



Wood plastic composite using graphene nanoplatelets



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ABSTRACT

This article presents the preparation and characterization of wood flour/polypropylene (PP) composites filled with graphene nanoplatelets (GNPs). The effects of GNPs, as reinforcing agent, on the mechanical and physical properties were also investigated. In order to increase the interphase adhesion, maleic anhydride grafted polypropylene (MAPP) was added as a coupling agent to all the composites studied. The morphology of the specimens was characterized using scanning electron microscopy (SEM) technique. The results of strength measurements indicated that when 0.8 wt.% GNPs were added, tensile and flexural properties reached their maximum values. At high levels of GNPs loading (3–5 wt.%), increased population of GNPs leads to agglomeration and stress transfer gets blocked. The addition of GNPs filler moderately increased the impact strength of composites. Addition of GNPs decreased the average water uptake and thickness swelling by 35% and 30%, respectively, compared to the control sample (without GNPs). It was observed that the composites filled with GNPs decomposed at higher temperatures compared to the pure PP and control. In all cases, the degradation temperatures shifted to higher values after the addition of GNPs. The improvement of physical and mechanical properties of composites confirmed that GNPs have good reinforcement and the optimum effect of GNPs was achieved at 0.8 wt.%.

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1. Introduction

Now-a-days, the use of wood polymer composites (WPCs) has tremendously increased due to their various advantages. They are used in different outdoor and indoor applications such as decking, railing, fencing, docks, landscaping timbers, and in a number of automobile industries [1]. The lignocellulosic fibers offer a combination of attractive properties such as low density, renewability, biodegradability, wide availability, and low cost, which make them alternatives to traditional synthetic fibers in many applications. However, WPCs exhibit lower physical (higher water absorption and thickness swelling), mechanical (less flexural and tensile strength) as well as thermal properties compared with traditional synthetic composites [2]. There are two major approaches to enhance physical and mechanical properties of WPCs through the use of fillers: by treating them with coupling agents, and by changing their particle size [1]. Chemical coupling agents are substances, typically polymers, which are used in small quantities to treat a surface so that bonding occurs between the treated surface and other surfaces, e.g. wood and thermoplastics [3]. The incorporation of nanoparticles as reinforcing fillers is another method for improving the overall properties of WPCs. The nanoparticles are

certainly one of the most promising reinforcements that are being studied by many scientists all over the world. This is mainly due to their high surface area, low density and high Young's modulus, among other properties that can be transferred to the polymeric matrix if a good dispersion of the filler is ensured in the polymer [4]. A nano-scale filler offers larger effective surface area than a micro-scale filler. As a result, there is more interphase region in a nanocomposite than in a microcomposite. Therefore, the properties of the polymer in the inter-phase region dominate the overall properties of the bulk nanocomposite. In other words, nanofillers can alter the properties of a large volume fraction of the polymer resulting in a significant change in properties at low loading [5].

Carbon is a unique and very versatile element which is capable of forming different architectures at the nano-scale. In this era of nanocomposites research, new members of the carbon nano-scale family have arisen, and more are going to come [6]. Until recently, carbon nanotubes (CNTs) were dominant nanosized carbon fillers for polymer composites [4,7]. However, effective utilization of CNT is hindered by the complexity of their dispersion in polymer matrix and high cost. One of the alternatives is graphene, the material which has become one of the hottest topics in physics and materials science [8–12]. As a matter of fact, from a geometric perspective (Fig. 1), carbon nanostructures like CNTs can be obtained by the coiling up of the covalent graphene building units resulting in a three dimensional structure [13].

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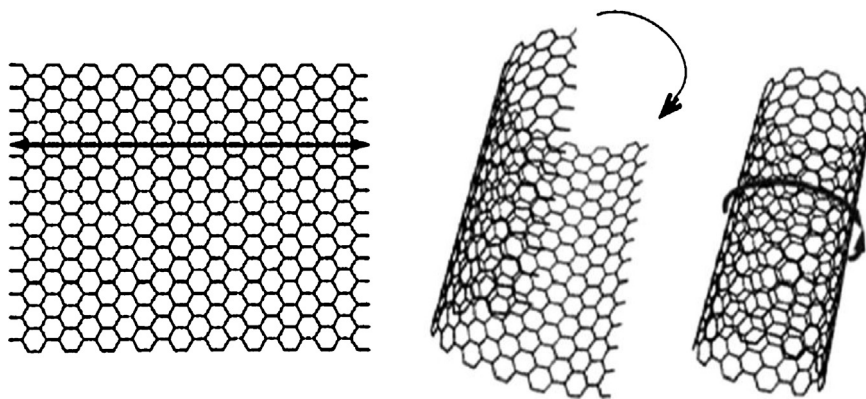


Fig. 1. Rolling up of graphene sheet to form carbon nanotubes.

Graphene has attracted tremendous attention from the experimental and theoretical scientific communities in recent years due to its unique mechanical and physical properties [14–17]. It is two-dimensional allotrope of carbon, formed by single layer of carbon atoms, bonded by sp^2 orbitals into hexagonal two-dimensional crystal lattice. Besides these ideal structures, controlled reduction of graphite oxide yields reduced graphene oxide sheets, which contain a few layers of graphene. The structures with several layers of carbon atoms are also named graphene nanoplatelets (GNP). They are much easier to produce, compared with CNTs and controlled reduction allows preparing nanoplatelets with different oxidation levels and different electronic properties [18]. Graphene, with its unique physical properties is a multifunctional nanofiller that can improve electrical, thermal, mechanical, or gas barrier properties of composites at extremely small loadings. The superior properties of graphene compared to polymers are also reflected in polymer/graphene nanocomposites. Polymer/graphene nanocomposites show superior mechanical, thermal, gas barrier, electrical, and flame retardant properties compared to the neat polymer [19,20]. It was also reported that the improvements in mechanical and electrical properties of graphene based polymer nanocomposites are much better in comparison to that of clay or other carbon filler-based polymer nanocomposites [12,21,22]. Liang et al. [23] demonstrated a solution-processed poly(vinyl alcohol) (PVA)–graphene oxide (GO) nanocomposites and a 76% increase in tensile strength and 62% improvement of Young's modulus were achieved at only 0.7 wt.% of GO. Rafiee et al. [24] compared reinforcement efficiency of graphene with single- or multi-walled carbon nanotube (CNT) and the superiority of graphene platelets over CNT was fully exemplified at a nanofiller weight fraction of 0.1–0.002%.

As mentioned earlier, previous studies have examined the application of GNPs in polymers. However, no published reports are available in the open-access literatures which evaluate the use of GNPs in WPCs. The main objective of this work was to study the effects of GNP as a reinforcing agent on the physico-mechanical properties of WPCs.

2. Materials and methods

2.1. Materials

Poplar (*Populus deltoides*) flour (PF) was collected from a local sawmill in Karaj, Iran. The PF was initially washed with 1% NaOH solution, followed by washing with cold water. The treated PF was milled down to particle size of 60 mesh and then dried at $105 \pm 5^\circ\text{C}$ till the attainment of constant weight. The sieved wood flour was kept in a container for subsequent use.

Polypropylene (PP), with trade name V30S, obtained from Arak Petrochemical Co. (Iran), was used as the polymer matrix. The coupling agent used in the preparation of composites was Maleic anhydride grafted polypropylene (MAPP), which was supplied by Arkema (France). Both PP and MAPP were in the form of pellets. Table 1 shows the typical properties of PP and MAPP.

The GNP powder (grade AO-4) was obtained from the Graphene Supermarket Co., USA. The specific surface area was $<15\text{ m}^2/\text{g}$, and the purity was 99.2%. The average thickness and lateral size of GNPs were about 12 nm and $4.5\text{ }\mu\text{m}$, respectively. Electronic microphotographs of GNPs (Fig. 2) were taken with scanning electronic microscope (MIRAI TESCANA).

2.2. Compounding of materials

Table 2 shows the blend design for compounding of materials. Totally 9 sets of blends and composite samples were fabricated. Compounding of PF, PP, MAPP, and GNPs was performed in a Brabender compounder (CW Brabender Instrument Inc.) with an internal mixer. The mixing was carried out at 185°C with a rotation speed of 40 rpm and a blending time of 5 min. For each batch, the total starting mass of materials was 45 g. The fiber volume fraction was kept constant at 20 wt.% for all the composites. Following compounding, all the blends were cooled to room temperature and ground into 10-mesh powder with a Thomas-Wiley mill.

Table 1
Physical and mechanical properties of used PP and MAPP.

Typical properties	Test method (ASTM)	Unit	Value	
			PP	MAPP
MFI @ 230°C	D1238	g/10 min	18	10
Density	D1505	g/cm^3	0.92	0.90
Vicat softening point, 10 N	D1525	$^\circ\text{C}$	154	147
Flexural modulus	D790	MPa	1550	880
Tensile strength	D638	MPa	33	22
Elongation at yield	D638	%	12	12

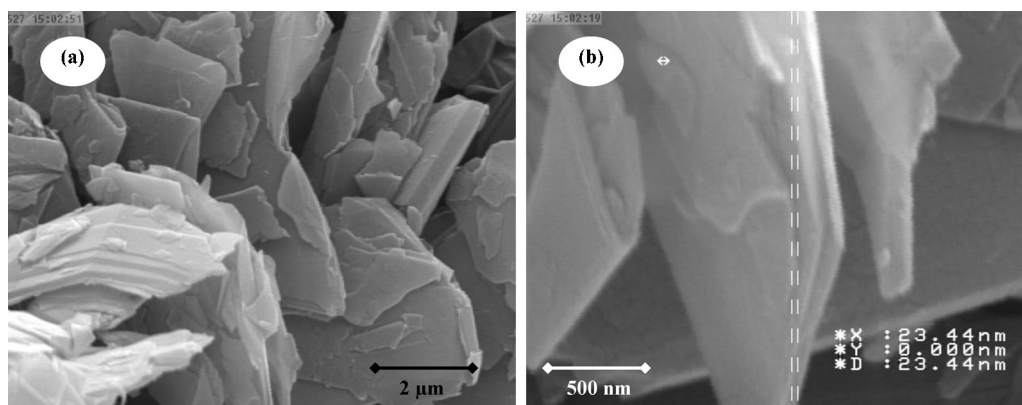


Fig. 2. SEM microphotographs of pristine GNP powder.

2.3. Preparation of composites

The WPCs were prepared by compression molding process using a square steel mold with dimensions of 152.4 mm × 152.4 mm. The blends were first added into the cold mold. The platens were preheated to 185 °C and a minimum pressure (close to zero) was applied during the preheating step to maintain the contact between the platens and mold. The pressure was then increased slowly to 2 MPa in 2 min and held at this pressure for an additional 10 min. The mold was then removed from the hot press and cooled to room temperature in a separate cold press under the same pressure (2 MPa). The target thickness of plaques was 1.5 mm.

2.4. Characterizations

2.4.1. Mechanical property

Mechanical properties of WPCs in terms of tensile and flexural tests were carried out using Universal Testing Machine (Instron, model 8112) according to ASTM D638 and D790, respectively. The specimens were tested at crosshead speed of 5 and 1.6 mm/min at room temperature for tensile and flexural, respectively. The Izod impact strength test according to ASTM D256 was carried out on notched samples using Santam pendulum impact tester (model SIT-20D) at room temperature. All the reported values for the tests were the average values of 4 specimens.

2.4.2. Dimensional stability tests

The thickness swelling (TS) and water absorption (WA) tests were conducted in accordance with ASTM D 570. Before testing, the weight, and dimensions, i.e. length, width and thickness of each specimen were measured. Conditioned samples of each composite type were soaked in distilled water at room temperature for 2 h. Samples were removed from the water, patted dry and then measured again. Each value obtained represented the average of 5 samples.

Table 2

Formulations of the used composites.

GNP (wt.%)	MAPP (wt.%)	PP (wt.%)	PF (wt.%)	Code
0.0	3	77.0	20	A0
0.2	3	76.8	20	A1
0.4	3	76.6	20	A2
0.6	3	76.4	20	A3
0.8	3	76.2	20	A4
1.0	3	76.0	20	A5
2.0	3	75.0	20	A6
4.0	3	73.0	20	A7
5.0	3	72.0	20	A8

2.4.3. Morphological study

Studies on the morphology of the composites were carried out using a scanning electron microscope (SEM). SEM micrographs of the surfaces of specimens were taken using SEM Model WEGA-II TESCAN. The specimen was coated with a thin film (25 nm) of gold to avoid electrical charge accumulation during the examination and then analyzed at an accelerating voltage of 25 kV.

2.4.4. Thermal property

Thermogravimetric analysis (TGA) was performed on the WPC samples to determine the weight loss as a function of temperature. Thermal properties were determined using a Polymer Laboratories Thermogravimetric Analyzer. The samples of approximately 4 mg of each composite were analyzed and heated at a rate of 10 °C/min up to 600 °C under nitrogen atmosphere.

3. Results and discussion

3.1. Tensile properties

Fig. 3a depicts the tensile strength and modulus of composites made with various GNPs contents. Samples filled with 0.8 wt.% GNPs showed the highest values among the other types of specimens. The tensile strength value was observed to be 26.6 MPa for composites made without GNPs, while maximum tensile strength was approximately 31.9 MPa for composites made with 0.8% GNPs. Tensile strength of pure PP decreased at least by 20% when PF was added. This is not surprising since it is known that when wood flour is used in thermoplastics, the tensile strength decreases [25]. Unlike tensile strength, tensile moduli of the samples increased by 41% (Fig. 3a). Sun et al. [26] reported that the tensile strength of composites is mainly influenced by filler fraction and the interfacial adhesion between particles and matrix. The possible reason propounded this kind of behavior may be the better interfacial adhesion between the matrix and PF. Better adhesion results in more restriction to deformation capacity of the matrix in the elastic zone and increased modulus. Similar observations were reported for other lignocellulosic fibers based PP composites [27].

As can be seen from Fig. 3a, both tensile properties were improved with the addition of GNPs. This result is consistent with the general observation that the introduction of nano-sized particles into a polymer matrix increases its tensile properties [28]. The enhancement is easily understood because filler in GNPs form can carry more tensile load. As mentioned earlier, the GNP is much stiffer than polymer matrix and as a result it adds stiffness to the composites. Similar results have been reported by Kordkheili et al. [2], who studied the properties of polymer/wood flour/CNT nanocomposites. Their data showed that the tensile modulus of

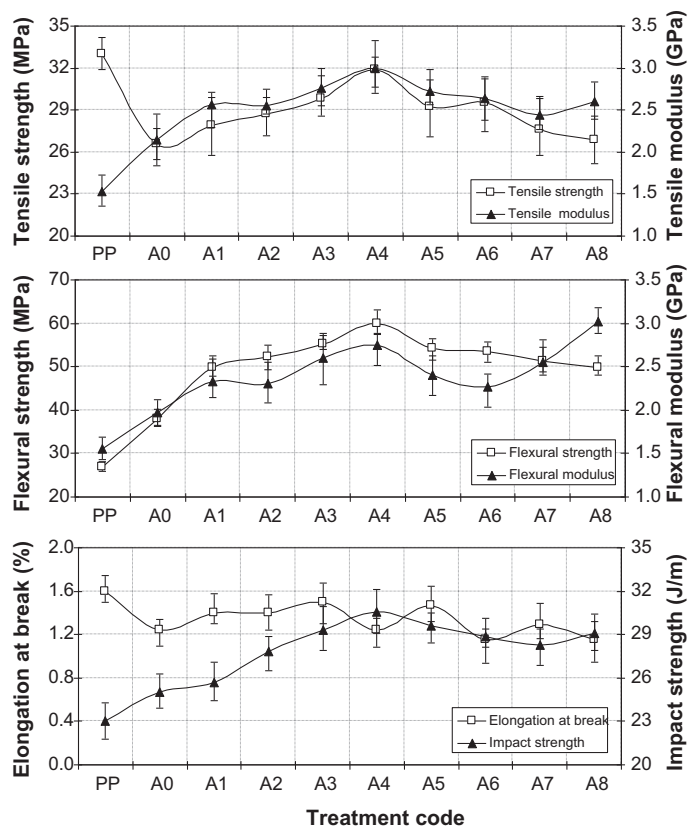


Fig. 3. Comparison of (a) tensile, (b) flexural and (c) impact properties of the composites as function of GNPs loading.

a polymeric material is remarkably improved when nanocomposites are formed with CNTs. However, Fig. 3a clearly illustrates that further addition (more than 0.8 wt.%) of GNPs cannot considerably improve tensile properties. This could be explained by the agglomeration of nanoparticles. The importance of dispersions and their effect on mechanical properties of composites made with nanoparticles has been discussed in many research works [29–31]. To achieve an effective reinforcement by adding carbon nanotubes, CNTs should be dispersed uniformly into the resin.

3.2. Flexural properties

The flexural properties of the composites vary significantly with GNPs contents (Fig. 3b). Similar with the trend results of tensile properties, composites made with 0.8 wt.% GNPs showed the maximum strength and modulus of flexural, whereas neat PP and composites without GNPs exhibited the lowest properties. The flexural modulus in composites is mainly a function of the modulus of individual component. Flexural modulus of GNPs is considerably higher than PF and PP, respectively. Increased flexural properties for 0.8 wt.% GNPs loading are attributable to the high stiffness of GNP with high aspect ratio. In addition, the increase in flexural properties was expected due to the improved adhesion between components in the composites.

As it can clearly be seen from Fig. 3b, with increase in GNPs contents from 0.8 to 5 wt.%, the flexural properties are considerably decreased. Sheshmani and Amani [17] reported that the modulus of composites at higher nanoparticles loading might not be decreased because of the GNPs agglomerate. One of the most important parameters in fabricating composites is GNPs dispersion in the matrix. Nanoparticles aggregation is harmful to physical and mechanical properties of the resultant nanocomposites. Use of

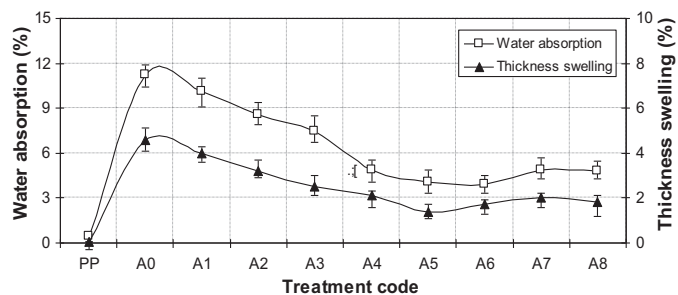


Fig. 4. Water absorption and thickness swelling of the composites as function of GNPs loading.

a graphene oxide is the best method to prevent the GNPs from aggregating together, and improves good dispersion of GNPs in nanocomposites [17].

3.3. Izod impact strength

Impact strength shows the strength of material against breakage and the start of cracking in the weakest point of the composite, which is the connecting point between lignocellulosic material and polymer matrix. Fig. 3c presents the gradual increase of notched impact strength with the incorporation of GNPs from 0.2 to 5 wt.%. It can be easily seen that the samples filled with GNPs had significant higher impact strengths in comparison with the neat PP and the control samples (without GNPs). The highest increment in impact strength was observed with the addition of 0.8 wt.% of GNPs, where the impact strength increased by 33% compared with that of neat PP.

3.4. Elongation at break

The elongations at break values of the composites are plotted against cellulosic type and content in Fig. 3c. The two cellulosic materials have different effects on this property as well, i.e. they influence the deformation and failure of the composites slightly differently. Composites filled with wood flour show brittle behavior, with 40% lower elongation at break than hybrid composites. It is interesting to note that the deformability of composites containing the wood pulp and those prepared by hybrid material is similar, although interfacial adhesion must be different in the two cases. As observed from the graph, the hybrid composites exhibited a positive elongation at break effect with the addition of wood pulp fibers. The positive hybrid effect of the elongation at break in hybrid composites was also observed by Hariharan and Khalil [32], who concluded that in a hybrid composite, the addition of high elongation fibers with low elongation fibers often increased the elongation at break of the hybrid composite compared to the composite made from low elongation fibers.

3.5. Water uptake and dimensional stability

The results of water absorption and thickness swelling of composites with different percentage of GNPs loading are shown in Fig. 4. As it can be seen, neat PP does not absorb moisture due to its hydrophobic nature, indicating that moisture is absorbed by the hydrophilic woody component in the composite as well as voids and micro-gaps at the interface. The value of water uptake was suddenly increased after the addition of PF to the blend. The hydrophilic nature of wood flour caused an increase in the water uptake. As PF content was constant (20 wt.%) in all blends, the different water absorptions among all the manufactured composites can be attributed to the role of GNPs. The water uptake was decreased

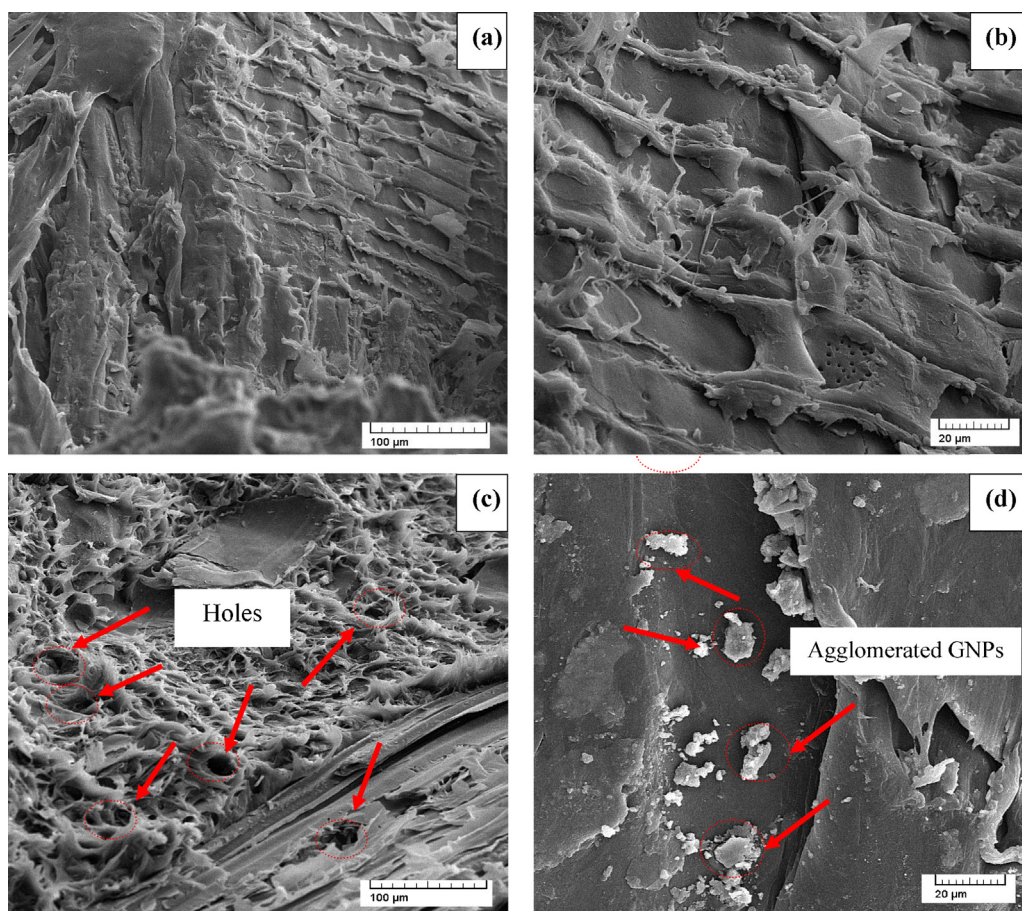


Fig. 5. SEM micrographs of the composites filled: (a) without GNPs (control), (b) with 2 wt.%, GNPs, (c) with 3 wt.%, and (d) with 5 wt.% GNPs.

after the addition of GNPs. WPC loaded with 1 wt.% showed lowest water uptake followed by WPC with 0.8 wt.%. The graphene layers provided tortuous path and increased the barrier property for water transport [16]. According to Das et al. [33], initially, water saturates the cell wall (via porous tubular and lumens) of the fiber, and next, water occupies void spaces. Since composite voids and the lumens of PF were filled with GNPs, the penetration of water by the capillary action into the deeper parts of composite was prevented. Another reason for less water absorption could be the hydrophobic and water repelling nature of GNPs surface that tends to immobilize some of the moisture, which inhibits the water permeation in the polymer matrix. Both the mentioned mechanisms may suggest that the water absorption has occurred in the surface layer.

Fig. 4 shows thickness swelling of the composites after 2 h immersion in distilled water. It can be seen that the composite without GNPs exhibited the highest thickness swelling values among other samples. Like water uptake results, the composites containing GNPs showed less thickness swelling percentage as compared with those made without it. The trend and explanation of thickness swelling of different samples were similar to those of samples taken for water uptake study.

3.6. Morphological characteristics

The significant improvements in mechanical properties of the composites with incorporation of GNPs were further supported by SEM micrographs. Representative SEM micrographs of the tensile fracture surfaces in the two different formulations are reported in Fig. 5. At 0.8 wt.% GNPs content (Fig. 5a and b), filler particles become the main component and few traces are seen where the

filler particles have been pulled-out. This is a clear indication that GNP has an effective interaction with PP matrix. As can be seen, there is no separation of the fibers from the matrix and a very good interaction between the components can be inferred from the image. The strong adhesion that is observed at the interface has been already discussed in mechanical properties of the composites and is related to the coupling agent, which encapsulated fibers in the matrix and caused strong bonding.

As it can be seen from Fig. 5c, the composites filled with 5 wt.% GNPs had many holes and cavities remained after the fillers were pulled out of the matrix. The presence of these holes means that the interfacial bonding between the filler and the matrix polymer is weak and therefore the GNPs could not provide an efficient stress transfer from the matrix. As shown in Fig. 5d, high content of GNPs were easily agglomerated, which is the characteristic of this nanofiller. The presence of these agglomerates results in the generation of flaws, resulting in the creation of voids between the filler and the matrix polymer. This causes the mechanical properties of the composites to be reduced, as compared with the composites filled with lower content of GNPs.

3.7. Thermal properties

The thermal stability of the composites is a very important parameter for the processing and usage of these materials. The manufacture of such composites requires the mixing of fibers and matrix at high temperatures, so the degradation of the cellulosic materials can produce undesirable effects on the properties. The weight losses of various samples at different temperatures are illustrated in Fig. 6. In all the cases, a decrease in weight loss below

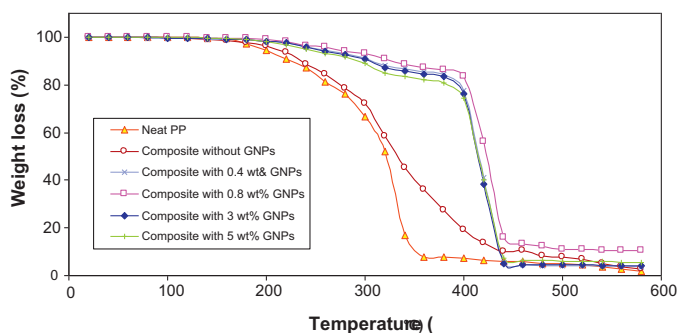


Fig. 6. TGA thermogram of neat PP and composites made with different GNPs contents.

100 °C was observed which was due to the removal of moisture. The TGA of neat PP showed a single-mass loss step with maximum degradation rate centered at 360 °C. For the control composite (sample A0), it was verified that the maximum degradation rate was shifted to a higher temperature (around 400 °C) showing that the presence of the wood flour improved the thermal stability of the polymer compared with the neat PP. This could be explained by the high thermal stability of lignin in chemical composition of wood flour. In an interesting research, Yildiz and Gümüşkaya [34] reported that lignin is thermally the most stable components of wood. It is widely accepted that the primary thermal decomposition of cellulosic materials occurs between 200 and 400 °C [35].

Another important feature observed was the higher degradation temperature of the polymer matrix in the composites containing the GNPs. The temperature of degradation of the polymer matrix increased from 330 to 430 °C in comparison to the control composite. This indicates that the thermal stability of the composites increased after using GNPs. The maximum improvement in thermal stability was observed by the inclusion of 0.8 wt.% GNPs. This might be attributed to the presence of graphene flakes which acted as a barrier and delayed the decomposition of volatile products [17]. The lowered thermal stability exhibited by WPCs filled with high content of GNPs was due to the poor dispersion and agglomeration of the graphene.

4. Conclusions

This study investigated the effect of GNPs on the physical and mechanical properties of WPCs. Some conclusions are as follows:

1. Although incorporating GNPs into the PP matrix effectively improves mechanical properties, this improvement comes at proper nanofiller loading (0.2–1.0 wt. %).
2. Addition of GNPs decreased the average water uptake and thickness swelling by 35% and 30%, respectively, compared to control (without GNPs).
3. The optimal loading of GNPs, at which the least water uptake was observed, was 0.8–1 wt.%.
4. SEM micrographs showed that high contents (3–5 wt.%) of GNPs were easily agglomerated. This caused the mechanical properties of the composites to be reduced.
5. The thermal degradation behavior of the composites was characterized with TGA. In all cases, the degradation temperatures shifted to higher values after the addition of GNPs. The highest improvement on the thermal stability of composites was achieved when 0.8 wt.% of GNP was added.

6. Finally, before achieving high performance of GNPs, the followings must be considered: (a) homogeneous dispersion or full exfoliation of graphene sheets in polymer matrix, and (b) strong interfacial interaction between the nanosheets and the surrounding polymer host, which is responsible for the effective transfer of external load.

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