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Study of the effect of the introduction of plant fibers on the properties of composites

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Abstract

To reduce the massive consumption of HDPE, which causes environmental problems, we offer biodegradable composite materials, at low cost and with low density. In this context, a study devoted to the valorization of lignocellulosic waste, a particular interest has been brought to the pistachio shell filler, as vegetable filler in the manufacture of high-density polyethylene matrix composite materials with a filler rate of 5% to 15%. The HDPE/FCP composites were initially mixed in a calendar before preparing the different sample shapes with a thickness of 2 and 3mm by compression at 190°C. The latter were characterized by various techniques: rheological, mechanical, physical tests, and morphological.

Keywords *Pistachio shell fillers, High-density polyethylene, composite materials, mechanical properties, Physical properties.*

I. Introduction

For several years, the emergence of concepts such as sustainable development, industrial ecology, and green chemistry has been accompanied by the development of new generations of materials. Among these materials, composites continue to evolve into high-performance and cost-effective products while meeting environmental constraints and regulatory requirements regarding recycling. Polymer matrix-based composite materials reinforced with natural fillers have garnered increasing attention due to their resulting final properties, allowing broad access to various high-value-added application areas. This is in contrast to materials derived from petroleum resources, which have environmental constraints, not to mention the limited availability of raw materials that appear to be depleting and whose cost is very high [1].

The depletion of petroleum resources, along with the rise in environmental awareness that emerged in 1990, has made various populations realize the importance of their actions on the health of our planet. It has become necessary to reduce our CO2 emissions and waste quantities, improve recycling processes, and limit our dependence on fossil-based materials. As a result, many environmental standards have emerged in recent years to ensure the sustainability of our planet. These changes are not without impact on our daily lives and those of industries.

To better address these environmental concerns, the composite materials sector has turned to a new range of more environmentally friendly products: bio-composites. These materials have the advantage of incorporating all or part of bio-based elements and are therefore recyclable and biodegradable [2].

Incorporating natural fibers as fillers or reinforcements in plastics could also be a feasible solution to reduce cost, improve performance, reduce weight, and address environmental concerns [3].

Natural fiber-reinforced composites have, therefore, garnered increasing attention due to their low cost, low density, biodegradability, availability, ease of processing, high specific modulus, and ability to be recycled [4].

Like all natural fibers, pistachio shell fiber is hydrophilic because it is composed of lignocellulose, which contains hydroxyl groups. Therefore, it is incompatible with hydrophobic thermoplastics, which is a weak point for its use as a polymer reinforcement. It is necessary to create interactions at the interface between the polymer matrix and the fibers to avoid compromising mechanical properties. Among the numerous methods used to improve fiber-matrix compatibility, alkaline treatment [5, 6] and maleic anhydride grafting onto the polymer matrix [7, 8] are the most commonly used.

The objective of this study is the development and characterization of a new biocomposite material based on high-density polyethylene (HDPE) reinforced with waste pistachio shell fibers.

II. Material and methods

The high-density polyethylene of type PEHD 5502, with a melt flow index of 0.35g/10min is produced by the CP2K complex (Skikda). In addition, Pistachio shell flour is used as a filler in the composite preparation.

1.1 Blend preparation

The pre-mixing of HDPE/PSF composites with different pistachio shell fiber content (5%, 10%, and 15%) is carried out in a two-roll mixer from the IQAP LAP brand at the "CP2K, Skikda" unit. The rotation speed of the two rolls is set at 32 rpm, and the temperature is maintained at 170°C. After 10 minutes, composite sheets with a thickness of 3 mm are obtained, which will be used to prepare various samples through the compression molding process.

After the mixing process, we obtained a blend of HDPE and pistachio shell flour, which we cut into small pieces (2 to 3 cm) using a manual cutter. These pieces are then placed into molds to fabricate samples in the form of dumbbells and squares, which will later be used in various characterization tests.

The films obtained from the mixing process are cut and then placed between the plates of a CARVER hydraulic press. They are heated to a temperature of 190°C for 14 minutes to prepare 3 mm thick sheets, which will be used to cut samples for various characterization tests.

Table 1: Formulations of HDPE/ PSF.

Percentage of pistachio shell fibers (%).	0	5	10	15
Parentage of HDPE (%).	100	95	90	85

2 Characterization

2.1 Tensile test

The mechanical properties at the break of the samples are measured using a TesT GmbH tensile testing machine at room temperature. The samples are cut in a dumbbell shape of type "H" with dimensions (3 x 20 x 100) mm³, following the ASTM D638 standard. The strain rate is set at 20 mm/min.

2.2 Shore D hardness test

According to the ASTM D-2240 standard, 3 mm thick plate samples are prepared using a press and then placed under the penetrator. By operating the lever arm until the steel ball or diamond cone penetrates, the value indicated by the device can be read. Three tests are conducted at different points, and the average value is calculated.

2.3 IZOD impact test

The most common method is known by the reference IZOD (ISO 180). It is widely used in the United States (ASTM D-256), however in France, it is limited to the characterization of polystyrene (T51-911, identical to the ISO 180 standard and similar to the ASTM D256 standard).

2.4 Measurement of the fluidity index

The principle of this test is to measure the mass of molten thermoplastic material passing through a die over a certain period under the action of a defined load applied to the piston. The melt flow index, expressed in g/10min, provides information about the polymer's viscosity and, therefore, its molecular mass.

The melt flow index was determined using an extrusion plastometer (rheometer) containing a heated barrel at 190°C. A 3g sample of the material to be analyzed is introduced into the barrel, and it is pushed toward the die by a piston loaded with a weight of 2.16 kg, according to the standard: ISO 1133.

2.5 Water absorption test

The water absorption test involves drying the samples in an oven at 80°C for 24 hours. After drying, the samples are weighed using an analytical balance with a precision of 0.0001g. The samples are then immersed in distilled water at room temperature. The percentage increase in their weight is measured every 24 hours, representing their water absorption percentage during that period. Every 24 hours, the samples are removed from the water and quickly dried to remove excess surface water. Their weight is recorded within 10 to 15 seconds to minimize errors due to evaporation. The difference between the weight after each 24-hour immersion and the initial dried weight from the oven, relative to the initial dried weight, is used to determine the water absorption of the composite for that day. This value is recorded for 30 days to observe the trend in water absorption in the composites.

2.6 Determination of density

The apparent density is measured by the pycnometric method, according to the NFT 51-063 standard. Distilled water is used as the displacement solvent, ensuring the good wettability of the sample.

To study the morphology of the materials and verify the dispersion of fibers in the composite materials, optical microscopy is used to take surface photos of the obtained films. The equipment used is an optical microscope from OPTIKA Microscopes, ITALY.

III. Results and discussion

III.1 Mechanical Properties

1.1. Tensile test

- **Evolution of the Stress at Break**

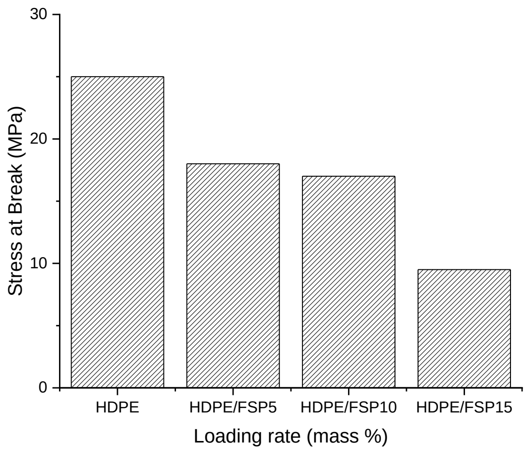

Figure 1: Evolution of the stress at break of HDPE/PSF composites as a function of the loading rate.

According to Figure 1, a decrease in the stress at break is observed with the increase in loading rate. This decrease becomes more significant as the amount of pistachio shell flour increases. These results are predictable and are consistent with many studies, such as those by S.M.B. Nachtigall et al. [9], Demir et al. [10], Khalid et al. [11], and Kaci et al. [12], who attributed this reduction to a weakening of the bond strength between the fiber and the matrix. This weakening obstructs the propagation of stress. This can be explained by the tendency of pistachio shell flour particles to cluster together, forming agglomerates that introduce heterogeneities and lead to uneven stress transfer within the matrix. As a result, the composite material becomes more brittle and fragile.

- **Evolution of elongation at break**

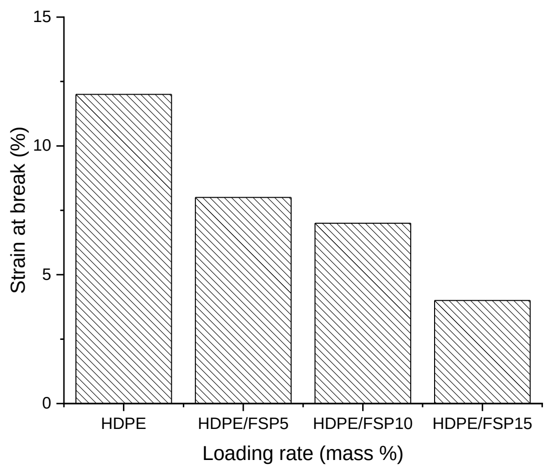

Figure 2: Variations in elongation at break of HDPE/PSF composites developed depending on the charging rate.

A significant decrease in the elongation at break is observed as the loading rate increases. Pasquini et al. [13] explained this decrease, from one perspective, by the hydrophilic nature of pistachio shell flour, which absorbs more moisture and causes swelling within the PEHD matrix, leading to material weakening. On the other hand, it is due to the increasing volume occupied by the filler particles, which creates defects in the system and reduces the inter-chain interactions. This results in a transition from ductile to brittle behavior in the material.

- **Evolution of Young's modulus**

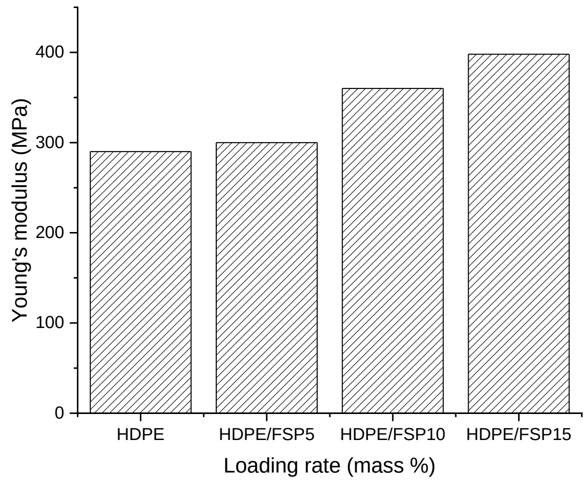

Figure 3: Variations in Young's modulus of HDPE/PSF composites as a function of rate dump.

According to Figure 3, a significant increase in the elastic modulus is observed with the increase in the loading rate. This mechanical property depends on the rigidity of the pistachio shell flour incorporated into the HDPE matrix [14].

1.2. Shore D hardness

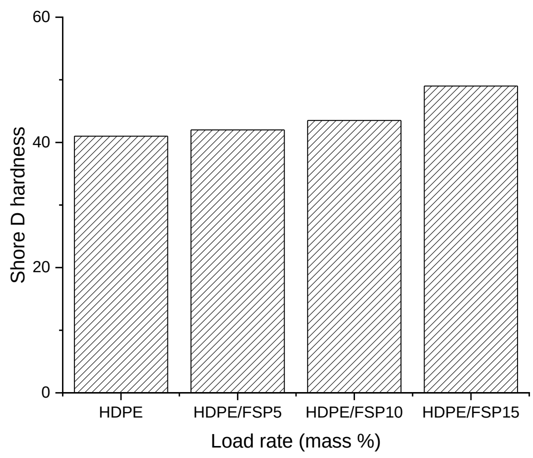

Figure 4: Evolution of the Shore D hardness of HDPE/PSF composites as a function of the charge rate.

We can observe that the incorporation of pistachio shell flour into the high-density polyethylene matrix (Figure 4) is accompanied by an increase in the hardness of the PEHD/PSF composites. This increase is more significant as the loading rate rises. These results are predictable since pistachio shell particles are composed of microfibrils of cellulose, which are classified as hard fibers. This results in greater resistance to penetration by the durometer needle into the composite material. This outcome was confirmed by N.Stark, and R.E. Rowlands [15].

1.3. IZOD impact resistance

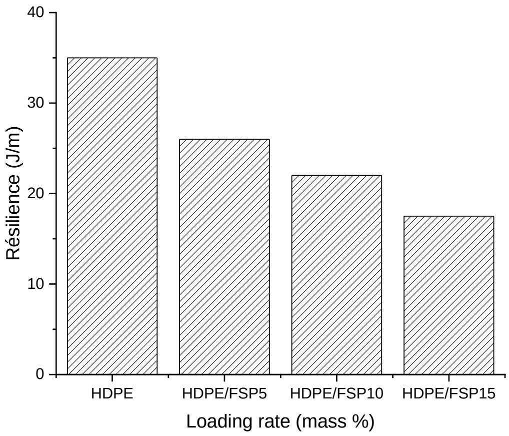

Figure 5: Evolution of the resilience of HDPE/PSF composites as a function of the rate of charge.

Figure 5 shows the variations in the impact resistance of PEHD/PSF composites as a function of the loading rate. According to this figure, the incorporation of pistachio shell flour into the PEHD matrix led to a decrease in the impact resistance of the composites. This can be explained by the incompatibility between the matrix and the pistachio shell particles reduces the adhesion between the two phases, which weakens the system, resulting in a decrease in the impact resistance of the composites. Additionally, the addition of FCP fibers, which are stiffer than PEHD, significantly increases the rigidity of the composite material, which also contributes to the reduction in impact resistance. These results are consistent with the ones presented by Petchwattana et al. [16] and Zheng et al. [17].

III.2 Rheological characterization

2.1Melt Flow Index: MFI

Figure 6 illustrates the evolution of the melt flow index of PEHD/PSF composites as a function of the loading rate. A decrease in the melt flow index is observed with the increase in pistachio flour content. This is due to the agglomeration of pistachio shell particles, which create obstacles to the free movement of the polymer chains and prevent the flow of the material. These results are consistent with M.N. ICHAZO et al [18] ones.

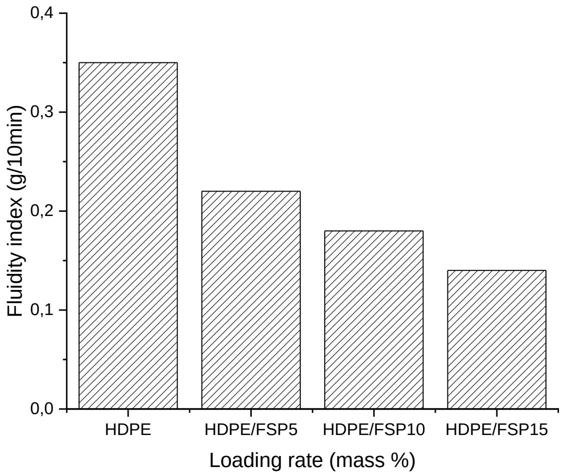

Figure 6. The fluidity index of HDPE/PSF composites evolution according to the rate dump.

Figure 6 illustrates the evolution of the melt flow index of PEHD/PSF composites as a function of the loading rate. A decrease in the melt flow index is observed with the increase in pistachio flour content. This is due to the agglomeration of pistachio shell particles, which create obstacles to the free movement of the polymer chains and prevent the flow of the material. These results are consistent with M.N. ICHAZO et al [18] ones.

III.3 Physical characterizations

3.1. Density

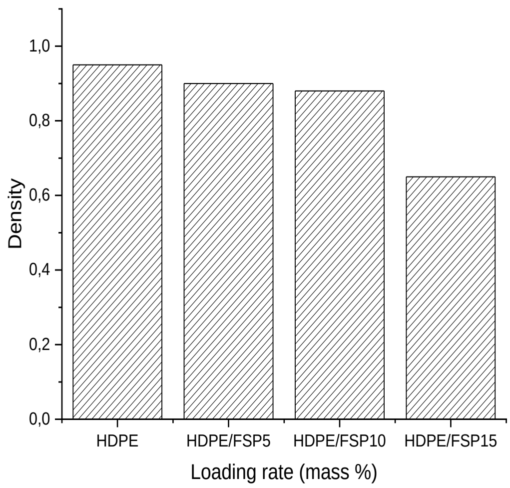

Figure 7: Evolution of the density of HDPE/PSF composites as a function of the charge rate.

The results of the density test for the composites PEHD, PEHD/PSF 5%, PEHD/PSF 10%, and PEHD/PSF 15% are presented in Figure 7. The measurement reveals that the density of pistachio shell particles is around 0.6254. In this regard, the incorporation of pistachio shell fibers into polyethylene results in composites with low density. The density profiles of the PEHD/PSF composites decrease as the loading rate increases, ranging from 0.95 for pure PEHD to 0.89, 0.85, and 0.63 for PEHD/PSF composites with 5%, 10%, and 15% pistachio flour, respectively [19].

III.4 Water absorption test

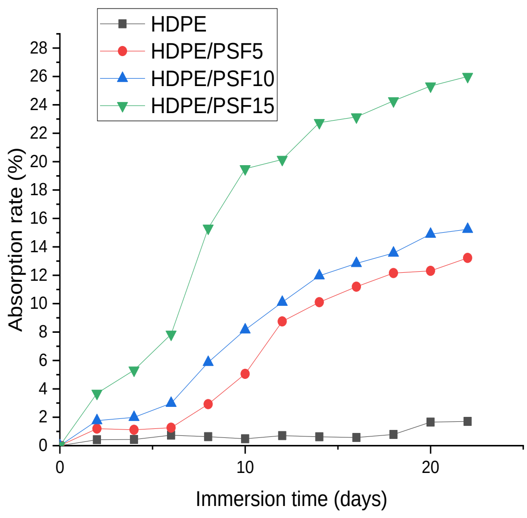

Figure 8: Evolution of the water absorption rate of HDPE/PSF composites depending on the immersion time.

We can observe an increase in the water absorption rate with immersion time and the pistachio shell flour content in the high-density polyethylene matrix. This is entirely expected since pistachio shell flour is highly rich in hydroxyl groups, which form hydrogen bonds with water molecules. Consequently, the higher the flour content, the greater the OH concentration, and thus, the water absorption rate increase. It is also noticeable that the water absorption rate of PEHD/FCP samples is initially rapid, and then slows down as time progresses until saturation is reached, where the water absorption rate becomes constant. For composites with 5%, 10%, and 15% pistachio shell flour, the maximum swelling rates are estimated at 13.7443%, 14.9659%, and 24.2842%, respectively. For virgin PEHD, a very low water absorption rate is recorded due to the non-polar nature of this polymer, which gives it hydrophobic characteristics. Water absorption is approximately 0.082% within 24 hours and does not exceed 0.033% after 15 days. These values are confirmed by S. Boufi et al. [20] and D. PASQUINI et al. [21].

III.5 Analysis of morphology by optical microscopy

The micrographs of the composites (Figure 10) related to the incorporation of 5%, 10%, and 15% by mass of pistachio shell particles into the HDPE matrix clearly show a heterogeneous and irregular surface, as well as the presence of pistachio shell particle aggregates completely separated from the high-density polyethylene matrix. These aggregates increase in number with the rise in the percentage of pistachio shell powder incorporated into the HDPE matrix. This is due to the incompatibility of the two phases resulting from the weak interfacial adhesion between the hydrophilic pistachio shell particles and the hydrophobic HDPE. These results are in good agreement with those reported by El-Shekeil et al [22], Al Maadeed et al [23], and Panaitescu et al [24].

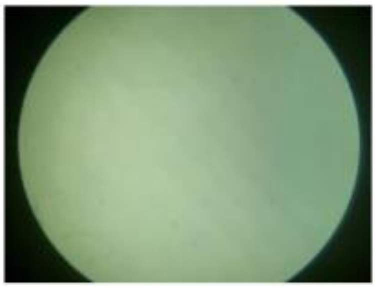

Figure 9: Optical microscopy micrograph of the Virgin HDPE surface.

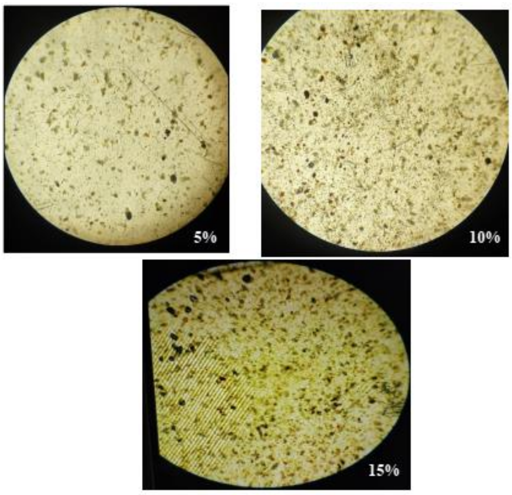

Figure 10: Optical microscopy micrograph of the surfaces of HDPE/PSF composites at different charging rates (5%, 10%, and 15% respectively).

IV Conclusion

This study aimed to develop and study filler/matrix composite materials, produced by conventional transformation processes (kneading followed by molding by compression). These composites consist of a high-density polyethylene matrix and
Pistachio shell charge with dimensions less than 90 µm and rates varying 5%, 10%, and 15% by weight. The composites obtained are then characterized by determining the rheological properties, physical, mechanical, and morphological.
The analysis of the experimental results allowed us to draw the following conclusions:

- Rheological characterization by measuring the fluidity index made it possible to deduce that:

- The HDPE/FCP composites fluidity index is decreased as a function of loading rate.

- The physical characterization by measuring the water absorption rate made it possible to deduce that :

- The increase in water absorption rate depends on the immersion time and the concentration of pistachio shell fillers.

- The mechanical characterization of HDPE/FCP composites made it possible to deduce that there are:

- An increase in the Shore D hardness of composites compared to the value of HDPE virgin.
- A decrease in the resilience of HDPE/PSF composites with the increase in rate dump.

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