Research Article

Preparation of LLDPE-PP- Coconut Shells Powder Composite Materials and Characterization of Their Mechanical Properties

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Abstract

Composite materials are characterized by a number of physical and mechanical properties that make them without rival in some applications requiring low weight and suitable strength. The interest in natural fibers reinforced composite materials is significantly growing due to their contribution in reducing long range environmental side- effects, minimizing the consumption of industrial raw materials and their low cost. The present work covered the preparation and study of PP- LLDPE- Coconut shell powder composite materials, the determination of the optimal preparation conditions and the characterization of their physical and mechanical properties.

The results of this work showed that the optimal conditions for the preparation of these composite materials were mixing at 170 °C and 50 rpm rotation speed and pressing at 170 °C and 20 tons pressure. The results confirmed also that coconut shell powder content can be increased up to 50 wt. % while preserving a suitable range of mechanical properties where tensile strength varied between 10- 30 MPa. This confirm the possibility to vary these composite constituents according to the requirement of the envisaged application without using compatibilizers and interfacial promoting bonding agents (silane compounds) between coconut shell powder and PP- LLDPE polymer blends.

Keywords: LLDPE; PP; Powder coconut husks; Mechanical properties; Composite materials

Abbreviations: PP: Polypropylene; HDPE: High Density Poly Ethylene; LDPE: Low Density Poly Ethylene

Introduction

Composite materials have evolved to meet the needs of modern technologies, especially aeronautics and aerospace requirements for light weight, excellent mechanical properties, fatigue resistance and durability. Composite materials consist of high-resistance and stiffness granular or fibrous reinforcement providing load bearing capability and polymeric, metallic or ceramic matrix ensuring matrix- fibers stress transfers and reinforcement protection against harmful environment. Reinforcements comprise glass, carbon and Kevlar fibers. Matrix materials include, mainly polymers, light metals and alloys and some inorganic materials. One of the most important advantages of composite materials is the design flexibility, so that the reinforcing fibers are placed in high load directions, complying with design requirements and leading to final product that offers mechanical resistance and light weight [1].

With the remarkable development of the production and use of composite materials, the issue of their waste becomes of prime importance. While metallic materials are prone to recycling and reforming, thermosetting polymer composite materials are mostly non-recyclable [2]. This issue is due to the resistance of their components, especially polymers, to ageing and biodegradation. Within this context, and in order to reduce costs, research and development efforts in recent decades have focused on trying to use thermoplastic recyclable polymers together with biodegradable and low-cost natural fibers. The issue of recycling and reuse became of utmost importance these days in order to preserve natural resources suffering from continuous depletion due to excessive use, to protect the environment and to reduce the environmental damage resulting from landfill and burning.

In 2003, Richardson and colleagues used hemp (cannabis sativa) fibers to strengthen phenolic resin used in applications requiring fire resistance. They found that using two layers of hemp fibers to produce composite panels, their flexural strength, flexural modulus and the impact resistance improved significantly. Hemp fibers contributed to reduce phenolic resin brittleness [3]. In 2010, Matuanal M. and colleagues studied the mechanical properties of wood powder -polymer mixtures of different proportions. Results were compared with wood mixtures with Polypropylene (PP), High Density Poly Ethylene (HDPE). It was noted that tensile strength decreased by increasing the ratio of wood. Compared to wood mixtures with PP and HDPE, Poly-butene was found to have a lower tensile strength than other mixtures, while it showed greater elongation and impact resistance and transport [4].

In 2013, S. E. Salih and colleagues studied the mechanical and structural properties of HDPE-PP and linear low density polyethylene (LDPE) -PP mixtures. The results showed that HDPE-PP showed a stronger tensile strength, Young and deformation parameters at fracture and greater durability than LDPE-PP, and that the mixture of 80% HDPE-20 PP% had the highest mechanical properties [5]. In 2013, N. Sarifuddin and colleagues studied the mechanical and structural properties of HDPE mixtures with different percentages of hemp fibers. The results showed that the mixing ratio of polyethylene with 40% weight of cannabis fibers achieved relatively high mechanical properties [6]. In 2011, S.A. Hussain and his colleagues studied the mechanical properties of composites of long green coconut fibers and HDPE polymer. The results showed that increasing the fiber ratio up to 40% by weight increased the tensile strength which gradually decreases after this ratio [7].

This shows the recent trend of research towards complex materials of bio-biodegradable or post-life recyclable material, called green composite materials, in reference to the nature to be conserved [8]. This work deals with the preparation of a comprehensive range of PP and LLDPE polymer blends, composite materials from these blends and various ratios of coconut shell powder and characterization of their mechanical properties in order to obtain composite materials reinforced by environmentally friendly fibers, reduce the proportion of polymeric materials and increase the proportion of natural reinforcement materials to the larger extent possible, while maintaining an appropriate level of mechanical properties without the use of compatibilizers and surface bonding agents (silane compounds) between coconut shell powder and PP-LLDPE blends.

Materials and Methods

Materials

Linear low density polyethylene (LLDPE) and polypropylene (PP): LLDPE and PP polymers were supplied by Sabic- Saudi Arabia. Table (1) gives the characteristics of these polymers according to the supplier data [9,10].

Polymer	Melting point (°C)	Mass Flow Index MFI	Density (g/cm³)	Yield stress (MPa)	Fracture stress (MPa)
LLDPE	125	20	0.925	12	8.5
PP	165	16	0.905	27	

Table 1: Characteristics of LLDPE and PP polymers according to the supplier data.

Coconut shells powder: Coconut shells were washed and cleaned with water and dried at 95 °C in an oven for 24 hours. After that, they were grinded over a 10 min time by a Retch ball mill. To remove the impurities and large

grains, the powder was screened on a 63 micron sieve and the passing powder from this sieve was used to prepare the composite materials.

Equipment

Equipment and tools used comprised Metal Spatula, Plastic containers, Sartorius electronic balance- accuracy 0.001 g, Retsch Ball Mill, Retsch screens set, Brabender internal mixer, Tensile specimens cutting machine (Ceast), operating under 4 bar air pressure, Thermal press (Ceast), consisting of two bottom and upper platens with thermal sensor and temperature reader, maximum load 20 tons, maximum temperature 300 °C with water cooling circuit, general mechanical testing machine DY34 Adamel Lhomargy (10KN load).

Methods

Preparation of samples: Raw materials comprising PP, LLDPE and coconut shell powder were weighed prior to the start of the mixing process on a sensitive balance. The total sample weight was around 45 g. After weighing, the batch was added progressively to the mixer chamber at 50 rpm and 170 °C in order to melt the polymer blend and

reduce its viscosity. The PP top-melting point of 165 $^{\circ}$ C was first added to give it longer mixing time for melting and homogenization in the mixer, then, the LLDPE with a melting point of 125 $^{\circ}$ C was add.

Depending on its composition, the polymer batch was left under mixing for 2-3 minutes, then, coconut shell powder was progressively added in the batch to ensure homogenization and avoid its aggregation. Batch mixing continues for 7 minutes to achieve a total mixing time of 10 minutes. The curves of the torque and temperature variations with time are continuously recorded and displayed on the computer monitor to ensure that the batch under mixing attains the stability state and to maintain the mixing conditions. By this way, all polymers and composite batches were prepared.

PP-LLDPE blends

Table 2 gives the composition of LLDPE-PP blends.

	PEOP	PE10P	PE20P	PE30P	PE40P	PE50P	PE60P	PE70P	PE80P	PE90P	PE100P
PP wt. %	0	10	20	30	40	50	60	70	80	90	100

Table 2: Weight composition for PP-LLDPE blends.

Compositions of PP-LLDPE-coconut shell powder composite material: Coconut shell powder was added to each of the PP-LLDPE blends by 5%, 10%, 20%, 30%, 40% and 50% by weight. Composite materials were designated as PE%PXC, where P% refers to the weight proportion of polypropylene and XC to the weight proportion of coconut shells powder in the composite material.

PP-LLDPE blends and composite plate forming: The polymer blends or the composite batches were placed in a preheated metal mold coated with a thin film of antiadhesion silicone oil. The pressure was applied slightly and gradually to allow batch softening and mold filling. The pressure was then increased up to 20 tons and stabilized for 10 minutes at a temperature of 170°C. After that, the mold was cooled up to room temperature while keeping the pressure to maintain the product dimensions.

Determination of the of polymer blends specific weight: The specific weight was measured according to Archimedes method, where the sample is weighed in air (m_0) and then in water (m_1) . The specific weight is given by the following relation:

$$\rho = \frac{m_0.\rho_0}{m_0 - m_1}$$

Where, ρ_0 represents the specific weight of water at its temperature during measurement.

The specific weight of the PP-LLDPE blends was also calculated using the following rule of mixtures [11]:

$$\rho = \rho_1. \mathcal{V}_1 + \rho_2. \mathcal{V}_2$$

Where, ρ_1 and ρ_2 refer to the densities of PP and LLDPE respectively, and \mathcal{V}_1 and \mathcal{V}_2 the volume fraction of PP and LLDPE in the blend respectively.

Forming of tensile specimens: For tensile testing, specimens were cut from plates according to specific dimensions using a Ceast cut machine under 4 bar air pressure. Table (3) gives the dimensions of tensile specimens [12].

Tensile specimen dimensions (mm)					
Specimen length	75				
Specimen ends width	12.5 ± 1				
Length of the specimen active part	25 ± 1				
Width of the specimen active part	4 ± 0.1				
Outer radius	8 ± 0.5				
Internal radius	12.5 ± 1				
Thickness	2 ± 0.1				

Table 3: Dimensions of tensile specimens.

Tensile testing procedure: The width and thickness of the test specimen is measured before the test and entered into the computer with the test parameters, especially the specimen length of the active part (specimen initial

length), 25 mm; surface area of the specimen section in the active part; tensile deformation speed of 5 mm / min. The tensile test provides the tensile curve of the stress σ in terms of the strain ε , σ = f (ε). This tensile curve allows for the determination of the following mechanical properties [12]:

• Stress at fracture σ (MPa):

$$\sigma = F/S$$

Where, F: the tensile force at fracture (N) and S: the surface area of the specimen section (m^2) .

• Strain at fracture ε (%):

$$\varepsilon = \frac{\Delta L}{Lo} \times 100$$

Where, ΔL : elongation (mm) at fracture and L₀: the initial length of the specimen (mm).

• Young modulus E(MPa):

$$E = \frac{\Delta \sigma}{\Delta \varepsilon}$$

Where, $\Delta \sigma$: the stress difference between two points of the linear portion of the stress- strain curve and $\Delta \epsilon$: the strain difference between the same two points.

Results and Discussion

Behavior of the PP- LLDPE - coconut shell powder composite materials during mixing process

Figure (1) shows the torque curve of the composite material PE60P30C of PP, LLDPE and 30% wt. coconut shell powder.



Figure 1 shows the torque increase at the beginning of the addition of coconut shells powder from 20 Nm to reach the maximum value of 32 Nm. This indicates the increase of the composite material viscosity. When the coconut powder is added and the thermal homogeneity is attained, the viscosity and the torque decrease until a stability state is reached after three or four minutes of the mixing process depending on coconut powder content.

Figures. (2-a), (2-b), (2-c) show the change in the values of the maximum torque in terms of coconut powder fraction of the composite materials.



Figure 2-a: Maximum torque in terms of coconut powder fraction.





It is noted from Figure (2-a; 2-b; 2-c) that coconut shells powder clearly contributes to increasing the viscosity of the PP- LLDPE blends and, as a result, to increasing the torque required to be applied to the mixing screw in the mixer chamber. It is also noted that the torque increases steadily as the proportion of polypropylene increases in the PP- LLDPE blends. In Figure (2-a), there are obvious disturbances in the direction of the trend of maximum torque values change, but they disappear with the increase of polypropylene content as shown in Figures (2b) and (2-c). This is consistent with the study of LDPE-Kenaf core fibers- thermoplastic sago starch composites, which showed increased torque due to an increase in the proportion of Kenaf core fibers [6].

Results of the specific weight of the PP- LLDPE bends

Results of measurement of the specific weight of PP-LLDPE bends:

Table 3 gives the specific weight values for PP- LLDPE polymer blends.

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Polymer blends	Measured specific weight (g.cm ⁻³)
LLDPE	0.923
PP	0.905
PE10P	0.919
PE20P	0.918
PE30P	0.911
PE40P	0.908
PE50P	0.911
PE60P	0.907
PE70P	0.906
PE80P	0.907
PE90P	0.906

Table 3: Specific weight values for PP- LLDPE polymer blends.

Figure (3) shows the change in the specific weight in terms of polypropylene proportion in the PP-LLDPE blends.



Figure 3: Specific weight in terms of percentage of polypropylene in PP-LLDPE blends.

Figure 3 shows that the specific weight decreases with the increase of polypropylene in the blends. LLDPE being more crystalline than polypropylene due to chains uniformity, LLDPE specific weight is higher than that of polypropylene.

Results of calculation of the specific weight of LLDPE-PP bends: Table (4) presents the results of the calculation of the specific weight of PP- LLDPE polymer blends.

PP % wt.	0	10	20	30	40	50	60	70	80	90	100
Specific weight (g/cm ³)	0.925	0.923	0.921	0.919	0.917	0.914	0.912	0.910	0.908	0.906	0.905

ΕV

Table 4: Calculated specific weight of PP- LLDPE polymer blends based on polymers supplier data.

Figure 4 shows the comparison between the calculated and measured specific weight of PP- LLDPE blends in terms of PP content.





Figure 4 shows that the calculated and measured specific weight of PP- LLDPE blends decrease with PP ratio and that the measured specific weight values are lower than the calculated specific weight. This could be related to the possibility of formation of micro-porosity in the blends leading to a reduction in the specific weight. The porosity in the blends PE40P, with the largest difference between calculated and measured specific weight, is estimated at 0.98%. Neglecting specific weight measurement error, this indicates limited micro-porosity and good PP- LLDPE blends.

Results of mechanical characterization.

Results of the mechanical characterization of PP-LLDPE bends: Table 5 summarizes the mechanical properties of the PP-LLDPE polymer blends, as well as the mechanical properties of each polymer.

Sample	Young modulus (MPa)	Tensile strength (MPa)	Strain at break (%)
LLDPE	123(34)	10.45(1.21)	33.11(4.04)
РР	512(58)	32.22(4.31)	13.00(2.41)
PE10P	203(32)	14.98(1.64)	13.18(3.01)
PE20P	284(41)	17.20(2.02)	10.86(1.62)
PE30P	279(45)	11.60(1.48)	6.35(0.92)
PE40P	355(52)	14.58(2.32)	5.70(1.01)
PE50P	371(44)	15.31(3.12)	5.64(0.92)
PE60P	482(58)	19.87(3.31)	6.69(0.87)
PE70P	491(62)	24.44(4.11)	7.41(0.79)
PE80P	556(71)	28.13(3.52)	9.97(0.99)
PE90P	584(82)	29.60(3.83)	11.91(1.52)

Table 5: Mechanical properties of the PP-LLDPE blends. () : standard deviation.

Figure (5) shows the strain at break variation of PP-LLDPE in terms of PP ratio.



Figure 5 shows that strain at break of the PP-LLDPE blends decreases when PP is added to pure LLDPE up to 30%, where PP is distributed in LLDPE. In the range of 40%- 60% PP, where both polymers ratios are roughly equal, strain at break remains almost constant and then increases slightly by 70% PP when LLDPE is distributed in PP.

Figure 6 shows the tensile strength variation of PP-LLDPE blends in terms of PP ratio.

Figure 6 shows that tensile strength increases as the proportion of polypropylene increases in the blends. This is because polypropylene is more resistant than linear low density polyethylene. Polypropylene strengthens polyethylene up to 20%, and at 30%, polyethylene begins to weaken polypropylene resistance, and with the increase of polypropylene proportion, tensile strength of

PP-LLDPE blends approaches gradually the rule of mixture behavior (straight line- figure 6).



Figure (7) shows the variation of Young modulus for PP-LLDPE blends in terms of PP ratio.

Figure (7) shows that the Young modulus of PP-LLDPE blends increases steadily with the increase of PP in the blends up to 90% of PP. The measured values of PP-LLDPE blends Young modulus are greater than the calculated values by the rule of mixtures (the straight line in figure 7). It seems that mechanical behavior of PP-LLDPE blends exhibits three domains: Blends rich with LLDPE, intermediate blends and blends rich with PP.



In the blends rich with LLDPE, mechanical behavior of LLDPE prevails and PP improves PP- LLDPE blends tensile strength and reduces their plasticity. In the blends rich with PP, mechanical behavior of PP prevails and PP-LLDPE- blends mechanical properties approaches those of PP. In the intermediate blends, PP- LLDPE blends mechanical properties are lower than the rule of mixtures predictions by about 30 % in the blend PE30P. This behavior is related to the partial solubility of LLDPE and PP polymers and is consistent with the results of S. E. Salih et al. [5], J. Li et al. [13] and R. Strapasson et al. [14].

Results of mechanical characterization of PP-LLDPE-Coconut shell powder composite materials: Table (6) gives the mechanical properties of all PP-LLDPE- coconut composite materials with different percentages of PP and coconut shell powder.

The sample	MPa) (Young modulus	MPa) (Tensile strength	Strain at break (%)
PE10P	203(49)	14.97(1.62)	13.18(1.78)
PE10P5C	322(69)	15.19(2.79)	11.84(1.61)
PE10P10C	311(73)	15.613(2.11)	10.88(2.11)
PE10P20C	381(50)	17.04(3.21)	8.23(1.38)
PE10P30C	483(66)	14.79(1.86)	4.85(0.61)
PE10P40C	577(63)	15.24(2.07)	3.71(0.63)
PE10P50C			
PE20P	284(45)	17.20(2.71)	10.86(0.89)
PE20P5C	287(67)	12.82(1.66)	12.70(1.56)
PE20P10C	343(85)	13.59(1.68)	10.70(0.92)
PE20P20C	363(72)	13.89(2.77)	7.61(0.83)
PE20P30C	493(66)	13.85(1.73)	5.40(0.29)
PE20P40C	510(83)	15.11(1.86)	4.89(0.59)
PE20P50C	642(73)	13.11(1.94)	2.99(0.24)
PE30P	279(41)	11.60(1.5)	6.35(0.82)
PE30P5C	342(67)	14.54(2.63)	6.32(0.71)
PE30P10C	373(56)	14.34(1.02)	5.32(0.58)

PE30P20C	426(52)	14.62(2.33)	4.76(0.55)
PE30P30C	506(75)	15.00(1.86)	4.10(0.47)
PE30P40C	653(93)	14.10(1.69)	3.15(0.71)
PE30P50C	711(62)	9.25(1.35)	2.06(0.31)
PE40P	355(64)	14.58(1.32)	5.70(0.87)
PE40P5C	474(56)	17.54(2.77)	4.02(0.79)
PE40P10C	561(73)	17.66(2.06)	4.06(0.44)
PE40P20C	604(50)	15.63(2.88)	3.63(0.56)
PE40P30C	725(64)	18.01(3.31)	3.72(0.25)
PE40P40C	833(72)	13.57(1.93)	2.22(0.36)
PE40P50C	924(83)	15.57(0.96)	2.62(0.32)
PE50P	371(66)	15.31(0.88)	5.64(0.44)
PE50P5C	418(64)	11.87(0.52)	4.27(0.63)
PE50P10C	496(76)	14.42(0.73)	4.72(0.51)
PE50P20C	573(79)	15.05(1.98)	4.47(0.63)
PE50P30C	681(62)	14.16(2.59)	3.41(0.78)
PE50P40C	778(66)	15.88(1.69)	3.78(0.65)
PE50P50C	691(62)	10.65(2.74)	2.13(0.37)
PE60P	482(67)	19.86(4.9)	6.69(0.62)
PE60P5C	557(81)	18.81(2.82)	6.37(0.82)
PE60P10C	574(72)	19.39(3.34)	6.19(0.64)
PE60P20C	668(73)	18.31(2.41)	5.07(0.73)
PE60P30C	750(76)	16.13(2.84)	3.76(0.66)
PE60P40C	899(77)	15.28(1.67)	3.01(0.28)
PE60P50C	973(74)	12.87(2.74)	2.16(0.33)
PE70P	491(62)	24.44(3.75)	7.41(0.76)
PE70P5C	624(55)	26.75(2.18)	7.17(0.91)
PE70P10C	699(81)	27.17(2.31)	6.86(0.78)
PE70P20C	732(67)	24.53(3.23)	5.65(0.83)
PE70P30C	847(63)	22.65(2.4)	4.40(0.43)
PE70P40C	863(73)	19.00(1.92)	3.50(0.49)
PE70P50C	1036(81)	15.53(0.68)	2.28(0.42)
PE80P	556(60)	28.13(3.25)	9.96(1.92)
PE80P5C	728(54)	26.56(2.16)	6.33(0.71)
PE80P10C	772(76)	26.69(1.35)	5.93(0.31)
PE80P20C	829(72)	21.13(1.86)	4.25(0.28)
PE80P30C	997(85)	19.20(0.9)	3.49(0.23)
PE80P40C	1032(87)	16.56(1.79)	2.68(0.25)
PE80P50C	1135(69)	15.80(1.56)	2.33(0.36)
PE90P	584(61)	29.59(2.74)	11.91(1.3)
PE90P5C	728(71)	27.72(2.21)	7.74(0.33)
PE90P10C	831(77)	25.98(2.45)	4.71(0.43)
PE90P20C	847(83)	26.79(1.46)	5.33(0.33)
PE90P30C	913(69)	24.22(3.71)	4.36(0.38)
РЕ90Р40С	937(110)	22.57(1.53)	3.89(0.24)
PE90P50C	1012(82)	19.60(0.90)	2.46(0.27)

Table 6: Mechanical Properties of PP-LLDPE-Coconut shell powder composite Materials.

(): standard deviation.

Tensile strength of PP-LLDPE - Coconut shell powder composite material: Figure (8) shows the tensile strength variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 10%, 20%, 30% PP.



Figure 8: Tensile strength variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 10%, 20% , 30% PP.

Figure 8 shows that the tensile strength of PP-LLDPE -Coconut shell powder composite material at 10% polypropylene gradually increases up to 20% of coconut shell powder and then decreases slightly within 30-50%. This behavior is nearly the same for 20% and 30% polypropylene, as the tensile strength increases gradually and slightly (12- 15 MPa) up to 30% of coconut shell powder and then decreases sharply (9- 14 MPa) at 50% of coconut shell powder. At this proportion the polymer blend matrix ratio becomes not sufficient to bond the coconut shell powder particles and, thus, tensile strength falls down.

Figure (9) shows the tensile strength variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 40%, 50%, 60% PP.

Figure 9 shows that at 40% and 50% polypropylene, PP-LLDPE - Coconut shell powder composite materials retain their tensile strength, with a slight increase up to 30% of coconut shell powder, except PE50P5C, and a slight decrease in the range 40-50% (10.5-15 MPa). At 60% polypropylene, a gradual decrease in tensile strength is observed with the increase of coconut shell powder (20-13 MPa). This can be attributed to the increasing viscosity

of the polymer blends with the increase in polypropylene proportion. This makes it difficult to mix the coconut shell powder with the polymer blend.



Figure 9: Tensile strength variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 40%, 50% , 60% PP.

Figure 10 shows the tensile strength variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 70%, 80%, 90% PP.



Figure 10: Tensile strength variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 70%, 80% , 90% PP.

Figure (10) confirms what was previously observed in the composite material PP-LLDPE - coconut shell powder at 60% polypropylene, a gradual decrease in tensile strength with increasing coconut shell powder (16-30 MPa). All previous tensile strength results are consistent with the

tensile strength results of N. Sarifuddin et al. [6] and S.A.Hussain et al. [7] where tensile strength decreased with the increase in the proportion of hemp fibers and green coconuts fibers in the composite material with HDPE polymer.

Young modulus of PP-LLDPE-Coconut shell Powder composite materials: Figs. (11), (12), (13) show Young modulus variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder and PP proportions.



Figure 11: Young modulus variation of PP-LLDPE -Coconut shell powder composite materials in terms of coconut shell powder proportion at 10%, 20% , 30% PP.



Figure 12: Young modulus variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 40%, 50% , 60% PP.



Figure 13: Young modulus variation of PP-LLDPE -Coconut shell powder composite materials in terms of coconut shell powder proportion at 70%, 80% , 90% PP.

Figure (11), (12), (13) show that the Young modulus increases steadily with the increase in the proportions of polypropylene and coconut shell powder, starting at 200 MPa for 10% polypropylene and ending at 600 MPa for the proportion of 90% polypropylene. Young modulus is steadily increasing with the proportion of coconut shell powder up to more than 1100 MPa at 50% coconut shell powder and 80% polypropylene. These results are consistent with N. Sarifuddin et al. [6]. The highest value of Young modulus with Kenaf core fibers and HDPE reached 600 MPa at 40% weight of Kenaf core fibers, which was the highest percentage of fiber they achieved.

Strain at breakage of PP-LLDPE -Coconut shell powder composite materials: Figure (14), (15), (16) shows the strain at break variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder and PP proportions.



Figure 14: strain at break variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 10%, 20% , 30% PP.



Figure 15: strain at break variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 40%, 50% , 60% PP.



Figure 16: strain at break variation of PP-LLDPE - Coconut shell powder composite materials in terms of coconut shell powder proportion at 70%, 80% , 90% PP.

Figures (14-16) show that strain at break decreases with the increase in coconut shells powder ratio. This is due to the nature of the low-plasticity coconut shell powder and the increase of its proportion at the expense of the polymer blend matrix ratio.

Discussion of mechanical properties of PP-LLDPE-Coconut shell Powder composite materials: The overall analysis of tensile strength of 63 PP-LLDPE -Coconut shell powder composite materials shows that tensile strength of all composite materials is higher than that of LLDPE (10.45 MPa), except of PE30P50C material (9.25± 1.35). On the other hand, Coconut shell powder, up to a ratio of 20- 30 %, improves tensile strength of PP-LLDPE blends, or preserves it, except of PE20PXC, PE80PXC and PE90PXC materials which still exhibit tensile strength of 12.82 MPa, 15.80 MPa and 19.60 MPa respectively. It is of notice that mechanical behavior of PP-LLDPE - Coconut shell powder composite materials exhibits, in term of PP content, three domains: Composites rich with LLDPE, intermediate composites and composites rich with PP. In the composites rich with LLDPE, Coconut shell powder improves tensile strength of PP-LLDPE blends in synergy with PP effect. In the composites rich with PP, Coconut shell powder, up to 5-20 % wt., improves tensile strength of the composites PE70PXC and PE80PXC and decreases tensile strength of the composites PE90PXC (19.60-27.72 MPa).

Mechanical behavior of PP-LLDPE - Coconut shell powder composite materials is consistent with mechanical behavior of PP-LLDPE - wood powder composite materials which were studied by D. G. Dikobe et al. [15] with better tensile strength for the former.

Conclusion

The present study showed that the appropriate practical conditions for the preparation of PP-LLDPE - Coconut shell powder composite materials include blending at a temperature of 170 $^{\circ}$ C, 50 rpm rotation speed and thermal pressing at a temperature of 170 $^{\circ}$ C and a pressure of 20 tons. The results confirmed the possibility of increasing coconut shell powder proportion in PP-LLDPE blends up to 50% wt. allowing the preservation of an appropriate range of mechanical properties of the resulting composite materials.

The effect of coconut shell powder ratio on mechanical properties of PP-LLDPE- coconut shell powder composite materials (tensile strength, Young coefficient, strain at break) has been evaluated, the tensile strength of these composite materials ranged between 10-30 MPa in the wide range of variation of their constituents. This confirms the possibility of changing the constituent's proportions according to the requirements of the intended use of these composite materials.

This study showed that the cost of these composite materials could be reduced steadily with the increase in the proportion of coconut shell powder without the use of polymer blend compatibilizers, surface bonding agents (silane compounds) between coconut shell powder and PP-LLDPE blends and chemical treatment of coconut shell powder.

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