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Valorization of lignocellulosic fibers in the development of composite materials

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Abstract

Natural fibers have recently attracted the attention of scientists because of their low-cost, low-density, renewable, biodegradable and non-abrasive properties. The growing use of composite materials is giving rise to problems of waste management. As a result, there is a growing trend towards the use of biodegradable products, either by using biodegradable polymers, or by incorporating biodegradable fibers into polymeric materials.
In this context, various high-density polyethylene-based formulations have been prepared (HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran).
The various composites were initially blended in a calender before the various samples, with average thicknesses of 2 and 3 mm, were prepared by compression at 190°C and characterized by mechanical test.

Keywords: *Wood flour, Wheat bran, Composite materials, High-density polyethylene.*

I. Introduction

In today's world, the growing demand for more advanced materials has compelled humans to combine two or three different components to create new products with improved properties. The combination of components with complementary characteristics results in attractive and essential properties for specific applications. Materials obtained in this manner are called "composites," and they currently represent a privileged area of scientific research [1,2].
One of the most significant advantages of polymers is their ease of processing, productivity, and cost reduction. For many applications, the properties of polymers are modified by using reinforcements and fillers [3].
The use of natural resources in composite materials is becoming increasingly common. Studies are dedicated to the valorization of lignocellulosic materials derived from wood and natural fibers as fillers in plastic matrices [4].
As a result, considerable attention in the research community is focused on the use of natural fibers as reinforcements in the design of composite materials, allowing for a combination of materials that are strong, lightweight, non-abrasive, structured, and cost-effective. However, despite these advantages, natural fibers present a major drawback when combined with plastics, especially polyolefins (such as polypropylene, polyethylene, polystyrene). Wood-based materials have a strong affinity for water (strongly hydrophilic character), which creates an interface incompatibility between lignocellulosic materials and highly hydrophobic thermoplastics. This interfacial incompatibility affects the synergy between the different constituents of the composite, knowing that the interface is the preferred location for stress transfer between the reinforcement and the matrix. To address this issue, many research efforts have been conducted to reduce the surface tension between plant fibers and thermoplastics [5].
The incorporation of cellulosic materials into high-density polyethylene matrices affects a wide range of properties. However, it has been found that the quality of adhesion alone does not dominate the properties of the composites, and other factors, such as the fraction, nature, or size of the incorporated filler, also come into play [6].

II. Material and methods

II.1 Raw Materials

The polymer used is high-density polyethylene of the type HDPE 5502. It is produced by the CP2K complex in Skikda and is marketed in the form of a white powder.
The WF was extracted from Aleppo pine trees, growing in the Djelfa region in the south of Algeria. The average particle size was approximately 63 µm.
Wheat bran is obtained after wheat grinding. Subsequently, all the grains are removed to retain only the bran. The wheat bran powder is processed to achieve a fine filler with a

uniform particle size, with a diameter less than 63 µm (<63µm).

II.2 Preparation

The different formulations chosen for the preparation of our samples are provided in the following table:

Table 1 The mass compositions of the various formulations.

Compositions	HDPE (%mass)	Wood flour (% mass)	Wheat bran (% mass)
HDPE	100	0	0
HDPE/WF	80	20	0
HDPE/WB	80	0	20
HDPE/WF/WB	80	10	10

The pre-blends of HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran are prepared in a double-roll mixer (Calender) of the brand IQAP LAP at the "CP2K, Skikda" facility. The rotational speed of the two cylinders is approximately 32 rpm, and their temperature is set at 170°C. After 8 minutes, composite films with a thickness of 1mm are obtained.

After the mixing process, we obtained a mixture of HDPE/Wood Flour, HDPE/Wheat Bran and HDPE/Wood Flour/Wheat Bran, which we cut into small pieces (2 to 3 cm) using a manual cutter. These pieces are then placed in molds to create samples in the form of dumbbells and squares for subsequent use in various characterization tests.

The films obtained through calendering are cut into small pieces and then placed between two insulating Teflon sheets, which are interleaved between two metal plates in a hydraulic press. They are heated to a temperature of 190°C for a total dwell time of 15 minutes, with 7 minutes allocated for preheating and 8 minutes for compression and degassing.

Upon exiting the press, the samples are cooled in the open air and carefully removed from the mold. Various shapes of samples with a thickness of 3mm are obtained, which will be subjected to various characterizations.

III. Characterization

The measurement of mechanical properties at the point of rupture of the samples is conducted using a TesTGmbH tensile testing machine at room temperature. The samples are cut into dumbbell-shaped specimens of "H" type with dimensions of (92 x 13 x 3) mm³, following the ASTM D638 standard. The deformation rate is set at 20 mm/min.

Shore D hardness tests samples with a thickness of 6 mm are prepared using a press, and they are then placed beneath the needle. By operating the lever arm until the needle penetrates, three tests are conducted at different points, and then the average is calculated [7].

From a 3.17 mm thick plate, Izod impact test samples with dimensions L = 100 mm and l = 12.7 mm are prepared using a cutter. A central V-notch, 2.5 mm deep, is created on these samples. Three tests are conducted and the average value is recorded [7].

Morphological Characterization by Optical Microscopy to study the material's morphology and verify the dispersion of fibers in composite materials, optical microscopy is used to capture surface images of the obtained films. The equipment used is an optical microscope from OPTIKA Microscopes ITALY.

IV. Results and discussion

IV.1 Impact strength IZOD

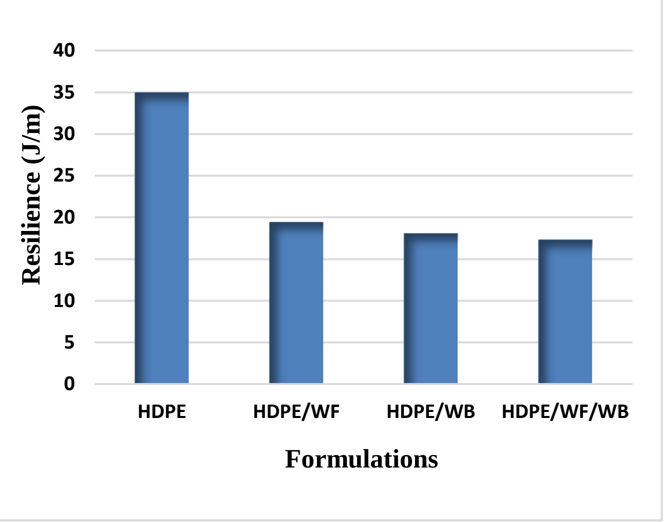

Figure 1. Evolution of the resilience of different composites developed with HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran.

According to figure 1, which illustrates the variations in the resilience of the HDPE/WF, HDPE/WB and HDPE/WF/WB composites, there is a clear decrease in impact resistance after the incorporation of Wood Flour and Wheat Bran into the HDPE matrix. This decrease is due to the increased rigidity and interfacial phenomena.

The HDPE/WF and HDPE/WB composites exhibit lower impact resistance values compared to pure HDPE. This indicates that wood flour and wheat bran have higher rigidity than high-density polyethylene, significantly increasing the composite material's rigidity, which, in turn, results in reduced impact resistance. This decline in impact resilience is also attributed to weak interactions (physical interactions) between the matrix and the filler, indicating poor interfacial adhesion. This has been previously confirmed by the work of Andrzej K. Bledzki et al and Ajay Karmarkar et al [8, 9].

IV.2 Shore D hardness test

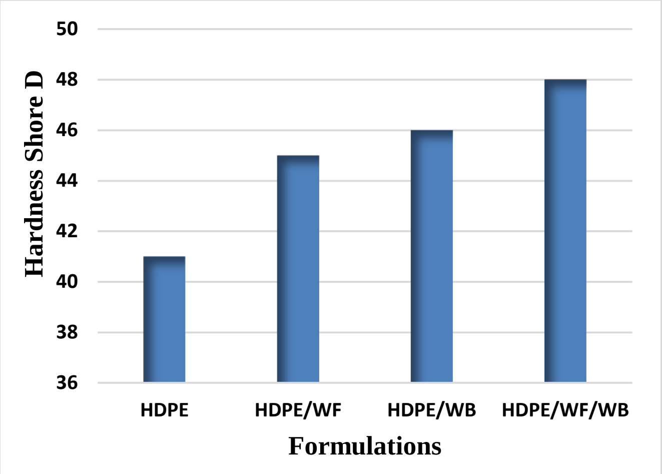

Figure 2. Evolution of the Shore D hardness of different composites developed with HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran.

According to figure 2, there is an increase in hardness with the incorporation of two types of fillers (Wood Flour and Wheat Bran) into the HDPE matrix. This increase is particularly significant for the hybrid composite (HDPE/Wood Flour/Wheat Bran).

These results are expected because the fillers consist of microcellulose fibrils, which are classified among hard fibers, making it more challenging for the durometer needle to penetrate the composite material. This outcome has been confirmed by Md. Rezaur Rabman et al and S. Th. Georgopoulos et al [10, 11].

IV.3 Tensile Test

IV.3.1 The evolution of tensile strength

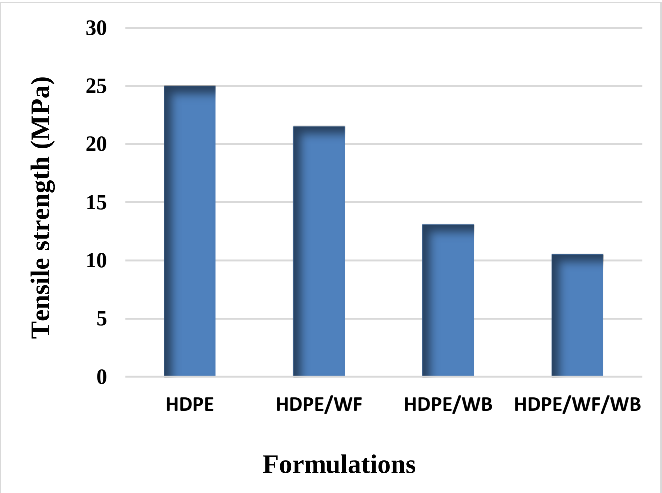

Figure 3. Evolution of the tensile strength of different formulations: HDPE, HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran.

A decrease in the breaking strength is observed for the various composites loaded with wood flour, wheat bran flour, and wood-wheat bran flour compared to the unloaded HDPE. These results are predictable and are in accordance with the findings of NM. Stark and R. E. Rowlands [12], B. Sanschagrin [13] and J. Simonsen [14]. They attributed this decrease to the reduction in the bond strength between the fillers and the matrix, which obstructs the transfer of stress and leads to a tendency to cluster together, forming agglomerates that induce heterogeneities and non-uniform stress transfer within the matrix, consequently resulting in a weakening of the composite material. In addition, there is a low interfacial adhesion between HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran due to the antagonistic nature (HDPE matrix being hydrophobic, while WF and WB fillers are hydrophilic). These results are consistent with those presented by K. Oksman and C. Clemons [15].

IV.3.2 Evolution of the Young's Modulus

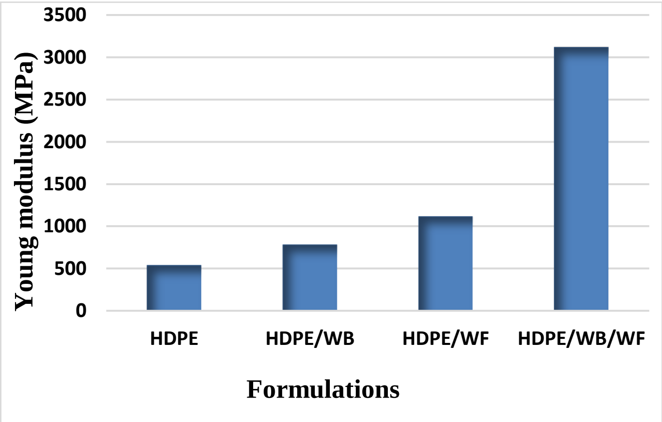

Figure 4. Evolution of the Young's Modulus of different formulations: HDPE, HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran.

The introduction of wood flour, wheat bran flour, and wood-wheat bran flour into the HDPE matrix increases the material's rigidity and reduces its elasticity. In other words, the Young's Modulus increases, and this increase is even more significant for the hybrid composite HDPE/Wood Flour/Wheat Bran. This can be explained by the fact that the rigid particles of wood flour and wheat bran flour tend to form a reinforcement within the composites, leading to greater strength. This is what NM. Stark and R. E. Rowlands [12] observed.

IV.3.3 Evolution of the elongation at break

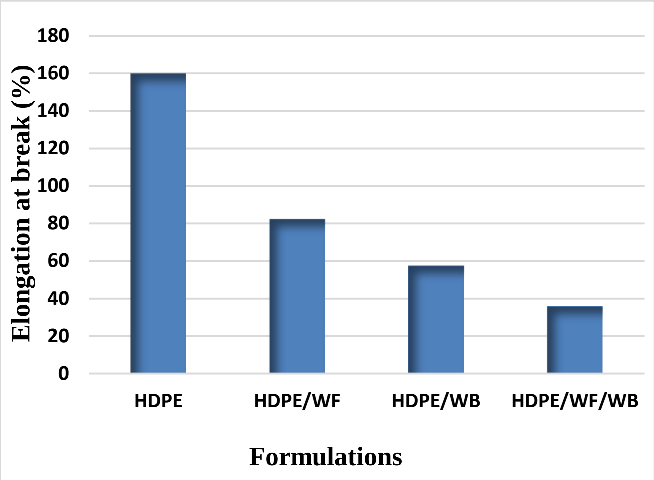

Figure 5. Evolution of the elongation at break of different formulations: HDPE, HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran.

There is a decrease in the elongation at break for the HDPE/Wood Flour and HDPE/Wheat Bran composites when compared to that of virgin HDPE, and this decrease is more significant for the hybrid composite HDPE/Wood Flour/Wheat Bran. This observation is in perfect agreement with many authors, such as B. A. Acha et al [16], T. T. L. Doan et al [17], S. J. Kim et al [18]. The deterioration of this property is primarily due to the incorporation of rigid wood flour and wheat bran flour into the HDPE matrix, which reduces the mobility of the polymer chains, leading to the premature failure of test specimens at low stresses.

IV.4. Morphology Analysis by Optical Microscopy

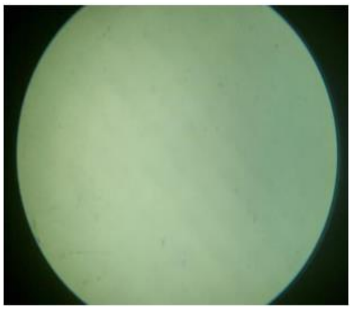

Figure 6. Optical microscopy micrograph of the surface of virgin HDPE.

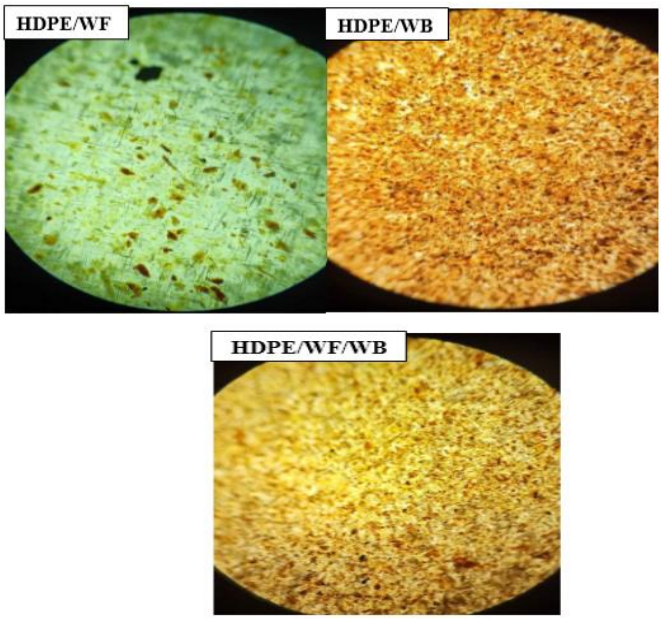

Figure 7. Optical microscopy micrograph of the surfaces of HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran composites.

The micrographs of the composites in figure 5 concerning the incorporation of wood flour, wheat bran flour and wood flour-wheat bran flour into the HDPE matrix, clearly depict a heterogeneous and irregular surface with the presence of aggregates that are completely separated from the high-density polyethylene matrix.
These aggregates increase in number in the hybrid composite HDPE/Wood Flour/Wheat Bran. This is due to the incompatibility of the two phases because of the low interfacial adhesion between wood flour and wheat bran flour, which have a hydrophilic nature and HDPE, which is hydrophobic.

V. Conclusions

The analysis of the experimental results has allowed us to draw the following main conclusions:

- The mechanical characterization of the developed composites has allowed for the deduction that:
  - ✓ He hardness of the developed composites HDPE/Wood Flour/Wheat Bran, is higher than that of virgin HDPE.
  - ✓ The resilience of the different developed composites HDPE/Wood Flour/Wheat Bran has decreased compared to that of virgin HDPE.
  - ✓ The mechanical behavior of the HDPE/Wood Flour/wheat Bran composites shows that both stress and elongation at the rupture decrease while the elasticity modulus increases progressively.
- The morphological characterization through optical microscopy of the HDPE matrix shows a homogeneous and regular surface, unlike the HDPE/Wood Flour, HDPE/Wheat Bran, and HDPE/Wood Flour/Wheat Bran composites, which clearly exhibit a heterogeneous and irregular surface with the presence of aggregates.

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