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EXPERIMENTAL STUDY AND SIMULATION BY METAHEURISTIC ALGORITHM OF THE DAMAGE OF BIO-COMPOSITE MATERIALS

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DÉDICACE

Je dédie cette modeste thèse : A mes très chers parents.

Aucun hommage ne pourrait être à la hauteur de l’amour dont ils ne cessent de me combler.

Que dieu leur procure bonne santé et longue vie.

À mes très chers sœurs et leurs époux ainsi que ma sœur cadette, mon frère et ma grande mère que dieu la garde.

À mes neveux et nièces.

À mes amis,

À toute ma famille.

À tous ceux qui ont contribué de près ou de loin pour que ce projet soit possible,

Je vous dis merci.

Abstract

This present work is focused on the elaboration of different types of bio-composites by recycling the waste plastic bottle caps made of high-density polyethylene (HD-PE) and using it as a matrix reinforced with Alfa fibers and Pozzolan powder, followed by a mechanical and morphological characterization. Furthermore, a numerical investigation was pursued to develop an analytical model based on a genetic approach, using the Weibull model, to study the effect of damage to the fibre-matrix interface of four biocomposite materials based on polylactic acid (PLA) reinforced with three types of natural fibres raffia, alfa, and sisal fibres.

The experimental results are in good agreement with the numerical investigation. indicate that natural fibers-based composite materials can represent a good potential in improving interfacial properties. The study shows an alternative for producing friendly biodegradable composites that can be a solution for plastic waste reduction and a substitute for petrochemical-derived products.

Keywords: Bio-composites, high-density polyethylene, Recycling, Alfa , Pozzolan, Genetic approach, Weibull model, polylactic acid, raffia, sisal.

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General Introduction

Over the last forty years, composite materials have been the dominant interest of many industrial sectors, and their domains of application have been growing. The adaptation ability of these modern materials, which comes with a suitable mixture of its interfacial constituents, can fulfill the needs of several technologies sectors linked with the automobile, electronics, biomedical, construction, and aerospace among other industries [12, 35].

A composite material is typically constituted by the combination of at least two immiscible phases with different materials. The first phase is the matrix which represents the continuous component and the second phase is the filler and which is the discontinued part [62, 150].

To have a composite material with improved features, the specific properties of each component must be combined in a synergetic way, which means an efficient interfacial structure. The composite performances are related directly to their constituent proportions, their dispersal, orientation, the morphology of each one, and their thermodynamic properties [95, 150].

There are various criteria to classify composites like their composition, their structure, or their proprieties. According to the matrix nature, there are commonly three groups of composites: The metal matrix composite (MMC), like iron; aluminum, or copper, this composites has in general good elastoplastic behavior; stiffness; yield strength; and can withstand temperatures to 1000°C.

The organic matrix composite (PMC), like organic polymers; thermosetting and thermoplastic resin, has Visco-elastoplastic behavior, it can be used in applications that do not pass the 200°C.

The ceramic matrix composite (CMC), can be used in applications with large temperatures.

Whereas we can classify composites by the filler form, fiber-reinforced composite (FRC) exists in short or long-form, and particle-reinforced composites (PRC) with a size that varies from 20 to 100 micrometers [14, 150].

The ability to select materials that are optimal for a visualized application is based on the knowledge of multiple parameters such as [14, 48],

- A better understanding of their behavior through fabrication and processing.
- The environmental reaction of the materials.
- The prospects of recyclability or other ways for energy recovery.
- The specific properties like corrosion resistance, stiffness, weight, strength, chemical resistance, etc.
- The cost of the whole realization and manufacturing process.

The objective of this work is to elaborate new biocomposite materials that can on the one hand be a solution for plastic waste that is polluting our environment and having a noxious effect on human lives along with other living creatures. On the other hand, replacing the synthetic-based PMCs with natural resources that have good specific properties.

This thesis is structured of three chapters:

The first chapter presents the state of the art for composite materials. A general definition of composite materials and their constituent. In this work, we have focused on PMCs. One of the major topics we undergo in this section is biocomposites and a few used chemical treatment methods for this kind of composite. In addition, we have discussed a few major damage scenarios that can cause failure in composite materials, followed by manufacturing methods of composite materials. finally, we have talked about recycling technics of PMCs.

The second chapter is dedicated to our study on recycling plastic bottle caps made of high-density polyethylene (rHD-PE) reincorporated, as a matrix and reinforced by alfa short fibers and natural pozzolan particles. Three types of biocomposite material have been produced and characterized, by initially using the Fourier transform infrared red (FTIR) spectroscopy to evaluate the structural properties of the biocomposite components. Then, a mechanical tensile test to obtain mechanical properties. Finally, scanning electron microscopy (SEM) was applied to acquire some information on the morphology of the composite.

The third chapter is an investigation study of the effect of three types of natural fibers on the polylactic acid (PLA) matrix, using a genetic algorithm to calculate interface damage. To carry out this simulation the three types of biocomposites were subjected to the same mechanical shear stress raging from 450 N/m^2 to 650 N/m^2.

Chapter 1

Préliminaires

To produce a Polymer Reinforced Composite material (PRCM) with improved mechanical properties and good thermal stability, the choice of reinforcements and the method of processing is crucial. Therefore, this chapter presents a literature review of PRCM, talking about the different polymeric matrices and reinforcements used in this type of composite materials, plus their damage mechanism, and different methods used to manufacture PRCM.

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1.1 Constituents of composite materials

Composite materials are very heterogeneous and anisotropic because of the resulting mixture of their components; which means the geometrical partition of the fibers into the matrix Figure.1.1. An efficient and coherent composite depends on the quality of its constituents (fiber and matrix), the strong adhesion between these two, and excellent interfacial characteristics, which means good compatibility between the two phases furthermore the manufacturing method used.

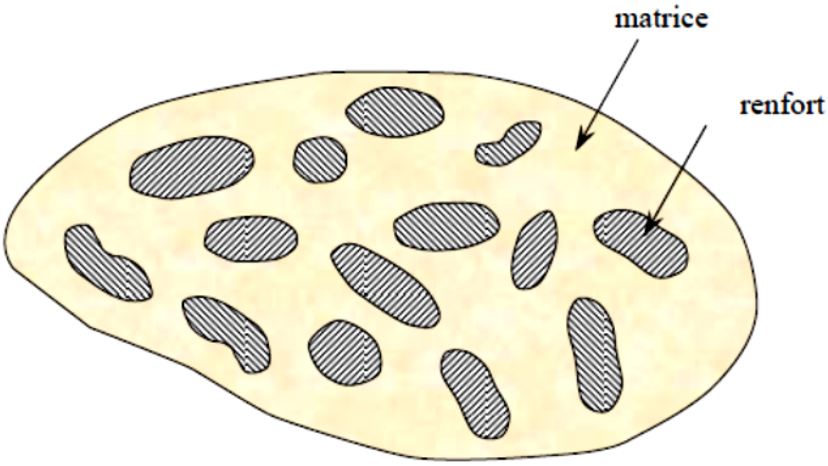

Figure 1.1: The schematization of composite components [17].

1.1.1 The matrix

The matrix, the constituent that binds the fibers and which provides preservation from external environmental factors like oxidation, ensures the cohesion between fibers and maintains the physical structure, its major function is to transfer also distribute the loads applied to the reinforcement [12].

There are several types of resins Figure.1.2, as we have mentioned before, organics, metallic, and ceramics. However, in this study, we will focus on the matrices made of polymers in general [84].

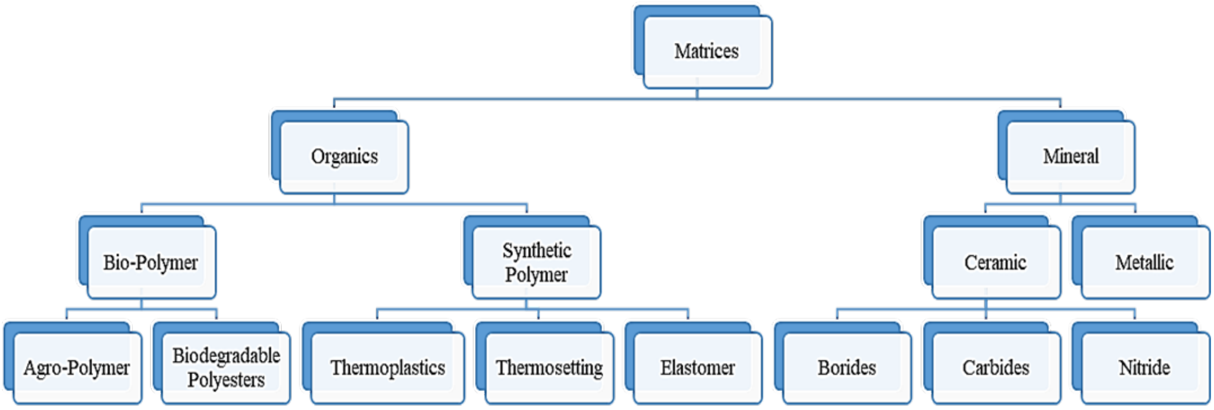

Figure 1.2: different types of matrices [118].

Organic matrices

We distinguish two groups of organic polymers; the petroleum-based synthetic polymers (the thermoplastics, the thermoset, and the elastomer). The second group is the biopolymers it gathered (the agro-polymers and the biodegradable polyesters).

a. Synthetic polymers

The matrix of composites materials are most often made of synthetic polymers, these polymers are constituent of macromolecular hydrocarbon chains with great chemical, physical and biological stability, and it can be classified based on the domain of application as follow [126, 153]:

Consumer polymers: it is used for their competitive overall cost also their low weight, for example, Polyethylene (PE), Polypropylene (PP), Polystyrene (PS)...etc [92].

The high-performance polymers: it is utilized in order to optimize the mechanical and thermal performance plus the weight reduction, for example, Polyether-ether Ketone (PEEK), Polymethylmethacrylate (PMMA)...etc [92].

After processing thermoplastics and thermosets form a three-dimensional network, yet these polymers differ in it, their manufacturing methods also by their mechanical, thermal, and chemical properties, besides that, each one represents advantages and disadvantages [92].

Thermoplastics polymers

Because of their ease of processing as it's just about the physical transformation, also their low cost, these polymers have taken place in various scopes, its consumption represents more than 80% of the total plastics consumption [22].

Thermoplastics exist in an amorphous or semi-crystalline form Figure.1.3 (figure1-3)15, its simple molecular structure allowed these polymers to be melted by heating to take forms and cooled to become a solid and can be fixed, these steps can be repeated multiple times without any risks of serious damage besides it is a short cycle because there is no cross-linking reaction, this operation is reversible [135].

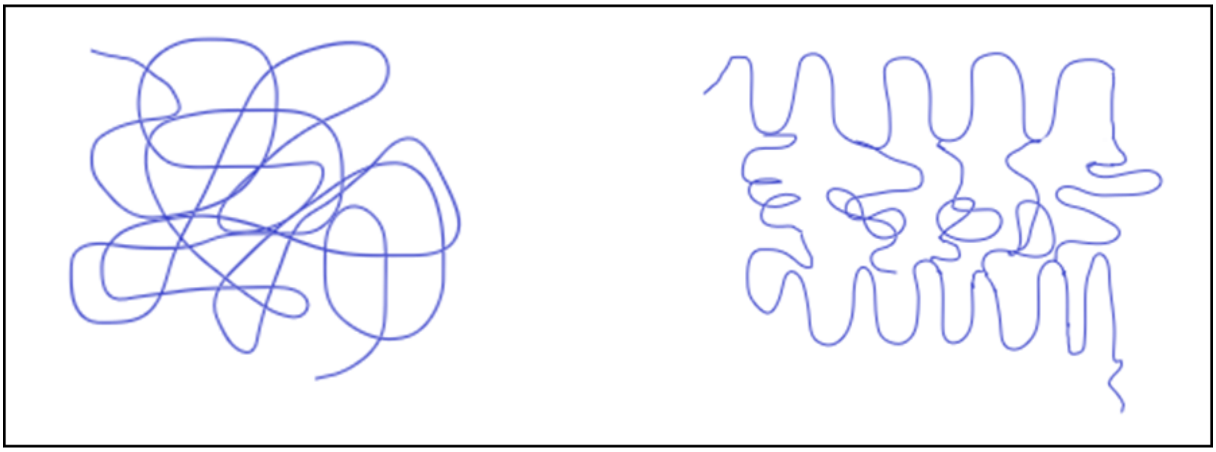

Figure 1.3: amorphous structure vs semi-crystalline structure [135].

More specifically the semi-crystalline thermoplastics have improved mechanical properties compared to the amorph thermoplastics and that is because of the organized structures of the crystalline part [135]. In Table.1.1 we can see some physical-mechanical properties of the most used thermoplastic resins.

Table 1.1: Characteristics of the most common thermoplastics [126].

Polymer	Tg (°C)	Tf (°C)	Density ρ (-)	σr (MPa)	E(GPa)
PP	5	165	0.92	30	1.2
PLA	50-60	150	-	40-60	3-4
HDPE	-100	134	0.95	28	1.1
PVC	75-105	160-220	1.39	58	2.9
PS	90-100	-	1.05	55	3.2
PET	70	255-265	1.30	47	2.6
ABS	90-120	-	1.05	50	2.5

These are the classified thermoplastics families Figure.1.4 16 based on their functioning [135]:

- Polyethylene (PE), Polypropylene (PP), Polyvinyl Chloride (PVC), Polystyrene (PS): commodity thermoplastics.

- Polyethylene (PE), Polypropylene (PP), Polyvinyl Chloride (PVC), Polystyrene (PS): commodity thermoplastics.

- Acrylonitrile-Butadiene-Styrene (ABS), Styrene-AcryloNitrile (SAN): copolymers with more specific applications.

- Polyamide (PA), Polycarbonate (PC), Polymethylmethacrylate (PMMA), Polyacetal (POM), Polyphenylene Ether (PPE), Polyethylene Terephthalate (PET), Polybutylene Terephthalate (PBT)...: engineering thermoplastics.

- Polysulfone (PSU), PolyetherImide (PEI), Polyphenylene Sulfide (PPS)...: engineering thermoplastics with more specific performances.
- Ethylene-tetrafluoroethylene (ETFE), Polyether-ether Ketone (PEEK): high-tech uses, limited consumption.

- Liquid Crystal Polymer (LCP), Polytetrafluoroethylene (PTFE), Perfluoroarlkoxy (PFA), Fluorinated Ethylene Propylene (FEP), Polyimides (PIs): high-tech uses, more limited consumption.

- PolyBenzimidazole (PBI): highly targeted uses and very restricted consumption.

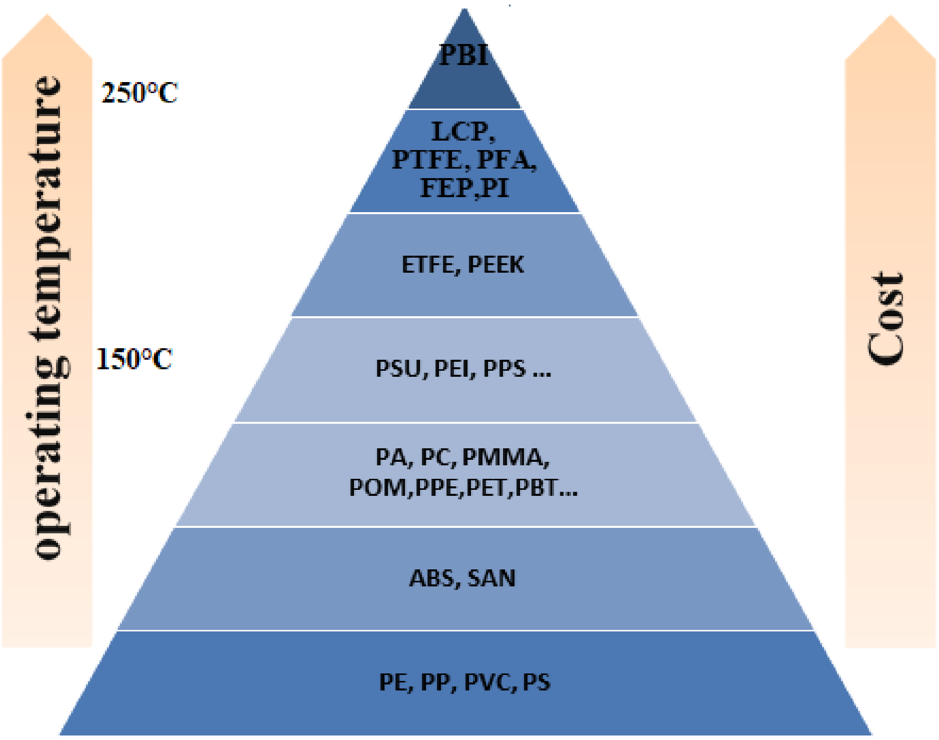

Figure 1.4: pyramid of quality for some thermoplastic families [27].

Thermosets polymers

In contrast to thermoplastic, thermosetting does not soften when heated it's an irreversible process for these polymers due to the chemical cross-linking, the curing of thermosetting results in a three-dimensional structure that has superior mechanical and thermomechanical properties compared to thermoplastic matrices [56, 130].

The dimensional stability gives these polymers a better general creep behavior and high modulus of elasticity except for the urethane, moreover simplicity of processing, and low price. There is one important drawback for this type of polymer concerning their waste management, they can be used as filler for polymer matrix composites but the methods of recycling are expensive, plus it is difficult to repair thermoset-based material composites [27].

overall, there are six thermosetting product families: Phenoplasts, Aminoplasts, Epoxies, Polyesters unsaturated, polyimides, and polyurethanes. In Table.1.2, we can see some of the most used thermosetting's physical-mechanical properties.

Some of the most common thermosets applications with classification Figure.1.5 are reported in the following section:

- Phenolic resins (PF) and Melamine (MF): high-temperature stability up to 300-350°C, high chemical stability, excellent fire retardance, low smoke, and toxicity emissions, and good friction properties. PF and MF resins are used in electronics, mine ventilation, rail, aerospace, decorative heat-resistant surfaces for kitchens

Table 1.2: Characteristics of the most common thermosetting [126].

Resin	Density ρ (-)	E(GPa)	σr (MPa)	Tmax (°C)
Epoxy	1.2	4.5	130	90-200
Urethane	1.1	0.7-7	30	100
Unsaturated Polyester	1.2	4	80	60-200
Urea Formaldehyde	1.2-1.5	6-10	40-80	140
Phenol-Formaldehyde	1.1-1.3	3-4	35-60	80-300

and furniture, etc [22, 70].

- Unsaturated Polyesters (UPs): A good basic resin for various uses at low cost. They are used in Toaster sides, iron handles, satellite dishes, breaker switch housing (with glass fiber), automotive body panels (with glass fiber), etc [34].

- Epoxy (EP): high adhesive properties (good adhesion with reinforcement), high chemical resistance, excellent mechanical properties, good fatigue resistance, low shrinkage, and high electrical resistance. They are used in Adhesive, automotive leaf springs (with glass fiber), bicycle frames (with carbon fiber), in high-performance applications the aerospace industry, etc [34, 70].

- Silicone (Si): High heat resistance up to 350°C, good resistance to tearing (flexibility). These resins are used as a mold release agent, in gaskets and sealants, and mainly for coating, etc [22, 70].

- Polyimides (PIs): Flame and chemical resistance, flexural strength. They are used in high-tech, especially in coating optical fibers in the electronics industry, etc [22, 70].

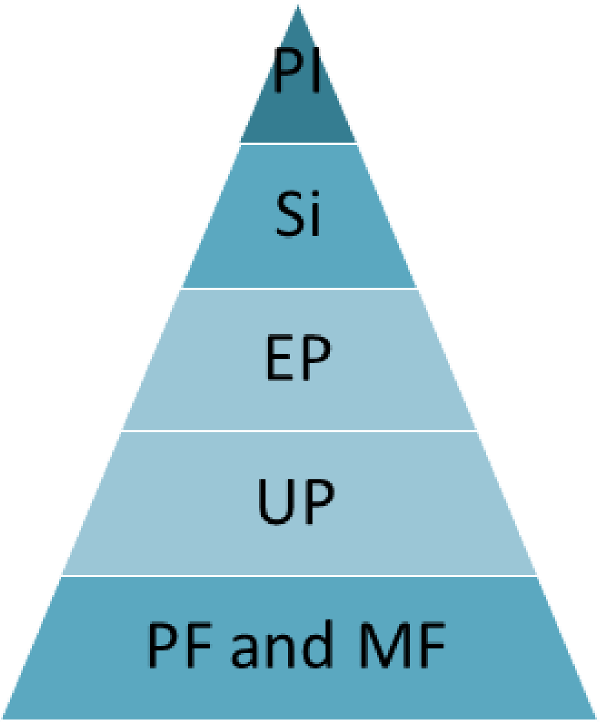

Figure 1.5: pyramid of quality for some thermoplastic families [22].

b. Bio-polymers

Developed from renewable resources, these types of polymers are usually made of biobased materials, which after their use these polymers can be readily recycled or disposed of chemically or biologically [105]. Biopolymers are obtained either by extraction from biomass, chemical, or biotechnological route of monomer polymerization. These biopolymers have found their place in various applications such as medicine, cosmetics, water purification, energy, and food storage, etc [73]. Based on their origin and synthesis as we can see in Figure.1.6, biopolymers are classified into four categories [41,140]:

- The first category is the agro-polymers or natural polymers, they are derived from plant or animal tissues by fractionation, e.g. polysaccharides and proteins [41,140].
- The second category is polyesters obtained by microbial fermentation, e.g. polyhydroxyalkanoate PHA [41,140].
- The third category is polyesters, chemically synthesized from monomers obtained from biomass, e.g. Polylactic acid PLA [41,140].
- The fourth category is a completely synthesized polyester by the petrochemical process, e.g. polycaprolactone PCL [72].

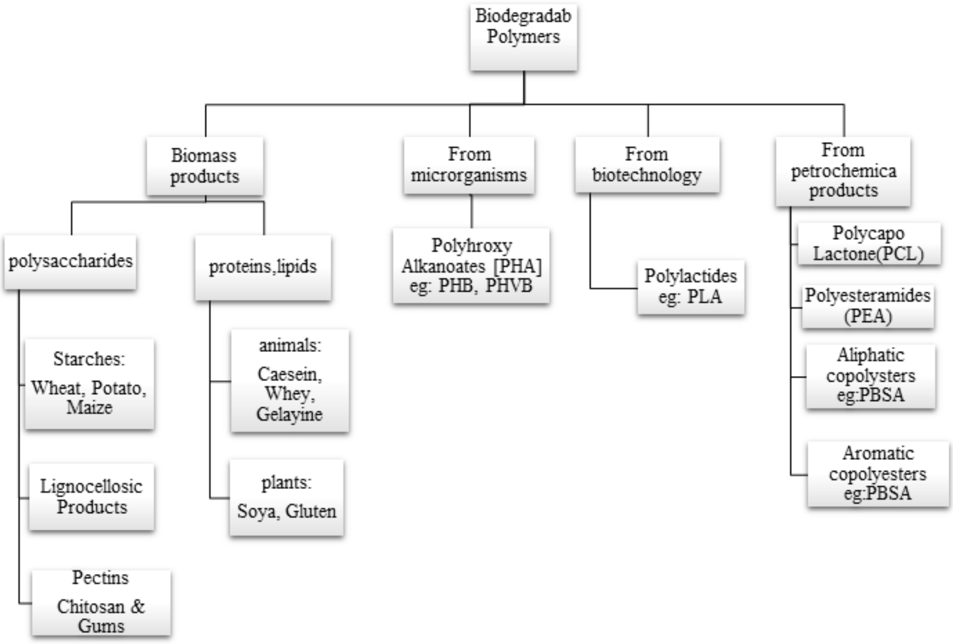

Figure 1.6: Classification of Biopolymers [118].

1.1.2 Reinforcement Materials

reinforcement materials represent the discontinue phase, it's the second important constituent for a material composite. This constituent improves the mechanical physical characteristics of the whole structure, the reinforcement operates as the skeleton of the composite. Its role is to sustain the majority of the mechanical stress applied to the composite material, thus because of its high modulus, rigidity, and ability to impede crack propagation. They can, however, impact other use aspects, such as thermal behavior (temperature resistance or fire resistance) [48, 92].

The choice of reinforcement materials involves the type of matrix and other factors such as shape, size, inherent properties (mechanical and environmental), cost, etc. The reinforcing material may also be split into three major categories based on size and form [66]:

- Flakes have two-dimensional geometry (flat shape), the type of materials used for this category of reinforcement is, mica, aluminum, and silver [124].

- Fillers come in the shape of particles or powders, they are Typical: calcium carbonate, aluminum oxide, lime (also known as calcium oxide), fumed silica, treated clays, and hollow glass beads [124].

- Fibers in the shape of rope or string, continue fibers, and they can be short like chopped fibers, or whiskers (elongated single crystals). Their cross-section can differ, circular, square, or hexagonal. The most used fibers are: glass, carbon, aramid, boron, alumina, and vegetal fibers [66, 124].

Reinforcement materials can be classified according to their chemical composition as it is represented in Figure.1.7:

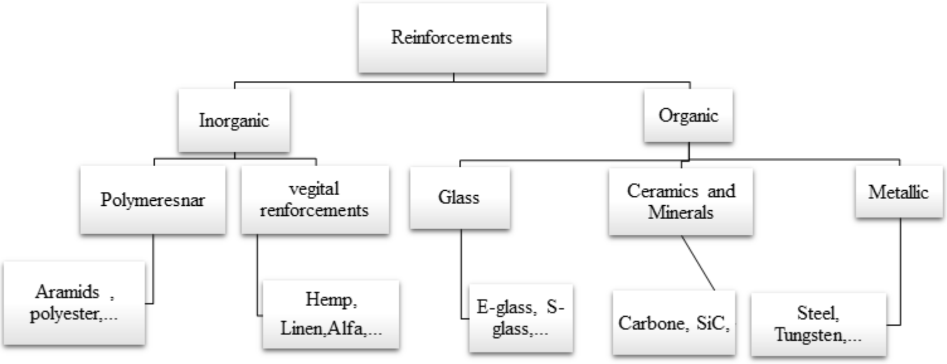

Figure 1.7: Classification of reinforsment material based on their nature. [118].

The following Table.1.3 compares the properties of some used organic and inorganic reinforcement materials:

Table 1.3: Characteristics of the most common thermosetting [12, 118].

Reinforsment material	Tensile strength (GPa)	Modulus (GPa)	Elongation (%)	Density (g/cc)
Carbon T300 (PAN)	3.53	230	1.5	1.75
Carbon P100 (Pitch)	2.41	758	0.32	2.16
Verre S	4.80	85.00	5.3	2.46-2.49
Kelvar 49	3.62	131	2.8	1.45
Boron	2.7-3.7	400	0.79	2.57
Steel	4.0	210	-	7.9
Hemp	0.4	35	1.6	1.07

Natural fibers

Natural fibers have been used since the dawn of civilization when ancient Egyptians had been using them as robes or yarns for textile weaving, hunting, fishing. . . etc. With the development of civilizations and over half a century, synthetic fibers were invented, driven by petrochemical origin these fibers have good characteristics such as durability, resistance,. . . etc, which made them more popular than natural fibers. The transition to a more sustainable and environmentally friendly economy has been driven primarily by changes in plant cultivation, food consumption, and energy production. Today, Many scientists are investigating novel applications for natural fibers in growing industries, there are around 2000 fiber species that are utilized for a variety of applications, and it has been demonstrated that natural fibers will be useful to humanity for a long time [73, 92, 134]. As the name implies, natural fibers are renewable materials that can provide refortification properties in composites and/or act as charges. In other words, because of their multiple advantages (biodegradability, CO2-neutral, low cost, important specific mechanical property,...etc), these fibers are a suitable alternative to synthetic fibers (glass, carbon, aramid, ...etc) [143].

Classification and structure

Natural fibers are classified based on their origin: animal, mineral, or vegetable, as it is shown in Figure.1.8. The composition and structure of these fibers depend on their origin.

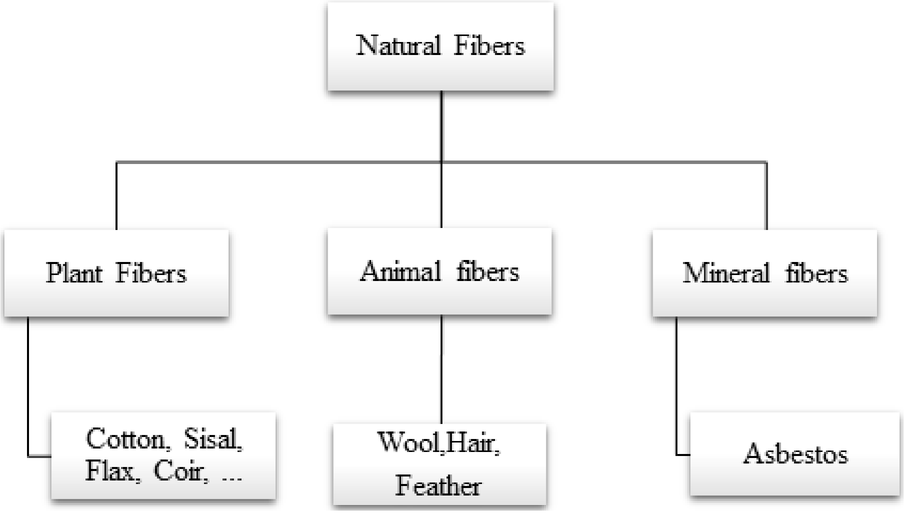

Figure 1.8: Classification of natural fibre [41].

- Plant fibers: composed mostly of cellulose, hemicelluloses, lignins, and pectins, plant fibers are the most used natural fibers, derived from biomass, these fibers can be extracted from a plant's fruit, stem, or leaf. Cellulosic fibers are mainly used in the manufacturing of paper and cloth, we can therefore cite cotton, jute, flax, ramie,...etc. This type of fiber will be further examined in the following section [38, 138].

- Animal fibers: consist mainly of proteins, these fibers represent barely 2% of the market of textile fibers, animal fibers include, hair, silk, and wool. There are Fibres taken from hairy mammals such as sheep wool, goat hair, etc, and we can find Secretions obtain from cocoons or spider webs to make silk fibers [38].

- Mineral fibers: Mineral fibers are primarily naturally occurring fibers or minimally modified fibers derived from minerals. Asbestos is a collection of minerals that naturally exist in the environment as bundles of fibers. These fibers are heat and fire-resistant, as well as poor conductors of electricity [38].

1.1.3 Fiber-matrix interface

The nature of the interaction between the distinct components during the composite system creation determines the behavior of composite materials [69]. (composite material components) these late are chemically, Mechanically, and physically linked and this interaction zone is referred to as the interface. The interface phenomenon occurring between the reinforcement and the matrix is manifested through a binding force [56]. At the micro-scale, the fiber-matrix interface is a finite volume extended area called the interphase where the properties of the overall composite are controlled [68]. only with good adhesion between the matrix and the reinforcement, the composite can sustain the transmitted stress across the interface.

adhesion is a complex notion because it can involve a large number of phenomena simultaneously from molecular to macroscopic. With the improvement in understanding the adhesion mechanism, several theories have been developed. From the literature, we state seven models of adhesion theories: mechanical interlocking, electronic theory, diffusion theory, chemical (covalent) bonding theory, adsorption (thermodynamic) or wetting theory, weak boundary layers, and acid-base theory [101].

Adhesion Theories

Mechanical interlocking

The mechanical adhesion model is a result of the formed bonding mechanism on all porous substrates, when the matrix penetrates through the roughness of fibers surfaces protuberance, and crevices create an interlocking between the two surfaces35. With the following methods figure.1.9, we can enhance the mechanical interlocking of composite materials. using Grafting nanoparticles to fibers to improve the roughness of fiber's surfaces [67, 68]. Anni Wang et al, have studied the effect of nano-clay grafting on flax fiber-reinforced epoxy composite durability. The results show that nano-clay grafting decreases saturation moisture uptake and the coefficient of diffusion of FFRP also increases the retention rate of the tensile modulus of the nano-clay grafted flax fiber-based FFRP [145]. For plant-based FRP composite materials, alkali treatment is often used to eliminate the noncellulosic component existing on the surface of the fiber; like wax and other amorphous components which can increase its roughness [67, 68]. As Wijang Wisnu Raharjo et al, have proved in their work about the effects of various alkali fiber treatments on the tensile and impact strengths of cantala fiber-reinforced recycled high-density polyethylene (rHDPE)composites, the experiments revealed the addition of alkali-treated cantala fibers to (rHDPE) matrix enhanced the tensile and impact strengths of the composite [127]. Polymer-grafted nanoparticles are a way to enhance the mechanical adhesion of composites by controlling the spatial arrangement of the nanoparticles in the polymer matrix [?, 68]. Joshua M. Kubiak and Robert J. Macfarlane, presented in their work a transformative aging strategy using anhydride crosslinking to produce rigid and highly filled composites in arbitrary geometries, the aging of the resulting composites enables the formation of covalent linkages between PGNPs, and depolymerization which increase the filler loading of the composite and the Tg of the remaining polymer, plus the raise of stiffness by 1-2 orders of magnitude [80].

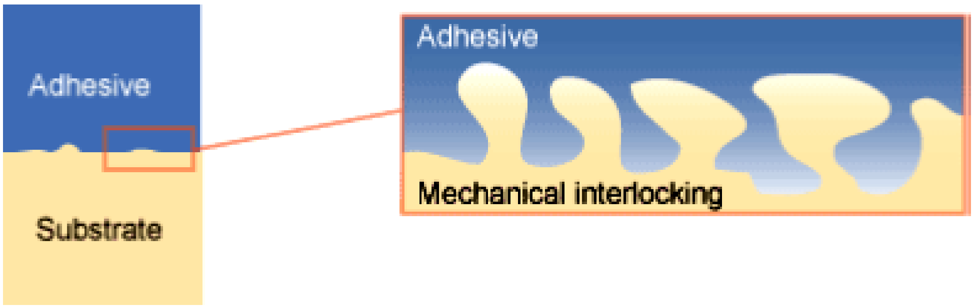

Figure 1.9: Schematic presentation of mechanical interlocking adhesion [93].

Electrostatic Adhesion

According to the electrostatic theory, adhesion is created by electrostatic forces between two charged layers, due to an imbalance of electrons or ions that caused a difference in electrostatic charge between the reinforcement and matrix surfaces at the interface. These layers are formed under the assumption that an electron exchange mechanism exists between the two surfaces. Few concepts demonstrate electrostatic adhesion, as Coulomb's Law describes the electrostatic interaction between electrically charged particles by [68, 113]:

$$|F| = K_e \frac{|q_1 q_2|}{r^2}$$

where: F is force, ke is Coulomb's constant, q1 and q2 are the charges and r is the distance between the charges.

Diffusion theory

the diffusion theory only applies when the substituents of the composite are soluble in one another, it involves the interdiffusion of macromolecules across the interface. the polymer materials constituting the composite are dissolving together and creating an interphase. The adherence energy or more specifically the molecular entanglement of the mixture is directly affected by time, temperature, and pressure of contact [121]. Healing as defined it Julien Avenet et al; is the diffusion of the macromolecular chains across the interface that enable bridging. In this study, the adhesion of thermoplastic composites is investigated. The goal is to characterize adhesion for conditions close to the industrial process, especially short residence time. For this characterization two experimental benches were developed, A short-time welding bench under controlled conditions designed for fast heating and cooling and A double cantilever beam setup to characterize adhesion quality. The correlation confirms that the mechanical quality is associated with macromolecule diffusion, It was also noted that for high-temperature thermoplastics, reticulation occurs, which inhibits molecular diffusion and increases the relaxation time [9] .

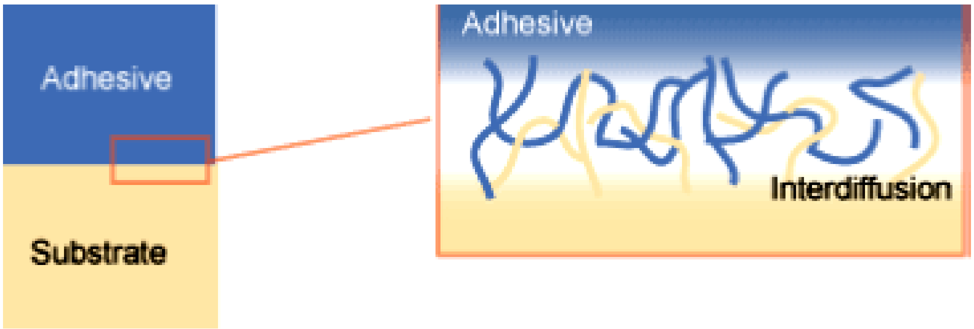

Figure 1.10: Schematic presentation of adhesion by diffusion [93].

Chemical bonding theory

This theory is based on the formation of effective chemical bonds between the active groups of the polymer chains and the surface of the substrate also called primary adhesion bonds. The efficiency of chemical coupling is affected by two parameters: the number of interfacial bonds and the length of the coupling links. These bonds use far more energy compared to secondary bonds like that of van der Waals and hydrogen bonds with 50 KJ/mol. Chemical bonds are classified into three types: covalent bonds with bond energies ranging from 60 to 700 kJ/mole, metallic bonds with energies ranging from 110 to 260 kJ/mole, and ionic bonds with energies ranging from 600 to 1000 kJ/mole [11]. To enhance chemical bonding in the case of plant base natural fibers reinforced polymer composites, coupling agents are used. Silane and malleated are two types of coupling agents, which are bifunctional molecules the two ends of these molecules can react with the compatible chemical groups of each part of the composite (fiber/matrix). M. Abdelmouleh et al investigate the effect of three types of silane coupling agents (c-methacryloxypropyltrimethoxy (MPS), c-mercaptoproyltrimethoxy (MRPS) and hexadecyltrimethoxy-silanes (HDS)) on cellulose fibers reinforcing two different matrices (low-density polyethylene and natural rubber). Results show that the composite materials based on cellulose fibers treated with MPS and MRPS using both matrices displayed good mechanical performances, compared to the incapacity of the HDS coupling agent to bring about covalent bonding with the matrix [1]. P. Mutje´ et al have explored another method to improve chemical bonding adherence. Maleated poly(propylene) (MAH-PP) was added as a coupling agent to hemp strand-based poly(propylene) composites. The addition of (MAH-PP) at 4% wt/wt to hemp strands, increases the ultimate tensile strength (σt) and flexural strength (σf) and the FT-IR spectroscopy results revealed an establishment of covalent bond [108].

Thermodynamic adhesion theory

thermodynamic adhesion theory is also known as the adsorption theory or simply the wetting model. This theory is applicable for solid-liquid composites, it involves the atomic and molecular interactions of the liquid surface onto a solid substrate, This interaction is created by the free surface energy (γ) characteristics of both liquid and solid. The interfacial forces generally considered of major importance are van der Waals and Lewis acid-based interactions [137].

The condition necessary for spontaneous wetting is given below:

$$\gamma_{sg} \geq \gamma_{sl} + \gamma_{lg}$$

Where: $\gamma_s g$, $\gamma_s l$ and $\gamma_l g$ are respectively the interfacial free energies for solid-gas, solid-liquid, and liquid-gas interfaces. If $\gamma_s l$ is insignificant, the criterion can be simplified to:

$$\gamma_{sg} \geq \gamma_{sl}$$

or

$$\gamma_{substrate} \geq \gamma_{adhesive}$$

which means that the adhesive will wet the surface of the adherend when the surface free energy of the substrate is greater.

Surface free energies of solids are generally determined by measuring the contact angles of appropriate probe liquids on the solid surface. to measure various types of substrates, various contact analysis techniques are used [101]. One is the Wilhelmy Plate technique which can be used to measure contact angles on thin plates and single fibers as shown in Figure.1.1134. The Wilhelmy equation is as follows:

$$F = \gamma_L P \cos\Theta + mg - \rho_L A h_g$$

where:

F=advancing or receding force on the sample in liquid.

γ_L=surface tention of the liquid.

P= perimeter of the wetted cross-section.

m= mass of the specimen.

g= acceleration due to gravity.

ρ_L= liquid density.

A= cross-sectional area of the specimen.

h= depth of immersion.

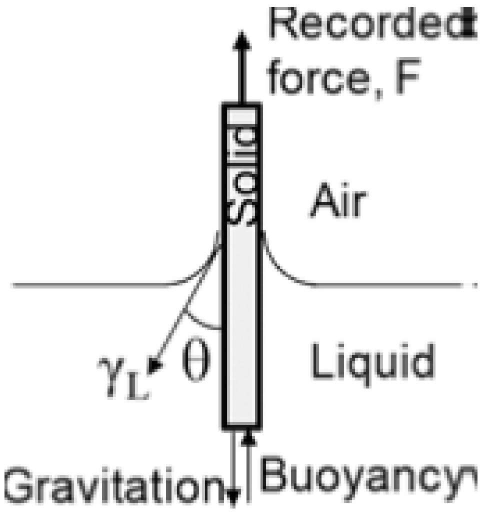

Figure 1.11: Schematic presentation of adhesion by diffusion [101].

by measuring the penetration rate of a liquid going through packed particles, Washburn's equation was used to determine the dynamic liquid contact angle into packed powders. The original Washburn's equation is [71]:

$$x^2 = \frac{R_{eff}\gamma_l \cos\Theta}{2\eta}t$$

Where:

x=the wetting distance in the packed powders as a function of time t.

R_{eff}=the effective radius of the interparticle capillaries in the porous packed powders.

γ_l=the surface free energy of the probe liquid.

Θ=the wetting contact angle.

η=the liquid viscosity.

weak boundary layers

At the joint of the adhesive and the substrate, interphases or boundary layers are created whose properties determine the joint strength. The First one to describe a weak boundary layer (WBL) was Bikerman, it states that the interphase can fail by either a cohesive break or a WBL. As we can see in figure.1.12 WBL is originally formed by the presence of air bubbles, impurities, and other agents near the bonding surface causing malformed links between the adhesive and the substrate [43, 137].

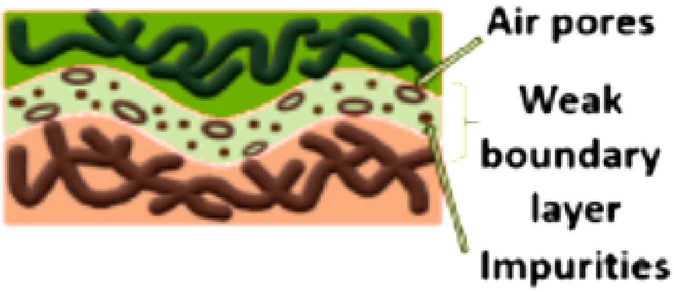

Figure 1.12: Schematic illustration of weak bondry layers [128].

Acid-base theory

The concept of acid-base was first proposed by Svante Arrhenius in 1884 based on the theory of ionization. the Arrhenius acids are the hydrogen-containing compounds that give H+ ions or protons on dissociation on dissociation in water, Arrhenius bases are the hydroxide compounds that give OH- ions on dissociation in water. in 1938 J. N. Bronsted and G. N. Lewis, has defined an acid as a substance that can accept an electron pair from a base; a base is a substance that can donate an electron pair [43, 106].

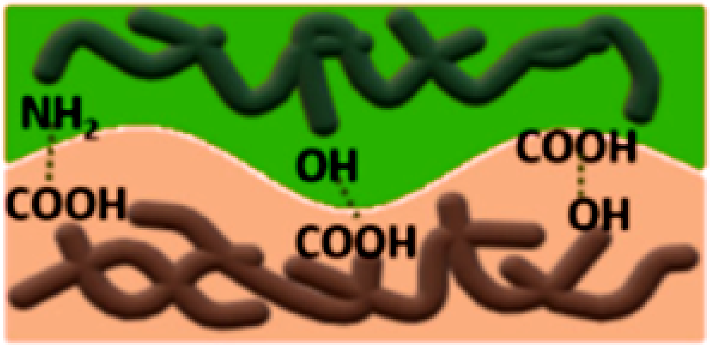

Figure 1.13: Schematic illustration of acid-base adhesion [128].

1.2 Bio-composite materials

Bio-composites or eco-friendly composites are interesting types of materials that have attracted great interest in different industry sectors such as automotive, medical field, construction, and aerospace, among others [79]. The considerable growth in using these materials in recent years is due to the increasing awareness of environmental protection since plastic waste has become one of the major and critical problems in different industries and a lifetime risk that threatens the environment. Furthermore, biocomposites gather multiple beneficial characteristics such as being abundant, cheap, recyclable, and biodegradable [2]. these materials are derived from renewable and non-renewable resources of biological origin [2]. these types of composites can combine hybrid materials that consist of a mixture of organic and inorganic components for example, synthetic and natural polymers reinforced with mineral or vegetable fibers [42].

1.2.1 Plant fibers

The speedy growth in the use of plant fibers over artificial filler is due to the advantageous features these fibers provide, such as nontoxicity, low cost and density, flexibility during processing, and high tensile and flexural modulus, along with others [154]. the basic chemical structure of plant fibers differs based on their type and origin. Generally, it's known that cellulose is a common component in all plant fibers, this polymer is a complex polysaccharide formed of glucose units that are linked in a unique way which makes cellulose resistant to breakdown, since these fibers are composites itself, the cellulose existing in a shape of fibrils inside plant fibers are covered by other components like hemicellulose and lignin and the content of these components vary through plants types, see Table.1.4 [23, 99].

Plant fibers are meanly classified as wood fibers and nonwood fibers, they are labeled as follows:

Nonwood fibers:

- Bast: hemp, flax, jute, kenaf, and ramie
- Leaf: sisal, pineapple, abaca
- Seed/fruit: coir, kapok, coconut
- Straw: wheat, rice, soy, and corn straw
- Grass/reed: switchgrass, miscanthus

Wood fibers:

- Soft and hard woods

Table 1.4: chemical composition of some plant fibers [19].

Type of fibers	**Chemical composition (%)**		
	Cellulose	**Hemi-cellulose**	**Lignin**
Jute	61-63	13	5-13
Banana	60-65	6-8	5-10
Coir	43	<1	45
Flax	70-72	14	4-5
Mesta	60	15	10
Pineapple leaf	80	-	12
Sisal	60-67	10-15	8-12
Wood	45-50	23	27
Sun hemp	70-78	18-19	4-5
Ramie	80-85	3-4	0.5

Fiber structure, cell dimensions, defects, crystallinity, and chemical composition are the primary variables that influence all fiber properties. In general, as cellulose content increases, so does the tensile strength and Young's modulus of the fibers, we can see in Table.1.555 the mechanical properties of different plant fibers [41].

Surface treatment methods of plant fibers

The optimization of the interfacial bonding between plant fibers and polymer matrix in the production of composite materials is an important step. Therefore, chemical modification of plant fibers is essential, since this late are hydrophilic; they absorb moisture and create performance problems with the hydrophobic polymer matrix.

To improve the performance of the final structure of the composite material and the adherence between natural fibers and polymer matrices, multiple approaches are used to modify the surface of lignocellulosic fibers, some of the most popular chemical treatments will be briefly introduced next [86].

Table 1.5: Mechanical properties of different types of plant fibers for composite applications [12, 118].

Natural fibers	Tensile (Mpa)	Elongation at break (%)	Young's modulus (GPa)
Jute	200-800	1.16-8	10-55
Banana	106-175	14.21-49	4-6
Flax	300-150	1.3-10	24-80
kenaf	295-119	3.5	2.86
Pineapple leaf	170-162	2.4	60-82
Sisal	80-840	2-25	9-38
Hemp	310-900	1.6-6	30-70
Ramie	348-938	1.2-8	44-128

Alkali treatment

One of the most cost-effect and widely used treatment techniques is also known as mercerization. This method involves treating the fibers with sodium hydroxide (NaOH) to remove and clean the surface of some lignin and hemicellulose while completely removing pectin, wax, oils, and other organic compounds [79]. The reaction is presented in Figure.1.14. A.M. Radzi et al [125] , investigate the effect of various sodium hydroxide(NaOH) concentrations on roselle (RF)/sugar palm fiber (SPF) reinforced thermoplastic polyurethane hybrid composites. Results show the highest tensile strength of 14.26 MPa, flexural strength is 14.05 MPa and impact strength is 23.76 kJ/M2) that was obtained from treatment 6 % NaOH concentration, (SEM) micrograph of hybrid composite tensile fractured revealed good adhesion bonding between fiber and matrix, FTIR spectroscopy analysis indicates lower water uptake and thickness swelling of the treated composites (7.97 % and 6.49 %) respectively obtained from 9 % NaOH concentration, the Thermogravimetric analysis exhibit improvement of the thermal stability for the treated composites.

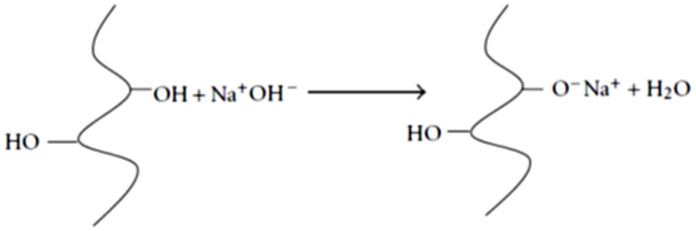

Figure 1.14: Alkali treatment reaction of cellulose-based fibers with NaOH [147].

Silane treatment

Silanes are widely used as coupling agents for lignocellulosic fiber-reinforced polymer composites. Organosilanes are the mean group of coupling agents, they are adopted to improve the interfacial bonds between polymers and natural fibers. The silanol then reacts with the hydroxyl group of the fiber see Figure.1.15, forming stable covalent bonds to the cell wall that are absorbed onto the fiber surface, thereby forming a polysiloxane network on the fiber surface that gives molecular continuity across the composite interphase [32, 86]. Y.Liu et al [91], examine the effect of different concentrations of silane on chemical, surface morphological and mechanical performances of Corn Stalk Fibre (CSF) as well as impact strength and impact fracture surface morphology of CSF reinforced polymer composite. The SEM images showed that the fiber's surface after treatment is rough and slightly clean. Regarding the composition of the fiber, part of the hemicellulose and pectin were removed from the CSF surface with silane treatment. Results show improvement of the CSF mechanical properties after treatment, the 5% silane-treated CSF presented the highest tensile strength of 223.33 MPa ± 41.22 MPa and Young's modulus of 7.05 GPa ± 1.07GPa among all the specimens.

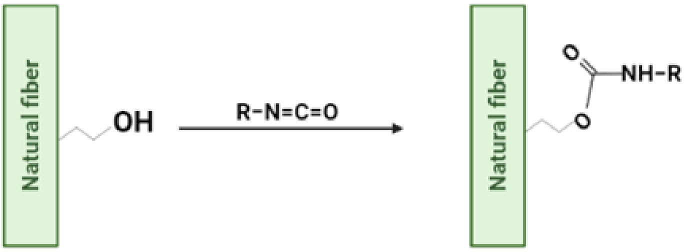

Figure 1.15: Silane treatment reaction of cellulose-based fibers [86].

Acetylation

Acetylation increased the hydrophobicity of natural fiber, it's a renowned esterification method figure.1.16, that occurs by introducing the acetyl functional group (CH3COO-) with acetic acid as a byproduct to the hydroxyl groups (-OH) on the fibers. the acetylation can be preceded by alkali pretreatment for better results, In some cases

[79, 120].

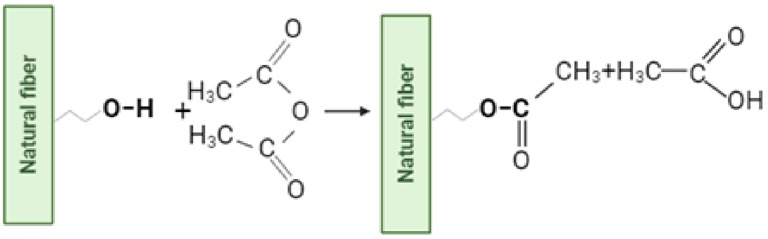

Figure 1.16: Acetylation treatment reaction of cellulose-based fibers [32].

H.U. Zaman and RH Khan, study the effect of fiber composition, mercerization of fiber as well as the incorporation of acetylation as a coupling agent on the properties of Nonwoven unidirectional matted banana empty fruit bunch fiber (BBF)-reinforced polypropylene (PP)composites. The results showed an improvement in the mechanical properties compared to the neat polymer which proves that the BBFs acted as reinforcing fillers, the acetylation modification after the alkali pretreatment revealed better interfacial shear strength and mechanical properties compared to the alkali-treated and untreated fiber composites plus the enhancement in thermal and flammability stability after acetylation-incorporation in the composites compared to the neat PP. The moisture absorption test, indicates less water absorption of the treated banana matted composites, than both alkali-treated and untreated composites [152].

Benzoylation

In the same concept as for acetylation treatment, Benzoyl is used to decrease the hydrophilic nature of the fiber, therefore increasing the adhesion with the hydrophobic matrix. Benzoyl ($C_6H_5C_5O$) is introduced to the cellulosic OH groups of the natural fibers figure.1.17, This reaction reduces the moister absorption of the plant fibers which results in better interaction between fiber/matrix and improves the thermal and mechanical properties of the composite [156].

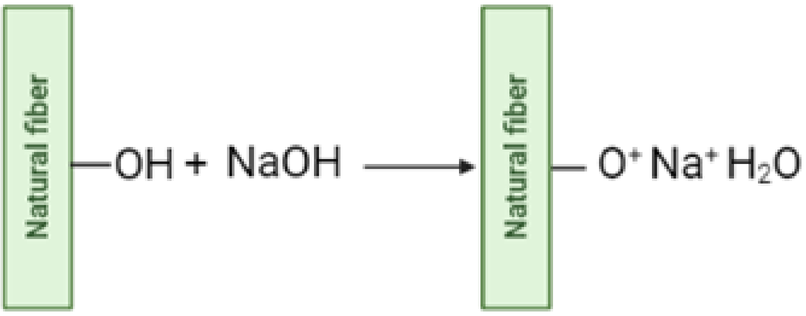

Figure 1.17: Benzoylation treatment reaction of cellulose-based fibers [88].

S. Mohd Izwan et al [103], have performed research to evaluate the mechanical and thermal properties of sugar palm fiber (SPF)- and kenaf fiber (KF)-reinforced polypropylene (PP) composites after benzoylation treatment of SPF/KF fibers. The results show that the treated composites show the best flammability properties using diffraction scanning calorimetry (DSC) and thermogravimetry analysis (TGA). Dynamic mechanical analysis (DMA) exhibits an increase in the storage modulus of the biocomposite which reveals the effective stress transfer from the fiber to the matrix at the interface. In addition, Thermomechanical Analysis (TMA) demonstrates better mechanical properties of the treated hybrid composites.

Maleated coupling treatment

This method is used to improve composite interfacial bonding and mechanical properties. Adding maleic anhydride (MA) to the composite mixture doesn't only modify the fiber's surface but the polymeric matrix too, thus what makes it different from other chemical treatments. This treatment works by grafting the MA agent to the polymer that is used to produce the final matrix, therefore ensuring great compatibility between the matrix and the coupling agent Figure.1.1865. Polypropylene (PP) is the most frequently used polymer in the maleated coupling treatment. The grafting of MA to PP results in grafted maleic anhydride polypropylene (MAPP). The MAPP reacts with hydroxyl groups (OH) in the amorphous region of the cellulose structure, which provides covalent bonds across the interface [79, 88]

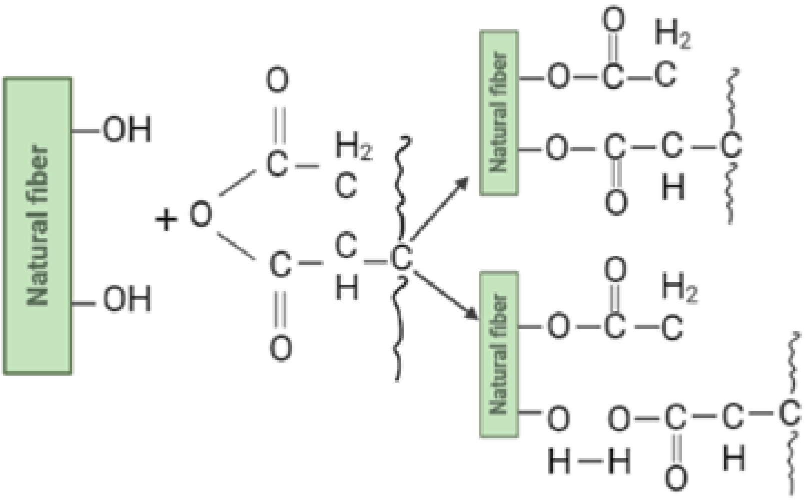

Figure 1.18: Maleated coupling treatment reaction of cellulose-based fibers [88].

In his study M.Poletto [122], have used maleic anhydride grafted soybean oil, to produce maleated soybean oil (MASO) as a coupling agent in wood-fiber-reinforced recycled polypropylene. Based on the obtained results, The usage of MASO as a coupling agent has clearly improved the interfacial adhesion between wood fibers and the polypropylene matrix and increased the mechanical and thermal properties.

1.3 Damage in composite materials

Damage can be defined as the phenomenon of the creation of new surfaces within a material under the effect of stress. A set of microstructural changes like the increase in the size or number of cracks causes a decrease in the mechanical characteristics of the system which leads to its ruin. Three mechanisms are generally observed during the failure processes in a material [78] :

- crack initiation: which corresponds to the appearance of the cavity within the material.
- Growth: during which the volumes of cavities increase.
- Failure: the phase that corresponds to the appearance of microcavities by coalescence of existing microcavities.

Composite materials are designed for a diversity of industrial applications, these materials can sustain different types of stress whether it was mechanical, thermal, or chemical that results from the specific functionalities used. Composite material undergoes different forms of degradation, it can be defects created during the manufacture of the composite (microcavities or microporous, bubbles,...) or damage resulting from loading the structure . The microcracks that develop are usually too small to cause damage in a composite. Thus, the accumulation of these microcracks will cause the final failure the nature of the composite constituent plays a very essential role in the type and mechanism of failure, and due to its heterogeneous nature, composites material's macroscopic properties are naturally anisotropic [59, 107].

During this work, we are interested only in the study of the damage under mechanical stresses. the dominant damage mechanisms of a heterogeneous material subjected to mechanical stress can be arranged into four main different modes depending on their function field, see figure.1.19 .

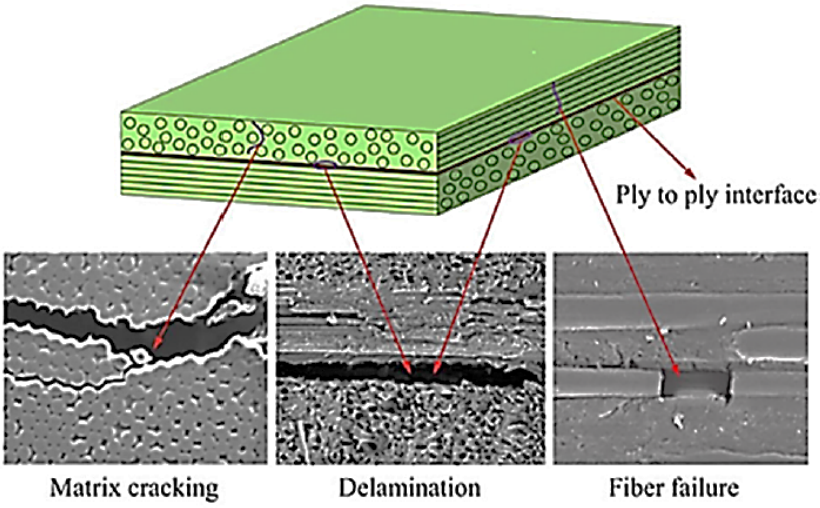

Figure 1.19: A representation of some failure modes of a composite materials [57].

1.3.1 Matrix cracking

Matrix cracking happens on a microscopic scale. Micro Cracks occur within the matrix areas where the heterogeneity is important or in the presence of porosity and inclusions, these inclusions lead to stress concentration, which introduces cracking of the matrix either by transverse cracking or by longitudinal cracking. This pattern failure occurs when the failure by decohesion energy is greater than the shear failure energy, which propagates in the perpendicular direction to the stress and can cause interface decohesion [107, 112].

1.3.2 Fiber breakage

Fiber breakage usually occurs at an advanced stage when the composite has gone through all other damage mods. This phenomenon leads to instability and above all a loss in the rigidity of the composite, which leads to the ruin of the structure and failure [10].

1.3.3 Fibre-matrix decohesion

The interphase is the zone of charge transfer between the matrix and the reinforcement, it is here where the fiber-matrix de-cohesion happens. This damage pattern is the first that appears in composite materials failure, which follows the appearance of microcracks in the matrix. The failure of the fiber-matrix interface depends on the quality of adhesion between the composite components [112]. After initiation, the propagation of the crack changes according to the nature of the fiber-matrix adhesion [78]:

- If the adhesion is very weak, the matrix cracking is deflected at the interface and the fiber-matrix decohesion propagates over a large distance.

- if the adhesion is very strong, there will be a propagation of the crack which will be initiated either by fibers crack or by matrix crack.

1.3.4 Delamination

Delamination is one of the most common failure modes of composite laminate materials. It may occur as a consequence of separation between two plies after loading the structure. This phenomenon can be observed on a macroscopic scale, besides it being caused by matrix cracking, bending cracks, and shear cracks, it may develop in anyone or any combination of the three basic modes of interlaminar fracture. These modes are mode I, the opening mode, or peel mode, mode II, the in-plane shear mode or sliding shear mode, and mode III, the out-of-plane shear mode, or twisting shear mode [40, 139].

1.4 Manufacturing of composite materials

There are several methods for fabricating composite materials, Therefore the choice of a method for a specific part will thus be determined by the materials used, the design of the part, and the end-use or application. Some of these processing techniques are the same used for standard materials, however, many were created to address specific design or manufacturing challenges. Indeed, the elaboration of a composite part works together with the development of the processing method [97, 109].

The main characteristics of composite materials elaborating processes that differentiate them from the other ordinary materials processes like metallic materials are [97]:

- The final shape of the product, as well as the geometry of the molded components, are obtained almost at the same time

- A variety of materials characterized by different properties, operating conditions, and costs are used

- The operating sequence has a predominant role in the mechanical characteristics, the geometry, and the manufacturing costs.

In this work, we are only interested in natural fibers base thermoplastic composites, as already munched in previous sections, thermoplastic polymers can be reinforced with vegetable fibers without any risk of degradation or loss in mechanical properties of the fibers since their thermal, mechanical, and rheological properties are reversible as these polymers exhibit minimal cross-linking [144]. In this case, fiber composites are most commonly fabricated by either impregnation with a matrix material in a preliminary process, resulting in a semi-finished product referred to as pre-preg (pre-impregnated), or by direct impregnation within the component production process, in order to wet out the fibers [5]. in Figure.1.20 the manufacturing methods suitable for this type of composite are described.

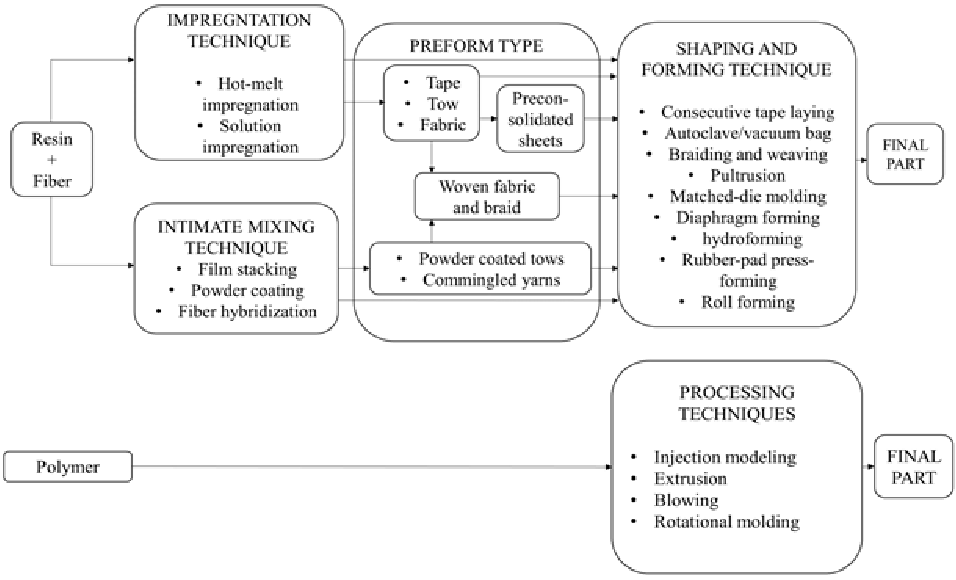

Figure 1.20: Routes for processing thermoplastic composites [144].

1.4.1 Pultrusion

Pultrusion is a continuous manufacturing process for long and short fiber-reinforced polymer composite profiles with a constant cross-section [132]. The resin-bath pultrusion (RBP) and the resin-injection pultrusion (RIP) processes are two common methods in this manufacturing technique, see Figure.1.21:

- The pre-impregnated (pre-pregs) rovings like (tows of fibers), mats, and fabrics are used depending on the composite material we need [64, 132].

- The pre-pregs pass through guides that shape and organize the layup into the profile. Depending on the application, the fiber material may be preheated before entering the pultrusion die to speed up the curing step [64, 132].

- After passing the guides, the fiber material is pulled into the impregnated apparatus [64, 132].

- the wet-out reinforcements are passed through The pultrusion die, which is actively heated and cooled to control polymerization (curing of thermosets) or crystallization (solidification of thermoplastics), the shape, and size, of the finished products [64, 132].

- Finally, A puller is used to drag the products through the die then a pelletizing system is used to cut the final products [64, 132].

Figure 1.21: A state-of-the-art resin-injection pultrusion line [132].

1.4.2 Compression molding

compression molding is a composite manufacturing process usually used to produce high-quality composite parts in high production volume. This technique offers short cycle times, good reproducibility, and excellent dimensional stability for biocomposites. Compression modeling also named hot press modeling, consist of placing the impregnated reinforcements in the mold cavity, the two part of the mold are hot pressed to give the final shape of the composite, as presented in Figure.1.22. The modeling parameter such as pressure, pressing cycles, and time duration are chosen based on the type of used polymer [52, 100].

Compression molding is the most suitable technique to achieve a high fiber content with good impregnation and mechanical characteristics [65].

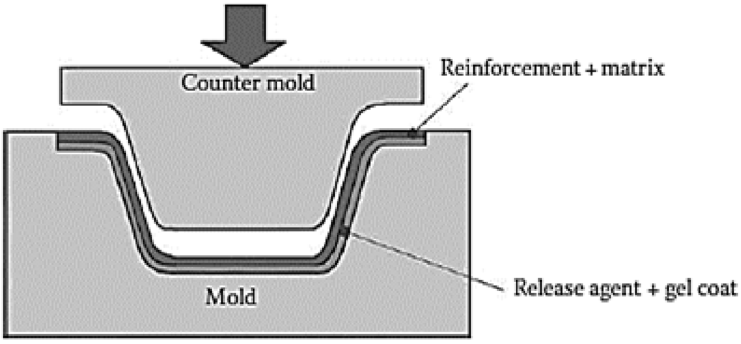

Figure 1.22: Compression molding [60].

1.4.3 Injection molding

injection molding is a widely used technique for short fibers reinforced thermoplastic composites, it is famous for mass production plus the production of complex shapes. Compared to other manufacturing processes, injection molding has low unit prices, short cycle times, and good surface aspects [52].

Before feeding the composite into the injection mold, the first step is the compounding of the polymer and the reinforcement. The polymer is first to be introduced to the twin-screw extruder as we can see in Figure.1.2386, secondly is the natural fibers, to have a homogenous mixt without the degradation of the fibers, resulting in composite pellets [123].

When the pellets are ready, They are heated up to polymer melt temperature and mixed all along their progress over the rotating screw. The material is injected into the mold by the movement of the screw into the heated mold at a given temperature. to avoid warpage, the material continues to be injected to maintain high-pressur inside the mold in order to reach a threshold. Finally, the ejection and cooling of the final product [123].

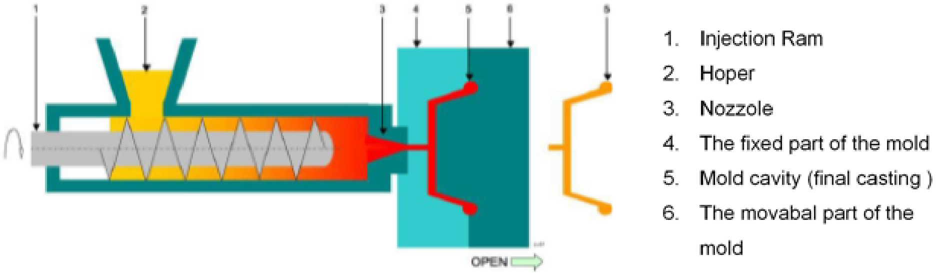

Figure 1.23: Injection molding process [65].

1.5 End of life and recycling of biocomposites

With the increasing demand for thermoplastic reinforced composites, there is an increase of waste for disposal, therefore to manage the waste which presents a risk towards the environment and our lives, investigations are taking place to develop new sustainable techniques of recycling since the traditional ones like incineration and landfills are becoming more restrictive [16]. The mean current used recycling processes for composite materials are reviewed.

1.5.1 Mechanical recycling

This route presents the advantage of recovering both matrix and fibers, without the use of hazardous chemicals and an absence of gas emissions, this technique is suitable for thermoplastic materials like PLA and is hence a desirable option given its ease of processing and parametric control [31], mostly used when it comes to recycling naturally reinforced thermoplastic composites [16]. Mechanical recycling consists in reducing the size of the scrap off-cuts or of a component at its end of life (EoL) by crushing or shredding followed by grinding following different stages [119]. Next, the resulting is separated into different size fractions using a sieve classifier. The resin-rich powder is commonly used as filler. The fibrous fraction is usually employed as a reinforcement [16].

Sujal Bhattacharjee and Dilpreet S Bajwa, have investigated the recyclability of oak wood flour (WF) filled high-density polyethylene (HDPE) composites. WF-HDPE composites with 30% and 50% oak wood flour, and 3% coupling agent MAPE were manufactured and reprocessed up to six times by extrusion followed by injection molding. Results show, with successive recycling, the strength and stiffness properties of the composites decreased which is related to the reduction of interfacial adhesion, but strain properties increased mainly due to the decrease in fiber length, and weight reduction of the polymer. The crystallinity of HDPE decreased but the thermal stability of the composite increased with the increased number of reprocessing cycles [20].

1.5.2 Chemical recycling

Recycling through chemical processes or feedstock recycling. In this process, the structural bonds of the polymer are broken and converted to monomers or partially depolymerized to oligomers utilizing chemical reactions, classified based on the solvent. The classification namely hydrolysis, glycolysis, and acid digestion, which are carried in the solution of water, glycols, and acid, respectively [136, 148].

1.5.3 Thermal recycling

The primary goal of thermal recycling is to separate the fibers from the matrix. and it can be achieved by different methods such as the pyrolysis technique, fluidized beds pyrolysis technique, and microwave pyrolysis techniques. These techniques operate at temperatures ranging from 450°C to 700°C and can recycle and recover glass and carbon fibers from materials composites [63].

Chapter 2

Mechanical and morphological characterization of recycled HD-PE bio-composites based on alfa fibers and natural pozzolan

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2.1 Introduction

Natural fiber-reinforced polymer composites (NFRPCs) are eco-friendly materials because they can be easily disposed of or composted. the current environmental regulation concerning plastic materials wastes has incited different industries to rely on these biomaterials. Moreover, NFRPCs are an excellent opportunity to reduce the dependency on petroleum-derived products [26, 33].

This chapter presents the experimental study of a biocomposite material composed of high-density polyethylene (HD-PE) reinforced by Alfa short fibers and natural Pozzolan particles. The objective of this study is to investigate the effect of different content of alfa short fibers and pozzolan particles on the mechanical and morphological properties of the recycled HDPE matrix.

2.2 Methods and Materials

In this work, we recycled waste plastic bottle caps made of High-Density Polyethylene (HD-PE) and used it as a matrix for different types of composite materials which are the following; natural Pozzolan powder/recycled HD-PE composite, short Alfa fiber/recycled HD-PE composite, and hybrid composite combining short Alfa fiber plus natural Pozzolan powder.

2.2.1 Materials

Alfa plant

Recently, an increasing interest has been focused on the usage of Alfa fibers as reinforcements in bio-composite materials. It is commonly used with polyolefin polymers especially thermoplastics due to their low price and their recyclability without any risks of serious damage, unlike thermoset polymers. Moreover, the thermoplastics processing temperature is less than 220°C which allows to avoid thermal degradation of natural fibers [74]. A study conducted by Sahar Salem et al [131], has shown that Alfa fibers-reinforced HD polyethylene composites possess higher performance than wheat straw, cornstalk, and corncob-reinforced HDPE composites concerning the tensile strength, its strength increases with increasing treated Alfa fibers content.

Alfa fibers are originally found in the arid and semi-arid regions of North Africa and Spain, Portugal, and the Balearic Islands (Alfa is the Arabic name of Esparto, Scientific is Stipatenacissima L). The Alfa plant belongs to the Gramancie family which is a xerophilous herb that exists in the form of bundles constituting stems that grow to a height of about one meter Figure 2.1, this plant does not need insecticides nor pesticides or much water to grow as it plays an important role in withstanding desertification. Hence, these species are neglected due to the misunderstanding of their economic and technical importance. This grass is mostly used in the production of handcrafted goods and paper of superior quality [7, 49].

Figure 2.1: Alfa fibers plant bundel in nature [153].

Morphology of Alfa fibers

Alfa grass grows naturally in bundles (tussocks). Green leaves, which can last up to 1.5 years and are continuously produced, form the bundle. Together with the dead leaves, they give the tussock its round shape. the leaf can open or close across the length depending on the relative humidity in the atmosphere, according to that it can fold along its long axis hiding The grooved surface [44].

The stems (leaves) have a length of 25 to 120 cm depending on weather conditions, it includes an underground section and another airline. The rhizome, the underground portion Figure 2.2, is made up of a complex network of deeply rooted seedlings that are 2 mm in diameter and 30 to 50 cm deep [49].

The aerial region is made up of several branches that are wrapped into one another and ended with long leaf blades. The upper face of the leaf has high ribs, and the underside is slightly brilliant. The cylinder-shaped, hollow stem is regularly interrupted at the node level by beam entanglements.

Generally, the esparto flowers towards the end of April at the beginning of May, they are green in color. The fruit is a caryopsis (a kind of grain) which measures 5 to 6 mm in length. The flower is protected by two glumes of equal length. The upper lemma seems partially separated into 2 parts and the lower lemma is finer [153].

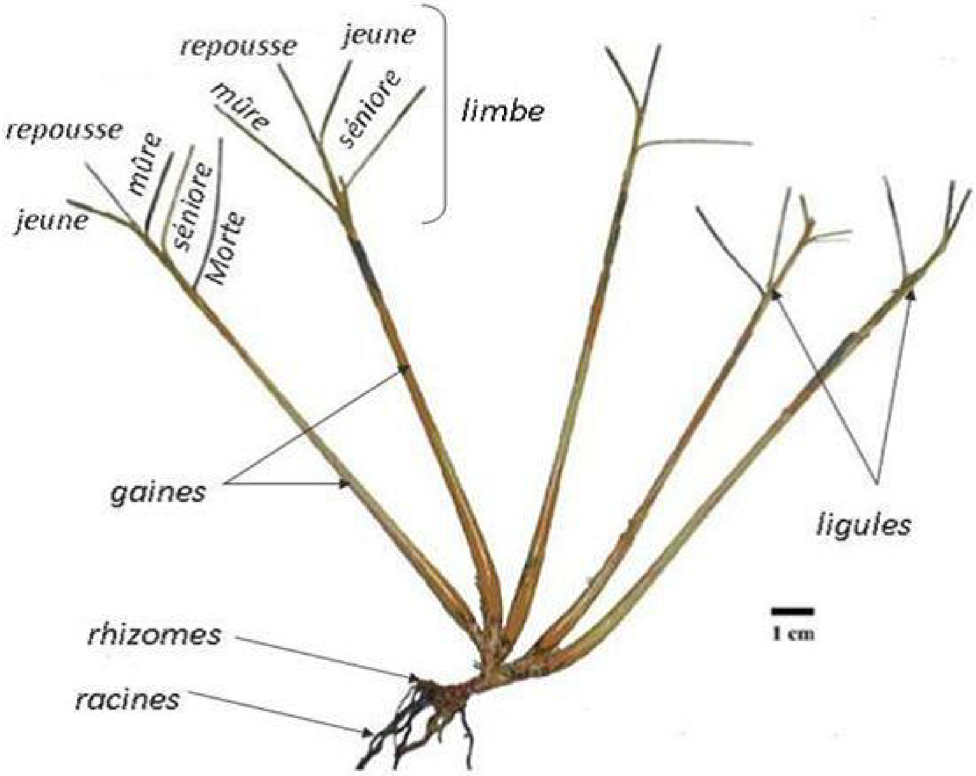


Figure 2.2: The morphologie of alfa plant [153].

Karama Elfehri Borchani et al [25], mentioned after an electronic scanning microscopic views of longitudinal and cross-sections of Stipatenacissima fiber that Alfa stem surfaces were found to be smooth, the long fiber is composed of bundles (nanotubes) bounded together with lignin and the diameter is about 200 μm in section. The SEM also shows that a singular fiber constituting the bundle consists of microfibrils.

A.Boukhoulda et al [29], have stated after a scanning electron microscope of the section and longitudinal part of Alfa leaf. the internal structure is composed of fibril bundles that are covered with layers of non-cellulosic components (trachoma, cuticles, and waxes) which are arranged and assembled. The wall layer is thick and each ultimate bundle is roughly polygonal or cylindrical with different diameters and contains a central lumen (holes), see Figure 2.3.

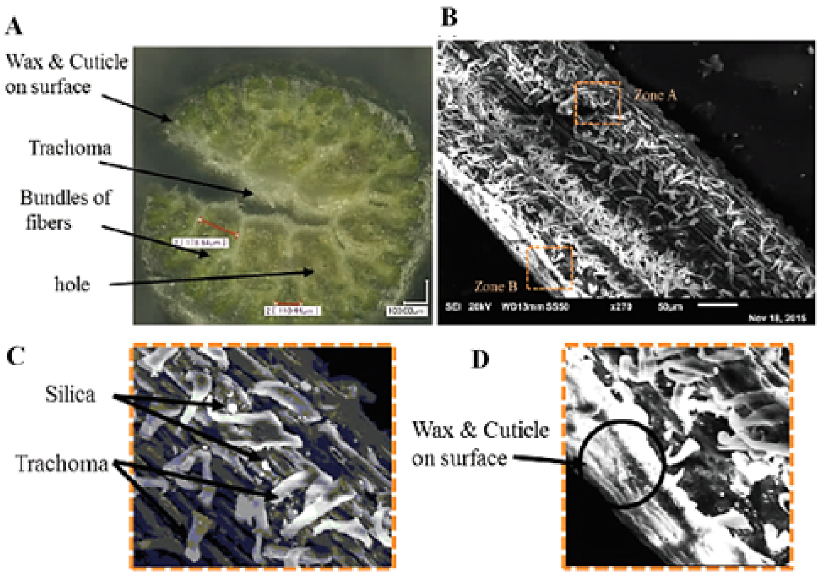


Figure 2.3: SEM and optical observations: (A) Longitudinal cut of the leaf, (B) SEM observation of Alfa bundle, (C) SEM observation of trachoma and silica (Zone A) and (D) SEM observation of waxes and cuticle (Zone B) [29].

Chemical composition and mechanical properties of Alfa fibers

Alfa leaves are composed of cellulose (43.8% to 47%), hemicellulose (22.15% to 28.4%), and lignin (17.7% to 24%) [96]. Table 2.1 presents the chemical composition and the percentage proportions of each substance (cellulose, lignin, hemicelluloses, pectin, and wax) of the most widely used vegetable fibers [38].

Table 2.1: Chemical composition (in %) of different plant fibers [38].

Fibers	Cellulose	Hémicelluloses	Lignine	Pectine	Cire
Coton	85-90	5-7	0.7-1.6	0-1	0.6
Lin	71	18.6-20.6	2.2	2.3	1.7
Chanvre	70-74	17.9-22.4	3.7-5.7	0.9	0.8
Jute	61.1-71.5	13.6-20.4	12-13	0.2	0.5
Ramie	68.6-76.2	13.1-16.7	0.6-0.7	1.9	0.3
Sisal	66-78	10-14	10-14	10	2
Coco	32-43	0.15-0.25	40-45	3-4	-
Alfa	45	24	24	5	2

Alfa fiber has Young's modulus which is about (18.42 GPa to 24.92 GPa) and a tensile strength of about (187.6 MPa to 308 MPa) [96]. Table 2.2 shows a comparison of the specific mechanical properties of the most used plant fibers.

Table 2.2: Tensile mechanical properties of some plant fibers [38].

Fibers	E(Gpa)	All(%)	σ (MPa)	Density
Coton	5.5-12.6	7-8	287-597	1.5-1.6
Lin	58	3.27	1339	1.53
Chanvre	35	1.6	389	1.07
Jute	26.5	1.5-1.8	393-773	1.44
Ramie	61.4-128	1.2-3.8	400-938	1.56
Sisal	9-21	3-7	350-700	1.45
Coco	4-6	15-40	131-175	1.15
Alfa	12.7	1.6	75-154	1.51

Natural pozzolan

as a mineral filler, the Pozzolan particles can be obtained from a pyroclastic rock (natural pozzolan) or artificially manufactured. Many industries have used inorganic fillers such as clay, black carbon, talc, and calcium carbonate for polymer-based composites due to their availability and low cost. They were found useful in improving certain mechanical properties and dimensional stability [45].

Natural pozzolans can be created when molten magma cools after being ejected into the atmosphere during violent volcanic eruptions, thus called volcanic origin pozzolan [151].

The second category of natural pozzolan includes diatomaceous earth and clays. The pozzolanic reactivity of clays is very low unless they are thermally treated. Diatomaceous earth, which is a sedimentary rock, consists basically of the fossilized remains of diatoms (a type of algae) [151].

Another type of natural pozzolan is called diagenetic, rich in amorphous silica, resulting from the weathering process of siliceous rocks [13].

Finally, the tuffs where volcanic glass has been transformed, entirely or partially, into zeolitic compounds [13].

Pozzolans have been in use for a very long time. The structures built using "Roman cement and mortars" made of pozzolan and lime, including pantheons, coliseums, stadiums, basilicas, aqueducts, bridges, and a variety of other buildings, have survived to the present day and represent priceless artifacts of Roman civilization [116].

Chemical composition of Natural pozzolan

Algeria has a significant amount of Pozzolanic materials of volcanic origin existing along 160 km between the Algerian-Moroccan border and the Sahel of Oran. The natural Pozzolan powder is extracted from a siliceous volcanic rock of variant colors ranging from red to black. Yassine Senhadji et al, study the use of local natural pozzolan (NP) as a mineral filler. The mineralogical composition of the natural pozzolan was determined by X-ray diffraction and is presented in Table 2.3 [133].

Table 2.3: Chemical composition of natural Pozzolan [116].

Composition	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	LOI
wt%	47.21	18.85	9.99	10.84	4.38	0.50	0.20	0.81	3.91

The physical extraction of Alfa short fibers and Pozzolan powder

The alfa fibers used in this study were extracted from the Stipatenacissima (Esparto grass plant) that have been harvested from the state of Ain Temouchent located in the North West region of Algeria. In the physical preparation, Alfa (esparto grass) were cut into small pieces of approximately 1 to 2 cm in length and then dehydrated in an oven (MEMMERT) at 105° for 48h. The little pieces were crushed with a blade crusher (MA garant) and recovered to get sifted in a vibratory sieve of 125 μm (RETSCH TYPE AS 200 amplitude 3 mm/g). Figure 2.4 shows the physical preparation of Alfa fibers.

Figure 2.4: Physical preparation of the Alfa fibres, (A) The Alfa stems gathered.(B) The Cut Alfa stems. (C) The Alfa fibres collected from a 250 μm diameter sieve.

the north-west of Algeria (reserves exceeding 18 million tons). The Pozzolan powder used in this study of was extracted from a volcanic rock that has 45–50% silica and 15–20% alumina. A (Retsch Jaw Crusher BB 100 w/Mn Steel Jaws & SS Plates, 220 V 60 Hz) has been used to crush the pozzolan rocks as illustrated in Figure 2.5 The obtained powder was sifted with a 250 μm diameter sieve (RETSCH TYPE AS 200 amplitude 3 mm/g).

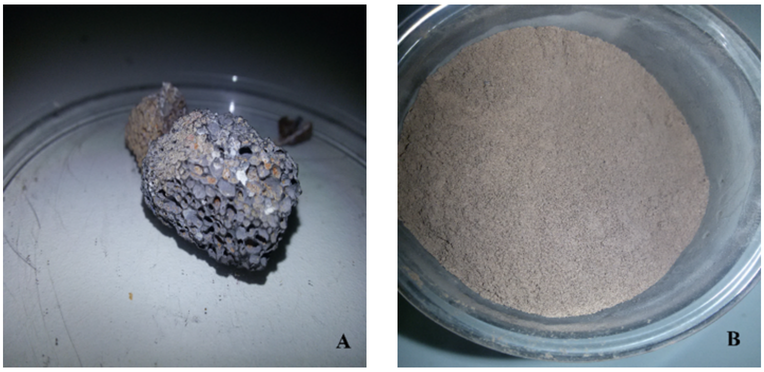


Figure 2.5: Physical preparation of Pozzolan, (A) Pozzolan rock (B) Pozzolan powder.

The preparation and elaboration of the bio-composites

The work was done manually where waste plastic bottle caps were reincorporated as a thermoplastic matrix for the bio-composites, these bottle caps that are made of high-density polyethylene were grind by an industrial knife crusher. The resulting shredded plastic bits were heated under 200°C on a laboratory hot plate. When the heated plastic takes the form of a paste, the heating temperature is reduced to 180°C. Then, the alfa short fibers along with the Pozzolan powder were added to the matrix and blended manually. The composites were finally molded at 50°C through the compression molding method. The experiment steps were repeated following the different fractions of each component for each composite as indicated in the following (Tables 2.4, and 2.5).

Table 2.4: list of the samples produced with their composition for PZ/HDPE composite and Alfa/HDPE composite.

	PZ/HDPE composite		**Alfa/HDPE composite**	
Sample code	**HDPE content (wt%)**	**PZ content (wt%)**	**HDPE content (wt%)**	**Alfa fibres content (wt%)**
95 :5	95	5	95	5
90 :10	90	10	90	10
85 :15	85	15	85	15
80 :20	80	20	80	20
75 :25	75	25	75	25
70 :30	70	30	70	30

HDPE : High DensityPoly-ethylene, **PZ**:puzzolana

Table 2.5: list of the samples produced with their composition for PZ+Alfa/HDPE hybrid composite.

Sample code	**HDPE content (wt%)**	**PZ content (wt%)**	**Alfa fibres content (wt%)**
0 :0	100	0	0
5 :25	70	5	25
10 :20	70	10	20
15 :15	70	15	15
20 :10	70	20	10
25 :5	70	25	5

HDPE : High DensityPoly-ethylene, **PZ**:puzzolana

The obtained molded specimens were machined using numerically controlled (NC) miller (KOSY A3S controlled with MegaNC-2D-2005) as shown in Figure 2.3

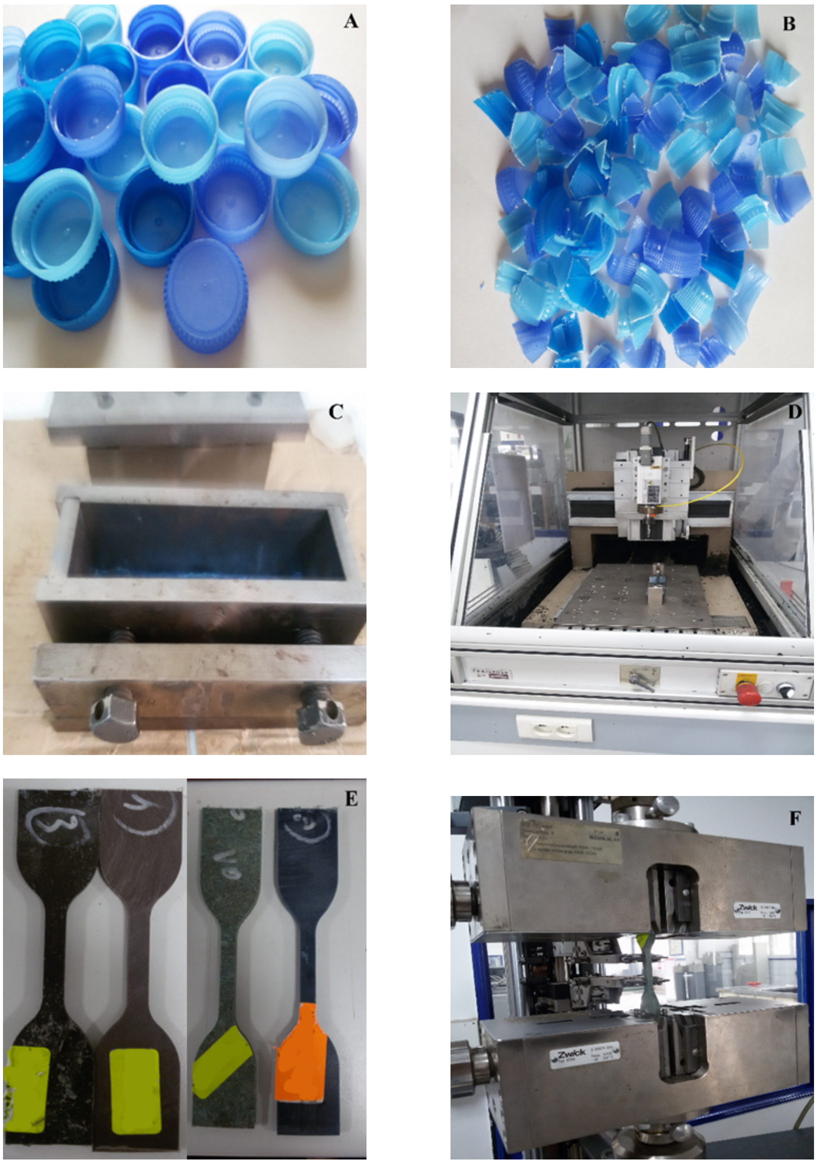


Figure 2.6: The processing steps of the Bio-composites. (A) Bottle caps waste. (B) Grinded bottle caps. (C) The molding step. (D) The machining of the platelets. (E) Samples of the produced bio-composites. (F) The tensile testing of samples.

2.3 Caracterisation of the biocomposites

Samples of the produced biocomposites were characterized, by initially using the Fourier Transform Infrared Red (FTIR) spectroscopy to evaluate the structural properties of the biocomposites components. Then, we obtained the mechanical properties of the samples by a Tensile Test. Finally, Scanning Electron Microscopy (SEM) was applied to acquire some information on the morphology of the composite.

2.3.1 Attenuated Total Reflection (ATR)-Fourier Transform Infra-Red (FTIR) analyses

The structural properties of the used fillers (Pozzolan and Alfa) were evaluated using A Thermo Scientific Nicolet Summit FTIR Spectrometer with the Everest ATR accessory configured for Specular Reflectance. The spectra were obtained with an accumulation of 16 scans with a resolution of 4 cm-1.

Tensile test

The tensile test was conducted on eighteen specimens according to the ISO 6259-1 standard. The test was performed on a universal testing machine ZWICK Z100 at a transversal cross head speed of 3 mm/m in an ambient temperature using a 10KN load sensor as illustrated in (Figure 2.6.F), wherein stress-strain curves were generated. The test ended when the specimens fractured.

The toughness (U_T) is calculated from equation (2.1) as follows:

$$U_T = \int_0^{\varepsilon_f} \sigma_e \, d\varepsilon_e \tag{2.1}$$

Where σ_e is the engineering stress, ε_e the engineering strain, and ε_f the fracture strain of the specimen, respectively.

Moreover, the modulus of resilience (MR) is calculated from equation(2.2):

$$M_R = \int_0^{\varepsilon_y} \sigma_e \, d\varepsilon_e \tag{2.2}$$

In which σ_e is the engineering stress, ε_e the engineering strain, and ε_y the yield strain at which linear elastic behavior ceases.

Scanning Electron Microscopy (SEM)

The morphology of the bio-composite was investigated using an electronic microscope Hitachi TM1000 electron. The tested specimens have been observed to analyze the fractured surfaces and the state of the components interface.

2.4 Results and discussion

2.4.1 ATR-FTIR Analyses

The Attenuated Total Reflectance (TAR) has been used to investigate the structure of the Alfa fibers and the Pozzolan powder in order to determine their composition.

FTIR spectroscopy allows measuring the variations of the Alfa compositions. The Alfa fibers contain respectively 46 % in weight of celluloses, 19.9 % of lignin, and 30.2 % of hemicelluloses. The Infra-Red Spectra of Alfa fibers are illustrated in (Figure 2.8.A). The intensive bands observed in the FTIR spectra in the alfa fibers are hydroxyl groups of H-O celluloses with wider bands of approximately 3334 cm-1 [75]. The vibration of C-H of CH and CH2 appears at 2918 cm-1 and 2850 cm-1 which is due to the celluloses- hemicelluloses [117].

The instance band at 1731 cm-1 corresponds to the stretch of the C=H of the carboxylic acids and ester groups [25]. A little peak is observed at 1620 cm-1 associated with the presence of water [55]. The other picks at 1515 cm-1 and 1455 cm-1 are attributed to the C=C vibration (aromatic skeleton) and to the deformation of C-H respectively which are present in the lignin [25]. The peak of about 1164 cm-1 is due to C-O-C asymmetric vibration of β-glycosidic linkage [3] . The wider band with a shoulder at 1030 cm-1 corresponds to stretched ring C-O in celluloses and hemicelluloses [25].

(Figure 2.8.B) shows the FTIR of the natural pozzolan used. A wide intensive band is observed at 3566 cm-1 and 1632 cm-1 due to the O-H bending of the absorbed water molecule H2O at the surface [4, 81]. The wide band at approximately 1434 cm-1 is associated with the stretching vibrations of O-C-O which is the result of atmospheric carbonation [4, 81]. Another wider band at 1004 cm-1 is related to the asymmetric stretching of the bridging oxygen atom in the T-O bond of the (Si-O-Si, Si-O-Al) of the aluminosilicate structure [81]. An absorption band located at 712 cm-1 corresponds to the asymmetric stretching of the AL-O bond in (AIV)-O-AIV [54].

2.4.2 Mechanical properties of composites

The mechanical properties of the three types of NFRTCs were determined using a tensile test and compared versus different content of reinforcement. The mechanical characteristics gathered from the tensile stress-strain curves are tensile strength, tensile strain, young modulus, toughness, and modulus of resilience.

Figure 2.10 illustrates the Stress-Strain curves of the three types of NFRTCs Alfa/recycled HD-PE, PZ/recycled HD-PE, and Alfa+PZ/recycled HD-PE.

In (Figures 2.10.A, and B) the addition of both reinforcement (PZ or Alfa) to the neat recycled HD-PE shows significant improvement in tensile strength of 35,73 MPa and 30,099 MPa for Alfa /recycled HD-PE and PZ/recycled HD-PE composites respectively, a gain of about 54% and 16% compared to the neat recycled HD-PE.

It can be observed from (Figure 2.10.C) that the hybrid composite Alfa+PZ/recycled HD-PE tensile strength indicates a considerable increase of 30.65 MPa for the sample

(10:20) this value represents a 19% increase in contrast to the neat recycled HD-PE.

It is interesting to see in (Figure 2.12.A) the high values of Young's modulus for the three types of composites compared to the neat recycled HD-PE. This enhancement in Young's modulus is the consequence of the strong and stiff natural reinforcement that was added to the matrix. The maximum results that were attended by the three types of composites are as follows; the PZ/recycled HD-PE composite has reached a value of 2193 MPa which is a difference of 39% compared to the neat recycled HD-PE. Alfa/recycled HD-PE composite represents a maximum value of 3696 MPa a change of about 134% in contrast to the neat recycled HD-PE of Young's modulus for the Alfa/recycled HPDE composite.

Otherwise, the hybrid composites reached a value of 3449.3 MPa of Young's modulus And it's 118% higher than that of the neat recycled HD-PE.

The improvement of Young's modulus of the three types of composites is associated with the stiffness of the reinforcements (PZ powder and Alfa fibers). It is also due to the good adherence to the interface.

The toughness (U_T) of a material is its ability to absorb energy in the plastic range above the yield stress without fracturing, and it is measured by integrating the area under the stress-strain curve. (Figure 2.12.B) represents the relation between U_T and (short alfa fiber, natural pozzolan powder) concentration. The three types of biocomposites showed a significant decrease in U_T when compared to neat HD-PE. This decrease in U_T may be attributed to the fact that short Alfa fibers and natural pozzolan powder acted as stress concentrators. On the other hand, it can be seen that PZ/HD-PE composites are tougher in contrast with Alfa/HD-PE and PZ+Alfa/HD-PE composites. This indicates that PZ/HD-PE composites are more ductile and have a greater total elongation.

The area up to the yield point in the stress-strain curve is called the Modulus of resilience (MR) and it is defined as the material's ability to absorb energy when deformed elastically. (Figure 2.12.C) exhibit the modulus of resilience as a function of pozzolan particles and alfa short fibers at different concentrations. The three types of biocomposites reveal an increase in MR by contrast to neat HD-PE. This increase in resilience is attributed to the increase in yield strength of these biocomposites. Higher MR values are shown for Alfa/HD-PE composites.

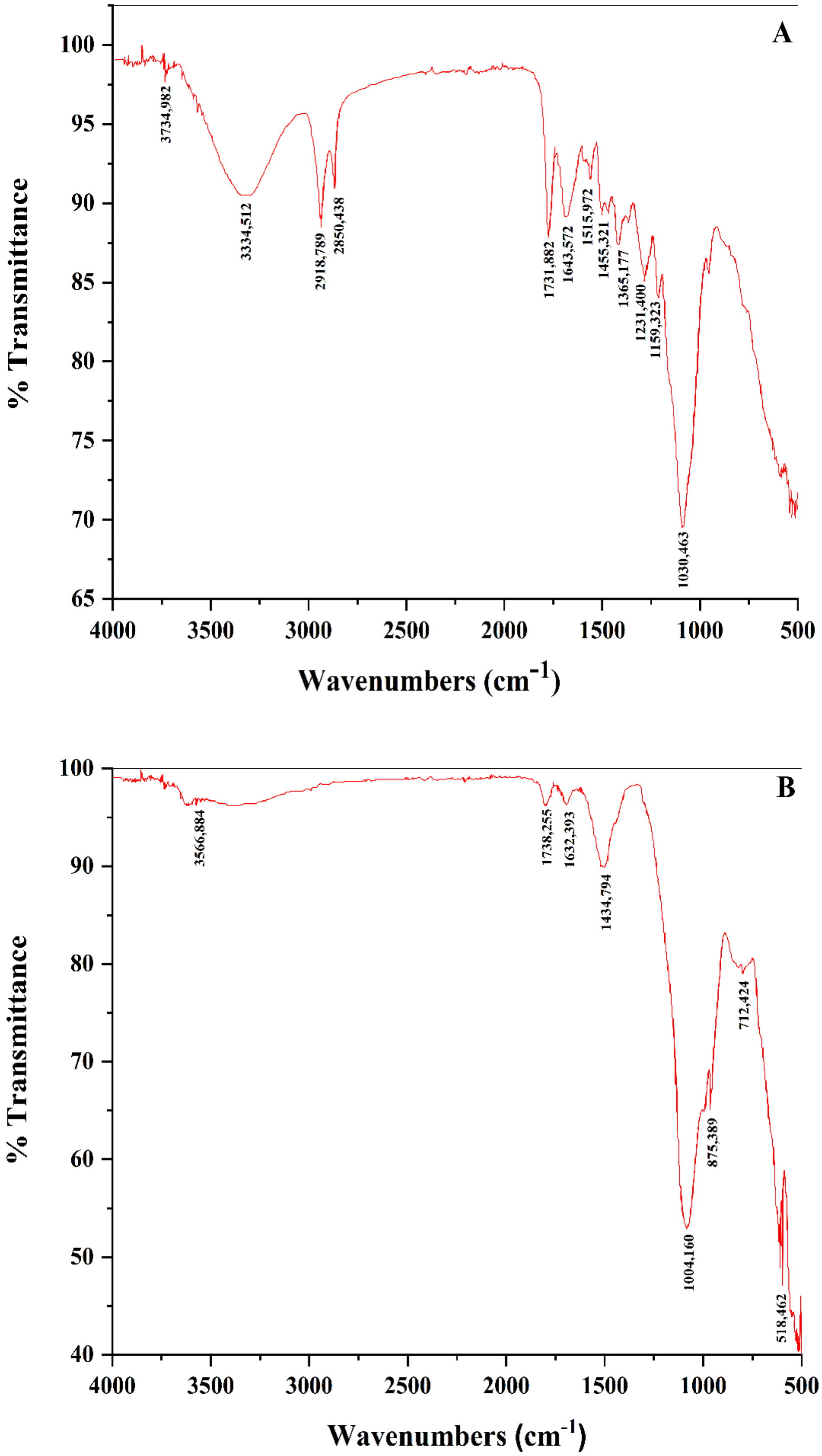
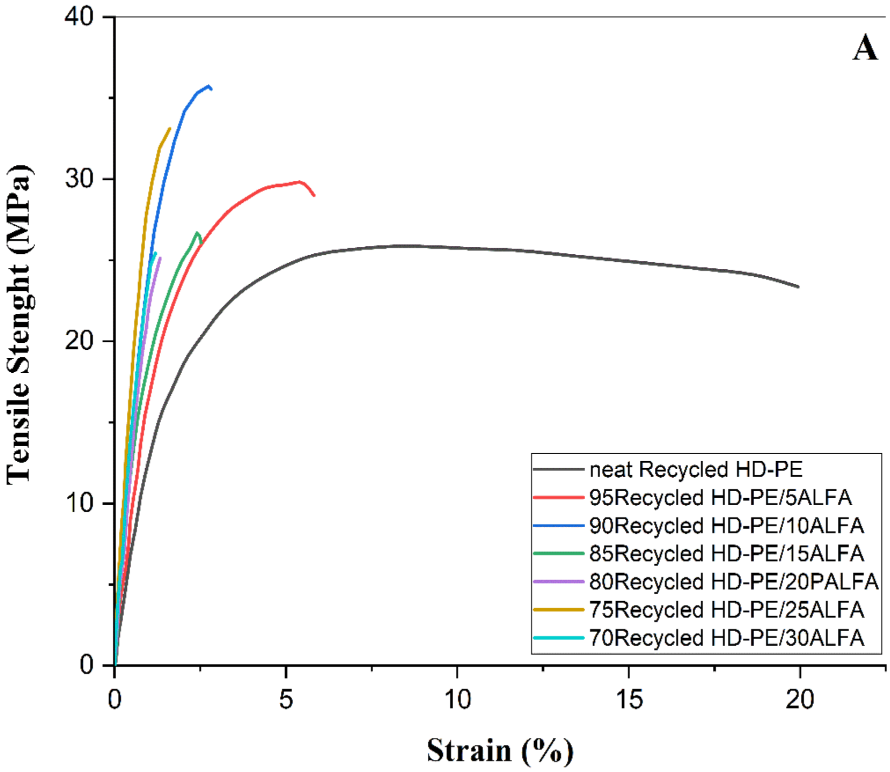


Figure 2.8: FTIR measurements of (A) raw Alfa fibers. (B) natural Pozzolan.


A
Tensile Stenght (MPa)
Strain (%)
0
10
20
30
40
0
5
10
15
20
neat Recycled HD-PE
95Recycled HD-PE/5ALFA
90Recycled HD-PE/10ALFA
85Recycled HD-PE/15ALFA
80Recycled HD-PE/20PALFA
75Recycled HD-PE/25ALFA
70Recycled HD-PE/30ALFA

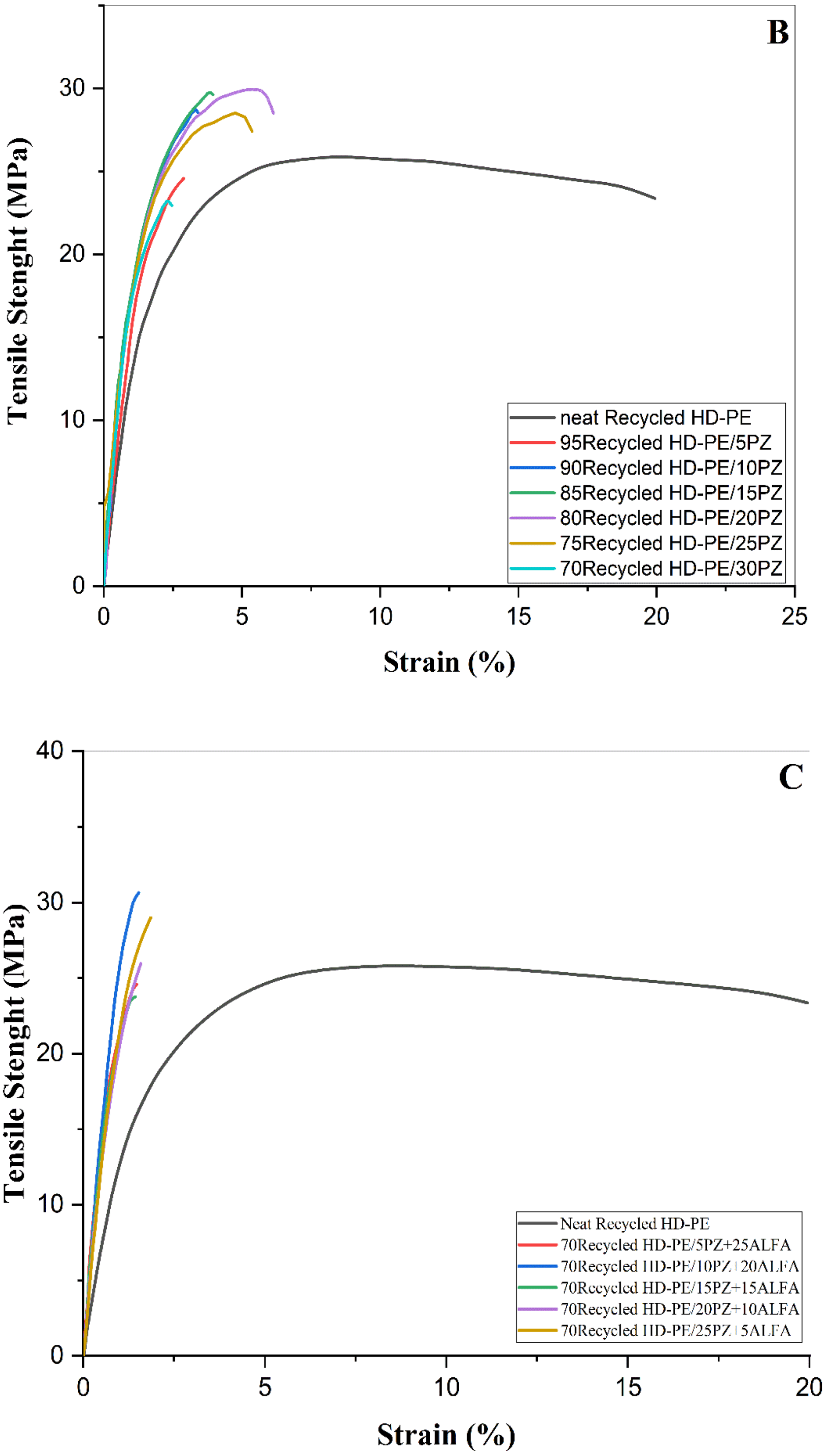
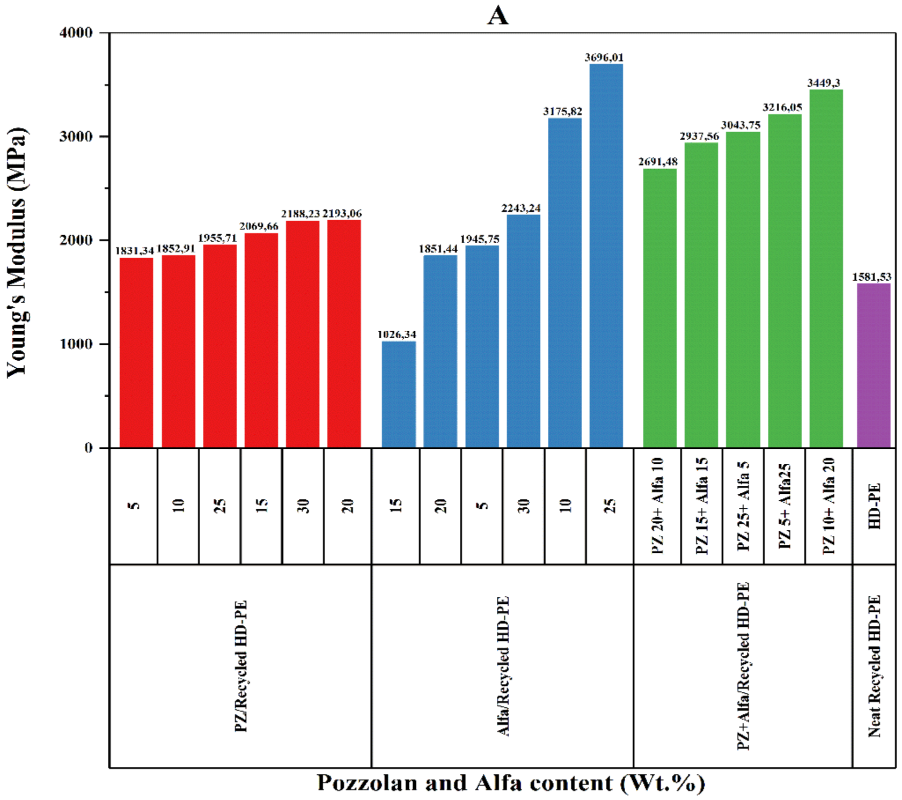
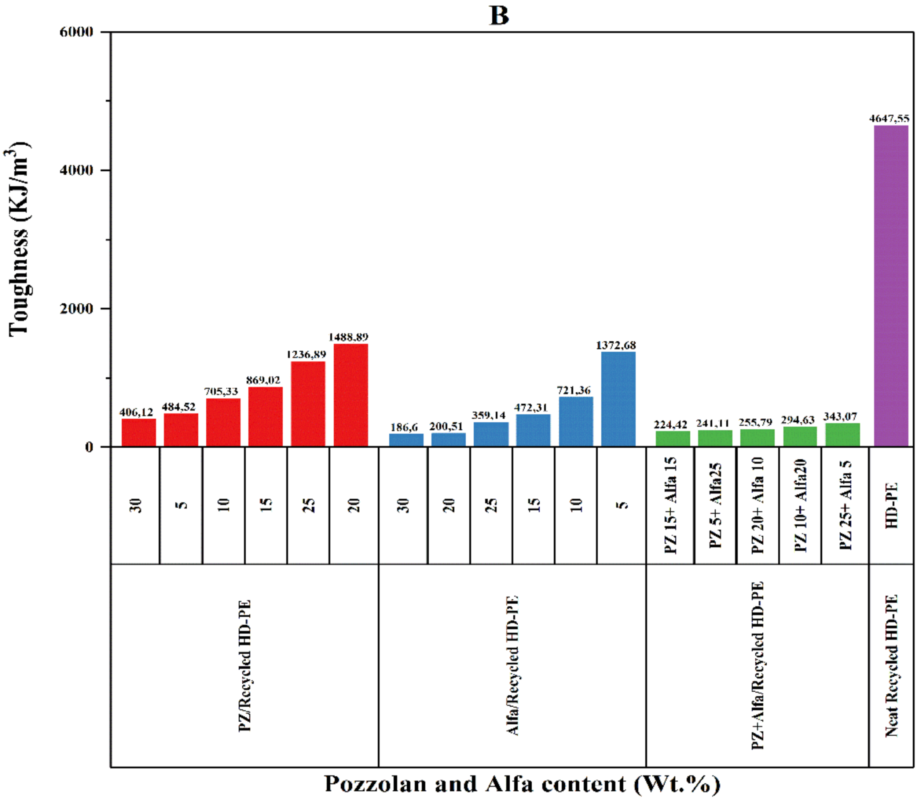


Figure 2.10: stress-strain curves of (A) Alfa/recycled HDPE composites, (B) PZ/recycled HDPE composites, (C) hybrid composites Alfa+PZ/recycled HDPE.


A
Young's Modulus (MPa)
0
1000
2000
3000
4000
1831,34
1852,91
1955,71
2069,66
2188,23
2193,06
1026,34
1851,44
1945,75
2243,24
3175,82
3696,01
2691,48
2937,56
3043,75
3216,05
3449,3
1581,53
5
10
25
15
30
20
15
20
5
30
10
25
PZ 20+ Alfa 10
PZ 15+ Alfa 15
PZ 25+ Alfa 5
PZ 5+ Alfa25
PZ 10+ Alfa 20
HD-PE
PZ/Recycled HD-PE
Alfa/Recycled HD-PE
PZ+Alfa/Recycled HD-PE
Neat Recycled HD-PE
Pozzolan and Alfa content (Wt.%)


B
Toughness (KJ/m3)
0
2000
4000
6000
406,12
484.52
705,33
869,02
1236,89
1488.89
186,6
200,51
359,14
472,31
721,36
1372,68
224,42
241,11
255,79
294,63
343,07
4647,55
30
5
10
15
25
20
30
20
25
15
10
5
PZ 15+ Alfa 15
PZ 5+ Alfa25
PZ 20+ Alfa 10
PZ 10+ Alfa20
PZ 25+ Alfa 5
HD-PE
PZ/Recycled HD-PE
Alfa/Recycled HD-PE
PZ+Alfa/Recycled HD-PE
Neat Recycled HD-PE
Pozzolan and Alfa content (Wt.%)

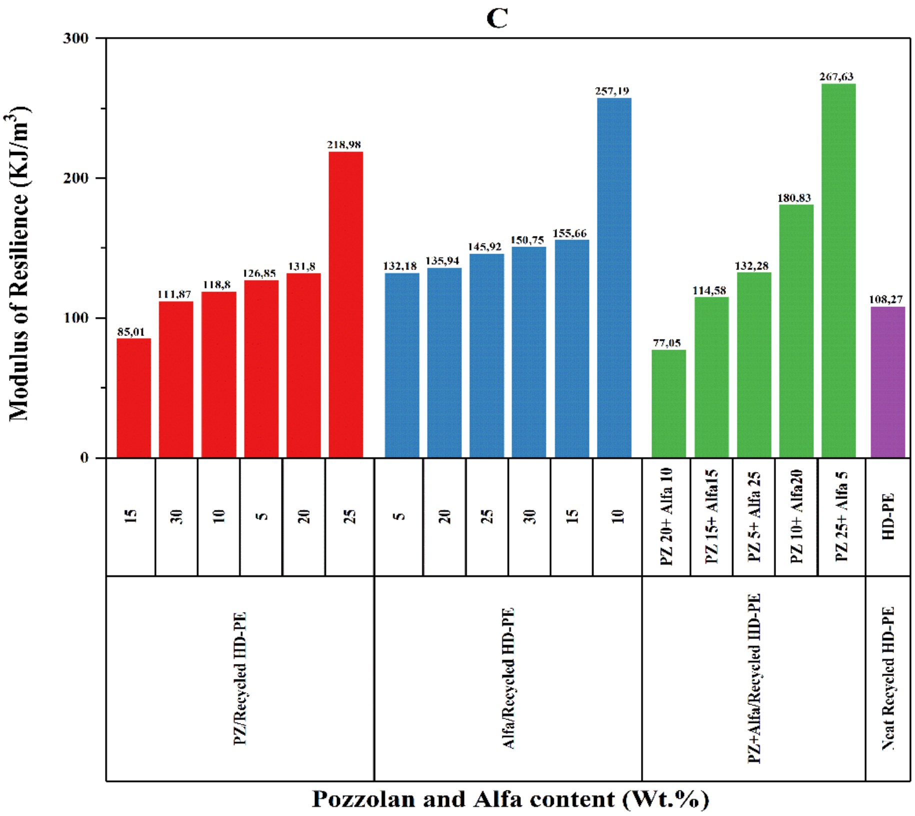


Figure 2.12: Effects of filler type and content on (A) Composites young's modulus, (B) Composites toughnes, (C) Composites modulus of resilience.

2.4.3 Morphological structure

A Scanning Electron Microscopy (SEM) analysis is carried out on the specimens of the three composites respectively alfa/HDPE, PZ/HDPE, and Alfa+PZ/HDPE to investigate the dispersion and distribution of the fillers in the matrix and the mechanical behavior of the bio-composites.

Figure 2.13 shows the micrographics of the fractured surfaces after the tensile test. Few voids have been observed due to the tearing provoked by the tensile load [6].

Otherwise, after the fracture of the specimen, most of the Alfa fiber residuals adhered to the matrix revealing the strong interfacial adhesion. This explains the improvement of the resistance to the traction as was reported in the mechanical analyses [28]. The SEM images also show the good dispersion of Pozzolan particles and Alfa fibers in the polymeric matrix.

In (Figure 2.13.B), it can be observed the penetration of the matrix through the fibrils presents a good interaction between the fibers and the recycled HDPE matrix as assured and illustrated in (Figure 2.13.C) showing the gluing between the polymeric matrix on the surface of the fibers [55].

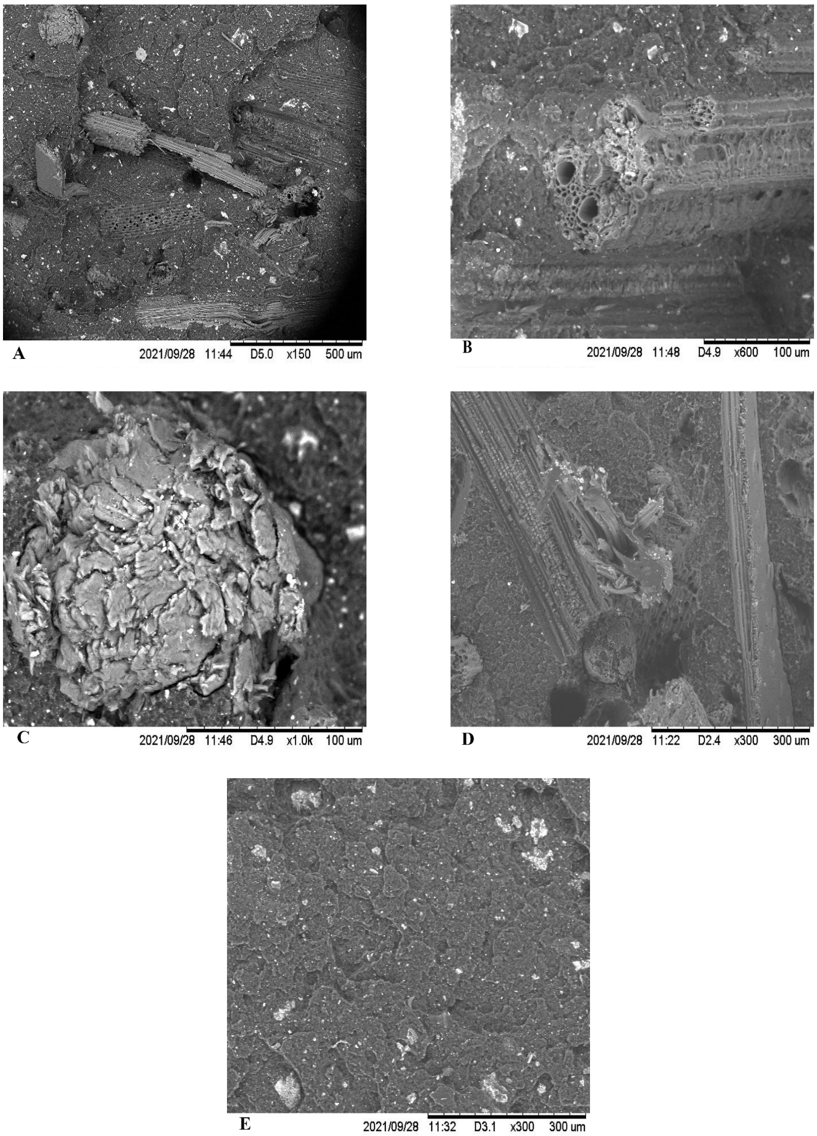


Figure 2.13: : SEM micrographs of fractured surfaces of samples : (A,B,C) Alfa+PZ/HDPE, (D) Alfa/HDPE, (E) PZ/HDPE.

2.5 Conclusion

The main aim of this experimental study is the reduction of plastic wastes and taking advantage of the existing abundant natural resources with excellent mechanical properties, to develop bio-composite materials with excellent and exceptional properties. Three types of new bio-composites have been successfully elaborated. waste plastic bottle caps made of high-density polyethylene (HD-PE) have been reused as a matrix for these bio-composites. The Alfa short fibers and natural Pozzolan powder have been incorporated as reinforcements with different fractions. Morphological and mechanical analyses were carried out to investigate the effects of Alfa short fibers and natural Pozzolan powder on the mechanical properties of the bio-composites.

The results of this study confirmed that the use of natural filler improves the mechanical properties of the recycled HPDE. The tensile tests showed an improvement in Young's modulus, tensile strength, and the modulus of resilience of the three developed bio-composites, on the other hand, a decries in toughness was noticed. The results vary with the variation of reinforcements content.

An increase in tensile strength reached the maximum of 35,73 Mpa plus an interesting improvement in young's modulus with a maximum value of 3696 MPa. The toughness of the neat recycled HD-PE decreased by 212% by adding the natural filler whereas the modulus of resilience exhibited an increase of 138% compared to the neat recycled HD-PE.

The obtained results indicate that the recycling of the HPDE reinforced with Alfa fibers and Pozzolan powder can represent a good potential in improving interfacial properties. The study shows an alternative for producing friendly bio-degradable composites that can be a solution for plastic waste reduction and a substitute for petrochemical-derived products.

Chapter 3

A numerical study for determining the effect of raffia and alfa fibers on the fiber-matrix interface damage of biocomposite materials

This chapter develops a part of the article

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3.1 Introduction

In composite materials, the interface fiber-matrix has an important role in ensuring the transfer of applied load from the matrix to the fiber, and for this reason, the choice of fiber quality becomes more than necessary, the fiber-matrix interfacere mains the most delicate area for controlling the resistance of composite material, understanding this part of the composite material and how it affects the final mechanical properties of the product is essential.

In the following chapter, an investigation of the effect of raffia, alfa and sisal fibers on fiber matrix interface damage of biocomposite materials was pursued; raffia/PLA (polylacticacid), alfa/PLA, and sisal/PLA subjected to the same mechanical shear stress, using a genetic algorithm. To calculate interface damage using fiber and matrix damage, the genetic operator crossing has been employed based on the Weibull approach.

3.2 Materials and Methods

In this study, Raffia, Alfa, and sisal fibers have been the subject of physical and mechanical characterization. Furthermore, three types of biocomposite materials were subjected to shear tests; raffia/PLA, alfa/PLA, and sisal/PLA. Finally, the genetic operator crossing is employed based on the fiber and matrix damage, to calculate the damage to the interface.

3.2.1 Materials

Raffia palms

Raffia palm fiber is a natural vegetable fiber like sisal, banana, and abaca, which is extracted from the leaflets of the raffia palm tree – Raphia Farinifera. There are 28 species in the species group Raphia, which is a member of the Arecaceae family, they are found only in tropical regions of Africa Figure. 3.1 [102]). Except one species named, R. Taedigera, which is endemic to Central and South America [149].

Raphia palms have a wide range of uses which include building materials, thatch roofing, and the production of palm wine. On the other side, Raffia leaves have several uses, ranging from the weaving of hats, baskets, mats, hammocks, and ceremonial costumes.

This palm tree can reach up to 20 meters in length, which is a record among palms, with 3–5 m long leaves/petiole [115]. The heart of young shoots is consumed in salads or ready meals in the country of origin. This palm tree owes its adjective 'Farinifera' to the flour which is obtained from it after processing and which is made into pan-

Figure 3.1: A sample of Raphia species. A: R. zamiana from northern Gabon. B. R. regalis from Gabon. C. R. vinifera from northwestern Cameroon, growing in maize and banana fields; D: Close up of a Raphiagabonica fruit. Photos Thomas Couvreur [102]

cakes [76, 111].

Raffia palm fibersare extracted from the leaflets of the raffia palm tree. These vegetal fibers are potential reinforcement materials for making cost-effective polymer-based composites, or to make geotextiles or lightening earthen bricks or concrete [21].

Chemical composition and morphology of raffia palm fibers

Raffia palm fibers(RPF)are composed mainly of cellulose (53 wt%), hemicellulose (13 wt%), and lignin (24 wt%). The remaining 10 wt% is attributed to other components (e.g. pectin, wax,and ash) [50].

From the SEM analysis of the RPF, it can be observed that the fibers are not roundish but longitudinal in shape [114]. The raffia textile fiber is made up of superimposing flat layers. In the longitudinal direction of the sample, The structure of the top surface the one in contact with air is a juxtaposition of parallel fibrils. The average width of these fibrils is about 10 μm and the average thickness is about 5 μm. Unlike other vegetable fibers, which are often smooth, these fibrils are formed by sorts of tiles or scales. Every tile is 6–20 μm long and covers the full width of the fibril. the part of the fiber, that is in contact with the body of the leaflet, has a honeycomb-like structure Figure 3.2. The diameter of the alveoli in the honeycomb network is in the range of 6–13 μm. They are separated by a wall about 0.5 μm thick [46, 47].

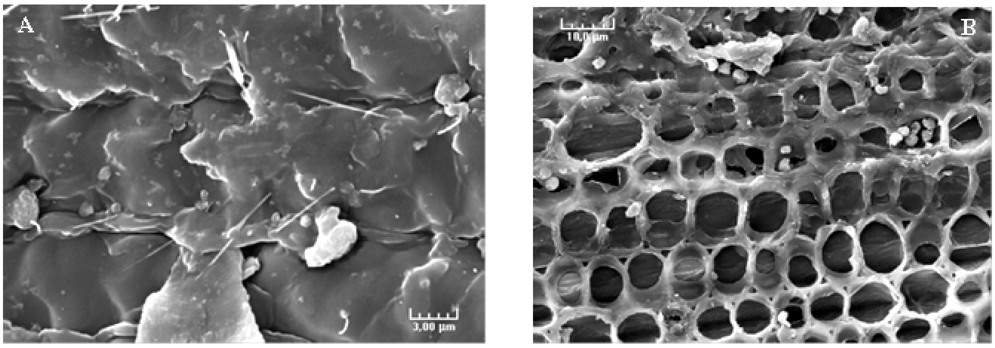


Figure 3.2: SEM examination of the structure of hookeri raffia fiber. (A) external face; (B) internal face.

Sisal plant

Sisal fibers complete name Agave Sisilana, originate from North and South America, Africa, and Asia. On average a single plant gives 5 to 6 leaves at a time and a single leaf contains 1000 fibers. A single leaf weighs about 600 g and reaches a length of 2 m, the leaf is composed of moisture (87.25%), fiber (4%), cuticle (0.75%), and other dry matter (8%). After two years of planting, the fibers can be extracted from the leaves. The plant has a 12-year productive life span, depending on the area, rainfall, and altitude, it can produce 120–240 leaves [70].

Sisal is a hard fiber that is tough, coarse, and resilient. It is one of the most widely used natural fibers in yarns, ropes, twines, cords, rugs, carpets, mattresses, mats, and hand crafted articles [53].

Sisa fibers are extracted by retting, scraping, or mechanical decortication. Mechanical decoration is the most used method Fig 3.3. The leaves are crushed using a blending knife the moisture and the fleshy pulp are removed from the fiber. After cleaning the fibers are dried in the sun.

Figure 3.3: extraction of sisal fiber: (A)Sisal plant, (B) Decortication , (C) Drying, (D) Raw fiber [70].

Chemical composition and morphology of sisal fibers

The length of the fiber varies between 1 and 1.5 m. The ends of the fiber are broad and blunt, the diameter is approximately 100±300 μm. Cells which are angular in shape are normally 500 to 6000 μm long and 5 to 40 μm wide. Calcium oxalate crystals are present in parenchyma circular and are often found packed with tiny globules as presented in Figure 3.4 [32].

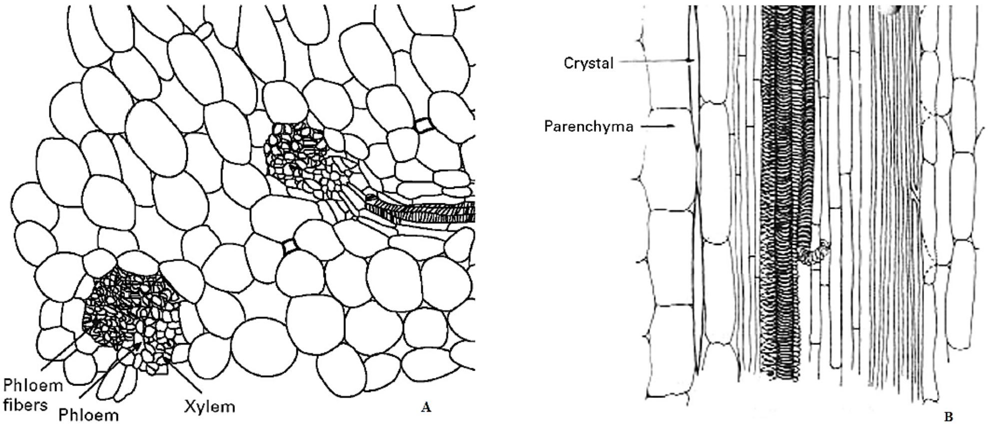


Figure 3.4: (A) The transverse section of sisal leaf shows phloem and xylem fibers; (B) longitudinal section shows parenchyma [32].

The fiber is a bundle of hollow sub-fibers, the cross-section of the fiber shows a built-up of about 100 fiber cells (each with a length of 2 to 5 mm),cell walls are reinforced with spirally oriented cellulose in a hemicellulose and lignin matrix. The fiber cells are linked together through middle lamellae, which consist of hemicellulose, lignin, and pectin.The external surface of the cell wall is composed of a layer of lignaceous material and waxy substances that bond the cells. Sisal fiber contains 78% cellulose, 8% lignin, 10% hemicellulose, 2% waxes, and about 1% ash by weight [89] Figure 3.5 present the section and transversal surface of sisal fiber.

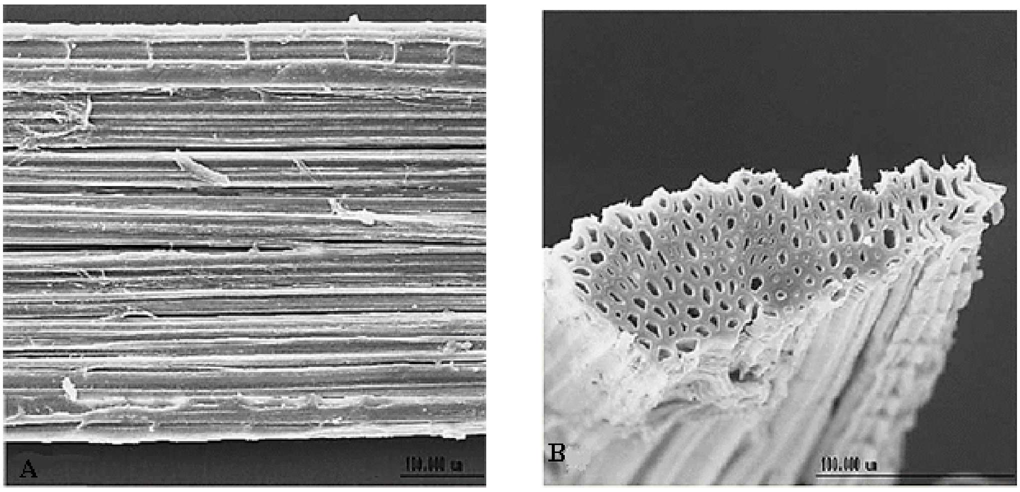


Figure 3.5: view of sisal fiber showing the porous nature of its micro-structure. (A) Surface and (B) Cross-section [142].

Mechanical properties of RPF, Sisal

Three vegetable fibers were used in this study, Table 3.1 shows the mechanical properties of Raffia, Sisal, and alfa fibers.

For RPF, Young's modulus is between 28-36 GPa while the tensile strength varies between 148-660 MPa, and the elongation at break is around 2%. The great variability of the stress at break can be explained by the presence of defects along with the sample (irregularity in thickness and width, presence of micro not cheslinked to handling during shelling or storage). On other plant fibers, a variation in mechanical properties was also observed, which is explained by the presence of defects and the variable structure of these materials. Indeed, the composition and structure of plant fibers depend on several factors: cultivation conditions, degree of maturity, method of extraction, length of the fiber, and water content [39, 47, 77].

For alfa fibers,Young's modulus is between 9-22 GPa while the tensile strength varies between 134-220 MPa and the elongation at break is around 1.5-2.4%. We have talked about this fiber in the previews chapter.

For Sisal fibers Young's modulus is between 13-18 GPa while the tensile strength varies between 80-840 MPa and the elongation at break is around 3–7%. Sisal fibers have the highest hardness, comparable to that of fiber glass; the close similarity with aramid fibers (kevlar, etc.) suggests good impact and breaking behavior. Therefore, its advantageous use for the manufacture of innovative energy absorption devices in various fields of industrial production such as automobiles, shipbuilding, and earth moving machinery [98].

Table 3.1: Mechanical properties of Raffia palms fibers, Alfa fibers, Sisal fibers.

Fibers	**Young's modulus (GPa)**	**Deformation at break (%)**	**Stress at break (MPa)**
Raffia palm [30, 47, 77]	28-36	2	148-660
Alfa [39, 77]	13-18	1.5-2.4	134-220
Sisal [18, 141]	9-22	3–7	80-840

Polylactic Acid Matrix

The PLA is synthesized through the polymerization of lactic acid, which was first discovered in 1780 by C.W. Scheelea Swedish chemist by examining curds and then commercialized for the first time in 1881 [87, 98]. Lactic acid is naturally present in many foods and can be produced by fermentation of various renewable resources such as corn, potatoes, beet sugar, and sugarcane [87, 98]. PLA has many interesting characteristics;its renewable resources, biocompatibility,biodegradation, good mechanical performance,and transparency of final material. Also, abroad spectrum of flow properties is available by simple architectural modification of PLA, thus allowing the use of this biopolymer in many transformation processes [51, 87]. PLA was then widely used in the biomedical, packaging, and textile fields [87, 110]. Thus, poly (lactic acid) is expected to compete with petrochemical polymers such as PET, PP, and PS. Unfortunately, the high cost, the lack of thermal and hydrolytic stability as well as the low resilience limit the use of this biopolymer. These faults are mostly overcome by mixing PLA with other polymers, and fillers, as well as chemical modification of the polymer. This literature study will assess recent developments made to improve the properties of PLA.

Chemical structure of PLA

Ring-opening polymerization (ROP), direct polycondensation in high boiling solvents (DP-S), and direct polymerization in bulk followed by chain extension with reactive additives are some of the process approaches that have been developed or are used on an industrial scale. ROP is the route that delivers by far the highest proportion of PLA chips available on the market. Polylactide or polylactic acid (PLA) is a synthetic, aliphatic polyester from lactic acid (LA), see Figure 3.6 [155].

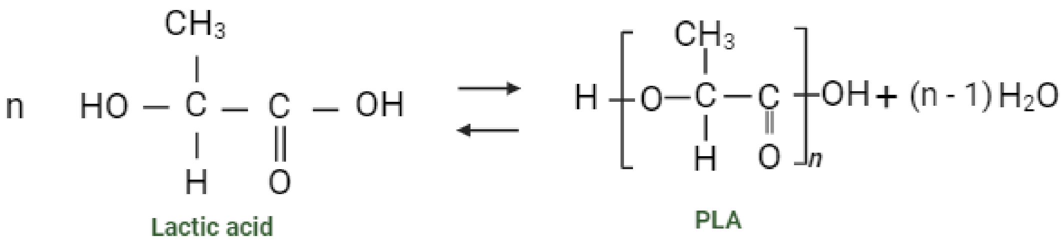
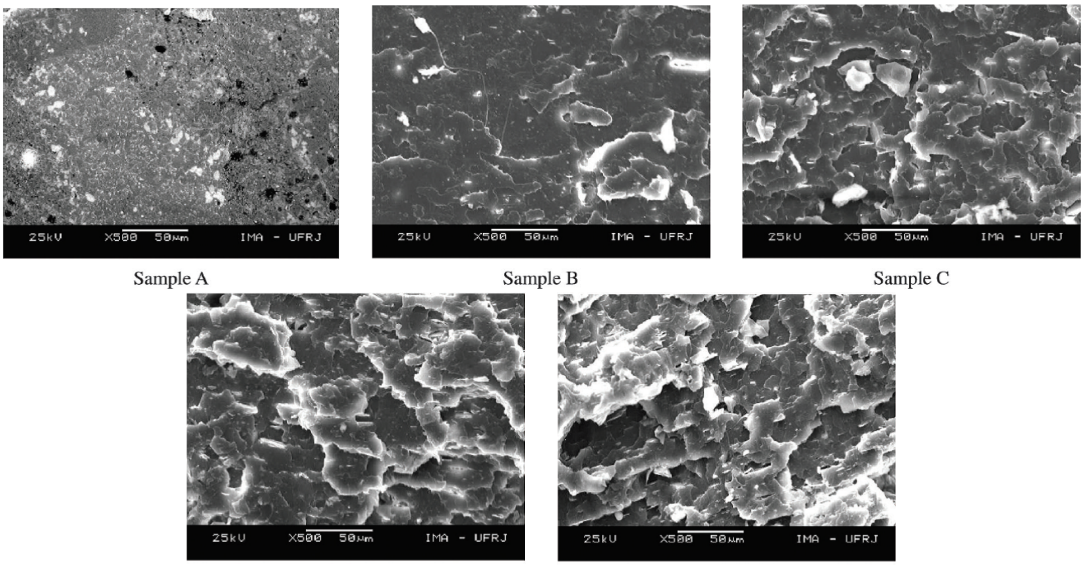


Figure 3.6: From lactic acid to polylactide [155].

Mechanical properties of PLA

PLA's Young's modulus and tensile strength are high, in the order of 3 GPa and 50-70 MPa, respectively. Such physical properties guarantee a wide range of applications to this material, comparable to that of polystyrene or PET and more important than HDPE and PP [58, 87]. Unfortunately, the mechanical properties of PLA are characterized by low impact resistance, as well as low elongation and brittle behavior. In Figure 3.7, Scanning Electron Microscope (SEM) Images of PLA fracture surfaces have been presented [36].



Sample A Sample B Sample C

Sample D Sample E

Figure 3.7: SEM imagesof PLA samples fracture surfaces [36]

3.2.2 Methods

Damage level

Chaboche (1988) [94] defined the damage of a finite element of a solid as follows:

S is the area representative volume element identified by its norm n

S_e is the effective resistance area if $S_e < S$

S_d is the damaged area where $S_d = S - S_e$

The mechanical measurement of local damage with n is characterized by:

$$D = \frac{S_d}{S}$$

If $D = 0$: the material is not damaged.

If $D = 1$: the volume element is broken into two parts along the plane normal to n.

If $0 < D < 1$: D characterizes the state of damage level.

Simulation of damage of raffia/PLA, alfa/PLA, and sisal/PLA composites

For the modeling of the fiber/matrix interface in the case of composites based on vegetable fibers, many analytical and numerical methods describe the shear of the fiber-matrix interface ("Pull-out" test, the release of a micro-drop, Diabolo compression test, Micro-indentation, and push-out test, Cox model, Kelly model, Weibull statistical approach,...) [113]. But to our knowledge, few methods and models have studied fiber-matrix interface damage.

In this study, to model the damage of the three types of composites, a probabilistic approach has been used which based on the damage to the fiber and the matrix proposed by Weibull and the genetic operators (selection, crossing, and mutation) in our approach to calculating the optimum values of the damage of the interface are employed [61].

The selection operator is used to choose the most appropriate solutions to have an optimal and convergent result. This operator is the application of the adaptation principle of Darwin's theory [61].

The Crossing operator, or crossing-over, is the result obtained when two chromosomes share their peculiarities. This allows the genetic mixing of the population and the application of the principle of inheritance of Darwin's theory [61].

The Mutation consists of altering a gene in a chromosome according to a mutation factor. This factor is the probability of mutation being made in an individual. This operator applies the principle of variation of Darwin's theory and allows, at the same time, to avoid a premature convergence of the algorithm towards a local extremum [61].

The Weibull distribution theory

Although it was first recognized by Fréchet 1927 and first used by Rosin and Rammler 1933 to describe the size distribution of particles, the continuous probability distribution known as the Weibull distribution is named after Waloddi Weibull, who described it in detail in 195. Since then, reliability engineering and other fields have frequently highlighted the Weibull distribution as one of the lifespan distributions. It accurately captures observed failure rates for a wide range of phenomena and component types [83].

This theory is without a doubt the most widely used statistical model today. It is of utmost interest to practitioners because of its ability to fit data from a variety of fields, including life data, weather data, observations made in economics and business administration, hydrology, biology, or the engineering sciences, as well as theory-oriented statisticians because of its numerous special features [129].

Weibull probabilistic formalism

Our results were obtained by a genetic simulation based on the Weibull equations. Damage to the matrix is given in equation (3.1) [104, 146]:

$$D_m = 1 - \exp\left\{-\frac{V_{eff}}{V_0}\left(\frac{\sigma_f}{\sigma_0}\right)^m\right\} \tag{3.1}$$

In which σ_f is mechanical stress, V_{eff} the matrix volume, (m and σ_0) the parameters of Weibull, V_0 the initial volume of the matrix. The second equation presents the law of the fracture of a fiber [82, 90].

$$D_f = 1 - \exp\left\{-A_f L_{equi}\left(\frac{\sigma_{max}^f}{\sigma_{0f}}\right)^{m_f}\right\} \tag{3.2}$$

Where $A_f = \pi a^2$, and σ_{max}^f is the maximum mechanical stress applied, σ_{0f} the initial stress, m_f is the parameter of Weibull, L_{equi} the fiber length at equilibrium.

Interface shear analytical model of Cox

Many analytical models have been proposed. One of the first solutions, that of Cox [37], gives the shape of the shear stress along the length of the fiber in the form of (3.3): The interface shear stress (IFSS - InterFacial Shear Strength) can be expressed as demonstrated by equation (3.3) [37]:

$$\tau = \frac{E_f * a * \varepsilon}{2}\left(\frac{2G_m}{E_f * r_f^2 * \ln\left(\frac{R}{r_f}\right)}\right)\left(\tanh\left(\left(\frac{2G_m}{E_f * r_f^2 * \ln\left(\frac{R}{r_f}\right)}\right) * \frac{\iota}{2}\right)\right) \tag{3.3}$$

with:

- (G_m) : shear modulus of the matrix;

- (E_f) : Young's modulus of the fiber;

- (ε) : deformation ;

- (a) : radius of the fiber;

- (R) : distance between fibers;

- (τ) : shear stress of the interface;

- (r_f) : the distance between the fiber and the matrix.

Genetic operators

Table.3.2, represents the mechanical properties of the three types of fiber according to the experimental studies, which will be used for the genetic algorithm method, In Table. 3.3, the main functions relating to the genetic operators (selection, crossing, and mutation) have been presented to determine the damage at the fiber-matrix interface of each material studied.

Table 3.2: Mechanical propertie of the three type of fibers used in Genetic operation.

Fibers	**Young's modulus (GPa)**	**Deformation at break (%)**	**Stress at break (MPa)**
Raffia palm [30, 47, 77]	32	2	404
Alfa [39, 77]	15.5	2	177
Sisal [18, 141]	15.5	5	460

Table 3.3: Mechanical propertie of the three type of fibers used in Genetic operation.

selection

```
function x = Gen_Pop_Init(Npop_init,To)
x(:,1) = rand(Npop_init,1);
x(:,2) =rand(Npop_init,1)*10e-5;
```

crossing

```
function[enf1, enf2]=cross(p1,p2)
nbVar = length(p1);
enf1=[]; enf2=[];
a = rand+0.2;
enf1 = p1*a + p2*(1-a);
enf2 = p2*a+p1*(1-a);
return
```

mutation

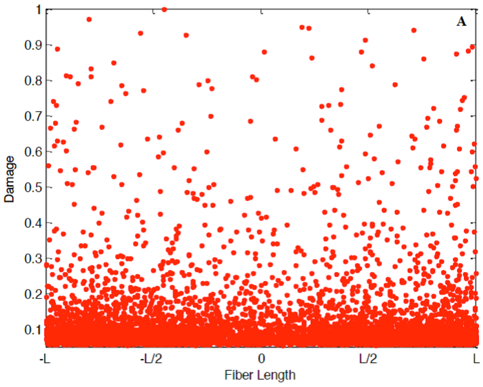
```
function [enf1] = mutate(enf1,probMut,To,Te)
bound2=[To Te];
bound1=[0 1000];
[nbenf  nbvr]=size(enf1);
df1 = bound1(2) - bound1(1); df2 = bound2(2) - bound2(1)
for k=1:nbenf
a = rand;
if a < probMut
mPoint = round(rand * nbenf);
if mPoint ≅ 0
    L=round(2*rand);
    if L ≅ 0
      if L ≅ 2
        enf1(k,L) = bound1(1)+rand*df1;
      else
        enf1(k,L) = bound2(1)+ rand*df2;
        end
     end
  end
  end
  end
```

3.3 Results and Discussion

The effect of raffia and alfa plant fibers on fiber-matrix interface damage of biocomposite materials (raffia / PLA, alfa / PLA, and sisal / PLA) has been investigated. The genetic simulation is based on the Weibull model (equations (3.1) and (3.2)), the operator crossing is used to calculate the interface damage of our materials. An initial population of 1000 individuals [8] was generated, then improved with a set of genetic operators (selection, crossing,and mutation), and in each case, the young modulus of each fiber is used Tables 3.2 and 3.3. The population consists of chromosomal genes representing the following variables: the shear stress (450, 550, and 650 N/m^2), Young's modulus, the modulus of the matrix shear, the fiber diameter, and the half distance R. The roulette selection and an equal mutation value of 0.25 have been chosen for optimizing our results (interface damage values).

3.3.1 Sisal/PLA Biocomposite Material

In figure.3.8, the damage (D) at the interface begins for a value of D = 0.18 for mechanical shear stress equal to 450 N/m^2 (Figure.3.8.A). It increases linearly when the stress increases and reaches a maximum value of 0.33 for a shear stress of 650 N/m^2 (Figure. 3.8.C). Symmetry of damage was observed in the middle of the interface. The red cloud explains that the damage is concentrated at the ends compared to the middle of the fiber, as has already been demonstrated by the micromechanical model of Cox [37].



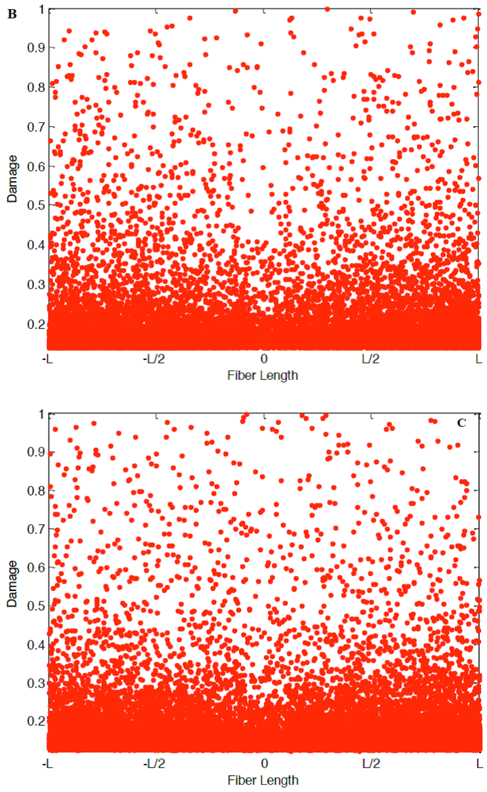


Figure 3.8: Interface fiber-matrix damage level of sisal/PLA for (A) σ = 450 N/m^2, (B) σ = 550 N/m^2, (C) σ = 650 N/m^2.

3.3.2 Alfa/PLA Biocomposite Material

Figure.3.9) show that the level of sheer damage begins for a damage value D = 0.21 Figure. 3.9.A) when σ = 450 N/m^2, and it reaches a maximum value D= 0.26 (3.9.C) for a maximum stress σ = 650 N/m^2. Symmetry of damage was observed in the middle of the interface. The red cloud explains that the damage is concentrated at the ends compared to the middle of the fiber, as has already been demonstrated by the micromechanical model of Cox.

The genetic results faithfully showed the real behavior of the two materials according to their mechanical properties, in particular, the values of the three Young's modulus. We have concluded that the fiber with the largest Young's modulus, its fiber-matrix interface, is the most resistant to mechanical stresses.

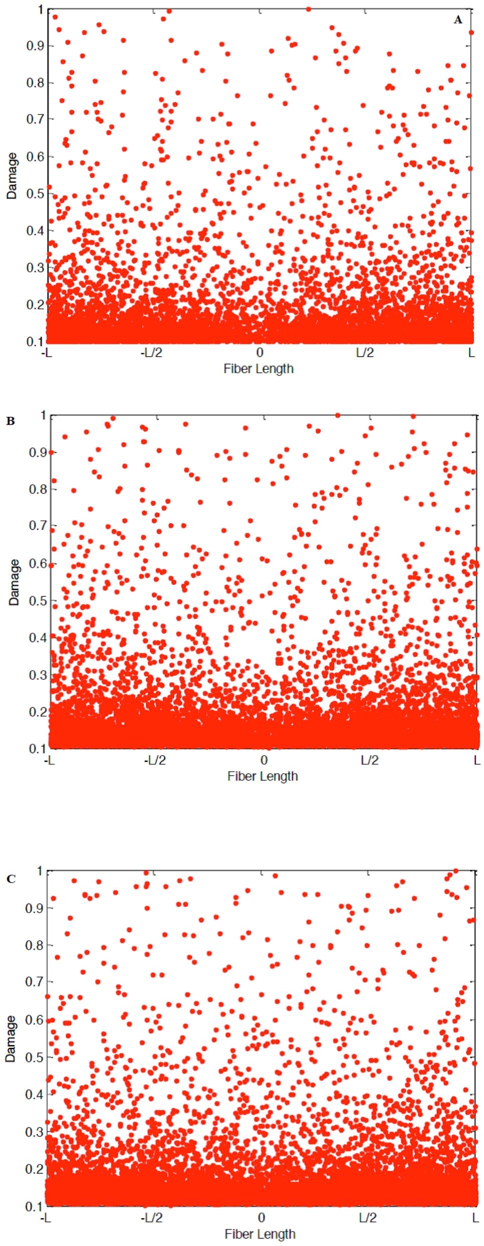


Figure 3.9: Interface fiber-matrix damage level of alfa/PLA for (A) σ = 450 N/m^2, (B) σ = 550 N/m^2, (C) σ = 650 N/m^2.

3.3.3 Raffia/PLA Biocomposite Material

Figure.3.10 show that the level of sheer damage begins for a damage value D = 0.12 (Figure.3.10.A) when σ = 450 N/m^2, and it reaches a maximum value D = 0.22 (Figure.3.10.C) for maximum stress of σ = 650 N/m^2. Symmetry of damage was observed in the middle of the interface. The red cloud explains that the damage is concentrated at the ends compared to the middle of the fiber, as has already been demonstrated by the micromechanical model of Cox.

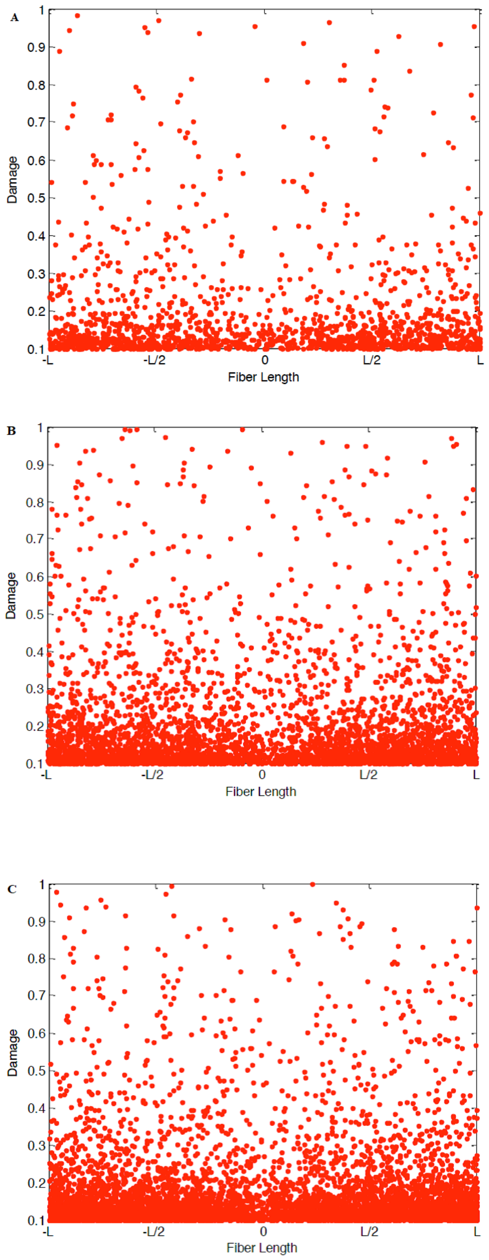


Figure 3.10: Interface fiber-matrix damage level of raffia/PLA for (A) $\sigma = 450\ N/m^2$, (B) $\sigma = 550\ N/m^2$, (C) $\sigma = 650\ N/m^2$.

3.4 Conclusion

This work focused on the study of the effect of raffia, alfa, and sisal fibers on fiber-matrix interface shear damage of the three biocomposite materials studied, which consisted of the same matrix of PLA. To calculate interface damage, the genetic operator crossing has been employed based on the Weibull approach. The results obtained by genetic modeling have shown that the raffia / PLA and alfa/PLA biocomposite materials have better mechanical properties compared to sisal/ PLA. This observation has been confirmed by the different values of interface damage of the biocomposites studied; the numerical results are similar and coincide perfectly with the analytical results of Cox [37], where he proved that Young'smodulus of fibers improves the resistance of the interface. These results are in good agreement with our results found by the genetic approach [15]. The experimental work by Antoine Le Duigou et al [85] and Bodros et al [24] showed that the use of natural fibers greatly improves the mechanical properties of biocomposite materials.

Conclusion and challenges

In conclusion, this thesis has explored the fascinating world of biocomposites, specifically focusing on those comprising a polymer matrix and natural fibers. Throughout this research journey, we have delved into various aspects of these innovative materials, from their composition and manufacturing processes to their mechanical and environmental performance.

The first part of this thesis is an experimental study with the main purpose of preparing biocomposite materials composed of high-density polyethylene (HD-PE) reinforced by Alfa short fibers and natural Pozzolan particles, to promote the recycling of plastic waste while taking advantage of the characteristics and properties that give these natural reinforcements. One of the most important points to emphasize and which prompted the development of this research is the fact of using simple and low-cost techniques compared to others.

In this work, we recycled waste plastic bottle caps made of high-density polyethylene (HD-PE) which was reincorporated as a matrix and reinforced by alfa short fibers and natural pozzolan particles. Using different weight percentages of both fillers of 5wt%up to 30wt%, three types of biocomposite materials have been produced; alfa short fibers/HDPE, pozzolan particles/HDPE, and alfa fibers pozzolan/HDPE. Specimens for each type of biocomposite were prepared through the compression molding method. During this experiment, the natural reinforcement did not go through any chemical treatment only ann mechanical extraction. Furthermore, we characterized samples of the produced biocomposites, by initially using the Fourier transform infrared red (FTIR) spectroscopy to evaluate the structural properties of the biocomposites components. Then, we obtained the mechanical properties of the samples by a tensile test. Finally, scanning electron microscopy (SEM) was applied to acquire some information on the morphology of the composite.

Tensile test results revealed an enhancement in the mechanical properties for the three types of biocomposites, an increase in tensile strength reached the maximum of 3573 MPa plus an interesting improvement in Young's modulus with a maximum value of 3696 MPa. The toughness of the neat recycled HD-PE decreased by 212% by adding the natural filler whereas the modulus of resilience. The obtained results indicate that the recycling of the HPDE reinforced with alfa fibers and pozzolan powder can represent good potential in improving interfacial properties. The study shows an alternative for producing friendly biodegradable composites that can be a solution for plastic waste reduction and a substitute for petrochemical-derived products.

The last part of this thesis is an investigation of the effect of raffia fiber on fiber matrix interface damage of biocomposite materials was pursued; raffia/PLA (polylactic acid), alfa/PLA, and sisal/PLA subjected to the same mechanical shear stress, using a genetic algorithm, To calculate interface damage using fiber and matrix damage, the genetic operator crossing has been employed based on the Weibull approach.

The results obtained by genetic modeling have shown that the raffia / PLA and alfa/PLA biocomposite materials have better mechanical properties compared to sisal/ PLA. This observation has been confirmed by the different values of interface damage of the biocomposites studied; the numerical results are similar and coincide perfectly with the analytical results of Cox [37], where he proved that Young'smodulus of fibers improves the resistance of the interface. These results are in good agreement with our results found by the genetic approach [15]. The experimental work by Antoine Le Duigou et al [85] and Bodros et al [24] showed that the use of natural fibers greatly improves the mechanical properties of biocomposite materials.

This present study can serve as a springboard for future research works. Indeed, it is possible to do more mechanical, thermal, and morphological analysis to extract more data about these biocomposites. Furthermore, investigate the treatment of the Alfa fiber and pozzolan powder or the addition of a coupling agent to improve the adhesion and the quality of the dispersion of the natural reinforcements within the matrix. It is also appropriate to exploit the durability of these composites and the effect of Alfa fibers, Raffia, and pozzolan powders treatment on the latter, as well as on the mechanical properties and water adsorption.

Bibliography

[1] Abdelmouleh, M., Boufi, S., Belgacem, M. N., and Dufresne, A. Short natural-fibre reinforced polyethylene and natural rubber composites: Effect of silane coupling agents and fibres loading. *Composites science and technology 67*, 7-8 (2007), 1627–1639. 17

[2] Ahmed, S., Ikram, S., Kanchi, S., and Bisetty, K. *Biocomposites: biomedical and environmental applications*. CRC Press, 2018. 21

[3] Alvarez, V. A., and Vázquez, A. Influence of fiber chemical modification procedure on the mechanical properties and water absorption of materbi-y/sisal fiber composites. *Composites Part A: Applied Science and Manufacturing 37*, 10 (2006), 1672–1680. 46

[4] Arabeche, K., Abdelmalek, F., Delbreilh, L., Zair, L., and Berrayah, A. Physical and rheological properties of biodegradable poly (butylene succinate)/alfa fiber composites. *Journal of Thermoplastic Composite Materials 35*, 10 (2022), 1709–1727. 46

[5] Arrabiyeh, P. A., May, D., Eckrich, M., and Dlugaj, A. M. An overview on current manufacturing technologies: Processing continuous rovings impregnated with thermoset resin. *Polymer Composites 42*, 11 (2021), 5630–5655. 30

[6] Arrakhiz, F., Benmoussa, K., Bouhfid, R., and Qaiss, A. Pine cone fiber/clay hybrid composite: Mechanical and thermal properties. *Materials & Design 50* (2013), 376–381. 52

[7] Arrakhiz, F., Elachaby, M., Bouhfid, R., Vaudreuil, S., Essassi, M., and Qaiss, A. Mechanical and thermal properties of polypropylene reinforced with alfa fiber under different chemical treatment. *Materials & Design 35* (2012), 318–322. 37

[8] Atig, K., Mokaddem, A., Meskine, M., Doumi, B., Belkheir, M., and El Keurti, M. Using genetic algorithms to study the effect of cellulose fibers ratio on the fiber-matrix interface damage of biocomposite materials. *Current Materials Science: Formerly: Recent Patents on Materials Science 12*, 1 (2019), 83–90. 67

[9] Avenet, J., Cender, T. A., Le Corre, S., Bailleul, J.-L., and Levy, A. Adhesion of high temperature thermoplastic composites. *Procedia Manufacturing 47* (2020), 925–932. 17

[10] Ayari, H. *Modélisation micromécanique de l'effet des chargements cycliques et de la vitesse de déformation sur l'endommagement de l'interface fibre-matrice dans les composites SMC*. PhD thesis, HESAM Université; Université de Sousse (Tunisie), 2020. 28

[11] Backelandt, A. *Etude des mécanismes d'adhésion à l'interface résine/ciment en vue de la réparation des ouvrages de génie civil*. PhD thesis, Lyon, INSA, 2005. 17

[12] Barbero, E. J. *Introduction to composite materials design*. CRC press, 2010. 1, 2, 5, 13, 23

[13] Barnes, P., and Bensted, J. *Structure and performance of cements*. CRC Press, 2002. 40

[14] BEIDA, M. D. *Application de l'algorithme génétique et les éléments finis à l'étude du comportement à l'endommagement des composites soumis à l'impact*. PhD thesis, Université de Mostaganem-Abdelhamid Ibn Badis. 2, 3

[15] Belkheir, M., Doumi, B., Mokaddem, A., and Boutaous, A. Using genetic algorithm for investigating the performance of carbonbasalt/polyester hybrids composite materials. *Current Materials Science: Formerly: Recent Patents on Materials Science 13*, 2 (2020), 120–128. 70, 73

[16] Bernatas, R., Dagréou, S., Despax-Ferreres, A., and Barasinski, A. Recycling of fiber reinforced composites with a focus on thermoplastic composites. *Cleaner Engineering and Technology 5* (2021), 100272. 33

[17] Berthelot, J.-M. Mécanique des matériaux et structures composites. *Institut Supérieur des Matériaux et Mécaniques Avancés* (2010), 176. ii, 5

[18] Bezazi, A., Belaadi, A., Bourchak, M., Scarpa, F., and Boba, K. Novel extraction techniques, chemical and mechanical characterisation of agave americana l. natural fibres. *Composites Part B: Engineering 66* (2014), 194–203. 61, 65

[19] Bharath, K. N., and Basavarajappa, S. Applications of biocomposite materials based on natural fibers from renewable resources: a review. *Science and Engineering of Composite Materials 23*, 2 (2016), 123–133. 1, 22

[20] Bhattacharjee, S., and Bajwa, D. S. Degradation in the mechanical and thermo-mechanical properties of natural fiber filled polymer composites due to recycling. *Construction and building materials 172* (2018), 1–9. 33

[21] BINICI, H., AKSOGAN, O., AND SHAH, T. Investigation of fibre reinforced mud brick as a building material. *Construction and building materials 19*, 4 (2005), 313–318. 57

[22] BIRON, M. *Thermoplastics and thermoplastic composites*. William Andrew, 2012. ii, 7, 10, 11

[23] BOCK, J. H., AND NORRIS, D. *Forensic plant science*. Academic Press, 2015. 21

[24] BODROS, E., PILLIN, I., MONTRELAY, N., AND BALEY, C. Could biopolymers reinforced by randomly scattered flax fibre be used in structural applications? *Composites Science and Technology 67*, 3-4 (2007), 462–470. 71, 73

[25] BORCHANI, K. E., CARROT, C., AND JAZIRI, M. Untreated and alkali treated fibers from alfa stem: effect of alkali treatment on structural, morphological and thermal features. *Cellulose 22* (2015), 1577–1589. 38, 46

[26] BORCHANI, K. E., CARROT, C., AND JAZIRI, M. Rheological behavior of short alfa fibers reinforced mater-bi® biocomposites. *Polymer Testing 77* (2019), 105895. 36

[27] BORGNA, T. *Études des propriétés de composite à matrice thermoplastique thermostable au-delà de leur température de transition vitreuse*. PhD thesis, Pau, 2017. ii, 9

[28] BOUJMAL, R., KAKOU, C. A., NEKHLAOUI, S., ESSABIR, H., BENSALAH, M.-O., RODRIGUE, D., BOUHFID, R., AND QAISS, A. E. K. Alfa fibers/clay hybrid composites based on polypropylene: mechanical, thermal, and structural properties. *Journal of Thermoplastic Composite Materials 31*, 7 (2018), 974–991. 52

[29] BOUKHOULDA, A., BOUKHOULDA, F., MAKICH, H., NOUARI, M., AND HADDAG, B. Elaboration and mechanical properties analysis of a composite based on polyester resin reinforced with natural alfa fibres. *Journal of Composite Materials 53*, 28-30 (2019), 3993–4001. ii, 38, 39

[30] BRAHIM, S. B., AND CHEIKH, R. B. Influence of fibre orientation and volume fraction on the tensile properties of unidirectional alfa-polyester composite. *Composites Science and Technology 67*, 1 (2007), 140–147. 61, 65

[31] CHAITANYA, S., SINGH, I., AND SONG, J. I. Recyclability analysis of pla/sisal fiber biocomposites. *Composites Part B: Engineering 173* (2019), 106895. 33

[32] CHAND, N., AND FAHIM, M. *Tribology of natural fiber polymer composites*. Woodhead publishing, 2020. ii, iii, 24, 25, 59

[33] CHAUHAN, V., KÄRKI, T., AND VARIS, J. Review of natural fiber-reinforced engineering plastic composites, their applications in the transportation sector and processing techniques. *Journal of Thermoplastic Composite Materials 35*, 8 (2022), 1169–1209. 36

[34] CHEREMISINOFF, N. P. *Condensed encyclopedia of polymer engineering terms*. Butterworth-Heinemann, 2001. 10

[35] CHUNG, D. D. *Composite materials: science and applications*. Springer Science & Business Media, 2010. 2

[36] CIPRIANO, T. F., SILVA, A. L. N. D., SILVA, A. H. M. D. F. T. D., SOUSA, A. M. F. D., SILVA, G. M. D., AND ROCHA, M. G. Thermal, rheological and morphological properties of poly (lactic acid)(pla) and talc composites. *Polímeros 24* (2014), 276–282. iii, 62

[37] COX, H. The elasticity and strength of paper and other fibrous materials. *British journal of applied physics 3*, 3 (1952), 72. 64, 67, 70, 73

[38] DALLEL, M. *Evaluation du potentiel textile des fibres d'Alfa (Stipa Tenacissima L.): Caractérisation physico-chimique de la fibre au fil*. PhD thesis, Université de Haute Alsace-Mulhouse, 2012. 1, 14, 15, 39, 40

[39] DATTA, R., TSAI, S.-P., BONSIGNORE, P., MOON, S.-H., AND FRANK, J. R. Technological and economic potential of poly (lactic acid) and lactic acid derivatives. *FEMS microbiology reviews 16*, 2-3 (1995), 221–231. 60, 61, 65

[40] DING, W., ET AL. *Delamination analysis of composite laminates*. University of Toronto, 2000. 29

[41] DO THI, V. V. D. T. *Matériaux composites à fibres naturelles/polymère biodégradables ou non*. PhD thesis, Université de Grenoble; Université des Sciences Naturelles d'Ho Chi Minh Ville, 2011. ii, 11, 14, 22

[42] DOS SANTOS ROSA, D., AND LENZ, D. M. Biocomposites: Influence of matrix nature and additives on the properties and biodegradation behaviour. *Biodegrad. Eng. Technol. Intech Rij. Croat* (2013), 433–475. 21

[43] EBNESAJJAD, S., AND LANDROCK, A. H. *Adhesives technology handbook*. William Andrew, 2014. 20

[44] EL-ABBASSI, F. E., ASSARAR, M., AYAD, R., BOURMAUD, A., AND BALEY, C. A review on alfa fibre (stipa tenacissima l.): From the plant architecture to the reinforcement of polymer composites. *Composites Part A: Applied Science and Manufacturing 128* (2020), 105677. 37

[45] EL ABBASSI, F. E., ASSARAR, M., AYAD, R., SABHI, H., BUET, S., AND LAMDOUAR, N. Effect of recycling cycles on the mechanical and damping properties of short alfa fibre reinforced polypropylene composite. *Journal of Renewable Materials 7*, 3 (2019), 253. 40

[46] ELENGA, R., DIRRAS, G., MANIONGUI, J. G., DJEMIA, P., AND BIGET, M. On the microstructure and physical properties of untreated raffia textilis fiber. *Composites Part A: Applied Science and Manufacturing 40*, 4 (2009), 418–422. 57

[47] Elenga, R., Djemia, P., Biget, M., Maniongui, J. G., and Dirras, G. Microstructure et propriétés physico-mécaniques de la fibre brute de raphia. *MATERIAUX–Dijon, France* (2006). 57, 60, 61, 65

[48] Elouaer, A. *Contribution à la compréhension et à la modélisation du comportement mécanique de matériaux composites à renfort en fibres végétales*. PhD thesis, Reims, 2011. 3, 12

[49] Essabir, H., Bouhfid, R., and Qaiss, A. Alfa and doum fiber-based composite materials for different applications. In *Lignocellulosic fibre and biomass-based composite materials*. Elsevier, 2017, pp. 147–164. 37

[50] Fadele, O., Oguocha, I. N., Odeshi, A., Soleimani, M., and Karunakaran, C. Characterization of raffia palm fiber for use in polymer composites. *Journal of Wood Science 64* (2018), 650–663. 57

[51] Fambri, L., and Migliaresi, C. Crystallization and thermal properties. *Poly (Lactic Acid) Synthesis, Structures, Properties, Processing, Applications, and End of Life* (2022), 135–151. 61

[52] Faruk, O., and Sain, M. *Biofiber reinforcements in composite materials*. No. 51. Elsevier, 2014. 31, 32

[53] Fiore, V., Scalici, T., Nicoletti, F., Vitale, G., Prestipino, M., and Valenza, A. A new eco-friendly chemical treatment of natural fibres: Effect of sodium bicarbonate on properties of sisal fibre and its epoxy composites. *Composites Part B: Engineering 85* (2016), 150–160. 58

[54] Firdous, R., Stephan, D., and Djobo, J. N. Y. Natural pozzolan based geopolymers: A review on mechanical, microstructural and durability characteristics. *Construction and Building Materials 190* (2018), 1251–1263. 46

[55] Fonseca-Valero, C., Ochoa-Mendoza, A., Arranz-Andrés, J., and González-Sánchez, C. Mechanical recycling and composition effects on the properties and structure of hardwood cellulose-reinforced high density polyethylene eco-composites. *Composites Part A: Applied Science and Manufacturing 69* (2015), 94–104. 46, 52

[56] Fradet, G. *Physico-chimie de l'interface fibres/matrice: applications aux composites Carbone/Carbone*. PhD thesis, Bordeaux 1, 2013. 9, 15

[57] Gao, X., Umair, M., Nawab, Y., Latif, Z., Ahmad, S., Siddique, A., and Yang, H. Mode i fatigue of fibre reinforced polymeric composites: a review. *Polymers 14*, 21 (2022), 4558. ii, 28

[58] Garlotta, D. A literature review of poly (lactic acid). *Journal of Polymers and the Environment 9* (2001), 63–84. 62

[59] GARNIER, C. *Etude du comportement dynamique des structures composites réalisées par LRI: application à l'impact et à la fatigue*. PhD thesis, 2011. 27

[60] GAY, D. *Composite materials: design and applications*. CRC press, 2022. ii, 32

[61] GAZUT, S. Les algorithmes génétiques peuvent-ils aider à identifier des biomarqueurs génétiques? 63

[62] GEHRING, F. Etude du comportement mécanique et de l'endommagement de composites thermoplastiques renforcés de fibres courtes de chanvre: Approche expérimentale et modélisation. *Université de Lorraine* (2013). 2

[63] GHARDE, S., AND KANDASUBRAMANIAN, B. Mechanothermal and chemical recycling methodologies for the fibre reinforced plastic (frp). *Environmental Technology & Innovation 14* (2019), 100311. 34

[64] GOPANNA, A., RAJAN, K. P., THOMAS, S. P., AND CHAVALI, M. Polyethylene and polypropylene matrix composites for biomedical applications. In *Materials for biomedical engineering*. Elsevier, 2019, pp. 175–216. 30, 31

[65] GOURIER, C. *Contribution à l'étude de matériaux biocomposites à matrice thermoplastique polyamide-11 et renforcés par des fibres de lin*. PhD thesis, Université de Bretagne Sud, 2016. ii, 31, 32

[66] HASHMI, M. S. J. *Comprehensive materials processing*. Newnes, 2014. 12, 13

[67] HORE, M. J., KORLEY, L. T., AND KUMAR, S. K. Polymer-grafted nanoparticles. *Journal of Applied Physics 128*, 3 (2020). 15

[68] HUANG, S., FU, Q., YAN, L., AND KASAL, B. Characterization of interfacial properties between fibre and polymer matrix in composite materials–a critical review. *Journal of Materials Research and Technology 13* (2021), 1441–1484. 15, 16

[69] IVANOV, Y., CHESHKOV, V., AND NATOVA, M. *Polymer Composite Materials—Interface Phenomena & Processes*, vol. 90. Springer Science & Business Media, 2001. 15

[70] JAWAID, M., THARIQ, M., AND SABA, N. *Mechanical and physical testing of biocomposites, fibre-reinforced composites and hybrid composites*. Woodhead Publishing, 2019. iii, 10, 58, 59

[71] JI, L., AND SHI, B. A novel method for determining surface free energy of powders using washburn's equation without calculating capillary factor and contact angle. *Powder Technology 271* (2015), 88–92. 19

[72] JOHN, M. J., AND THOMAS, S. Biofibres and biocomposites. *Carbohydrate polymers 71*, 3 (2008), 343–364. 11

[73] KALIA, S., AND AVÉROUS, L. *Biopolymers: biomedical and environmental applications*. John Wiley & Sons, 2011. 11, 14

[74] KANDOLA, B. K., MISTIK, S., PORNWANNACHAI, W., AND ANAND, S. Natural fibre-reinforced thermoplastic composites from woven-nonwoven textile preforms: Mechanical and fire performance study. *Composites Part B: Engineering 153* (2018), 456–464. 36

[75] KANI, E. N., AND ALLAHVERDI, A. Effect of chemical composition on basic engineering properties of inorganic polymeric binder based on natural pozzolan. *Ceramics-Silikaty 53*, 3 (2009), 195–204. 46

[76] KANKAM, C. Raffia palm-reinforced concrete beams. *Materials and Structures 30* (1997), 313–316. 57

[77] KELLER, A., LEUPIN, M., MEDIAVILLA, V., AND WINTERMANTEL, E. Influence of the growth stage of industrial hemp on chemical and physical properties of the fibres. *Industrial crops and products 13*, 1 (2001), 35–48. 60, 61, 65

[78] KHIAT, S. M. A. *Etude de la probabilité dèndommagement des composites unidirectionnels aux interface fibre/rèsine*. PhD thesis, USTOMB, 2012. 27, 28

[79] KORONIS, G., AND SILVA, A. *Green composites for automotive applications*. Woodhead Publishing, 2018. 21, 23, 25, 26

[80] KUBIAK, J. M., AND MACFARLANE, R. J. Polymer-grafted nanoparticles as single-component, high filler content composites via simple transformative aging. *Advanced Functional Materials 32*, 6 (2022), 2107139. 16

[81] LABIDI, K., KORHONEN, O., ZRIDA, M., HAMZAOUI, A. H., AND BUDTOVA, T. All-cellulose composites from alfa and wood fibers. *Industrial crops and products 127* (2019), 135–141. 46

[82] LADEVÈZE, P., AND LUBINEAU, G. Pont entre les «micro» et «méso» mécaniques des composites stratifiés. *Comptes rendus mécanique 331*, 8 (2003), 537–544. 64

[83] LAI, C.-D., AND LAI, C.-D. *Generalized weibull distributions*. Springer, 2014. 63

[84] LARIBI, M. A. *Caractérisation et Modélisation du comportement micromécanique des matériaux composites SMC sous chargement thermomécanique de type quasi-statique et fatigue.* PhD thesis, Paris, ENSAM, 2018. 5

[85] LE DUIGOU, A., KERVOELEN, A., LE GRAND, A., NARDIN, M., AND BALEY, C. Interfacial properties of flax fibre–epoxy resin systems: existence of a complex interphase. *Composites Science and Technology 100* (2014), 152–157. 71, 73

[86] LE MOIGNE, N., OTAZAGHINE, B., CORN, S., ANGELLIER-COUSSY, H., AND BERGERET, A. *Surfaces and interfaces in natural fibre reinforced composites: Fundamentals, modifications and characterization*. Springer, 2018. ii, 22, 24

[87] LEMAITRE, J., AND CHABOCHE, J.-L. *Mechanics of solid materials*. Cambridge university press, 1994. 61, 62

[88] LI, X., TABIL, L. G., AND PANIGRAHI, S. Chemical treatments of natural fiber for use in natural fiber-reinforced composites: a review. *Journal of Polymers and the Environment 15* (2007), 25–33. ii, 25, 26

[89] LI, Y., MAI, Y.-W., AND YE, L. Sisal fibre and its composites: a review of recent developments. *Composites science and technology 60*, 11 (2000), 2037–2055. 59

[90] LISSART, N., AND LAMON, J. Damage and failure in ceramic matrix minicomposites: experimental study and model. *Acta Materialia 45*, 3 (1997), 1025–1044. 64

[91] LIU, Y., LV, X., BAO, J., XIE, J., TANG, X., CHE, J., MA, Y., AND TONG, J. Characterization of silane treated and untreated natural cellulosic fibre from corn stalk waste as potential reinforcement in polymer composites. *Carbohydrate polymers 218* (2019), 179–187. 24

[92] MAHFOUDH, A. *Étude de la production et de la caractérisation de composites bois-plastiques*. PhD thesis, Université Laval, 2013. 6, 12, 14

[93] MAIA, B. S. *Study on the effect of surface energy of polypropylene/Polyamide12 polymer hybrid matrix reinforced with virgin and recycled carbon fiber*. University of Toronto (Canada), 2017. ii, 16, 17

[94] MANCINOA, A., MARANNANO, G., AND ZUCCARELLO, B. Implementation of eco-sustainable biocomposite materials reinforced by optimized agave fibers. *Procedia Structural Integrity 8* (2018), 526–538. 62

[95] MERCIER, J. P., ZAMBELLI, G., AND KURZ, W. *Introduction to materials science*. Elsevier, 2012. 2

[96] MESSAOUI, S., BORCHANI, K. E., GHALI, L., GUERMAZI, N., HADDAR, N., AND MSAHLI, S. Effect of a new composition ratio and of a new chemical treatment on natural alfa fiber/polypropylene composites manufacturing and their mechanical properties. *Journal of Natural Fibers 19*, 15 (2022), 10126–10141. 39

[97] MIHALUTA, M. *Etude de la mise en oeuvre industrielle d'un nouveau procédé de mise en forme de produits composites*. PhD thesis, Arts et Métiers ParisTech, 2011. 29

[98] MILITELLO, C., BONGIORNO, F., EPASTO, G., AND ZUCCARELLO, B. Low-velocity impact behaviour of green epoxy biocomposite laminates reinforced by sisal fibers. *Composite Structures 253* (2020), 112744. 60, 61

[99] MISRA, M., PANDEY, J. K., AND MOHANTY, A. *Biocomposites: design and mechanical performance*. Woodhead Publishing, 2015. 21

[100] Mitschang, P., and Hildebrandt, K. Polymer and composite moulding technologies for automotive applications. In *Advanced materials in automotive engineering*. Elsevier, 2012, pp. 210–229. 31

[101] Mittal, K. L. *Progress in adhesion and adhesives*. John Wiley & Sons, 2015. ii, 15, 18, 19

[102] Mogue Kamga, S., Brokamp, G., Cosiaux, A., Awono, A., Fürniss, S., Barfod, A. S., Muafor, F. J., Le Gall, P., Sonké, B., and Couvreur, T. L. Use and cultural significance of raphia palms. *Economic Botany 74* (2020), 207–225. iii, 56, 57

[103] Mohd Izwan, S., Sapuan, S., Zuhri, M., and Mohamed, A. Thermal stability and dynamic mechanical analysis of benzoylation treated sugar palm/kenaf fiber reinforced polypropylene hybrid composites. *Polymers 13*, 17 (2021), 2961. 25

[104] Mokaddem, A., Alami, M., Doumi, B., and Boutaous, A. Prediction by a genetic algorithm of the fiber–matrix interface damage for composite material. part 1. study of shear damage in two composites t300/914 and peek/apc2. *Strength of materials 46* (2014), 543–547. 64

[105] Mostafa, N., Farag, A. A., Abo-dief, H. M., and Tayeb, A. M. Production of biodegradable plastic from agricultural wastes. *Arabian journal of chemistry 11*, 4 (2018), 546–553. 11

[106] Munjal, S., and Singh, A. The arrhenius acid and base theory. In *Corrosion*. IntechOpen, 2020. 20

[107] Munoz Cuartas, V. *Identification de mécanismes d'endommagement de stratifiés carbone-époxyde par couplage de l'émission acoustique et de la thermographie infrarouge*. PhD thesis, 2015. 27, 28

[108] Mutjé, P., Vallejos, M., Girones, J., Vilaseca, F., López, A., López, J., and Méndez, J. Effect of maleated polypropylene as coupling agent for polypropylene composites reinforced with hemp strands. *Journal of Applied Polymer Science 102*, 1 (2006), 833–840. 17

[109] Nagavally, R. R. Composite materials-history, types, fabrication techniques, advantages, and applications. *Int. J. Mech. Prod. Eng 5*, 9 (2017), 82–87. 29

[110] Nampoothiri, K. M., Nair, N. R., and John, R. P. An overview of the recent developments in polylactide (pla) research. *Bioresource technology 101*, 22 (2010), 8493–8501. 61

[111] Ndenecho, R. Biogeographical and ethnobotanical analysis of the raphia palm in the west cameroon highlands. *Journal of the Cameroon Academy of Sciences 7*, 1 (2007), 21–34. 57

[112] Nechad, H. Evaluation de l'endommagement et de la rupture de matériaux hétérogènes par ultrasons et émission acoustique: Estimation de la durée de vie restante. *Thesea l'Institut National des Sciences Appliquées de Lyon* (2004). 28

[113] Nguyen, D. C. *Caractérisation de l'interface fibre/matrice: application aux composites polypropylène/chanvre*. PhD thesis, Troyes, 2016. 16, 63

[114] Odera, R., Onukwuli, O., Atuanya, C., et al. Characterization of the thermo-microstructural analysis of raffia palm fibers proposed for roofing sheet production. *Journal of Minerals and Materials Characterization and Engineering 3*, 04 (2015), 335. 57

[115] Oliveira Filho, E. G. d., Luz, F. S. d., Fujiyama, R. T., Silva, A. C. R. d., Candido, V. S., and Monteiro, S. N. Effect of chemical treatment and length of raffia fiber (raphia vinifera) on mechanical stiffening of polyester composites. *Polymers 12*, 12 (2020), 2899. 56

[116] Pacheco-Torgal, F., Shasavandi, A., and Jalali, S. Eco-efficient concrete using industrial wastes: A review. In *Materials Science Forum* (2013), vol. 730, Trans Tech Publ, pp. 581–586. 1, 40, 41

[117] Paiva, M., Ammar, I., Campos, A., Cheikh, R. B., and Cunha, A. Alfa fibres: Mechanical, morphological and interfacial characterization. *Composites Science and Technology 67*, 6 (2007), 1132–1138. 46

[118] Paris, C. *Étude et modélisation de la polymérisation dynamique de composites à matrice thermodurcissable*. PhD thesis, 2011. ii, 1, 6, 12, 13, 23

[119] Pegoretti, A. Towards sustainable structural composites: A review on the recycling of continuous-fiber-reinforced thermoplastics. *Advanced Industrial and Engineering Polymer Research 4*, 2 (2021), 105–115. 33

[120] Pickering, K. L., Efendy, M. A., and Le, T. M. A review of recent developments in natural fibre composites and their mechanical performance. *Composites Part A: Applied Science and Manufacturing 83* (2016), 98–112. 25

[121] Pocius, A. V., and Dillard, D. A. *Adhesion science and engineering: surfaces, chemistry and applications*. Elsevier, 2002. 16

[122] Poletto, M. Maleated soybean oil as coupling agent in recycled polypropylene/wood flour composites: Mechanical, thermal, and morphological properties. *Journal of Thermoplastic Composite Materials 32*, 8 (2019), 1056–1067. 26

[123] Proy, J., Massa, F., Notta-Cuvier, D., Lauro, F., Tison, T., and Spingler, G. Integrating fibers and injection molding process variability in short-natural-fiber-reinforced thermoplastics behavior: A review. *Materials Today Communications 29* (2021), 102785. 32

[124] QIN, Q.-H., AND YE, J. *Toughening mechanisms in composite materials*. Elsevier, 2015. 12, 13

[125] RADZI, A., SAPUAN, S., JAWAID, M., AND MANSOR, M. Effect of alkaline treatment on mechanical, physical and thermal properties of roselle/sugar palm fiber reinforced thermoplastic polyurethane hybrid composites. *Fibers and Polymers 20* (2019), 847–855. 23

[126] RAGOUBI, M. *Contribution à l'amélioration de la compatiblilité interfaciale fibres naturelles/matrice thermoplastique via un traitement sous décharge couronne*. PhD thesis, Nancy 1, 2010. 1, 6, 7, 10

[127] RAHARJO, W. W., KUSHARJANTO, B., AND TRIYONO, T. Alkali treatment effect on the tensile and impact properties of recycled high-density polyethylene composites reinforced with short cantala fiber. *Journal of Southwest Jiaotong University 55*, 3 (2020). 16

[128] RAYDAN, N. D. V., LEROYER, L., CHARRIER, B., AND ROBLES, E. Recent advances on the development of protein-based adhesives for wood composite materials—a review. *Molecules 26*, 24 (2021), 7617. ii, 20

[129] RINNE, H. The weibull distribution: a handbook crc press. *Boca Raton* (2009). 64

[130] SADOUN, S. Contribution À l'étude expérimentale d'un polyéthylène À haute densité (pehd)- effet de la température et de la vitesse de déformation. Master's thesis, 2007. 9

[131] SALEM, S., OLIVER-ORTEGA, H., ESPINACH, F. X., HAMED, K. B., NASRI, N., ALCALÀ, M., AND MUTJÉ, P. Study on the tensile strength and micromechanical analysis of alfa fibers reinforced high density polyethylene composites. *Fibers and Polymers 20* (2019), 602–610. 36

[132] SANDBERG, M., YUKSEL, O., COMMINAL, R. B., SONNE, M. R., JABBARI, M., LARSEN, M., SALLING, F. B., BARAN, I., SPANGENBERG, J., AND HATTEL, J. H. Numerical modeling of the mechanics of pultrusion. In *Mechanics of materials in modern manufacturing methods and processing techniques*. Elsevier, 2020, pp. 173–195. ii, 30, 31

[133] SENHADJI, Y., ESCADEILLAS, G., KHELAFI, H., MOULI, M., AND BENOSMAN, A. S. Evaluation of natural pozzolan for use as supplementary cementitious material. *European Journal of Environmental and Civil Engineering 16*, 1 (2012), 77–96. 41

[134] SEYDIBEYOGLU, M. O., MOHANTY, A. K., AND MISRA, M. *Fiber technology for fiber-reinforced composites*. Woodhead Publishing, 2017. 14

[135] SEYNI, A. *Propriétés physico-chimiques et d'usage de matériaux composites à charge dégradable produits par co-broyage*. PhD thesis, 2008. ii, 7

[136] Shanmugam, V., Mensah, R. A., Försth, M., Sas, G., Restás, Á., Addy, C., Xu, Q., Jiang, L., Neisiany, R. E., Singha, S., et al. Circular economy in biocomposite development: State-of-the-art, challenges and emerging trends. *Composites Part C: Open Access 5* (2021), 100138. 33

[137] Stokke, D. D., Wu, Q., and Han, G. *Introduction to wood and natural fiber composites*. John Wiley & Sons, 2013. 18, 20

[138] Tesinova, P. *Advances in composite materials: analysis of natural and man-made materials*. BoD–Books on Demand, 2011. 14

[139] Thomas, C. *Étude des mécanismes d'endommagement des composites fibres de carbone/matrice polyamide: application à la réalisation de réservoirs de stockage de gaz sous haute pression de type IV*. PhD thesis, École Nationale Supérieure des Mines de Paris, 2011. 29

[140] Thomas, S., Gopi, S., and Amalraj, A. *Biopolymers and their industrial applications: from plant, animal, and marine sources, to functional products*. Elsevier, 2020. 11

[141] Thomason, J., Carruthers, J., Kelly, J., and Johnson, G. Fibre cross-section determination and variability in sisal and flax and its effects on fibre performance characterisation. *Composites Science and Technology 71*, 7 (2011), 1008–1015. 61, 65

[142] Toledo Filho, R. D., Ghavami, K., Sanjuán, M. A., and England, G. L. Free, restrained and drying shrinkage of cement mortar composites reinforced with vegetable fibres. *Cement and concrete composites 27*, 5 (2005), 537–546. iii, 60

[143] Toupe, J. L. *Optimisation des propriétés mécaniques de composites à base de fibres naturelles: Application à un composite de fibre de lin avec un mélange de polyéthylène/polypropylène d'origine post-consommation*. PhD thesis, Université Laval, 2015. 14

[144] Vaidya, U., and Chawla, K. Processing of fibre reinforced thermoplastic composites. *International Materials Reviews 53*, 4 (2008), 185–218. ii, 30

[145] Wang, A., Xian, G., and Li, H. Effects of fiber surface grafting with nano-clay on the hydrothermal ageing behaviors of flax fiber/epoxy composite plates. *Polymers 11*, 8 (2019), 1278. 15

[146] Weibull, W. A statistical theory of strength of materials. *IVB-Handl.* (1939). 64

[147] Williams, T., Hosur, M., Theodore, M., Netravali, A., Rangari, V., Jeelani, S., et al. Time effects on morphology and bonding ability in mercerized natural fibers for composite reinforcement. *International Journal of Polymer Science 2011* (2011). ii, 24

[148] Wirawan, R. Recyclability of natural fiber-filled thermoplastic composites. 33

[149] Yapnaa, D. B., Talontsic, F. M., Kouamb, S. F., Hussaind, H., Ngadjuie, B. T., Greenf, I. R., and Krohng, K. Chemical constituents of the seeds of raphia farinifera. *Syllabus Review Science Series 4* (2013), 9–11. 56

[150] Yin, H., and Zhao, Y. *Introduction to the micromechanics of composite materials*. CRC press, 2016. 2

[151] Yuan, Q., Liu, Z., Zheng, K., and Ma, C. *Civil Engineering Materials: From Theory to Practice*. Elsevier, 2021. 40

[152] Zaman, H. U., and Khan, R. A. Acetylation used for natural fiber/polymer composites. *Journal of Thermoplastic Composite Materials 34*, 1 (2021), 3–23. 25

[153] ZEDDOUR MOHAMED BRAH, Z. *PRÉDICTION DES MÉCANISMES D'ENDOMMAGEMENT DANS UN MATÉRIAU COMPOSITE*. PhD thesis, 2018. ii, 6, 37, 38

[154] Zhou, Y., Fan, M., and Chen, L. Interface and bonding mechanisms of plant fibre composites: An overview. *Composites Part B: Engineering 101* (2016), 31–45. 21

[155] Zohuri, G. Polymer science: a comprehensive reference. iii, 61

[156] Zwawi, M. A review on natural fiber bio-composites, surface modifications and applications. *molecules 26*, 2 (2021), 404. 25

Abstract

This present work is focused on the elaboration of different types of biocomposites by recycling the waste plastic bottle caps made of high-density polyethylene (HD-PE) and using it as a matrix reinforced with Alfa fibers and Pozzolan powder, followed by a mechanical and morphological characterization. Furthermore, a numerical investigation was pursued to develop an analytical model based on a genetic approach, using the Weibull model, to study the effect of damage to the fibre-matrix interface of four biocomposite materials based on polylactic acid (PLA) reinforced with three types of natural fibres raffia, alfa, and sisal fibres.

The experimental results are in good agreement with the numerical investigation. indicate that natural fibers-based composite materials can represent a good potential in improving interfacial properties. The study shows an alternative for producing friendly biodegradable composites that can be a solution for plastic waste reduction and a substitute for petrochemical-derived products.

Keywords: Bio-composites, high-density polyethylene, Recycling, Alfa Problème singulier, Pozzolan, Genetic approach, Weibull model, polylactic acid, raffia, sisal.

Abstract

Ce présent travail se concentre sur l'élaboration de différents types de biocomposites en recyclant les déchets de bouchons de bouteilles en plastique en polyéthylène haute densité (HD-PE) et en les utilisant comme matrice renforcée de fibres Alfa et de poudre de pouzzolane, suivi d'une opération mécanique et caractérisation morphologique. Par ailleurs, une investigation numérique a été poursuivie pour développer un modèle analytique basé sur une approche génétique, utilisant le modèle de Weibull, pour étudier l'effet des dommages à l'interface fibre-matrice de quatre matériaux biocomposites à base d'acide polylactique (PLA) renforcé de trois types de fibres naturelles de raphia, d'alfa et de sisal.

Les résultats expérimentaux sont en bon accord avec l'investigation numérique. indiquent que les matériaux composites à base de fibres naturelles peuvent représenter un bon potentiel pour améliorer les propriétés interfaciales.

L’étude montre une alternative pour produire des composites biodégradables respectueux qui peuvent constituer une solution pour la réduction des déchets plastiques et un substitut aux produits dérivés de la pétrochimie.

Keywords: Bio-composites, polyéthylène haute densité, Recyclage, Alfa, Pouzzolane, Approche génétique, modèle Weibull, acide polylactique, raphia, sisal.

ملخص

يركز هذا العمل الحالي على إعداد أنواع مختلفة من المركبات الحيوية عن طريق إعادة تدوير أغطية الزجاجات البلاستيكية المصنوعة من البولي إيثيلين عالي الكثافة (س-ص‌ز) واستخدامها كمصفوفة معززة بألياف ألفا ومسحوق البوزولان، يليها عملية ميكانيكية وكيميائية. التوصيف المورفولوجي. علاوة على ذلك، تم إجراء بحث عددي لتطوير نموذج تحليلي يعتمد على المنهج الوراثي، باستخدام نموذج ويبل، لدراسة تأثير الضرر الذي يلحق بالواجهة الليفية لأربع مواد مركبة حيويا تعتمد على حمض البوليلاكتيك (ص‌ش‌ا) المقوى بثلاثة أنواع. من ألياف الرافية والألفا والسيزال الطبيعية.

النتائج التجريبية تتفق جيدا مع التحقيق العددي. تشير إلى أن المواد المركبة القائمة على الألياف الطبيعية يمكن أن تمثل إمكانات جيدة في تحسين خصائص الواجهة. وتظهر الدراسة بديلاً لإنتاج مركبات صديقة للبيئة وقابلة للتحلل والتي يمكن أن تكون حلاً لتقليل النفايات البلاستيكية وبديلاً للمنتجات المشتقة من البتروكيماويات. الكلمات المفتاحية: مركبات حيوية، بولي إيثيلين عالي الكثافة، إعادة التدوير، ألفا، بوزولان، مقاربة وراثية، نموذج ويبول، حمض متعدد اللبن، رافيا، سيزال.

الكلمات المفتاحية: : مركبات حيوية، بولي إيثيلين عالي الكثافة، إعادة التدوير، ألفا، بوزولان، مقاربة وراثية، نموذج ويبول، حمض متعدد اللبن، رافيا، سيزال.