

# HDPE-Coconut Flour Composites: Effect of Coupling Agents and Surface Modification

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**Summary:** This research work studies the mechanical, thermal and morphological behavior of coconut flour/polyethylene composites, with special emphasis on the influence of the surface modification of coconut flour and the presence of different coupling agents on the interfacial bonding. The different treatments of the composites with an EAA copolymer, with 5 and 18 wt% of NaOH and acetylation, confirm the better tensile behavior of these composites.

**Keywords:** acetylation; coconut flour; composites; coupling agent; surface modification

## Introduction

Interest in the use of lignocellulosic fibers as a reinforcing element in polymer matrices, to replace fiberglass,<sup>[1–7]</sup> has been gaining interest in recent years, because they are intended to produce a range of new materials for applications in the field of engineering. Some of the lignocellulosic fibers in thermoplastic-containing composites can be used in diverse areas, such as furniture manufacture, packaging, construction and in the automotive industry.

According to Nechwatal et al.,<sup>[8]</sup> TITK Rudolstadt has developed natural fiber reinforced composites. Topics relevant for this area include: findings in the field of natural fiber improvement; test methods for physical-mechanical properties of fibers and composites; non-woven natural fiber and hybrids of non-woven fiber; manufacture and processes related to natural fiber composites (compression molding, extru-

sion, injection molding), and thermoplastic granules reinforced with long natural fiber.

Furthermore, Wielage et al.<sup>[9]</sup> conducted studies by means of dynamic and mechanic analysis (DMA) on polypropylene reinforced with linen and canvas, determining that fiber conditioning, manufacture process and processing parameters are significant factors that influence the mechanical properties of the end product. The results also show that elastic properties (rigidity, storage module) of the composite depend on the kind of coupling agent. Other influencing parameters are specific surface and the content of added fiber. The effect of some of these parameters on thermoplastic composites with sisal, wood and pineapple fiber has been studied by a number of researchers, including Kuruvilla et al.,<sup>[10]</sup> Manikandan et al.,<sup>[11]</sup> Albano et al.,<sup>[3,4]</sup> etc. However, there are only few studies on coconut fiber, which is a material found in large quantities in Venezuela. Brahmakumar et al.<sup>[12]</sup> studied low density polyethylene sandwich composites and coconut fibers, 140 mm in length and 200  $\mu$ m in diameter, reporting that fiber wax provides a proper fiber-matrix bonding, increasing tensile properties.

It is worth highlighting the boost given by Daimler-Benz together with UNICEF, through the program Poverty and Environment in Amazonia (developed in Brazil), to

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promote the use of bio-composites in the manufacture of their Class E cars and Class A trucks. All these developments are perfectly harmonized with the environment without detriment to the quality required for Mercedes-Benz products.<sup>[13]</sup>

Pradhan et al.<sup>[14]</sup> studied a powdered ultra high molecular weight polyethylene (UHMWPE) and coconut flour composite compacted at 200 °C. A stiffer composite was obtained when the filler content was of 20% v/v or higher.

Ramaraj<sup>[15]</sup> worked on a PVA composite with 10, 20, 33 and 50% w/w of coconut fiber, studying the influence of adding filler on the humidity content, the moisture vapor transmission rate, solubility, swelling and thermal transition of composites.

Since studies on an effective interfacial adhesion between coconut flour and polymers is relevant for the processing and ultimate performance of composites,<sup>[16]</sup> this work studies the mechanical, thermal and morphological behavior of coconut flour/high-density polyethylene composites, with special interest on the influence of the surface modification of coconut flour and the presence of different coupling agents on the interfacial bonding.

## Experimental Part

High density polyethylene (MFI = 7.3 dg/min,  $\rho = 0.96 \text{ g/cm}^3$ ) was used. Coconut flour with a particle size smaller than 50 mesh and a density of  $0.0975 \text{ g/cm}^3$  was used as a filler. An ethylene-acrylic acid random block copolymer (EAA) with 15 wt% acrylic acid content and a titanate coupling agent (Lica 12) were used as interfacial agents at 5 and 1 wt% with respect to the filler content, respectively. Coconut flour was previously treated with 5% NaOH. Acetylation (with NaOH and acetic anhydride) was carried out as per the procedure developed by Bello et al.<sup>[17]</sup> and treatments with 5 and 18 wt% NaOH according to Ramaraj's procedure<sup>[15]</sup> were used to modify the surface of the coconut flour.

HDPE composites with 5, 10, 15 and 20 wt% of coconut flour were obtained in a co-rotating twin screw extruder with a temperature profile of 200, 200, 200, 200, 210, 210, 210 and 210 °C at 110 rpm. The specimens for tensile testing were injection molded in a Reed Prentice 100TE machine.

Tensile tests were done at 50 mm/min as per ASTM D 638 standard in a Lloyd Instrument Universal Assay Machine.

Differential scanning calorimetric analysis was carried out in a Mettler-Toledo DSC 822°. Samples (9–10 mg) were heated up to 170 °C and subsequently kept for about 3 min in order to erase the previous thermal history. This initial heating was performed at a rate of 20 °C/min. Then, the samples were submitted to a cooling step to room temperature and subsequently heated up to 170 °C, both at the same rate of 10 °C/min. The values of melting temperature ( $T_m$ ), crystallization temperature ( $T_c$ ) and crystallinity degree ( $X_c$ ) were determined from the thermograms from the cooling and second heating. Crystallinity degree was calculated using the following formula:

$$X_c(\%) = \frac{\Delta H_{m_{\text{exp}}}}{\Delta H_{m_{\text{theo}}}} \times 100 \quad (1)$$

Where  $X_c$  is the crystallinity degree,  $\Delta H_{m_{\text{exp}}}$  is the experimental melting enthalpy and  $\Delta H_{m_{\text{theo}}}$  corresponds to 100% crystalline PE,  $293 \text{ J/g}$ .<sup>[18]</sup> Since coconut flour is considered to be inert, the area of the peak was normalized according to the actual content of PE.

Thermal decomposition analyses were performed in order to elucidate the thermal stability of the samples evaluated. The Dharwadkar-Karkhanavala method<sup>[19]</sup> (modified Horowitz-Metzger) was employed to calculate the activation energy ( $E_a$ ). Samples (5–6 mg) were heated from room temperature up to 700 °C at 10 °C/min in a Mettler-Toledo TGA/STDA analyzer.

Dharwadkar-Karkhanavala method equations are as follows:

$$\ln[-\ln(1 - \alpha)] = \frac{E_a}{RT_i^2} \cdot \frac{100}{(T_f - T_i)} \theta + C \quad (2)$$

where  $T_i$  is the reaction starting temperature (K),  $T_f$  is the reaction end temperature and  $C$  is a constant.<sup>[20]</sup>

For reaction order different to one, the left member of the equation is equal to:

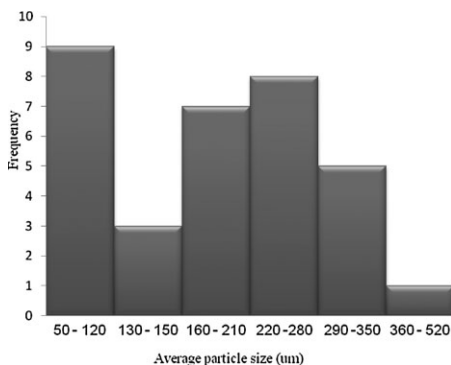
$$\ln \left[ \frac{1 - (1 - \alpha)^{1-n}}{1 - n} \right] \quad (3)$$

The morphology of the coconut flour particles with and without surface treatment, as well as the fracture surface of the composites, was analyzed using a scanning electron microscope (SEM), Hitachi S-2400.

## Results and Discussion

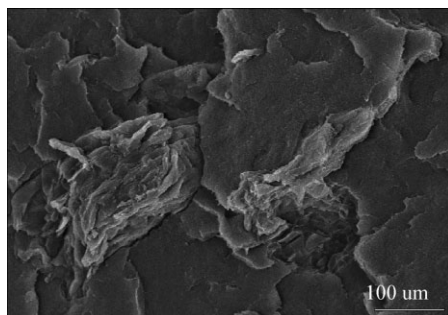
The different properties analyzed are presented in Table 1. The Young's modulus ( $E$ ) and the tensile strength ( $\sigma_B$ ) of the HDPE-untreated coconut flour composites increased from 717 to 882 MPa and from 12.1 to 19.4 MPa, respectively, with a maximum at 15 wt.% of coconut flour. This tensile behavior could be influenced by the particle size distribution (Figure 1), because their sizes range between 50–120  $\mu\text{m}$  for smaller particles, 220–280  $\mu\text{m}$  and 290–350  $\mu\text{m}$  for the larger ones.

Tensile strength values increase up to 60% for the composite with 15 wt.% of coconut flour in comparison to pure HDPE, perhaps because hydrogen interacts with the numerous hydroxyl groups that are present in the filler, thus forming a three-dimensional network among the coconut flour particles that enhances tensile properties.<sup>[21,22]</sup> The addition of more filler



**Figure 1.**  
Particle size distribution of coconut flour.

(20 wt.%) decreased both properties, due to the formation of agglomerates and to the stronger filler-filler interactions. Values of elongation at break ( $\epsilon_B$ ) sharply decrease due to the polymer-filler interface discontinuity, as shown in micrograph in Figure 2, corresponding to a HDPE with 15 wt.% of coconut flour.



**Figure 2.**  
SEM micrographs of the fracture surfaces of HDPE with 15 wt.% of coconut flour.

**Table 1.**  
Mechanical, thermal and thermodegradative behavior of the HDPE-coconut flour composites.

HDPE/coconut flour composites	Mechanical properties				Thermal properties		
	$E$	$\sigma_B$	$\epsilon_B$	$T_c$	$T_m$	$X$	$E_a$
	(MPa)	(MPa)	(%)	(°C)	(°C)	(%)	(kJ/mol)
100/0	717±36	12.1±1.9	153±37	111	135	65	325
95/5	716±21	11.2±0.6	119±4	114	136	71	270
90/10	692±17	12.2±1.7	37±5	114	136	73	253
85/15	882±38	19.4±0.9	12±1	114	136	70	281
80/20	733±23	18.7±0.4	12±1	115	137	72	306

Some theoretical models were used to analyze the tensile data. The Young's modulus was predicted using the Guth-Smallwood equation:<sup>[23,24]</sup>

$$\frac{E_c}{E_m} = 1 + 2.5\phi_f + 14.1\phi_f^2 \quad (4)$$

where  $E_c$  is the Young's modulus of the composite (MPa),  $E_m$  is the Young's modulus of the matrix (MPa) and  $\phi_f$  is the filler volume fraction. To calculate  $\phi_f$ , the values of 0.9595 g/cm<sup>3</sup> as the HDPE's density<sup>[18]</sup> and 0.09975 g/cm<sup>3</sup> as the density of the coconut flour (determined experimentally) were used.

Furthermore, to predict the theoretical values of the tensile strength, the equation of Nicolais-Narkis was employed:<sup>[25]</sup>

$$\frac{\sigma_c}{\sigma_m} = \left(1 - 1.21\phi_f^{2/3}\right) \quad (5)$$

where  $\sigma_c$  is the composite's tensile strength (MPa) and  $\sigma_m$  is the matrix tensile strength (MPa).

On the other hand, the equation of Nielsen<sup>[26]</sup> was used to predict the values of  $\varepsilon_c/\varepsilon_m$  and to compare them to the experimental ones. This equation indicates the existence of perfect adhesion, because the fracture tends to proceed from particle to particle. The equation is:

$$\frac{\varepsilon_c}{\varepsilon_m} = \left(1 - \phi_f^{1/3}\right) \quad (6)$$

where  $\varepsilon_c$  is the elongation at break of the composite (%) and  $\varepsilon_m$  is the elongation at break of the matrix (%).

Table 2 shows the dependence of the Young's modulus, tensile strength and elongation at break values ( $E_c$ ,  $\sigma_c$ ,  $\varepsilon_c$ ) on the composites normalized to the respective values of the Young's modulus, tensile

strength and elongation at break of the matrix on the weight fraction of the filler.

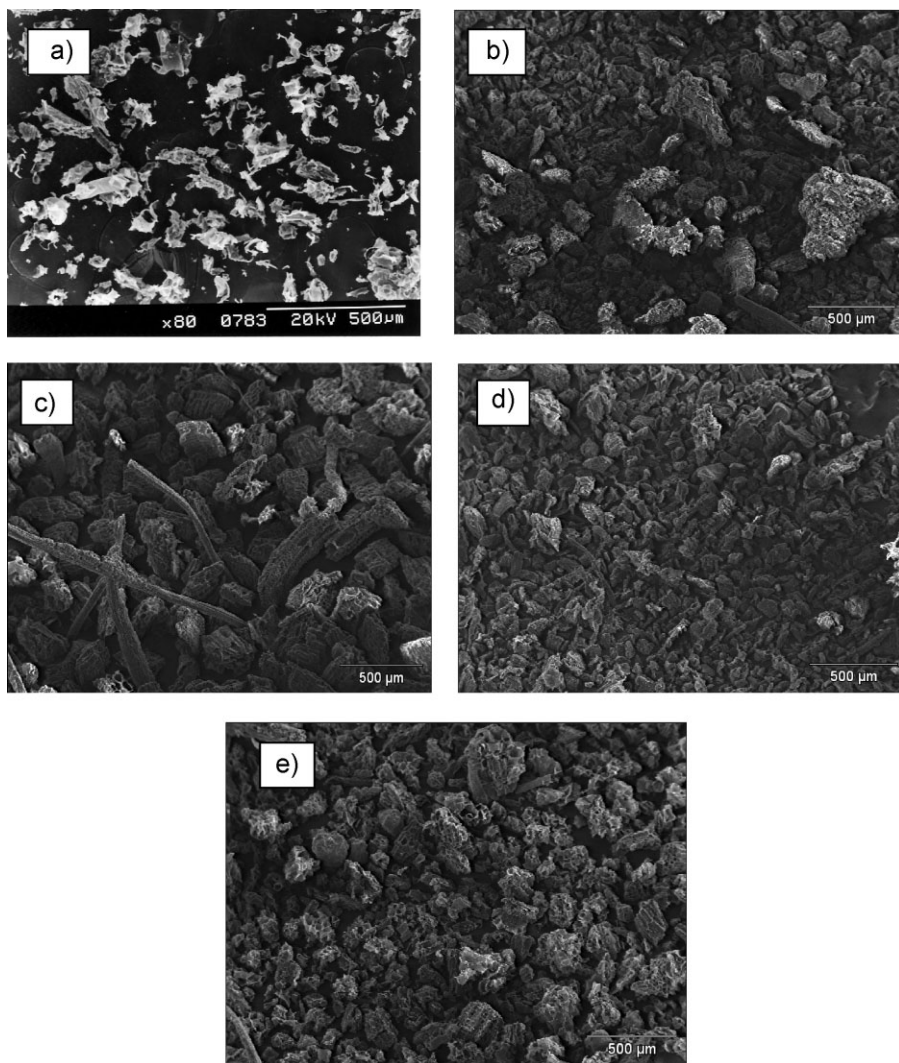
When the results presented are analyzed, it can be concluded that the values of  $E_c/E_m$  of the composites obtained through Equation (4) are reasonably well related to the experimental results, indicating that the incorporation of coconut flour increases HDPE rigidity, through the restriction in mobility of the polymer molecules. Slight differences observed are due to some assumptions that were made by the authors of the equation at the time of putting it forward, which are not fulfilled in the composites analyzed in this work. They are: the equation takes into account that the filler must be evenly distributed, there must not be filler-filler interactions and the particles must be rigid and spherical. These assumptions are not exactly true in this case as can be seen in the micrographs (Figures 2 and 3a), because coconut flour particles are not spherical and rigid and tend to agglomerate. It can be inferred that filler-filler interactions could be possible in the composites when the filler content increases. The equation does not consider crystallinity as a factor that can modify Young's modulus.

The tensile strength of the composites was analyzed using Nicolais-Narkis' theoretical model to understand the generation of discontinuities or weak points in the structure of these two-phase systems. The predicted values of the  $\sigma_c/\sigma_m$  ratio are smaller than the experimental ones throughout the range of coconut flour contents, except for the composite containing 5 wt.% of filler. The Nicolais-Narkis equation describes structures where the adhesion is poor, because the weight factor

**Table 2.**

Comparison of experimental data with the theoretical predictions from the Equation 4, 5 and 6 for HDPE with different content of coconut flour.

Composites	$E_c/E_m$ theo.	$E_c/E_m$ exp.	$\sigma_c/\sigma_m$ theo.	$\sigma_c/\sigma_m$ exp.	$\varepsilon_c/\varepsilon_m$ theo.	$\varepsilon_c/\varepsilon_m$ exp.
95/5	1.0136	0.9986	0.9632	0.9256	0.8256	0.7778
90/10	1.0292	0.9651	0.9402	1.0083	0.7776	0.2418
85/15	1.0433	1.2302	0.9235	1.6033	0.7485	0.0784
80/20	1.0593	1.0223	0.9073	1.5454	0.7232	0.0784



**Figure 3.**

Morphology of the coconut flour particles: a) untreated, b) with NaOH at 5%, c) with NaOH at 18%, d) Lica 12, e) acetylated.

(1.21) is believed to be dependent on the adhesion quality between the matrix and the inclusion. A value of 1.21 of the weight factor is stated to be valid for the extreme case of poor adhesion and spherical inclusions.<sup>[27]</sup>

Table 2 presents a steeper decrease in the  $\varepsilon_c/\varepsilon_m$  values than the one that Nielsen's equation predicts. This indicates that the filler produces discontinuities in the stress transfer and that the composite is weaker, bringing about the fracture at lower strains.

These results contradict those obtained for the tensile strength. This could be explained by the formation of agglomerates of coconut flour (shown in Figure 2) resulting from interactions between hydroxyl groups, which strongly reduce the values of elongation at break.

The crystallinity degree (X) increased from 65 to 70% and crystallization temperature ( $T_c$ ) from 111 to 115 °C (Table 1), indicating that the coconut flour could be acting as a nucleating agent. Similar results



were observed by Habibi et al.<sup>[7]</sup> in their studies on LDPE with cotton fibers. It is also observed that independently of the content of coconut flour, there are no appreciable differences in  $T_c$  and  $X$ , while melting temperature ( $T_m$ ) remained constant around 135–137 °C.

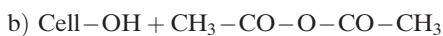
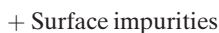
On the other hand, activation energy determined according to the Dharwadkar-Karkhanavala method decreased slightly when the filler was added, thus indicating that coconut flour, due to the polar groups (OH) it contains, could be accelerating the HDPE degradation process. Reaction order ( $n$ ) value is 1.5 for all composites and for HDPE alone, implying that the decomposition process is complex and that it is likely that several decomposition mechanisms are occurring simultaneously or in series. Furthermore, the value of “ $n$ ” means that the decomposition reactions are not elemental.

Based on these results, HDPE with 15 wt% of coconut flour was selected as the appropriate proportion to study the different treatments previously mentioned.

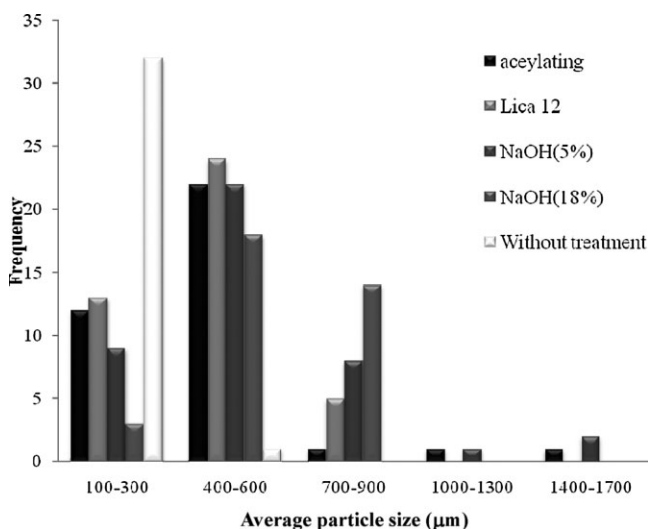
Figure 3 shows the morphology of the treated coconut flour particles. It can be seen that the particles tend to agglom-

erate regardless of the surface treatment or the interfacial agent used. Figure 4 shows the particle size distribution of the coconut flour particles treated with the different additives. It can be observed that the largest amount of particles is found within the 400–600  $\mu\text{m}$  range.

Treatment with  $\text{NaOH}^{[15]}$  (a) and acetylation<sup>[17]</sup> (b) are obtained from the following reactions:



In the case of treatment with  $\text{NaOH}$ , it removes lignin, wax, etc., which serve to cover the outer part or the cell wall that protects the content of the vegetable cell; therefore, this treatment depolymerizes the initial cellulosic structure. As a consequence, the inner region is less dense and rigid, thereby increasing surface roughness, which, in turn, could contribute to a better



**Figure 4.**

Particle size distribution of coconut flour with different treatments.

polymer-filler interaction thus increasing the modulus and tensile strength.<sup>[10]</sup>

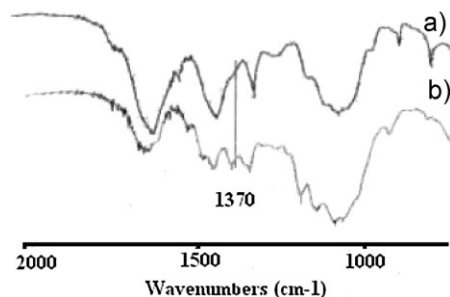
Concerning coconut flour acetylation, Figure 5 presents FTIR spectra of acetylated and not-acetylated coconut flour, showing bands corresponding to the C=O groups at  $1640\text{ cm}^{-1}$ ,  $\text{CH}_3$  asymmetric absorption of the  $\text{CH}_3\text{CO}$  group at  $1430\text{ cm}^{-1}$ , and  $\text{CH}_3$  symmetric absorption of the  $\text{CH}_3\text{CO}$  group at  $1370\text{ cm}^{-1}$ , C–O–C asymmetric stretching at  $1245\text{ cm}^{-1}$  and C–O–C symmetric absorption at  $1060\text{ cm}^{-1}$ . This means that the acetylated coconut flour contains OH, C=O,  $\text{CH}_3$  and C–O–C groups.  $\text{CH}_3$  and C=O groups are the ones that best indicate that coconut flour was acetylated, because they take part in the acetylation reaction, according to the results reported by Sulcahuaman et al.<sup>[28]</sup> Within the band at  $1370\text{ cm}^{-1}$  in the sample with not-treated coconut flour, a slight shoulder is observed that converts into a band with a certain intensity after acetylation (Figure 5), due to the  $\text{CH}_3$  group absorption by the coconut flour. Substituting OH groups in coconut flour with the  $\text{CH}_3\text{CO}$  group, implies a lower coconut flour polarity, therefore moisture absorption should be less. Since fiber does not absorb moisture, it does not swell and the composite material does not contract. This has a positive effect on the polymer-filler interface. This behavior can be observed when micrographs of the sample fracture surface are compared to acetylated coconut

flour and those treated with NaOH (Figure 6).

Figure 3 shows that coconut flour particles treated with Lica 12 present a more uniform size in comparison to coconut flour submitted to other surface treatments, because molecules of neoalkoxy triorganofunctional titanate coupling agent form a monolayer over the filler surface, giving rise to particulate deagglomeration and also to the removal of air and water.<sup>[29]</sup>

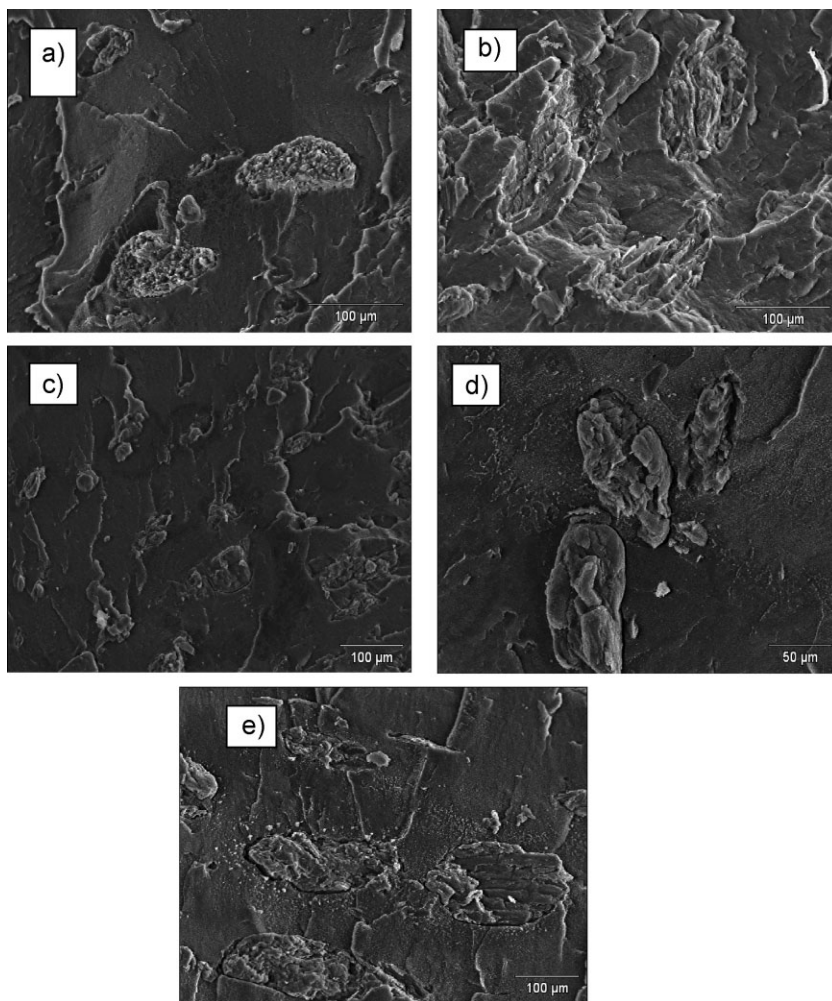
With respect to HDPE tensile properties with treated filler (Table 3), Young's modulus increased approximately 40% with the surface treatments used, being this effect less notorious when the filler was treated with NaOH at 18% and with Lica 12. Similar behavior was observed for the tensile strength, being the increase of up to 70%. Basically, the different interfacial agents employed and the different surface modifications done to the coconut flour improved the interfacial bonding. Concerning elongation at break of composites with filler, a decrease occurs when surface is modified, because polymer-filler interaction increases; as a consequence, the composite is less ductile because interactions increase composite rigidity. Vallejos<sup>[30]</sup> reports that acetylation improves effort transfer efficiency through the interface. Results similar to those obtained in this research work were obtained by Ichazo et al.<sup>[31]</sup> in their PP composites with acetylated sisal fiber and by Abdelmouteh et al.<sup>[21]</sup> in LDPE with coconut flour and a silane coupling agent.

When the ethylene-acrylic acid copolymer was added to the HDPE composite with 15 wt% of coconut flour, the largest increase is observed in Young's modulus and in tensile strength, due to several reasons: 1) the coconut flour particle size in the HDPE matrix is smaller when compared to that obtained when the different surface modifications were carried out (Figure 6), 2) the interaction between acid acrylic functional groups with OH groups and those produced by the treatment of the coconut flour with NaOH ( $\text{O}^-$  and  $\text{Na}^+$ ), because it was previously treated with NaOH. The acrylic



**Figure 5.**

FTIR of the coconut flour non acetylated (a) and acetylated (b).



**Figure 6.**

SEM micrographs of the fracture surfaces of HDPE with 15 wt.% of coconut flour with the different treatments and the interfacial bonding: a) acetylated, b) Lica 12, c) copolymer ethylene-acrylic acid d) NaOH 18%; e) NaOH 5%.

**Table 3.**

Mechanical, thermal and thermodegradative behavior of the HDPE with 15 wt% of coconut flour with interfacial agents and surface modification.

Different treatments	Mechanical properties				Thermal properties		
	E	$\sigma_B$	$\epsilon_B$	$T_c$	$T_m$	X	Ea
	(MPa)	(MPa)	(%)	(°C)	(°C)	(%)	(kJ/mol)
NaOH 5%	1219 ± 18	32.2 ± 0.7	5.2 ± 0.2	112	132	63	266
NaOH 18%	1045 ± 16	30.7 ± 0.5	6.6 ± 0.2	112	132	67	263
Cop. EAA	1282 ± 27	33.3 ± 1.5	5.3 ± 0.4	112	133	63	314
Lica 12	790 ± 48	26.4 ± 2.4	6.5 ± 0.6	111	133	64	213
Acetylating	1251 ± 20	31.3 ± 0.6	6.6 ± 0.3	112	133	65	222



part, in turn, favor polarization in the copolymer and, therefore, results in higher dispersion of coconut flour in the PE matrix; and 3) the ethylene part of the copolymer that is related to the HDPE, giving rise to higher dispersion and filler distribution in the HDPE matrix.

Composites of HDPE with 15 wt.% of coconut flour submitted to different treatments were analyzed through the theoretical models already mentioned (Table 4). It is observed that  $E_c/E_m$  experimental values are higher than the one corresponding to the same composite not treated, except for the case when Lica 12 was used as coupling agent. This means that these composites show more rigidity resulting in some cases form the polymer-filler interaction and also from the better load dispersion over the polymer matrix; these factors are not considered in the development of the Guth-Smallwood model.<sup>[23,24]</sup>

The predicted values of the ratio  $\sigma_c/\sigma_m$  are smaller than the experimental ones in all the treatments analyzed, which again confirms a higher polymer-filler interaction in all cases. Due to these results, a P factor was introduced in equation (5) instead of 1.21, being the values of P negative as seen in Table 5, because the composite's tensile strength is higher than that of the matrix. According to Maiti & Lopez,<sup>[27]</sup> the polymer-filler adhesion improves as the "P" values decrease. Hence, it can be concluded that some sort of interface interaction is present in these composites. This fact was confirmed when the Kunori & Geil equation was used.<sup>[32]</sup> This equation relates the tensile strength with a proportionality parameter "a", which is a stress concentration

**Table 5.**

Values of the parameters "P" and "a" from Equation 5 and 7 for HDPE with 15 wt.% of coconut flour with different treatments.

Treatments	"P"	"a"
NaOH 5%	−10.4346	−31.8675
NaOH 18%	−9.2118	−28.8673
Cop. EAA	−11.3314	−33.9901
Lica 12	−5.7064	−19.3768
Acetylating	−9.7009	−30.0846

parameter. A higher value of "a" corresponds to a stronger stress concentration. The Kunori & Geil equation is as follows:

$$\frac{\sigma_c}{\sigma_m} = \exp(-a\Phi_f) \quad (7)$$

The values of "a" are shown in Table 5. As it can be seen, those values are negative, thus leading to the same conclusion as before.

"P" and "a" values obtained for HDPE composites with coconut flour treated with Lica 12 are the highest, implying that although Lica 12 is having an effect on the polymer-filler interface, its action is not as effective as the addition of copolymer and the other treatments.

The analysis of  $\epsilon_c/\epsilon_m$  values shows that experimental values are lower than those predicted by Nielsen's theory, because with a stronger polymer-filler interaction, composites are more fragile and, therefore, the experimental  $\epsilon_c/\epsilon_m$  ratio decreases.

Table 3 shows that Tc, Tm and X values of the composites submitted to different treatments dropped from 114 to 111 °C, 136 to 132 °C and 70 to 63%, respectively. These values support the presence of an interaction between the two components,

**Table 4.**

Comparison of experimental data with the theoretical predictions from the Equation 4, 5 and 6 for HDPE with 15 wt.% of coconut flour.

Treatments	$E_c/E_m$ theo.	$E_c/E_m$ exp.	$\sigma_c/\sigma_m$ theo.	$\sigma_c/\sigma_m$ exp.	$\epsilon_c/\epsilon_m$ theo.	$\epsilon_c/\epsilon_m$ exp.
NaOH 5%		1.3821		1.6598		0.4333
NaOH 18%		1.1848		1.5825		0.5500
Cop. EAA	1.0433	1.4535	0.9235	1.7165	0.7485	0.4417
Lica 12		0.8957		1.3608		0.5417
Acetylating		1.4184		1.6134		0.5500

which further reduces the close packing of the HDPE chains.

Figure 6 shows evidence of an improvement in the polymer-coconut flour interaction for the treatments with acetylation, titanate and the EAA copolymer. This confirms the better tensile behavior of these composites. Although a clear interaction is not observed in the treatment with NaOH, the good mechanical properties obtained could be due to the better dispersion of the treated particles.

According to Table 3, when the filler surface is modified with different treatments,  $E_a$  falls from 281 kJ/mol to 213 kJ/mol, because the filler surface characteristics accelerate the HDPE degradation process. Although the acrylic acid carboxylic groups are less stable because the copolymer activation energy is 144 kJ/mol, polymer-filler interaction increases when ethylene-acrylic acid-based copolymer is added, due to the good dispersion of coconut flour which results from the copolymer interaction and the ethylene part of the copolymer that is akin to HDPE. This means that the composite with an  $E_a$  value of 314 kJ/mol is more stable.

## Conclusion

Coconut flour in composites with HDPE increases Young's modulus and tensile strength, but reduces elongation at break. When Guth-Smallwood, Nicolais-Narkis and Nielsen equations are applied, it can be inferred that discrepancies exist in the Young's modulus between the experimental values and those obtained by the model, because the assumptions on which the experimental values are based, such as particles are spherical and they do not form agglomerates, are not fulfilled in the composites under study. Furthermore, this model does not consider crystallinity as a factor that could modify Young's modulus. It was also demonstrated that OH groups present in coconut flour accelerate the composite degradation process.

Treating coconut flour surface with NaOH and acetylation, as well as using ethylene-acrylic acid based copolymer, increase Young's modulus and tensile strength and decrease elongation at break, due to the greater polymer-filler interaction obtained when these additives are used. This was confirmed by the SEM micrographs. When Guth-Smallwood, Nicolais-Narkis and Nielsen equations are applied to these composites, it can be inferred that "a" and "P" clearly show an interaction at the interface.

Thermal studies show that crystallization temperature and crystallinity increase, meaning that coconut flour acts as a nucleating agent. However, when coconut flour was treated with the different additives, it was observed that crystallization and fusion temperatures and crystallinity drop, due to the significant effect that polymer-filler interactions have on these properties.  $E_a$  of composites with treated coconut flour decreases because when the filler is treated, groups that are more susceptible to the formation of radicals, which accelerate the composite decomposition process, are found at the interface. However, thermal stability increased with the use of copolymer, due to the bond that occurs between the filler OH groups and the AAc carboxylic group and to the affinity between the ethylene part of the copolymer and the composite matrix (HDPE).

These results lead us to infer that treating the filler, as well as the use of copolymer, improve tensile behavior, a characteristic that is useful to applications in both industrial products and in items like lighter furniture.

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