3

Composition of all investigated systems using weathered bags (PA/PE Ph) and bags exposed to the contact with the olive oil (PA/PE FS).

	PA/PE Bags Ph, %	PA/PE Bags FS, %	GnPs, %
PA/PE Bags Ph ^a	100	–	–
PA/PE Bags Ph +5% GnPs	95	–	5
PA/PE Bags FS ^b	–	100	–
PA/PE Bags FS +5% GnPs	–	95	5

^a Ph = photo-oxidized samples.^b FS = samples exposed to a food simulant.**Fig. 1.** – Scheme and visual images during exposure to food simulant (olive oil).

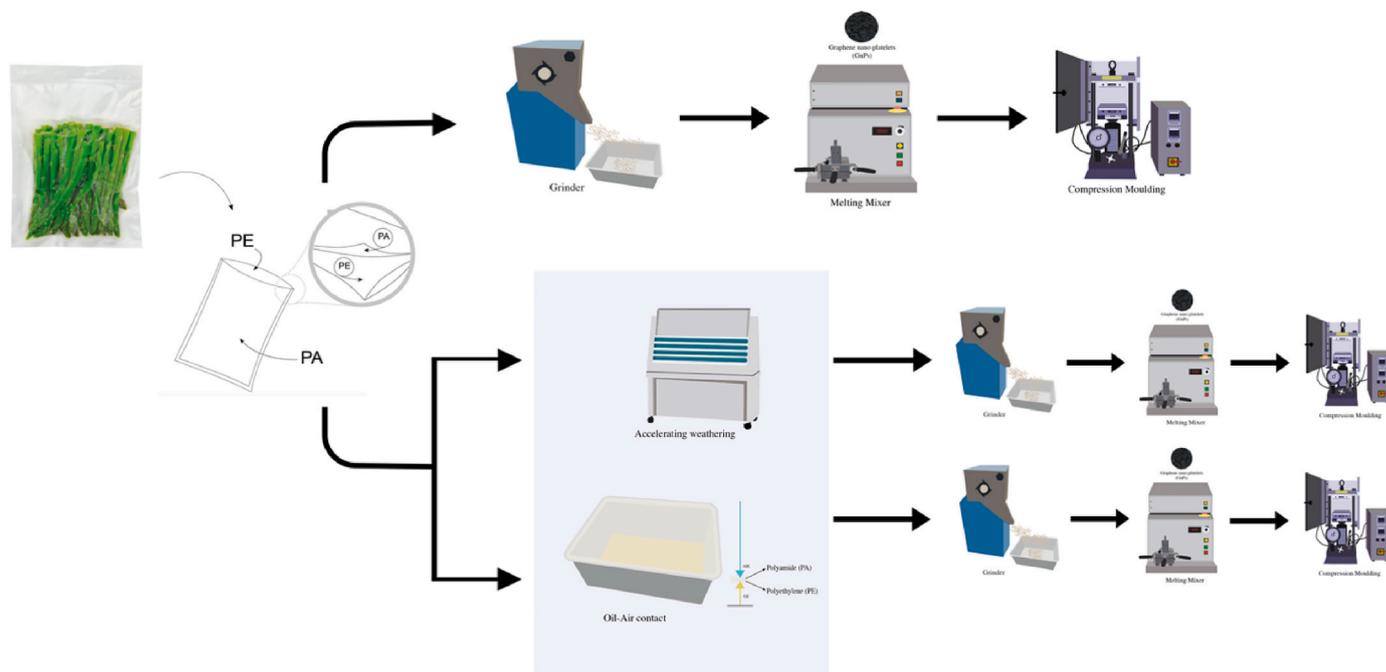


Fig. 2. Scheme of the methodology adopted in this work.

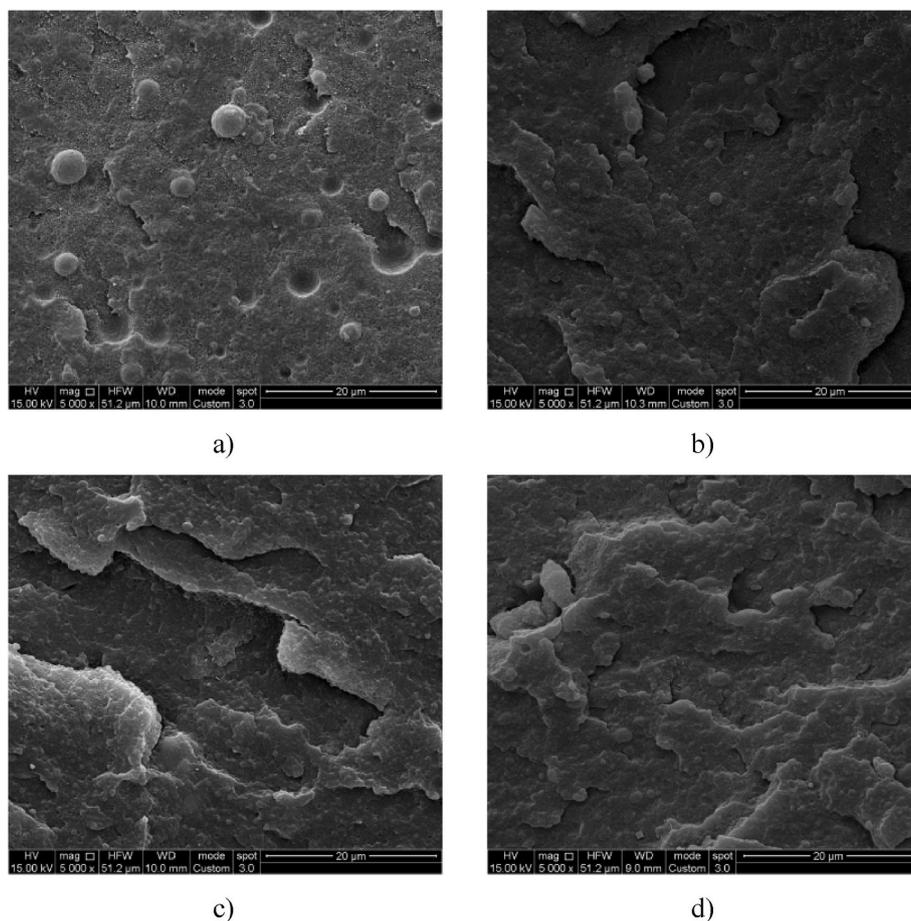


Fig. 3. – SEM micrograph: a) PA/PE Bags, b) PA/PE Bags +1% GnPs, c) PA/PE Bags +3% GnPs, d) PA/PE Bags +5%GnPs.

rheometer ARES G2 (TA Instruments, New Castle, DE, USA) (plate diameter 25 mm). Measurements were performed over an angular frequency range from 0.1 to 100 rad/s at a temperature of 240 °C.

2.5.2. Tensile mechanical characterization

Tensile tests were conducted at room temperature using an Instron universal testing machine (Instron, mod. 3365, High Wycombe, UK)

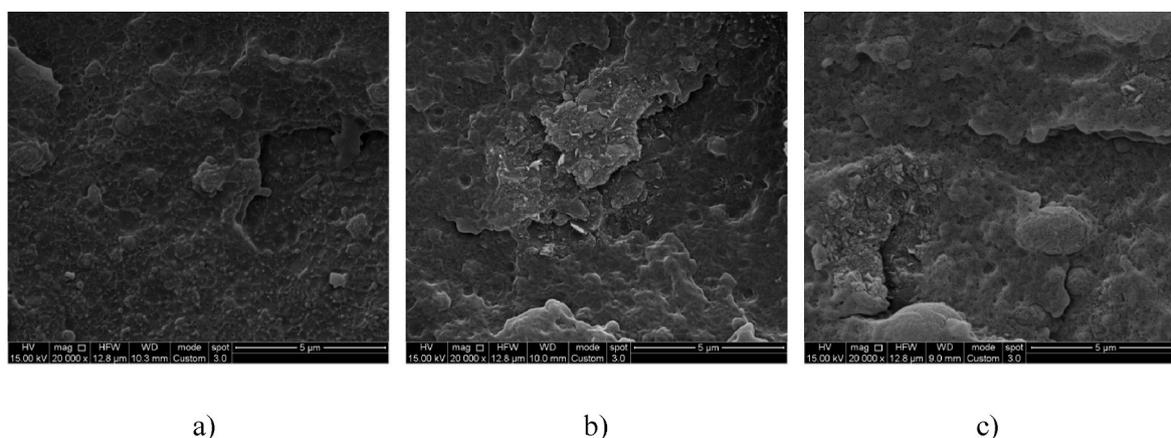


Fig. 4. – High magnification SEM micrograph: a) PA/PE Bags +1% GnPs, b) PA/PE Bags +3% GnPs, c) PA/PE Bags +5%GnPs.

according to ASTM D638-14. Rectangular specimens (length = 90 mm, width = 10 mm and thickness = 0.5 mm) were tested at a rate of 5 mm/min until failure. The mean value of the mechanical tests with the corresponding standard deviation is the result of ten measurements. Elastic modulus, E , tensile strength, TS, and elongation at break, EB, were the outcomes of the tests.

2.5.3. Morphological analysis

The fracture surfaces, obtained by soaking the specimens in liquid nitrogen for about 30 min and broken in a brittle manner, were observed with a Quanta 200 F (FEI Co., Hillsboro, OR, USA) scanning electron microscope (SEM). Prior to analysis, the samples were gold sputtered to make them conductive. Image analysis was conducted using ImageJ software, which is freely available and in the public domain.

2.5.4. Thermal analysis

Thermal analysis was performed using differential scanning calorimetric (DSC) using a Chip-DSC 10 (Linseis Messgeraete GmbH, Selb, Germany). The amount of sample placed in the DSC aluminum pans was about 7 ± 2 mg, while the heating rate was from 10 °C/min up to 280 °C.

3. Result and discussion

3.1. Recycle of pre-consumer packaging

In Fig. 3a–d the SEM micrographs, at 5.000x magnification, of PA/PE Bags and nanocomposites with 1, 3 and 5% of GnPs are reported.

Fig. 3a shows the typical morphology of an immiscible and incompatible blend. In particular, it can be seen that the particles of the non-dispersed phase are quite distinguishable (4.5 ± 0.3 μm), and a poor interfacial adhesion is evidenced by the presence of voids caused by the detachment of the PA particles during breakup. By adding GnPs, see Fig. 3b–d, there is a general improvement in morphology: interfacial adhesion is improved, as evidenced by the absence of voids, and the spherical particles of the non-dispersed phase are less and less visible as the GnPs content increases (from 4.5 ± 0.3 μm for PA/PE Bags to less than 1.2 ± 0.1 μm for PA/PE Bags +5% GnPs). This behavior has already been found in other systems [28–31] and has been attributed to the interfacial localization of the nanofiller that prevents coalescence of droplets during mixing. In fact, nanofillers can act as a shield and stabilize the morphology of the blend.

High magnification micrograph of nanocomposites, see Fig. 4, provide further direct evidence of the effect of GnPs on plastic bags.

Fig. 5 shows the complex viscosity, η^* , curves as a function of angular frequency for PA/PE Bags and nanocomposites.

As visible in Fig. 5, the complex viscosity curves show that GnPs clearly influence the response of nanocomposites. In particular, it is

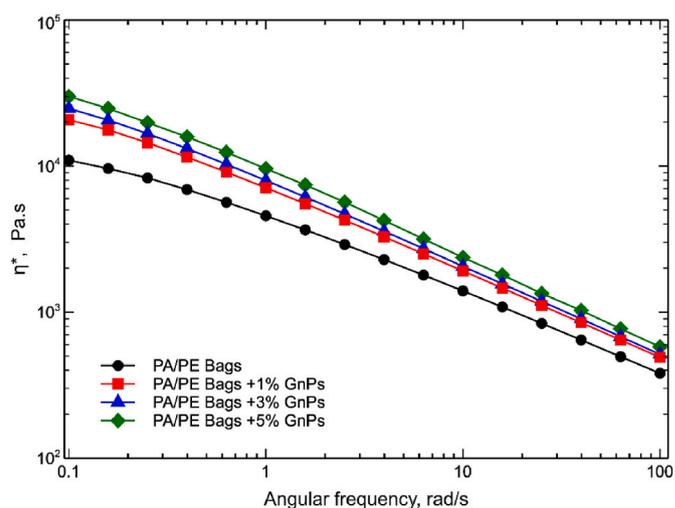


Fig. 5. – Flow curves with complex viscosity (η^*) as a function of angular frequency of PA/PE Bags and nanocomposites.

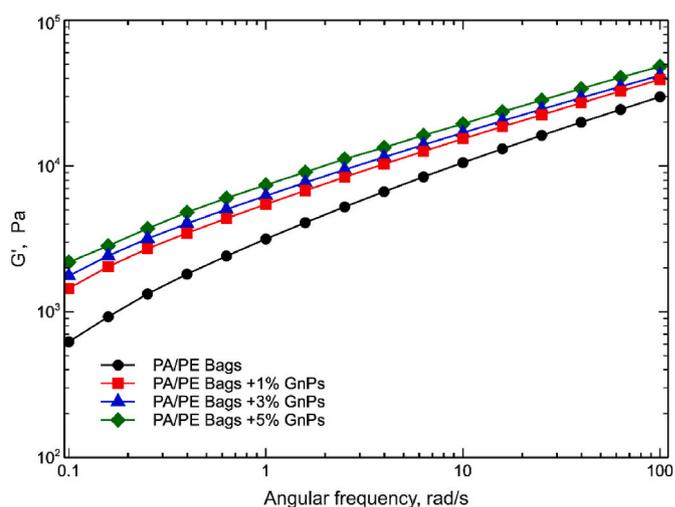


Fig. 6. – Storage modulus (G') curves as a function of angular frequency of PA/PE Bags and nanocomposites.

observed that the complex viscosity increases as the amount of GnPs increases at low frequencies, with slightly more pronounced non-Newtonian behavior at high frequencies compared to the pristine

Table 4

Mean tensile test results and the relative standard deviation of PA/PE Bags and nanocomposites.

	E, MPa	TS, MPa	EB, %
PA/PE Bags	312 ± 33	13.8 ± 1.2	95 ± 18
PA/PE Bags +1% GnPs	331 ± 37	14.0 ± 1.4	87 ± 16
PA/PE Bags +3% GnPs	351 ± 41	14.2 ± 1.5	72 ± 13
PA/PE Bags +5% GnPs	375 ± 44	15.2 ± 1.9	58 ± 9

Table 5

Differential scanning calorimetry (DSC) analysis of PA/PE matrix and relative increases for nanocomposites at 1, 3, and 5 wt% GNPs.

	Scan	T _{PE} , °C	T _{PA} , °C	Δ H _{PE} , J/g	Δ H _{PA} , J/g
PA/PE	1	125.6	223.0	-90.5 ± 0.9	-18.7 ± 0.8
	2	104.2	186.0	81.6 ± 0.5	9.0 ± 0.7
	3	123.9	223.1	-89.6 ± 0.7	-18.3 ± 0.4
PA/PE +1% GnPs	1	125.7	224.5	-91.2 ± 1.1	-18.5 ± 0.7
	2	97.4	183.9	86.9 ± 1.2	9.37 ± 1.5
	3	124.4	224.4	-90.3 ± 1.0	-18.5 ± 0.5
PA/PE +3% GnPs	1	126.4	225.6	-91.3 ± 1.3	-18.7 ± 0.7
	2	95.4	186.9	87.0 ± 1.8	9.63 ± 1.9
	3	127.1	227.3	-90.4 ± 1.3	-19.2 ± 0.7
PA/PE +5% GnPs	1	127.8	226.5	-91.6 ± 1.5	-19.0 ± 1.0
	2	95.3	188.2	89.6 ± 1.2	10.1 ± 1.2
	3	128.2	228.3	-90.5 ± 1.4	-21.0 ± 1.7

matrix. This behaviour, commonly ascribed to the interaction between the dispersed filler and the polymeric matrix, which limits the mobility of the polymer chains, is in agreement with other nanocomposite systems [32,33] including polymer/GnPs nanocomposites [24,34].

Fig. 6 shows the storage modulus, G' as a function of angular frequency for PA/PE Bags and nanocomposites.

The viscoelastic behavior of the three nanocomposites, differs from that of the pristine PA/PE Bags matrix. The modulus values are slightly higher, with slight pseudo-solid behavior at lower angular frequencies. Based on the scientific literature [35,36], it can be stated that the presence of GnPs lead to an intensification of the solid behavior of the material, reinforcing what has already been said about the increase in viscosity.

The results of the tensile properties measured on the compression molded specimens are shown in Table 4.

It is observed that the addition of GnPs leads to an increase in elastic modulus, which is more evident in the nanocomposites with 5 wt% GnPs. More in detail, the elastic modulus of PA/PE Bags +5% GnPs increased by approximately 20%. The tensile strength also showed a significant increase, with a high improvement observed in the same nanocomposites, where the tensile strength increased by 10% compared to the pristine matrix, while a slight improvement was observed in the nanocomposites with 1 and 3 wt% GnPs. However, the presence of GnPs decreases the elongation at break for all samples analyzed. In particular, the higher the tensile strength, the more the elongation at break is reduced. The sample with 5 wt% GnPs showed the highest reduction (-39%), but good ductility was still observed.

Table 5 provides a summary of the results from the differential scanning calorimetry (DSC) analysis. Firstly, the peak temperatures confirm the information stated in the technical data sheet regarding the presence of the two components, polyethylene and polyamide. Secondly, the nanocomposites containing GnPs particles exhibit slight increases in ΔH compared to the pristine matrix. Notably, the ΔH of both components, polyethylene and polyamide, demonstrates an upward trend with increasing filler concentration. Moreover, a marginal elevation in melting temperature is evident for both components with the augmentation of GnPs concentration. This phenomenon is well-documented in scientific literature and is attributed to the presence of

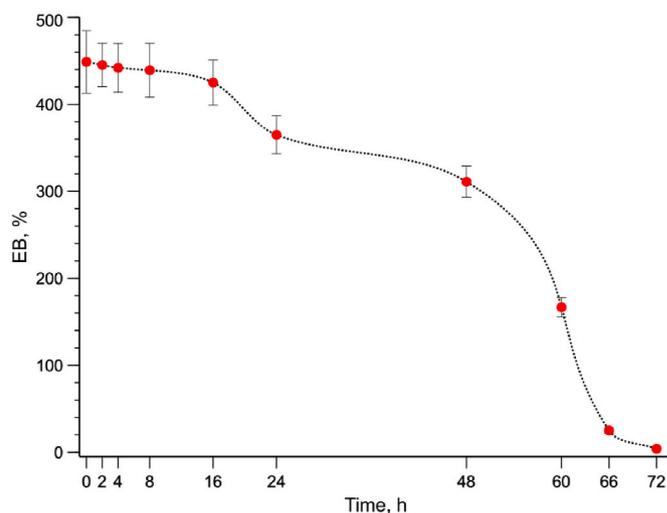


Fig. 7. - Elongation at break, EB, as a function of weathering time of the PA/PE Bags.

the nanofiller, which serves as a nucleating agent [37].

3.2. Recycle of post-consumer packaging

In an attempt to recycle these materials after use, the objective is to replicate the behavior of naturally weathered bags. Therefore, to assess the comparability of exposure to bags used in practical applications, the bags were artificially aged by placing them inside laboratory chambers that simulate the main degradation characteristics of materials in order to accelerate the degradation phenomenon and predict the outside impact of degradation. To simulate real food, the bags were placed inside trays in contact with a food simulant. As the latter depends on the type of food to be simulated, the simulant selected was olive oil, which simulates fatty food products.

All bags were characterized (section 3.2.1 and 3.2.2) and, subsequently, blended with and without GnPs to evaluate the possibility of recycling (section 3.2).

3.2.1. Accelerating weathering

As already reported [38], elongation at break serves as a reliable parameter for assessing the durability of polymeric materials under accelerated aging conditions, as it is highly sensitive to molecular and morphological alterations. In this regard, the durability of multilayer packaging to weathering was monitored by elongation at break.

Fig. 7 show the evolution of elongation at break, EB, as a function of weathering time.

It can be observed that elongation at break rapidly decreases with exposure time. In fact, a decrease of about 30% from the initial value is observed at 48h. Usually, the time at which the elongation at break reaches half of its initial value, i.e. $t_{1/2}$ [23] is adopted as a parameter useful to assess the durability of a polymer film subjected to accelerated ageing. However, in this case it was observed that, the material properties were practically compromised at $t_{1/2}$. Therefore, a specific time frame was determined during which the material properties exhibited characteristics that allowed for recycling and, furthermore, enabled the material to be processable. This time frame was identified as 48 h since, at this time point, the elongation at break had decreased to a level that still allowed for the material to be mechanically processed and recycled efficiently.

3.2.2. Contact with fatty food simulant (olive oil)

The used food simulant, i.e., olive oil, was chosen to reproduce the characteristics of a high-fat food, in order to subject the packaging to a stress similar to that to which they could be exposed under real

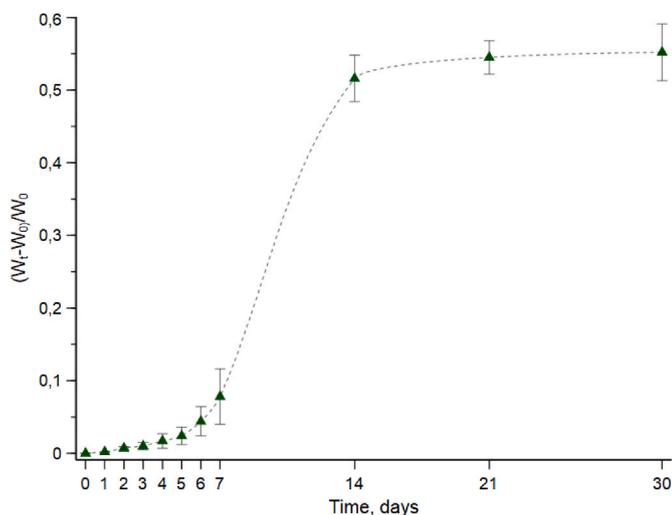


Fig. 8. – Percentage of weight gain during treatment with food simulant (olive oil).

Table 6
Mean tensile test results and the relative standard deviation of PA/PE Bags and PA/PE Bags FS.

	E, MPa	TS, MPa	EB, %
PA/PE Bags	403 ± 25	30.8 ± 2.9	449 ± 36
PA/PE Bags FS	297 ± 23	23.7 ± 1.6	508 ± 34

conditions of use. This is particularly important considering that the inner layer of the bag is made of polyethylene, a polymer material that is particularly prone to interact with fats. To measure the absorption level

of the food simulant, the weight gain of the PA/PE bags as a function of the contact time was collected and the results are displayed in Fig. 8.

The amount of absorption was observed to be slow for the first 7 days and proceed at a rapid rate thereafter (7–14 days) reaching a plateau at 30 days. This time was defined as the maximum contact time.

Table 6 shows the mean tensile test results and the relative standard deviation of the film before and after contact with food simulant.

As can be seen from Table 5, the treatment does not induce a significant deterioration of the mechanical properties. There was a slight increase in flexibility attributable to the fact that, as the contact time increases, the food simulant migrates, probably, through the polyethylene layers reaching the intermediate layers and producing plastification and mechanical relaxation.

3.3. Properties of recycled post-consumer PA/PE bags

Based on the above results, this section presents the results of recycling PA/PE Bags subjected to 48-h accelerated weathering and 30-day exposure to the food simulant with and without GnPs.

Fig. 9 shows the micrograph of recycled PA/PE Bags Ph and PA/PE Bags FS with and without GnPs compared with recycled pre-consumer PA/PE Bags.

As previously described, the recycled pre-consumer PA/PE Bags, see Fig. 9a, show a surface relatively sharp, with the typical morphology of an immiscible and incompatible blend. In the micrograph of weathered samples, see Fig. 9b', a reduction in the size of PA domains and voids can be observed, with less sharp interface surfaces. As for the nanocomposite, see Fig. 9b'', the interface surfaces are slightly sharper and the morphology becomes more homogeneous, such that the PA domains in the polyethylene matrix are almost undetectable in the micrographs.

As regard the samples exposed to food simulant, it can be observed that for PA/PE Bags FS samples, no significant changes in morphology occurred. On the contrary, for the nanocomposite, the addition of GnP leads to a reduction in the size of PA domains and voids.

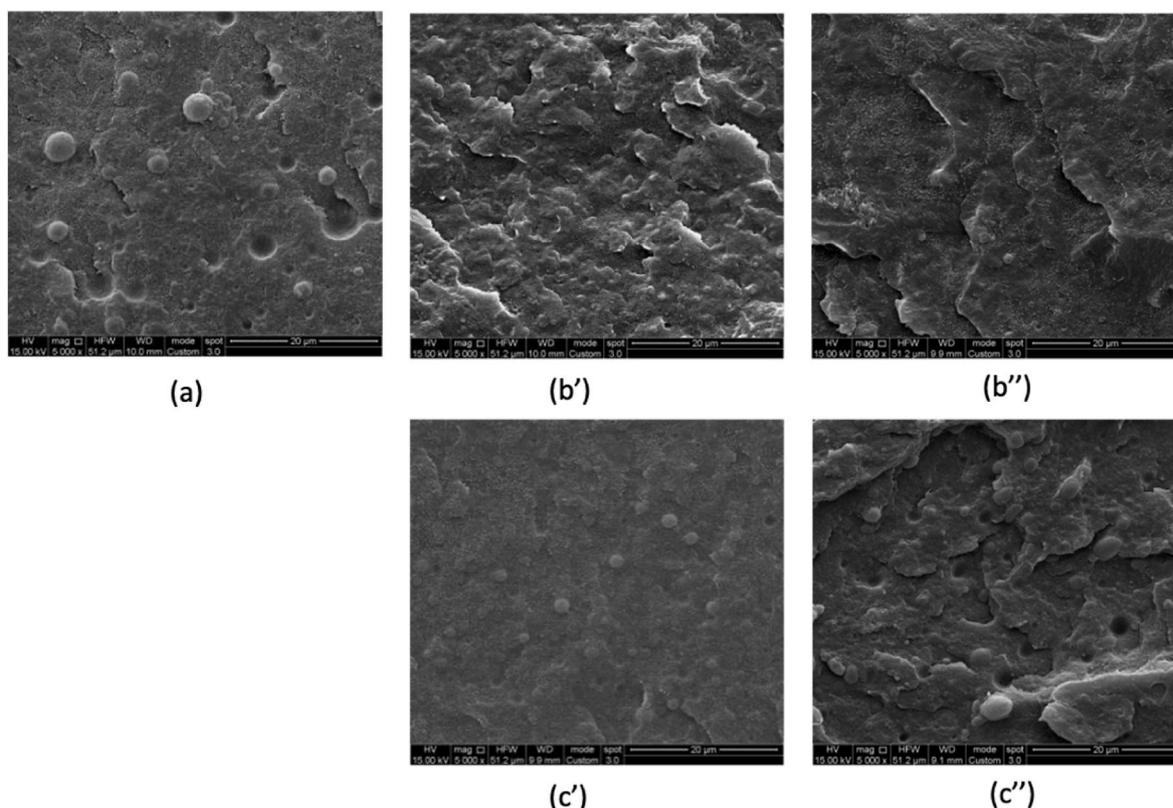


Fig. 9. - SEM micrographs: a) PA/PE Bags b') PA/PE Bags Ph b'') PA/PE Bags Ph +5% GnPs, c') PA/PE Bags FS, c'') PA/PE Bags FS +5% GnPs.

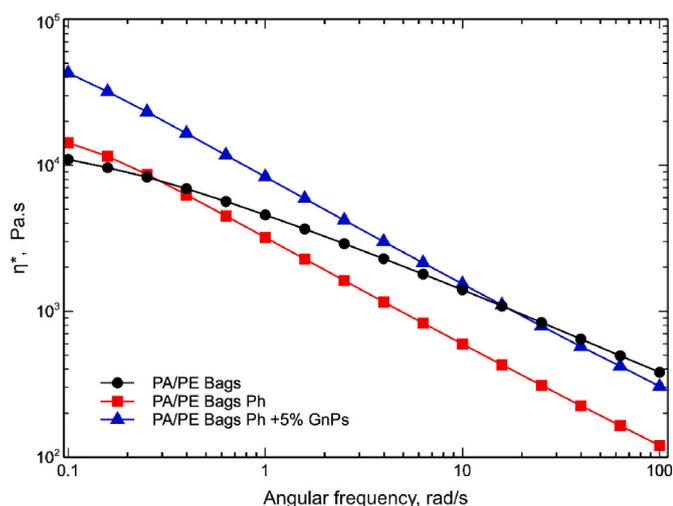


Fig. 10. – Flow curves with complex viscosity (η^*) curves as function angular frequency using PA/PE Bags Ph.

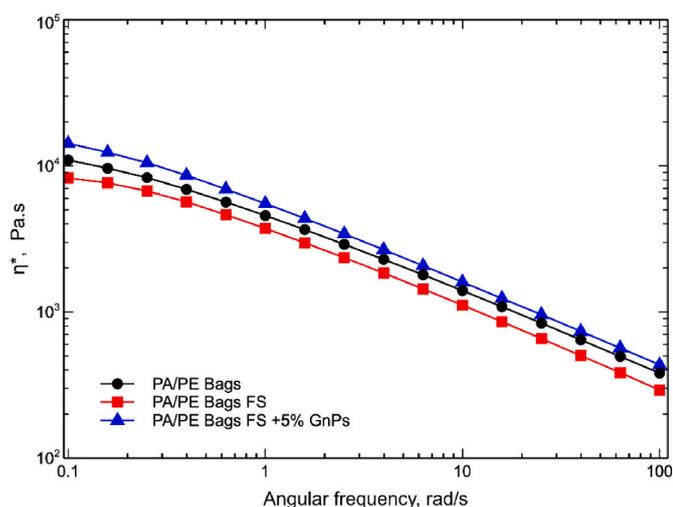


Fig. 11. - Flow curves with complex viscosity (η^*) curves as function angular frequency using PA/PE Bags FS.

Figs. 10 and 11 shows the complex viscosity curves of 5% GnPs nanocomposites using photo-oxidized PA/PE Bags and PA/PE Bags exposed to food simulant as matrix, respectively.

As expected, the viscosity curve of the photo-oxidized sample, PA/PE Bags Ph, is lower than that of the PA/PE Bags matrix. This provides clear evidence of chain breaking resulting from photo-oxidative processes, which in turn leads to a decrease in molecular weight. Additionally, the high viscosity value at low frequencies of the photo-oxidized sample suggests the presence of branched and/or cross-linked structures. Upon adding 5% of GnPs to PA/PE Bags Ph, viscosity increases compared to the respective matrix. However, the flow curve exhibits a similar behavior to that of PA/PE Bags Ph without GnPs. Conversely, the viscosity of the sample exposed to the food simulant contact decreases due to the plasticizing effect of oil, without any significant effect on the curve slope. Adding 5% GnPs to PA/PE Bags FS causes an increase in viscosity as the presence of the filler prevails over the plasticizing effect of the oil, with a slightly more pronounced non-Newtonian behavior.

Figs. 12 and 13 show histograms of the tensile properties for PA/PE Bags Ph and PA/PE Bags FS, with and without GnPs, respectively, and compare them to PA/PE Bags.

The results shows that photo-oxidation, see Fig. 12, clearly affected the mechanical properties of recycled weathered samples. Specifically, a

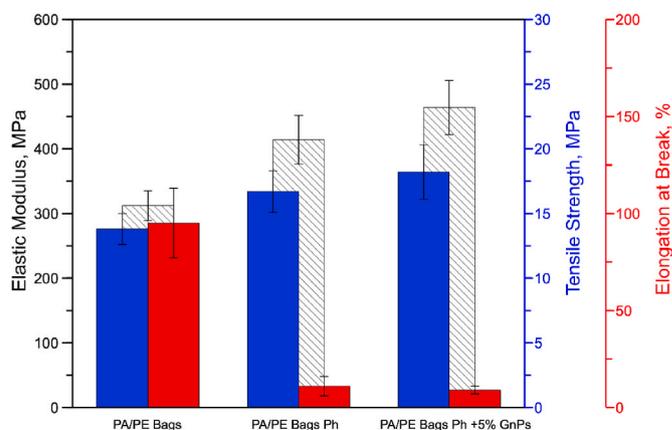


Fig. 12. – Bar chart of tensile properties using PA/PE Bags Ph.

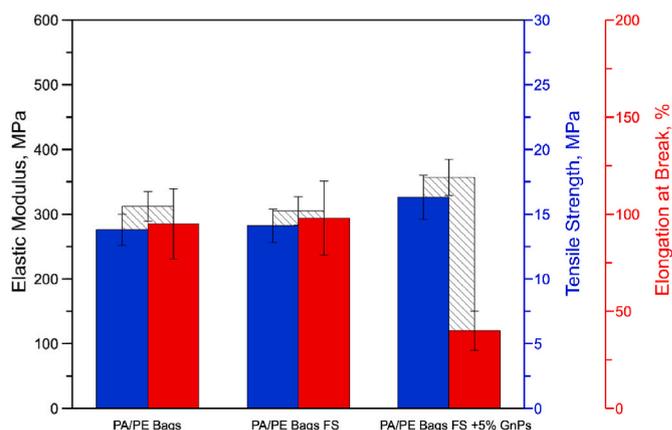


Fig. 13. - Bar chart of tensile properties using PA/PE Bags FS.

drastic increase in elastic modulus was observed in PA/PE Bags Ph samples compared to the untreated PA/PE Bags together with a strong reduction of the elongation at the break. These results, can be attributed to the decrease in molecular weight which led to an increase in the crystallinity degree. Adding GnPs led to stiffening of the material and a slight further reduction of the ductility. Specifically, the elastic modulus of PA/PE Bags Ph increased from 414 ± 38 MPa to 464 ± 42 MPa upon adding 5% GnPs. However, while tensile strength slightly increased, elongation at break decreased significantly. More in detail, the elongation at break of PA/PE Bags was $95 \pm 18\%$, while that of PA/PE Bags Ph and PA/PE Bags Ph + 5% GnPs was $11 \pm 5\%$ and $9 \pm 2\%$, respectively.

In contrast to photo-oxidation, exposure to food simulant, as mentioned earlier, had minimal impact on the mechanical properties of PA/PE Bags. The values between PA/PE Bags and PA/PE Bags FS were very similar, as shown in Fig. 13. However, the addition of GnP resulted in an overall stiffening of the structure, leading to a slight increase in both the elastic modulus and the tensile strength, but a non-negligible decrease in elongation at break compared to that of PA/PE Bags FS.

4. Conclusions

In this work, it was studied the mechanical recycling of multilayer barrier plastic bags used for food vacuum packaging. In particular, it was evaluated the use of a bilayer PA/PE packaging as polymeric matrix for producing nanocomposites filled with GnPs. The incorporation of the nanofiller led to an improvement of the polymer blends morphology, i.e. a reduction in the size of the dispersed phase and an increase in viscosity was observed throughout the studied frequency range. On the other hand, with the increase in filler content, an increase in elastic modulus

and tensile strength has been observed, while there has not been a drastic reduction in elongation at break due to good interfacial adhesion between phases. The samples obtained from post-consumer PA/PE Bags, i.e. multilayer films exposed to accelerated weathering or to contact with an olive oil, exhibited reduced performances because of simulated life conditions, particularly in the case of weathered samples. However, again, the use of GnPs allowed to obtain nanofilled polymer blends with good morphology and mechanical performance, especially in terms of improved elastic modulus and tensile strength, thus demonstrating that the use of GnPs can be a viable method to recycle multilayer packaging.

CRedit author statement

Maria Chiara Mistretta: Data curation, Visualization, Writing - Review & Editing, Supervision. **Vincenzo Titone:** Investigation, Data curation, Writing—original draft preparation, Writing - review & editing. **Francesco Paolo La Mantia:** Visualization, Writing - Review & Editing, Supervision. **Valeria Pellitteri:** Investigation. **Luigi Botta:** Conceptualization, Investigation, Methodology, Writing—original draft preparation, Writing - Review & Editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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