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RESEARCH ARTICLE

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Effect of thermomechanical pulp fibers and clay on the characteristics of high- and low-density polyethylene biocomposites

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Abstract

This study focused on the performance evaluation of clay and thermomechanical pulp (TMP) fiber to reinforce low- and high-density polyethylene (LDPE and HDPE) biocomposites. A 2^3 factorial experiment was designed using two levels of clay, TMP fibers, and PE as variables. Mechanical properties, thermal behavior, melt flow index, and water absorption were evaluated. In HDPE, the partial replacement of TMP fiber with 10 wt% clay increased the melting point. Clay also reduced the main polymer degradation temperature in both matrices (LDPE and HDPE). The mechanical properties of the samples with 20 wt% fiber and 10 wt% clay were similar to or better than those containing 30 wt% TMP, that is, tensile strength and modulus of 34 and 2700 MPa, compared to 30 and 2400 MPa, respectively. Although the water absorption increased with the addition of TMP fiber and clay, the water absorption of the composite with 20 wt% TMP and 10 wt% clay was relatively low and similar to the biocomposite containing 30 wt% TMP, that is, 1.15 and 1.07% after 30 days, respectively. The comparable properties of biocomposites with 30 wt% TMP and biocomposites with 20 wt% TMP and 10 wt% clay demonstrate the potential of clay to reduce the cost of the final product.

Highlights

- Clay enhances the tensile modulus and strength, and reduces the color darkening, compared to TMP.
- TMP fibers and clay reduce the melt flow index, elongation, and impact toughness.
- TMP fibers and clay increase the melting point and reduce the degradation temperature.
- Reduction in production costs of biocomposites by adding inorganic clay filler.

KEYWORDS
cellulose fibers, clay, impact energy, polyethylene biocomposites, tensile properties

1 | INTRODUCTION

Wood fibers can be obtained through well-established mechanical or chemical pulping processes. Thermomechanical pulping (TMP) is a high-yield production process where more than 95% of the woody biomass that enters the process line is converted into lignocellulosic pulp. Cellulose, hemicellulose, and lignin are the main components of the obtained fiber. The TMP fibers are relatively cheap, compared to chemical-thermomechanical pulp (CTMP) and chemical pulp fibers.[1,2]

TMP fibers are commonly applied in the production of folding and boxboards, newsprint, molded pulp, etc. Additionally, in the last years, TMP fibers have been applied as reinforcement in thermoplastic biocomposites.[3] Overall, it has been shown that the addition of wood fibers to thermoplastics such as polylactic acid (PLA)[4] and polyethylene (PE),[5] enhances the mechanical properties, reduces the global warming potential,[6] and improves the degradability of biodegradable thermoplastics such as (polyhydroxyalkanoates) PHAs.[7]

Cost reduction in biocomposites could be achieved using fillers (layered silicates: clay or talc, calcium carbonate: $CaCO_3$, or titanium dioxide: TiO_2).[8] In Europe, standard $CaCO_3$ is the cheapest material (~250 USD/mT), and the most expensive is TiO_2 (2500–3000 USD/mT, rutile grade).[9] According to CEPI statistics, the most widely used mineral fillers in the European pulp and paper industry during 2023 were $CaCO_3$ (52.4%) and clay (17.0%). However, the decision on a specific filler will not only depend on the cost, but on its effect on the final properties. Clay particles have been an important ingredient of pulp furnishings to increase the gloss of printing paper. Clay is also cheaper than pulp fibers which implies economic benefits.[10] The cost of clay particles varies from roughly 1/3 to less than 1/5 of the cost of pulp fibers, when considering, for example, TMP and chemical pulp fibers, respectively.

Clay has a layered structure (lamellar geometry)[11,12] so during the extrusion process, the flakes tend to orient in the direction of extrusion.[13] $CaCO_3$ has a more rhombohedral, spherical, and needle-like structure,[14] which impacts the mechanical properties.[15] The layered clay structure leads to an increase in dimensional stability and improvement of some mechanical properties.[15–18] Leong et al.[15] evaluated the influence on the mechanical properties and melt flow index of three different mineral fillers ($CaCO_3$, talc, and clay) added to PP (10–40 wt%). The results showed higher melt flow index (MFI) values when calcium carbonate was used, however, increased filler fractions deteriorated the tensile modulus and strength. On the other hand, both talc and kaolin clay at a loading of 10% presented similar MFI. The tensile properties were maximized with talc, although tensile strength at 10% filler presented similar values.[15] Post et al.[16] found similar results for the same mineral fillers but with biodegradable matrices.[16] In wood fiber-based biocomposites, when comparing the mechanical properties after adding talc or $CaCO_3$ as a third component in an LDPE-kenaf fiber system, improved effects were observed when increasing the talc fraction, and a reduction in mechanical properties when $CaCO_3$ was used.[17] Additionally, it has been reported that clay in thermoplastic matrices can have a smoother surface finish, reduced water absorption, and improved mechanical properties.[19] Lin and Renneckar[20] reported on clay adsorbed onto wood fiber surfaces by applying a time-consuming layer-by-layer process before compounding with polypropylene. Despite the efforts to modify the surface of the wood fibers, the authors measured a decrease in the tensile strength and modulus, and increased water uptake, as the fraction of clay particles increased.

A previous study demonstrated increased tensile strength and modulus when adding TMP fibers in PE matrices.[3] However, the combined effects of clay and TMP fibers in LDPE- or HDPE-biocomposites have been scarcely explored. Hence, the purpose of this study was to demonstrate the potential of a furnish commonly used for printing paper (TMP and clay particles), as reinforcement in biobased PE (bioPE) for injection molding products. Two grades of bioPE (low-density and high-density PE) were used, with two levels of TMP fibers (20 and 30 wt%). The assessment included mechanical, thermal, and water absorption properties.

2 | EXPERIMENTAL

2.1 | Materials

Low-density polyethylene SPB681/59 (LDPE, with melt-flow index (MFI) 3.8 g/10 min and density of 0.922 g/cm^3) and high-density polyethylene SHL7260 (HDPE, with MFI 20 g/10 min and density of 0.955 g/cm^3) were kindly provided by Braskem (Brazil). Yparex RENEW 0H17 biobased compatibilizer based on maleic anhydride grafted polyethylene (bioMAPE) was provided by YPAREX

TABLE 1 Morphological characteristics of TMP fibers.

Fiber length (mm)	Fiber width (μm)	Fines (%)	Fibril area (%)
1.47 ± 0.01	32.8 ± 0.10	41.0 ± 0.60	13.2 ± 0.10

(The Netherlands). According to the YPAREX, the RENEW 0H17 bioMAPE has a bio-based content >97% and a maleic anhydride content of <2%. The TMP fibers (Norway spruce) and clay SIBELCO particles were provided by Norske Skog Saugbrugs (Norway) (Figure S1). According to the supplier, 98% of the clay particles have an equivalent spherical diameter of less than 20 μm. The TMP fibers were collected from the reject press. The morphological characteristics of the TMP fibers, analyzed with an L&W Fiber Tester Plus (code 912. Software: version 4.0–3.0, Kista, Sweden), are indicated in Table 1. The L&W Fiber Tester Plus automatically quantifies the morphology of the TMP fibers by image analysis of optical images and is based on some 10,000 quantified fibers. Fines content refers to the sum of length averaged fibrous objects with length <0.2 mm. The fibril area is the percentage of the fiber perimeter that is created by the external fibrils. Also, a Hitachi microscope (SU3500 model) was used (5 kV and 6 mm acceleration voltage) for a pre-inspection of the TMP fibers (100× magnification) and the clay (1000× magnification).

2.2 | Compounding

The TMP fibers were dried for 1 h at 105°C. The LDPE and HDPE were dried for 4 h at 80°C, and the bioMAPE was dried for 2 h at 75°C. The polymers, fibers, and clay were compounded in a Coperion twin-screw compounder at 170–180°C, equipped with a water bath and pelletizing unit. All compounds contained 4 wt% bioMAPE. After the first compounding step, the pellets were dried for 4 h at 80°C and compounded again. The pellets were compounded twice to secure a good dispersibility of the fillers in the polymer matrix. Approximately 3 kg of pellets were obtained (diameter ~ 5 mm). The series was produced according to a 2^3 ANOVA factorial design (Table 2). Images of the biocomposite pellets were acquired with an Epson Perfection Pro scanner using a resolution of 1200 dpi.

2.3 | Melt flow index

The melt flow index (MFI) was measured with a Melt Flow Index-Deluxe Presto Stantest Private Limited (Faridabad, India). The temperature was 200°C with a preheating time of 5 min and using 5 kg weight. Three replicate measurements were undertaken for each sample.

TABLE 2 Compounds produced in this study. Four weight percent of the total formulation was bioMAPE in all the series.

Series	PE type	TMP load (wt%)	Clay (wt%)
LD20T	LDPE	20	0
LD20T10C	LDPE	20	10
HD20T	HDPE	20	0
HD20T10C	HDPE	20	10
LD30T	LDPE	30	0
LD30T10C	LDPE	30	10
HD30T	HDPE	30	0
HD30T10C	HDPE	30	10

2.4 | Preparation of test specimens

All test specimens except the Charpy impact specimens, were injection molded using an Xplore Instruments BV system (Sittard, The Netherlands). Four dogbone specimens (type 1A of ISO 527-2) and four circular tablets (target diameter 10 mm, height 4 mm) were injection-molded (10 bar pneumatic pressure, 190°C melting temperature, and 30°C mold temperature). The specimens for Charpy impact testing were machined from ISO 527 type 1A specimens, which were injection molded by ISO 294-1, using an injection molding machine with 500 kN clamping force (Battenfield, Kottingbrunn, Austria). The Charpy specimens were notched using a CEAST Motorized Notchvis (Instron, Norwood, MA, USA). The most common notch geometry of ISO 179 was used (type 1eA; 45°, 2 mm deep, 0.25 mm notch tip radius). The specimens were conditioned for 7 days at 23°C and 50% RH before testing.

2.5 | Bulk density

The bulk density of tablets was calculated as the ratio between the weight and volume of tablets at room temperature. The sample dimensions were measured using a digital caliper and the weights were determined by a Sartorius Analytic (Mettler Toledo, Switzerland) scale. The reported values correspond to the average of four measurements.

2.6 | Mechanical properties

Tensile testing was conducted with a Zwick Roell Zmart Pro (Ulm, Germany), using a load cell of 2.5 kN and a clip-on extensometer. The test speed was 2 mm/min.

Instrumented Charpy impact tests were performed using an Imatek/Rosand Drop Weight Impact (Imatek

Systems Ltd., Old Knebworth, England, UK). The impactor speed, mass, and energy were 2.9 m/s, 8.507 kg, and 36 J, respectively. The tests were performed at room temperature (22 ± 2°C) and −30°C, using five repeat specimens.

2.7 | Thermo-gravimetric analysis and differential scanning calorimetry

Thermo-gravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed with a Netzsch Jupiter F3 instrument. The analysis was performed between 35°C and 800°C under a nitrogen atmosphere and a heating rate of 10°C/min.

2.8 | Fourier-transformed infrared spectroscopy

Fourier-transformed infrared spectroscopy (FTIR) spectra were collected using an FTIR-ATR Perkin Elmer Spectrum (Waltham, United States). Each spectrum was collected in the wavenumber range from 4000 to 652 cm^{-1} with a resolution of 4 cm^{-1}.

2.9 | Scanning electron microscopy

Test specimens (dogbones) were embedded in epoxy resin and prepared for scanning electron microscopy (SEM) cross-sectional imaging in backscatter electron imaging (BEI) mode. The prepared cross-section corresponded to the middle area (2 × 4 mm) of the dogbone specimen. A Hitachi microscope (SU3500 model) was used, using 5 kV and 6 mm acceleration voltage, and working distance, respectively.

2.10 | Water absorption

Water absorption was measured according to the ASTM D570 standard. Four circular tablets (target diameter 10 mm, height 4 mm) were immersed in water for 24 h, 7 days, 10 days, 15 days, and 30 days. The water absorption value was estimated according to Equation I:

$$\text{WA}\,(\%) = \frac{(m_\text{t} - m_0)}{m_0} \times 100 \qquad (1)$$

where WA is the water absorption percentage, and m_0 and m_t are the weight of the specimen before and after immersion, respectively.

2.11 | Statistical analysis

Analysis of variance (ANOVA) was performed to evaluate differences in mechanical properties and water absorption values. ANOVA was performed using Statgraphics Plus statistical software.

3 | RESULTS AND DISCUSSION

3.1 | Biocomposite morphology

Figure 1 shows the images of the biocomposite pellets after the second extrusion. Interestingly, compared to the HDPE-based pellets, the LDPE-based pellets had a wider diameter and appeared to be fluffier, and this was especially the case with the series containing clay (LD20T10C and LD30T10C). Keep in mind that both neat LDPE and HDPE are white, while the biocomposite colors vary from light to dark brown. Greater color changes were observed in HDPE containing 30 wt% TMP (sample HD30T). Generally, the addition of clay reduced the darkening of the specimens. The color change increases with increasing TMP fraction, and it is associated with hydrolysis and oxidation of wood components.[21,22] The darkening of wood fiber at a given extrusion temperature is mainly associated with hemicellulose (pentosans) thermal degradation[23] and with the release of acetic and formic acids.[24]

SEM of dogbones cross-sections revealed the structural composition of the biocomposites containing TMP as reinforcement (Figure 2, left) and those containing TMP and clay (Figure 2, right). The clay particles appear as white particles compared to the gray fibers. In addition to the large tubular gray TMP fibers, we observed a large amount of smaller gray particles. These are fines that have been quantified to be about 46% in the same TMP as used in this study.[25] The quantification of fines is length-weighed and defined as fibers under 0.2 mm in length. The SEM micrographs revealed a good dispersion of the TMP and clay particles in the PE matrix. Additionally, the micrographs revealed a tubular shape of the TMP fibers, indicating that the fibers are mostly aligned parallel to the main flow direction in the injection molded specimens.

3.2 | FTIR analysis

The IR absorption spectra of the LDPE and HDPE matrices combined with fibers and clay are shown in the supporting information (Figures S2 and S3, respectively). The maximum absorbances reached in different areas of the spectrum correspond to functional groups of the fiber, polymers, and clay. The peaks referring to the

FIGURE 1 Biocomposite pellets of the series described in Table 1. The scale bar (lower-right image) corresponds to 10 mm.

fraction of TMP fibers are similar in all cases, the detected areas involve vibrations and stretching peaks from cellulose, hemicellulose, and lignin chemical groups. The symmetric and asymmetric stretching bands for C—H were observed at 2848 and 2916 cm^{-1}: attributed to cellulose, hemicellulose, and lignin groups.[26] In the samples containing clay, peaks appear between 3600 and 3700 cm^{-1}. These correspond to the valence absorption bands of hydroxyl groups (3692 cm^{-1}), surface hydroxyls of the octahedral layer interacting with the core oxygens of the adjacent tetrahedral layer (3650–3670 cm^{-1}), and internal layers (3620 cm^{-1}) of the structure.[27,28] At lower wavenumbers, it was possible to visualize bands related to Si—O bonds (around 695 cm^{-1}) and OH around 900–1000 cm^{-1} related to inner OH deformation in clay,[29] being more predominately in LDPE, which may be due to a more restricted vibration of OH groups.[30] The specific IR absorption bands of the polymers did not show variations, with two maxima at 2915 cm^{-1} and 2849 cm^{-1} of the CH_2 asymmetric and symmetric stretching vibrations, CH_3 umbrella mode: at 1472 cm^{-1} in LDPE, and two peaks in HDPE 1464 cm^{-1}, and 1472 cm^{-1} due to the crystalline splitting. Finally, around 720 cm^{-1} the split CH_2 vibration in both.[31]

3.3 | DSC and TGA

The thermal properties of the pure polymers and the biocomposites are shown in Table 3. The melting temperatures of the pure polymers are somewhat lower than the melting temperatures of the corresponding biocomposites. The biocomposites present two degradation temperatures (supporting information, Figure S4), one in the range 317–339°C, which corresponds to the degradation of the TMP fibers, and one in the range 462–469°C which corresponds to the degradation of polyethylene, where the degradation temperature of LDPE is a few degrees lower than that of HDPE. As expected, the residual mass of the biocomposites is higher than that of the pure polymers. These results agree with previously reported data on PE/wood fiber biocomposites.[32] Moreover, the samples containing clay had between 7% and 9.5% higher residual mass than those without clay, which agrees with the clay fractions applied in this study.

3.4 | MFI

Figure 3A shows the MFI values obtained after adding TMP and clay fillers to the LDPE and HDPE matrices. Compared to the corresponding neat LDPE and HDPE values, the MFI values decreased in both matrices and with the 20 wt% TMP and 30 wt% TMP fiber load. The analysis indicated a statistically significant reduction of the MFI when adding the clay filler ($p < 0.05$) to the biocomposite samples. Generally, adding fillers to polymer matrices decreases MFI,[33] probably due to the reduced mobility of the molecular polymer structure. Using wood fibers as reinforcement in polymer biocomposites leads to a significant reduction of the MFI.[32,34] Although the MFI

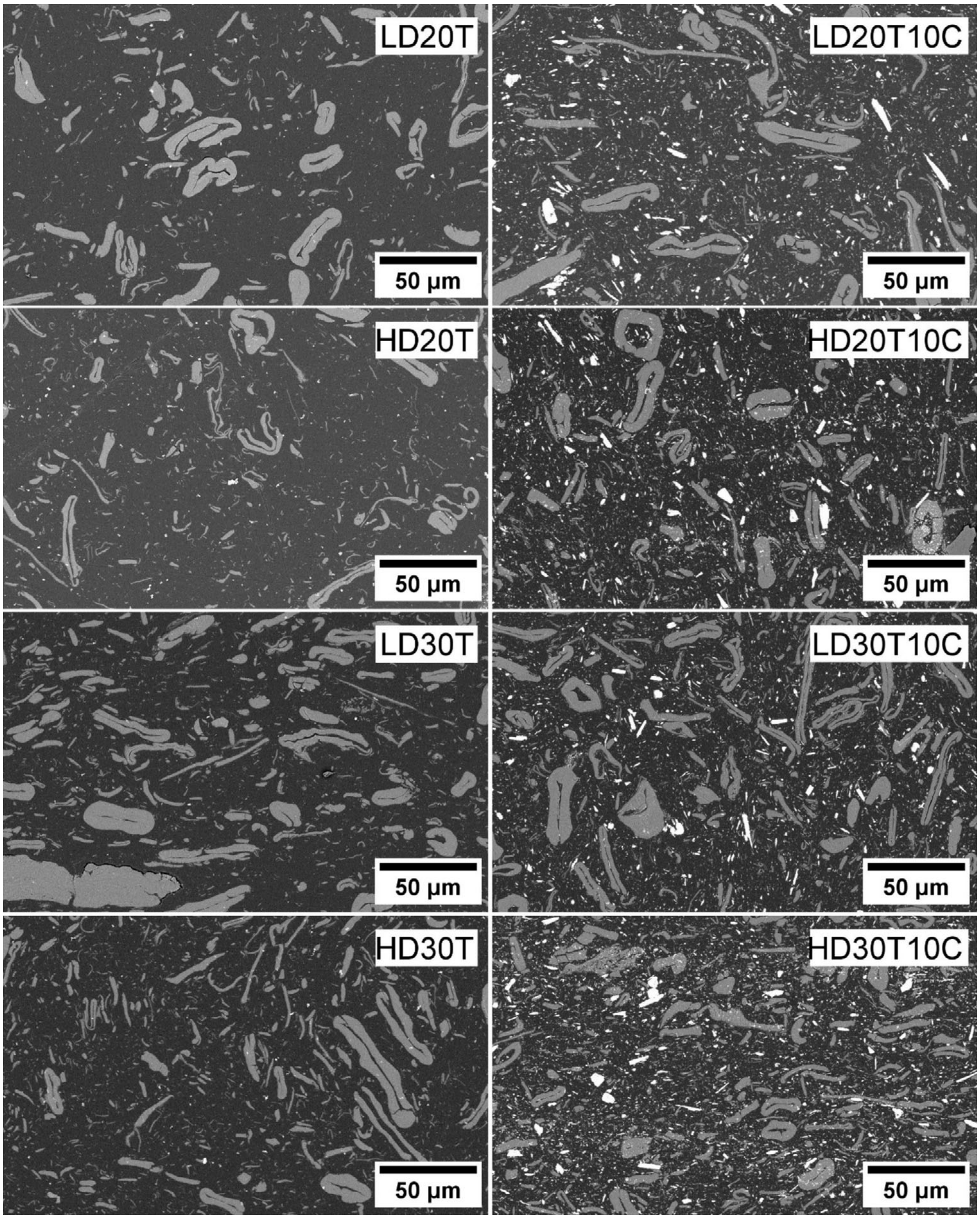

FIGURE 2 SEM of cross-sections of biocomposite specimens. The dark gray background corresponds to the PE, the gray tubular structures are TMP fibers, and the white particles (right column) are clay particles.

TABLE 3 Thermal properties of the biocomposites. The TGA and DSC analyses are provided in Figures S4 and S5.

Sample	Melting temperature (°C)	Degradation temperature 1 (°C)	Degradation temperature 2 (°C)	Residual content (%)
LDPE	111.4	–	467.7	0.14
HDPE	128.2	–	468.2	0.80
LD20T	118.8	317.4	461.9	3.23
LD20T10C	117.0	338.7	462.9	12.69
HD20T	139.6	326.7	466.1	3.19
HD20T10C	140.9	321.3	466.8	10.49
LD30T	118.3	323.8	461.7	5.48
LD30T10C	117.9	322.3	462.8	13.20
HD30T	138.9	323.1	464.7	4.12
HD30T10C	140.0	324.5	466.8	13.09

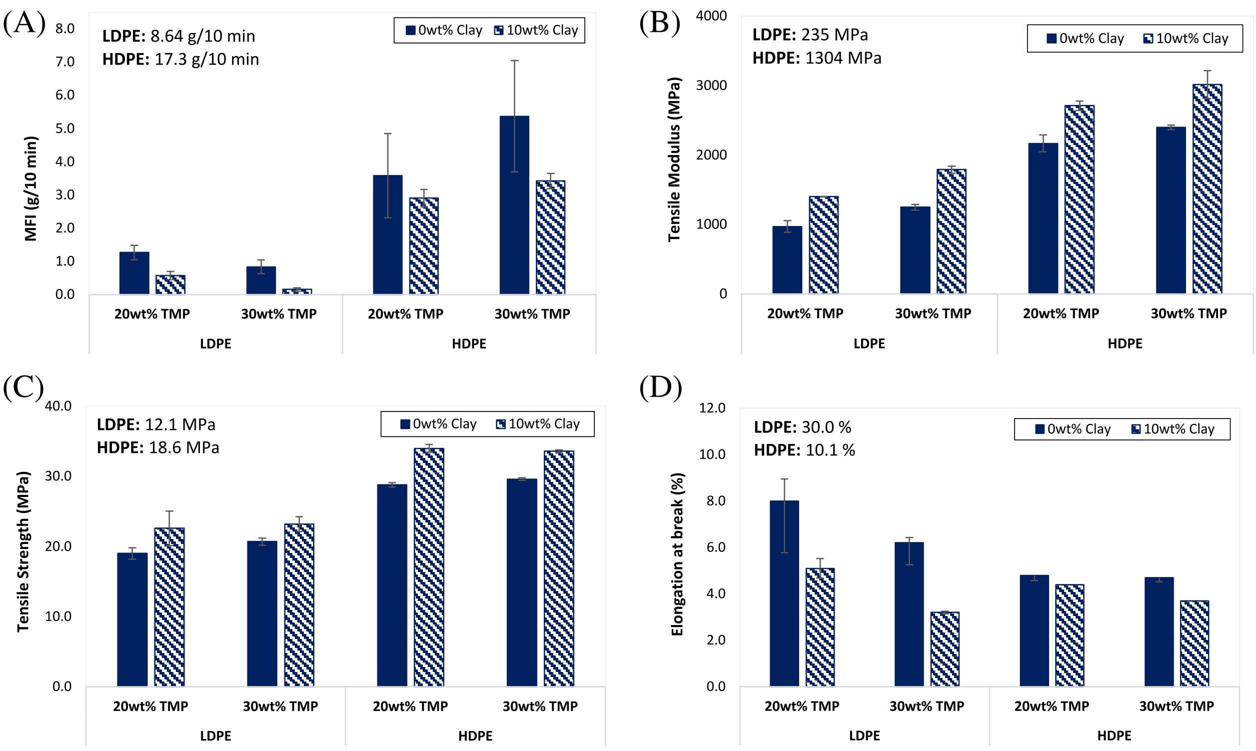

FIGURE 3 The effect of clay addition on (A) MFI, (B) tensile modulus, (C) tensile strength, and (D) elongation at break.

increased when the fraction of fibers was raised from 20 to 30 wt% TMP in the HDPE matrix, the relative increment was not considered significant, as the MFI is still approx. 1/3 of the neat HDPE matrix (17.3 g/10 min). Additionally, it has been observed that clays also reduce the MFI of polymer composites.[35] Biocomposites with the same amount of filler (30 wt% TMP vs. 20 wt% TMP plus 10 wt% clay) show that a filler combination with clay gives a larger reduction in MFI than using only TMP fibers. A study with polypropylene (PP) showed increments in viscosity after the clay addition. The authors attributed the increase to the clay size and its specific surface area, as well as the dispersion of the particle in the matrix which led to an additional restriction in the polymer chain.[36]

3.5 | Tensile strength properties

The tensile strength, tensile modulus, and elongation at the breakpoint of the biocomposites are shown in Figure 3B–D. The ANOVA analysis indicated significant individual effects of TMP fiber load, clay load, and matrix type on the tensile strength and tensile modulus ($p < 0.05$). The interaction effects between fiber load-matrix, and clay load-matrix were also observed for the tensile strength ($p < 0.05$ in both cases). The clay increased the tensile strength in all cases. Moreover, adding 10 wt% clay to the biocomposite with a 20 wt% TMP allowed reaching strength values that were higher than the biocomposites containing 30 wt% TMP fiber. This is considered beneficial in terms of costs since clay is less expensive than wood pulp.[8] An increase in the tensile strength of biocomposites containing cellulose fibers has not been observed with all inorganic fillers. For example, when talc-based fillers were added to biocomposites, a decrease in tensile strength was observed in HDPE/wood flour biocomposites[37] and PHBV/wood fibers.[38] On the other hand, it is well known that talc increases the stiffness of PP, as also demonstrated in CNF/PP biocomposites.[39]

The reduction of tensile strength due to inorganic filler addition has also been reported for precipitated calcium carbonate as filler in bamboo plastic biocomposites.[40] Adding inorganic fillers is associated with an increase in brittleness, leading to early failure. The different effects of talc (in the studies mentioned above) and clay are probably related to particle size. Clay can achieve smaller particle sizes and variable shapes, providing greater surface area and better distribution, which enhance the stress transfer in all biocomposites.[41]

Additionally, the enhanced mechanical properties revealed in this study (tensile strength and modulus) may be related to the dispersion of the clay,[42] which has been shown to also depend on the type of mixing method performed during the extrusion stage.[43] Nunes et al.

demonstrated a 15% increase in tensile strength in recycled HDPE biocomposites with 20 wt% of coir fibers and 3 wt% clay.[44] Additionally, Lei et al. demonstrated the synergic effect of clay and MAPE on HDPE/wood biocomposites production, reaching a 20% increase in tensile strength.[45] The beneficial effect of clay on tensile properties was also observed in studies with other matrices such as PLA.[46] On the other hand, the fiber load significantly reduced the elongation at break ($p < 0.05$), also previously reported by some authors.[47,48] In addition, a significant effect on elongation at break ($p < 0.05$) was observed regarding the interaction of the matrix with the clay and fiber loads. The decrease in elongation at break was more pronounced for the LDPE matrix.

3.6 | Charpy impact toughness properties

The impact toughness values of biocomposites are shown in Figure 4. The statistical analysis shows a significant difference between the individual factors ($p < 0.05$) and between the double interactions ($p < 0.05$). At 23°C, a reduction in the impact toughness/energy when adding fillers is the largest for the LDPE-based composites. This effect in LDPE is probably due to its branched, less crystalline, and less dense polymer structure. This greater flexibility permits more molecular movement of the structure, achieving a high ability to absorb the energy and plastically deform without failure.[49] After filler addition, the reduced mobility leads to a lower ability of the material to withstand the impact.

The increase in fiber load and the inclusion of clay reduced the impact toughness in LDPE-biocomposites (Figure 4A). The inclusion of clay simultaneously leads to an increase in the peak force (Figure 4B). However, impact testing revealed that the 20 wt% TMP and 10 wt% clay samples were similar or tougher than the 30% TMP sample. This is considered advantageous since it would reduce the cost of the biocomposite formulation to achieve the same characteristics in terms of impact toughness. Adding organic and inorganic fillers in LDPE matrices generally reduces the impact toughness as the reinforcement load increases.[50–52] Atuanya et al. observed a decrease in impact toughness after incorporating wood sawdust in LDPE matrices, resembling impact performance studies when coconut fibers[52] and kenaf[51] were used.

In the HDPE, a moderate impact toughness decrease was observed after the addition of fibers (Figure 4A). However, there was no significant difference between the loads. A non-significant effect of fiber content was also reported by Ferreira et al.[53] The authors concluded that the decrease after fiber addition was due to the fiber being a rigid material, reducing ductility, and, therefore, the impact energy.[53] The clay addition demonstrated a small increase in impact toughness ($p < 0.05$). Since the HDPE with TMP fiber fractures at a small displacement, the increase is likely associated with the stiffness increase

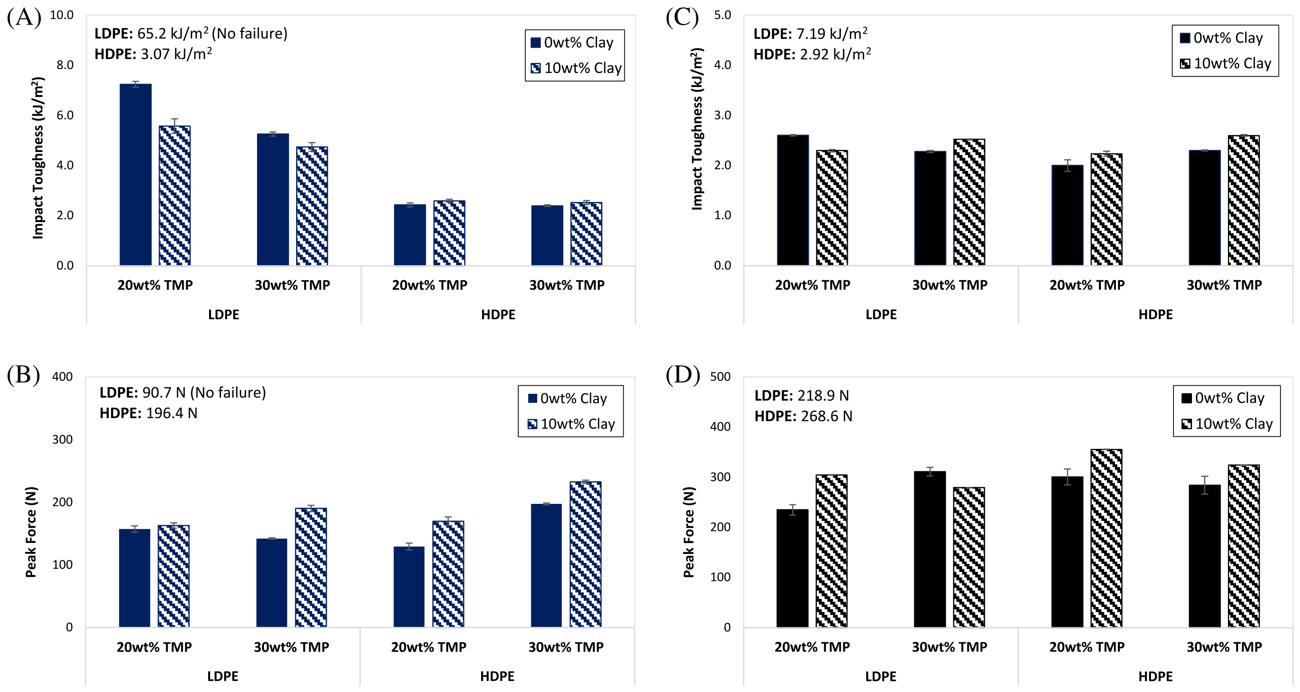

FIGURE 4 Impact strength and peak force, at 23°C (A, B) and at −30°C (C, D).

(a higher peak force, Figure 4B). An increase in the impact toughness of PP/HDPE-based composites with 15% or less of clay was reported.[54,55] Our interpretation is that the increase may be related to the improvement in the distribution of the stress applied over an area, preventing the propagation of cracks.[56] Similar results were obtained with other inorganic fillers like talc and calcium carbonate.[57]

The low-temperature impact tests (−30°C) impart lower impact toughness values for all materials (Figure 4C). Note that the LDPE specimen failed at −30°C, which was not the case at 23°C. The impact toughness decrease is associated with the ductile-to-brittle transition of LDPE, which also allows less displacement. The addition of TMP fiber ($p < 0.05$) and clay ($p < 0.05$) reduced the impact toughness even more. The influence of the clay addition was observed in the maximum force (Figure 4D), probably related to effects on stiffness, but without producing strong effects on impact toughness.

3.7 | Water absorption

The water absorption of biocomposites is a critical factor regarding outdoor applications. Figure 5 shows the water absorption values for neat polymers and biocomposites based on LDPE (Figure 5A), and HDPE (Figure 5B) during the first 30 days (1, 7, 10, 15, and 30 days). Note that equilibrium is not yet reached. However, the test helps to evaluate the absorption kinetics of the biocomposites in their first contact with water. The addition of fillers led to an increase in water absorption in all biocomposites. The ANOVA was carried out with the water absorption values after 30 days as a response. The fiber load, clay load, and matrix type significantly influenced the water absorption values ($p < 0.05$). Significant differences were also observed in the two-way interactions ($p < 0.05$). In cellulose-based biocomposites, the presence of water causes swelling of the cellulose fiber, leading to the development of stresses and, therefore, loss of material performance.[58]

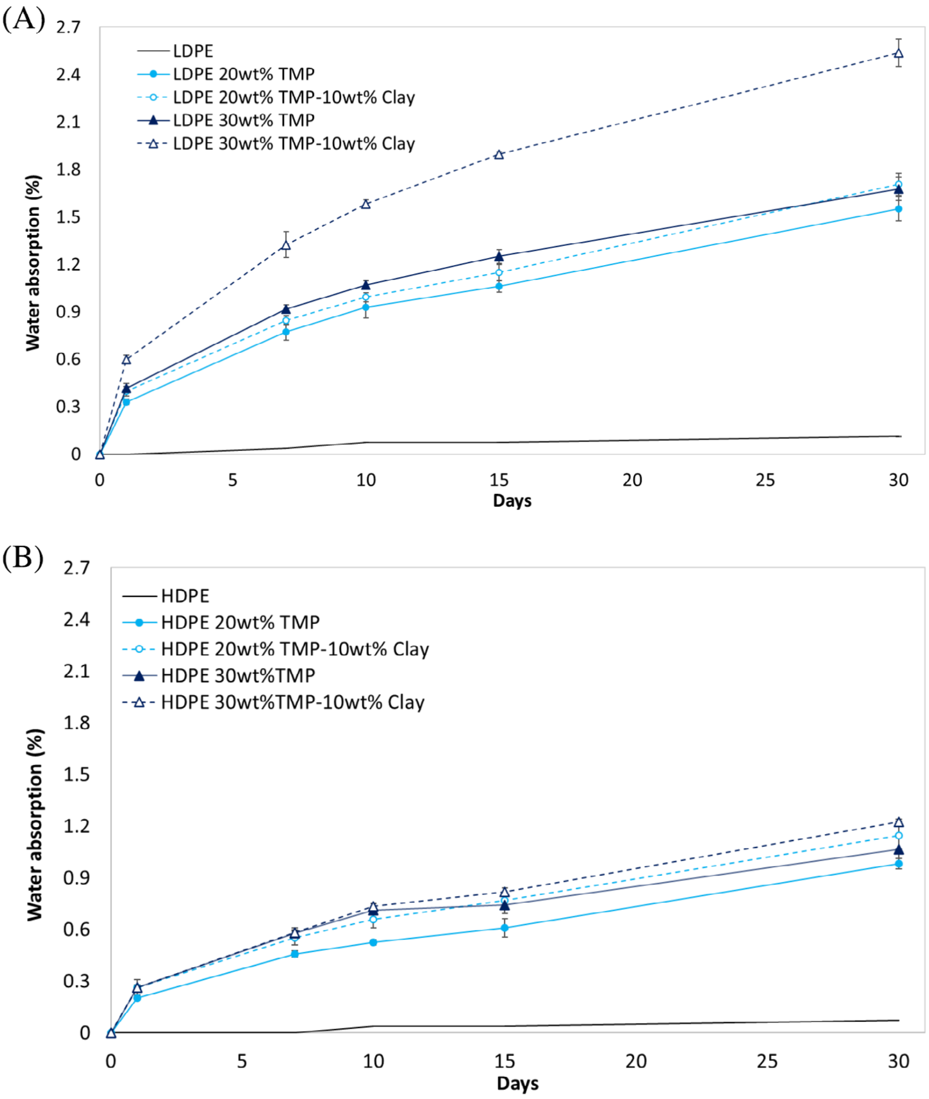

FIGURE 5 Water absorption (A) LDPE and (B) HDPE, and the corresponding biocomposites.

Clay increases water absorption, due to the greater hydrophilicity of clay compared to the polymers.[59] When adding fibers, the increase is even greater, both due to the nature of the fiber and that of the clay as demonstrated by the interaction effect in the statistical analysis. Notice that the effect of filler on water absorption was higher in LDPE than in HDPE. This is possibly due to the greater number of interstitial spaces of the material, due to its high branching structure and low density. It has been reported that water absorption increased due to the dispersion of the clay in the matrix, large aggregates, and random distribution of clay components.[42] As for wood fibers, some strategies to improve the dispersion and compatibilization of clay in polymers involve chemical modification[60,61] or the addition of compatibilizers.[62]

Table S1 shows the initial bulk density values of the injection-molded tablets. Bulk density refers to the mass of the biocomposite that can occupy a given volume. The addition of clay significantly increased the bulk density of the samples ($p < 0.05$). The bulk density increase was also demonstrated in HDPE combined with milled poplar wood fiber and red pottery clay.[63] The authors attributed this behavior to the gaps filling, which decreased the porosity and led to a denser and more compact structure.[63] This could be perceived as contradictory considering that decreased porosity is expected to decrease water absorption. However, in our study clay caused an increase in water absorption after 30 days. Keep in mind that the water absorption in biocomposites is influenced by the diffusivity and the nature of the filler (TMP fiber + clay).[59,64] The higher increase in samples with clay at 30 days, compared with the sample only with TMP fiber, can be attributed to a clay swelling effect, which may cause water to penetrate between clay layers.[65,66] The determination of swelling values will be considered in future studies.

Although the TMP fiber + clay biocomposites developed in this study absorb water, the level of water absorption is considered relatively low compared to previous studies. In this study, the biocomposites were compounded twice, and this caused a good dispersion of TMP fibers and clay particles as revealed by SEM (Figure 2). The good dispersion most probably contributed to limiting the contact of the hydrophilic components (TMP and clay) with water and thus reduced the water absorption. Figure 6 compares water absorption measurements (at room temperature) in biocomposites (30 wt% filler load) from this study with results obtained previously by other authors using a range of matrices.[67–78] Note that the water absorption of our biocomposites containing clay is relatively low compared to other matrices with the same fiber load. PLA biocomposites containing 30 wt% softwood flour (Figure 6, PLA/CP-30F and PBAT/PLA-30F) in commercial matrices (PLA, Ecovio®, Bioflex, and Tenite propionate) absorbed water rapidly reaching roughly 1% water absorption after 24 h,[68] which is approximately 2–3 times the water absorption levels of the PE biocomposites developed in this study (Figure 5). Similarly, the addition of 30 wt% bleached kraft softwood fibers in PLA matrices led to a high-water absorption (5.27%), at the saturation point (20 days).[75] Even for 100 wt% PLA a water absorption value of 0.97% was

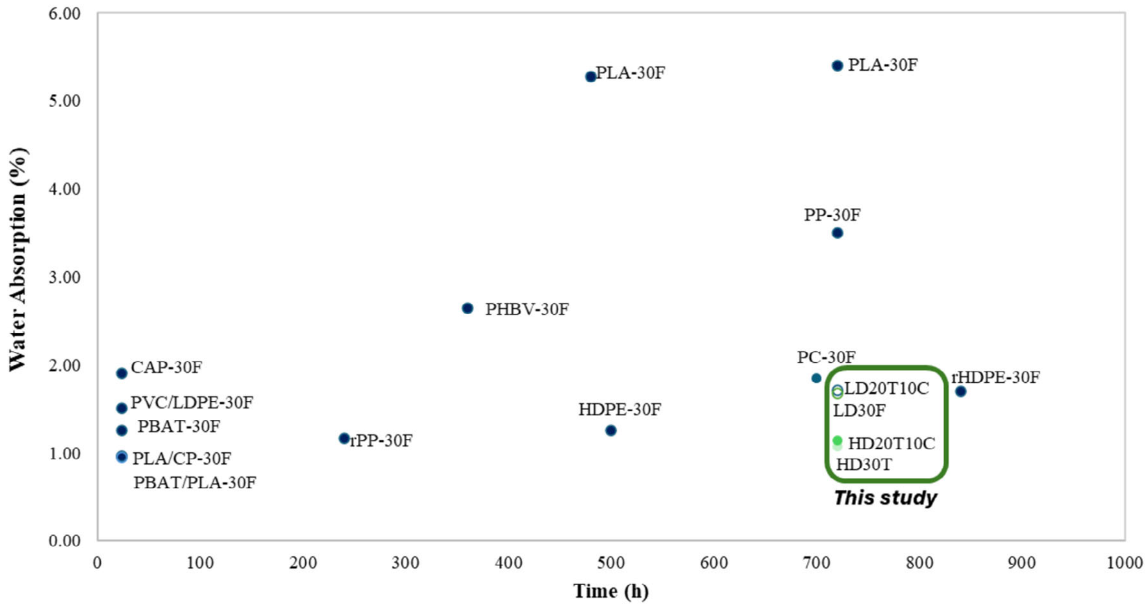

FIGURE 6 Comparison of water absorption for a range of matrices reinforced with 30 wt% cellulose fibers. The following polymers were included: Polypropylene (PP), recycled polypropylene (rPP), polylactic acid (PLA), polycarbonate (PC), polyhydroxybutyrate-co-valerate (PHBV), poly(vinyl chloride) (PVC), polybutylene adipate terephthalate (PBAT, EcoFlex®), copolyester (CP), and cellulose acetate propionate (CAP, Tenite propionate).[57–67]

reached.[75] A similar value was obtained after the addition of 30 wt% walnut shell flour, reaching an increase in water absorption of 5.40% and saturation point at 15 days.[77]

Furthermore, virgin and recycled PP matrices have shown increments in water absorption by incorporating fibers as reinforcement.[74,76,78] For a similar period that the samples evaluated in this study, virgin PP reinforced with wood powder showed twice as high absorption value.[74,78] To conclude, the HDPE and LDPE biocomposites developed in this study have demonstrated improved mechanical properties and relatively low water absorption, a trend also observed in previous studies with virgin and recycled PE and 30% fiber.[69,72]

4 | CONCLUSIONS

This study revealed the effects of clay particles and TMP fibers in composites with polyethylene matrices (HDPE and LDPE). Generally, both TMP and clay increased the mechanical strength of the biocomposites. The tensile strength increased from 19 MPa for neat HDPE to 30 MPa with 30 wt% TMP. Interestingly, the tensile strength of the biocomposite containing 20 wt% TMP and 10 wt% clay was significantly higher (34 MPa) than that of the biocomposite containing 30 wt% TMP (30 MPa). A similar trend was observed for the tensile modulus and impact toughness. Although there are still some water absorption challenges for the biocomposites developed in this study, the measured levels of water absorption were relatively low (roughly 1–2% after 30 days), compared to biocomposites with other polymer matrices (e.g., PP and PLA). Finally, based on the lower price of clay compared to TMP fibers, we expect that using clay in the formulation may imply a significant cost reduction of the final biocomposites.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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