ONLINE ISSN 2069-7430
ISSN-L 1841-4737
PRO LIGNO
www.proligno.ro
Vol. 14 N° 4 2018
pp. 131-139

SELECTED PROPERTIES OF CHASTE TREE AND SUNFLOWER HUSK FILLED THERMOPLASTIC COMPOSITE

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Abstract:
In this study, mechanical and physical properties of high density polyethylene (HDPE) based composite filled with two different lignocellulose fibers were investigated. Polymer composite were prepared by using HDPE as a polymer matrix and sunflower seed husk (Helianthus annuus L.) or chaste tree (Vitex negondu L.) as lignocellulosic filler at 15% and 30% loading. The effect of filler types and concentration on the mechanical properties of high density polyethylene (HDPE) based composites were investigated. Pellets were first manufactured through single screw extruder. Then, manufactured pellets were turned into composite samples by using injection molding machine. Tensile, flexural and impact properties were determined according to ASTM D638, ASTM D790 and ASTM D256, respectively. Addition of chaste tree fiber to thermoplastic composite provided better mechanical and physical properties compare to sunflower husk. All manufactured groups provided necessary values required by the ASTM D 6622 standards.

Keywords: *HDPE; Injection molded; sunflower husk; chaste tree; mechanical properties.*

INTRODUCTION

Composite can be defined as a material that combines two or more different components together to form an individual product with better properties (Mengeloglu and Karakus 2008b). Manufactured new composite materials may provide better performance than one component can provide by itself (Vuorinen 2007). Generally composites consist of two phases; matrix and reinforcement. The matrix is the continuous phase of the composite. Composites materials may be called with different names depending on the matrix preferences. When matrix is polymer, the resulting composite is called polymer-composites. Polymers can be thermoset or thermoplastic in nature. Many of the well-known composite materials are produced using thermoset polymers (plywood, particleboard, medium density fiberboard etc.). Thermoplastic based composites are also gaining new application areas over the years.

Polymer industry utilizes several additives including fillers in their manufacturing. The primary reasons for using additives are property modification or enhancement, overall cost reduction, and improving and controlling of processing characteristics.

WPC can be defined as composite that combines lignocellulosic flour/fiber (such as wood and non-wood) and plastic matrix (Panthapulakkal et al. 2006). Reinforcing of filling material can be agricultural waste or various wood flour/fiber while Plastic matrix can be polypropylene (PP), polyethylene (PE), polystyrene and polyvinyl chloride (PVC) (Wolcott 2001). There is a need for additives also (Haider et al. 2012).

There is an interest on finding new renewable filler for plastic industry. Billions of tones of agricultural crop waste (grass, cotton, corn stalk, bagasse and cereal straw, etc.) are generated all over the world and they are a good candidate for it. Agricultural waste represents cheap, easily existent and huge amount of lignocellulosic materials. Only small amount of this wastes are used as household fuel or animal feed and rest of them is burned in the field causing environmental pollutions (Sain and Panthapulakkal 2006). Utilizing agricultural residues to make thermoplastic composites can improve the some properties of the composites and may decrease material costs. Different kinds of agricultural wastes can be utilized for thermoplastic composites (Wu et al. 2003; Nyambo et al. 2010). Several agricultural wastes were used as filler in composite materials. Some of them are wheat straw (Mengeloglu and Karakus 2008b), rice husk (Chaudhary et al. 2004), cotton (Kim et al. 2008), sunflower husk (Cosereanu et al. 2014), bamboo (Manickavasagam et al. 2015) etc.

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Sunflower (Helianthus annuus L.) is an annual agricultural crop and has versatile usage including seed oil production. The sunflower seed consist of a hull and a kernel. Globally the sunflower business is substantial and growing continuously (Gertjejanson et al. 1972). The two sorts of sunflowers become commercially in the United States are the birdseed and confectionery varieties and, the oil-bearing varieties (Robinson et al. 1967). Sunflower hull or husk is interesting raw material for composite manufacturing (Gonzalez-Perez et al. 2002; Schroeder et al. 1996). This waste material was generated during sunflower oil production as a by-product. This by-product was mostly burned as a fuel to generate energy for the same oil production plant. The fiber of this plant and its by-product waste is digestible and has fiber length similar to the wood fiber. This presents an opportunity to be utilized as a raw material for the composite industry (Barcas and Anonima 1996; U.S point document).

Chaste tree (Vitex nigondu L.) is in Verbenaceae family. It is a small tree or large shrub with much branched stem. Plant grows to three - to nine feet-tall. This plant usually grows in Mediterranean countries and central Asia and is generally used in medical purposes (Mahalakshmi et al. 2010). The root or stem of chaste tree plant can be an interesting raw material in the manufacture of filled thermoplastic composite due to its lignocellulosic nature.

In this study, possible utilization of Sunflower husk and Chaste tree flours in high density polyethylene (HDPE) based composites were investigated. Mechanical, physical and morphological properties were evaluated.

MATERIAL AND METHODS

Materials

High-density polyethylene (HDPE) was used as thermoplastic matrix and chaste tree and sunflower husk fiber were used as fillers. Chaste tree was collected from the local place in Erbil, Iraq. Sunflower husk (SH) also obtained from a factory in Erbil, Iraq. Paraffin wax (K.130.1000) was used as a lubricant. Maleic anhydrite grafted polyethylene (Licocene PEMA 4351 by Clarient, MAPE) was utilized as coupling agents. Description of coupling agent was given in Table 1. The important chemical components of lignocellulosic materials are presented in Table 2.

Table 1

Descriptions of the coupling agents used in this study

Description	Licocene PEMA 4351 (MAPE)
Appearance	White fine grain
Softening point	43 mg KOH/g
Acid value	0.99 g/cm^3
Density at 23°C	300 mPa.s

Table 2

Chemical characteristics

Chemical Components	Chaste Tree	Sunflower Seed Husk
Hemicellulose (%)	30.17	24.62
Cellulose (%)	39.47	34.37
Lignin (%)	39.61	30.26
Ash (%)	4.71	15.48

Methods

Composite Manufacturing

Chaste tree (CT) wood, collected from the local place in Erbil-Iraq, was first cleaned from dust and dirt and later granulated the into flour form in Wiley Mill. Sunflowers were obtained from a factory in Erbil-Iraq, SHs were separated from seed and granulated into flour form using a Wiley mill. Both flours were screened and CT flours 0.25-0.42mm and for SH flours 0.18-0.42mm were used in this study. The classified fillers were dried in oven at 103°C (±2) until their moister content decreased to about 1% for 24h before manufacturing.

The experimental design of the study is presented in Table 3. Depending on the formulation, given amount of HDPE, SH, CT, MAPE and paraffin wax were dry-mixed in a high-intensity mixer to produce a homogeneous blend. These blends were compounded in a single-screw extruder at 40 rpm screw speed in the temperatures (barrel to die) of 170-175-180-185-190°C. Extruded samples were cooled in water pool and subsequently granulated into pellets. The pellets were dried in oven at 103°C (±2) for 24 hours. Dried pellets were injection molded into standard test samples using an HDX-88 Injection Molding Machine.

Table 3

Description of the manufactured samples

ID	HDPE (%)	Sunflower Husk Flour (SH) (%)	Chaste Tree Flour (CT) (%)	MAPE (%)	WAX (%)
HDPE	100	0	0	3.0	3.0
PE-SH-15	85	15	0	3.0	3.0
PE-SH-30	70	30	0	3.0	3.0
PE-CT-15	85	0	15	3.0	3.0
PE-CT-30	70	0	30	3.0	3.0

Mechanical Property Testing

Testing of the samples was conducted in a climate-controlled testing laboratory. Tensile, flexural and impact properties of all samples were determined according to ASTM D 638, ASTM D 790, and ASTM D 256, respectively. Tensile and flexural testing were performed on Zwick 10KN while a HIT5.5P by Zwick™ was used for impact testing on notched samples. The notches were added using a Polytest notching cutter by RayRan™. For mechanical property determination, five samples were tested.

Physical Property Testing

The thickness swelling (TS) and water absorption (WA) of the thermoplastic composites were carried out according to ASTM D 570. Before testing, specimens were conditioned and thickness and weight of samples were measured. Measured samples were entirely immersed for 1-day, 7-days, and 28-days under distilled water at room temperature. At the end of the waiting period, samples were taken out of water and their surface water was removed with a clean dry cloth. The specimens were weighed to the nearest 0.01g and measured to the nearest 0.001mm immediately. The resulting weights and thickness were used to calculate the WA and TS values. Five replicates were tested for each group.

Scanning electron microscope (SEM) study

Fractured surfaces of the samples were studied using a ZEISS scanning electron microscope (SEM, Model EVO LS10 5500LV) at 20 kV accelerating voltage. First, samples were dipped into liquid nitrogen and then broken in half to prepare the fractured surfaces. Finally, samples were mounted on the sample stub and were sputtered with gold to provide electrical conductivity.

RESULTS AND DISCUSSION

In this study, two different lignocellulosic fillers (chaste tree flour (CT) and sunflower husk (SH)) and two different concentrations of filler (15% and 30%) were used. HDPE based CT or SH flour filled composites were produced in the density range of 0.95-1.04 g/cm^3. Mean density values are presented in Table 4. Both lignocellulosic flour filled composites provided slightly higher density values compared to neat polymer. This increase was believed to be due to the higher cell wall density of lignocellulosic materials (Mengeloglu and Karakus, 2008b). The composites produced with CT flour had slightly higher density values than SH.

Table 4

Density of the manufactured composites

ID	Density (g/cm^3)
HDPE	0.96 (0.007)*
PE-SH-15	0.99 (0.007)
PE-SH-30	1.02 (0.006)
PE-CT-15	1.00 (0.004)
PE-CT-30	1.04 (0.008)

* Values in parenthesis are standard deviations.

Mechanical Properties

Mechanical properties (Flexural, tensile and impact strengths) of the CT and SH filled polymer composites were determined and their results were summarized in Table 5. The arithmetic mean and standard deviation values were given for each group in the table. ANOVA (analysis of variance) were performed and Interaction graphs (Fig. 1-3) were prepared.

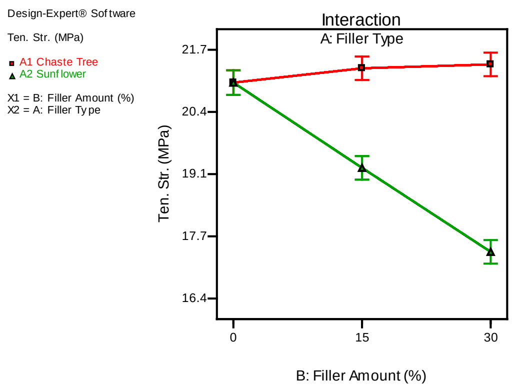
Table 5

Mechanical properties of polymer composites

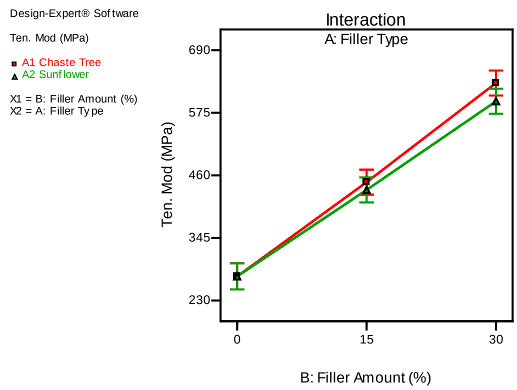
ID	Tensile strength (MP)	Tensile modulus (MPa)	Elongation at break (%)	Flexural strength (MP)	Flexural modulus (MPa)	Impact strength (kj/m^2)
HDPE	20.97 (0.34)*	277.9 (27.6)	454,6 (0,41)	21,8 (0,34)	621,1 (19,98)	3,87 (0,35)
PE-SH-15	19.18 (0.15)	433.2 (47.5)	10.24 (1.33)	24.79 (0.59)	816.3 (64.56)	3.55 (0.24)
PE-SH-30	17.39 (0.59)	596.07 (16.66)	5.13 (0.42)	30.86 (0.40)	1269.8 (22.7)	3.29 (0.31)
PE-CT-15	21.30 (0.17)	447.42 (19.61)	8.55 (0.37)	28.02 (0.23)	931.2 (16.06)	2.99 (0.17)
PE-CT-30	21.38 (0.37)	629.63 (46.01)	5.17 (0.26)	33.91 (0.99)	1287.5 (50.57)	2.84 (0.31)

* Values in parenthesis are standard deviations.

Interaction graphs for tensile properties of the manufactured composites were given in Fig. 1. Statistical analysis showed that both filler type and filler amount had a significant effect on tensile strength, tensile modulus and elongation at break values. Even though 3% MAPE was used in the formulation, tensile strength values were significantly reduced with the rise of SH flour concentration. This reduction might be due to the lack of compatibility between nonpolar polymer matrix and polar lignocellulosic fillers (Mengeloglu and Karakus 2008b). However, the slight increase has been observed on tensile strength with the rise of CT flour loading. That increase may be due to the morphology of CT structure. CT has thin and long fiber and this might improve stress transfer by minimizing space between fiber and polymer matrix. It was also reported usage of coupling agents (MAPE) has good effect on tensile strength (Chukwujike et al. 2015).



(a)



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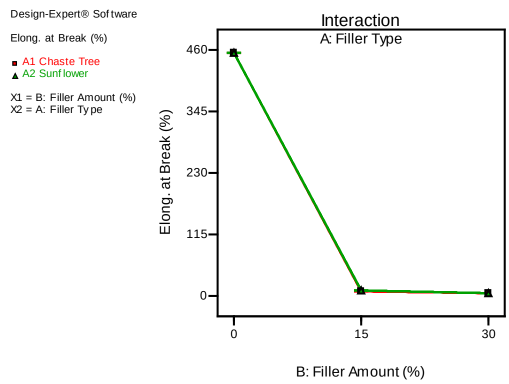


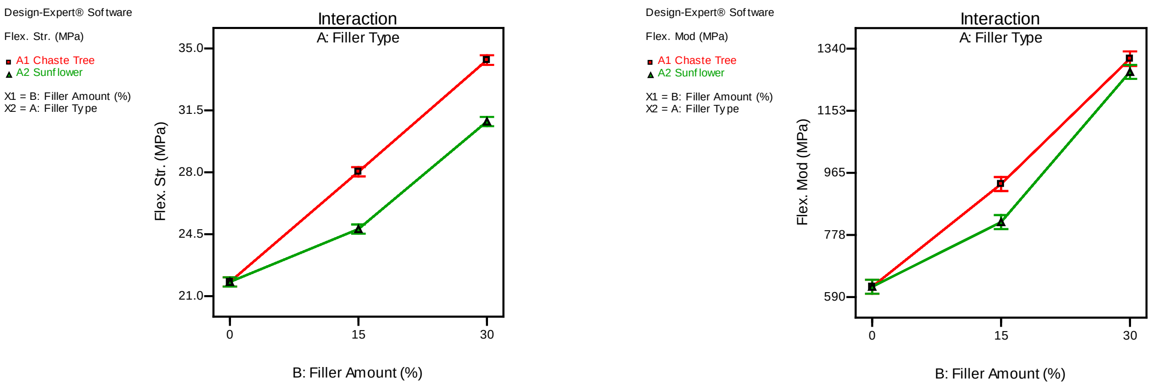
(c)

Fig. 1.
Interaction graphs of tensile properties a) tensile strength, b) tensile modulus, and c) elongation at break.

Interaction graphs of tensile modulus and elongation at break are shown in Fig. 1b and Fig. 1c, respectively. Tensile modulus values were significantly increased with the rise of filler concentration for both composites. Similar results were also reported on other studies on wood flours filled polymer composites (Wang et al. 2003; Qiu et al. 2004; Mengeloglu et al. 2007). The filler has significant effect on elongation at break values for both composites. Significant reduction by addition of filler in elongation at break values was determined for both composites.

Flexural properties include flexural strength and flexural modulus. The results showed that the flexural strengths are significantly affected by filler amount. Interaction graph of flexural strength is shown in Fig. 2a. The results of the statistical analysis show that the rate of lignocellulosic filler was effective on flexural strength. Compared to control samples, both SH and CT flour filled composites provided higher flexural strength values. Once again, CT having longer and fiber like morphology provided better flexural strength than composite produced with SH. Similar results were also reported by Mengeloglu and Karakus (2008a).

Interaction graph of flexural modulus is shown in Fig. 2b. With the rise of fillers loading increased the flexural modulus for both composites. This increase was statistically significant (P<0.0001). Addition of the lignocellulosic filler improves tensile modulus of the thermoplastic composites. This could simply be explained by the rule of mixtures (Matuana et al. 1998). Since lignocellulosic fillers have higher modulus of elasticity values than polymer, resulting composites are expected to have higher modulus values than polymer. Therefore, flexural modulus values were increased with the rise of lignocellulosic filler amount.



a b

Fig. 2.
Interaction graphs of flexural properties: a - Flexural strength; b - Flexural modulus.

In this study, produced composite materials were usually considered as an alternative to the polyolefin-based plastic lumber decking boards. For polyolefin-based plastic lumber decking boards, ASTM D 6662 (2001) standard requires the minimum flexural strength of 6.9MPa. All composites produced in this study provided flexural strength values between 24.79 and 33.91MPa that are well over the requirement by the standard. ASTM D 6662 (2001) standard requires the minimum flexural modulus of 340MPa for polyolefin-based plastic lumber decking boards. All composites produced in this study provided flexural modulus values (816-1287MPa) well over the required standards.

Interaction graph of impact strength is shown in Fig. 3. The results show that pure HDPE has higher impact. Impact strength reduced when lignocellulosic fillers were added to polymer matrix. Filler is significantly effective on impact strength. This usually arises from increasing of brittleness of the composite material (Mengeloglu and Karakus 2008b).

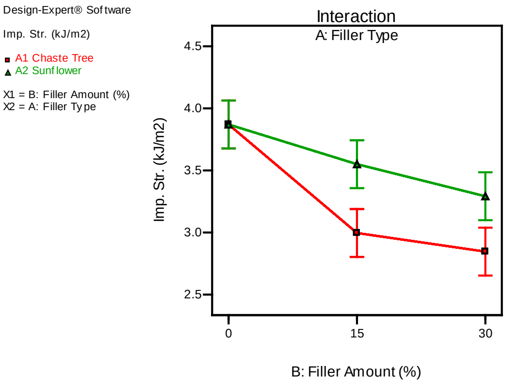


Fig. 3.
Interaction graphs of impact properties.

Morphology of the produced samples was also studied. SEM images of neat HDPE samples (sample ID: HDPE) was presented in Fig. 4. SEM images of 100 mesh size SH flour and CT flour was shown in Fig. 5a and Fig. 5b, respectively. Furthermore, SEM images of composites with highest SH flour and CT flour rate (sample ID: PE-SH-30, PE-CT-30) was shown in Fig. 6a and Fig. 6b, respectively.

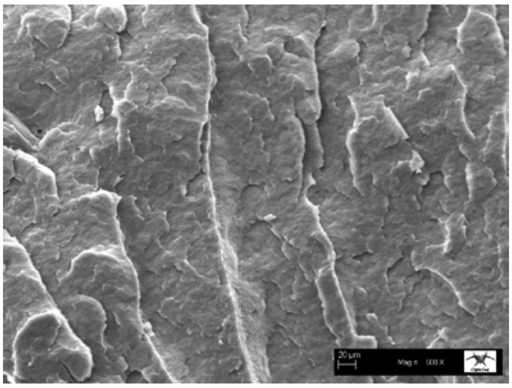
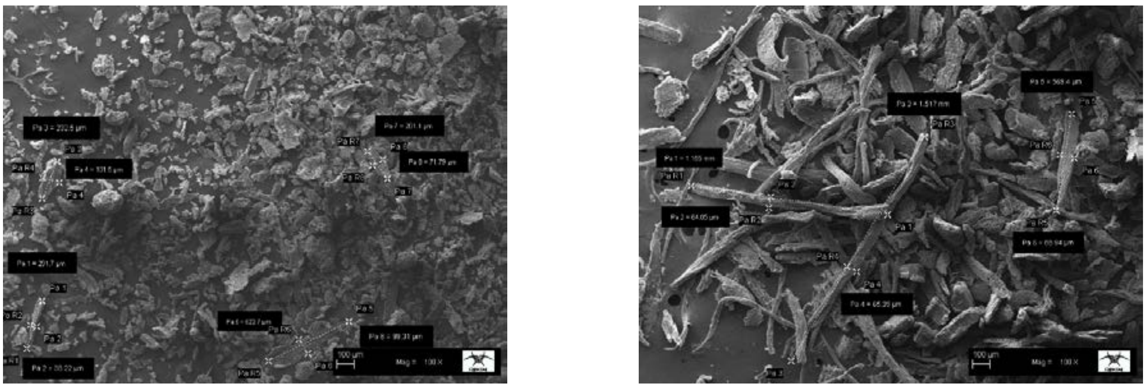


Fig. 4.
SEM images of neat-HDPE samples.



a b

Fig. 5.
SEM images of 100 mesh lignocellulosic flour: a - SH flour; b - CT flour.

ONLINE ISSN 2069-7430
ISSN-L 1841-4737
PRO LIGNO
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Vol. 14 N° 4 2018
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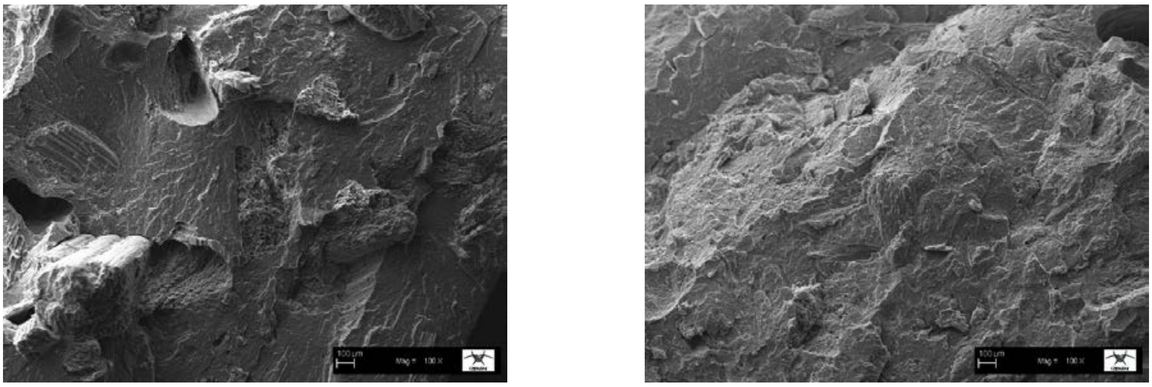

a b

Fig. 6.
SEM images of composites with highest lignocellulosic flour rate: a - Composite with SH flour; b - Composite with CT flour.

From SEM images, it is clear that polymer matrix and lignocellulosic flours were successfully mixed. In both composites, there are individual flours pulled out of the matrix indicating the lack of adhesion between the hydrophilic lignocellulosic filler and hydrophobic polymer matrix.

Physical Properties

Thickness swelling (TS) and water absorption (WA) values of all samples were determined using five samples with a dimension of 2cm×2cm for each group. The graphs of TS and WA properties are given in Fig. 7-8, respectively. Fig. 7 and Fig. 8 shows that samples' dimension are still changing even after 21 days of testing. The rate and extent of water uptake varied with fiber type. The composite made with CT flour were slower to absorb water compare to SH filled composites.

The rates of TS and WS were higher in case of higher loaded flour in HDPE composite due to their higher possibility of water absorption by hydrophilic nature of fibers, which proportionally increases the rate of water absorption with higher fiber loading. However composite made with SH flour showed higher TS and WS compared to composite with CT flour. The samples having higher plastic content had less water absorption, as expected.

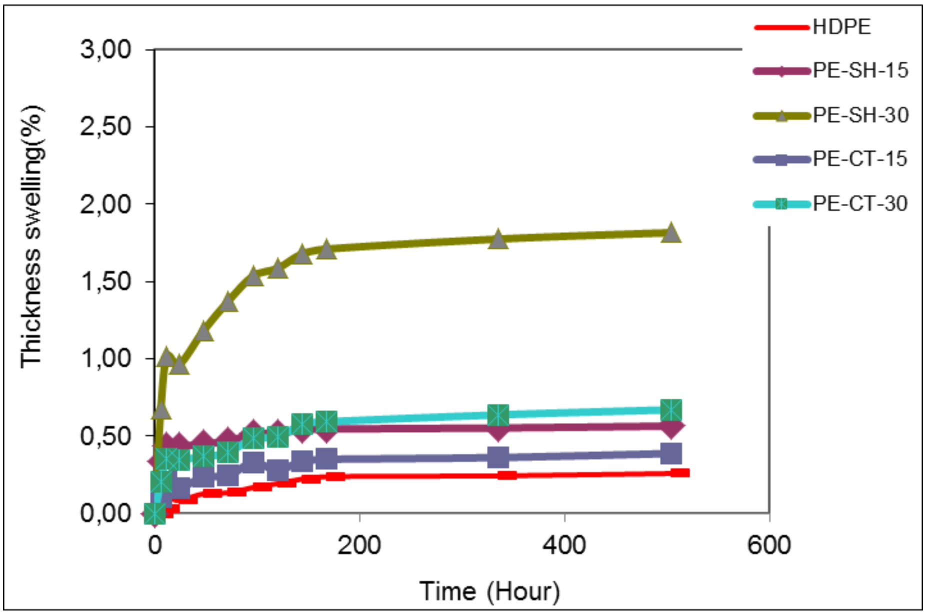

Fig. 7.
Thickness swelling properties.

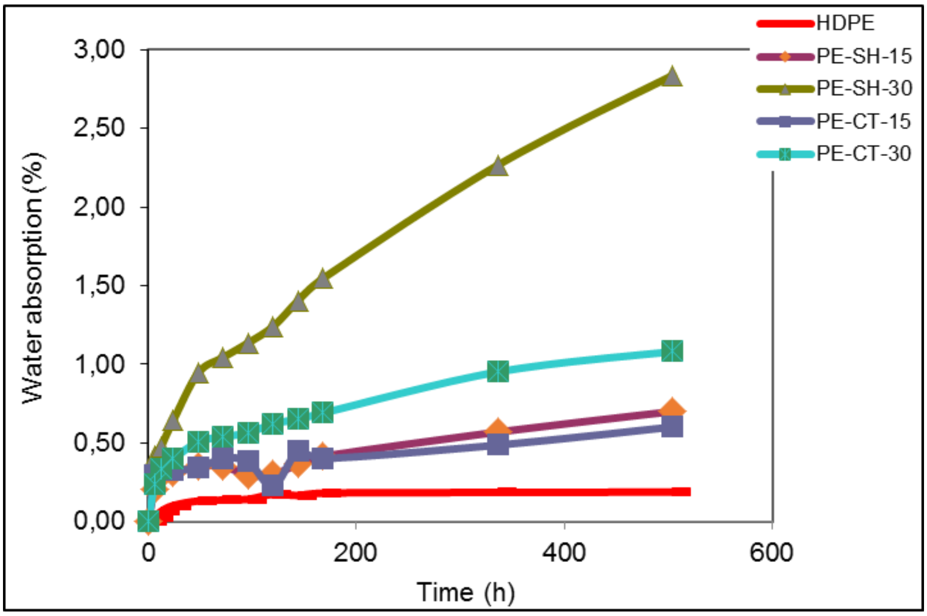

Fig. 8.
Water absorption properties.

CONCLUSION

Sunflower husk (SH) and chaste tree (CT) flour filled high-density polyethylene (PE) based polymer composites are manufactured through injection molding. The mechanical (tensile strength, tensile modulus, flexural strength, flexural modulus, elongation at break and impact strength) and physical (thickness swelling and water absorption) properties of the produced composites were determined.

The composites were successfully produced and the following conclusions were reached;

1. HDPE based composites with CT flour filled provided better mechanical and physical properties than composites produced with SH flour.
2. Addition of CT flour into polymeric matric improved strength, modulus values while reducing elongation and impact values.
3. Addition of SH flour into polymeric matric, by contrast with CT flour, reduced tensile strength. On the other hand, SH flour increased flexural strength, modulus values. Elongation at break and impact values also were got worse with loading of SH flour in the HDPE based composites.
4. Through SEM studies, lack of sufficient adhesion between filler and polymer matrices were determined.
5. HDPE and PP based polymer composites provide adequate mechanical properties according to ASTM D 6662 (2001).

As a result, CT and SH flour might be utilized as filler for HDPE based thermoplastic composites.

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