

# Thermomechanical and Structural Analysis of Biocomposites and Gamma Irradiation and Photoaging on Mechanical and Viscoelastic Properties

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**Abstract:** The replacement of petrochemical fibres with natural fibres for reinforcing polyvinyl chloride matrix biomaterials is currently being researched and applied in the technical and technological fields. Natural fibres offer environmental advantages combined with economic advantages related to comparatively lower cost and lower energy consumption. It is in this context that typha stem fibres extracted from the typha plant found in south-saharan Africa are used in this study for the manufacture of biocomposites. The objective is to evaluate the thermogravimetric (TG/DTG) and viscoelastic behaviour of polyolefin matrix biocomposites reinforced with relatively high volume fractions, 25, 35 and 45%, of typha stem powder. The incorporation of typha stem powder slightly reduces the thermal stability of the biomaterials by decreasing the thermal degradation onset temperature and the DTG peak temperature compared to pure matrices. The limit for the practical application of these composites could be set at 270°C, before the onset of major weight loss. Monitoring of the different materials by rheological measurements during photoaging allowed to understand the mechanisms of photodegradation. The shear thinning behaviour observed on the complex viscosity points to possible small changes at the molecular level. Photochemical degradation over the photo-aging cycles resulted in cut-off and recombination phenomena. Dynamic storage moduli (G') and loss moduli (G'') tend to increase with the proportion of typha powder. The dynamic storage modulus (G') and loss modulus (G'') tend to increase with the proportion of typha powder. We observed a rheofluidic behaviour by shearing of the melt. The dynamic storage (G') and loss (G'') moduli tend to increase with the proportion of typha powder. From a thermal and rheological point of view, HDPE-based biocomposites show interesting properties for use in applications. The influence of gamma irradiation leads to a competition between two mechanisms (chain breaks and recombination) of photooxidation that take place together within the material. Mechanical properties such as tensile and flexural strength are improved with increasing gamma radiation dose up to 75 kGy.

**Keywords:** Thermogravimetric Analysis (TGA), Thermal Stability, Photoaging, Mechanical and Viscoelastic Properties

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## 1. Introduction

Today's growing population and pollution mean that the environmental situation in the world is rather unfavourable. This ever-increasing population requires more and more food, consumer goods and housing. All this leads to the growth of cities, industry and an increase in the volume of municipal solid waste. It is in this context that scientific research is stimulated with a view to designing and proposing new materials for the construction, packaging, furnishing and automotive industries. This collaboration between industry and the research community has led to satisfactory improvements in environmentally friendly materials in the field of polymer science through the development of biocomposites [1, 2]. The search for new materials, combining natural fibres with biodegradable or non-biodegradable polymers, appears to be a viable solution to current environmental problems. Natural fibres can provide reinforcement to composites and, in addition, the use of fibres derived from residues can minimise environmental pollution and reduce the costs of the materials produced. Natural fibre reinforced composites are gaining acceptance in structural applications due to their relatively low cost and recyclability. The introduction of natural fibres such as sisal, flax, jute, *grewiaoptiva*, *hibiscus sabdariffa* and pine needles into composite materials promises to produce new environmentally friendly materials [3, 4]. Natural fibres are biodegradable and renewable, and have low cost and low density [5, 6]. Natural fibres are also considered as biocomposites due to their components such as cell, lignin, starch and polylactic acid [7, 8]. Many studies have been conducted to investigate the suitability of natural fibres for use as reinforcing components. Many researchers [9, 10] have already investigated natural fibres as a replacement for glass fibres to improve the environmental performance of products. In general, the mechanical properties of the polymer matrix are improved with the use of natural fibres [11].

In the last decade, the thermoplastic composites industry has moved from the use of advanced high performance composites to cost effective engineering composites using different fibres. Polyethylene (PE) is one of the most widely used thermoplastics in the world due to its good usable properties such as toughness, near-zero moisture absorption, excellent chemical inertness, low coefficient of friction, ease of processing and low electrical conductivity [12]. Polyethylene is used in many applications such as pipes, sheets, containers and other products. The mechanical and physical properties of PE are highly dependent on variables such as the extent and type of branching, crystal structure and molecular weight.

New composites that include PE as a matrix are widely used in many applications with better mechanical and physical properties compared to the polymer alone. Polyethylene composites can be used in packaging, electrical, thermal energy storage, automotive, biomedical and space applications [13].

Much work has been done to develop PE composites using various additives. Improving the interfacial bond strength is one of the main challenges facing scientists, especially for the addition of natural fibres. High-density polyethylene consists of long chains, without major branching, with less than one side chain per 200 carbon atoms in the main chain. The long linear chains result in regular packing and high crystallinity due to the stereoregularity of PE molecules. High density polyethylene is more rigid, due to its higher crystallinity. High density polyethylene is used in many products and packaging such as milk jugs, detergent bottles, margarine tubs, rubbish containers and water pipes [14]. PP is a commonly used polymer and is widely used in a variety of applications due to its non-toxicity, low cost, low density and ease of processing. In addition, it has good mechanical properties and chemical stability [15], but the main drawback of this polymer is that it retains its non-renewable origin. Blending polyolefins with plant fibres has been reported as an effective way to reduce production costs [16] while maintaining or increasing matrix properties [17]. Interestingly, polypropylene composites reinforced with plant fibres have been shown to have good recycling properties [18]. In the manufacture of composites various natural fibres have been used as reinforcement, however, there are other types of fibres in the biomass that are not yet known in the field.

In the Saint Louis region of Senegal, typha is one of the most abundant fibres and is used in the manufacture of bags, mats, handicrafts and in the construction sector. However, the fibre can also be used as a structural reinforcement for polymer matrix composites.

In this paper, the incorporation of plant species (Typha) into the polypropylene matrix and high density polyethylene to develop composite biomaterials is discussed. The comparison of viscoelastic properties, crystallization kinetics and thermal stability will be analysed. The relationship between the structure of the biomaterials and their properties will be discussed in this review article.

## 2. Experimentations

### 2.1. Materials

HDPE was supplied in pellet form by Nova Chemicals. Its melt index was 7.0 g/10 min at 190°C and its density at room temperature was 0.962 g/cm<sup>3</sup>. While polypropylene (PP) was generously provided by Solvay Co (International Chemical and Pharmaceutical Group). The latter has a density of 0.900g/cm<sup>3</sup> and a melt index of 2.5g/10 min.

### 2.2. Composite Manufacturing Methods

Thus, prior to any mixing operation, the wood powder was oven dried at 105°C for 24 hours to remove moisture below 1%. No other treatment took place on the polymer before its use. The composite materials were obtained by mixing in a 7 cm<sup>3</sup> co-rotating twin screw minilab mixer. For the extraction of noxious gases, the fume hood was opened. In a first step,

the polymer was introduced for 5 min at 180°C and the screw speed was set at 100 rpm. In a second step, the typha stem powder is introduced. The system is mixed for 10 min at 180°C at 100 rpm. After this phase, each sample is sandwiched for 1 min between two Teflon sheets and pressed at 175°C and 180°C under a pressure of 200 bar to obtain films of 100 to 200 microns thickness for spectroscopic analyses, or of a few millimetres thickness for the study of different macroscopic properties. With the polymer matrix (HDPE), four formulations were produced with different contents of typha stem powder at the laboratory of the Institut de Chimie de Clermont-Ferrand (ICCF) in France (Table 1). The temperature profile in the machine was kept below 180°C to minimise thermal degradation of the powder.

**Table 1.** Composition and code of biocomposite samples.

(1- a),

Echantillons	PEHD	Typha
PEHD 25TT	100	0
PEHD 25TT	75	25
PEHD 35TT	65	35
PEHD 45TT	55	45

(1- b),

Echantillons	PP	Typha
PP PUR	100	0
PP 25 TT	75	25
PP 35 TT	65	35
PP 45 TT	55	45

(a), (b) manufactured by twin-screw extrusion (University of Clermont Auvergne, France).

### 2.3. Characterization of Composites

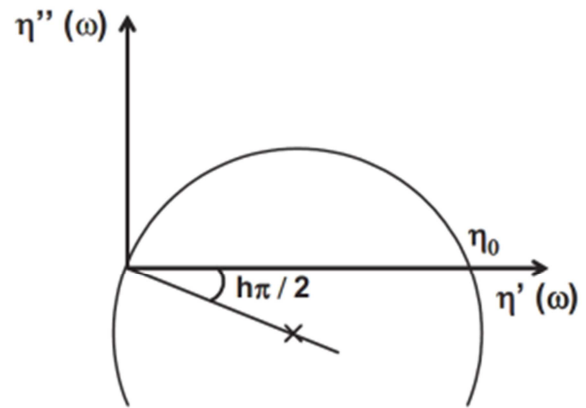
#### 2.3.1. Thermogravimetric Analysis (TGA)

To study the thermal degradation process of biocomposites, a TGA analysis was performed. A Mettler Toledo machine operating under a nitrogen atmosphere (20 ml/min) has been used for this purpose. The samples were analyzed at 30°C - 600°C with a heating rate of 10°C/min and a mass of  $\approx 20$  mg was used for each sample.

#### 2.3.2. Rheological Characterization

The viscoelastic behaviour of molten polymers can be determined using oscillatory rheological experiments such as dynamic mechanical tests, which provide a convenient means of assessing the frequency dependence of polymer mechanical properties. A useful representation is to plot the data points of the experimental frequency sweep in the complex plane. This means that the values of the imaginary part (of the complex viscosity)  $\eta''$  are plotted along the abscissa (X-axis) and the real values ( $\eta'$ ) on the ordinate (Y-axis) as schematised in Figure 1; where  $\eta'$  is the loss viscosity,  $\eta''$  is the storage viscosity and  $\omega$  is the frequency pulsation:  $\omega = 2\pi N$ . Usually, the experimental points are located on an arc of a circle characteristic of a Cole distribution. Extrapolation of this arc to the zero ordinate value gives the Newtonian viscosity which is related to the average molecular weight  $M_w$  of the polymer considered by

a power law: ( $\eta_0 = K (M_w)^{3,4}$ ) [18, 19]. Melt rheological measurements of pure HDPE, PP PUR and their composites were performed with an ARES Rheometer Scientific mechanical spectrometer in oscillatory frequency sweep mode with a parallel plate measuring cell. The diameter of the plates was 8 mm and the gap was 1 to 1.5 mm. The frequency was between 0.1 rad/s and 100 rad/s. The strain was kept constant at 5% over the entire frequency range to ensure linearity. This strain was selected from a dynamic strain sweep test, in which, within a range of 1% to 10% of strains, at a fixed frequency of 10 rad/s, the drift deviation from linearity was monitored; the frequency sweep test was then performed at a constant temperature of 180°C.



**Figure 1.** Cole-cole schema.

#### 2.3.3. Mechanical Properties

The apparatus used is a Lloyd EZ 50 traction bench. The results are analyzed using the software: NEXYGEN. The tensile strength and elongation at break of the composite were measured according to the ASTM D638-10 method at a crosshead speed of 30 mm/min. Each sample was subjected to at least 5 repetitive tests to obtain an average.

#### 2.3.4. Irradiation

The biomaterials and virgin HDPE were irradiated using a Co-60 gamma source. For gamma irradiation, a Co-60 (25kCi) model 650 source is loaded with the GBS-98 source which consists of 36 double encapsulated capsules. The type C-252 loaded with Co-60 pellets was used. Biomaterials were pre-treated with gamma radiation at different doses (5-100kGy).

## 3. Results and Discussion

### 3.1. Thermal Analysis

The thermal degradation of pure polypropylene and their bio-composites reinforced with typha fibres was analysed in detail by performing thermogravimetric analyses. Figure 3a shows the thermogravimetric (TG) and differential thermogravimetric (DTG) curves of each natural fibre at a heating rate of 10K / min. The TG / DTG curves in an air atmosphere were performed to understand the thermal degradation behaviour of typha fibres, polymers and

biocomposites with 10, 20 and 30% typha fibres (Figure 3a). The mass loss up to 220°C was attributed to the evaporation of water, glycerol and other volatile compounds. Thermal degradation of typha and hemicelluloses occurs between 250 and 300°C [20], while cellulose is thermally degraded between 240 and 350°C.

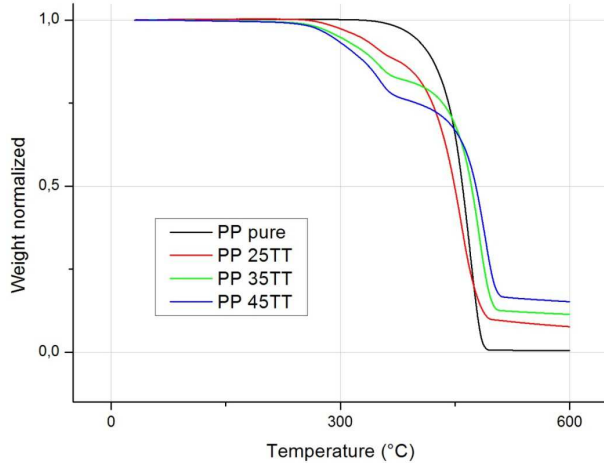


Figure 2. ATG thermograms of pure PP and its composites.

However, for biocomposites, according to Figure 2, the decomposition of PP starts at about 400°C (breakage of the CC chains of the main chain) [21, 22], which is a higher breakage temperature than that of the fibres [23, 20]. Pure PP has a one-step decomposition process from 400°C to 500°C. While composites clearly show a two-step decomposition process. The first step corresponds to the decomposition of the fibre and the second step to the decomposition of the matrix. The addition of fibres therefore reduces the stability of the polypropylene in the composite. On the other hand, since typha fibres are characterised by different particle sizes and geometries, small fibres seem to slightly decrease the thermal stability compared to larger fibres. This is because the amount of interface with the fibres is important for small fibres.

When the small fibres start to form (before the PP), the surface area of the PP that can start to degrade is then higher and decomposition is accelerated so that it takes place at a lower temperature. In general, 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, so the degradation of typha fibre can accelerate the decomposition of Polypropylene [24, 25].

The DTG curves of biocomposites show two zones. The first zone is characterised by a small variation in mass loss. The second zone of characteristic mass loss is between 400°C and 500°C. In its temperature range, the initial samples (reduced to 100%) lose about 80% of their initial masses. In these zones, if we examine the DTG spectrum, we observe a peak with a maximum noted at about 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 [26, 27].

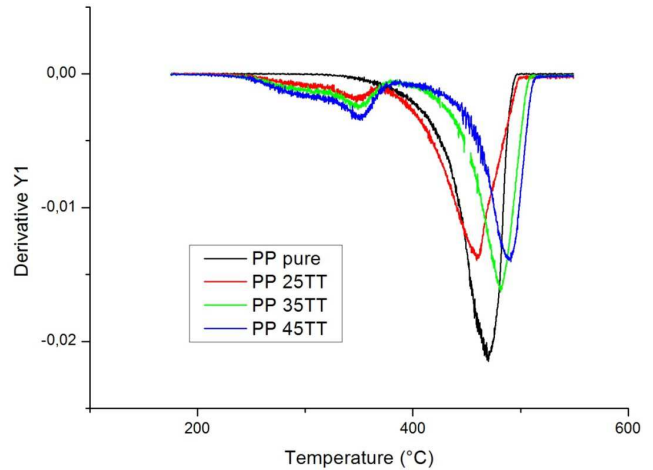


Figure 3. DTG thermograms of pure PP and its composites.

However, for HDPE/Typha stem biocomposites, thermogravimetric analysis allows the variation of the mass of a sample to be followed as a function of temperature, and thus to access the decomposition parameters of a material. It is convenient to plot the derived curve (DTG) of the TGA, which allows easy identification of the characteristic temperatures related to each mass loss phenomenon.

Figures 4 and 5 show the ATG and DTG thermogravimetric behaviour of high density polyethylene (HDPE), Typha fibres and composites (HDPE/Typha).

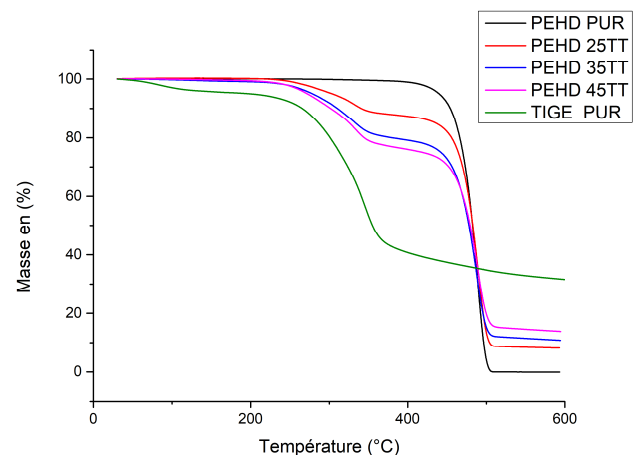


Figure 4. ATG thermograms of pure HDPE and its composites.

Many studies [28-30] on the thermal degradation of polyolefins show that above 500°C the polymer chains are completely degraded. In the case presented, it is observed that the decomposition starts at around 370°C up to 410°C. The decomposition is completely completed above 500°C.

For typha fibres, three regions appear in the mass loss thermograms (Figure 4):

1. 57-100°C corresponding to dehydration.
2. 210-400°C identified as matrix degradation (HDPE).
3. > 400°C corresponding to the degradation of the plant fibre (typha).

In a lignocellulosic material subjected to a temperature rise programme, hemicelluloses generally decompose first at

temperatures between 150°C and 350°C, followed by cellulose by random cleavage of these glycosidic bonds between 280°C and 350°C and lignin decomposition taking place between 250°C and 500°C [31, 32]. These reactions take place mainly in the amorphous region of the plant fibres and continue to occur until the degree of polymerisation becomes constant [33].

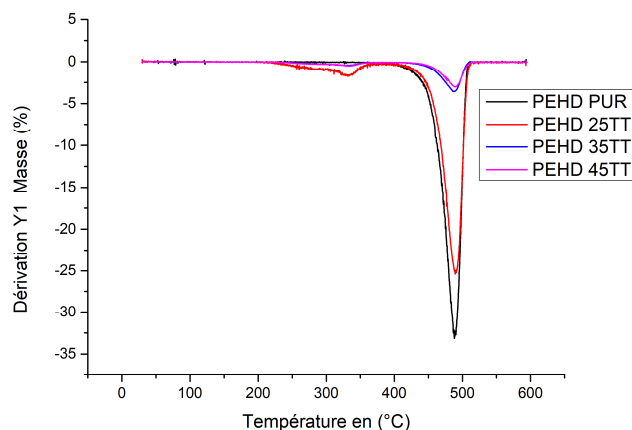


Figure 5. DTG decomposition curves for pure HDPE and its composites.

Also due to the high flammability of cellulose, the addition of wood makes the composite less thermally stable. It has been observed that its molecular weight decreases sharply due to the degradation of its macromolecular chains as confirmed by this work [34, 35].

The degradation mechanism of wood/HDPE composites has not been sufficiently understood. However, possible reasons for the increased thermal stability of our blends can be explained by the fact that the amorphous polymer phase is initially protected by the surrounding polymer chains. The degradation temperature range (about 450°C) of this polymer (HDPE) is shifted to lower temperatures.

Figure 5 shows the same result as Figure 4 in its derived form (DTG). A two-stage thermal degradation is observed. The temperature ranges of the first stage correspond as mentioned before to a dehydration of the wood, a (loss of bound water). The temperature at the start of dehydration is taken as a measure of the thermal stability of CBP [26, 36, 37]. The loss of mass as a function of time or temperature is an irreversible process due in part to thermal degradation.

The composites clearly show a two-stage process. The mass loss in the first stages is 8%, 11% and 16.7% respectively for the 25%, 35% and 45% typha composites. The second stages correspond to the decomposition of the polymer matrix chains. The sample loses a total of 92% of its mass around 600°C. From the curve (Figure 4), an initial degradation temperature of 210°C is observed. The mass loss information obtained by thermogravimetric analysis often does not allow the specification of the types of decomposition products. This is only possible if the ATG device is coupled to an infrared spectrometer or a mass spectrometer.

From the observations, it appears that the incorporation

of typha decreases the overall thermal stability of HDPE. This may be due to the fact that the extrusion process favours the splitting of polymer chains, thus increasing the fragility of the composite materials. The most likely hypothesis is the reaction of fibre degradation products on the matrix.

### 3.2. Rheological Characterization

Rheological characterisation is considered a crucial feature that allows distinguishing the degree of dispersion of samples [38]. Based on a dynamic frequency sweep test, the rheological characterisation was carried out 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 assessed 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 6). In this diagram, the imaginary viscosity component ( $\eta''$ ) is plotted against the real viscosity component ( $\eta'$ ). The graph should look like a semicircle if the system describes a single relaxation. Due to the heterogeneous nature 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 [39]. Cole-Cole graphs of pure PP revealed a semicircle shape related to a single relaxation time. In contrast, 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.

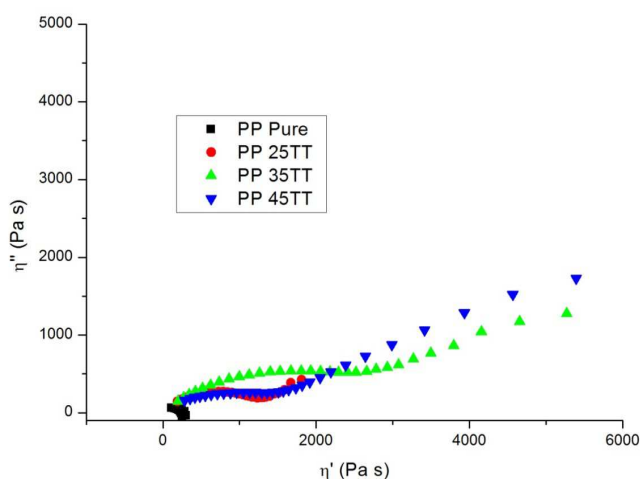
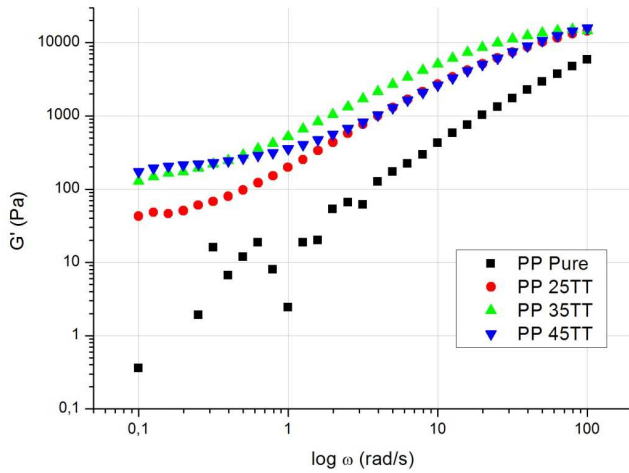


Figure 6. Cole-cole diagram of pure PP and its composites at  $T = 190^\circ\text{C}$ .

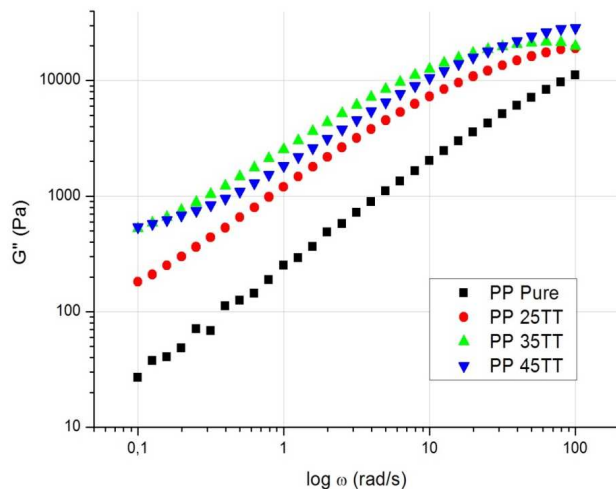
Furthermore, it is observed that the  $G'$  and  $G''$  values of pure PP and biocomposites increase with the rate of reinforcements. This increase has been observed previously and could be related to agglomeration phenomena of Typha fibres that reduce the sliding or flow between them inside the biocomposite. In fact, at low frequencies, the viscosity



increases strongly with the Typha powder content, as shown by the storage modulus of the composites ( $G'$ ) (Figure 7) since  $G'$  is higher than the pure matrix due to the intrinsic stiffness of the Typha fibre. 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 the elastic behaviour, we can conclude that the composites have an elastic behaviour [40]. 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 7 and 8). 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 fibre can influence the rheological characteristics of polypropylene, which is in agreement with the work of several authors [41, 42].



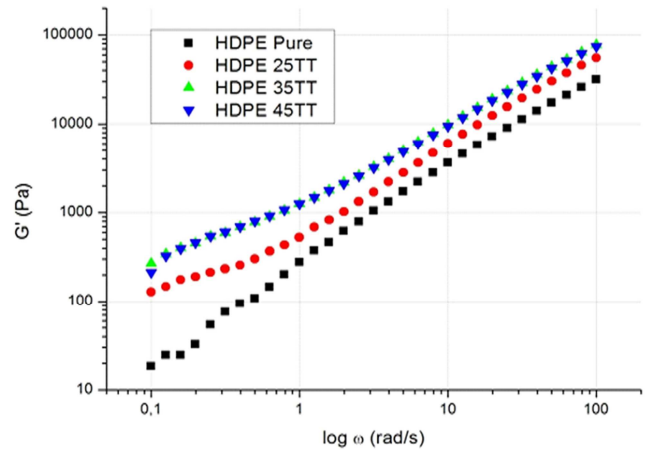
**Figure 7.** Frequency variations of storage modulus ( $G'$ ) for pure PP and its composites at  $T = 190^\circ\text{C}$ .



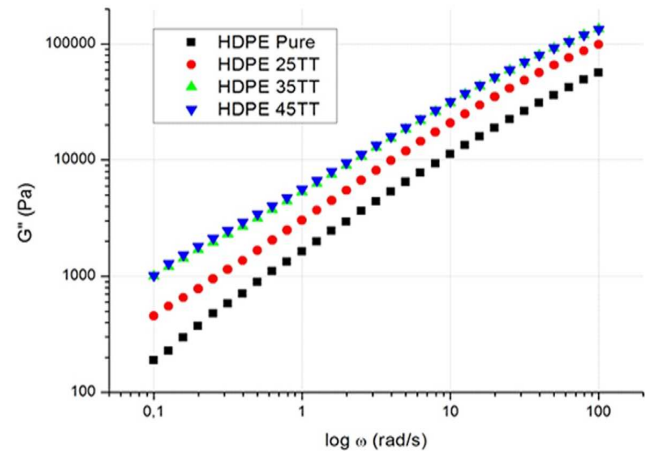
**Figure 8.** Frequency variations of the loss storage modulus ( $G''$ ) for pure PP and its composites at  $T = 190^\circ\text{C}$ .

### 3.3. Rheological Study of HDPE/Typha Composites

The rheological properties of the samples in the molten state were determined by dynamic mechanical measurements. The storage modulus  $G'$ , the loss modulus  $G''$  and the dynamic viscosity  $\eta^*$  were measured as a function of swept frequencies between 0.1 and 100 rad/s. Figures 9, 10 and 11 show the viscoelastic behaviour of the HDPE/TYPHA materials via the dynamic moduli ( $G'$ ) and ( $G''$ ) and the complex viscosity.

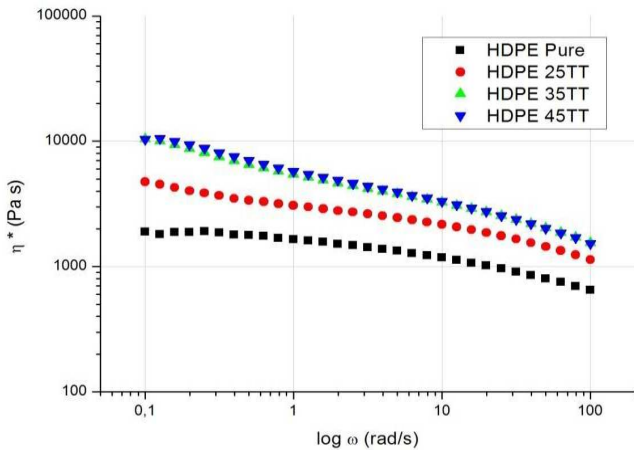


**Figure 9.** The variation of dynamic modulus ( $G'$ ) as a function of frequency with different percentages of typha stem.



**Figure 10.** The variation of the loss modulus ( $G''$ ) as a function of frequency with different percentages of typha stem.

The dynamic storage ( $G'$ ) and loss moduli ( $G''$ ) tend to increase with the proportion of typha powder (Figure 9 and 10). This results in a more elastic behaviour for high wood masses. Obviously, the complex viscosity of the pure polymer increases progressively with the increase of the powder content from 25% to 45% (figure 11). This is because in filled systems, the wood particles disturb the flow of the pure polymer and hinder the mobility of the chain segments in the direction of flow. These particles often occur in the form of aggregates.



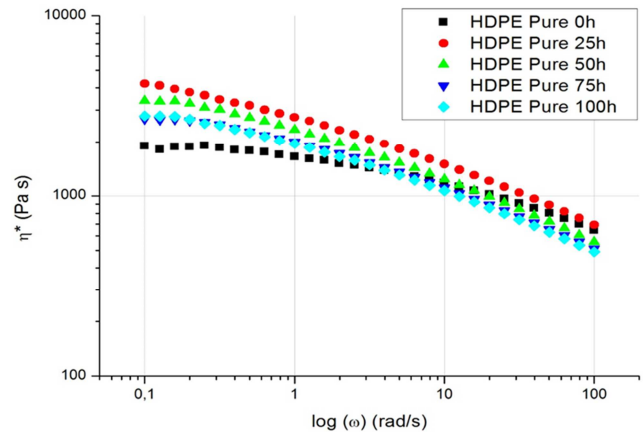
**Figure 11.** Variation of complex viscosity as a function of frequency of pure HDPE and its composites at  $T = 180^{\circ}\text{C}$ .

This is also due to the inhomogeneous dispersion and alignment of the cellulose phase in the composites. These observations are confirmed by those of other authors [32, 43, 44]. Moreover, the moduli  $G'$  and  $G''$  tend to have the same values above 35% (Figures 9 and 10). We observed a rheofluidic behaviour by shearing of the melt. The viscosity of the composites is strongly influenced by the shear rate. The composites show approximately the same viscosity above 35%. This is probably due to the alignment of the fibres at high shear rates. This reduces collisions between particles [33, 45]. In effect, the fluidity of the mixture is increased by increasing the oscillation frequency, which decreases the complex viscosity. This reduction in viscosity demonstrates the pseudoplastic nature of the materials in the molten state.

In the low frequency range, the viscosity increases significantly with increasing fibre content in the composite material (Figure 11). The addition of fibres affects the stiffness, density and viscoelastic behaviour of a polymer [46-48]. The highest values of complex viscosity were observed with the composite containing 45% by weight of typha fibres (Figure 11). Furthermore, the presence of agglomerates (due to their high fibre content) causes flow resistance and an increase in the viscosity of the composites. This behaviour can be explained by a higher interaction rate between the polymer matrix and the content. This forces the mixture to have a higher shear stress and longer relaxation times to the flow of the composites [49]. The rheological properties of high density polyethylene show the same shear rheofluidising behaviour. The moduli of elasticity show an improvement in the dynamic behaviour of the composites.

In Figure 12 below we show the effect of photo-wetting on the rheological properties. It represents the complex viscosity of (HDPE 35 TT) films irradiated in SEPAP 12-24. We observe that with irradiation the viscosity of the biocomposite increases at low frequencies but with the increase of the oscillatory frequencies, the value of the viscosity decreases;

This decrease in viscosity is also due to the increase in exposure time.



**Figure 12.** Complex viscosity curve of (HDPE 35 TT) films irradiated in SEPAP 12-24.

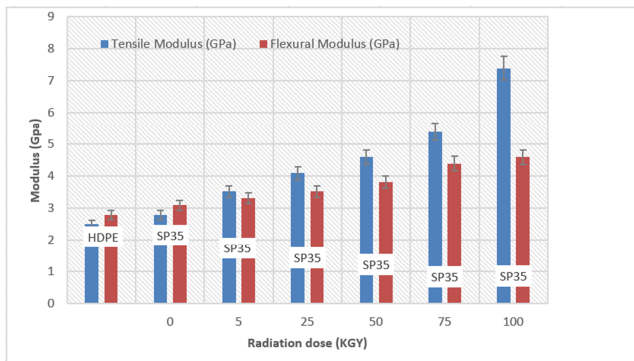
These behaviours observed during photodegradation reflect chain breaks in a first step and the formation of a cross-linking network due to chain recombination in a second step. It is interesting to observe that the two mechanisms (chain splitting and recombination) of photooxidation take place together within the material. Chain splitting predominates for a population that can then be assimilated to the HDPE chains in the biocomposite. The cross-linkages of this population are predominant up to 75h of irradiation, and then become a minority compared to the macromolecular chain splittings [50, 51]. Under irradiation, the lignin component of the plant fibre considered as well as the key structures involved in its degradation generate phenoxy radicals, and phenols are also oxidised. The influence of the presence of fibres in composites is expressed in the durability of the biocomposite by the modification of the kinetics of photooxidation. Indeed, the presence of fibres leads to an increase in the oxidation rate of the polymer matrix after the induction period. This effect is increasingly accentuated as the amount of fibre in the composite increases. This can be explained by the radical species produced by photooxidation [52]. Thus, the fibre acts as an initiator and/or accelerator of photooxidation [53].

### 3.4. Influence of Gamma Irradiation on Mechanical Properties of Biocomposites HDPE / Typha Stem

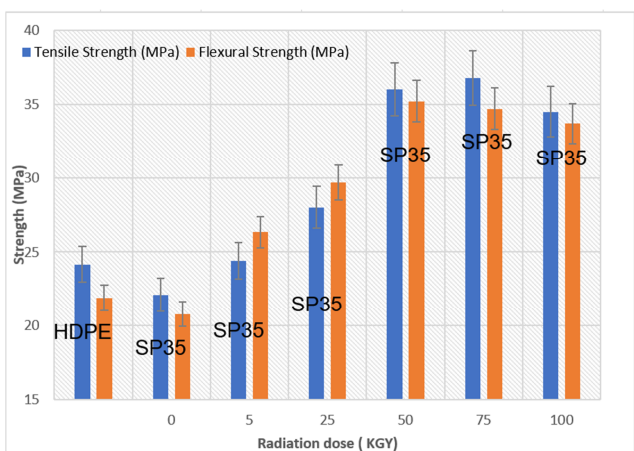
The effect of gamma irradiation on the mechanical properties of HDPE/Typha fibre composites in terms of Young's modulus, tensile strength and flexural strength are shown graphically in Figures 13 and 14. It was observed that the Young's modulus values of pure HDPE (Figure 5) increase with gamma dose from 5 to 75kGy.

The tensile and flexural strength of the composites increases from 5 to 75kGy. This could be due to the cross-linking effect of gamma radiation, as gamma radiation generates polarsites/free radicals in the HDPE chain in the form of carbonyl groups due to its oxidation [54]. Gamma irradiation also produces free radicals in the wood flour. These free radicals can react to change the chemical structure, which can be attributed to the formation of cross-

linking between the wood and HDPE [55] resulting in an increase in the tensile strength of the composite.



**Figure 13.** Tensile and flexural moduli of virgin HDPE and composites irradiated with gamma dose variations.



**Figure 14.** Tensile and flexural strength of pure HDPE and composites irradiated with varying gamma radiation doses.

This substantial increase in tensile and flexural strength could be due to competition between cross-linking and chain scission [56]. The decrease in tensile and flexural strength above 75 kGy could be due to the predominant chain-splitting effect of gamma radiation for HDPE at a higher dose. This decrease in strength at higher gamma dose could be explained by fibre agglomeration in the matrix, as high gamma dose also leads to increased fibre-fibre interaction [57].

## 4. Conclusion

The study of the physico-chemical, mechanical and thermal properties of the samples as well as the effects of chemical treatment on them was carried out in this paper. The incorporation of typha fibres with a reinforcing character leads to a loss of ductile behaviour but induces a very important gain in stiffness and mechanical strength. Thermogravimetric analysis showed that the incorporation of typha flour in the polymer matrix leads to a decrease in the temperature at which the composites start to decompose. Gamma irradiation leads to an improvement in the mechanical properties. The gamma dose is increased due to

the stress transfer of the polymer from the semi-crystalline to the amorphous structure. Mechanical properties such as tensile and flexural strength are improved with increasing gamma radiation dose up to 100 kGy. The melt viscoelastic behaviour of Typha fibre biocomposites shows the rheofluidizing phenomenon by melt shear. It was found that  $G'$  and  $G''$  increased with the increment of the filler content. These reflect a more elastic behaviour for high amounts of Typha.

## References

- [1] De Lemos, A. L., Mauss, C. J., & Santana, R. M. C. (2017). Characterization of natural fibers: wood, sugarcane and babassu for use in biocomposites. *Cellulose Chemistry and Technology*, 51 (7-8), 711-718.
- [2] Suoware, T. O., Edelugo, S. O., & Ezema, I. C. (2019). Impact of hybrid flame retardant on the flammability and thermomechanical properties of wood sawdust polymer composite panel. *Fire and Materials*, 43 (4), 335-343.
- [3] Thakur VK, Thakur MK, Gupta RK. Graft copolymers from cellulose: Synthesis, characterization and evaluation. *J Carbohydr polym* 2013; 97 (1): 18-25.
- [4] Singha AS, Thakur VK. Fabrication and Characterization of H. sabdariffa Fiber-Reinforced Green Polymer Composites. *J Polym plast technol eng* 2009; 48 (4): 482-487.
- [5] Singha AS, Thakur VK. Mechanical, Thermal and Morphological Properties of Grewia Optiva Fiber/Polymer Matrix Composites. *J Polym plast technol eng* 2009; 48 (2): 201-208.
- [6] Alves C, Ferrao P. M. C, Silva A. J, Reis L. G, Freitas M, Rodrigues L. B, Alves D. E. Ecodesign of automotive components making use of natural jute fiber composites. *J Clean Prod* 2010; (18): 313-327.
- [7] Ali R, Iannace S, Nicolais L. Effect of processing conditions on mechanical and viscoelastic properties of biocomposites. *J Appl Polym Sci* 2003; 88: 1637-42.
- [8] Garkhail SK, Meurs E, Van de Beld T, Peijs T. Thermoplastic composites based on biopolymers and natural fibres. ICCM12 (12th international conference on composite materials), Paris; 1999. p. 1175.
- [9] Puglia D, Biagiotti J, Kenny JM. A review on natural fiber based composites Part II: application of natural reinforcements in composite materials. *J Nat Fibers* 2004; 1 (3): 23-65.
- [10] Suddell B C, Evans W J, Isaac DH, Crosky A. A Survey into the Application of Natural Fiber Composites in the Automotive Industry. In: *International Symposium on Natural Polymers and Composites*, Sao Pedro, Brazil, 2002.
- [11] Yuan XW, Jayaraman K, Bhattacharyya D. Plasma treatment of sisal fibers and its effects on tensile strength and interfacial bonding. *J Adhes Sci and Technol* 2002; 16 (6): 703-27.
- [12] Klyosov A.: 'Wood-plastic composites'; 2007, John Wiley & Sons, Hoboken, New Jersey, Canada. [Crossref, Google Scholar].



- [13] Vasile C. and Pascu M.: 'Pascu: Practical guide to polyethylene'; 2005, iSmithers, Rapra Publishing Shawbury, Shrewsbury, UK. [Google Scholar].
- [14] Huang X., Ke Q., Kim C., Zhong H., Wei P., Wang G., Liu F. and Jiang P.: *Polym. Eng. Sci.*, 2007, 47, (7), 1052–1061. [Crossref, Web of Science®, Google Scholar]
- [15] Xu M, Shi X, Chen H, Xiao T. Synthesis and enrichment of a macromolecular surface modifier PP-b-PVP for polypropylene. *Appl Surf Sci* 2010; 256: 3240–4. doi: 10.1016/j.apsusc.2009.12.012.
- [16] Faruk O, Bledzki A., Fink H-P, Sain M. Biocomposites reinforced with natural fibers: 2000-2010. *Prog Polym Sci* 2012; 37: 1552–87.
- [17] Ausias G, Bourmaud A, Coroller G, Baley C. Study of the fibre morphology stability in polypropylene-flax composites. *Polym Degrad Stab* 2013; 98: 1216–24.
- [18] Bourmaud A, Baley C. Investigations on the recycling of hemp and sisal fibre reinforced polypropylene composites. *Polym Degrad Stab* 2007; 92: 1034–45.
- [19] Ndiaye, D., Diop, B., Thiandoume, C., Fall, P. A., Farota, A. K. and Tidjani, A. (2012) Morphology and thermo mechanical properties of wood/polypropylene composites. *Polypropylene*, 4, 730-738.
- [20] AlMaadeed, M. A., Kahraman, R., Khanam, P. N., & Madi, N. (2012). Date palm wood flour/glass fibre reinforced hybrid composites of recycled polypropylene: Mechanical and thermal properties. *Materials & Design*, 42, 289-294.
- [21] Awal, A., Ghosh, S., & Sain, M. (2010). Thermal properties and spectral characterization of wood pulp reinforced bio-composite fibers. *Journal of thermal analysis and calorimetry*, 99 (2), 695-701.
- [22] Li, X., Lei, B., Lin, Z., Huang, L., Tan, S., & Cai, X. (2014). The utilization of bamboo charcoal enhances wood plastic composites with excellent mechanical and thermal properties. *Materials & Design*, 53, 419-424.
- [23] Guo, Y., Zhu, S., Chen, Y., & Li, D. (2019). Thermal properties of wood-plastic composites with different compositions. *Materials*, 12 (6), 881.
- [24] Farhadinejad, Z., Ehsani, M., Khosravian, B., & Ebrahimi, G. (2012). Study of thermal properties of wood plastic composite reinforced with cellulose micro fibril and nano inorganic fiber filler. *European Journal of Wood and Wood Products*, 70 (6), 823-828.
- [25] Monteiro, S. N., Calado, V., Rodriguez, R. J., & Margem, F. M. (2012). Thermogravimetric stability of polymer composites reinforced with less common lignocellulosic fibers—an Overview. *Journal of Materials Research and Technology*, 1 (2), 117-126.
- [26] Keener T and Brown T. Epolene™ maleated polyethylene coupling agents. In: *Proceedings of seventh international conference on wood-fibre-plastic composites*; 2002; Wisconsin, USA.; 2002.
- [27] Matuana LM, Balatinecz JJ, Sodhi RNS, Park CB. Surface characterization of esterified cellulosic fibers by XPS and FTIR spectroscopy. *Wood Science and Technology* 2001; 35 (3): 191-201.
- [28] J. L. Thomason et J. L. Rudeiros-Fernández, «Thermal degradation behaviour of natural fibres at thermoplastic composite processing temperatures», *Polymer Degradation and Stability*, vol. 188, p. 109594, 2021.
- [29] P. Gañán et I. Mondragon, «Thermal and degradation behavior of fique fiber reinforced thermoplastic matrix composites», *Journal of Thermal Analysis and Calorimetry*, vol. 73, no 3, p. 783-795, 2003.
- [30] M. Sood et S. Singh, «Recent advancements in thermal properties and behavior of modified natural fiber composites», in *Hybrid Natural Fiber Composites*, Elsevier, 2021, p. 73-89.
- [31] N. V. Penava, V. Rek, et I. F. Houra, «Effect of EPDM as a compatibilizer on mechanical properties and morphology of PP/LDPE blends», *Journal of Elastomers & Plastics*, vol. 45, no 4, p. 391-403, 2013.
- [32] H.-S. Kim, H.-S. Yang, H.-J. Kim, et H.-J. Park, «Thermogravimetric analysis of rice husk flour filled thermoplastic polymer composites», *Journal of thermal analysis and calorimetry*, vol. 76, no 2, p. 395-404, 2004.
- [33] B. Wielage, T. Lampke, G. Marx, K. Nestler, et D. Starke, «Thermogravimetric and differential scanning calorimetric analysis of natural fibres and polypropylene», *Thermochimica Acta*, vol. 337, no 1-2, p. 169-177, 1999.
- [34] A. M. Badji, D. Ndiaye, A. K. Diallo, N. Kebe, et V. Verney, «The effect of poly-ethylene-co-glycidyl methacrylate efficiency and clay platelets on thermal and rheological properties of wood polyethylene composites», *Advances in Chemical Engineering and Science*, vol. 6, no 4, p. 436-455, 2016.
- [35] D. Ndiaye, A. M. Badji, et A. Tidjani, «Physical changes associated with gamma doses on wood/polypropylene composites», *Journal of composite materials*, vol. 48, no 25, p. 3063-3071, 2014.
- [36] F. Michaud, «Rhéologie de panneaux composites bois/thermoplastiques sous chargement thermomécanique: aptitude au postformage», 2003.
- [37] S. Mathurosemontri, K. Okuno, Y. Ogura, S. Thumsorn, et H. Hamada, «Investigation on Fracture Behavior of Glass Fiber Reinforced Thermoplastic and Thermosetting Composites», in *Key Engineering Materials*, 2017, vol. 728, p. 235-239.
- [38] P. Takkalkar, M. Ganapathi, C. Dekiwadia, S. Nizamuddin, G. Griffin, N. Kao, Preparation of square-shaped starch nanocrystals/polylactic acid based bio- nanocomposites: morphological, structural, thermal and rheological properties, *Waste Biomass Valoriz.* (2018) 1–15.
- [39] Osswald, T.; Rudolph, N. *Polymer Rheology. Fundamentals and Applications*; Hanser Publications, Munich 2015, ISBN 9781569905173.
- [40] Ma, P., Jiang, L., Ye, T., Dong, W. and Chen, M. (2014) Melt Free-Radical Grafting of Maleic Anhydride onto Biodegradable Poly (Lactic Acid) by Using Styrene as a Comonomer. *Polymers*, 6, 1528-1543.
- [41] Mazzanti, V., & Mollica, F. (2020). A review of wood polymer composites rheology and its implications for processing. *Polymers*, 12 (10), 2304.

- [42] Ogah, A. O., Afiukwa, J. N., & Nduji, A. A. (2014). Characterization and comparison of rheological properties of agro fiber filled high-density polyethylene bio-composites. *Open Journal of Polymer Chemistry*, 2014.
- [43] S. Mohanty, S. K. Verma, et S. K. Nayak, «Rheological characterization of PP/jute composite melts», *Journal of applied polymer science*, vol. 99, no 4, p. 1476-1484, 2006.
- [44] C. Zhao, H. Qin, F. Gong, M. Feng, S. Zhang, et M. Yang, «Mechanical, thermal and flammability properties of polyethylene/clay nanocomposites», *Polymer Degradation and Stability*, vol. 87, no 1, p. 183-189, 2005.
- [45] K. J. Wilczynski, «Determination of viscosity curves of wood polymer composites based on limited rheological measurements», *Polimery*, vol. 63, no 3, p. 213-218, 2018.
- [46] T. Yokohara, S. Nobukawa, et M. Yamaguchi, «Rheological properties of polymer composites with flexible fine fibers», *Journal of Rheology*, vol. 55, no 6, p. 1205-1218, 2011.
- [47] N. Sombatsompop et R. Dangtangee, «Effects of the actual diameters and diameter ratios of barrels and dies on the elastic swell and entrance pressure drop of natural rubber in capillary die flow», *Journal of applied polymer science*, vol. 86, no 7, p. 1762-1772, 2002.
- [48] H. Qin et al., «Thermal stability and flammability of polypropylene/montmorillonite composites», *Polymer Degradation and Stability*, vol. 85, no 2, p. 807-813, 2004.
- [49] N. E. Marcovich, M. M. Reboredo, J. Kenny, et M. I. Aranguren, «Rheology of particle suspensions in viscoelastic media. Wood flour-polypropylene melt», *Rheologica Acta*, vol. 43, no 3, p. 293-303, 2004.
- [50] S. Gaudin, F. Fraisse, V. Verney, S. Commerceuc, R. Guyonnet, et A. Govin, «Etude rhéologique de nouveaux biocomposites bois - polymères biodégradables», p. 5.
- [51] J. Tian, R. Zhang, Y. Wu, et P. Xue, «Additive manufacturing of wood flour/polyhydroxyalkanoates (PHA) fully bio-based composites based on micro-screw extrusion system», *Materials & Design*, vol. 199, p. 109418, 2021.
- [52] Ndiaye, D., Badji, A. M., & Tidjani, A. (2014). Physical changes associated with gamma doses on wood/polypropylene composites. *Journal of composite materials*, 48 (25), 3063-3071.
- [53] D. Ndiaye, «Contribution à l'étude de la caractérisation, des propriétés physico-mécaniques et du processus de photo vieillissement des composites bois polymères.», 2012.
- [54] Motaleb, K. Z. M., Milašius, R., & Ahad, A. (2020). Influence of gamma radiation on mechanical properties of jute fabric-reinforced polymer composites. *Fibers*, 8 (9), 58.
- [55] Rahman, H., Alimuzzaman, S., Sayeed, M. M., & Khan, R. A. (2019). Effect of gamma radiation on mechanical properties of pineapple leaf fiber (PALF)-reinforced low-density polyethylene (LDPE) composites. *International Journal of Plastics Technology*, 23 (2), 229-238.
- [56] Islam, J. M., Hossan, M. A., Alom, F. R., Khan, M. I. H., & Khan, M. A. (2017). Extraction and characterization of crystalline cellulose from jute fiber and application as reinforcement in biocomposite: Effect of gamma radiation. *Journal of Composite Materials*, 51 (1), 31-38.
- [57] Zaman, H. U., & Beg, M. D. H. (2015). Improvement of physico-mechanical, thermomechanical, thermal and degradation properties of PCL/gelatin biocomposites: Effect of gamma radiation. *Radiation Physics and Chemistry*, 109, 73-82.