Contribution to the Study of the Thermal, Rheological and Morphological Properties of Biocomposites Based on Typha/PP

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Abstract: Biocomposites based on polypropylene (PP) and typha fibre were prepared using a twin-screw extruder and different characterisation techniques, namely thermal, mechanical, rheological and morphological analysis, to evaluate the effects of typha stem powder on the final properties of the biomaterials. The rheological characterisation showed that the G' and G'' values of pure PP and biocomposites increase with the rate of reinforcement and are related to agglomeration phenomena of Typha fibres which reduce the sliding or flow between them inside the biocomposite. The viscosity of the composites is strongly influenced by the shear rate. Shear thinning behaviour of the melt was observed. The results of the mechanical tests show an increase in tensile young's modulus up to 45% and an increase in tensile strength up to a critical value of 25% for typha stems, which can be attributed to the good interfacial adhesion between the matrix and the filler. Differential Scanning Calorimetry (DSC) measurements indicate the presence of crystalline phases and a slight difference of about 3°C between the melting temperatures. Typha acts as a nucleating agent. Micrographs show the diffuse aspect of the fibre distribution in the matrix. The good wettability of typha fibres by polypropylene contributes to the reduction of microcavities, which has a positive effect on the mechanical properties up to a certain level of reinforcement. Finally, the thermogravimetric analysis shows that typha fibre decreases the thermal stability of the biomaterials.

Keywords: Wood Polymer Composites, Rheology, DSC, SEM

1. Introduction[1]

Currently, the environmental situation in the world is rather unfavorable. A constantly growing population requires more and more food, consumer goods and housing. All of this leads to the growth of cities, industry and an increase in the volume of municipal solid waste [1]. Due to its concerns related to the environment and sustainability issues, industrialists in collaboration with researchers have made remarkable improvements in green materials in the field of polymer science through the development of biocomposites [2, 3]. Biocomposite materials often mimic the structure of the natural fibers involved in the process and retain the reinforcing properties of the matrix used. They represent a category of materials that can be easily processed and adapted to a wide range of applications, such as packaging, building (roof structure, bridge, window, door and green

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[1] I would like to dedicate this work to the memory of our dear Prof. Diène Ndiaye.

kitchen), automotive, aerospace, military applications, consumer products and medical devices [4]. The addition of natural fibers in the polymer matrix helps to reduce the density and cost of the products, while improving the performance of the virgin material. It is well known that the main components of lignocellulosic fibers are cellulose, hemicellulose and lignin. Cellulose is a group of microfibrils made up of d-glucose units linked by β-1,4-glycosidic bonds and controlled between chains by hydrogen bonding, which gives it a highly ordered structure and good properties. Hemicellulose is a group of pentose and hexose polysaccharides, and plays an important role in supporting cellulose microfibrils. Lignin is a phenolic constituent with amorphous structures which stabilizes and protects these fibrils [6-8]. There is no doubt that the great interest in plant fibers is linked to their physical, chemical and mechanical properties, such as their lightness, their profitability, their inexhaustible availability and their biodegradability. Among the natural fibers used in the manufacture of biocomposites we can mention the fibers of bamboo, jute, coconut, rice, etc.

The scientific community has focused on the development of biocomposite materials reinforced with natural fibers.

Due to the fact that the latter have attractive characteristics such as their high specific rigidity, impact resistance, reduction of energy consumption, non-irritation of the skin, renewable, recyclable and biodegradable character. They also offer thermal and acoustic insulation for specific applications.

In the Saint Louis region of Senegal, typha is one of the most abundant fibers, it is used in the manufacture of bags, mats, handicrafts and in the construction sector.

However, this fiber can also be used as a structural reinforcement of polymer matrix composites [9, 10].

PP is a commonly used polymer and it is widely used in a variety of applications due to its non-toxicity, low cost, low density and its ease of processing. In addition, it has good mechanical properties and good chemical stability [11], but the main disadvantage of this polymer is that it retains its non-renewable origin. Blending polyolefins with plant fibers has been reported to be an effective way to reduce production costs [12] while maintaining or increasing matrix properties [13]. Interestingly, polypropylene composites reinforced with vegetable fibers have been shown to have good recycling properties [14].

This high stability of the mechanical properties of composites containing plant fibers is strongly correlated with the value of the aspect ratio of the fiber (L/d) which is defined by the ratio between its length (L) and its diameter (d). The higher the fiber aspect ratio, the better the stiffness of the composite [15].

In this article it is about the incorporation of plant species (Typha) in the Polypropylene matrix to develop composite materials.

The variation in the rate of reinforcement on the thermomechanical properties will be monitored. The morphological characterization of the samples will be studied by scanning electron microscopy (SEM). At the end we will examine the viscoelestic properties of our samples.

The final object is to develop a new generation of biocomposites by extrusion, having good properties to consider several applications.

2. Experimentations

2.1. Materiels

Polypropylene (PP) has been generously provided by Solvay Co. (International Group of Chemistry and Pharmaceuticals). The latter has a density of $0.900g/cm^3$ and a melt index of 2.5g/10 min.

2.2. Composite Manufacturing Methods

The composite materials were obtained by mixing in a co-rotating bis-screw extrusion.

For the extraction of noxious gases, the hood was open. First, the polymer is introduced for 5 min at 180° C and the screw speed set at 40 rev/min. In a second step, the wood fibers are introduced. The system is mixed for 10 min at 180°C at 90 rpm.

After the treatment phase, each sample is sandwiched for 1 min between two sheets of Teflon and pressed at 175°C under a pressure of 200 bars to obtain films with thicknesses between 100 to 200 microns for the study of the different macroscopic properties.

Table 1. The percentage samples of wood and polymer.

Echantillons	PP	Wood
TTPPC0	100	0
TTPPC1	75	25
TTPPC2	65	35
TTPPC3	55	45

2.3. Characterization of Composites

2.3.1. Thermogravimetry

Thermogravimetric analysis. The thermogravimetric analysis was carried out in a TGA50 - Shimadzu equipment under an N2 atmosphere with a purge gas flow rate of 50 $cm^3.min^{-1}$ from 25 to 600°C.

About 10 mg of each wood sample was used. The analysis was carried out at four different heating rates (10°C. min^1). This technique makes it possible to measure weight loss as a function of temperature.

2.3.2. Mechanical Testing of the Composites

The static tensile tests were carried out using an electromechanical testing machine (MTS/100 equipped with a 10 KN load cell. All tests were carried out at 23°C and 50% of relative humidity in a laboratory. The tensile test was carried out in accordance with ISO 527. All the loading speeds were 1 mm.mn^{-1}, except for the determination of the strain at break of the PP, which was obtained with a loading speed of 10 mm.mn. An MTS extensometer was used with a nominal length of 50 mm. The tests were performed at least five times for each specimen and the results were averaged arithmetically.

2.3.3. Morphological Characterization

A HIROX SH4000M scanning electron microscope (SEM) was used. The samples were fractured in liquid nitrogen and after sputter-coated with gold under an argon gas flow for 15 min. The coated samples were left to dry at room temperature before SEM could be performed. The SEM analyses were done at 20 kV and at a range of magnification equal at ×80 to ×150.

2.3.4. Differential Scanning Calorimetry (DSC) Analysis

To study the effect of wood fibers on the thermal properties of PP, 8 to 12 mg of each type of RDPPC granules were used for DSC analysis with the METTLER TOLEDO DSC 822e system.

Scanning electron microscopy (SEM) is used to obtain cracks on the surfaces of micro photographs of composites. It is carried out using an FEI Quanta 400 microscope operating at a voltage of 30 kV. This technique makes it possible to visualize the penetration and adhesion of the molten polymer in the lumen of the wood cells.

The samples were heated to 200°C at a rate of 10°C/min. They were held at this temperature for 5 minutes, then cooled to 50°C at a rate of 10°C/min to follow the crystallization process.

They were again heated up to 200°C to record their melting behaviors. The heat of fusion was calculated from the area of the melting peak. The maximum of the endotherm and the minimum of the exotherm were considered as the melting temperature (Tm) and the crystallization temperature (Tc), respectively. The non-isothermal crystallization behavior has also been investigated in programmed cooling.

TA Universal Analysis software was used to analyze the measurement data. The enthalpy change was recorded, the crystallization temperature (Tc), the melting temperature (Tm) and the degree of crystallinity (χc) were examined. The χc was calculated using the following equation:

$$\frac{\Delta H_m - \Delta H_c}{\Delta H_0 X(1 - \%WTT)} X100$$

Where ΔHm is the measured enthalpy of fusion, ΔHc is the measured enthalpy of crystallization, ΔH_0 is the enthalpy of 100% crystalline PP (209 J/g) [5], and w% TT represents the weight fraction of typha stem in samples.

2.3.5. Rheological Characterization

The rheological characterization was carried out using a rheometer with parallel plates with smooth surfaces (ARES, TA Instruments) in dynamic oscillation and controlled deformation mode. The samples, in the form of filammant, were cut from extruded profiles using a cutting punch. Then they are placed between the parallel plates with a space of 2.5 mm to perform the rheological measurements. In the strain sweep test, the response of the material to increasing strain amplitudes was measured at constant frequency and temperature. This experiment helps to determine the LVR (Linear Viscoelasticity Domain or Region) of materials, which is important to properly perform the frequency sweep test. For each material, three different frequencies (1, 3, 10 rad/s) were evaluated, and the shear strain was between 0.02 and 5%.

The viscoelastic behavior of molten polymers can be determined using oscillatory rheological experiments such as dynamic mechanical testing, which offers a convenient way to assess the frequency dependence of mechanical properties of polymers. A useful representation is to plot the experimental frequency sweep data points in the complex plane. That means that imaginary part (of the complex viscosity) η' values are reported along the abscissa (X axis) and the real ones (η") on the ordinate (Y axis) as schematized in Figure 1; where η' is the loss viscosity, η" is the storage viscosity and ω is the pulsation of the frequency: ω = 2πN. Usually, the experimental points are located on an arc of circle characteristic for a Cole-Cole distribution. The extrapolation of this arc of circle to the zero ordinate value gives the Newtonian viscosity which is related to the average molecular weight Mw of the considered polymer through a power law: (η_0 = K (Mw)3.4) [10].
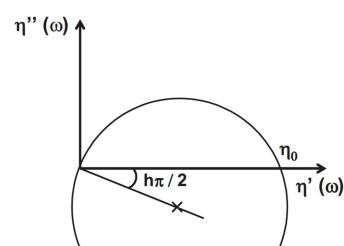

Figure 1. Cole-cole schema.

3. Results and Discussion

3.1. Morphological Characterisation

After breaking up the composites, the facies are analyzed under a scanning electron microscope. The results are shown in the table below. This microstructure reveals voids and holes resulting from a phenomenon of detachment of the fibers during the tensile test.

We also note the presence of broken wood particles, indicating the break in the wood and not at the interface. This observation confirms the good mechanical strength of the fiber/matrix interface. Indeed, an increase in the content of typha fiber and the large contact surface of the fibers contributed to the improvement of the interfacial adhesion.
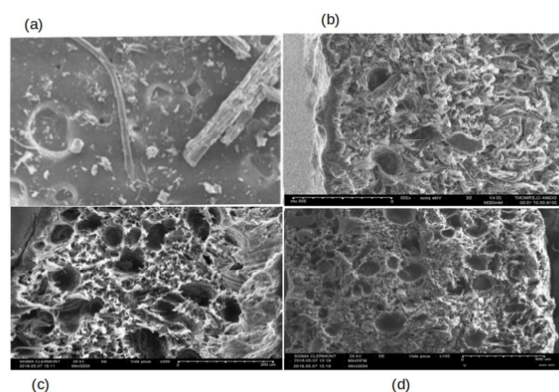
(a) PP/25 TT; (b) PP/35TT (c) PP/45TT

Figure 2. SEM images of PP biocomposite.

Figure 2 shows that the wood particles are not oriented, which results in good adhesion of the fiber/matrix interface. The rupture seems to be adhesive, we observe a loosening of the fibers leaving holes on the facies of the ruptures. The micrographs make it possible to visualize the diffuse aspect of the distribution of fibers in the matrix. The good wettability of the typha fibers by the polypropylene contributes to the reduction of microcavities which has a positive impact on the mechanical properties.

3.2. Characterisation of the Thermal Stability of the Composite

Thermogravimetric analysis (TGA)

The thermal degradation of pure polypropylene and their bio-composites reinforced with typha fibers has been analyzed in detail by performing thermogravimetric analyzes. Figure 3a shows the thermogravimetric (TG) and differential thermogravimetric (DTG) curves of each natural fiber at a heating rate of 10K/min. TG/DTG curves in an air atmosphere were carried out to understand the thermal degradation behavior of typha fibers, polymers and biocomposites with 10, 20 and 30% of typha fibers (Figure 4). The mass loss up to 220°C has been attributed to the evaporation of water, glycerol and other volatile compounds. Thermal degradation of typa and hemicelluloses occurs between 250 and 300°C [16], while cellulose degrades thermally between 240 and 350°C.
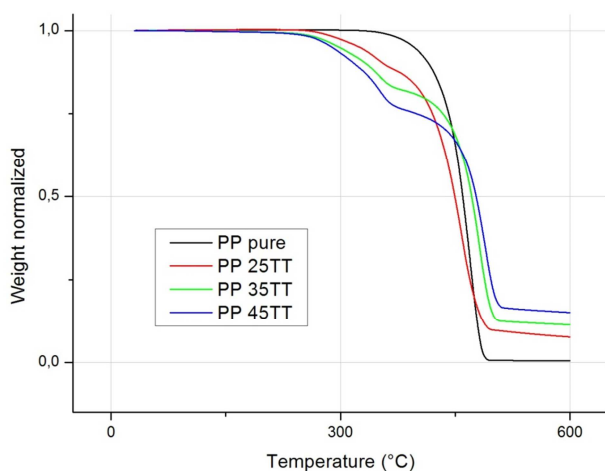

Figure 4. *ATG thermograms of pure PP and its composites.*

According to figure 3 the decomposition of PP, begins at about 400°C (breakage of CC chains of the main chain) [17, 18], this breaking temperature is higher than that of fibers [19, 20]. Pure PP has a one-step decomposition process of 400°C to 500°C. While composites clearly show a two-step decomposition process. The first step corresponds to the decomposition of the fiber and the second step to that of the matrix. The addition of fiber therefore decreases the stability of the Polypropylene in the composite. On the other hand, since typha fibers are characterized by particles of different size and geometry, small fibers seem to slightly decrease thermal stability compared to larger fibers. Indeed, the amount of

interface with the fibers is important for small fibers.

When these start to form (before the PP) the surface area of the PP that can start to degrade is then higher and the decomposition is accelerated so that it takes place at a lower temperature. In general, the addition of typha fiber decreases the thermal stability of biocomposite, because the thermal stability of fibers being lower than that of PP, the degradation of the typha fiber can accelerate the decomposition of Polypropylene [21, 22].
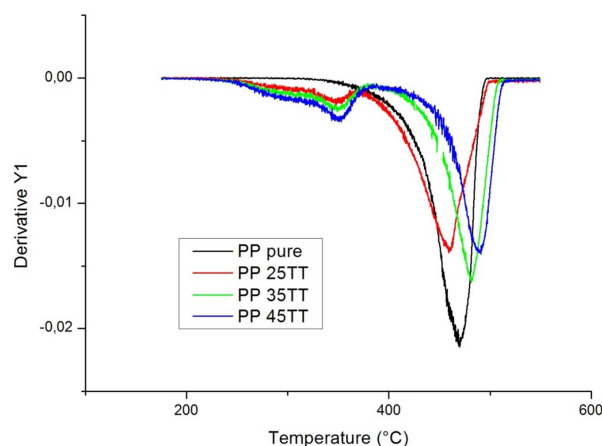

Figure 5. *DTG thermograms of pure PP and its composites.*

The DTG curves of the biocomposites show two zones

The second Characteristic Mass Loss Zone ranges between 400°C and 500°C. In its temperature pages, the initial samples (reduced to 100%) lose approximately 80% of their initial masses. In its areas, if we examine the DTG spectrum, we observe a peak with a maximum noted at approximately 464°C, 480°C, and 500°C, respectively for biocomposites containing 25%, 35%, 45% by mass of powder. These results obtained are confirmed by other research works [23, 24].

### 3.3. Differential Scanning Calorimetry (DSC)

The DSC measurements were carried out on the various samples produced. We have collected a number of data from these measurements, namely the melting temperature (Tf), the crystallization temperature (Tc), the enthalpy of crystallization (ΔHc (J/g)), the enthalpy of fusion (ΔHc (J/g)) and the percentage of crystallinity XC (%). In Figure 3 are shown thermograms of two composite samples of PP containing different percentage of Typha. Like the thermograms in Figure 5 and Figure 6, the thermograms of all samples show exothermic peaks between 161.74°C and 164.51°C during heating and peaks endothermic between 116.96°C and 121.50°C during cooling.

As shown by the two thermograms in Figure 3, there is a slight difference of about 3°C between the melting temperatures. The existence of a difference of 3°C between the two melting temperatures has already been observed. This deviation is probably due to the reorganization in the molten state, to the variation of crystal dimensions or to the presence of crystallites of different stability. The appearance of a small shoulder is observed in the samples of

biocomposites. In fact, the melting behavior of semi-crystalline polymers with semi-rigid chains is generally qualified as multiple because it is characterized by the appearance of several endothermic melting peaks during heating. To describe the origin of this phenomenon, several models have been proposed by the authors. Multiple melting peaks can be explained by the presence of crystals of different thicknesses in the semi-crystalline structure [25, 26]. Therefore, the thinner crystals melt first at a lower temperature. These crystals would then be unable to reorganize into more perfect crystals melting at a higher temperature. In this case, the final melting peak is mainly associated with the thicker crystals formed during the initial crystallization. It is often shown that the number of melting peaks depends on the experimental conditions of crystallization [27, 28].

Table 2. *Neat PLA thermal properties and its composites.*

ECHANTILLON	Tc (°C)	$\Delta H_C^{(J.g-1)}$	Tf(°C)	$\Delta H^{(J.g-1)}$	Xc (%)
PP PUR	116,96	80,54	161,74	30,54	24
PP 25	119,22	73,66	164,48	29,09	86
PP 35	121,23	66,35	164,35	24,43	58
PP 45	121,50	54,52	164,51	18,31	31
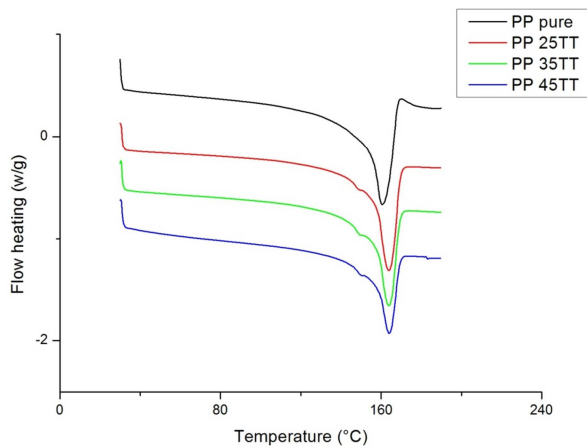
Figure 5. *DSC fusion thermograms neat PP and its composites containing different rates of typha stem, melting.*
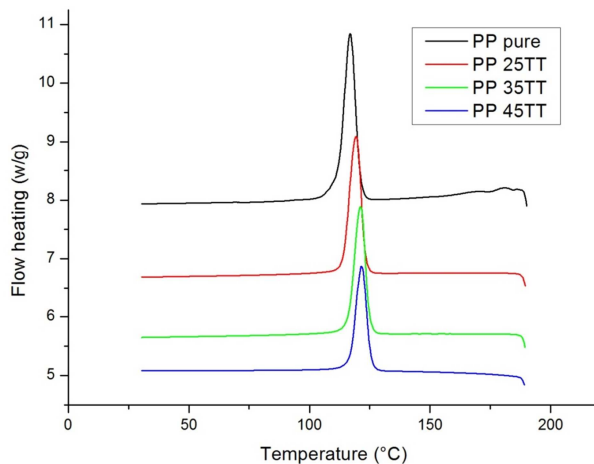

**Figure 6.** *DSC cooling thermograms of neat PP and its composites containing different rates of typha stem.*

3.4. Rheological Characterisation

Rheological characterisation is considered to be a crucial feature that allows the degree of dispersion of samples to be distinguished [29]. Based on a dynamic frequency sweep test, rheological characterisation was performed at a temperature of 190°C and a frequency of 0.05-100 rad/s. The linear viscoelastic behaviour of pure PP and all formulated composites was evaluated by oscillatory measurements.

In order to further investigate the addition of Typha fibres on the rheological and structural behaviour of the materials, the Cole-Cole diagram was used (Figure 7). In this diagram, the imaginary viscosity component (η'') is plotted against the real viscosity component (η'). The graph should look like a semicircle if the system describes a single relaxation. Due to the heterogeneous character of our fibre/matrix blends containing agglomerated fibres, the semicircle shape of the Cole-Cole graph is modified, the elastic component of the viscosity, and the relaxation time increases [30]. Cole-Cole graphs of pure PP revealed a semicircle shape related to a single relaxation time. On the other hand, the addition of Typha fibres generated an increase in the elastic behaviour and relaxation time of the structure, visualised by increases in the values of the viscosity components. This behaviour indicates the presence of agglomerated fibres and decreases progressively with increasing shear rate.
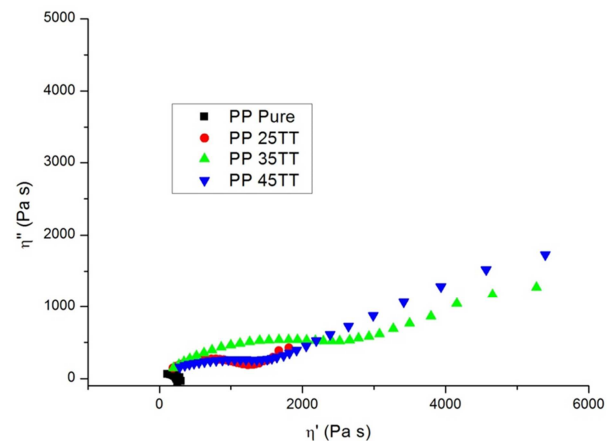
Figure 7. *Cole-cole diagram of pure PP and its composites at T = 190°C.*
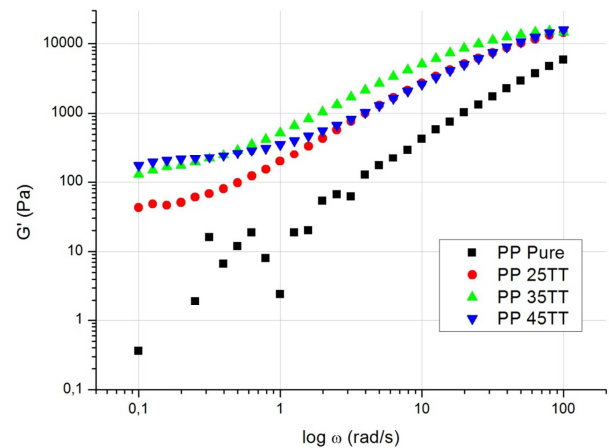
**Figure 8.** *Frequency variations of storage modules (G ') for pure PP and its composites at T = 190°C.*
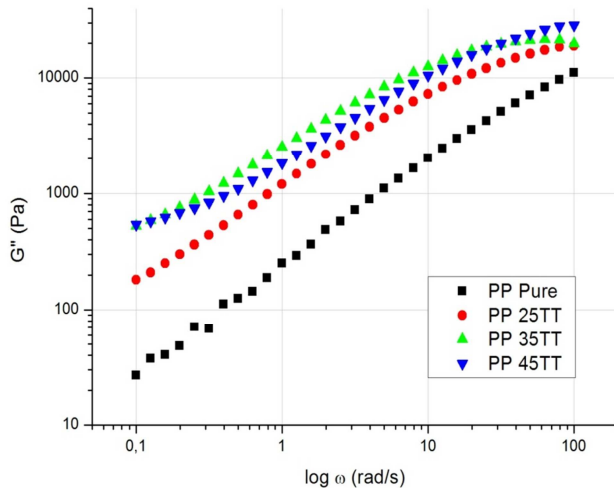
Figure 9. *Frequency variations of the loss storage modules (G '') for pure PP and its composites at T = 190°C.*

Furthermore, it is observed that the G' and G" values of pure PP and biocomposites increase with the rate of reinforcements. This increase has been observed previously and could be related to agglomeration phenomena of typha fibres which reduces the sliding or flow between them inside the biocomposite. In fact, at low frequencies, the viscosity increases strongly with the Typha powder content, as evidenced by the storage modulus of the composites (G') (Figure 9) since G' is higher than the pure matrix due to the intrinsic stiffness of the Typha fiber. Shear thinning behaviour of the melt was observed, the viscosity of the composites is strongly influenced by the shear rate. The storage modulus, especially for the highest powder content, shows that the matrix transfers a large amount of stress to the load and based on the data of G', serving as a measure of elastic behaviour, we can conclude that the composites have elastic behaviour [31]. The energy change that occurs during the shearing process is attributed to the variations of the dynamic moduli (G′ and G″), which is strongly related to the synergy of the interphases of the matrices in the blends (Figures 9 and 10). In addition to this, it should be noted that the shape of the filler, the concentration and density of the filler and the degree of interaction of the polymer and the typha fiber can influence the rheological characteristics of the polypropylene, which is in accordance with the work of several authors [32-34].

3.5. Mechanical Properties of Biocomposites PP/Typha Stem

Figures 8 and 9 show the tensile modulus and ultimate tensile strength. In Figure 8, the Young's modulus of biocomposites containing 25% to 45% TT is increased compared to virgin polymer. The tensile modulus of the samples varies between 1268 MPa to 1480 Mpa, which is accompanied by a transfer of stress from the matrix to the fibres depending on the amount of wood incorporated. It has been shown that the increase in wood leads to an increase in the crystallinity content of the composites during cooling.

The crystallites have much higher tensile moduli than the amorphous parts and can increase the contribution of the polymer matrix to the modulus of the composite [35].
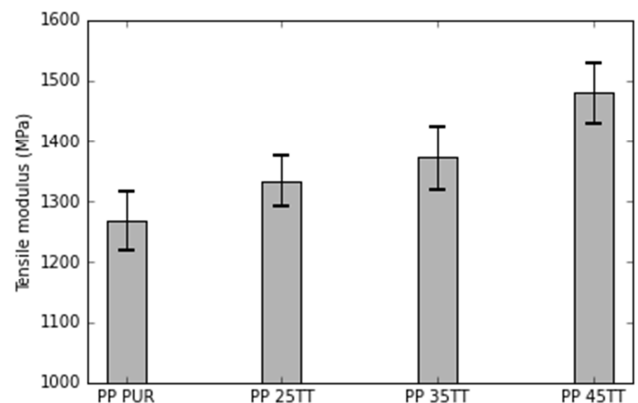
Figure 10. *The tensile modulus as a function of TSP concentration.*
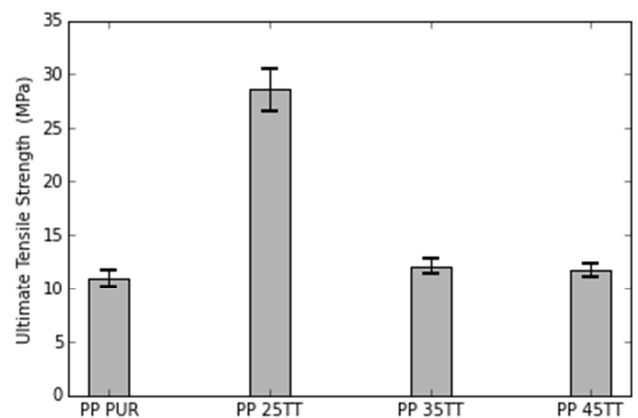
Figure 11. *The Ultimate Tensile Strength as a function of TSP concentration.*

The increase in tensile strength up to this critical value can be attributed to the good interfacial adhesion between the matrix and the filler. The incorporation of filler can have a positive effect, if there is interaction between the matrix and the Typha fibre, due to a fairly homogeneous dispersion of the fibre in the polymer matrix, these for values not exceeding 25%. The decrease in tensile strength (Figure 11) is strongly influenced by the presence of microcracks, pores, inclusions of brittle particles (internal defects) and by the presence of notches (macro cracks) resulting from manufacturing defects. These results observed for biocomposites beyond the weight loading are related to both the conditions of crack propagation in the biomaterials as a function of the applied forces. In the presence of Typha fibre and under the effect of the stresses, the microcracks become unstable and lead to failure when the applied force exceeds a critical value. This observation is in agreement with the authors [36].

4. Conclusion

The incorporation of typha fibre into polypropylene resulted in an increase in the tensile modulus of

biocomposites containing 25% to 45% TT compared to the virgin polymer. The tensile modulus of the samples varied between 1268 MPa to 1480 Mpa, which is accompanied by a transfer of stress from the matrix to the fibres depending on the amount of wood incorporated. However, the tensile strength increases up to this critical value, which can be attributed to the good interfacial adhesion between the matrix and the filler. The incorporation of filler can have a positive effect, if there is interaction between the matrix and the Typha fibre, due to a fairly homogeneous dispersion of the fibre in the polymer matrix, these for values not exceeding 25%.

The thermogravimetric analysis reveals that the addition of typha fibre decreases the thermal stability of the biocomposite, as the thermal stability of the fibres is lower than that of PP, the degradation of the typha fibre can accelerate the decomposition of the Polypropylene.

The DSC analysis shows a slight difference of about 3°C between the melting temperatures. The existence of a 3°C gap between the two melting temperatures has already been observed. This difference is probably due to the reorganisation in the molten state, to the variation of the crystalline dimensions or to the presence of crystallites of different stability. We note the appearance of a small shoulder in the biocomposite samples. typha acts as a nucleating agent. The SEM images show voids and holes resulting from the detachment of the fibres during the tensile test. The presence of broken wood particles, indicating failure in the wood and not at the interface, is also noted. This observation confirms the good mechanical strength of the fibre/matrix interface.

Finally the dynamic rheology shows the speudoplastic nature of the blends with shear thinning behaviour, it should be noted that the shape of the filler, the concentration and density of the filler and the degree of interaction of the polymer and the typha fibre can influence the rheological characteristics of the polypropylene.

5. Recommendation

The development of structural elements based on biocomposites could be carried out in order to make a comparative study with standard injection and extrusion processes. The focus will be on new methods in the process of manufacturing short and continuous fibres for which the procesś will have to be optimised. The instantaneous performances will be tested and validated in order to establish behaviour laws and thus enable the dimensioning of prototype parts for applications.

Conflicts of Interest

All the authors do not have any possible conflicts of interest.

Data Availability Statement

All the data (figures, tables, graphs) that support the results of this study are available from the corresponding author, Babacar NIANG, and will be given to you if necessary in the publication of this article.

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