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Investigation of Mechanical, Thermal, and Surface Properties of LDPE/TPS/Cellulose Additives Composites towards Using in Packaging Application

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ABSTRACT

The widespread use of petroleum products and non-biodegradable materials in packaging has caused serious damage to the environment. The growing demand for durable packaging has encouraged researchers to explore non-toxic, compatible, and biodegradable materials. Cellulose as organic natural sustainable polymer became more usable in the fields of medical and environmental applications. In this work, the mechanical, thermal history, and water absorption characteristics of low-density polyethylene LDPE / thermoplastic starch (TPS) composite materials reinforced with different cellulose materials, including 2.5 and 5% of each of sawdust, powder cellulose, and crystalline nanocellulose CNC are examined. In order to assure well dispersion of Cellulose in composites, initially cellulose was added to TPS and then, blending in twin-screw extruder with LDPE at 190°C, and 50 rpm. Thermal history, tensile strength, Young modulus and contact angle were examined for the samples. Numerically simulation using material design-Ansys version 2021 software based on the RVE model was applied to check and validate the mechanical properties with experimental test. The experimental results show that the tensile strength, Young modulus and hardness were increased in general with the increasing of cellulose additives compared with LDPE/TPS and the 5% cellulose indicates significant increasing. The melting point, degree of crystallinity, and enthalpy increase with the increasing of cellulose additions in the DSC data, which supports the reading of tensile strength. The contact angle decreases, and water absorption increases with increasing of cellulose, but it remains within the acceptable percentage compared to LDPE/TPS. On the other hand, the electronic scanning images showed the surface characteristics and the adhesive interference of the samples, the microstructural gave a semi-homogeneous images and no significant cellulose agglomeration or cracks were observed. The numerical results proved a comfortable match with the experimental results regarding the Young's modulus and there was acceptable agreement with hardness, water absorption and melting point of the previous studies.

1. INTRODUCTION

For a number of decades, non-biodegradable plastics made of petroleum-based compounds have been extensively utilized in a variety of applications, especially food packaging, Low-Density Polyethylene (LDPE) is one of the common petroleum-based or fossil plastics used in food packaging [1]. It is used because of its unique qualities: high strength, durability, flexibility, lightweight and low cost. However, these materials are not prone to degradation or available for recycling after using, causing seriously polluting the environment [2]. According to the data, there will be nine billion people on the planet by 2050, which would significantly raise the need for plastics in order to meet human needs. As a result, there is no question that it will lead to a rise in plastic trash, which directly affects pollution air, water, and global warming [3]. An affordable and flexible way to improve the biodegradability of petroleum-based polymers is to blend natural and synthetic polymers that degrade naturally. This results in partly biodegradable, sustainable plastics with a variety of appealing qualities. Amongst the biodegradable polymers, starch is a naturally occurring, renewable, inexpensive, and plentiful biopolymer which is increasingly used to create biodegradable composites and blends [4]. Despite the fact that starch is a biodegradable polymer that can be produced in vast numbers at a reasonable cost, handled with ease, and forms film products with low oxygen permeability. The main problem with native starch is that it is brittle and hydrophilic; these restrict its range of uses, including the production of food packaging. Glycerol, glycol, and sorbitol are an example of plasticizers that can be used to plasticize starch to increase its flexibility and ease of processing [5]. The process of turning starch into thermoplastic starch (TPS) at a temperature that is higher than that of starch's gelatinization, usually between 65 and 90°C. By boosting chain mobility and enhancing the biopolymer's extensibility and flexibility, the

plasticizers lessen intermolecular stresses [4]. The primary drawback of LDPE/TPS composites lies in the incompatibility between polar TPS and nonpolar LDPE, which inevitably leads to a decline in the mechanical properties of the composites. The incompatibility between TPS and LDPE phases prevents the formation of strong interfacial hydrogen bonds. Numerous studies have been conducted to enhance the compatibility of TPS and LDPE by modifying starch [6, 7] and incorporating compatibilizers (or coupling agents) into LDPE/TPS composites [8-10]. Compatibilizers are widely recognized in industrial applications as an effective means to improve the weak interfacial bonding and compatibility of immiscible polymer blends.

For LDPE/TPS composites, LDPE grafted with maleic anhydride (LDPE-g-MA) has been identified as one of the most commonly used and practical compatibilizers [1], as it can form hydrogen bonds with the hydroxyl groups of TPS. While LDPE/TPS/LDPE-g-MA films exhibit improved degradability, their mechanical properties remain suboptimal. A promising approach to enhance mechanical performance involves incorporating various cellulose materials to reinforce the blend and produce biodegradable composites [9].

In recent years, cellulose has gained significant attention as one of the most abundant natural polymers on Earth, playing a crucial role in providing mechanical strength and structural support. Due to its linear and highly regular molecular structure with multiple hydroxyl groups, cellulose polymers can form well-organized crystalline regions stabilized by hydrogen bonds. These crystalline regions contribute to the excellent mechanical properties of cellulose [11].

Cellulose nanocrystals (CNCs), which typically have a width of 3 to 20 nm and a length of approximately 50–500 nm, are obtained through the acid hydrolysis of wood or other cellulose-containing materials [12]. CNCs exhibit a remarkable combination of biophysicochemical properties, including biocompatibility, biodegradability, lightweight nature, non-toxicity, stiffness, renewability, sustainability, optical transparency, low thermal expansion, gas impermeability, adaptable surface chemistry, and enhanced mechanical properties [13]. However, extracting CNCs from amorphous cellulosic materials requires extensive and costly pretreatments, making them a high-value product with an estimated cost of around €50/kg [6].

A particularly promising alternative is powdered cellulose, which is generated as a byproduct of various pulping processes used to extract high-value compounds from wood, such as lignin (via extraction with acetic acid-containing water). In contrast, organosolv cellulose, which is extracted using ethanol or methanol, contains predominantly alcohol functional groups [6].

Among plant fibers, sawdust is a noteworthy material composed of lignin, cellulose, and hemicellulose. The reinforcement of thermoplastics with sawdust has garnered significant interest due to its numerous advantages over traditional reinforcing materials. These benefits include weight reduction in composites, reuse of waste materials, renewability, high specific strength and stiffness, and significant cost-saving potential. However, the improper disposal or incineration of sawdust and its derivatives poses a serious environmental threat [14, 15].

In this study, six LDPE/TPS/Cellulose composites were prepared by incorporating 2.5% and 5% of sawdust, CNC, and cellulose powder into the LDPE/TPS blend. A twin-screw extruder was used for melting and mixing the composites at a speed of 50 rpm and a temperature of 190°C.

Differential scanning calorimetry (DSC), a universal tensile testing machine, scanning electron microscopy (SEM), and a contact angle measurement device were employed to analyze the thermal, mechanical, microstructural, and surface properties of the composites, respectively. Additionally, numerical simulation using Material Design-Ansys 2021 software, based on the Representative Volume Element (RVE) model, was conducted to verify the mechanical properties obtained from experimental tests. Furthermore, the hardness, water absorption, and melting point of the composites were correlated with findings from previous studies.

2. EXPERIMENTAL PART

2.1 Materials

The materials used in this work are as following:

1- LDPE used as a matrix, was provided from Saudi Basic Industries Corporation (SABIC) and had the following characteristics: MFI (190°C /2.16 kg; ISO 1133-1)=2.1 g/10 min, ρ=0.91 g/cm^3, Tm=112°C, purity=99.

2- Maleic anhydride-grafted-polyethylene LDPE_g_MA, (ρ=0.92 g/cm^3, Tm=105°C) as a coupling agent was purchased from Coace chemical company limited _China, purity=99.

3- Starch was purchased from Arab food industries company, Jordan. contain 25% amylose and 75% amylopectin.

4- Sawdust obtained from sawmills and lumber markets, from poplar wood of Russian origin, using sieved with apertures of 180 µm, ρ=250 kg/cm^3.

5- Cellulose powder was purchased from himedia company, India. Particle size=18.24 µm, ρ=1.5 g/cm^3, purity=95.

6- Cellulose nanocrystal CNC was purchased from Nanografi company, Turkey, with the following characteristics: ρ=1.49 g/cm^3, crystallinity=92%, particle size =10_20 nm wide, 300_900 nm length, purity=99.

2.2 Preparation of composites

Composites of LDPE/TPS with various cellulose materials performed as following:

1- Various cellulose materials were prepared:

•Sawdust was dried at 60°C for two hours. then sieved using standard sieve (H-4325 U.S.A) with apertures of 180 µm. It was then mixed (2.5 g) with distilled water of (60 ml)

•The powder cellulose (2.5 g) was combined with ethanol alcohol (50 mL).

•CNC (2.5%wt) mixed with distilled water to obtain homogenous suspension.

•The cellulose materials in a, b, and c were dispersed for 20 min, at 27°C and power 500 watt using Ultrasonic (SJIA-1200W MTI).

2- TPS polymer was prepared by mixed the starch with 30% Glycerol at 140°C for 8 minutes using mechanical stirring.

3- The cellulose materials were mixed with the TPS polymer, for 30 min, at 27°C. The mixed solution was standing in bags made from plastic one-night then it was dried by placing in an oven at 40°C for 24 hours after that the mixture was pelletized to 40–60 mesh size.

4- The samples produced in item (3) were mixed with LDPE granules and LDPE-g-MA coupling agent according to the proportions in Table 1. The obtained samples melted using twin-screw extruder (SLJ-30A.chine) of 50 rpm and at 190°C [16].

5- The composites in item 4 produced as a sheet from twin-

screw extruder and subjected to the hot pressing at 190°C, pressure of 5 Mpa for 5 minutes in a pressing device, then the samples become ready for different tests.

Table 1. The proportions of Cellulose, LDPE-g-MA, TPS, and LDPE in different LDPE/TPS/ Cellulose additives composites (wt.%)

Sample Code	Type of Cellulose	Cellulose wt.%	LDPE wt.%	TPS wt.%	LDPE-g-MA wt.%
LDPE/TPS/ LDPE-g-MA	-	0	85	13	2
L1S2.5	Sawdust				
L1P2.5	Powder	2.5	82.5	13	2
L1N2.5	CNC				
L2S5	Sawdust				
L2P5	Powder	5	80	13	2
L2N5	CNC				

2.3 Characterization

2.3.1 DSC test

The melting temperature and the thermal history of the composites are measured by Differential scanning calorimetry (DSC). Using DSC I (CW-05 G) (ASTM D3418) on a temperature range from 25°C to 250°C with a 10°C/min heating rate to carry out the measurement [17]. The degree of crystallization was considered by the equation as shown below:

$$\chi c\ \% = \frac{\Delta Hm}{\Delta H0} \times 100\%$$

where, ΔHm is the melting enthalpy, and ΔH0 is a theoretical value of the melting enthalpy of 100% crystalline LDPE. The value of ΔH0 = 293 J/g was used in a degree of crystallinity calculations [18].

2.3.2 Tensile test

At least ten specimens measuring 40 × 6 × 2 mm^3 were created from the composites of each formulation. Were used to test the tension properties according to ASTM 638-IV [16], the tensile strength was measured at 19 ± 2°C with a relative humidity of 62 ± 5%, using an Instron 5556 testing machine at a tensile speed of 15 mm/min [19], a graph paper acquired the relationship between stress and strain from the device.

2.3.3 Hardness

Utilizing a Shore D hardness tester and ASTM D-2240 as a basis to examine the impact of the LDPE content on the hardness of the LDPE cellulose additions, the hardness of each composite was tested seven times for each composite [20].

2.3.4 Contact angle

Using an optical contact angle measurement device (Data Physics aSL200B at KINO Industry Co., Ltd., USA), to assess the impact of additives on the wettability of pure materials. A droplet of 5 µl distilled water was placed on the films' surface. For every type of film, three were utilised, and 10 measurements were made at various locations on the film's surface following the drop deposition. After that, the mean values were determined and presented.

2.3.5 Water absorption

Using 3 × 1 mm^2 strips with a 1 mm thickness, the composite samples' water absorption was calculated using the ASTM D570-98 technique. The vacuum oven was used to dry the samples were in at 70°C for 24 hours earlier than measurement, and they were then promptly weighed. For three days, the samples were soaked in distilled water to determine the water absorption. After removing each sample from the container, its water absorption was measured by weighing it. This is how the water absorption (%) was determined [21]:

$$Water\ absorption = \frac{W_f - W_0}{W_0} \times 100\%$$

where, W_f = Weight after immersed in water, W_0 = Weight before immersed in water

2.3.6 Surface morphology

Using a 5.0 kV SEM microscope to analyze the cellulose's dispersion in the LDPE sample's polymer matrix, the surface morphology of several specimens was investigated.

3. NUMERICAL PART

3.1 Material designer

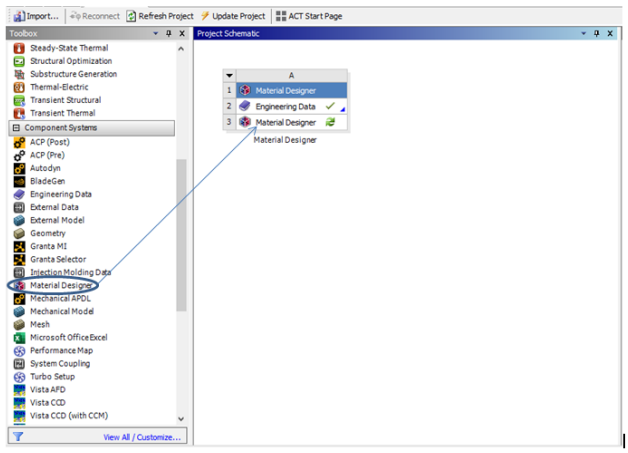

Figure 1. Material designer in Workbench Ansys 2021

Ansys software based on the finite element was used through material design element to obtain the final mechanical properties of the composites in order to validate with the experimental behavior [22]. The material design analysis of the composite is a powerful tool for mechanical investigation according to the following procedures:

1- Creation of the material data and the engineering data (Figure 1).

2- Adding the material properties of the Cellulose additives as fillers and the LDPE as matrix (Table 2).

Table 2. Properties of Cellulose additives and LDPE

Material	Density (g\cm^3)	E (G pa)	Poisson Ratio	G (G pa)

LDPE	0.91	0.3	0.34	2.238
CNC	1.49	100	0.31	7.62235
Sawdust	2.50	7	0.3	5.846
Powder cellulose	1.5	6	0.3	5.769

3.2 Numerical simulation

The material designer can setup a unit cell of our composite and calculate the homogenized material properties to use in this analysis.

Material designer uses the Ansys space claim direct modeler interface and define a representative volume element (RVE) of its microstructure. Unidirectional composite as chosen as RVE type (Figure 2).

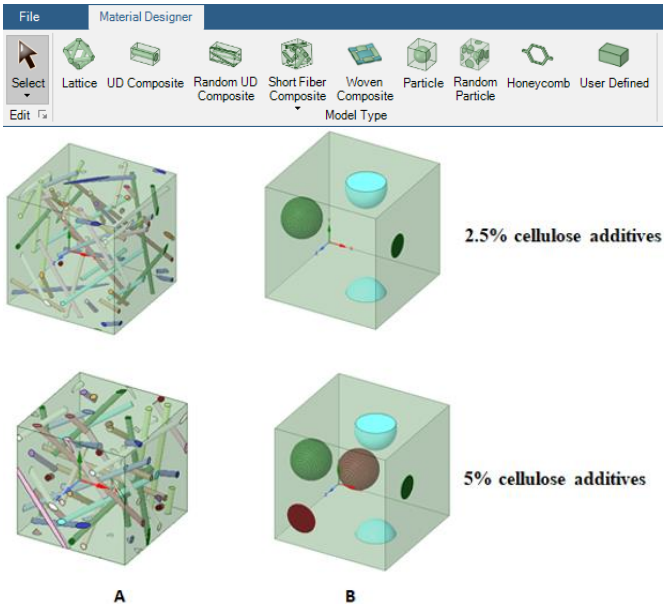

Figure 2. RVE Geometry of LDPE composite at 2.5 and 5% of each of (A) CNC and sawdust (B) powder cellulose

3.3 Mesh attributes

Before any volume meshing, the particles and matrix volume were given the required material properties (Figure 3).

3.4 Solution

The Ansys Mechanical APDL solver is launched in the background and will solve a number of different setups depending on what type of material properties setting.

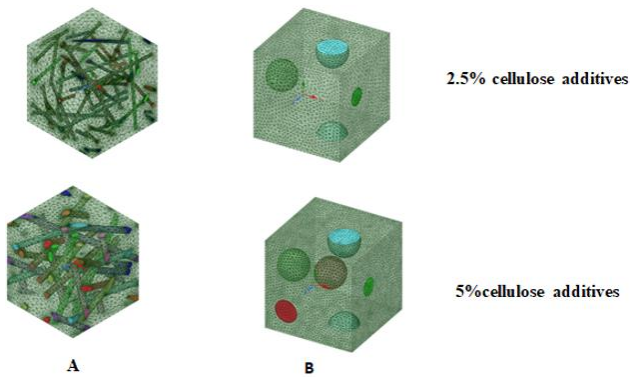

Figure 3. Mesh of LDPE composite at 2.5 and 5% of each of (A) CNC and sawdust, (B) Powder cellulose

4. RESULTS AND DISCUSSION

4.1 Experimental results

4.1.1 DSC

Figure 4(A) and Table 3 indicate that addition of TPS and LDPE-g-MA to the LDPE may produce multiple melting peaks or a broader melting range. TPS has a lower melting point and its addition may obtain a reduction in the overall crystallinity of the composite to about 3.863, leading to a decrease in the melting temperature of LDPE from about 112 to 105.82℃. The presence of may slightly shift the melting peaks due to improved interfacial adhesion by LDPE-g-MA.

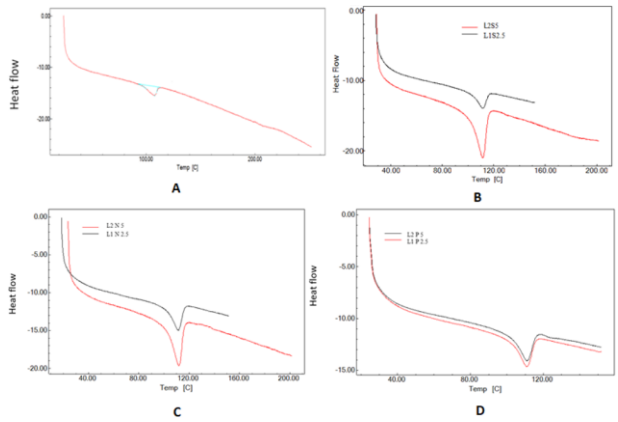

Figure 4. The DSC thermograms of (A) LDPE/TPS (B)LDPE/TPS/Sawdust (C) LDPE/TPS/CNC (D) LDPE/TPS /Powder

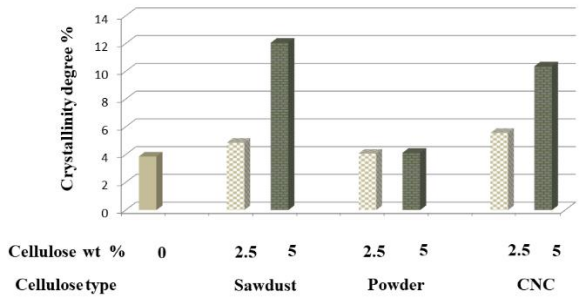

Figure 5. The crystallinity degree of LDPE/TPS/various cellulose additives

Figure 4(B)-(D) indicates the DSC results for 2.5, and 5% of each of sawdust, powder, and CNC adding to the LDPE/TPS. The melting temperature (Tm) corresponds to the temperature at which crystalline regions melt into a liquid phase. The onset, peak, and melting temperature can be determined from the melting peak in DSC thermograms. The melting temperatures, melting enthalpy, and crystallization degree increased with the increasing of cellulose proportions, compared with the original LDPE/TPS sample, this enhances the ability to achieve higher mechanical performance specifications. The arrangement of the composites for increasing in melting enthalpy, crystallization degree and melting temperatures were occurs due to the sawdust, CNC, and powder adding respectively. From the other hand the arrangement of composite from high to low melting temperature due to the 2.5% proportion was CNC, sawdust, and powder. The 5% proportions show significant change in

melting point for sawdust, while a slightly change in melting temperature for CNC and powder composites was observed. This means that the increasing of sawdust concentration may be produce further increasing in the melting temperature, crystallizing degree and melting enthalpy, as illustrates in Table 3 and Figure 5.

Table 3. The values of Tm, onset, end set enthalpy and crystallinity degree of LDPE/TPS/various cellulose additives

Sample	Tm °C	Onset °C	End Set °C	ΔHmJg^{-1} Melting Enthalpy	X_c (%) Crystallinity Degree
LDPE/TPS	105.82	105.85	115.02	11.32	3.863
L1 S 2.5	106.30	105.82	107.05	14.20	4.846
L2 S 5	111.17	103.19	115.45	35.31	12.051
L1 P 2.5	110.83	104.66	115.28	11.93	4.071
L1 P 5	110.96	104.40	115.92	12.09	4.126
L1 N 2.5	111.45	104.20	116.17	16.28	5.556
L2 N 5	111.82	103.77	115.91	30.34	10.354

4.1.2 Tensile test

The tensile strength values of LDPE/TPS/cellulose composites are shown in the Figure 6 for the cellulose-free composites, the presence of TPS content significantly reduced the tensile strength. It might be related to TPS and LDPE's incompatibility which is less rigid than LDPE. However, the presence of LDPE-g-MA as a compatibilizer can modify interfacial adhesion between LDPE and TPS, partially compensating for the decreasing in tensile strength.

The overall tensile strength will depend on the balance between these effects.

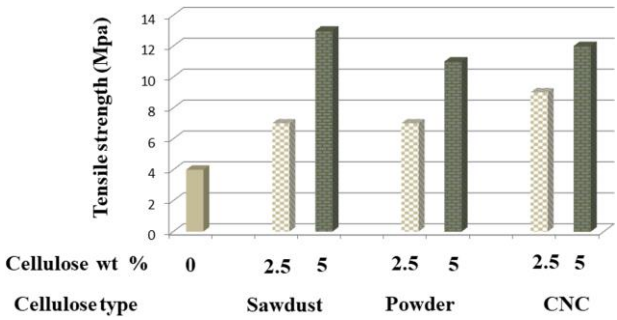

Figure 6. Tensile behavior of LDPE/TPS /Cellulose additives

However, the addition of 2.5 and 5% by weight of sawdust, powder and CNC in LDPE leads to an overall increase in the tensile strength of the composites, these are consistent with the results of DSC, which is suitable for packaging applications. at 2.5% cellulose loading the tensile strength of CNC is increased by 55%, sawdust and powder is increased by 42 at 5% higher cellulose content. The tensile strength increased by 69% for sawdust and 66% for CNC and 63% for powder compared to the sample without cellulose. The optimum cellulose content was found to be 5%. First, the excellent compatibility of TPS and CNC resulting from the presence of hydroxyl groups, which form hydrogen bonds and reinforce TPS and the final LDPE/TPS/cellulose composites, could be the reason for the reinforcing impact of cellulose on LDPE/TPS. Second, cellulose may enhance TPS-TPS stress transmission by acting as a binder [4]. Thirdly, because of the hydrogen bonds that develop between cellulose and TPS, consistent cellulose dispersion in the TPS also contributes to improved reinforcing of the completed composites.

4.1.3 Hardness

Hardness, which measures a material's resistance to local deformation, is a crucial factor in the packaging sector as well as many other applications using polymers and composites. It can be enhanced by Homogeneous addition and dispersion of suitable particles in a Polymer [23].

Figure 7 illustrates the hardness decreasing of LDPE/TPS due to the addition of TPS, which is generally softer than pure LDPE. The extent of this decrease will depend on the degree of compatibility and dispersion of TPS within the LDPE matrix facilitated by LDPE-g-MA.

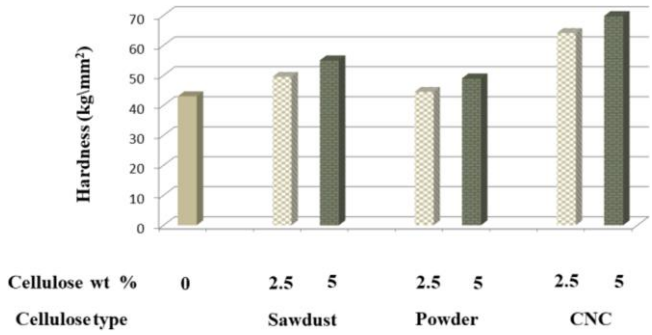

Figure 7. Hardness behavior of LDPE/TPS/Sawdust, LDPE/TPS/Powder, and LDPE/TPS/CNC

The hardness values increased almost linearly with the increase in cellulose content in the mixture for all types of cellulose (sawdust, CNC, and powdered cellulose) at both 2.5% and 5% concentrations, compared to the sample without cellulose. The increase in hardness was attributed to the addition of CNC, sawdust, and powdered cellulose, respectively.

Cellulose, when added to LDPE, acts as a reinforcing filler, enhancing the mechanical properties of the composite, including hardness. It improves the rigidity and strength of the material, resulting in greater hardness compared to LDPE/TPS alone, which aligns with the findings in reference [4].

Hardness plays a crucial role in evaluating the efficiency of food packaging materials. Harder materials exhibit better barrier properties due to their denser and less permeable structure, reducing the transmission of oxygen, carbon dioxide, and moisture—key factors in food spoilage. However, the hardness must remain within an appropriate range for the specific type of packaged material. Excessive hardness can lead to reduced flexibility, increased brittleness at low temperatures, or difficulties in container formation and sealing.

This highlights the importance of plasticizers and a well-controlled mixing process, which can enhance mechanical properties without exceeding the permissible hardness limit. Therefore, the cellulose content in LDPE composites must be carefully considered, as its impact on hardness is significant.

4.1.4 Contact angle

Contact angle is an important parameter characterizing the surface wetting properties of materials. It represents the angle at which a drop of liquid contacts the surface of a solid material. A larger contact angle indicates that the liquid is less likely to spread on the surface, indicating hydrophobicity, while a smaller contact angle indicates better wettability and hydrophilicity [24].

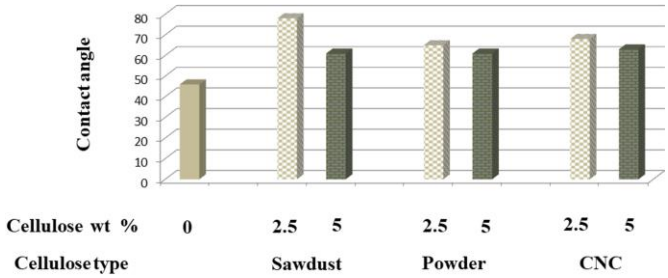

Figure 8. Contact angle behavior of LDPE/TPS /Cellulose additives

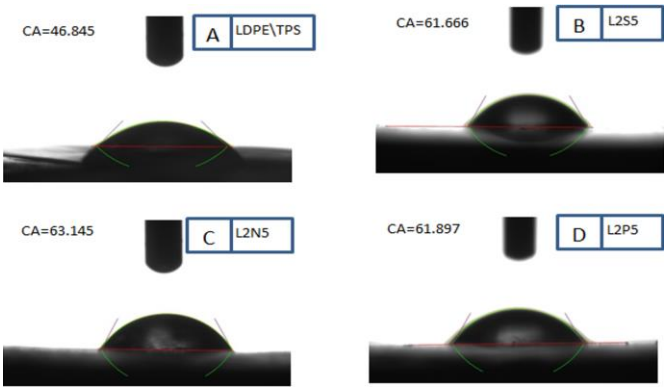

Figure 9. Contact angle of (A) LDPE\TPS (B) 5% sawdust (C)5% CNC and (D) 5% Powder cellulose

Figures 8 and 9 show that the contact angle of LDPE/TPS composite without cellulose will decrease, compared with LDPE, which Exhibits a high contact angle due to its hydrophobic nature. The increasing in surface hydrophilicity attributed to the presence of TPS. The exact reduction in contact angle will depend on the surface distribution of TPS and the effectiveness of LDPE-g-MA in plasticizer the blend.

It can be observed that the contact angle increases for composite contain cellulose with no discernible variation in the contact angle values. of the composites containing 2.5 and 5% cellulose for all type of cellulose (sawdust, CNC and powder cellulose), this phenomena may be explained by the fact that cellulose has a higher crystallinity than starch chains, which reduces their mobility, this lead to increase contact angle and reduce wettability and adhesion

4.1.5 Water absorption

Since water absorption behavior can limit the applications of LDPE/TPS composites, it is considered a crucial characteristic [25]. Figure 10 presents the moisture absorption values for the tested samples.

For the LDPE/TPS blend without cellulose, water absorption increases significantly due to the hydrophilic nature of starch, in contrast to the very low water absorption of LDPE, which is inherently hydrophobic. The presence of TPS introduces polar groups into the matrix, making it more susceptible to water absorption.

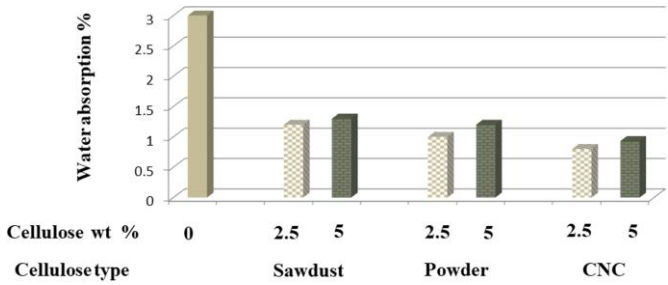

Figure 10. Water absorption behavior of LDPE/TPS/Cellulose additives

LDPE-g-MA can help in decreasing the extent of water absorption by improving the dispersion of TPS, but it will not hide the hydrophilic nature of starch. Compared to LDPE/TPS blends, cellulose-containing LDPE/TPS composites showed substantially less moisture absorption, and there was no discernible difference in the moisture absorption values of composites containing 2.5 and 5% cellulose across all cellulose types (sawdust, CNC and powder cellulose), these correspond to the contact angle results. surprisingly, adding cellulose greatly mitigated the negative effects of TPS concentration on moisture absorption. It's interesting to note that the addition of cellulose significantly lessened the detrimental effects of TPS concentration on moisture absorption. This phenomenon might be the result of strong interactions between the TPS phase, which is responsible for the high water uptake, and cellulose. Strong chemical interactions between TPS's hydroxyl groups and cellulose may create a compact structure and stabilize the TPS phase, which would decrease the flow of water molecules into the composite. This would be consistent with [1, 26]. Increasing the contact angle helps to prevent the penetration of water and moisture, reduces the adhesion of contaminants to the surface of the packaging material, and reduces water absorption. Therefore, using compounds of hydrophilic and hydrophobic materials in calculated proportions may achieve the required balance between water resistance and mechanical specifications

4.1.6 Surface morphology

Figure 11 presents the SEM analysis of LDPE composites containing cellulose and TPS, highlighting changes in surface morphology compared to pure LDPE. The incorporation of cellulose and TPS contributes to the formation of a heterogeneous structure with increased surface roughness and irregularities.

Variations in surface roughness, particle distribution, and interfacial interactions between TPS and LDPE can be observed through SEM imaging. Figure 11(A) demonstrates that the SEM images reveal a more heterogeneous structure due to the dispersed TPS particles, in contrast to the relatively homogeneous surface of the pure LDPE matrix. The modification of LDPE with LDPE-g-MA enhances interfacial adhesion between LDPE and TPS, promoting a more uniform distribution and potentially reducing voids or defects.

Figures 11(B)-(D) illustrate the dispersion of CNC, powdered cellulose, and sawdust throughout the LDPE/TPS matrix, with no significant agglomeration observed. This uniform distribution contributes to enhanced mechanical and thermal properties, as well as improved reinforcing effects.

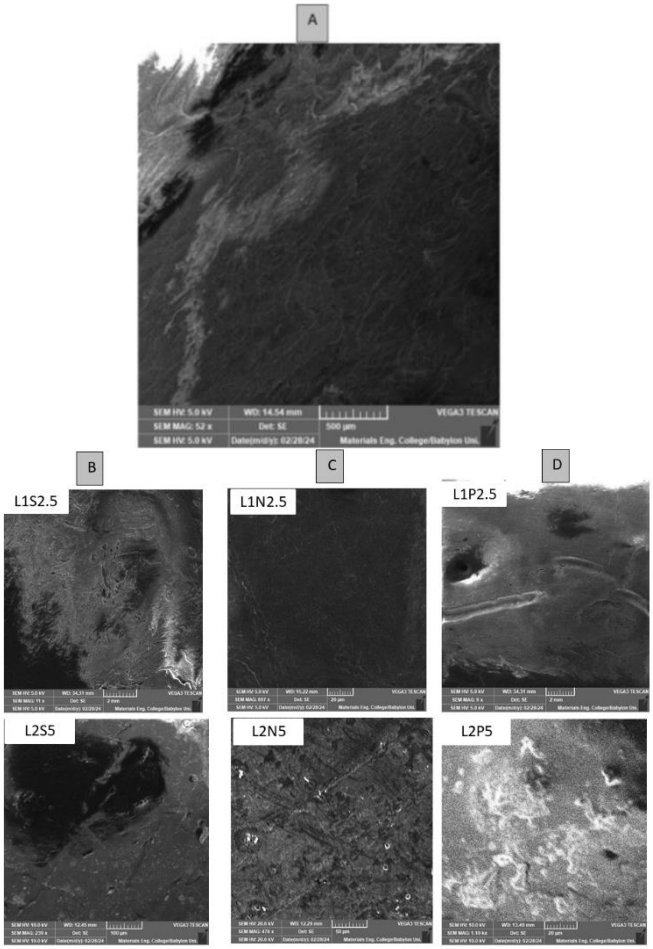

Figure 11. The SEM images of (A) LDPE/TPS (B) LDPE/TPS/Sawdust (C) LDPE/TPS/CNC (D) LDPE/TPS/Powder

Among the tested composites, those containing 5% sawdust, CNC, and powdered cellulose exhibit the most uniform microstructure, attributed to the appropriate material proportions and effective mixing methods. This observation aligns with the mechanical and physical results, indicating that a 5% weight ratio is optimal for achieving uniform dispersion under the applied material boundary conditions in this study. The use of a twin-screw extruder and the premixing process of LDPE/TPS with cellulose additives further supports the effectiveness of this ratio.

4.2 Numerical result correlation

The elastic modulus of polymer composites such as LDPE/TPS (low-density polyethylene/thermoplastic starch) blends with different reinforcements (crystalline nanocellulose, powdered cellulose, and sawdust) is affected by the concentration and nature of the fillers. Both experimental and numerical studies provide insights into these effects, and a correlation between them can help in predicting and optimizing material properties.

Sawdust can increase the modulus to a certain limit compared to CNC. The irregular shape and heterogeneous nature of sawdust particles can lead to low stress transfer. Increasing sawdust content increases the modulus up to a specific value, beyond which the composite modulus may decrease. Crystalline nano cellulose (CNC) typically increases the elastic modulus significantly due to its high stiffness and strong interfacial interaction with the polymer matrix. As, the modulus increases with the CNC concentration increases due to better load transfer and reinforcement. Powdered cellulose (PC) also improves the modulus, but the extent may be lower than that of CNC due to lower stiffness and potentially less interfacial adhesion. Higher concentrations of PC increase the modulus, although the improvement rate may decrease at higher loadings due to agglomeration or low dispersion.

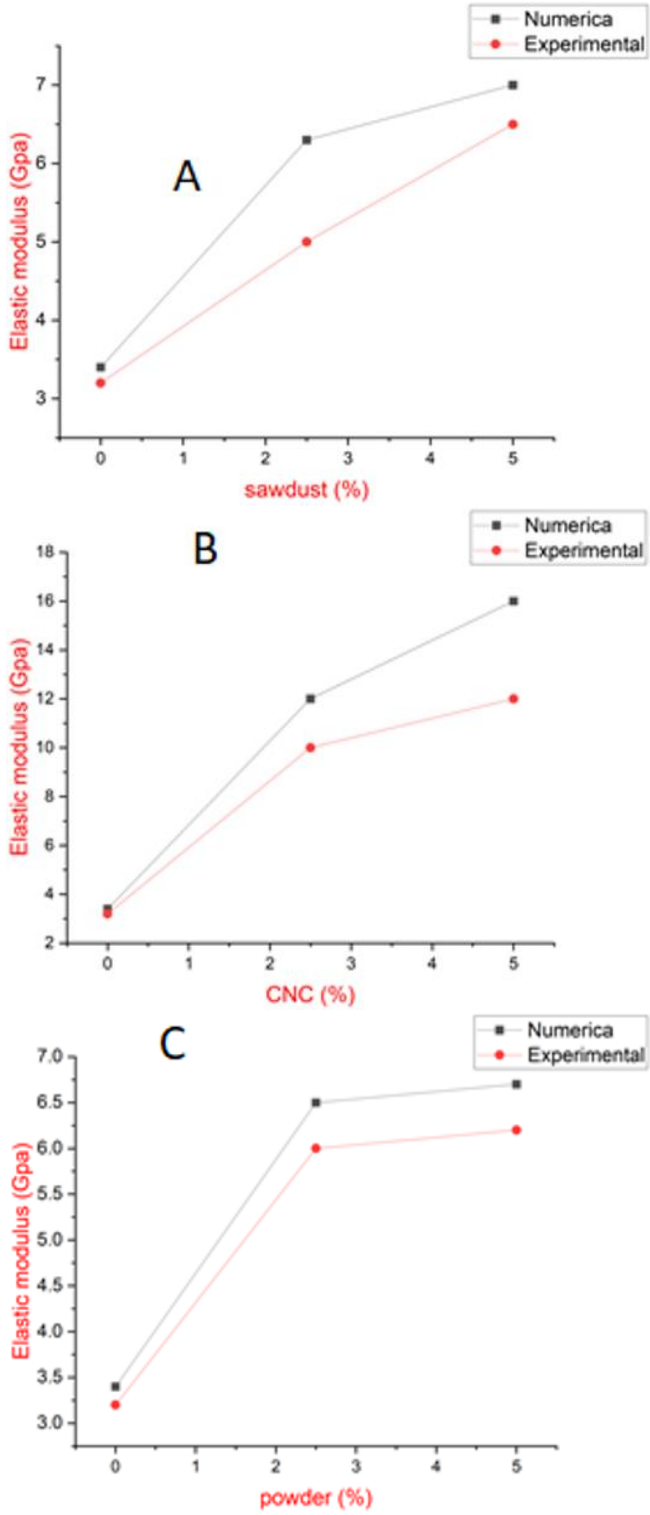

Figure 12. Experimental and numerical elastic modulus of (A) LDPE/TPS/Sawdust (B) LDPE/TPS/CNC (C) LDPE/TPS/Powder

Numerical simulations using ANSYS software based on finite element analysis can predict the elastic modulus of composites by considering material properties, cellulose

content, and distribution. The correlation between numerical and experimental results depends on the accuracy of input data, model assumptions, and boundary conditions.

LDPE/TPS/Sawdust (Figure 12(A)):

The models must account for the irregular shape and distribution of sawdust particles. Discrete element methods or the inclusion of heterogeneous particles in Representative Volume Elements (RVEs) may be used. However, due to the variability in sawdust properties, the correlation between numerical and experimental results may be less precise. Nevertheless, numerical models should predict an increase in modulus with sawdust content, which aligns with the findings in reference [27].

LDPE/TPS with CNC (Figure 12(B)):

RVEs reinforced with CNC particles demonstrate a good correlation between numerical and experimental results when the CNC distribution, orientation, and interfacial properties are accurately represented. Numerical models should predict a significant increase in modulus as CNC content increases.

LDPE/TPS with Powdered Cellulose (Figure 12(C)):

Numerical predictions indicate an increasing trend in modulus with powdered cellulose (PC) content, though the absolute values may be lower compared to CNC-reinforced composites. The accuracy of these predictions depends on the proper representation of dispersion and interfacial bonding.

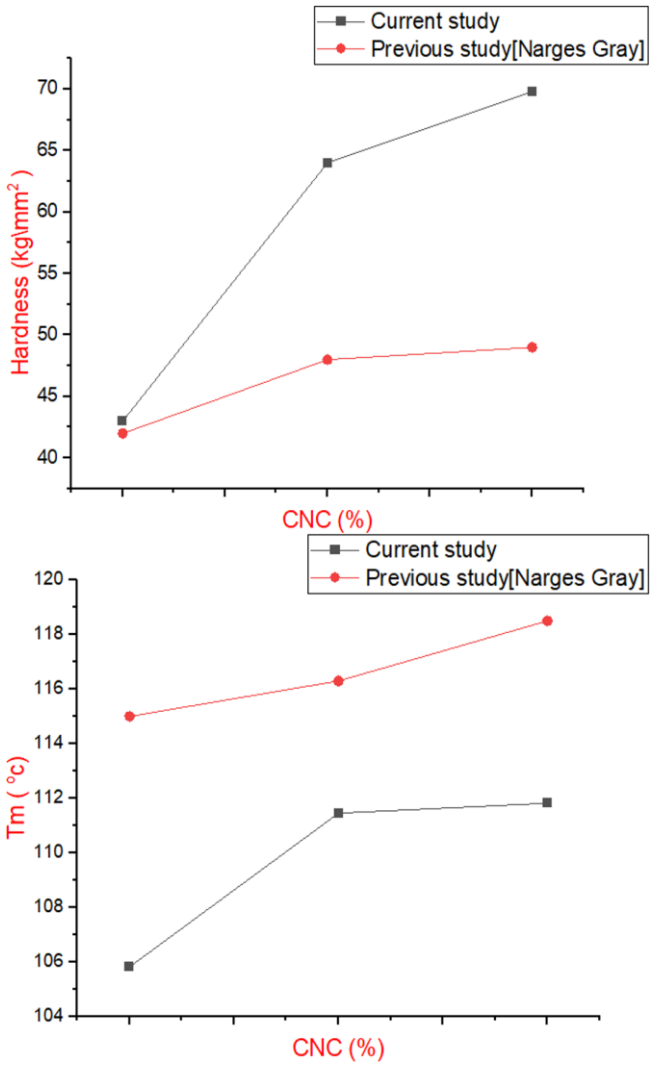

Figure 13. Validation between current study and previous study of Gray et al. [4], regarding water absorption, hardness and melting temperature behavior of LDPE/CNC

The numerical model shows a close match with experimental results at 0% and 2.5% concentration, while some divergence occurs at 5%. Validation of assumptions regarding CNC dispersion and interaction was found to be acceptable up to 2.5%.

4.3 Validation

The results of the current study confirm that the addition of cellulose to LDPE/TPS enhances the hardness, water absorption, and melting point of the LDPE/PE-g-MA/CNC composite. These findings align closely with those of Narges Gray et al. [4], as illustrated in Figure 13.

Figure 13 shows that the previous study reported higher water absorbency and lower hardness compared to the present study. This difference is primarily attributed to the starch content, which was 30% in the previous study compared to 13% in the current study. The higher starch content in the previous research led to a reduction in mechanical properties and an increase in water absorbency.

The inverse relationship between hardness and water absorption observed in both studies further supports the conclusions of this research, reinforcing the impact of cellulose content on composite performance.

5. CONCLUSION

Interest in the packaging industry has recently become one of the main areas of research because of its relationship to food, human health and the environment. Therefore, it was necessary to reduce reliance on plastic materials that are harmful to human health and the environment by using sustainable natural materials such as polysaccharide. In this work, the amount of LDPE used for food packaging was reduced by adding about 20% of starch and cellulose, while at the same time maintaining the mechanical, thermal, and physical specifications required in the process. Packaging. Adding starch initially reduced the mechanical, thermal, and physical specifications, but it reduced the amount of plastic and its impact on the environment. After that, when adding different types of cellulose, the mechanical, thermal, and physical specifications improved and became more compatible with the packaging specifications, and their expected ability to resist bacteria increased. Adding 2.5 and 5%. of sawdust, CNC and powder cellulose to the LDPE/TPS mixture, it achieved varying percentages of increased tensile strength and hardness, improved crystallization ratio and contact angle, and reduced water absorption. The degree of uniformity of cellulose distribution appeared in the electronic scanning examination, and LDPE-g-MA played an important role in the mixing process and balancing the specifications. In this work, the compatibility between the practical and numerical studies was also studied regarding the increase of elastic Modulus with the percentages of cellulosic materials. The role of the numerical study was also proven to reach the best percentage of additives with less time and effort. Also, compatibility between the current research and previous research has been achieved to some extent. There was agreement regarding water absorption and hardness with the previous study [4], with a difference in values because they added 30% of TPS, while the addition in this work was 13%, which explains the increased of water absorption and lower hardness compared to our results.

Recommendations:

1- Studying the effect of different concentration sizes,

shapes, and distribution of cellulose additives on the LDPE properties.

2- Studying the effect of different mixing speed and temperature on the films production for packaging applications.

3- Improving the LDPE/Cellulose composites to ward using for packaging of pharmaceutical materials. 4-Studying the effect of other organic polymers like lignin on the LDPE, PP, HDPE, and PS composites for packaging applications.

REFERENCES

[1] Kaboorani, A., Gray, N., Hamzeh, Y., Abdulkhani, A., Shirmohammadli, Y. (2021). Tailoring the low-density polyethylene-thermoplastic starch composites using cellulose nanocrystals and compatibilizer. Polymer Testing, 93: 107007. https://doi.org/10.1016/j.polymertesting.2020.107007

[2] Radfar, R., Hosseini, H., Farhoodia, M., Średnicka-Toberd, D., Ghasemie, I., Fathif, M., Khaneghahg, A.M. (2020). Factors affecting mechanical properties of low-density polyethylene (LDPE)/starch blends: A review. Carpathian Journal of Food Science and Technology, 12(4): 6-26. https://doi.org/10.34302/crpjfst/2020.12.5.1

[3] Norizan, M.N., Shazleen, S.S., Alias, A.H., Sabaruddin, F.A., et al. (2022). Nanocellulose-based nanocomposites for sustainable applications: A review. Nanomaterials, 12(19): 3483. https://doi.org/10.3390/nano12193483

[4] Gray, N., Hamzeh, Y., Kaboorani, A., Abdulkhani, A. (2018). Influence of cellulose nanocrystal on strength and properties of low density polyethylene and thermoplastic starch composites. Industrial Crops and Products, 115: 298-305. https://doi.org/10.1016/j.indcrop.2018.02.017

[5] Zhong, Y., Godwin, P., Jin, Y., Xiao, H. (2020). Biodegradable polymers and green-based antimicrobial packaging materials: A mini-review. Advanced Industrial and Engineering Polymer Research, 3(1): 27-35. https://doi.org/10.1016/j.aiepr.2019.11.002

[6] Scholten, B.V., Özen, M.B., Söyler, Z., Thomassin, J.M., Wilhelm, M., Detrembleur, C., Meier, M.A.R. (2020). Rheological and mechanical properties of cellulose/LDPE composites using sustainable and fully renewable compatibilisers. Journal of Applied Polymer Science, 137(22): 48744. https://doi.org/10.1002/app.48744

[7] Oner, B., Gokkurt, T., Aytac, A. (2019). Studies on compatibilization of recycled polyethylene/thermoplastic starch blends by using different compatibilizer. Open Chemistry, 17(1): 557-563. https://doi.org/10.1515/chem-2019-0064

[8] Yasim-Anuar, T.A.T., Ariffin, H., Norrrahim, M.N.F., Hassan, M.A., Andou, Y., Tsukegi, T., Nishida, H. (2020). Well-dispersed cellulose nanofiber in low density polyethylene nanocomposite by liquid-assisted extrusion. Polymers, 12(4): 927. https://doi.org/10.3390/polym12040927

[9] Arman Alim, A.A., Baharum, A., Mohammad Shirajuddin, S.S., Anuar, F.H. (2023). Blending of low-density polyethylene and poly (butylene succinate) (LDPE/PBS) with polyethylene–graft–maleic anhydride (PE–g–MA) as a compatibilizer on the phase morphology, mechanical and thermal properties. Polymers, 15(2): 261. https://doi.org/10.3390/polym15020261

[10] Shazleen, S.S., Foong Ng, L.Y., Ibrahim, N.A., Hassan, M.A., Ariffin, H. (2021). Combined effects of cellulose nanofiber nucleation and maleated polylactic acid compatibilization on the crystallization kinetic and mechanical properties of polylactic acid nanocomposite. Polymers, 13(19): 3226. https://doi.org/10.3390/polym13193226

[11] Verma, J., Petru, M., Goel, S. (2024). Cellulose based materials to accelerate the transition towards sustainability. Industrial Crops and Products, 210: 118078. https://doi.org/10.1016/j.indcrop.2024.118078

[12] Qasim, U., Osman, A.I., Al-Muhtaseb, A.A.H., Farrell, C., et al. (2021). Renewable cellulosic nanocomposites for food packaging to avoid fossil fuel plastic pollution: A review. Environmental Chemistry Letters, 19: 613-641. https://doi.org/10.1007/s10311-020-01090-x

[13] Trache, D., Hussin, M.H., Haafiz, M.M., Thakur, V.K. (2017). Recent progress in cellulose nanocrystals: Sources and production. Nanoscale, 9(5): 1763-1786. https://doi.org/10.1039/C6NR09494E

[14] Ramesh, M., Rajeshkumar, L., Sasikala, G., Balaji, D., Saravanakumar, A., Bhuvaneswari, V., Bhoopathi, R. (2022). A critical review on wood-based polymer composites: Processing, properties, and prospects. Polymers, 14(3): 589. https://doi.org/10.3390/polym14030589

[15] Kuforiji, C., Durowaye, S., Kassim, K., Lawal, G. (2023). Influence of sawdust particles reinforcement on physical and mechanical properties of High-Density Polyethylene (HDPE) matrix composites. Kathmandu University Journal of Science, Engineering and Technology, 17(1): 62382. https://doi.org/10.3126/kuset.v17i1.62382

[16] Xu, Y., Wu, Z., Li, A., Chen, N., Rao, J., Zeng, Q. (2024). Nanocellulose composite films in food packaging materials: A review. Polymers, 16(3): 423. https://doi.org/10.3390/polym16030423

[17] Najim, A.S., Hadi, N.J., Mohamed, D.J. (2014). Study the effect of $CaCO_3$ nanoparticles on the mechanical properties of virgin and waste polypropylene. Advanced Materials Research, 1016: 23-33. https://doi.org/10.4028/www.scientific.net/AMR.1016.23

[18] Gaska, K., Xu, X., Gubanski, S., Kádár, R. (2017). Electrical, mechanical, and thermal properties of LDPE graphene nanoplatelets composites produced by means of melt extrusion process. Polymers, 9(1): 11. https://doi.org/10.3390/polym9010011

[19] Jabir, R.K., Hadi, N.J., AL-Zubiedy, A.A.A. (2020). Rheological and mechanical investigation of PET, LDPE and HDPE wastes blended for re-use in bio-pipes. IOP Conference Series: Materials Science and Engineering, 987(1): 012006. https://doi.org/10.1088/1757-899X/987/1/012006

[20] Nwapa, C., Okunwaye, O.J., Okonkwo, C.L., Chimezie, O.W. (2020). Mechanical properties of high density polyethylene and linear low density polyethylene blend. SSRG International Journal of Polymer and Textile Engineering, 7: 22-28. https://doi.org/10.14445/23942592/IJPTE-V7I1P103

[21] Ramli, N. (2014). A study on low density polyethylene (LDPE) sawdust composites. Universiti Teknologi PETRONAS.

[22] Latef, Z.B., Hadi, N.J., Saad, N.A. (2020). The rheological properties of LDPE nanocomposites per a

capillary rheometer: an experimental and numerical study. IOP Conference Series: Materials Science and Engineering, 987(1): 012007. https://doi.org/10.1088/1757-899X/987/1/012007

[23] Müller, K., Bugnicourt, E., Latorre, M., Jorda, M., et al. (2017). Review on the processing and properties of polymer nanocomposites and nanocoatings and their applications in the packaging, automotive and solar energy fields. Nanomaterials, 7(4): 74. https://doi.org/10.3390/nano7040074

[24] Palencia, M. (2017). Surface free energy of solids by contact angle measurements. Journal of Science with Technological Applications, 2: 84-93. https://doi.org/10.34294/j.jsta.17.2.17

[25] Lomelí-Ramírez, M.G., Valdez-Fausto, E.M., Rentería-Urquiza, M., Jiménez-Amezcua, R.M., Hernández, J.A., Torres-Rendon, J.G., Enriquez, S.G. (2018). Study of green nanocomposites based on corn starch and cellulose nanofibrils from Agave tequilana Weber. Carbohydrate Polymers, 201: 9-19. https://doi.org/10.1016/j.carbpol.2018.08.045

[26] Zaman, H., Beg, M.D.H. (2021). Study on binary low-density polyethylene (LDPE)/thermoplastic sago starch (TPS) blend composites. Progress in Applied science and Technology, 11(1): 53-65. https://doi.org/10.14456/past.2021.5

[27] Saigal, A., Pochanard, P. (2019). The application of a representative volume element (RVE) model for the prediction of rice husk particulate-filled polymer composite properties. Materials Sciences and Applications, 10(1): 78. https://doi.org/10.4236/msa.2019.101008