



Article

# Investigation of Mechanical Properties of Coffee Husk-HDPE-ABS Polymer Composite Using Injection-Molding Method

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**Abstract:** Waste biomass-based natural fibers are being extensively researched nowadays as a composite material with various waste-based high-density polyethylene (HDPE) to utilize the waste biomass and recycle the plastic waste in an effective approach. In this study, chemically modified spent coffee husk (CH) has been applied with different ratios of HDPE to produce composite material and characterized comprehensively to determine the mechanical stability of the products. The injection molding method was used for composite development containing HDPE with untreated and 10 wt% NaOH-treated CH weight ratios of 0%, 15%, 20%, and 25% together with 10 wt% coupling agent and filler materials of acrylonitrile butadiene styrene (ABS) and kaolin clay, respectively. Physicochemical characteristics of untreated CH, 10 wt% NaOH treated CH, pristine HDPE and HDPE-CH composites have been analyzed comprehensively in this study. Adding 25 wt% fiber with 65 wt% HDPE and 10 wt% of ABS (7 wt%)-kaolin clay (3 wt%) increased the tensile and bending properties significantly. This composite presented the maximum tensile, flexural, and impact strengths, which were 36 MPa, 7.5 MPa, and 2.8 KJ/m<sup>2</sup>, respectively. The tensile strength and bending strength of NaOH-treated coffee husk fibers (CHF) were enhanced by 32% and 29%, respectively. The microstructural characteristics of HDPE with treated and untreated CHF composites analyzed by scanning electron microscopy (SEM) demonstrated the fibers' and matrix's excellent adhesion and compatibility. Thus, HDPE polymer-treated CH composite presented excellent stability, which can be expected as a new addition for construction, food packaging, and other industrial applications.

**Keywords:** composite development; coffee husk; HDPE utilization; mechanical properties; injection molding



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## 1. Introduction

Natural fibers have been extensively researched and used for various composite development (as polymeric composite reinforcement) in various industrial applications, from food packaging to construction applications [1]. In this study, residual fibers obtained from the coffee husk (CH), an agribusiness waste typically burned or disposed of in the environment, have been emphasized. In many earlier research studies, natural hydrophilic fibers with hydrophobic polymers exhibited adhesion problems. Therefore, this adhesion issue needs to be improved before applying composite development. This study presented an alkaline treatment (by sodium hydroxide (NaOH)) of spent coffee husk to improve the texture of coffee husk fibers (CHF) and composite development. Further, the efficiency of the surface NaOH treatment applied to waste CHF has been studied to improve the mechanical performance of high-density polyethylene (HDPE) polymer composites. Social awareness

has grown considerably as an associated outcome of environmental problems such as waste creation, fuel scarcity, high material prices, and an increased need to reduce carbon [2]. As a result of these changes, the construction and automotive business has shifted to many ecologically friendly chemical compound materials and composites [3]. The environmental consciousness movement has fueled a search for alternative renewable resources. In several studies, vegetable fibers, also known as natural lignocellulosic fibers (NLF), were used as reinforcing fillers in polymeric composites [1,4–7]. Creating novel materials from replenished resources or even waste materials has attracted much interest [8,9]. Coffee is another significant and widely used source of agricultural waste [10]. The coffee husk has a thickness of 3–5 mm and a 0.20 m. It has been studied in different forms as a source of antioxidants and valuable chemicals and as a filler in composite formulations of green composites. Green composites can successfully demonstrate technological advantages over conventional petroleum-derived polymer composites, such as cost reduction, lower density, no toxicity, balanced mechanical properties, and, most importantly, lower environmental impacts [11]. HDPE is the most widely used polymer worldwide, and a large amount of HDPE comes from waste plastic materials. Recycling of waste-plastic-based HDPE and HDPE utilization has become a significant concern worldwide to the circular economy and effective waste management nowadays [12]. The advantage of recycling HDPE and HDPE utilization is that it has almost similar qualities to its petrochemical equivalent, which has strong mechanical resistance, high quality, and increased water resistance [12,13]. Within the final long time, squanders have been broadly utilized as fortifications in polymer composites [14,15].

HDPE forms excellent mechanical properties, high chemical resistance, and good processability and includes a broad run of applications, counting electrical fabric applications. Therefore, HDPE is considered a suitable material for composite development. It has a few impediments, such as its softening temperature, warm steadiness, and high combustion efficiency [16]. The mixing of polymers is regularly a vital course for obtaining unused and made strides polymeric materials, which are troublesome to obtain by coordinate polymerization handle [16,17]. Mixing is an appealing strategy for making unused materials with enhancement and adaptability in execution and significantly better properties than the existing polymers [17]. With the expanded toll of materials and natural contamination, creating elective and naturally inviting common fiber materials for thermoplastic composites is crucially required [18]. Characteristic filaments offer specialized and natural benefits when utilized to strengthen plastics [19]. Fillers are used alongside various commodities as planning polymers to form strides within the properties and decrease the fetch. The change of integrity filler materials into plastic moves forward different properties of the materials, such as mechanical properties and physical properties. The mechanical characteristics of chemical compound particle materials heavily depend on the living, shape, and conveyance of filler particles inside the chemical compound lattice and the enlargement grip between the filler and the framework [20,21]. Acrylonitrile butadiene styrene (ABS) is a high-impact modifier once used in CH powder. ABS is a common thermoplastic polymer. Its glass transition temperature is approximately 221 °F (105 °C). ABS is amorphous and therefore has no actual melting point. ABS has high rigidity, good impact resistance, good abrasion and resistance, and high dimensional stability [22].

The coupling agent, ABS, is employed to regulate the interface. This research aimed at evaluating the mechanical characteristics of spent CH with an HDPE-ABS infusion for target applications such as construction, food packaging, and automotive industries. Kaolin clay features high compressibility and also very low strength. A composite may be a heterogeneous fabric that comprises two or further stages that are insoluble in each other. Composites are outlined to autonomously have mechanical properties and exhibitions predominant to constituent materials [22]. Natural fortifications in thermoplastic lattices have been used for many years. The unique fiber composites are essential to provide natural advantages, such as reduced dependency upon vital sources and non-renewable resources [23–27]. In this study, alkali-treated (NaOH) and untreated CHF were ground

and then extruded with ABS, kaolin clay, and HDPE, and the resulting compounded pellets were injection molded into pieces. The mechanical properties of HDPE with various coffee husk powder contents were determined to analyze the influences of coffee husk-derived lignocellulosic filler on the final performance of the manufactured composite pieces and determine their potential for the desired application.

## 2. Materials and Methods

### 2.1. Materials

The coffee waste utilized in this experiment was gained from the Arabica low process at Ilubabor (Mattu) and Buno Bedele, both in southwest Ethiopia. Yangzi Petrochemical Co., Ltd. (Nanjing, China) is a subsidiary of Sinopec. High-density polyethylene (HDPE) (5000 s, soften flow index of 0.940 g/min, density 0.961 g/cm<sup>3</sup>, melting point 128 °C) was used to make the composite components. The acrylonitrile butadiene styrene (ABS) (Changzhou Kesi Success Plastics Materials Co., Ltd, Shanghai, China) was from China, and kaolin clay was sourced from Borana Shakiso, Oromia, Ethiopia.

### 2.2. Methods

#### Fiber Treatment and Preparation of the Composites

Composites of the coffee husk fibers (CHF) (80–120 mesh) were prepared using the following process (Table 1). The CHF was dried under vacuum at 103 °C for 24 h to reduce the water content. A mixer (SJ45, Nanjing Rubber-Plastic Machine Ltd., Nanjing, China) was used to mix and compound the ABS as a coupling agent, kaolin as filler, CHF as reinforcement material, and the HDPE as a matrix. HDPE, ABS, kaolin, and CHF were mixed in specific ratios (Table 1) in a high-speed mixer for 10 min. Each composition was then extruded using a twin-screw/single-screw extruder system to produce CHF/HDPE composite sheets. The melting, pumping, and die zone processing temperatures were set at 160, 170, and 180 °C, and the mold temperature was set at 60 °C. These were extruded through a round die to produce strands and pelletized using an air-knife unit. Resultant pellets were shaped in an injection molding machine, Meteor 270/75, from Mateu and Sole (Barcelona, Spain), at 190 °C using a mirror-finishing steel mold with standard geometries for sample characterization. A low injection time of 1 s was employed, whereas the applied clamping force was 20 tons, and the pieces were cooled for 10 s in the mold. As a result, 3 mm pieces were produced that, before characterization, were stored for 48 h. The rotation speeds of the twin-screw and single-screw extruders were 40 and 20 rpm, respectively. The composites were composed of four components: HDPE as the matrix, CH as reinforcement, Shakiso kaolin clay as the corresponding filler ad, and ABS as the coupling agent. Table 1 shows the formulation of the composites. This study created six distinct composites (3 treated and 3 untreated CH) with weight proportions of 25/65, 20/70, and 15/75 for treated and untreated CH samples, respectively.

**Table 1.** Formulations for the composites according to the weight percentage (%).

Sample	CHF (%)	Kaolin (%)	ABS (%)	HDPE (%)
HDPE/(CH)	25	3	7	65
HDPE/(CH)	20	3	7	70
HDPE/(CH)	15	3	7	75
HDPE		100	0	0

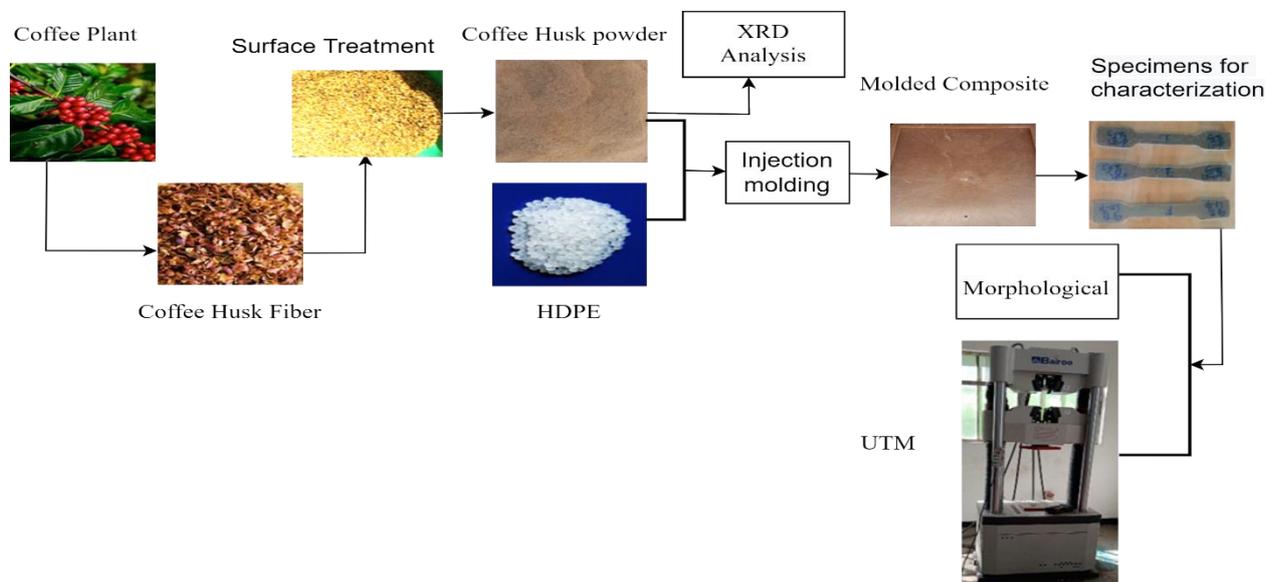
Kaolin was created to increase material interface compatibility and deliver an effective, high-quality product. The kaolin was utilized in the study obtained from Borana Shakiso to increase the abrasion resistance, physical properties, and flow properties. The CHF was washed correctly, and the composite was prepared using the injection molding method. The CH has a thickness of 3–5 mm and a length of 0.20 m. They were washed with clean

water up to a dirty of the husk was removed and oven-dried for 24 h. For 2 h, the fiber was immersed in a 10% NaOH solution. To remove impurities from the fiber's surface, the husks were washed frequently with normal water and then with clean water before drying at room temperature for 24 h. The CH fiber has been treated with 10% NaOH alkali. Lastly, the treated CH was grounded with a milling machine of 0.18 mm to obtain the powder.

The injection molding machine was used to create the samples with dimensions of 200 mm × 200 mm × 3 mm. Three composite samples were prepared: 25/65, 20/70, and 15/75 of treated and untreated CH. The composition of the constituents in percentage is 25/65, 20/70, and 15/75, describing 7 wt% kaolin, 3 wt% ABS, 15 wt% CHF, and 65 wt% HDPE. The kaolin and ABS are constant, while the fiber and HDPE percentages vary.

A removing agent was added to the surface to keep the fibers from adhering to the mold.

A release agent, also known as a mold release agent, is a chemical applied to the mold's surface to prevent materials from bonding to the mold during production. Rests of material on the mold surface have a negative impact on the finished product's quality. Release agents aid in the separation of the mold from the material used in mold release, plastic release, die-cast release, and other processes. Some mold release agents are sprayed directly on the mold's tool steel. Some of these include silicone and fluorocarbon-based materials. Other mold release agents are coated or sprayed onto the molding powder's surface. This is done in batch blenders just before packing out to avoid passing through an extruder. Most release agents that are melt blended with the polymer can be used in this manner. The composite was prepared and condensed for 24 h with a 30 kg weight. This was done to ensure that the sample decreased porosity. The samples were cured for 48–72 h to strengthen them. Following this procedure, test samples were prepared following ASTM standards. Figure 1 represents the processing sequence to produce untreated composite (UC) and treated composite (TC) composites with their characterization.



**Figure 1.** Representation of the processing sequence to produce UC and TC composites with their characterization.

### 2.3. Characterization of Mechanical Properties Characterization

Following the recommendations of ASTM standards, the planned mechanical tests were performed on the prepared test specimens, which included impact, flexural, and tensile tests [1,11,28–30]. The specimen size and shape for the corresponding tests are explained in the following sections. Three specimens were tested for each set of samples, with the mean points used for analysis.

### 2.3.1. Tensile Strength

Tensile testing was used to measure the breaking force when the stress was applied to the molten plastics molecules, which were always oriented in the direction of flow, with the interpretation that the mechanical properties of the composite can be achieved when the orientation of the molecules is in the flow direction [29,30]. According to ASTM D3039 standards [31,32], the specimen dimension,  $l \times w \times t$ , used was 250 mm  $\times$  25 mm  $\times$  3 mm. The test was performed on a 50 kN capacity machine (Bairoe, Shanghai, China Universal test machine) with a crosshead speed of 5 mm/min and a gauge length of 150 mm.

### 2.3.2. Flexural Strength

The material's resistance to deformation under a given load was investigated using flexural strength measurements. To investigate the interlaminar shear failure, a three-pointed test was performed using an ASTM D790 standard [32,33] on UTM (Wp310 universal material tester, Gut, Barshuttel, Germany) at test speeds ranging from 1 mm/min. The dimensions of the specimen were 127 mm  $\times$  13 mm  $\times$  3 mm. The following formula was used to calculate flexural strength:

$$\sigma = \frac{3F_{max}L}{2bh^2} \quad (1)$$

where  $\sigma$  represents the flexural strength,  $F_{max}$  is the maximum load at failure,  $b$  is the sample width,  $h$  is the sample thickness, and  $L$  is the sample length between the two support points.

### 2.3.3. Impact Load Test

The Charpy impact strength was determined via a Tinius Olsen (Horsham, PA, USA) model 104 impact tester with a pendulum of 246 g (1.24 J). The impact test is a one-point test that determines a material's resistance to impact load from a swaying pendulum on the specimen until it fractures. The specimen size for this test was 65 mm  $\times$  13 mm  $\times$  3 mm. All specimens were notched in the center of the longitudinal side by an automatic sample notcher, and the impact strength was calculated using a Charpy impact tester (Ceast Torino, Torino, Italy) following the ASTM D256 standard used [34].

### 2.3.4. Absorption of Water

Water absorption tests on treated and untreated coffee husk polymer composites were recommended at room temperature. The test was carried out following the ASTM D570 standard [35], with samples drawn from water at regular intervals and weighed immediately before being cleaned with a dry cloth. The samples were weighed using an electric balance to determine how much water had been absorbed. All the samples were dried in an oven until they reached a constant weight before being re-immersed in water. Equation (2) was used to calculate the percentage of water absorption.

$$WA = \frac{W_2 - W_1}{W_2} \quad (2)$$

where  $WA$  is the water absorption,  $W_1$  is the initial (dry) weight of the composite, and  $W_2$  is the final (wet) weight of the composite after immersion in water, respectively.

### 2.3.5. Morphological Studies of Composites by SEM

The SEM technique was used to study the structural morphology of the composite fracture surface. Before being examined in an SEM at 20 kV, the samples were thoroughly cleaned and coated with thick platinum in a JEOL sputter iron coater. The morphology of the fracture surface of the composite samples was examined. SEM images usually reveal the microstructure of composite surfaces with comprehensive morphological details [36].

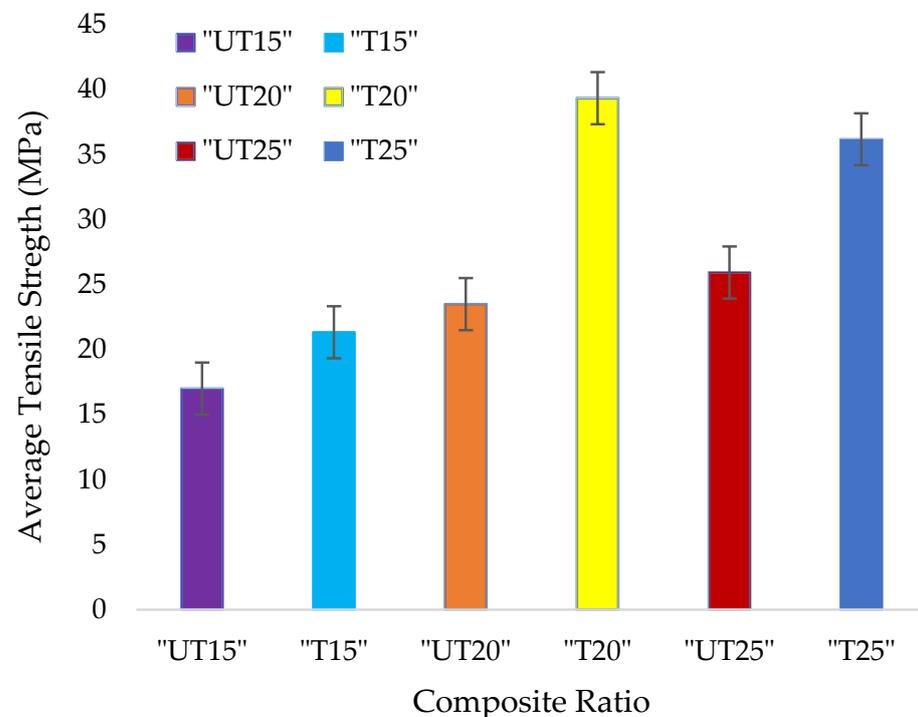
### 2.3.6. X-ray Diffraction (XRD) Analysis

The structures of CH were examined using the X-ray diffraction method. The Shimadzu Corporation (Kyoto, Japan) XRD-7000 diffractometer was used to demonstrate the crystallinity analysis with a tension of 40 kV and 40 mA, which was applied to the samples for the measurement of Cu anode materials at a temperature of 25 °C. The XRD patterns of the samples were measured in the continuous scanning mode with a scan speed of 5° /min and a scan range of 10 to 60°.

## 3. Results

### 3.1. Tensile Test

The following are the experimental results for the tensile strength of fabricated treated and untreated composite materials. As shown in Figure 2, treated coffee husk (CH) polymer composites have higher tensile strength (36 MPa) than untreated CH polymer composites. In Figure 2, UT15, UT20, UT25, T15, T20, and T25 represent 15% of untreated CH, 20% of untreated CH, 25% of untreated CH, 15% of treated CH, 20% of untreated CH and 25% of untreated CH in the composite mixtures, respectively.

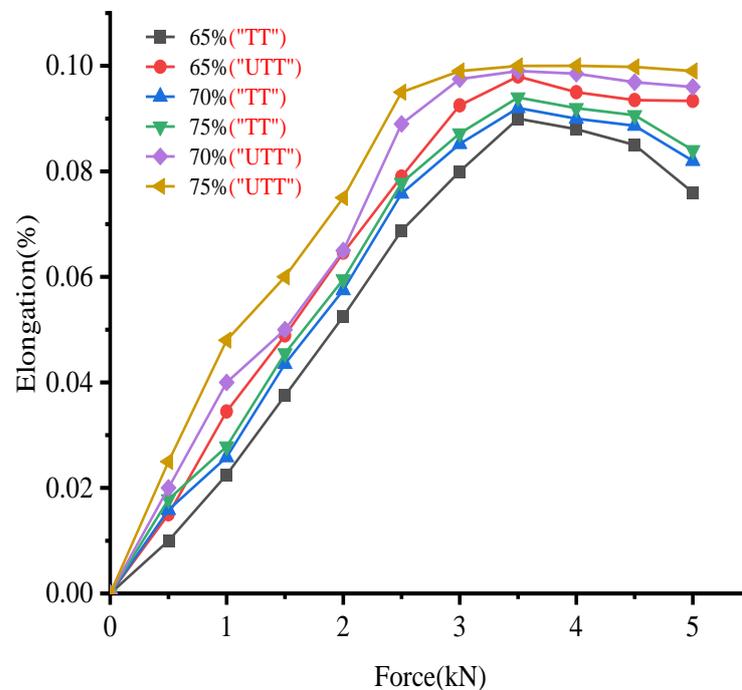


**Figure 2.** Tensile strength test results. UT15, UT20, UT25, T15, T20, and T25 represent 15% of untreated CH, 20% of untreated CH, 25% of untreated CH, 15% of treated CH, 20% of untreated CH and 25% of untreated CH in the composite mixtures, respectively.

The tensile strength of the untreated CH polymer composite is lower (43.3 MPa) than that of the treated composite. Compared to the treated coffee husk fiber (CHF) composite, the tensile strength of treated and untreated CH composites with volume ratios of 25/65, 20/70, and 15/75 improved by 46.7%, 43.3%, and 32.2%, respectively. The tensile strength increased as the percentage of CH in the composite increased. A higher lignin content weakens the interfacial bond between fiber and polymer components, resulting in a weak composite. As a result, most researchers used fiber treatment to reduce lignin concentration to strengthen the interfacial connection and improve composite mechanical properties [27]. As a result, most researchers proceed with different pre-treatments of the fibers to reduce lignin concentration, which consequently strengthens the interfacial connection and improves composite mechanical properties [27]. Reducing

the hydrophilicity of the fiber enhances the bonds of functional groups, which provides additional strength to the fiber matrix and increases the mechanical properties of the composites. Therefore, this study presented an enhancement of the tensile strength in the fibers as well as composites after reducing lignin content.

The tensile strength of treated CHF is higher than that of untreated composites with the same (25/65, 20/70, and 15/75) ratios. This is most likely due to matrix integrity loss and insufficient wetting between the fiber and matrix in composites. When treated fiber was added to the composite, the tensile strength increased. This is because CHF is more polymer compatible, and the tensile strength of composite material is determined mainly by the strength and modulus of its fibers. Figure 3 depicts the force with elongation graphs for coffee husk/high-density polyethylene (CH/HDPE) 25/65, 20/70, and 15/75. Here, 65% TT, 65% UTT, 70% UTT, 70% TT, 75% TT and 75% UTT represent 65% of treated CH, 65% of untreated CH, 70% of untreated CH, 70% of treated CH, 75% of untreated CH and 75% of treated CH in the composite mixtures, respectively. It is shown that all of the curves increase linearly at first and then increase exponentially up to the maximum force.



**Figure 3.** Tensile force-elongation diagram of treated and untreated CH composites. Here, 65% TT, 65% UTT, 70% UTT, 70% TT, 75% TT and 75% UTT represent 65% of treated CH, 65% of untreated CH, 70% of untreated CH, 70% of treated CH, 75% of untreated CH and 75% of treated CH in the composite mixtures, respectively.

### 3.2. Flexural Test

The maximum load was applied in the middle of a freely supported beam specimen in the three-point bending test, and the average flexural test results from three samples were analyzed. As shown in Figure 4, treated CH polymer composites have higher flexural strength (7.5 MPa) than untreated CH polymer composites. The flexural strength of the untreated CH polymer composite is lower (4.5 MPa) than that of the treated composite. As observed, the flexural strength of the treated CH-reinforced composite improved by 32% with a 25/65 weight ratio composite. The flexural strength values progressively increased as the CH content peaked at 25 wt% CH content and decreased afterward. The reinforcement improved the flexural strength of the composites reinforced with 25 wt% CH powder.

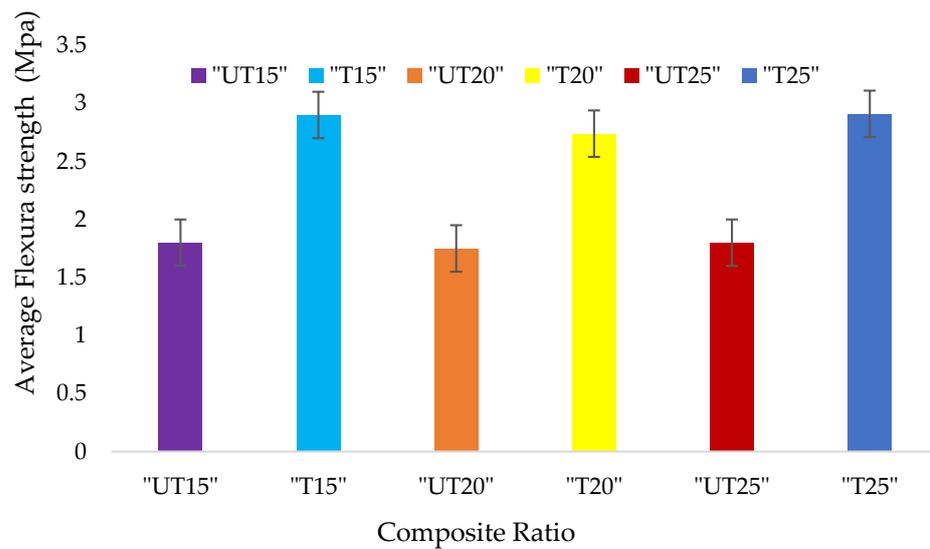


Figure 4. Flexural strength results.

### 3.3. The Impact Test

The impact strength of the created composites was determined using the Charpy impact test. The impact resistance of treated CH slightly increased by 8%, 18%, and 32% with 15 wt%, 20 wt%, and 25 wt% of fiber. The highest value was recorded at 25 wt% of treated CH, as demonstrated in Figure 5. Figure 5 depicts the results of testing V-notched specimens with a depth of 1.15 mm. Compared to treated CH-reinforced and untreated CH composites, treated CH-reinforced polymer composites had a significant impact strength of 21 kJ/m<sup>2</sup>, increasing the impact strength. However, the impact strength of the untreated CH-reinforced composite was lower, but it improved as more CH fiber was added. The data show that the optimum value (18.11 kJ/m<sup>2</sup>) is found in the 25/65 treated reinforced fibers. The optimal values of the composite improved by 12.45% compared to the untreated husk-reinforced composite.

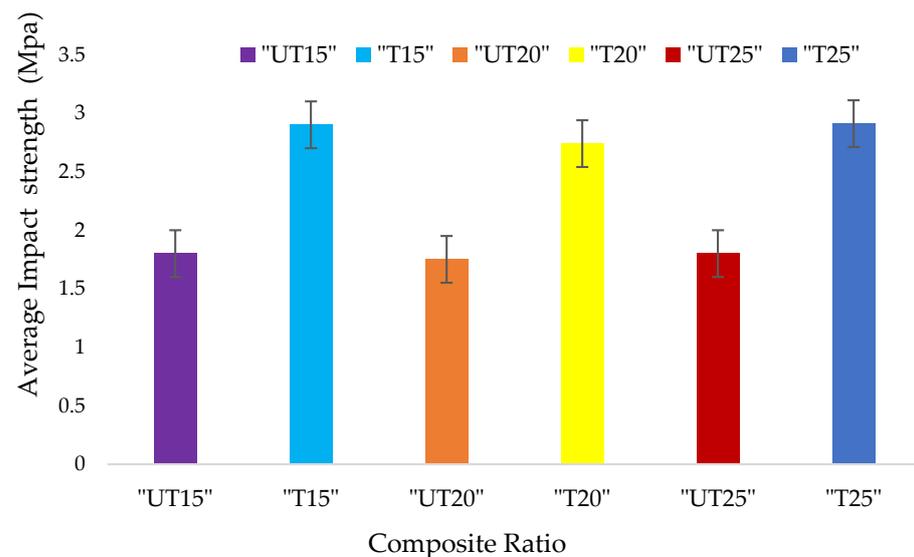
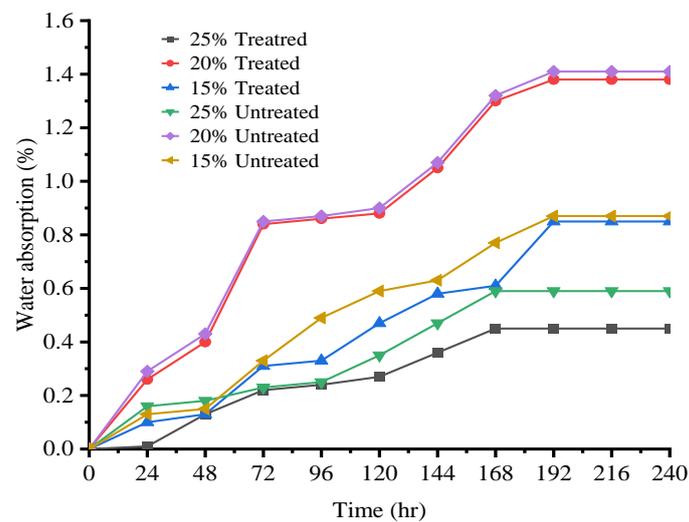


Figure 5. Impact strength test results of CH composite.

### 3.4. Moisture Absorption

Figure 6 depicts reflective plots of the water uptake of treated and untreated CH over time. According to the data, untreated CH absorbs more water than treated CH at a

15/75 CH-reinforced composite ratio. The water absorption properties of all husks increased for the first 12 h, then varied from 4% to 5%, and remained relatively constant after 88 h. Chemical fiber treatments can aid in reducing water uptake in the fiber. As a reinforcement in composite materials, the expansion of the fiber after water absorption induces microcracks in the composite material, impacting its mechanical properties [30]. The fiber-matrix compatibility can help to reduce water absorption in natural fiber composites without any chemical treatment of the fibers.



**Figure 6.** Moisture absorption test results of CH composites.

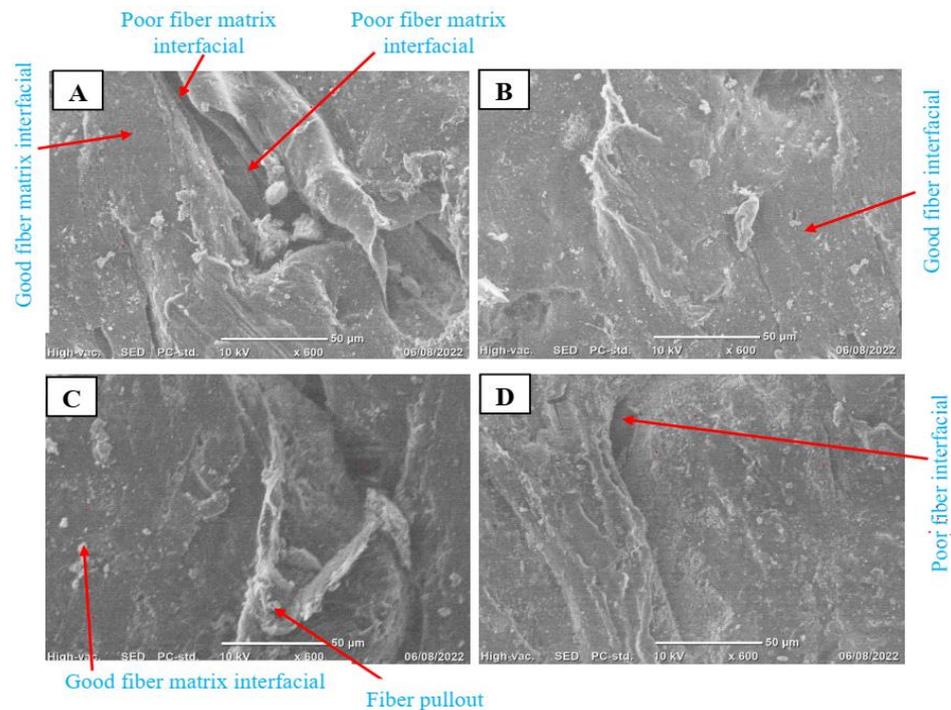
SEM was used to assess the ruptured surfaces of the fabricated composites after tensile testing. The SEM image of CH/HDPE and the 25/65 and 15/75 CH composites was demonstrated in Figure 7. Figure 7 illustrates SEM image data of fabricated composites cured with MXD harder. According to elemental analysis, the HDPE and ABS contents of CH-filled composites ranged between 43.43 and 75.23% and 26.23 and 31.75%, respectively. SEM images clearly show morphological variations between treated CH and untreated CH with the matrix, as illustrated in Figure 7. Composites prepared with treated polymers were more homogeneous than composites prepared with untreated polymers. SEM images revealed that the micro-voids and holes in the composite structure were reduced due to improved filled-matrix interaction via chemical treatment and surface coating [36].

The Images show that the treated CH composite has better bonding. While a robust interfacial bond is required to create materials for interior and structural applications, a weak interfacial link increases toughness by promoting the pullout effects required in energy absorption, i.e., impact strength. The foremost variability within the middle of treated and untreated samples was found for the elastic response of the fabric, attributable to a micro-sized dimension of the CH that performed as a defect in HDPE films. The morphological structure presented by SEM images in Figure 7 shows unit visible changes within the final crystallinity of the composites. The filler material and coupling agent is directive to upgrade the mechanical properties similarly as causative allowable lead to different properties; the SEM images can be used to determine the composite's failure mechanism. The SEM image (Figure 7) depicts some of the observed failure mechanisms of the composite, while the peaks indicate the structures of treated and untreated CH with HDPE composites (Figures 8 and 9). The measurements of the conditions used were: CuK $\alpha$  radiation, a tension of 40 kV, a current of 30 mA and 0.05 (20/5 s) scanning from values of 2 $\theta$  as it enters 10 to 85 $^\circ$ (2 $\theta$ ) [37,38]. The crystallinity index, CI, was calculated because of

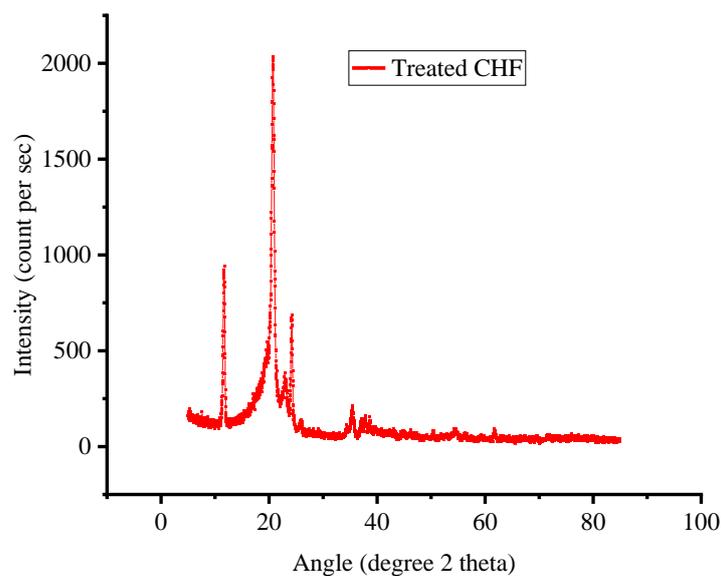
the magnitude relation of the intensity variation within the peak position at 18° and 22° according to Equation (3) [37,38].

$$CI = \frac{I_{22} - I_{18}}{I_{22}} \quad (3)$$

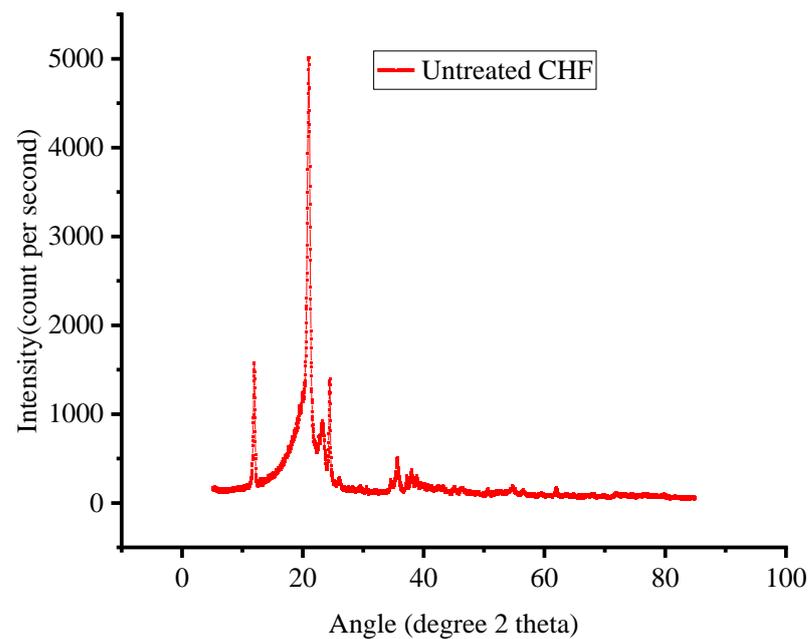
where  $I_{22}$  represents the highest intensity of the 002-lattice reflection of the cellulose  $I$  and  $I_{18}$  represent the highest intensity of X-ray scattering wide band attributable to the amorphous fraction of the sample.



**Figure 7.** SEM image of (A) 75 wt% treated, (B) 65 wt% treated, (C) 75 wt% untreated, and (D) 65 wt% untreated CH composites.



**Figure 8.** Diffraction of X-rays of NaOH-treated composites.



**Figure 9.** Diffraction of X-rays of untreated composites.

The crystallographic nature of CHF was characterized by exploitation diffraction. The diffractograms for CHF are shown in Figures 8 and 9, and these composites have semi-crystalline properties. The optical phenomenon peak for  $2\theta$  was determined to be between 20 and 24, which corresponds to the polysaccharide crystallographic planes (002) [39,40]. The fabric diffraction peaks are caused by crystallinity scattering, whereas disordered areas cause the diffuse background. Fibers demonstrated 50% crystallinity using this approach. Figure 8 illustrates an XRD pattern of treated CHF with a broad and robust peak centered at  $2\theta = 20^\circ$ , and Figure 9 depicts a weak peak at  $42^\circ$  [39,40]. When compared to the diffraction peaks of treated and untreated CHFs and composites, it is clear that composites exhibit diffraction (XRD) patterns similar to the matrix. The composites have firm peaks of about  $20\text{--}24^\circ$ , corresponding to treated and untreated samples. On the other hand, a very modest peak at  $2\theta = 38^\circ$  belonging to CHF was seen exclusively for composites containing 25 wt% fiber. Every result showed that the CHF particles were well disseminated in the chemical systems.

#### 4. Conclusions

Composites of high-density polyethylene/coffee husk (HDPE/CH) reinforcement material made from post-industrial waste coffee husk (CH) were prepared and analyzed. The mechanical properties of CH polymer composites, such as tensile, flexural, and impact strength and water absorption, were investigated in this study. The coffee husk fibers (CHF) were successfully manufactured at 25 wt%, 20 wt%, 15 wt% fiber and 65 wt%, 70 wt%, and 75 wt% polymers. Based on the findings of the initial study, the following conclusions can be reached:

- We prepared and analyzed HDPE functionalized organic reinforcement CH from a coffee industry post-industrial residue.
- The results demonstrate an appropriate reinforcement effect, as evidenced by increased tensile modulus and tensile strength.
- These composites' improved mechanical properties make them suitable for various applications.
- The organic reinforced composites behaved differently, increasing melting temperature and decreasing crystallization.
- ABS was grafted into HDPE macromolecules to enhance matrix-filler compatibility.
- The SEM image can be used to determine the failure mechanism of composites.

- The effects of CHF loading content and the addition of a coupling agent, acrylonitrile butadiene styrene (ABS), on composite processing and properties were investigated.
- This study's findings can help determine the best weight ratio for developing CH polymer composites for various applications.

**Author Contributions:** The idea, experimental work, the analysis of data, and the writing throughout the work were carried out by B.T.A. Examination and correction and supervision throughout the work were carried out by H.A. and G.S.T. Correction of the final manuscript, review and editing, proofreading, and funding acquisition for APC cover were carried out by N.H. All authors have read and agreed to the published version of the manuscript.

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**Data Availability Statement:** The data described in this study are accessible on request from the corresponding author.

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**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. Gonçalves, B.M.; Camillo, M.D.; Oliveira, M.P.; Carreira, L.G.; Moulin, J.C.; Fantuzzi Neto, H.; de Oliveira, B.F.; Pereira, A.C.; Monteiro, S.N. Surface treatments of coffee husk fiber waste for effective incorporation into polymer biocomposites. *Polymers* **2021**, *13*, 3428. [[CrossRef](#)] [[PubMed](#)]
2. Liminana, P.; Garcia-Sanoguera, D.; Quiles-Carrillo, L.; Balart, R.; Montanes, N. Development and characterization of environmentally friendly composites from poly(butylene succinate) (PBS) and almond shell flour with different compatibilizers. *Compos. Part B Eng.* **2018**, *144*, 153–162. [[CrossRef](#)]
3. Lascano, D.; Quiles-Carrillo, L.; Balart, R.; Boronat, T.; Montanes, N. Toughened poly (lactic acid)-PLA formulations by binary blends with poly(butylene succinate-co-adipate)-PBSA and their shape memory behaviour. *Materials* **2019**, *12*, 622. [[CrossRef](#)] [[PubMed](#)]
4. Zhang, Z.; Cai, S.; Li, Y.; Wang, Z.; Long, Y.; Yu, T.; Shen, Y. High performances of plant fiber reinforced composites—A new insight from hierarchical microstructures. *Compos. Sci. Technol.* **2020**, *194*, 108151. [[CrossRef](#)]
5. da Luz, F.S.; da Costa Garcia Filho, F.; del-Río, M.T.G.; Cassiano Nascimento, L.F.; Pinheiro, W.A.; Monteiro, S.N. Graphene-incorporated natural fiber polymer composites: A first overview. *Polymers* **2020**, *12*, 1601. [[CrossRef](#)]
6. Mokhothu, T.H.; John, M.J. Review on hygroscopic aging of cellulose fibres and their biocomposites. *Carbohydr. Polym.* **2015**, *131*, 337–354. [[CrossRef](#)]
7. Sood, M.; Dwivedi, G. Effect of fiber treatment on flexural properties of natural fiber reinforced composites: A review. *Egypt. J. Pet.* **2018**, *27*, 775–783. [[CrossRef](#)]
8. Tahir, N.; Bhatti, H.N.; Iqbal, M.; Noreen, S. Biopolymers composites with peanut hull waste biomass and application for Crystal Violet adsorption. *Int. J. Biol. Macromol.* **2017**, *94*, 210–220. [[CrossRef](#)]
9. Quiles-Carrillo, L.; Montanes, N.; Jorda-Vilaplana, A.; Balart, R.; Torres-Giner, S. A comparative study on the effect of different reactive compatibilizers on injection-molded pieces of bio-based high-density polyethylene/poly lactide blends. *J. Appl. Polym. Sci.* **2019**, *136*, 47396. [[CrossRef](#)]
10. Kallel, F.; Bettaieb, F.; Khiari, R.; García, A.; Bras, J.; Chaabouni, S.E. Isolation and structural characterization of cellulose nanocrystals extracted from garlic straw residues. *Ind. Crops Prod.* **2016**, *87*, 287–296. [[CrossRef](#)]
11. Ortiz-Barajas, D.L.; Arévalo-Prada, J.; Fenollar, A.O.; Rueda-Ordóñez, Y.J.; Torres-Giner, S. Torrefaction of coffee husk flour for the development of injection-Molded green composite pieces of polylactide with high sustainability. *Appl. Sci.* **2020**, *10*, 6468. [[CrossRef](#)]
12. Önal, E.; Uzun, B.B.; Pütün, A.E. Bio-oil production via co-pyrolysis of almond shell as biomass and high density polyethylene. *Energy Convers. Manag.* **2014**, *78*, 704–710. [[CrossRef](#)]
13. Torres-Giner, S.; Torres, A.; Ferrándiz, M.; Fombuena, V.; Balart, R. Antimicrobial activity of metal cation-exchanged zeolites and their evaluation on injection-molded pieces of bio-based high-density polyethylene. *J. Food Saf.* **2017**, *37*, e12348. [[CrossRef](#)]
14. Corrado, A.; Polini, W. Measurement of high flexibility components in composite material by touch probe and force sensing resistors. *J. Manuf. Process.* **2019**, *45*, 520–531. [[CrossRef](#)]
15. Chen, J.; Gao, X. Directional dependence of electrical and thermal properties in graphene-nanoplatelet-based composite materials. *Results Phys.* **2019**, *15*, 102608. [[CrossRef](#)]
16. Posada, J.C.; Jaramillo, L.Y.; Cadena, E.M.; García, L.A. Bio-based composites from agricultural wastes: Polylactic acid and bamboo *Guadua angustifolia*. *J. Compos. Mater.* **2016**, *50*, 3229–3237. [[CrossRef](#)]

17. Saha, D.; Majumdar, M.K.; Das, A.K.; Sarwaruddin Chowdhury, A.M.; Ashaduzzaman, M. Structural nanocomposite fabrication from self-assembled choline chloride modified kaolinite into poly(Methylmethacrylate). *J. Compos. Sci.* **2019**, *3*, 83. [[CrossRef](#)]
18. Faruk, O.; Bledzki, A.K.; Fink, H.P.; Sain, M. Biocomposites reinforced with natural fibers: 2000–2010. *Prog. Polym. Sci.* **2012**, *37*, 1552–1596. [[CrossRef](#)]
19. Azmi, M.; Zameri, M.; Saman, M.; Sharif, S. Proposed framework for End-Of-Life vehicle recycling system implementation in Malaysia. In Proceedings of the 11th Global Conference on Sustainable Manufacturing, Berlin, Germany, 23–25 September 2013; Volume 6, pp. 187–193.
20. Thio, Y.S.; Argon, A.S.; Cohen, R.E.; Weinberg, M. Toughening of isotactic polypropylene with CaCO<sub>3</sub> particles. *Polymer* **2002**, *43*, 3661–3674. [[CrossRef](#)]
21. Da Silva, A.L.N.; Rocha, M.C.G.; Moraes, M.A.; Valente, R.C.A.R.; Coutinho, F.M.B. Mechanical and rheological properties of composites based on polyolefin and mineral additives. *Polym. Test.* **2002**, *21*, 57–60. [[CrossRef](#)]
22. Mohseni Tabar, M.; Tabarsa, T.; Mashkour, M.; Khazaeian, A. Using silicon dioxide (SiO<sub>2</sub>) nano-powder as reinforcement for walnut shell flour/HDPE composite materials. *J. Indian Acad. Wood Sci.* **2015**, *12*, 15–21. [[CrossRef](#)]
23. Mustafa, S.N. Effect of Kaolin on the Mechanical Properties of Polypropylene/Polyethylene Composite. *Diyala J. Eng. Sci.* **2012**, *05*, 162–178. [[CrossRef](#)]
24. Goulart, S.A.S.; Oliveira, T.; Teixeira, A.A.; Miléo, P.C.; Mulinari, D.R. Mechanical behaviour of polypropylene reinforced palm fibers composites. *Procedia Eng.* **2011**, *10*, 2034–2039. [[CrossRef](#)]
25. Sobczak, L.; Lang, R.W.; Haider, A. Polypropylene composites with natural fibers and wood—General mechanical property profiles. *Compos. Sci. Technol.* **2012**, *72*, 550–557. [[CrossRef](#)]
26. Awal, A.; Cescutti, G.; Ghosh, S.B.; Müssig, J. Interfacial studies of natural fibre/polypropylene composites using single fibre fragmentation test (SFFT). *Compos. Part A Appl. Sci. Manuf.* **2011**, *42*, 50–56. [[CrossRef](#)]
27. Bekele, A.E.; Lemu, H.G.; Jiru, M.G. Exploration of Mechanical Properties of Enset—Sisal Hybrid Polymer Composite. *Fibers* **2022**, *10*, 14. [[CrossRef](#)]
28. Vázquez Fletes, R.C.; Rodrigue, D. Effect of wood fiber surface treatment on the properties of recycled hdpe/maple fiber composites. *J. Compos. Sci.* **2021**, *5*, 177. [[CrossRef](#)]
29. Azam, F.A.A.; Royan, N.R.R.; Yuhana, N.Y.; Radzuan, N.A.M.; Ahmad, S.; Sulong, A.B. Fabrication of porous recycled HDPE biocomposites foam: Effect of rice husk filler contents and surface treatments on the mechanical properties. *Polymers* **2020**, *12*, 475. [[CrossRef](#)]
30. Yiga, V.A.; Pagel, S.; Lubwama, M.; Epple, S.; Olupot, P.W.; Bonten, C. Development of fiber-reinforced polypropylene with NaOH pretreated rice and coffee husks as fillers: Mechanical and thermal properties. *J. Thermoplast. Compos. Mater.* **2020**, *33*, 1269–1291. [[CrossRef](#)]
31. Nemeth, I. Hotavvezetetek hoszigeteles alatti korrozioja elleni vedelem. *Korroz. Figy.* **2010**, *35*, 36–38.
32. Batu, T.; Lemu, H.G. Investigation of mechanical properties of false banana/glass fiber reinforced hybrid composite materials. *Results Mater.* **2020**, *8*, 100152. [[CrossRef](#)]
33. ASTM D790-07; Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials. Annual Book of ASTM Standards. ASTM International: West Conshohocken, PA, USA, 2002; Volume 14, pp. 1–12.
34. ASTM D256-97; Standard Test Methods for Determining the Izod Pendulum Impact Resistance of Plastics. Annual Book of ASTM Standards. ASTM International: West Conshohocken, PA, USA, 1998; Volume 8, pp. 14–18.
35. ASTM D570; Standard Test Method for Water Absorption of Plastics. ASTM International: West Conshohocken, PA, USA, 2010; Volume 98, pp. 25–28.
36. Vincenzo, A.; Cataldo, A.; Cavallaro, G. Coffee grounds as filler for pectin: Green composites with competitive performances dependent on the UV irradiation. *Carbohydr. Polym.* **2017**, *170*, 198–205.
37. Rosa, M.; Medeiros, E.; Malmonge, J.; Gregorski, K.; Wood, D.; Mattoso, L.; Glenn, G.; Orts, W.; Imam, S. Cellulose nanowhiskers from coconut husk fibers: Effect of preparation conditions on their thermal and morphological behavior. *Carbohydr. Polym.* **2010**, *81*, 83–92. [[CrossRef](#)]
38. Collazo-Bigliardi, S.; Ortega-Toro, R.; Chiralt Boix, A. Isolation and characterisation of microcrystalline cellulose and cellulose nanocrystals from coffee husk and comparative study with rice husk. *Carbohydr. Polym.* **2017**, *191*, 205–215. [[CrossRef](#)] [[PubMed](#)]
39. Reis, R.S.; Tienne, L.G.P.; Souza, D.D.H.S.; Marques, M.D.F.V.; Monteiro, S.N. Characterization of coffee parchment and innovative steam explosion treatment to obtain microfibrillated cellulose as potential composite reinforcement. *J. Mater. Res. Technol.* **2020**, *9*, 9412–9421. [[CrossRef](#)]
40. Pujol, D.; Liu, C.; Gominho, J.; Olivella, M.; Fiol, N.; Villaescusa, I.; Pereira, H. The chemical composition of exhausted coffee waste. *Ind. Crops Prod.* **2013**, *50*, 423–429. [[CrossRef](#)]