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DEVELOPMENT AND INVESTIGATION OF NATURAL FIBER REINFORCED POLYMER HYBRID COMPOSITES FOR SUSTAINABLE APPLICATIONS

Doctoral dissertation
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KAUNO TECHNOLOGIJOS UNIVERSITETAS

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LIST OF ABBREVIATIONS

ALG – Alginate
AR – Aramid
ASTM – American society for testing and materials
B – Banana
B – Basalt
C – Caryota
CA – Acetate
CB – Montmorillonite
CLF – Chlorofibre
CLY – Lyocell
CMCs – Ceramic matrix composites
CNC – Calcined nanoclay
CTA – Triacetate
CUP – Cupro
CV – Viscose
E – E-glass
ED – Rubber
EDX – Energy dispersive X-ray
EFBB – Empty fruit bunch banana
EP – Epoxy polymers
F – Flax
FEA – Finite element analysis
FRCM – Fiber reinforced composite materials
FRCs – Fiber reinforced polymer composites
FRP – Fiber reinforced polymers
FTIR – Fourier transform infrared spectrometer
G – Graphite
GTEV – Green thermoplastic elastomer vulcanizate
H – Hemp
HCl – Hydrochloric acid
HDPE – High-density polyethylene
HIPS – High-impact polystyrene
LDPE – Low-density polyethylene
MAC – Modacrylic
MAPE – Maleate polyethylene
MAPP – Maleic anhydride grafted polypropylene
MF – Melamine formaldehyde
MMCs – Metal matrix composites
MMT – Montmorillonite nanoclay
NaOH – Sodium hydroxide
NF – Noil flax
NH – Noil hemp

OMCs – Organic matrix composites
P – Pineapple
PA – Polyamide
PA 6 – Nylon 6
PA 6,6 – Nylon 6,6
PAR – Acrylic
PC – Polycarbonate
PC – Polycarbonates
PE – Polyethylene
PES – Polyester
PET – Polyethylene terephthalate
PET – Polythylene terephthalate
PF – Phenol formaldehyde
PHA – Polyhydroxyalkanoates
PLA – Polylactic acid
PMMA – Poly methyl methacrylate
POM – Polyoxymethylene
PP – Polypropylene
PP – Polypropylene
PS – Polystyrene
PTFE – Flurofibre
PVAL – Vinylal
PVC – Polyvinyl chloride
RAFT – Reversible addition-fragmentation chain transfer
RF – Resorcinol-formaldehyde
ROM – Rule of mixtures
RPU – Rigid thermoset polyurethane
RTM – Resin transfer molding
S – Sisal
SEM – Scanning electron microscope
T-F – Treated flax
T-H – Treated hemp
T-N-F – Treated noil flax
T-N-H – Treated noil hemp
TS – Thermosets
U-F – Untreated flax
UF – Urea formaldehyde
U-H – Untreated hemp
U-N-F – Untreated noil flax
U-N-H – Untreated noil hemp
UPE – Unsaturated polyester
VARTM – Vacuum assisted resin transfer molding technique

INTRODUCTION

In recent years, there has been significant interest in the development of composites using natural fibers reinforced with polymer matrices. Natural fiber hybrid composites (NFCs) are a class of materials that combine natural fibers reinforcement with a polymer resin matrix [1]. These materials have gained attention for their ability to absorb carbon dioxide, reducing environmental pollution, and their emission of no harmful gases [2, 3]. Natural fibers offer numerous advantages over synthetic fibers, such as biocompatibility [4], low cost [5], high strength [6], and low environmental impact [7]. The potential use of natural fibers in the global market is projected to increase at a compound annual growth rate of 9.4% from 2017 to 2022, especially lingocellulosic fibers, including banana, pineapple, hemp, flax, sisal, kneaf, and jute, have emerged as potential substitutes for traditional glass fibers [8]. Various industries, including construction, biomedical, automotive, microfluidics, energy, marine, and military sectors, utilize natural fiber composites [9]. For instance, in the microfluidcs sector, natural fiber composites used to produce lightweight parts, such as cell culture, drug delivery, and point-of-care diagnostics [10]. However, due to their complexity, they were not designed in the current research.

In general, composite materials are often composed of two or more constituent materials, such as a matrix and reinforcement. The reinforcement is bound by a matrix, which acts as a binding substance [11]. The materials that are included in the matrix are known as reinforcements [12], and they provide composite with extra strength [13]. The importance of composites lies in their ability to deliver superior physical, chemical, and mechanical qualities compared to their individual elements. Composites can be classified as polymer, ceramic, or metal matrix composites depending on the matrix [14, 15]. The performance of natural fiber composites mainly depends on the reinforcement length, arrangement, shape, origin of available and interfacial bonding between fiber and matrix material. Fibers can be classified as continuous fibers and discontinuous fibers based on their length-to-diameter ratio. The arrangement of the fiber-reinforced phase is typically categorized as either woven or non-woven [16]. They come in a variety of forms, including laminate, fibrous, and particle composites [17], depending on the reinforcement. The reinforcements are divided into two categories: natural and synthetic, based on their origin and availability [18].

Recent studies have demonstrated a significant impact of the hybridization development of composite material properties [19]. These studies have explored innovative approaches to combine different materials with the aim of enhancing their overall qualities. The examples include combining two distinct types of natural fibers, combining two different natural fibers with synthetic fibers, or even combining two different types of synthetic fibers within the same matrix material. [20]. The mechanical properties of these composites have been studied to ensure an improvement over single fiber composites. By altering the size, type of filler, and orientation of fibers, different forms of hybrid composites can be created. The hybridization using different types of fiber material may be able to overcome the

shortcomings of one component [21, 22]. The hybrid effect observed in composite material typically depends on the mechanical and chemical characteristics of the fibers; as a result, the effect is reduced compared to the composites made of individual fibers [23]. Extensive research has been conducted in recent decades to explore the blending of synthetic and plant fibers in composite materials [24–27]. However, natural fiber-reinforced hybrid composites have received less research attention recently with the majority of studies focusing on the mix of natural and synthetic fibers. The utilization of carbon, glass, and aramid in conjunction with hemp, flax, bananas, and pineapples is the best illustration of such composites [28–30].

The single fiber composites are considered inferior to hybrid fiber-reinforced composites. Due to their superior mechanical qualities and affordable price, supplementary applications can be found [31]. Hence, the artificial fiber composites have been replaced by hybrid composites due to their superior qualities [32], especially in load-bearing and building applications [33, 34]. The use of products made from natural fiber composites in a variety of applications, including household appliances, sports equipment, tables, packing shelves, and lightweight goods, has increased [35–37]. Most natural fiber composites are short fiber composites with irregular reinforcing length and orientation distributions [38, 39]. Discontinuous fiber composites are known to have a modest elastic modulus and strength compared to the continuous fiber-reinforced composites [40]. However, there is more flexibility in the choice of fabrication techniques. Discontinuous fiber composites can be made by using a variety of plastic processing techniques. This opens up the potential for simplified mass production [41].

In composites, both the reinforcement and matrix maintain their unique physical and chemical properties. However, when combined, they create a distinctive set of properties that cannot be achieved by either component alone [42]. The major intention of these materials is to achieve a balance of properties through a combination of various constituents. Coupling agents, coatings, and fillers are additional components that may be present in composites. Coupling agents and coatings are employed to the fibers to enhance their compatibility with the matrix, promoting better wetting and bonding at the reinforcement-matrix interface [43]. The addition of fillers to the composite material in the form of nanoparticles is considered highly promising for improving the mechanical and physical properties of composites [44].

Despite their superior mechanical properties, natural fiber composites have some drawbacks, such as poor adhesion properties [45], ductile nature [46], moisture sensitivity [47], internal stresses [48], etc. The presence of these drawbacks may weaken the strength of the composite material and adversely affect its mechanical properties. In order to overcome these drawbacks, the attempts have been made to use various natural fiber reinforced hybrid polymer composites. Alkaline and heat treatments were conducted on the reinforcement materials, and various fabrication techniques were developed for the purpose of understanding fabrication errors. Hybrid composites with various weight fractions were fabricated to improve the mechanical properties and propose their use in sustainable applications, which are intended to use

in microfluidics, such as disposable devices, various components of microfluidic systems (e.g., substrates), and device packaging.

Aim

The aim of the doctoral dissertation is to develop hybrid fiber-reinforced polymer composites and investigate their mechanical, morphological, and wettability characteristics for sustainable applications.

Objectives

In order to accomplish the primary aim of the thesis, a set of specific objectives has been developed, which are as follows:

1. To fabricate hybrid composites by reinforcing natural fibers with an epoxy matrix material and investigate how the incorporation of hybridization affects the mechanical properties of the composites.
2. To determine the effect of chemical treatments on the mechanical properties of natural fiber-reinforced hybrid composites and compare their performance with untreated fiber composites.
3. To develop a numerical method and conduct experimental analysis to investigate the influence of various matrix materials on the properties of hybrid polymer composites.
4. To evaluate the influence of nanoparticles on hybrid fiber composites intended for the use in microfluidic applications.

Research methods

As mentioned earlier, the aim of this dissertation is to develop and characterize the mechanical properties of hybrid fiber-reinforced composites for sustainable applications, considering the influence of the matrix material, chemical treatments, incorporation of nanoparticles, and varying weight fractions of fibers. The composites were developed by using the hybridization concept with Caryota, sisal, banana, pineapple, hemp, flax, E-glass, and Basalt fibers as reinforcements and epoxy resin, EcoPoxy, and polyester resins as matrix materials. Graphite was used as the filler material for nanoparticle incorporation. The composites with different weight fractions were fabricated using hand layup, compression molding, vacuum-assisted resin transfer technique, and vacuum-assisted free lamination techniques. NaOH and HCl solutions were used for the chemical treatment purposes. The experimental studies assessed the mechanical properties, including tensile tests, flexural tests, and interlaminar shear tests conducted on Tinus Olsen H10KT, impact tests conducted on Coesfeld Magnus impact tester, and hardness tests conducted on Vickers hardness tester. Wettability analysis was conducted by using contact angle measurement analysis. Additionally, the morphological studies were conducted by using SEM analysis conducted on Hitachi S-3400N, and SEM-EDX analysis was conducted on Bruker Quad 5040. The theoretical studies were conducted using the modified Halpin–Tsai model.

Novelty of the research work

These are the key findings and results of the research work conducted on natural fiber-reinforced hybrid composites:

1. The mechanical properties of natural fiber hybrid composites are several times higher than those of individual fiber composites.
2. The experimental analysis suggests that the mechanical properties of natural fiber reinforced hybrid composites primarily depend on the volume fraction of reinforcement materials.
3. The interlaminar shear strength between the natural fibers and epoxy composites is two times stronger than the composites with EcoPoxy matrix material, as determined by both theoretical and experimental methods.
4. Alkaline treatments to the natural fibers are well responsive to improve the cleanliness, increased surface roughness, enhanced fiber-matrix adhesion, and greater compatibility with polymer matrices, all of which can impact the overall performance of composite materials.
5. The addition of graphite nanoparticles shows a reduction in the porosity content and improving moisture resistance of the hybrid composites.
6. The mechanical properties of the natural fiber hybrid composites are comparable to, or even superior to, those of synthetic fiber composites. This finding is supported by the morphological studies and wettability analysis, which further indicate excellent interfacial adhesion properties.

Statements for the defense

1. The morphological properties of hybrid composites can be noticeably improved through the introduction of vacuum assisted free lamination and resin transfer models during the fabrication process.
2. The use of alkaline treatments to improve the surface properties of natural fibers is a novel and effective method for enhancing the mechanical properties of natural fiber composites.
3. The study on interlaminar shear strength and shear energy of hybrid composites is a novel approach to assessing the interfacial bonding between the reinforcement and matrix, and it can provide insight into the durability and longevity of the composite material.

Practical value

The practical value of this research work, while closely related to the scientific novelty, can be summarized as follows.

1. The research on natural fiber hybrid composites contributes to the development of eco-friendly solutions that reduce the reliance on non-renewable resources and mitigate environmental impact.
2. The optimization of materials selection and fabrication techniques has allowed for the enhancement of mechanical performance of a composite material. This can lead to the development of stronger and more durable composite materials for use in lightweight structural applications.

3. The study on alkaline treatments and the use of graphite nanoparticles highlights the techniques to improve the performance of natural fiber composites. This knowledge can be applied in practical manufacturing processes to enhance the quality and properties of composite materials.
4. The development of natural fiber hybrid composites can be used to fabricate microfluidic devices, such as channels, chip substrates, chambers, mixers, stirrers, and valves.

Research approbation

The scientific results that have been obtained during the course of current research were published in various publications. These publications and presentations of research findings demonstrate the significance and impact of the research conducted during the PhD studies.

The results of the dissertation were published in 7 scientific papers, 6 of which were published in the international journals listed in the ISI Web of Science database, and have an impact factor. Notably, five out of the seven published papers were featured in Quartile 1 journals, while one was published in a Quartile 2 journal. One paper was published as a conference proceeding in an international conference.

The results were as well presented at 4 international conferences and 5 international research workshops.

Author's contribution

[A1]: **Atmakuri, Ayyappa**; Palevicius, Arvydas; Vilkauskas, Andrius; Janusas, Giedrius. Review of hybrid fiber based composites with nano particles–material properties and applications // Polymers. Basel : MDPI. ISSN 2073-4360. 2020, vol. 12, iss. 9, art. no. 2088, p. 1-30, **Impact factor 4.967, Q1.**
Ayyappa Atmakuri led the conceptualization and design of the research project, including developing the research question, study design, and data analysis plan. He as well played a key role in conducting the formal analysis and drafting the original manuscript.

[A2]: **Atmakuri, Ayyappa;** Palevicius, Arvydas; Kolli, Lalitnarayan; Vilkauskas, Andrius; Janusas, Giedrius. Development and analysis of mechanical properties of Caryota and sisal natural fibers reinforced epoxy hybrid composites // Polymers. Basel : MDPI. ISSN 2073-4360. 2021, vol. 13, iss. 6, art. no. 864, p. 1-19. **Impact factor 4.967, Q1.**
Ayyappa Atmakuri was responsible for collecting and curating the data as well as conducting the experimental analysis and creating visualizations of the results. He as well contributed to the writing and editing of the manuscript.

[A3]: **Atmakuri, Ayyappa**; Palevicius, Arvydas; Siddabathula, Madhusudan; Vilkauskas, Andrius; Janusas, Giedrius. Analysis of mechanical and wettability properties of natural fiber-reinforced epoxy hybrid composites // Polymers. Basel : MDPI. ISSN 2073-4360. 2020, vol. 12, iss. 12, art. no. 2827, p. 1-15. **Impact factor 4.967, Q1.**

Ayyappa Atmakuri led the conceptualization and design of the research article as well as conducting the investigation and analysis of the data. He was responsible for drafting the original manuscript and provided feedback and revisions of the manuscript.

[A4]: **Atmakuri, Ayyappa;** Palevicius, Arvydas; Janusas, Giedrius; Eimontas, Justas. Investigation of hemp and flax fiber-reinforced EcoPoxy matrix biocomposites: morphological, mechanical, and hydrophilic properties // Polymers. Basel : MDPI. ISSN 2073-4360. 2022, vol. 14, iss. 21, art. no. 4530, p. 1-21. **Impact factor 4.967, Q1.**

Ayyappa Atmakuri contributed to the experimental investigation and morphological analysis. He as well contributed to the methodology, formulation and writing of the original draft.

[A5]: **Atmakuri, Ayyappa**; Palevicius, Arvydas; Vilkauskas, Andrius; Janusas, Giedrius. Numerical and experimental analysis of mechanical properties of natural-fiber-reinforced hybrid polymer composites and the effect on matrix material // Polymers. Basel : MDPI. ISSN 2073-4360. 2022, vol. 14, iss. 13, art. no. 2612, p. 1- 18. **Impact factor 4.967, Q1.**

Ayyappa Atmakuri was responsible for collecting and curating the data as well as conducting the experiments, and he contributed to the conceptualization, methodology, writing of the original draft preparation, and visualization.

[A6]: **Atmakuri, Ayyappa**; Kolli, Lalitnarayan; Palevicius, Arvydas; Urbaite, Sigita; Janusas, Giedrius. Influence of filler materials on wettability and mechanical properties of Basalt/E-glass woven fabric–reinforce composites for microfluidics // Micromachines. Basel : MDPI. ISSN 2072-666X. 2022, vol. 13, iss. 11, art. no. 1875, p. 1-14. **Impact factor 3.523, Q2.**

Ayyappa Atmakuri was involved in the fabrication and research methodology (mechanical properties, wettability properties, and morphological studies), formal analysis, investigation (experimental), visualisation (scanning electron microscopic investigation) writing of the original draft, and formation of the results.

1. STATE OF THE ART

1.1. Natural fiber reinforced hybrid composites

In recent decades, there has been significant interest in utilizing natural fibers, both renewable and non-renewable, such as banana, okra, pineapple, hemp, sisal, flax, bamboo, and jute, to produce composite materials [49]. Cellulose fiber-producing plants can be categorized into various types, including bast fibers (flax, hemp, jute, and kenaf), seed fibers (kapok, coir, and cotton), leaf fibers (abaca, pineapple, and sisal), grass and reed fibers (wheat, rice, and corn), core fibers (hemp, jute, and kenaf) as well as other miscellaneous sources (roots and wood) [50–52].

Natural fibers offer numerous advantages compared to the synthetic fibers [53]. These include their renewable nature, biodegradability, low cost, minimal damage to processing equipment, lightweight properties, improved surface finish of composite molded parts, flexibility during processing, favorable mechanical properties, such as tensile modulus and flexural modulus, and reduced health hazards. The benefits of natural fibers surpass those of synthetic fibers in multiple aspects [54].

In 1960s, Toyota made a significant breakthrough by incorporating fenders and deck lids made from soy protein-based eco plastic into the cars [55]. In the past, the research and development of composite materials derived from renewable and sustainable sources were regarded as relatively insignificant compared to the synthetic composites sector. However, nowadays, the utilization of eco composites has become a crucial element in the future of industrial practices. The growing emphasis on sustainable growth is the primary driving force behind the increasing importance of composite materials [56].

The growing societal demand for environmentally-friendly and biodegradable materials has created a strong impetus for advancements in composite science. The biodegradability of natural fibers is a particularly noteworthy aspect of their application in polymeric materials [57]. By designing a blend of lightweight and robust biomaterials, it is possible to develop advanced lightweight composites that can serve as alternatives to the conventional synthetic structures or metal components used in sustainable applications [58]. Despite the favorable characteristics of natural fibers, there are certain drawbacks that make their use in composites for semi-structural or structural applications inconvenient. These drawbacks include high susceptibility to moisture absorption, thermal and mechanical degradation during manufacturing, poor compatibility with certain polymeric matrices, and low resistance to microbial degradation [59, 60]. In order to address these challenges, the development of natural fiber hybrid composite materials presents a potential solution. Hybrid composites, which combine two different fibers, either natural or synthetic fibers, offer a promising strategy for composite materials. The concept behind hybridization is to introduce different types of fibers in order to overcome the limitations of composites reinforced with only one type of fiber [61–63].

1.2. Reinforcement and matrix interfacial adhesion concerns

The interfacial adhesion between the reinforcement and matrix material plays a crucial role in the mechanical performance of a composite material [64]. The inadequate adhesion between the natural fibers and matrices remains a significant constraint in the industrial utilization of natural fibers. The distinctive chemical composition, hydrophilic nature, and surface characteristics of natural fibers hinder their ability to establish a strong interfacial adhesion with hydrophobic polymer matrices [65]. This weak adhesion at the natural fiber/matrix interface can create a pathway for water penetration, leading to the exposure of hydrophilic fiber groups to water molecules. Consequently, this can result in the degradation of the composite and a decline in its mechanical properties [66–68].

Achieving the best mechanical performance in a composite material relies on maximizing the ability to transfer stress across the fiber-matrix interface, which is closely linked to the level of adhesion. The adhesion at the interface can be attributed to various factors, including interaction forces, chemical bonds, and residual stresses [69]. In hybrid composites, there are two types of interfaces, namely, interplay interface and intraplay interface. The quality of adhesion at both types of interfaces plays a significant role in determining the mechanical properties of the hybrid composite. However, the interplay interface has been specifically recognized as being fundamental in hybrid composites, as it influences the overall performance of the material [70]. Recent studies have highlighted that surface modification of fibers can enhance the adhesion properties of composite materials. One effective method of surface modification is chemical treatment using alkaline or alkane solutions. These treatments have been found to have a significant impact on the morphology of the fibers [71].

1.3. Surface modification treatments

The primary challenge in the industrial utilization of natural fibers lies in their poor compatibility with polymer matrices. The unique chemical composition and surface characteristics of natural fibers hinder their ability to establish a strong interfacial adhesion with hydrophobic polymer matrices, resulting in composites with inferior mechanical properties [72].

The main constituents of plant fibers include cellulose, hemicellulose, wax, lignin, and pectin. The relative composition of these components can vary depending on the growing conditions [73]. Cellulose, hemicellulose, and lignin are the key contributors to the physical properties of the plant fibers. Among them, cellulose is the most important organic constituent, providing rigidity and strength to the fiber [74].

In order to enhance the adhesion of natural fibers to matrix materials, various physical and chemical treatment methods can be employed to modify the fiber surfaces. One approach is to reduce the amount of hydrophilic hydroxyl groups that are present on the fiber surface [75]. The physical treatments, such as surface fibrillation, cold plasma treatment, and electric discharge method, can alter the surface properties of fibers and promote better mechanical bonding between the fibers and the

matrix [76]. Chemical treatment is another widely used method to modify the surfaces of natural fibers. It aims to reduce the hydrophilic nature of the fibers and improve their compatibility with the matrix. Different chemical treatments have been employed, including alkalization, acetylation, salinization, benzoylation, and peroxidation. These treatments effectively modify the fiber surfaces and enhance their properties in terms of adhesion and compatibility with the matrix [77–79].

1.4. Mechanical properties

The diverse range of properties exhibited by the natural fibers is primarily influenced by the factors, such as plant species, growth (atmospheric conditions) conditions, and the method used for fiber extraction [80]. Additionally, the properties of natural fibers are influenced by the fiber cell geometry specific to each type of cellulose and its degree of polymerization. These factors play a crucial role in determining the unique characteristics and performance of natural fibers [81, 82]. Bast fibers, such as hemp, flax, banana, pineapple, are highly favored among non-wood lignocellulosic fibers due to their exceptional technical properties and the ease of their extraction from raw resources [83], typically through retting and manual extraction techniques [84]. The manual extraction method results in high-quality fibers, but it can be a time-consuming and labor-intensive process [85]. These fibers are sometimes referred to as stem fibers, because they are derived from the outer cell layers of the plant's pseudo-stem, which is a part of the stem that contains fibers.

Extensive research has been carried out to examine the impact of fiber content and orientation on the mechanical properties of various natural fiber-reinforced polymer composites. These studies encompass a wide range of composites, including both thermoplastic and thermosetting materials [86–89]. Alhazmi et al. [90] studied the tribological and mechanical properties of epoxy reinforced with hybrid nanoparticles. The results demonstrated that the incorporation of nanoparticles into the epoxy resin enhanced its wear resistance. However, it has been observed that there were some drawbacks associated with this reinforcement, including the reduction of tensile strength and modulus of elasticity. Karimzadeh et al. [91] studied the effect of stacking sequence on the mechanical properties and moisture absorption characteristics of hybrid pineapple/glass fiber composites. The findings revealed that hybrid composites exhibited superior flexural strength, tensile strength, and tensile modulus compared to the pure composites. Additionally, it has been observed that pure pineapple composites absorbed a higher percentage of water compared to the hybrid composites. In a study conducted by Senthil et al. [92], the mechanical performance of biocomposites made from hemp and sisal fibers were investigated. The experimental results indicated that hybrid composites exhibited only minor variations in the mechanical properties when the stacking sequence was altered. Among the hybrid composites, the one with an intercalated arrangement demonstrated the highest tensile modulus. Based on the review of relevant literature studies, it has been observed that alkaline-treated natural fibers have demonstrated improved mechanical properties due to the enhanced adhesion between the reinforcement and matrix materials. While epoxy resin materials are commonly used and flexible in

composite fabrication, they possess certain drawbacks, such as non-biodegradability, chemical harshness, and limited skin-friendliness [93, 94]. In order to tackle the mentioned challenges, the current research investigates the use of both epoxy and bio-inspired resin materials as potential alternatives. Additionally, a variety of natural fibers, including banana, pineapple, hemp, flax, okra, Caryota, and sisal fibers, are employed in the fabrication process for comparative analysis. By utilizing different types of fibers, the aim is to evaluate their suitability and performance in composite materials, considering factors such as mechanical properties, adhesion, and overall sustainability.

The detailed description of various natural fibers, synthetic fibers, chemical and mechanical properties, fabrication techniques, thermoset resins, thermoplastic resins, and applications were explained clearly in the review article titled “The review of hybrid fiber composites with nano particles - material properties and applications," which was published in the *Polymers* journal in 2020. Figure 1 shows the graphical abstract of the published article.

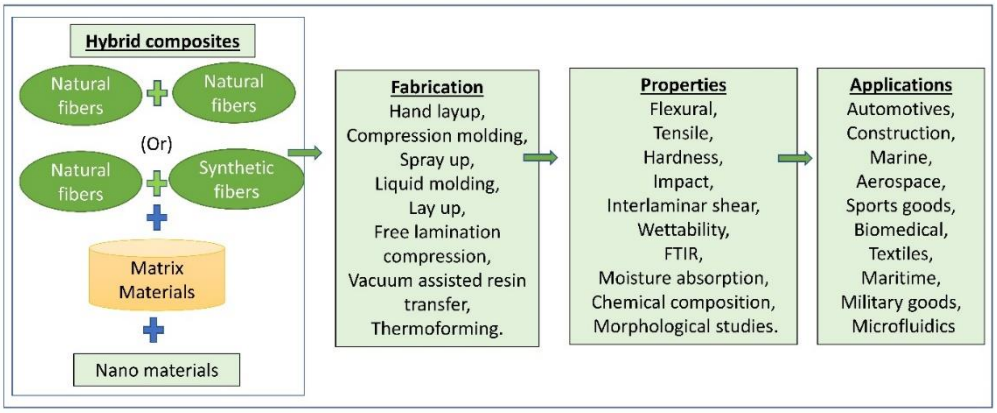


Fig. 1. Overview of the hybrid composites: fabrication, properties, applications

The review as well emphasizes the positive impact of filler materials on the overall strength of composite materials. Moreover, the article discusses the drawbacks of natural fibers, such as their inconsistent mechanical properties, flame retardancy, and moisture absorption, which can be addressed through chemical treatments. The review motivates further research to improve the fabrication techniques and the use of nanofillers to enhance the properties and expand the applications of fiber-based hybrid composites; furthermore, it provides valuable insights for formulating the aim and objectives of the dissertation.

2. OVERVIEW OF THE PUBLICATIONS

2.1. "Development and analysis of mechanical properties of Caryota and sisal natural fibers reinforced epoxy hybrid composites" (scientific publication no. [A2])

This sub-section is based on a scientific paper titled "Development and analysis of mechanical properties of Caryota and sisal natural fibers reinforced epoxy hybrid composites," which was published in the *Polymers* journal in 2021. It covers the 1st objective of the research work. This paper presents the fabrication of Caryota and sisal natural fiber hybrid composites in the presence of epoxy resin along with the hardener and the investigation of mechanical properties.

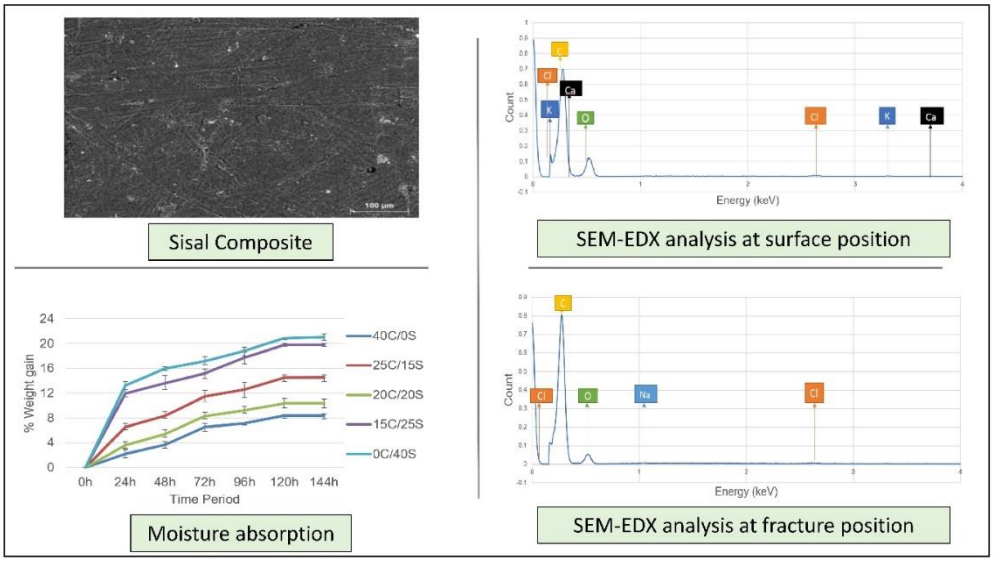


Fig. 2. Visual representation of the key results

The article reports on the development and characterization of hybrid composites using carbon and sisal fibers as reinforcements and epoxy resin as the matrix material. The composites were prepared by using the hand layup method, and their mechanical and moisture absorption properties were evaluated. Tensile, flexural, and impact tests were conducted, and the results showed that the hybrid composites exhibited superior mechanical properties compared to the pure composites. The tensile strength and modulus of the hybrid composites were observed to be up to 38.2 MPa and 6.44 GPa, respectively, whereas in the pure composites, they were up to 25.7 MPa and 5.66 GPa, respectively. The hybrid composites as well exhibited higher flexural strength and impact energy compared to the pure composites. Moisture absorption tests showed that all the composites responded to moisture over time, and the rate of absorption was higher in pure composites than in the hybrid composites. The SEM analysis revealed that the weight percentage of carbon was higher at the surface than the fracture position, while the weight percentage of oxygen was higher

at the fracture position than the surface, indicating that the concentration of matrix material was higher at the surface position, while the fiber percentage was higher at the fracture (inside) position. The study highlights the potential of using hybrid composites in various industrial applications and suggests further research to optimize the mechanical properties of composites by using various natural fibers and fabrication techniques. It was recommended to investigate the hybrid composites with different natural fibers and fabrication techniques further in order to improve their mechanical properties. Overall, this article highlights the potential of natural fiber hybrid composites as a sustainable and environmentally friendly alternative to the traditional composite materials, and it can be used as a packaging material for microfluidic devices. The material's biocompatibility and low cost make it a suitable option for disposable devices.

2.2. "Analysis of mechanical and wettability properties of natural fiber-reinforced epoxy hybrid composites" (scientific publication no. [A3])

This sub-section is based on a scientific paper titled "Analysis of mechanical and wettability properties of natural fiber-reinforced epoxy hybrid composites," which was published in the *Polymers* journal in 2020. It covers the 1st objective of the research work. It presents the development of hemp, flax, banana, and pineapple natural fiber hybrid composites in the presence of epoxy resin along with the hardener, and the mechanical properties of two sets of hybrid composites were investigated, and the results were compared.

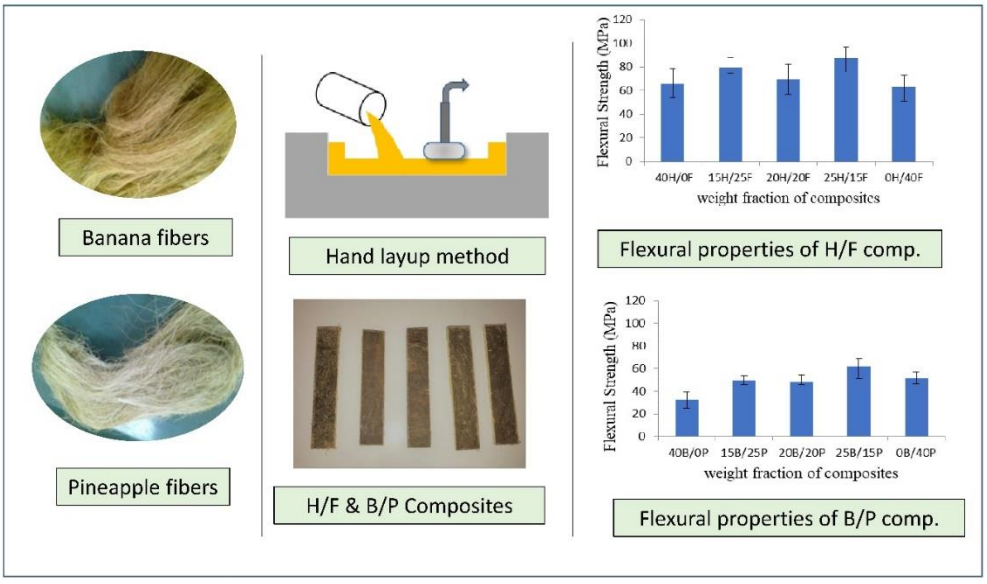


Fig. 3. Graphical abstract of the research outcomes

Hybrid composites were fabricated by using a combination of hemp, flax, banana, and pineapple fibers as reinforcements and an epoxy resin as the matrix material. The composites were fabricated based on the concept of hybridization,

resulting in two sets of hybrid composites (H/F and B/P) with five different weight fractions. Mechanical and wettability properties were tested and compared between the two sets of composites. The mechanical properties were evaluated by measuring the flexural and interlaminar shear strength properties. The flexural properties of the hemp/flax and banana/pineapple composites ranged from 60 to 88 MPa and from 38 to 69 MPa, respectively, with the 25H/15F hybrid composite demonstrating the highest flexural strength at 87.57 MPa and the 40B/0P composite, showing the least strength at 39.36 MPa. The interlaminar shear strength properties of the individual fiber composites were found to be superior to the hybrid composites in both cases. The wettability analysis indicated that all the composites had a contact angle lower than 90º, indicating hydrophilic surface properties.

2.3. "Investigation of hemp and flax fiber-reinforced EcoPoxy matrix biocomposites: morphological, mechanical, and hydrophilic properties" (scientific publication no. [A4])

This sub-section is based on a scientific paper titled "Investigation of hemp and flax fiber-reinforced EcoPoxy matrix biocomposites: morphological, mechanical, and hydrophilic properties," which was published in the *Polymers* journal in 2022. It covers the 2nd objective of the research work. It presents the investigation of mechanical and morphological properties of hemp, flax, noil hemp, and noil flax natural fiber hybrid composites in the presence of EcoPoxy resin along with the hardener. This outcome was achieved due to the collaboration of research teams from two international institutions.

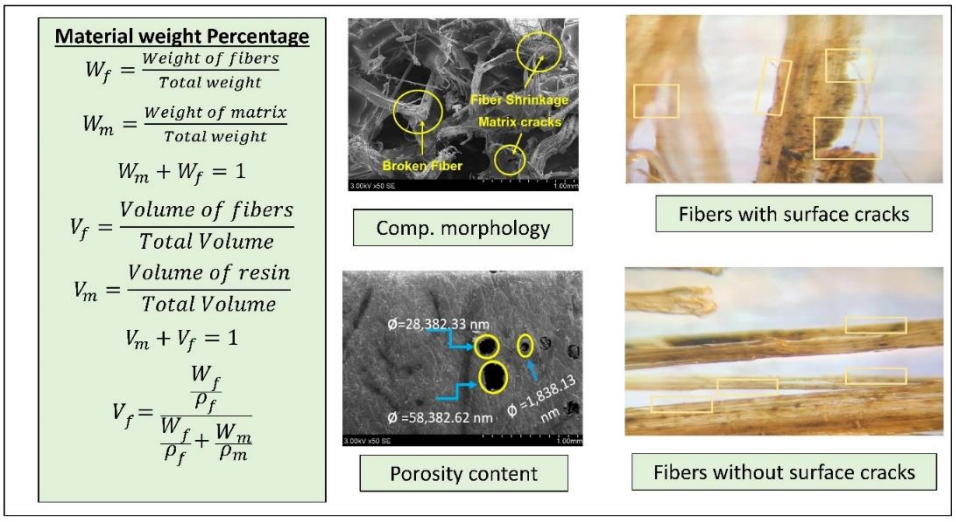


Fig. 4. Overview of the main findings

In this article, the mechanical and morphological properties of hemp, flax, noil hemp, and noil flax natural fiber hybrid composites in the presence of EcoPoxy resin along with the hardener were investigated. The composites were developed by using the natural fibers as reinforcements and EcoPoxy resin as the matrix material. In order

to enhance the fiber surface properties and adhesion properties, an alkaline treatment was carried out with NaOH solution, and the results were observed with microscopic analysis and FTIR analysis. The impact of chemical treatment on the natural fiber was observed through the elimination of surface cracks, kink bands, and surface smoothness presented in the natural fibers. Subsequently, it enhanced the fiber adhesion properties. The contact angle was observed lower than 90º in both normal and noil fiber cases. However, in the noil case, the hybrid composite exhibited a contact angle close to 90º, which means that the composites tend to show the hydrophobic surface properties. It has been observed from the mechanical properties that the hemp/flax natural fiber hybrid biocomposites showed superior mechanical properties over the noil fiber hybrid composites. The strong surface properties and mechanical properties make these composites a suitable option for packaging material in microfluidic devices. Additionally, the biocompatibility and low cost of the material make it a suitable option for disposable microfluidic devices.

2.4. "Numerical and experimental analysis of mechanical properties of natural-fiber-reinforced hybrid polymer composites and the effect on matrix material" (scientific publication no. [A5])

This sub-section of the thesis is based on the scientific paper titled "Numerical and experimental analysis of mechanical properties of natural-fiber-reinforced hybrid polymer composites and the effect on matrix material," which was published in the *Polymers* journal in 2022. It covers the 3rd objective of the research work. This article presents the impact of matrix material on the mechanical and morphological properties of natural fiber reinforced hybrid composites. This outcome was achieved due to the collaboration of research teams from two international institutions.

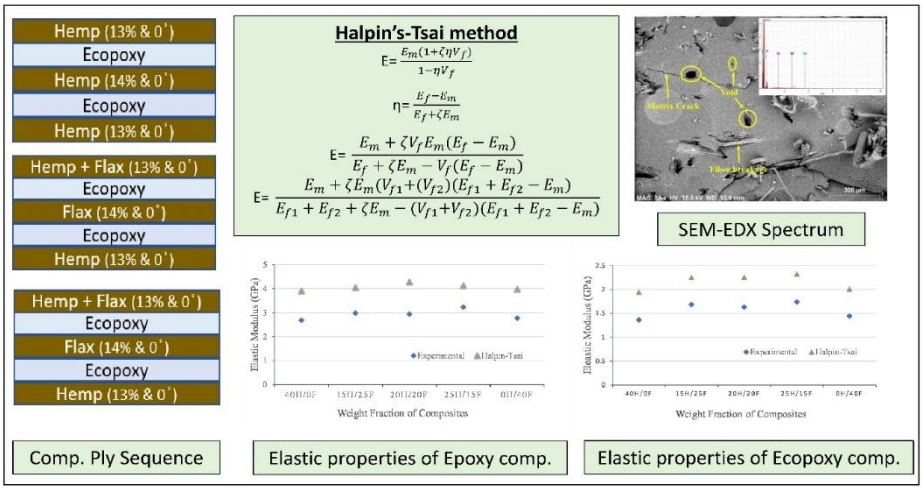


Fig. 5. Summary of the research methodology and key findings

This research article investigated the effect of matrix material on the mechanical properties of natural fiber hybrid composites. The composites were fabricated by using hemp and flax fibers as reinforcements and epoxy resin and EcoPoxy resin with

a hardener as matrix materials. The mechanical properties of the composites, such as flexural strength, interlaminar shear strength, and wettability, were analyzed and compared between the two types of matrix materials. The Halpin–Tsai approach was used for the theoretical analysis of the composites. The results showed that the epoxy-based composites exhibited superior mechanical properties compared to the EcoPoxy matrix-based hybrid composites. Additionally, both types of hybrid composites showed better mechanical properties than the pure composites. SEM-EDX analysis revealed that the weight percentage of carbon was higher at the surface over the fracture position in both cases, while the weight percentage of the oxygen was higher at the fracture position. The percentage of oxygen was higher in the case of EcoPoxy matrix material due to its bioactivity. The modified Halpin–Tsai method was used for the theoretical evaluation, but it has been found that the error percentage was higher in this method compared to the numerical methods. Therefore, Halpin–Tsai method may not be suitable for the use in natural fiber hybrid composites.

2.5. "Influence of filler materials on wettability and mechanical properties of Basalt/E-glass woven fabric–reinforce composites for microfluidics" (scientific publication no. [A6])

This sub-section of the thesis is based on a scientific paper titled "Influence of filler materials on wettability and mechanical properties of Basalt/E-glass woven fabric–reinforce composites for microfluidics," which was published in the *Micromachines* journal in 2022. It covers the 4th objective of the research work. This article presents the impact of nano filler material on the mechanical and morphological properties of Basalt, E-glass fiber reinforced hybrid composites. This outcome was achieved due to the collaboration of research teams from two international institutions.

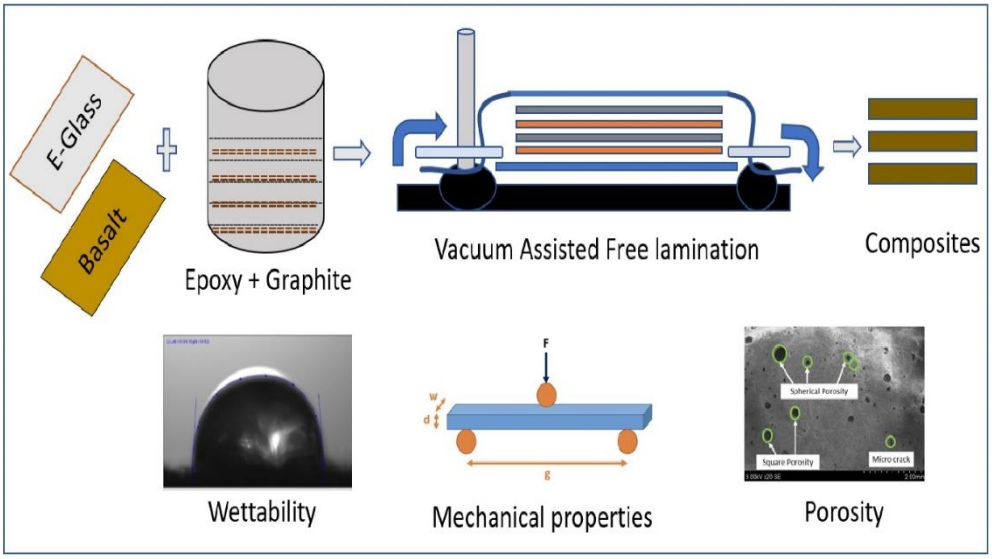


Fig. 6. Schematic representation of the research process

In this scientific publication, the influence of filler particles on the mechanical properties of hybrid composites were investigated. A new method of fabrication technique was developed for the fabrication of Basalt/E-glass fiber hybrid composites. Graphite powder nanoparticles were employed as a filler material in the composites. These nanoparticles were prepared through the mechanical milling process and had particle sizes ranging from 50 to 80 nanometers. These nanoparticles were purchased from Sigma Aldrich Chemicals Private Limited, located in Bangalore, India. The impact energy levels of hybrid composites with 5%, 10%, and 15% weight concentrations of graphite filler particles were 3.8 KJ/m2, 4.0 KJ/m2, and 3.9 KJ/m2, respectively, which were higher than those of the pure Basalt and E-glass composites. SEM analysis showed that the addition of filler particles improved the interfacial bonding between the fibers and the matrix, resulting in better mechanical properties. However, when the weight concentration of filler particles exceeded 10%, the mechanical properties of the hybrid composites decreased due to the agglomeration and poor dispersion of filler particles. The results suggest that the addition of a small amount of filler particles can significantly improve the mechanical properties of hybrid composites, but the weight concentration of the filler particles should be carefully controlled to avoid adverse effects on the properties of the composite material. Overall, the findings of this research have important implications for the development of high-performance composite materials with improved mechanical properties, and they can be used as a substrate material for microfluidic chips. The material's low cost and ease of manufacturing make it an attractive option for the microfluidic chip fabrication.

3. CONCLUSIONS

After the overview of the five research papers, the following findings, as formulated by the aim and objectives, can be derived based on the four objectives that were established at the beginning of the study:

1. The hybrid composites based on the natural fibers and epoxy matrix materials were fabricated by using various fabrication techniques. The Caryota and sisal fiber hybrid composites exhibited improved mechanical properties compared to the single fiber composites with a tensile strength of 38 MPa, flexural strength of 89.16 MPa, and flexural modulus of 3.40 GPa. The hemp and flax fiber composites showed an interlaminar shear strength ranging between 6.30 and 13.50 MPa. The 25H/15F hybrid biocomposites had a maximum flexural strength and modulus of 87.50 MPa and 3.45 GPa.

2. The alkaline treatment with a 5% NaOH solution on the reinforcement material showed an adverse effect on the mechanical properties. The morphological study revealed the elimination of surface cracks, kink bands, and improved surface roughness in chemically treated fiber materials, which ultimately affected the mechanical properties. The FTIR analysis indicated the changes in functional groups and the intensity of hydroxyl groups (-OH). The intensity of (-OH) hydroxyl peaks that were observed before the treatment was around 2,700–2,900 cm^{-1}, whereas after treatment, it was around 2,200–2,500 cm^{-1}. Additionally, a reduction in lignin and hemicellulose peaks was observed at 1,130 cm^{-1} and 960 cm^{-1}, respectively. These changes in the chemical structure indicated functional group transformations, impurity removal, and improved cellulose crystallinity, thereby enhancing the fiber surface properties.

3. It has been observed that the epoxy-matrix-based hybrid composites exhibited two times the mechanical properties compared to EcoPoxy-matrix-based hybrid composites. The maximum flexural and shear strengths for epoxy composites were 78 MPa and 13 MPa, respectively, while for EcoPoxy composites, they were 30 MPa and 10 MPa, respectively. The elastic properties of the composites were determined by the theoretical and numerical analyses, and it has been observed that the modified Halpin–Tsai method had higher errors.

4. Basalt and E-glass woven-fiber-based epoxy hybrid composites with 10% graphite filler material exhibited superior mechanical properties compared to the individual Basalt and E-glass fiber composites. The B/E/G15 composite showed the highest flexural strength of 43.81 MPa and flexural modulus of 3.57 GPa, while the epoxy-based B/E/10G composites displayed the highest tensile properties. The porosity content in hybrid composites with filler material was lower than in those without filler material.

Based on the findings, natural fiber hybrid composites have potential as a substitute for the artificial fiber composites in sustainable applications, including chip substrates, packaging devices, microfluidic channel, and valve materials.

Proposal for further investigations in the future

The study's findings suggest several potential areas for the future research.

1. Further investigation of the effect of different chemical treatments on the natural fibers to improve their surface properties and mechanical properties.
2. Further exploration of the effect of different filler materials on the mechanical properties of natural fiber hybrid composites.
3. The moisture absorption behavior of natural fiber hybrid composites, including the effect of different environmental conditions and storage time.
4. The investigation of the use of natural fiber hybrid composites in other microfluidic device applications, besides substrates, packaging devices, and microfluidic chips. Future research could explore their potential in other areas, such as microfluidic pumps, valves, and sensors.

4. ABSTRACT

The synthetic fibers have been dominant in the field of composite development for many years. However, there is currently a growing interest in the environmental awareness within the industrial applications. In the modern era of advancement, the researchers are increasingly focused on developing the biodegradable materials that align with sustainable principles, aiming to prevent the use of non-biodegradable resources. The fibers that derive from organic sources (agricultural waste) offer several advantages in comparison to the traditional synthetic fibers, including lower cost and density, non-toxicity, minimal environmental impact, and easier waste disposal. In addition, they offer comparable features to the artificial fiber composites but at a lower cost and with higher sustainability, and ease of production, making them a promising alternative for many sustainable applications. Despite the many benefits of natural fibers as reinforcements, there are some downsides to consider. These include issues, such as fiber separation, poor surface properties, variable mechanical performance, poor adhesion properties, moisture uptake behavior, porosity content, and delamination, which can collectively have a significant impact on the mechanical properties and final failure of the composite. This PhD research aims to investigate the causes behind these issues and explore the ways to improve the mechanical properties of natural fiber hybrid composites.

The preparation of reinforcement materials for fabrication is crucial in creating a strong bond between the reinforcement and matrix materials. Previous studies have indicated that the chemical treatments (e.g., alkaline or alkane treatments), and heat treatments of natural fibers can enhance their surface properties. In this study, the reinforcement materials were created by using the hand-sitting process, and the fibers were separated by using a cotton comb from the thick stems and small pallets. The separated fibers underwent an alkaline treatment and a heat treatment process to dry them. A 5% NaOH solution at 60 °C was used for hemp, flax, noil hemp, and noil flax fibers for alkaline treatment. Caryota and sisal fibers were treated with a 2% HCl solution to remove the gelatinous substance. The optical microscopic analysis and FTIR analysis were used to evaluate the effect of alkaline treatment on hemp and flax fibers before and after the chemical treatment. The results indicated that the chemical treatment improved the fiber performance by eliminating surface microcracks and fiber kink bands, resulting in a smoother surface finish. The FTIR analysis as well showed that the treated fibers exhibited stronger OH groups of cellulose particles, enhancing their chemical properties.

In order to improve the mechanical performance, the study explored the concept of hybridization in composite material preparation. Hybridization refers to the creation of composite materials using two or more reinforcement materials under the same matrix material. The study used Caryota, sisal, hemp, flax, banana, pineapple, Basalt, and E-glass fibers as reinforcement materials with epoxy, EcoPoxy, and polyester resin used as matrix materials for comparison. Three fabrication techniques were employed for the production of hybrid composites: hand layup followed by compression molding, vacuum-assisted resin transfer techniques, and vacuum-

assisted free lamination process. Each set of composites included five different weight fractions with 250 to 300 samples fabricated for the comparison. Furthermore, one set of hybrid composites was created by incorporating filler material into the composite material, using nano graphite filler material with varying weight percentages added through the resin transfer technique.

Continuing the study on the impact of resin material on natural fiber hybrid composites, EcoPoxy (a bioactive flow cast resin) matrix material was introduced in the composite fabrication. The results were compared with those of epoxy matrix-based hybrid composites. As it can be seen from the mechanical properties, such as flexural properties and interlaminar shear strength properties, the epoxy resin matrix-based natural fiber hybrid composites are twice as strong as EcoPoxy matrix composites. Additionally, the wettability analysis showed that the epoxy-based composites always have a contact angle of less than 90º, which means that they tend to exhibit hydrophilic surface properties, whereas in EcoPoxy matrix composites, the contact angle is almost close to (nearly 90º) hydrophobic surface properties. Furthermore, the amount of moisture absorbed by the Ecopoxy composites is more than that absorbed by the epoxy composites, which weakens the strength of the composite material.

The introduction of filler (nano graphite) materials through the resin transfer process could preferably help in reducing the porosity content of composite material. The presence of porosity content in a composite material weakens its mechanical properties and serves as storage cells for moisture content. Additionally, the significance of the nano filler particles needs to be further explored, as they could pave the way for composites to be studied from a distinct perspective compared to the conventional method of looking at them in the light of hybrid composite structures. Moreover, the presence of nano particles in a composite material reduces the internal stress and improves the adhesion properties of the reinforcement and matrix material. The addition of nano graphite filler particles further improves the mechanical properties of the hybrid composites. Based on these findings, natural fiber hybrid composites have potential as a substitute for artificial fiber composites in various sustainable applications.

5. SUMMARY

Pastaraisiais metais išryškėjo didelis susidomėjimas kompozitų, naudojant natūralų pluoštą, sustiprintą polimerinėmis matricomis, kūrimu. Natūralaus pluošto hibridiniai kompozitai (NFC) yra medžiagų klasė, kurioje natūralaus pluošto armatūra derinama su polimerinės dervos matrica [1]. Šios medžiagos susilaukė dėmesio dėl savo gebėjimo sugerti anglies dioksidą, mažinti aplinkos taršą ir neišmesti kenksmingų dujų [2] [3]. Natūralūs pluoštai turi daug pranašumų, palyginti su sintetiniais pluoštais, pavyzdžiui, biologinis suderinamumas [4], maža kaina [5], didelis stiprumas [6] ir mažas poveikis aplinkai [7]. Buvo numatyta, , kad potencialus natūralių pluoštų panaudojimas pasaulinėje rinkoje nuo 2017 m. iki 2022 m. padidės 9,4 % metinio sudėtinio augimo tempo, ypač lignoceliuliozės pluoštai, įskaitant bananus, ananasus, kanapes, linus, sizalį, šerpetą ir džiutą. galimi tradicinio stiklo pluošto pakaitalai [8]. Įvairios pramonės šakos, įskaitant statybą, biomediciną, automobilių, mikrofluidikos, energetikos, jūrų ir karinius sektorius, naudoja natūralaus pluošto kompozitus [9]. Pavyzdžiui, mikroskysčių sektoriuje natūralaus pluošto kompozitai naudojami lengvoms dalims, tokioms kaip ląstelių kultūros, vaistų pristatymo ir diagnostikos taškas, gaminti [10]. Tačiau dėl jų sudėtingumo jie nebuvo sukurti atliekant dabartinį tyrimą.

Paprastai kompozicines medžiagas dažnai sudaro dvi ar daugiau sudedamųjų medžiagų, tokių kaip matrica ir armatūra. Armatūra yra surišta matrica, kuri veikia kaip rišamoji medžiaga [11]. Medžiagos, kurios yra įtrauktos į matricą, žinomos kaip sutvirtinimai [12] ir suteikia kompozitui papildomo stiprumo [13]. Kompozitų svarba slypi jų gebėjime užtikrinti geresnes fizines, chemines ir mechanines savybes, palyginti su atskirais elementais. Pagal savo matricą, kompozitai gali būti klasifikuojami kaip polimero, keramikos arba metalo matricos kompozitai [14][15]. Natūralaus pluošto kompozitų eksploatacinės savybės daugiausia priklauso nuo armatūros ilgio, išdėstymo, formos, turimos krypties ir pluošto bei matricos medžiagos sąsajos sujungimo. Pagal jų ilgio ir skersmens santykį pluoštai gali būti skirstomi į ištisinius pluoštus ir nepertraukiamus pluoštus. Pluoštu sustiprintos fazės išdėstymas paprastai skirstomas į austinę arba neaustinę [16]. Pagal armatūrą, jie būna įvairių formų, įskaitant laminatą, pluoštinį ir dalelių kompozitą[17]. Sutvirtinimai skirstomi į dvi kategorijas: natūralius ir sintetinius, atsižvelgiant į jų kilmę ir prieinamumą [18].

Naujausi tyrimai parodė, kad kompozitų hibridizacija turi įtakos medžiagų savybių raidai [19]. Daugelyje tyrimų buvo siekiama rasti naujų būdų, kaip derinti medžiagas, kad būtų pagerintos jų savybės;, pavyzdžiui, dviejų skirtingų natūralių pluoštų, dviejų atskirų natūralių ir sintetinių pluoštų arba dviejų skirtingų sintetinių pluoštų derinimas toje pačioje matricinėje medžiagoje[20]. Šių kompozitų mechaninės savybės buvo ištirtos tam, kad būtų užtikrintas geresnis tvirtumas, palyginti su vieno pluošto kompozitais. Keičiant pluošto dydį, užpildo tipą ir orientaciją, galima sukurti įvairių formų hibridinius kompozitus. Hibridizacija naudojant skirtingų tipų pluoštines medžiagas gali padėti įveikti vieno komponento trūkumus [21][22]. Hibridinis poveikis paprastai skiriasi pagal pluošto mechanines ir

chemines savybes; dėl to efektas sumažėja lyginant su kompozitais iš atskirų pluoštų [23]. Per pastaruosius kelis dešimtmečius buvo atlikta daugybė sintetinių ir augalinių pluoštų maišymo tyrimų [24–27]. Tačiau pastaruoju metu natūraliu pluoštu sustiprinti hibridiniai kompozitai sulaukė mažiau mokslinių tyrimų dėmesio, o dauguma tyrimų buvo skirti natūralių ir sintetinių pluoštų mišiniams. Anglies, stiklo ir aramido panaudojimas kartu su kanapėmis, linais, bananais ir ananasais yra geriausias tokių kompozitų pavyzdys [28–30].

Vieno pluošto kompozitai laikomi prastesniais už hibridiniu pluoštu sustiprintus kompozitus. Dėl puikių mechaninių savybių ir prieinamos kainos galima rasti papildomų pritaikymų [31]. Dėl šios priežasties dirbtinio pluošto kompozitai buvo pakeisti hibridiniais kompozitais, kurie pasižymi puikiomis savybėmis [32], ypač nešančiųjų ir kitų statybinių konstrukcijų srityse [33], [34]. Gaminiai, pagaminti iš natūralaus pluošto kompozitų, naudojami įvairioms reikmėms, įskaitant palatų lenteles,langus, automobilių durų plokštes, sporto įrangą, stalus, pakavimo lentynas ir lengvas prekes [35–37]. Dauguma natūralaus pluošto kompozitų yra trumpo pluošto kompozitai, turintys netaisyklingą armatūros ilgį ir orientacijos pasiskirstymą [38], [39]. Žinoma, kad nepertraukiamo pluošto kompozitai turi nedidelį tamprumo modulį ir stiprumą, palyginti su ištisiniu pluoštu armuotais kompozitais [40]. Visgi yra daugiau lankstumo pasirenkant gamybos būdus. Nepertraukiamo pluošto kompozitai gali būti pagaminti taikant įvairius plastiko apdirbimo būdus. Tai atveria supaprastintos masinės gamybos galimybes [41].

Kompozituose armatūra ir matrica išlaiko savo unikalias fizines ir chemines savybes. Tačiau derinamos jos sukuria išskirtinį savybių rinkinį, kurio negali pasiekti vienas iš komponentų [42]. Pagrindinis šių medžiagų tikslas yra pasiekti savybių pusiausvyrą derinant įvairias sudedamąsias dalis. Jungiamosios medžiagos, dangos ir užpildai yra papildomi komponentai, kurių gali būti kompozituose. Pluoštams naudojamos jungiamosios medžiagos ir dangos, siekiant pagerinti jų suderinamumą su matrica, skatinant geresnį drėkinimą ir sukibimą pluošto bei matricos sąsajoje [43]. Manoma, kad nanodalelių pavidalo užpildų pridėjimas prie kompozicinės medžiagos yra labai perspektyvus gerinant kompozitų mechanines ir fizines savybes [44].

Nepaisant puikių mechaninių savybių, natūralaus pluošto kompozitai turi tam tikrų trūkumų, tokių kaip prastos sukibimo savybės[45], plastiškumas[46], jautrumas drėgmei[47], vidiniai įtempiai[48] ir kt. Dėl šių trūkumų gali susilpnėti sudėtinės medžiagos stiprumas ir pablogėti jos mechaninės savybės. Siekiant pašalinti šiuos trūkumus, buvo bandoma naudoti įvairius natūraliu pluoštu sustiprintus hibridinius polimerinius kompozitus. Buvo atliktas armatūros medžiagų šarminis ir terminis apdorojimas, o siekiant suprasti gamybos klaidas sukurti įvairūs gamybos būdai. Hibridiniai kompozitai su įvairiomis svorio frakcijomis buvo pagaminti siekiant pagerinti medžiagos mechanines savybes ir pasiūlyti juos naudoti tvariose srityse, mikrofluidikoje, pavyzdžiui, vienkartiniams prietaisams, įvairiems mikroskysčių sistemų komponentams (pavyzdžiui, substratams) ir prietaisų pakuotėms gaminti.

Tikslas ir uždaviniai:

Tikslas:

Daktaro disertacijos tikslas – sukurti hibridinius pluoštu armuotus polimerinius kompozitus ir ištirti jų mechanines, morfologines bei drėkinimo charakteristikas tvariems pritaikymams.

Uždaviniai:

Pagrindiniam baigiamojo darbo tikslui pasiekti buvo sukurtas konkrečių uždavinių rinkinys:

1. Pagaminti hibridinius kompozitus sutvirtinant natūralius pluoštus epoksidine matricine medžiaga ir ištirti kaip hibridizacijos įterpimas paveikia kompozitų mechanines savybes.
2. Nustatyti cheminio apdorojimo poveikį natūraliu pluoštu armuotų hibridinių kompozitų mechaninėms savybėms ir palyginti jų veikimą su neapdorotais pluošto kompozitais.
3. Sukurti skaitmeninį metodą ir atlikti eksperimentinę analizę, siekiant ištirti įvairių matricinių medžiagų įtaką hibridinių polimerinių kompozitų savybėms.
4. Įvertinti nanodalelių įtaką hibridiniams pluošto kompozitams, skirtiems naudoti mikrofluidikos srityje..

Tyrimo metodai:

Kaip minėta anksčiau, šios disertacijos tikslas yra sukurti ir apibūdinti hibridinių pluoštu armuotų kompozitų mechanines savybes tvariam pritaikymui, atsižvelgiant į matricos medžiagos įtaką, cheminį apdorojimą, nanodalelių įterpimą ir kintamą pluošto masės frakciją. Kompozitai buvo sukurti taikant hibridizacijos koncepciją, naudojant *Caryota*, sizalio, bananų, ananasų, kanapių, linų, epoksidinio stiklo ir bazalto pluoštus kaip sutvirtinimus, o epoksidines dervas, ekopoksidą ir poliesterio dervas kaip matricines medžiagas. Grafitas buvo naudojamas kaip užpildas nanodalelėms įterpti. Įvairių svorio frakcijų kompozitai buvo gaminami taikant rankinį klojimą, po to buvo taikomas formavimas suspaudžiant, vakuuminio dervos perdavimo technika ir laisvojo laminavimo vakuuminiu būdu metodai. Cheminiam apdorojimui buvo naudojami NaOH ir HCl tirpalai. Eksperimentiniais tyrimais įvertintos mechaninės savybės, įskaitant tempimo bandymus, lenkimo bandymus ir tarpsluoksninius šlyties bandymus, atliktus su *Tinus Olsen H10KT*, smūgio bandymus, atliktus su *Coesfeld Magnus* smūgio testeriu, ir kietumo bandymus, atliktus su *Vickers* kietumo testeriu. Drėgnumo analizė atlikta taikant kontaktinio kampo matavimo analizę. Be to, atlikti morfologiniai tyrimai taikant *Hitachi S-3400N* SEM analizę ir *Bruker Quad 5040* SEM-EDX analizę. Teoriniai tyrimai atlikti taikant modifikuotą Halpin-Tsai modelį.

Tyrimo darbo naujumas

Tai yra pagrindinės mokslinio darbo, atlikto su natūraliu pluoštu sustiprintais hibridiniais kompozitais, išvados ir rezultatai:

1. Natūralaus pluošto hibridinių kompozitų mechaninės savybės yra kelis kartus geresnės nei atskirų pluoštinių kompozitų.
2. Eksperimentinė analizė rodo, kad natūraliu pluoštu armuotų hibridinių kompozitų mechaninės savybės pirmiausia priklauso nuo armatūros medžiagų tūrio dalies.
3. Natūralaus pluošto ir epoksidinių kompozitų tarpsluoksnio šlyties stipris yra du kartus stipresnis nei kompozitų su ekopoksidine matricine medžiaga, nustatyta tiek teoriniais, tiek eksperimentiniais metodais.
4. Natūralių pluoštų apdorojimas šarminiu būdu pastebimai gerina švarą, didina paviršiaus šiurkštumą, pluošto matricos sukibimą ir leidžia juos geriau suderinti su polimerinėmis matricomis – visa tai gali turėti įtakos bendram kompozicinių medžiagų veikimui.
5. Grafito nanodalelių pridėjimas rodo, kad hibridinių kompozitų poringumas sumažėja ir padidėja atsparumas drėgmei.
6. Natūralaus pluošto hibridinių kompozitų mechaninės savybės yra panašios arba netgi pranašesnės už sintetinio pluošto kompozitų savybes. Šią išvadą patvirtina morfologiniai tyrimai ir drėkinamumo analizė, kuri dar geriau parodo puikias sąsajos sukibimo savybes.

Ginamieji teiginiai

1. Hibridinių kompozitų morfologines savybes galima pastebimai pagerinti gamybos proceso metu įdiegus vakuuminio laisvojo laminavimo ir dervos pernešimo modelius.
2. Apdorojimo šarminiu būdu taikymas natūralių pluoštų paviršiaus savybėms pagerinti yra naujas ir veiksmingas būdas pagerinti natūralaus pluošto kompozitų mechanines savybes.
3. Hibridinių kompozitų tarpsluoksnių šlyties stiprio ir šlyties energijos tyrimas yra naujas metodas armatūros ir matricos sąsajų sujungimui įvertinti. Jis gali suteikti informacijos apie kompozicinės medžiagos patvarumą ir ilgaamžiškumą.

Praktinė vertė

Praktinę šio tiriamojo darbo vertę, nors ir glaudžiai susijusią su moksline naujove, galima apibendrinti taip.

1. Natūralaus pluošto hibridinių kompozitų tyrimai padeda kurti ekologiškus sprendimus, kurie mažina priklausomybę nuo neatsinaujinančių išteklių ir poveikį aplinkai.
2. Medžiagų parinkimo ir gamybos metodų optimizavimas leido pagerinti kompozicinės medžiagos mechanines charakteristikas. Tai gali padėti sukurti tvirtesnes ir patvaresnes kompozicines medžiagas, skirtas naudoti lengvose konstrukcijose.
3. Šarminio apdorojimo ir grafito nanodalelių įterpimo tyrime pabrėžiami būdai, kaip pagerinti natūralaus pluošto kompozitų veikimą. Šios žinios gali būti

pritaikytos praktiniams gamybos procesams, siekiant pagerinti kompozitinių medžiagų kokybę ir savybes.

4. Natūralaus pluošto hibridinių kompozitų kūrimas gali būti taikomas mikrofluidiniams įrenginiams, tokiems kaip kanalai, lustų substratai, kameros, maišytuvai ir vožtuvai, gaminti.

Tyrimo aprobavimas

Šios publikacijos ir tyrimų rezultatų pristatymai parodo doktorantūros studijų metu atliktų tyrimų reikšmę ir poveikį mokslo pasauliui.

Disertacijos rezultatai paskelbti 7 moksliniuose straipsniuose, iš kurių 6 publikuoti tarptautiniuose žurnaluose, įrašytuose į *ISI Web of Science* duomenų bazę ir turi citavimo indeksą. Pažymėtina, kad penki iš septynių paskelbtų straipsnių buvo paskelbti „1 kvartilio“ žurnaluose, o vienas buvo paskelbtas „2 kvartilio“ žurnale. Vienas pranešimas buvo paskelbtas kaip konferencijos tęsinys tarptautinėje konferencijoje.

Rezultatai taip pat pristatyti 4 tarptautinėse konferencijose ir 5 tarptautiniuose moksliniuose seminaruose.

LEIDINIŲ APŽVALGA

2.1 Natūralaus pluošto *Caryota* ir sizalio epoksidinių hibridinių kompozitų mechaninių savybių kūrimas ir analizė (mokslinė publikacija nr. [A2])

Straipsnyje aprašoma hibridinių kompozitų, naudojant anglies bei sizalio pluoštus kaip sutvirtinimus ir epoksidinę dervą kaip matricinę medžiagą, kūrimą bei apibūdinimą. Kompozitai buvo paruošti rankinio klojimo metodu, įvertintos jų mechaninės ir drėgmės sugėrimo savybės. Buvo atlikti tempimo, lenkimo ir smūgio bandymai, kurių rezultatai parodė, kad hibridiniai kompozitai pasižymi geresnėmis mechaninėmis savybėmis, palyginti su grynais kompozitais. Pastebėta, kad hibridinių kompozitų tempiamasis stipris ir modulis buvo atitinkamai iki 38,2 MPa ir 6,44 GPa, o grynųjų kompozitų – atitinkamai iki 25,7 MPa ir 5,66 GPa. Hibridiniai kompozitai taip pat pasižymėjo didesne lenkimo jėga ir smūgio energija, palyginti su grynaisiais kompozitais. Drėgmės sugerties bandymai parodė, kad visi kompozitai laikui bėgant reagavo į drėgmę, o grynųjų kompozitų absorbcijos greitis buvo didesnis nei hibridinių kompozitų. SEM analizė atskleidė, kad anglies masės procentas buvo didesnis paviršiuje nei lūžio padėtyje, o deguonies masės procentas buvo didesnis lūžio padėtyje nei paviršiuje, tai rodo, kad matricos medžiagos koncentracija buvo didesnė paviršiaus padėtyje, bet štai skaidulų procentas buvo didesnis lūžio (vidinėje) padėtyje. Tyrime pabrėžiamas hibridinių kompozitų panaudojimo įvairiose pramonės srityse potencialas ir siūlomi tolesni tyrimai, siekiant optimizuoti kompozitų mechanines savybes, naudojant įvairius natūralius pluoštus ir taikant įvairius gamybos būdus. Rekomenduota toliau tirti hibridinius kompozitus su skirtingais natūraliais pluoštais ir gamybos būdus, siekiant pagerinti jų mechanines savybes. Apskritai šiame straipsnyje pabrėžiamas natūralaus pluošto hibridinių kompozitų, kaip tvarios ir aplinkai nekenksmingos alternatyvos tradicinėms kompozicinėms medžiagoms,

potencialas ir natūralus pluoštas gali būti naudojamas kaip mikrofluidinių prietaisų pakavimo medžiaga. Dėl šios medžiagos biologinio suderinamumo ir mažos kainos ji yra tinkama vienkartiniams prietaisams.

2.2 Natūraliu pluoštu sustiprintų epoksidinių hibridinių kompozitų mechaninių ir drėkinamumo savybių analizė (mokslinė publikacija nr. [A3])

Hibridiniai kompozitai buvo pagaminti naudojant kanapių, linų, bananų bei ananasų pluoštus kaip sutvirtinimus ir epoksidinę dervą kaip matricos medžiagą. Kompozitai buvo pagaminti remiantis hibridizacijos koncepcija, todėl gauti du hibridinių kompozitų rinkiniai (H/F ir B/P) su penkiomis skirtingomis svorio frakcijomis. Mechaninės ir drėkinimo savybės buvo išbandytos ir palygintos tarp dviejų kompozitų rinkinių. Mechaninės savybės buvo įvertintos išmatuojant lenkimo ir tarpsluoksnių šlyties stiprumo savybes. Kanapių/linų ir bananų /ananasų pluošto kompozitų lenkimo savybės svyravo atitinkamai nuo 60 MPa iki 88 MPa ir nuo 38 MPa iki 69 MPa. 25H/15F hibridinis kompozitas pasižymi didžiausiu lenkimo stipriu – 87,57 MPa, o 40B/0P kompozitas rodo mažiausią stiprumą esant 39,36 MPa. Nustatyta, kad abiem atvejais grynųjų kompozitų tarpsluoksninės šlyties stiprumo savybės yra pranašesnės už hibridinių kompozitų. Drėkinamumo analizė parodė, kad visų kompozitų kontaktinis kampas buvo mažesnis nei 90˚, o tai reiškia hidrofilines paviršiaus savybes.

2.3 Kanapių ir linų pluoštu sustiprintų *EcoPoxy* matricinių biokompozitų tyrimas: morfologinės, mechaninės ir hidrofilinės savybės (mokslinė publikacija nr. [A4])

Šiame straipsnyje ištirtos kanapių ir linų lino natūralaus pluošto hibridinių kompozitų mechaninės ir morfologinės savybės, esant ekopoksidinei dervai kartu su kietikliu. Kompozitai buvo sukurti naudojant natūralius pluoštus kaip sutvirtinimus ir ekopoksidinę dervą kaip matricinę medžiagą. Siekiant pagerinti pluošto paviršiaus savybes ir sukibimo savybes, buvo atliktas šarminis apdorojimas NaOH tirpalu, o rezultatai buvo stebimi atliekant mikroskopinę analizę ir FTIR analizę. Cheminio apdorojimo įtaka natūraliam pluoštui buvo stebima pašalinant paviršiaus įtrūkimus, įlinkimus ir paviršiaus nelygumą, esantį natūraliuose pluoštuose. Vėliau jis pagerino pluošto sukibimo savybes. Sąlyčio kampas buvo pastebėtas mažesnis nei 90˚ tiek įprasto, tiek nelygaus pluošto atvejais. Tačiau nešvarumų atveju hibridinio kompozito kontaktinis kampas buvo artimas 90˚, o tai reiškia, kad kompozitai linkę parodyti hidrofobines paviršiaus savybes. Remiantis mechaninėmis savybėmis, buvo pastebėta, kad kanapių/linų natūralaus pluošto hibridiniai biokompozitai pasižymi geresnėmis mechaninėmis savybėmis, palyginti su hibridinio pluošto kompozitais. Dėl gerų paviršiaus savybių ir mechaninių savybių šie kompozitai yra tinkamas pasirinkimas pakuoti medžiagas mikrofluidiniuose įrenginiuose. Be to, dėl biologinio suderinamumo ir mažos medžiagos kainos ji yra tinkama vienkartiniams mikrofluidiniams prietaisams.

2.4 Natūraliu pluoštu sustiprintų hibridinių polimerų kompozitų mechaninių savybių ir poveikio matricinei medžiagai skaitmeninė ir eksperimentinė analizė (mokslinė publikacija nr. [A5])

Šiame moksliniame straipsnyje buvo tiriama matricinės medžiagos įtaka natūralaus pluošto hibridinių kompozitų mechaninėms savybėms. Kompozitai buvo pagaminti naudojant kanapių ir linų pluoštus kaip sutvirtinimus ir epoksidinę dervą bei ekopoksidinę dervą su kietikliu kaip matricines medžiagas. Buvo išnagrinėtos ir palygintos dviejų tipų matricinių medžiagų mechaninės kompozitų savybės, tokios kaip atsparumas lenkimui, tarpsluoksninis šlyties stiprumas ir drėgnumas. Halpin-Tsai metodas buvo taikomas teorinei kompozitų analizei. Rezultatai parodė, kad epoksidiniai kompozitai pasižymi geresnėmis mechaninėmis savybėmis, palyginti su hibridiniais kompozitais ekopoksidinės matricos pagrindu. Be to, abiejų tipų hibridiniai kompozitai pasižymėjo geresnėmis mechaninėmis savybėmis nei grynieji kompozitai. SEM-EDX analizė atskleidė, kad anglies masės procentas buvo didesnis paviršiuje virš lūžio padėties abiem atvejais, o deguonies masės procentas buvo didesnis lūžio padėtyje. Deguonies procentas buvo didesnis ekopoksidinės matricos medžiagos atveju dėl jos biologinio aktyvumo. Teoriniam vertinimui taikytas modifikuotas Halpin-Tsai metodas, tačiau nustatyta, kad šio metodo klaidų procentas buvo didesnis lyginant su skaitiniais metodais. Todėl Halpin-Tsai metodas gali būti netinkamas taikymui natūralaus pluošto hibridinių kompozitų atveju..

2.5 Užpildo įtaka bazalto/E-stiklo audinio – mikrofluidikai skirtų armatūros kompozitų drėgnumui ir mechaninėms savybėms (mokslinė publikacija nr. [A6])

Šioje mokslinėje publikacijoje buvo ištirta užpildo dalelių įtaka hibridinių kompozitų mechaninėms savybėms. Bazalto/ E-stiklo pluošto hibridinių kompozitų gamybai buvo sukurtas naujas gamybos technikos metodas. Grafito miltelių nanodalelės buvo naudojamos kaip kompozitų užpildas. Šios nanodalelės paruoštos mechaninio malimo būdu ir jų dydis svyravo nuo 50 nanometrų iki 80 nanometrų. Šios nanodalelės buvo įsigytos iš *Sigma-Aldrich Chemicals Private Limited*, esančios Bangalore, Indijoje. Hibridinių kompozitų su 5%, 10% ir 15% masės grafito užpildo dalelių smūgio energijos lygiai buvo atitinkamai 3,8 KJ/m^2, 4,0 KJ/m^2 ir 3,9 KJ/m^2, didesni nei grafito užpildo dalelių. grynojo bazalto ir E-stiklo kompozitų atveju. SEM analizė parodė, kad užpildo dalelių įterpimas pagerino pluoštų ir matricos sąsają, todėl pagerėjo mechaninės savybės. Tačiau užpildo dalelių masės koncentracijai viršijus 10%, hibridinių kompozitų mechaninės savybės pablogėjo dėl aglomeracijos ir prastos užpildo dalelių sklaidos. Rezultatai rodo, kad pridėjus nedidelį užpildo dalelių kiekį galima žymiai pagerinti hibridinių kompozitų mechanines savybes, tačiau reikia atidžiai kontroliuoti užpildo dalelių masės koncentraciją, kad būtų išvengta neigiamo poveikio kompozicinės medžiagos savybėms. Apskritai šio tyrimo išvados turi svarbių pasekmių kuriant puikios kokybės kompozicines medžiagas, pasižyminčias patobulintomis mechaninėmis savybėmis, ir šias medžiagas galima naudoti kaip mikrofluidinių lustų substratą. Dėl mažos medžiagos kainos ir gamybos paprastumo tai yra patraukli galimybė gaminti mikrofluidiniams lustams.

IŠVADOS

Iš penkių mokslinių straipsnių apžvalgos galima padaryti šias išvadas kaip suformuluotus tikslus ir uždavinius, remiantis keturiais tikslais, kurie buvo nustatyti tyrimo pradžioje:

1. Hibridiniai kompozitai iš natūralių pluoštų ir epoksidinių matricų medžiagų, buvo pagaminti taikant įvairius gamybos būdus. *Caryota* ir sizalio pluošto hibridiniai kompozitai pasižymėjo geresnėmis mechaninėmis savybėmis, palyginti su vieno pluošto kompozitais, kurių tempiamasis stipris buvo 38 MPa, stipris lenkiant 89,16 MPa ir lenkimo modulis 3,40 GPa. Kanapių ir linų pluošto kompozitų tarpsluoksninis šlyties stipris svyravo nuo 6,30 MPa iki 13,50 MPa. 25H/15F hibridinių biokompozitų didžiausias lenkimo stipris ir modulis buvo 87,50 MPa ir 3,45 GPa,
2. Armatūros medžiagos šarminis apdorojimas 5 % NaOH tirpalu parodė neigiamą poveikį mechaninėms savybėms. Morfologinis tyrimas atskleidė, kad chemiškai apdorotose pluoštinėse medžiagose pašalinami paviršiaus įtrūkimai, sulenktos juostos ir pagerėjo paviršiaus šiurkštumas, o tai galiausiai paveikė mechanines savybes. FTIR analizė parodė funkcinių grupių pokyčius ir hidroksilo grupių (-OH) intensyvumą. (-OH) hidroksilo smailių intensyvumas, pastebėtas prieš apdorojimą, buvo apie 2700-2900 cm^{-1}, o po apdorojimo apie 2200-2500 cm^{-1}. Be to, lignino ir hemiceliuliozės smailės sumažėjo atitinkamai ties 1130 cm^{-1} ir 960 cm^{-1}. Šie cheminės struktūros pokyčiai parodė funkcinių grupių transformacijas, priemaišų pašalinimą ir pagerėjusį celiuliozės kristališkumą, taip pagerinant pluošto paviršiaus savybes.
3. Pastebėta, kad epoksidinės matricos pagrindu pagaminti hibridiniai kompozitai pasižymėjo du kartus geresnėmis mechaninėmis savybėmis, palyginti su hibridiniais kompozitais ekopoksimatricos pagrindu. Epoksidinių kompozitų didžiausias lenkimo ir šlyties stipris buvo atitinkamai 78 MPa ir 13 MPa, o ekopoksidinių kompozitų – atitinkamai 30 MPa ir 10 MPa. Kompozitų tamprumo savybės buvo nustatytos atlikus teorinę ir skaitinę analizę, pastebėta, kad modifikuotas Halpin-Tsai metodas turėjo didesnes paklaidas.
4. Bazalto ir E-stiklo pluošto pagrindu pagaminti epoksidiniai hibridiniai kompozitai su 10 % grafito užpildo pasižymėjo geresnėmis mechaninėmis savybėmis, palyginti su atskirais bazalto ir E-stiklo pluošto kompozitais. B/E/G15 kompozitas pasižymėjo didžiausiu lenkimo stipriu – 43,81 MPa, lenkimo moduliu – 3,57 GPa, o epoksidinės dervos pagrindu pagaminti B/E/10G kompozitai pasižymėjo geriausiomis tempimo savybėmis. Akytumas hibridiniuose kompozituose su užpildu buvo mažesnis nei tuose, kuriuose nėra užpildo.

Remiantis išvadomis, natūralaus pluošto hibridiniai kompozitai gali pakeisti dirbtinio pluošto kompozitus tvariose srityse, įskaitant lustų substratus, pakavimo įrenginius, mikrofluidinius kanalus ir vožtuvų medžiagas.

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LIST OF PUBLICATIONS

Publications indexed in the Web of Science with Impact Factor (JCR SCIE) International (foreign) publishers

A1. [S1; CH; OA] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Vilkauskas, Andrius; Janusas, Giedrius. Review of hybrid fiber based composites with nano particles–material properties and applications // Polymers. Basel : MDPI. ISSN 2073-4360. 2020, vol. 12, iss. 9, art. no. 2088, p. 1-30. DOI: 10.3390/polym12092088. [Science Citation Index Expanded (Web of Science); Scopus; Academic Search Complete] [IF: 4,329; AIF: 4,658; IF/AIF: 0,929; Q1 (2020, InCites JCR SCIE)] [FOR: T 009] [Input: 0,250].

A2. [S1; CH; OA] **Atmakuri, Ayyappa;** Palevicius, Arvydas; Kolli, Lalitnarayan; Vilkauskas, Andrius; Janusas, Giedrius. Development and analysis of mechanical properties of Caryota and sisal natural fibers reinforced epoxy hybrid composites // Polymers. Basel : MDPI. ISSN 2073-4360. 2021, vol. 13, iss. 6, art. no. 864, p. 1-19. DOI: 10.3390/polym13060864. [Science Citation Index Expanded (Web of Science); Scopus; MEDLINE] [IF: 4,967; AIF: 5,162; IF/AIF: 0,962; Q1 (2021, InCites JCR SCIE)] [FOR: T 009] [Input: 0,200].

A3. [S1; CH; OA] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Siddabathula, Madhusudan; Vilkauskas, Andrius; Janusas, Giedrius. Analysis of mechanical and wettability properties of natural fiber-reinforced epoxy hybrid composites // Polymers. Basel : MDPI. ISSN 2073-4360. 2020, vol. 12, iss. 12, art. no. 2827, p. 1-15. DOI: 10.3390/polym12122827. [Science Citation Index Expanded (Web of Science); Scopus; Academic Search Complete] [IF: 4,329; AIF: 4,658; IF/AIF: 0,929; Q1 (2020, InCites JCR SCIE)] [FOR: T 009] [Input: 0,200].

A4. [S1; CH; OA] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Janusas, Giedrius; Eimontas, Justas. Investigation of hemp and flax fiber-reinforced EcoPoxy matrix biocomposites: morphological, mechanical, and hydrophilic properties // Polymers. Basel : MDPI. ISSN 2073-4360. 2022, vol. 14, iss. 21, art. no. 4530, p. 1-21. DOI: 10.3390/polym14214530. [Science Citation Index Expanded (Web of Science); Scopus; MEDLINE] [IF: 4,967; AIF: 5,162; IF/AIF: 0,962; Q1 (2021, InCites JCR SCIE)] [FOR: T 006, T 009] [Input: 0,250].

A5. [S1; CH; OA] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Vilkauskas, Andrius; Janusas, Giedrius. Numerical and experimental analysis of mechanical properties of natural-fiber-reinforced hybrid polymer composites and the effect on matrix material // Polymers. Basel : MDPI. ISSN 2073-4360. 2022, vol. 14, iss. 13, art. no. 2612, p. 1- 18. DOI: 10.3390/polym14132612. [Science Citation Index Expanded (Web of Science); Scopus; MEDLINE] [IF: 4,967; AIF: 5,162; IF/AIF: 0,962; Q1 (2021, InCites JCR SCIE)] [FOR: T 009] [Input: 0,250].

A6. [S1; CH; OA] **Atmakuri, Ayyappa**; Kolli, Lalitnarayan; Palevicius, Arvydas; Urbaite, Sigita; Janusas, Giedrius. Influence of filler materials on wettability

and mechanical properties of Basalt/E-glass woven fabric–reinforce composites for microfluidics // Micromachines. Basel : MDPI. ISSN 2072-666X. 2022, vol. 13, iss. 11, art. no. 1875, p. 1-14. DOI: 10.3390/mi13111875. [Science Citation Index Expanded (Web of Science); Scopus; MEDLINE] [IF: 3,523; AIF: 5,908; IF/AIF: 0,596; Q2 (2021, In Cites JCR SCIE)] [FOR: T 009] [Input: 0,200].

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B1. [P1a; US] **Atmakuri, Ayyappa**; Janušas, Giedrius; Siddabathula, Madhusudhan; Palevičius, Arvydas. Wettability and moisture analysis on natural fiber reinforced epoxy resin hybrid composites // 15th international conference of mechatronic systems and materials MSM'2020: the book of selected, peer reviewed papers, July 1-3, 2020, Bialystok, Poland / edited by: Z. Kulesza, A. Mystkowski, J. Pauk, A. Idzkowski. Piscataway, NJ : IEEE, 2020. ISBN 9781728169576. eISBN 9781728169569. p. 1-6. DOI: 10.1109/MSM49833.2020.9202320. [Conference Proceedings Citation Index - Science (Web of Science); Scopus; INSPEC] [FOR: T 009] [Input: 0,250].

Presentation of research results at the international conferences

C1. [T3; LT; OA] **Atmakuri, Ayyappa**; Siddabathula, Madhusudan. Investigation of mechanical properties of banana and pineapple natural fibre hybrid composites // IRWBM 2019: International research workshop in biomechanical microsystems 2019, October 23, 2019, Kaunas University of Technology, Lithuania: program and abstracts. Kaunas: [KTU]. 2019, p. [13]. [FOR: T 009].

C2. [T3; PL; OA] **Atmakuri, Ayyappa**; Janusas, Giedrius; Siddabathula, Madhusudan; Palevicius, Arvydas. Wettability and moisture analysis on natural fiber reinforced epoxy resin hybrid composites // 15th International Conference Mechatronic Systems and Materials (MSM 2020), 1-3 July 2020, Białystok, Poland: book of abstracts. Bialystok: Bialystok University of Technology. 2020, p. 19. [FOR: T 009].

C3. [T2; LT] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Janusas, Giedrius. Investigation of mechanical properties of okra-banana fiber reinforced polyester composites for microfluidic applications // Mechanika 2021: proceedings of the 25th international scientific conference, May 21, 2021 Kaunas University of Technology, Lithuania. Kaunas: Technologija. ISSN 2783-5677. 2021, p. 6-7. [FOR: T 009].

C4. [T2; LT] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Janusas, Giedrius. Evaluation of wettability and water absorption properties of natural fiber reinforced polyester hybrid composites for microfluidic applications // Advanced materials and technologies: book of abstracts of 23rd international conference-school, 23-27 August 2021, Palanga, Lithuania. Kaunas: Kaunas university of technology. ISSN 2669-1930. 2021, A-P125, p. 158. [FOR: T 009].

C5. [T3; LT] Madhusudan, Siddabathula; **Atmakuri, Ayyappa**. Estimation of hygric strain – an experimental approach // IRWBM 2021: 2nd international research workshop in biomechanical microsystems 2021: October 22, 2021, Kaunas University of Technology, Lithuania: program and abstracts. Kaunas: Kaunas University of Technology. 2021, p. 10. [FOR: T 009].

C6. [T3; LT] **Atmakuri, Ayyappa**. Development and characterization of bio-based fiber reinforced eco-friendly composite materials // IRWBM 2021: 2nd international research workshop in biomechanical microsystems 2021: October 22, 2021, Kaunas University of Technology, Lithuania: program and abstracts. Kaunas: Kaunas University of Technology. 2021, p. 14. [FOR: T 009].

C7. [T2; LT] **Atmakuri, Ayyappa**; Palevicius, Arvydas; Janusas, Giedrius. Damage analysis on natural fiber reinforced epoxy matrix hybrid composites: bolted, adhesive joints // Mechanika 2022: proceedings of the 26th international scientific conference, 20 May 2022, Kaunas university of technology, Lithuania. Kaunas: Technologija. ISSN 2783-5677. 2022, p. 7-8. [FOR: T 009].

C8. [T3; LT] **Atmakuri, Ayyappa**. Study of mechanical performance of nano-graphite fillers adapted epoxy hybrid composites // IRWBM 2022: 3rd international research workshop in biomechanical microsystems 2022: October 21, 2022, Kaunas university of technology, Lithuania: program and abstracts. Kaunas: Kaunas university of technology. 2022, p. 10. [FOR: T 009].

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Review

Review of Hybrid Fiber Based Composites with Nano Particles—Material Properties and Applications

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Abstract: The present review article provides an overview of the properties of various natural and synthetic fibers for the fabrication of pure natural composites and the combination of both natural/synthetic fibers-based hybrid composites, bio-based resins, various fabrication techniques, chemical and mechanical properties of fibers, the effect of chemical treatment and the influence of nanoparticles on the composite materials. Natural fibers are becoming more popular and attractive to researchers, with satisfactory results, due to their availability, ease of fabrication, cost-effectiveness, biodegradable nature and being environmentally friendly. Hybrid composites made up of two different natural fibers under the same matrix material are more popular than a combination of natural and synthetic fibers. Recent studies relevant to natural fiber hybrid composites have stated that, due to their biodegradability and the strength of individual fibers causing an impact on mechanical properties, flame retardancy and moisture absorption, natural fibers need an additional treatment like chemical treatment for the fibers to overcome those drawbacks and to enhance their better properties. The result of chemical treatment on composite material properties such as thermal, mechanical and moisture properties was studied. Researchers found that the positive influence on overall strength by placing the filler materials (nanoparticles) in the composite materials. Hybrid composites are one of the fields in polymer science that are attracting consideration for various lightweight applications in a wide range of industries such as automobile, construction, shipping, aviation, sports equipment, electronics, hardware and biomedical sectors.

Keywords: natural and synthetic fibers; hybrid composites; thermosets and thermoplastic resins; fabrication techniques; chemical treatment; nanoparticles; applications

1. Introduction

In recent years, many researchers have been showing an interest in the utilization of natural fibers for the preparation of hybrid composites because of its abundance, low density, recyclability and great strength [1,2], Natural fibers received a great deal of consideration as a potential elective alternative for synthetic fibers [3] as a support of different reinforcement for innovative applications due to their properties, for example, their availability in nature, low cost and they are environmentally friendly. Their resources do not emit any harm full gasses and absorb carbon dioxide, which reduces environmental pollution [4–6].

One of the primary drawbacks of using natural fibers as reinforcement in composites is the hydrophilic in nature, which makes them inconsistent with hydrophobic matrices. For refining the inconsistency, the surface treatment of natural fiber is required to advance the global properties of the composite [7,8]. Apart from this, natural fibers are readily accessible to the flame, to overcome this drawback, various fire retardants have been added to them to advance its fire resistance.

Surface modifications by using various chemical compounds tend to increase their mechanical and physical properties. And also, structure modification of the composite materials leads to extra stability [9,10].

Over the previous years, there has been an ever-expanding enthusiasm to fuse at least two fillers into a typical matrix. There has been a consistent increment in the quantity of distributed works that are identified with hybrid composites from the past ten years. in view of hybrid composites (between 2010 to 2020) found in the Web of Science and Scopus via looking for the words 'hybrid composites' with 26,450 research yields. The development came about because of one goal, to progress the properties of the subsequent composite material to accomplish the wanted properties and improved execution of such composites.

Figure 1 shows the total research outcomes on hybrid composites in recent years and the input was from the Web of Science. Most of the research articles are from China, followed by India. The hybridization process intersects the most research articles, for example; material multi-disciplinary, chemical industry, polymeric science, nanoscience, nanotechnology, material science engineering, electrochemistry, mechanics and mechanical engineering so on.

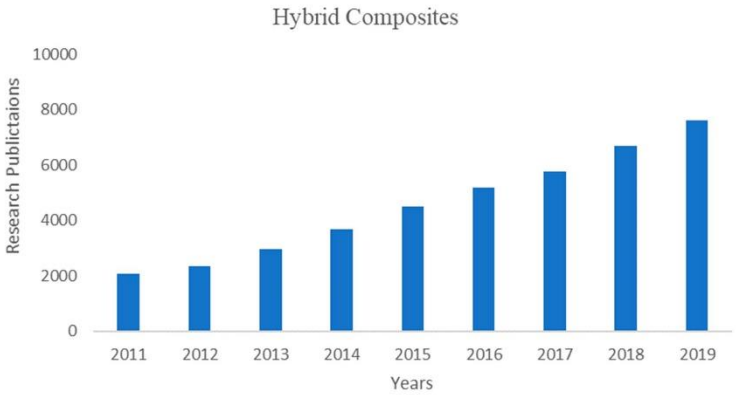


Figure 1. Recent publications on hybrid composites.

Figure 2 shows the total research publications on natural fiber hybrid composites. The input was from the Web of Science by searching for "natural fibers and hybrid composites." There is a total of 1460 research yields, with most of the researchers from material science and polymeric science engineering. The process of the hybridization of natural fiber composites started in early 2008 and then the interest slightly increased. The main aim is to improve the composite properties by adding two or three different filler materials under the same matrix material. Many researchers proved that the orientation of filler material also plays a key role in the composite [11–13].

Hybrid Composites are the materials that are made up of twofold or more constituent materials, namely matrix and reinforcement [14,15]. A matrix is a coupling material which binds the reinforcement. Reinforcements are the materials which are embedded in the matrix. These give the additional strength to the composite. The significance of the composites is to provide the desired physical, chemical and mechanical properties in an improved manner, which differs from the individual constituents [16]. Based on the matrix, composites are categorized as polymer, ceramic and polymer. Depending upon the reinforcement these are varied as laminate, fibrous and particulate composites. Depending upon the type of availability, reinforcements are classified as either natural or synthetic [15,17].

If the compound material is metal fiber then it is called a metal matrix composite, if it is a polymer matrix then it is called a polymer matrix material [17,18]. The fiber reinforced polymers (FRP) consist of a polymer matrix and high-quality fibers joined with an unmistakable crossing point among them. In this shape, the two matrices and fibers hold their chemical and physical properties.

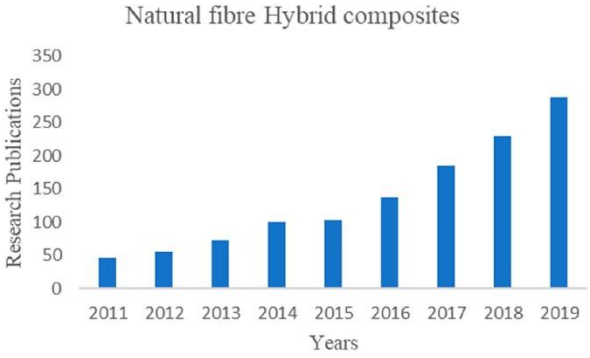


Figure 2. Recent publications on natural fiber hybrid composites.

2. Natural Hybrid Fiber Composites

Natural hybrid composite materials contained fibers of great quality and a modulus introduced in a matrix material. Along these lines, every one of the individual materials (fiber and matrix) holds its chemical and physical properties and they give a combination of properties which would not be conceivable if they were utilized alone. Here, the fibers act as the central load-carrying agents and the matrix material encompasses them in position alongside, acting as a load carrying part between them, protecting the fibers from the natural harms of raised temperature and humidity. Accordingly, it could be properly said that the fiber gives reinforcements to the matrix; thus, the name Fiber Reinforced Composite Materials (FRCM).

Reinforced hybrid materials are made by joining at least two distinct varieties of fibers in a distinctive matrix material. Hybridization of two sorts of filaments with particular lengths and widths offers a couple of central focuses over the use of both of the strands alone in a lone polymer lattice. Most studies are on the hybridization of regular filaments with glass strands to upgrade the properties. They have average calorific regard and cause little stress, similar to prosperity, wellbeing and protecting Also, they show amazing mechanical properties, have a low thickness and are modest [7,14,19–21]. The benefits of composite materials by confessional materials, such as greater rigidity and durability, make them more flexible to use. The availability and cost of these natural fibers attract researchers and also have promising results of composite fabrication for real-life applications. Composite materials are comprised of at least two materials with physically detachable phases.

2.1. Characterization of Composites

Composite materials classified on the basis of a matrix serve as a binder and a reinforcement, which acts as a filler material. Figure 3 shows the classification of composites.

2.2. Metal Matrix Composites (MMCs)

Metal matrix composites (MMCs) have numerous advantages over solid metals such as great strength, modulus and better-quality properties at raised temperatures. As a result of these qualities, metal matrix composites are getting looked at for a wide scope of applications. On account of their

high thickness, they are not greatly requested. The most commonly used metals in the MMCs are steel, tungsten, boric and, molybdenum [22,23].

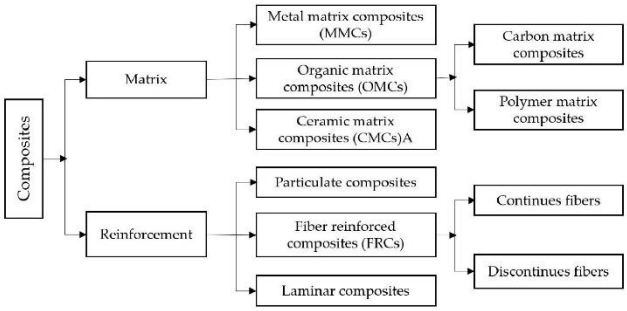


Figure 3. Classification of composites [22].

2.3. *Ceramic Matrix Composites (CMCs)*

Ceramic matrix composites (CMCs) and carbon-based composites are examples of inorganic (non-metallic) composite materials. The main aim of CMCs is to improve the toughness of the composite materials [24] and they are intended to overcome the problems of monolithic ceramics. The problem caused by the matrix materials is less than that of the failure of fiber materials, so it has a great impact on composite fabrication. To avoid the general drawbacks during the processing (early failure), there are several techniques implemented such as gas-phase and liquid-phase fabrication of CMCs.

2.4. *Organic Matrix Composites (OMCs)*

The organic matrix composites (OMCs) are broadly classified into polymer matrix composites and carbon-carbon matrix composites. Polymer composites are the most popular and commonly used matrix materials. The mechanical properties of polymeric composites are easily applicable to a wide range of structures because of their desirable mechanical properties, such as low mass, high stiffness and high-temperature resistance. When compared to the metal and ceramic composites, polymer composites have less strength and stiffness; to overcome this problem, composites are reinforced with other materials. The handling of polymer composites need not include high temperature and pressure. Likewise, the equipment required for the fabrication of composites is more straightforward. Therefore, polymer matrix composites quickly became famous and of interest for many researchers in terms of basic applications. Composites are utilized in light of the fact that the general properties of the composites are better than those of the specific segments.

Polymer composites are further classified as thermosets and thermoplastic composites. Thermoplastic materials are solids at room temperature; heating is required to soften or melt the material to place it into the mold and then it is cured to get the desired shape. These are recyclable. Thermosetting polymers are in both liquid and solid form and these are not recyclable. In thermoplastics, the atoms are bonded by covalent bonds whereas thermosetting polymers have cross-linked bonds, because these thermosetting materials are stronger and stiffer than thermoplastics. Thermoplastic materials include polyamides, polyoxymethylene (POM), polycarbonates (PC) and acrylics. Thermo setting materials include epoxies (EP), thermoset polyamides, amino plastics and polyester thermosets (TS) [24].

2.5. Fiber Reinforced Polymer Composites (FRCs)

Fiber Reinforced Polymer Composites (FRCs) are the most widely used composites and are composed of fiber and matrix. Fiber serves as a reinforcement and acts as a stiffness agent. The matrix material glues all fibers together and acts as a stress distributor among the fibers. Sometimes, the filler material is added to smooth the fabrication process. Depending upon the availability, reinforcements are classified into natural and synthetic fibers. Though there are many advantages to synthetic composites, there are some disadvantages. Synthetic fibers burn more quickly than natural fibers. Extra electrostatic charge is produced by rubbing them with natural fibers [23,25].

3. Classification of Fibers

3.1. Natural Fibers

Natural fibers are produced from animal (protein is the main component), plant (cellulose is the main component) and mineral based resources. Natural fibers have advanced physical and mechanical characteristics when compared to the other fibers. They are readily available in nature and also biodegradable [26,27]. Because of its lightweight, high strength, low cost and corrosion resistance properties it is drawing attention for use in industrial applications [28]. On the other side, natural fibers have few disadvantages, like high water absorption, high anisotropy, less homogeneity when compared to the synthetic fibers, development of stresses among the fibers in a composite [28]. The following Figure 4 shows the classification of natural fibers.

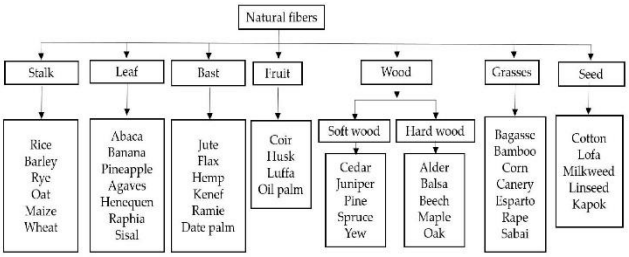


Figure 4. Classification of natural fibers [22–24].

The chemical composition plays a significant role in fiber materials and it depends on the fiber source. It varies from fiber to fiber and also within the family as shown in Table 1. Their performance is mainly dependent on the aspect/ratio and cellulose crystallinity of the fibers. Previous studies stated that these fibers are biodegradable because of their chemical constitutes. For example, UV degradation because of lignin and hemicellulose is accountable for thermal and biodegradation and also water absorption. The physical and mechanical properties of the natural fibers are shown in Table 2. Cellulose, hemicellulose and lignin are the main components in the natural fibers. Due to the nature of development conditions, the percentage of these components varies accordingly. To consider the behavior of these fibers, it is important to consider the structure of the components. Cellulose is one of the major building blocks of the natural fibers, it makes up around 50% of plants. The presence of cellulose in the plant fibers has an impact on moisture absorption. Hemicellulose is the second major component followed by cellulose and it is less remarkable as a filler material. Phenylpropane derivatives is comprised of lignin and it is a formless normal polymeric material that controls the transaction of fluid and also combines the hard cells in the plant. Lignin acts as a cementing material and it influences the structure, properties and morphology [29]. Some of the advantages and disadvantages of natural fibers are explained in Table 3.

Table 1. Natural fibers chemical composition [22,24–26].

Fiber Type	Cellulose (%)	Hemi-Cellulose (%)	Lignin (%)	Pectin (%)	Wax (%)	Ash (%)	Moisture (%)	Microfibrillar-Angle (°)
Abaca	56–63	20–25	7–9	-	3	-	-	20–25
Bamboo	26–43	30	1–31	-	10	-	9.16	-
Banana	63–83	-	5	-	11	-	10.71	11–12
Coir	36–43	41–45	0.15–0.25	3–4	-	-	11.36	30–49
Cotton	83–91	3	-	0.6	8–9	-	7.85–8.5	-
Flax	64–72	64–72	2–2.2	1.8–2.3	-	-	8–12	5–10
Hemp	70–74	0.9	3.7–5.7	0.8	1.2–6.2	0.8	6.2–12	2–6.2
Jute	61–72	18–22	12–13	0.2	0.5	0.5–2	12.5–13.7	8
Kenaf	45–57	8–13	22	0.6	0.8	2–5	6.2–12	2–6.2
Nettle	86	5.4	4.0	0.6	3.1	-	-	-
Rachis	43	-	26	-	-	-	-	28–37
Ramie	69–91	5–15	0.4–0.7	1.9	-	-	-	69–83
Rice husk	38–45	-	-	-	-	20	-	-
Sisal	78	10	8	-	2	1	11	-
Hardwood	38–49	17–23	23–30	-	-	-	-	-
Softwood	40–45	19–22	26–34	-	-	-	-	-

Table 2. Natural fibers origin, physical and mechanical properties [22,26–28].

Fiber Type	Origin	Diameter (μm)	Density g/cm^3	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation (%)	Specific Strength (S/ϱ)	Specific Modulus (E/ϱ)
Abaca	Leaf	-	1.5	430–813	31.1–33.6	2.9–10	267	8
Bagasse	Grass	-	0.89	350	22	5.5	-	-
Bamboo	Grass	240–330	0.6–1.25	290	11–17	-	454	32.6
Banana	Leaf	50–250	1.35	529–914	27–32	2.6–5.9	444	13.2
Coir	Fruit	-	1.15–1.25	131–220	4–6	15–40	146	3.3–5
Cotton	Seed	-	1.51	400	12	3–10	179–373	3.44–7.9
Flax	Stem	-	1.4	800–1500	60–80	1.2–1.6	535–1000	18.4–53
Hemp	Stem	-	1.48	550–900	70	1.6–4.0	372–608	47.3
Jute	Stem	40–350	1.31–1.48	393–800	13–26.5	1.16–1.8	269–548	6.85–20.6
Kenaf	Stem	70–250	1.4	284–930	21–60	1.6	641	36.55
Nettle	Bast	-	1.51	650	38	1.7	-	87–115
Pineapple	Leaf	-	1.44	413–1627	60–82	14.5	-	-
Ramie	Stem	50	1.5	500	44	2	147–675	29.3–85
Sisal	Leaf	50–300	1.3–1.4	390–450	12–41	2.3–2.5	366–441	6.5–15.2
Softwood	Wood	-	1500	1000	40	-	667	26.67
Hardwood	Wood	-	1200	950	37.9	-	-	-

Table 3. Merits and demerits of natural fibers.

Merits	Demerits
Lightweight	Flammable
Recyclable	Dimensional instability
Improved specific mechanical properties	High moisture absorption
Eco-friendly	Anisotropic behavior
Do not generate any harmful gasses during processing	Limited processing temperature (~200–230 °C)
Good thermal properties	Sensitive to UV
Good acoustic properties	Fugal attack and microbial
Low cost, availability	Low strength than synthetic fibers

3.2. Synthetic Fibers

There are various man-made or synthetic fibers that have been brought into recent research networks with respect to their remarkable properties. The mechanical properties of synthetic fibers are shown in Table 4. Among all synthetic fibers, glass and carbon fibers are the most used filler materials in the hybrid composites. These fibers are manufactured by using the polymerization concepts, which includes joining monomers to form a great chain. The merits and demerits of synthetic fibers explained in Table 5. Below Figure 5 shows the classification of synthetic fibers.

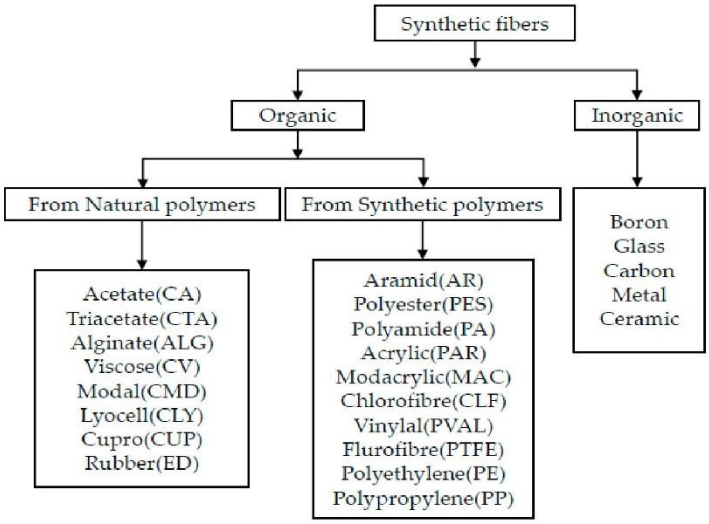


Figure 5. Classification of synthetic fibers [15].

Table 4. Mechanical properties of synthetic fibers [26–28].

Type of Fiber	Density (g/cm^3)	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation (%)
Aramid	1.44	3000	124	2.5
Carbon	1.4	400	230–240	1.4–1.8
E-Glass	2.55	2000–3500	63–67	2.5
S-Glass	2.5	4570	70–73	1.8–3.2
Silicon carbide	3.16	-	360–440	-

Table 5. Advantages and disadvantages of synthetic fibers.

Advantages	Disadvantages
Long lasting	Flammable
Readily pick-up to various dyes	Prone to heat damage
Stretchable	Melt easily
Waterproofing	Not eco-friendly
Non biodegradability	Cause for microplastic pollution
Moisture resistance	Not suitable for hot washing
Strain and wear resistance	Poor insulation capacity

4. Chemical Treatment of Natural Fibers

Natural fibers are not defect-free materials even though they have great qualities when compared to the other advanced fibers. They have strong polar bonds, which may affect the bonding capability in the polymer matrix [30]. To avoid this, some sort of chemical treatment is required to improve it. Chemical treatment is one that gives additional strength to the composite material to develop a relationship between fiber and matrix material during the curing process. It also improves the surface quality and decreases the amount of water absorbed (Moisture content) by the composite material [31]. Some of these treatment techniques are listed down in Table 6.

Table 6. Chemical treatment processes and its importance on hybrid fiber composites.

Chemical Treatment	Fibers	Advantages	Reference
Alkaline treatment	Flax Jute Sisal	Improves the surface roughness Better mechanical interlocking, and cellulose content.	[32–34]
Silane treatment	Glass	Prevent swelling of composite Improve thermal stability.	[35,36]
Acetylation treatment	Sisal	Improve the bio-resistance of a composite and fiber-matrix interactions.	[37,38]
Benzoylation treatment	Flax Sisal	Reduces the moisture absorption, improve its thermal stability and strength.	[39–41]
Acrylonitrile grafting and Acrylation	Oil palm Glass	It improves the tensile strength and young's modulus.	[42]
Malleated coupling gents	Cellulose fibers Banana Hemp	Growth in flexural strength, hardness, impact strength and flexural modulus.	[43,44]
Permanganate treatment	Cellulose fibers	It improves the hydrophilic tendency of the composite.	[45]
Peroxide treatment	Oil palm	Improves tensile properties.	[45]
Isocyanate treatment	Leaf fibers	Enhance better mechanical properties.	[46,47]

5. Selection of Matrix Materials for Fabrication

A matrix material is one which plays an adverse role in the composite material. The shape, durability and surface properties depend on matrix (resin) material. Based on processing techniques, resin materials are classified into two types, such as thermosets and thermoplastics [48]. A matrix is used as a load distributer among the reinforcement material when external pressure is applied. Thermoplastics are widely used resin material over thermosets because of its ready pick-up shape to the required mold (moldability). Thermosets are not recyclable because it does not come back to its original state when the resin is converted from liquid state to solid state after the curing process. Only a heating process is required for the thermoplastics to form a new shape.

5.1. Thermoplastics

Researchers are showing interest in using thermoplastics as matrix materials because of their integral properties, such as low cost, moldability, less weight and recyclability. The main advantages and disadvantages of these thermoplastic polymer materials are discussed below in Table 7. The viscosity range is more than 500 times that of uncured thermosets during the melting process. Recent results proved that the addition of chemical treatment to the natural fibers may lead to improving the bonding properties between matrix and reinforcement [49], chemical treatments such as; alkaline, silane, acetylation, benzoylation treatments and many others. Due to the addition of chemical and biological treatments, the overall properties of composites are improved [50]. Among all synthetic polymers, thermoplastics are the most commonly used commercial purpose materials. The most widely used thermoplastics are polyvinyl chloride (PVC), polystyrene (PS), polypropylene (PP), polyethylene and natural rubber. The mechanical properties of thermoplastic resins are explained in Table 8.

Table 7. Thermoplastic resins advantages and disadvantages.

Type of Resin	Advantages	Disadvantages
Polystyrene (PS)	Moisture resistance Weather resistance Good chemical resistance	Flammable Brittle Low impact resistance
Polypropylene (PP)	Good fatigue resistance High thermal resistance High dielectric resistance Excellent chemical resistance Low cost	Hard to process Comparatively high cost Limited availability
Polyethylene (PE)	Lightweight Low moisture absorption Good fatigue resistance Good impact strength High ductility	Flammable High thermal expansion Poor weather resistance
Polyvinyl chloride (PVC)	Versatility High impact strength Fire retardant Advance in chemical resistance Less cost Excellent dimensional stability	Poor resistance to UV and temperature
Natural rubber	Good tensile strength High resilience Tear resistance Low cost Water resistance	Poor resistance to hydrocarbons Chances for swelling Complete dissolution

Kasama et al. investigated the mechanical and thermal properties of sisal and glass fibers as reinforcement and polypropylene as a matrix material [51], the authors reported that the addition of polypropylene to the hybrid composites causes improvement in thermal stability, moisture resistance

and also interfacial adhesion between reinforcement and matrix improved. Schmidt et al. reported the mechanical properties of the hybrid composites by using the RTM (resin transfer molding) technique [52]. They analyzed the woven and non-woven composites by in-plane permeability of the hybrid composites and draw a contrast between both of them for comparison purposes and reported that improved mechanical properties and flow media of sisal fibers and glass mats by adding pp to the hybrid composite. Nayak et al. summarized the dynamic mechanical and rheological properties of bamboo glass fiber hybrid composite PP as a resin material by using an injection molding technique [53]. Interestingly, the researcher found that the addition of Maleic anhydride grafted polypropylene (MAPP) in place of normal PP has given advanced results. Vega-Hernandez et al. developed the composites grounded on blue agave bagasse by using polystyrene as a resin matrix [54]. They have invested in the mechanical performance of the hybrid composites by in situ reversible addition-fragmentation chain transfer (RAFT).

Haneefa et al. worked on the tensile and flexural properties of banana and glass fiber reinforced PS composites [55]. It was discovered that the rigidity and modulus of the composite increases by increments in the volume part of the glass fiber. This is expected due to the more noteworthy similarity of glass fiber than banana fiber to polystyrene. In any case, the stretching at break diminishes in increments in the volume portion of glass filaments—this is because of the lower prolongation at the break estimation of glass fiber. Experiments proved that chemical modifications of composites exhibit 30% greater mechanical properties than normal composites. Hao X et al. examined the effects of fiber geometry and alignment on the anisotropy of the flexural and thermal expansion of natural fiber polyethylene hybrid composites [56]. Samples were fabricated by using the extrusion process and, to find out the fiber orientation effectiveness, composites are cut by various off-set angles for comparison purposes and the results showed that at zero angle, composites showed the best mechanical properties whereas at 90 degrees some deviation was observed. Robledo-Ortiz et al. developed green polyethylene natural fiber bio composites by using a rotational molding technique [57]. In this work, composites were prepared by using the rotomolded (rotational molding) technique to develop the bio composites. Natural fibers were combined in a low-density green polyethylene medium and after fabrication, samples were taken for testing mechanical properties. Maleate polyethylene (MAPE) was utilized for surface treatment with the purpose of increasing its adhesion properties. Results indicated that its less elastic nature was because of its poor bonding capacity between reinforcement and matrix. Paran et al. examine the dynamic properties of the green thermoplastic elastomer vulcanizate (GTEV) nanocomposites based on poly vinyl chloride (PVC) and rice straw as reinforcements [58]. The surface morphological studies exposed that the GTEV nanocomposite showed a great surface roughness fracture and specified the presence of some relations between the matrix material and natural fibers [59]. The effect of loading on fibers was foreseen through using the parallel and serious equations [60]. Boussehel et al. worked on the impact of chemical treatment on polyvinyl chloride natural fiber hybrid composites [61]. In this research, the authors drew a contrast between the PVC with and without the chemical treatment of natural hybrid composites and the results showed a slight variation in the mechanical properties [62]. But there is a change of structure for natural fibers after chemical treatment due to the formation of hydroxyl bonds [63]. Haseena et al. carried out research on the interfacial adhesion of natural hybrid fiber natural rubber composites by using the equilibrium technique [64]. The swelling coefficient was found to diminish with an expansion in fiber stacking and penetrant size. The holding operator added blends demonstrated more noteworthy limitation to growth because of the more grounded interfacial grip and the resultant higher cross-linked density [65]. The recent research on various fabrication techniques of thermoplastic resins is discussed in Table 9.

Table 8. Mechanical properties of thermoplastic resins.

Polymer	Density (g/cm^3)	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation (%)	Glass Transition Temp.(°C)
Polyethylene (PE)	0.9	18.0	0.5	350.0	−78.0
High-density polyethylene (HDPE)	0.9–1.0	32.0–38.2	1.3	150.0	−110.0
Low-density polyethylene (LDPE)	0.9	10.0–11.6	0.2–0.3	400.0	−110.0
Polyethylene terephthalate (PET)	1.5–1.6	55.0–159.0	2.3–9.0	300.0	67.0
Polystyrene (PS)	1.04	34.0	3.0	1.6	100.0
High-impact polystyrene (HIPS)	1.0	42.0	2.1	2.5	88.0–92.0
Polycarbonate (PC)	1.2	69.0	2.3	200.0	147.0
Polypropylene (PP)	0.9–1.3	35.8	1.6	80.0	−20.0
Poly methyl methacrylate (PMMA)	1.1–1.2	72.4	3.0	2.5	125.0
Polyvinyl chloride (PVC)	1.3–1.5	52.0–90.0	3.0–4.0	50.0–80.0	82.0
Nylon 6 (PA 6)	1.1	81.4	2.8	60.0	47.0
Nylon 6,6 (PA 6,6)	1.1	82.7	2.8	60.0	70.0

Table 9. Recent research on thermoplastic hybrid composites and fabrication techniques.

Matrix	Reinforcement	Fabrication Process	Properties Observed	Reference
Polyethylene (PE)	Coir fibers	Rotational molding	Water absorption SEM Mechanical measurements	[66]
Polyethylene (PE)	Cupula fibers	Screw extruder	Tensile, flexural properties Thermogravimetric Differential scanning calorimetry	[67]
Polyolefin	Coir coconut fibers	Injection molding	Mechanical Viscoelastic Thermal properties	[68]
LDPE	Pineapple fibers	Compression molding	Flexural and Izod impact Water absorption Thermal resistance	[69]
LDPE	Jute fibers	Compression molding	Mechanical properties Moisture intake	[70]
HDPE	Jute fibers	Sandwich technique	Fracture morphology Mechanical performance	[71]
HDPE	Oil-palm	Injection molding	Thermal properties Dynamic mechanical properties	[72]
PET	Moringa olefin fruit fibers	Hot press	Thermal Mechanical Morphological	[73]
PET	Hemp	Vacuum infusion	Flexural Tensile	[74]
HIPS	Sisal	Compression molding	Water absorption Tensile Fracture	[75]
Polycarbonate (PC)	Carbon fiber	Injection molding	Thermomechanical Tensile Flexural Izod	[76]
Polypropylene	Kenaf Pineapple	Hot press molding compression	Tensile Flexural Water absorption Surface morphology	[77]
Polypropylene	Flax	Hot press technique	Water absorption Sound transmission loss test Tensile	[78]
Polypropylene	Coir fibers	Melt-blending	Microscopic analysis Friction and wear Tensile	[79]
Polypropylene	Banana glass	Twin-screw extruder	Flexural Impact	[80]
PVC	Wood flour Zinc borate	Twin screw extruder + melt press	Water absorption Thermal properties Mechanical properties	[81]
Polypropylene	Marble waste	Injection molding	Moisture absorption Surface morphology Mechanical properties	[82]

5.2. Thermosets

Thermosets are well known for their exclusive uniqueness of making 3-dimensional bonds after curing. Thermoset resin far differs from thermoplastics and these have improved mechanical properties, thermal stability, resistance to the creep and biodegradability [83]. The merits and demerits of some of the thermoset resins are explained in Table 10. Unlike thermoplastics, thermosets are not reusable materials [84]. Epoxy resin, vinyl ester, polyester, phenolic, urea-formaldehyde and many others are some of the examples for thermoset polymers. Among all the resins, epoxy and vinyl ester are widely used resins because of their availability, flexibility and low cost [85]. The primary worry for thermosets is the long length related to handling. Moreover, with the low recyclability of these composites, they require new procedures of structuring maintainable reusing components. For instance, they can be crushed and utilized again as filler materials. The primary target has been the adjustment of the durability of the thermosets and augmenting the handling temperatures. Because of the enhancing mechanical properties as shown in Table 11, thermoset based composites have a huge market for commercial purposes such as the aerospace industry, sports equipment, lightweight objectives, construction field, automobile, biomedical and many other real-life applications. Amongst all these, epoxy resin along with hardener is mostly used in hybrid composite fabrication. There are several processing techniques (Table 12) available in the present time, including the hand lay-up technique, the combination of hand lay-up with compression molding, pultruded, hot press molding, twin-screw extruder and the vacuum assisted resin transfer molding technique (VARTM). The hand lay-up process is mostly used because of cost-effectiveness and easy procedure with improved results. Although there are some drawbacks like curing process, nonuniformity of resin material, vacuum voids in the composite and these are considerably lower compared to the other techniques.

Table 10. Merits and demerits of thermoset resins.

Type of Resin	Advantages	Disadvantages
Epoxy resin	High water resistance High thermal and mechanical properties Low curing time Durability	Difficult to process Corrosive amine hardener More expensive than vinyl ester
Vinyl ester	High chemical resistance High mechanical properties than polyester	High curing shrinkage High styrene content Needs post curing More expensive
Polyester	Lowest cost Easy to use	Limited access moderate mechanical properties Huge curing shrinkage
Phenolic	High fire retardance	High gas emissions Hard to develop

Table 11. Mechanical properties of thermoset resins.

Polymer	Density (g/cm^3)	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation (%)	Glass Transition Temp. (°C)
Epoxy resin (EP)	1.2–1.3	600.0	80.0	1.3	70.0–167
Urea formaldehyde (UF)	1.5–1.6	65.0	9.0	0.8	130.0
Melamine formaldehyde (MF)	1.5–1.6	65.0	12.0	0.6	150.0
Phenol formaldehyde (PF)	1.2	45.0	6.5	1.2	170.0
Unsaturated polyester (UPE)	1.1	60.0	3.4	2.0	110.0–188.0
Rigid thermoset polyurethane (RPU)	1.2	60.0	2.2	90.0	−150.0–0.0

Yorseng et al. developed the bio-based natural fibers hybrid bio epoxy composites for the application of semi-structured components and their mechanical behavior under various circumstances was observed [86]. Surface morphology was conducted by using contact angle measurement properties

to find the wettability of the composites. The results showed that a decrease in mechanical properties because of the bio epoxy degradation by accelerated weathering conditions and the ductile nature of the composite are increased and it causes breakdown [87–89]. Thermal studies stated that bio-epoxy based natural fiber composites withstand up to 295 °C temperature, which means these composites are valid for high-temperature applications. Alves et al. examined the dynamic mechanical properties of hybridized jute and biaxial glass fiber reinforcement and epoxy as matrix material composites [90]. The vacuum infusion technique was used for the best impregnation level for both fibers during the fabrication process. This paper offered the woven and non-woven fabrics reinforced epoxy resin composites and the effect of fiber arrangement on mechanical properties. Asim et al. investigated the dynamic and mechanical characteristics of Kenaf and pineapple fibers reinforced phenolic hybrid composites [91]. Test samples were prepared by using the hydraulic hot press and fibers were ground up to 0.1 to 0.8 mm and compressed for 8 min to get the desired shape and then used for testing purposes. From the outcomes, it is seen that kenaf composites showed advanced mechanical properties whereas pineapple showed the least stability under phenolic resin but the difference is not very comparative [92]. The wear resistance of natural corn stalk fiber reinforced phenolic resin composites were prepared by using the wet granulation technique [93]. The effect of phenolic resin on the brake friction materials was observed by varying the percentage of resin matrix in the composite weight. Results showed that the wet granulation method improves the surface morphology of the friction materials and also stated that tribological behavior also increased.

Singha et al. worked on the synthesis and characterization of pine needles reinforced resorcinol-formaldehyde (RF) biocomposites [94]. Composite sheets by varying the fiber loadings were fabricated by using the compression molding technique and allowed for mechanical properties testing. Results showed that pine needle composites showed more improved mechanical properties than neat RF resin composites. Composite strength increased along with fiber loadings so it has the potential as a replacement for synthetic fibers. Singha et al. investigated the mechanical, thermal and morphological properties of grewia optiva fiber reinforced resorcinol-formaldehyde polymer composites [95]. Composites were fabricated by using the compression molding technique. Fibers were taken in the form of particles for the fabrication and test specimens allowed for material properties testing and composites were prepared by varying the fiber loadings (by weight). Results showed that an increase in mechanical properties up to 30% fiber loadings and then a slight decrease in it due to weak bonding between matrix and reinforcement material in the composite. Thakur et al. studied the mechanical properties of hibiscus sabdariffa fiber reinforced urea-formaldehyde (UF) green composites [96]. Results suggested an improvement in mechanical properties and there is a huge difference between the fiber composites and neat UF resin composites. Fiber composites are much stronger than pure resin composites. Singha et al. worked on the fabrication and characterization of saccharum cilliare fiber-reinforced resorcinol formaldehyde green composites [97]. Samples were fabricated on a compression molding machine. These test specimens were allowed for mechanical properties testing. Results suggested that saccharum cilliare fibers composite were higher than the matrix composites and these fibers have the potential to be used in composite fabrication.

Table 12. Recent research on thermoplastic hybrid composites and fabrication techniques.

Matrix	Reinforcement	Fabrication Process	Properties Observed	Reference
Epoxy	Hemp fibers Flax fibers	Hand layup	Mechanical Moisture analysis Contact angle measurement	[98]
Epoxy	Jute fibers glass fibers	Vacuum infusion	Compressive strength Shear strength	[90]
Epoxy	Crab shell Sisal fibers	Compression molding	Mechanical Chemical Morphological	[99]
Epoxy	Woven cotton Bamboo	Compression molding	Flexural Tensile Impact	[100]
Epoxy	Basalt fiber UHMWPE	Compression molding	Impact Tensile Compression	[101]
Epoxy	Glass Basalt woven	Resin transfer molding technique	Mechanical properties Impact behavior	[102]
Epoxy	Pineapple Basalt	Compression molding	Mechanical properties Tensile Flexural Damping	[103]
Epoxy	Sisal Banana Coir	Compression molding	Hardness Tensile Flexural Water absorption	[104]
Epoxy	Kevlar Cocos nucifera	Compression molding + hand layup	Thermal Mechanical	[105]
Epoxy	Carbon Flax	Hand layup	Impact strength	[106]
Epoxy	Jute Basalt	Hand layup	Mechanical properties Water absorption	[107]
Epoxy	Bamboo Kenaf	Hand layup	Thermal properties	[108]
Epoxy	Banana Flax	Hand layup	Tensile strength Flexural strength Thermal properties	[109]
Epoxy	Neem Banyan leaf	Hand layup	Thermal stability Thermogravimetry	[110]
Epoxy	Coir Kevlar	Hand layup	Mechanical properties	[111]
Vinyl ester	Glass	The Electroless plating method	Thermal wear analysis Mechanical properties	[112]
Vinyl ester	Flax Basalt	Resin infusion and hand layup	Impact strength Mechanical characteristics	[113]
Vinyl ester	Flax Basalt	Vacuum assisted resin infusion	Interlaminar strength Fracture toughness	[114]
Vinyl ester	Flax Basalt	Vacuum infusion	Fracture toughness Water absorption	[115]
Epoxy	Napier Glass	Cold Pressing	Tensile properties	[116]
Epoxy resin	Sisal Coir	Cold Pressing	Moisture absorption Fire retardation	[117]
Epoxy resin	Long kenaf Woven glass	Cold Pressing	Fracture toughness Water absorption	[118]
PLA	Sisal Hemp fibers	Injection molding+ Extrusion	Mechanical properties Thermal stability	[119]

5.3. Bio-Based Resins

Bio-based polymeric resins are produced from renewable sources. These are classified into three types based on their physical properties. The first one is fully bio-based and biodegradable resins those are polyhydroxyalkanoates (PHA) and starch, the second one is partial bio-based resins those are polylactic acid (PLA) and cellulose and the last one is partial non-biodegradable resins those are polyethylene terephthalate (PET) and polyethylene. Most of these bio-based resins are originated from plants and plants-based sugars. The merits and demerits of bio-based resins are discussed in Table 13.

Table 13. Merits and demerits of bio-based resins.

Resin	Merits	Demerits
Cellulose	Inexpensive Easily available Ease to adjust Intermediate impact resistance Intermediate heat resistance Eco-friendly	Relative low decomposition High moisture absorption
Polyhydroxy alkenoates (PHA)	Biodegradable High molecular weight	Low stability Fragile Low decomposition costly
Polylactic acid	Nontoxic nature High strength High modulus Low cost	Brittle Poor impact strength Low thermal degradation
Starch	Low cost Biodegradable	Hard to process Water sensitive Brittle

6. Integration of Nanoparticles in Natural Fiber Hybrid Composites

Hybrid composites are not defected free materials because of the fabrication errors, voids in the composite, weak bonding between matrix and reinforcement, environmental conditions, errors during the development and curing period, scratches on the fiber materials, impurities, unbalanced proportion of matrix and reinforcement affect the total performance of the composite materials [120]. Limiting the reasons for corrupting components can assist with keeping the presentation of composite materials. The addition of new constituents and material advancement quality control techniques is necessary to progress the performance of the hybrid composites. One of the best ways to overcome these problems is the integration of nanoparticles or nano constituents. Recent reports state that the addition of nanofillers in the composite materials showed greater improvement in mechanical properties such as tensile strength, flexural strength, creep strength, tensile strain, shear modulus, impact strength and also thermal properties than individual polymer-based hybrid composites [121].

Nowadays researchers are using three types of nanofillers to improve the performance of the composite material. They are classified into three main groups such as nanotubes, nanoparticles and nanolayers. Nanofillers are used based on the particle sizes—one dimensional, two dimensional and three dimensional. Nanomaterials are larger than macro-level particles, individual atoms and also have a huge surface area per unit volume. Most of them are produced from metals, carbon, polymers and metal oxides. Nano clay, carbon nanotubes, carbon black and graphene are the best example of metal-based nanofillers and also widely used nanomaterials.

Pappu et al. worked on unused materials, like fly ash and sisal fibers, to prepare the micro and nanocomposites for multifunctional applications [122]. The hand layup technique along with compression molding used for the preparation of composites, fly ash and sisal fibers were used as reinforcement and epoxy used as a matrix material. They prepared a total of two composites fabricated for comparison purposes. Results indicated that the presence of natural fiber and nanofiller boost the

performance of the composite, particularly the cellulose content in the sisal fiber and silica along with alumina in fly ash leads to the potential growth of mechanical properties. Majeed et al. reported on food packaging materials from natural fiber hybrid composites, which have nanofiller as nano clay [72] and developed a new method to enhance the bonding between matrix and reinforcement. The addition of nano clay to the hybrid, composite showed good progress in mechanical and thermal properties. But these are not suitable for all types of packaging applications because of higher biodegradability and moisture absorption properties than that of synthetic fibers. Essabir et al. investigated the mechanical and thermal properties of clay and palm fiber hybrid composites [123]. They reported that the hybridization of oil-palm incorporated with clay particles showed improved tensile and young's modulus properties. Similar tests were reported by Arrakhiz et al. In this work, they reported the effect of mechanical and thermal properties of pine fiber reinforced with clay particles hybrid composites [124]; the results were similar to those of the previous research. The addition of clay particles leads to the elimination the voids in the composite surface and adversely affect the surface properties and overall performance of the composite.

Nanoclay particles are one of the most commonly used filler materials due to its availability and inexpensiveness. It comes under the silicate group nanomaterials. Nanoclay particles in the composite material can increase the mechanical, physical and chemical properties. The size and shape of the particles also plays an important role in the composite material for the strength and bonding between reinforcement and matrix material. Toyota automobile industries first started using the nanoclay particles in the polymer composite fabrication [125]; in this research work, the author has shown the importance of clay particles as a filler material to create the interfacial bond between the matrix and reinforcement and an improvement in the physical and fracture toughness of the composite material. Due to advancements in the material properties by using the nanoclay particles, the materials have been used in several functional and structural applications. The microstructure of the nanoparticles present in the material plays a key role in the improvement of mechanical properties.

Generally, nanoclays are available in several forms such as nanoclay platelets, calcined nanoclay and natural montmorillonite (CB). Assaedi et al. investigated the effect of nanoclay platelets (also called cloisite 30B) on the thermal and mechanical properties of fly ash geopolymer [126]. In this study, the platelets are added by varying the weight percentages such as 1%, 2% and 3% loadings. Results showed that adding the nanoclay platelets to the geopolymer shows a slight increase in mechanical properties. Flexural strength and flexural modulus were increased significantly with an increase in nanoclay loading up to 2% weight loading. After 2% weight, the properties of the materials slightly decreased due to porosity and poor dispersion. It is evident that continuous adding of the filler materials to the geopolymers does not increases its mechanical properties. On the other hand, Assaedi also stated the conditions for the temperature variant; he studied this by using the thermo-gravimetric analysis in terms of weight. It was observed that most of the weight loss occurred at 3% nanoclay loading due to extra friction.

Hakamy et al. studied the effect of calcined nanoclay (CNC) on mechanical and microstructural properties of hemp fiber reinforced Portland cement nanocomposites [127]. Calcined nanoclay was prepared by using the heat treatment process of nanoclay particles at 800, 850 and 900 °C temperature for 2 h. It was found that nanoclay converted into a calcined state at 900 °C. Experimental results suggest that the mechanical properties, such as flexural strength and fracture toughness, were improved by adding the CNC to the composite material. Calcined nanoclay particles were added by varying its weight percentages such as 1%, 2% and 3%. The optimum hemp fabric content was found as 6.9 % and CNC was found as 1%.

Eesaee et al. worked on the effect of nanoclay particles in the reinforced glass/woven fiber composites with phenolic resins [128] and mainly dealt with the natural montmorillonite (CN) and artificial montmorillonite (CB). The filler material improved the composite performance because of the proper dispersion of nanoclay particles on the composite surface, which lead to better interfacial bonding with the matrix and reinforcement. It was exposed that the amalgamation of 2.5 wt.% of the

filler material boosts the elastic modulus up to 38% for CN and 43% for CB. On the other side, aging with water molecules and hydroxyl groups of the phenolic matrix leads to a decrease in the strength of the composite.

Avil et al. observed the effect of porosity and failure strength in the presence of nanoclay in glass fiber reinforced epoxy composites [129]. The results showed that the additional filler material caused the reduction of porosity by 73.2% at 10 wt.%. Binu et al. studied the effect of nanoclay, cloisite 15A, on the mechanical performance and thermal properties of polyester-based glass fiber composites [130]. The results showed a maximum mechanical performance observed at 1% loading of the filler materials to the composite. According to this study, the glass transmission temperature failed in between 95 and 110 °C. John et al. observed the dynamic mechanical and thermal properties of cyanate ester syntactic foams besides the nanoclay material [131]. The studies showed that a significant improvement in mechanical properties like tensile, flexural strength moduli was detected for the foams on the integration of nanoclay particles. Nanoclay hikes the toughness of the overall composite and, on the other hand, thermal properties are not much prejudiced by the addition of filler materials.

Maharsia et al. [132] studied mechanical properties in syntactic foams through nanoclay reinforcement. Results indicated that the presence of nanoclay (2% and 5% by weight) showed an improvement in mechanical properties though there was a reduction in modulus due to improper bonding between nanoclay particles and matrix material. Withers et al. [133] observed the effect of cloisite 25A montmorillonite (MMT) nanoclay filler material in epoxy-based composites. The nanocomposites were fabricated by varying the weight percentages of nanoparticles in it. According to the results, the optimum mechanical properties are shown at 2 wt.% of MMT clay addition. The improved surface properties are observed at the low percentage loadings of MMT clay particles. Mo-lin Chan et al. [134] studied the mechanism of reinforcement in nanoclay polymer composite. Different mechanical tests were conducted by varying the content of nanoparticles in the composite material. Interestingly the results are showed that tensile strength and modulus of the composite increased by 34% and 25% correspondingly. The establishment of limitations between the nanoclay clusters and epoxy resin can filter the matrix grains and additionally improve the strength of the composite. Some of the research publications on nanofillers in the composite materials are discussed in Table 14.

Table 14. Various research publications on nanofillers in fiber composite materials.

Fibers/Nanofillers	Polymer	Investigations	Reference
Glass fibers/Silica	Epoxy resin	Elastic properties	[135]
Pineapple/Silica	Epoxy resin	Tensile fatigue Fracture toughness	[136]
Rubber/Silica	Epoxy resin	Vibration and damping	[137]
Glass/Kevlar/Silica	Epoxy resin	Fracture toughness Tensile strength Flexural strength	[138]
Banana/Sisal/Glass/Redmud	Polyester resin	Tensile strength Flexural strength Impact strength	[139]
Silica/Glass/Graphite	Polypropylene	Thermo Mechanical properties and elastic properties	[140]
Basalt/Clay	Polypropylene	Impact Properties	[141]
Carbon fiber/Carbon nanotube	Polyethylene	Mechanical properties	[142]
Rubber/Carbon nanotube	-	Stress-strain analysis	[143]
Carbon fiber/Carbon nanotube	-	Mechanical properties	[144]
Bamboo/Clay	Polyvinyl alcohol	Thermal, Physical and mechanical properties	[145]
Pineapple/Carbon/Jute/Watermelon peel nano particles form	Epoxy resin	Tensile, flexural and fracture toughness	[146]
Sugar palm/Clay	Polyester	Thermal and mechanical properties	[147]
Glass/Clay	Polyester	Mechanical properties	[148]
Glass/Clay	Vinyl ester	Thermal and flame retardancy studies	[149]
Glass/Silica	Vinyl ester	Mechanical properties	[150]
Glass/Clay	Vinyl ester	Mechanical, thermal and vibration properties	[151–153]
Glass/Clay	Vinyl ester	Mechanical properties	[154]
Basalt/graphene oxide	Epoxy resin	Interlaminar shear strength Fracture toughness	[155]

7. Applications

Fiber-reinforced composites are better than pure polymer composites. There should be an occurrence of auxiliary applications on account of its high mechanical properties and lower expense. Due to an account of advanced properties, hybrid composites are a replacement for synthetic composites, particularly when it comes to the load-bearing and construction applications [24]. A recent study stated that goods made up of natural fiber composites have been increased and used in several applications such as ward boards, windows, car door panels, tables, packing selves and lightweight goods [156]. The utilization of natural fiber composites in place of synthetic and petroleum-based fiber composites has increased drastically in various sectors such as sports equipment, aerospace, office machinery, automobile, and shipping industries are one of the main application sectors for lightweight composite materials. The researchers found interest in the replacement of bio fiber-based composites due to its abundance, cost-effectiveness, recyclability, ecofriendliness, low fabrication cost and biodegradability [157,158].

A recent survey stated that the fabrication of fiber-reinforced polymer composites has become more popular in commercial applications, especially in the aerospace industry. The utilization of fiber-based composite caused a reduction in overall weight by 35% of the aircraft, and shows a positive effect on fuel consumption and efficiency of the overall performance. Due to these properties, aircraft industries such as Boeing, Dreamliner and Airbus companies have started using the composites in the majority of their components [159]. The main crucial factor in composite fabrication is weight reduction. Some of the natural fibers serve as a natural filler material in the composite fabrication in the presence of bio-based and biodegradable matrix material designed for the extrusion film blowing of thin packages [160].

Santos da luz et al. worked on natural fiber composites in the replacement of synthetic fiber composites for military applications [161]. They compared natural fiber (pineapple) epoxy composite material and synthetic (aramid) fiber composite material in a hard armor. Results showed that the armor plate made from natural fibers has a stronger impact strength than synthetic fiber plates. Natural fiber hybrid nanocomposites are used in various industries because of its improvement in mechanical, thermal and moisture absorption along with light weight and less cost. Nowadays, these composites are in construction, biomedical, filtration, drug delivery, tissue template, wound dressing and cosmetics. Some of the potential applications are listed below [162].

1. Filter media: Liquid, gas and molecule filtration.
2. Nano-sensors: Biochemical, thermal and piezoelectric sensor.
3. Cosmetics: Skin cleaning and healing.
4. Life science: Drug delivery carrier and wound dressing.
5. Tissue engineering scaffolding: Porous membrane for skin, 3D scaffolds for bone and cartilage regeneration.
6. Industrial applications: Micro/nano electronic devices, electrostatic dissipation, LCD devices, lightweight space craft materials.

8. Conclusions and Future Work

In this review paper, fiber-based hybrid composites have been evaluated, specifically on account of their properties such as surface morphology, thermal stability, dynamic mechanical, mechanical, moisture absorption, fire retardance and applications. Hybridization became famous, not just because of the improved presentation of the subsequent items, but also due to the possibility of conquering the confinements that hamper the relevance of common filaments in specialized structures. There is an immense chance that a huge level of natural fibers can be consolidated in ordinary engineered fortified composites items, which is a major advance from biological and affordable perspectives. The quality natural fibers rely on the development conditions and development time affects the performance of the composite. Although there are so many advantages to using natural fibers in commercial

applications, it is bounded to some of the applications because of its biodegradability and flammable properties compared to synthetic fiber composites. To overcome this problem, the researchers started to work on chemical treatment of natural fibers under various conditions. These modifications improved the fiber strength and also bonding capacity between matrix and reinforcement. On the other hand, the researchers found an interest in the integration of nanofillers into composite materials. The addition of nanoparticles to the hybrid composites played a crucial role in advancing their properties. The surface properties, fracture toughness, void elimination, amount of water absorbed is decreased and the dynamic mechanical properties are increased because of these nanofiller materials in the hybrid composite. It is worth saying that hybrid composite materials bring a revolution in the real-life commercial applications such as biomedical, aerospace, shipping industry, automobile, biomedical, construction, drug delivery, wound dressing and gas filtration applications. In the future, the alterations in fabrication techniques and the addition of nanofillers may lead to improvement as well as prediction of the results is possible, which attracts researchers to use it in multiple applications.

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Development and Analysis of Mechanical Properties of Caryota and Sisal Natural Fibers Reinforced Epoxy Hybrid Composites

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Abstract: In recent years, natural fiber reinforced polymer composites have gained much attention over synthetic fiber composites because of their many advantages such as low-cost, light in weight, non-toxic, non-abrasive, and bio-degradable properties. Many researchers have found interest in using epoxy resin for composite fabrication over other thermosetting and thermoplastic polymers due to its dimensional stability and mechanical properties. In this research work, the mechanical and moisture properties of Caryota and sisal fiber-reinforced epoxy resin hybrid composites were investigated. The main objective of these studies is to develop hybrid composites and exploit their importance over single fiber composites. The Caryota and sisal fiber reinforced epoxy resin composites were fabricated by using the hand lay-up technique. A total of five different samples (40C/0S, 25C/15S, 20C/20S, 15C/25S, 0C/40S) were developed based on the rule of hybridization. The samples were allowed for testing to evaluate their mechanical, moisture properties and the morphology was studied by using the scanning electron microscope analysis. It was observed that hybrid composites have shown improved mechanical properties over the single fiber (Individual fiber) composites. The moisture studies stated that all the composites were responded to the water absorption but single fiber composites absorbed more moisture than hybrid composites.

Keywords: Caryota and sisal fibers; epoxy resin; hybrid composites; mechanical properties, moisture absorption; SEM analysis

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1. Introduction

A matrix material plays an important role in composite fabrication. A matrix is used as a load distributer among the reinforcement material when the external pressure is applied. Based on the matrix material polymer composites are classified as thermoset polymers and thermoplastic polymer composites [1,2]. Unlike thermoplastics, thermosets are not recyclable because it does not come back to its original state when the resin is converted from liquid state to solid state after the curing process. Just heating process is required for thermoplastics to get a new shape. Many researchers found interest in using the thermoset polymer resins over thermoplastic resin due to its wide range of applications. Among all the thermoset resins, epoxy resins have gained much popularity due to their adhesive properties, low shrinkage and curing time, good permeability resistance to moisture [3–7]. Due to their high adhesive properties, epoxy resins are vastly suited for bonding with various materials, such as fibers, steel, plastics, and wood [8,9]. To create a strong network-like structure between reinforcement and matrix material, a curing agent (hardener) is usually added with the epoxy resin. Though their great cross-linking density leads to the crack propagation and fracture toughness of the composites. Many researchers have shown interest to improve its structural stability by using chemical and physical modifications, the addition of nanoparticles, and inert pigments such as glass, iron oxide, and basalt particles [10–14]. Based on matrix material composites are classified into three

types such as organic matrix composites, ceramic-based composites, and metal matrix composites [15,16].

Natural fiber hybrid composites became popular over synthetic fiber composites in recent years due to their lightweight, abundance, low cost, and high strength. These are mainly derived from plants, animals, and mineral resources. Sisal, palm, abaca, bamboo, hemp, flax, hemp, banana, and pineapple are some of the examples of natural (plant-based) fibers. Glass, silica, carbon, and ceramic are some of the examples of synthetic fibers. Synthetic fiber composites are not biodegradable, affect the environment by releasing harmful gases, and also cost-effective. Whereas natural fibers are readily available in nature and do not poses any harm to the environment [17–19]. The physical and mechanical properties of natural fibers and their composites are still being investigating.

In a broad sense, composite materials contain solid mass conveying elements is called reinforcement fill in delicate material is called matrix. Reinforcement affords rigidity and inflexibility, supporting the basic load. The matrix or else binder keeps up the location and introduction about reinforcement. Fundamentally, the content of the composite their personal, physical, and chemical properties are still well balanced they deliver a blend of characteristics that singular contents longing in fit for creating only [20,21]. The composite materials bear benefits over other regular materials because of their greater properties, for example, impact, elastic, and flexural strength, fatigue along stiffness properties, that empower the basic plan to be higher adaptable. Because of the benefits, they are generally eased on mechanical engineering applications like machine components, thermal control, internal combustion engines, electronic packaging, marine applications, construction industry, military applications, sports equipment, and the aerospace industry [22,23].

Chemical composition plays a significant role in plant-based natural fibers and varies from fiber to fiber. The fiber performance mainly depends on the aspect/ratio and cellulose content in the fiber materials. In plant-based fibers cellulose, hemicellulose, and lignin content play a prominent role in overall performance [24].

Reinforced hybrid materials are made by joining at least two distinct varieties of fibers in a distinctive matrix material. Hybridization of two sorts of filaments having particular lengths and widths offers a couple of central focuses over the use of both of the strands alone in a single polymer composite. By far most of the examinations are on the hybridization of natural fibers with glass strands to upgrade the properties [25]. Here the fibers act as the central load-carrying agents and the matrix material encompassing them in a position [26]. As reported by numerous scientists, the hybridization of different fibers under variable weight fractions tends to a gradual upsurge in mechanical, physical properties, and also cost-effective composite materials. The main benefit is mixing of the fibers can be done in various methods such as intermixed continuous fibers, intermixed discontinuous fibers, intermingled particulate fibers, aligned short fibers, and sandwich layers [27]. Sallam et al. [28] investigated the mode II fracture toughness of hybrid fiber reinforced concrete materials. The authors worked on the fracture toughness of four different hybridization patterns. The results showed that the fracture toughness improved in the hybrid composites. Though there are some advantages, it was sensitive to the hybridization patterns of fibers. The authors have given the suitable reasons for this behavior in a discussion part of the following paper concerning the readers' doubts [29]. Sallam et al. [30] worked on the long-term behavior of normal-weight concrete contained hybrid nanoparticles subjected to gamma radiation. The experimental results showed that the addition of nanoparticles to the hybrid composites showed improvement in physical properties and mechanical properties.

The mechanical performance of epoxy resin-based hybrid composites with natural fibers such as hemp, flax, banana, pineapple, jute, sisal, bamboo, palm, abaca, and okra has been extensively studied by many researchers. Studies stated that natural fiber composites are conventional replacements for synthetic fiber composites. Alamri et al. [31] investigated the mechanical properties and water absorption behavior of recycled cellulose fiber reinforced epoxy composites. The results stated that the epoxy-based hybrid composites

mechanical properties increased as fiber content increased. The effect of absorption on the mechanical properties was investigated by using SEM and FTIR analysis. The porosity and bonding between matrix material and reinforcement were observed. Sallam et al. [32] investigated the tribological and mechanical properties of epoxy reinforced by hybrid nanoparticles. Results showed that the addition of nanoparticles to the epoxy resin improved the wear resistance of epoxy and also there were some drawbacks such as reduction of tensile strength and modulus of elasticity. Colomer-Romero et al. [33] studied the comparison of mechanical properties of hemp-fiber bio composites fabricated with biobased and regular epoxy resins. The authors have stated that composites with normal epoxies exhibited improved results whereas bio-based epoxy resins caused fabrication difficulties due to their high viscosity nature and lead to a decrease in performance. Saba et al. [34] investigated the dynamic mechanical properties of oil palm nanofiller kenaf fiber epoxy hybrid composites. It has been found that the incorporation of nanofillers to the epoxy hybrid composites enhances its storage modulus and loss modulus. The authors concluded that the hybridization results in environmentally friendly nanocomposites, possessing superior damping properties and dynamic modulus. Jawaid et al. [35] studied the effect of oil palm and jute fiber treatment on the mechanical performance of epoxy hybrid composites. Results indicated that mechanical properties such as flexural and impact strength properties of altered fiber–reinforced hybrid composites enhanced as compared to unprocessed hybrid composites due to better fiber/matrix interfacial bonding, which was confirmed by scanning electron microscopy. Hanan et al. [36] investigated the mechanical performance of oil palm kenaf fiber-reinforced epoxy-based bilayer hybrid composites. Results showed that the hybridization of kenaf/palm fiber reinforced epoxy composites increased tensile and flexural properties. The inter bonding between matrix and filler material also improved and the results were observed by using SEM analysis. Though there was an improvement in the mechanical properties also poor bonding was observed in the scanning electron microscope analysis, this could be improved by using the chemical treatment of the fibers. Cordeiro et al. [37] studied the effect of composition and fiber modification on mechanical and dynamic properties of epoxy resin-based natural fiber peach palm composites. The authors concluded from the results that a very good mechanical response was observed for these composites even when 70 wt.% of fiber was used. Hanan et al. [38] worked on the characterization of hybrid oil palm empty fruit bunch woven kenaf fabric reinforced epoxy composites. In this research, they investigated the mechanical, physical, morphological properties of hybrid composites. Results obtained show that improvement in tensile and flexural properties whereas reduction in impact strength and is due to improper bonding or fabrication errors. The scanning electron microscopy (SEM) analysis results clearly show the various failure modes of the tensile fractured samples. The hybridization of palm fibers with the kenaf fibers has not given the proper results due to a lack of inter-bonding capabilities. Ramesh et al. [39] Studied the mechanical properties of glass/sisal/jute fiber hybrid composites. The results stated that glass/sisal fiber epoxy resin hybrid composites showed superior properties to jute/glass fiber composites. The addition of sisal to the glass fiber enhances the mechanical properties and also there are few drawbacks due to the presence of glass (synthetic) fibers such as biodegradability, moisture absorption properties. To overcome these problems an attempt has been made on Caryota and sisal natural fiber hybrid composites in the current work.

In this study, Caryota (C) and sisal (S) fiber-reinforced epoxy resin hybrid composites with varying weight fractions were fabricated by using the hand lay-up technique. The fibers were allowed for chemical treatment after the retting process. The fabricated samples were allowed for mechanical testing such as tensile properties, flexural properties, hardness, impact strength, and moisture properties. The scanning electron microscope was used to analyze the surface morphology, chemical composition, and porosity content in the composite samples. The mechanical performance of the fabricated samples was investigated and the results were compared.

2. Materials and Methods

2.1. Materials

Caryota and sisal fibers used as reinforcement materials and epoxy resin used as a matrix material. Sisal fibers are a type of leaf fiber extracted from the sisal plant. Caryota fibers are derived from fishtail palm trees. Both the fibers are readily available in nature. These plants are mainly cultivated in Asian countries since they always needed warm weather conditions. The chemical and mechanical properties of both sisal and Caryota fibers are given in Table 1. The thermoset polymer epoxy resin along with a hardener was used as a matrix material. The wood material was used for mold preparation for the fabrication process.

Table 1. The chemical composition and mechanical properties of natural fibers [40,41].

Property	Sisal Fibers	Caryota Fibers
Cellulose (%)	50–78	37–47
Hemicellulose (%)	10–14	25–34
Lignin (%)	8–11	18–23
Density (g/cm^3)	1.45	0.7–1.55
Young's modulus (GPa)	9.4–22	1–9
Elongation at break (%)	3–7	2–4.5
Moisture (%)	11	13–15
Microfibrillar angle (°)	20–25	14–18

2.1.1. Preparation of Fibers

Fiber extraction is one of the foremost parts of the research. Caryota fibers were extracted from a fishtail palm tree by using a scraping machine. The scrapping machine is a combination of three rollers such as feed roller, serrate roller, and leaf scratching roller. Sisal fibers were extracted from the leaves of the agave sisalana plant. Once these fibers were extracted from resources, both were washed with still water and dried in sunlight. Then fibers were allowed for the chemical treatment process to increase the interface adhesion between fiber and matrix material. 2% HCl solution was prepared and each fiber was kept in the solution for 3–4 h separately and drawn out again washed in still water and dried at room temperature. Then the fiber laminates were separated by using mechanical combs by hand sitting patiently. After separating them, fibers were made into fine pieces using scissors according to the mold dimensions. The fibers used for fabrication purpose are shown in Figure 1.



(**a**) (**b**)

Figure 1. Sisal fibers (**a**) Caryota fibers (**b**) used for the fabrication of composites.

2.1.2. Weight Fraction of Materials

The weight fraction of reinforcements and matrix material used for the development of hybrid composites were considered based on the hybridization concept. It is defined as the fabrication of composites with two or more different fibers under the same resin matrix with a 0.4 weight fraction ratio (0.4 wf.). In this current research, a total of five different (varying weight fractions) composites were fabricated and the weight proportions are discussed in the following Table 2.

Table 2. Fiber weight fractions.

Composite Type	Caryota Fiber (%)	Sisal Fiber (%)	Total Fiber Volume (%)	Total Resin Volume (%)
40C/0S	40	0	40	60
25C/15S	25	15	40	60
20C/20S	20	20	40	60
15C/25S	15	25	40	60
0C/40S	0	40	40	60

2.1.3. Preparation of Matrix Material

Epoxy resin along with hardener was used as a matrix material for fabrication purpose. It was purchased from Sigma Aldrich. The weight proportions were considered as 10:1 ratio as per instructions. The required amount of epoxy and hardener were taken into a plastic container and stirred with a plastic stirrer for 3 to 4 min to get proper mix and left for 30 s. The chemical and mechanical properties of epoxy resin and hardener are given in Table 3.

Table 3. The chemical and mechanical properties of Epoxy and Hardener [42].

Property	Epoxy Resin	Hardener
Type	Araldite LY 556	Ardur HY 951
Mixing proportion	10	1
Color	Pale	Brown
Specific gravity	1.14	1.02
Density (g/cm^3)	1.15–1.18	0.97–0.99
Viscosity at 25 °C (MPa)	550	600
Curing time (h) at 23 °C	24–32	24–32
Pot life (min) at 23 °C	35	35

2.1.4. Fabrication of Composites

Caryota and Sisal fibers are measured for proportionate weight ratios as discussed above. The fibers were laid uniformly inside the mold before applying any resin to it. After arranging the fibers uniformly, they are compressed for a few minutes in the mold. The compressed fibers were laid over the coat of epoxy resin, ensuring uniform distribution of fibers. The epoxy resin mixture has been poured over the fibers uniformly and compressed for a curing time of 24 h, with a constant load of 5 kg. All samples are fabricated according to ASTM D 638M standards. The fabricated samples of Sisal/Caryota fiber hybrid composites are shown in Figure 2.

2.2. Methods

2.2.1. Tensile Properties

Tensile tests were performed to find out the in-plane tensile properties of polymer composites fabricated with matrix and reinforcements. In a broad sense, the tensile test is a measurement of the ability of a material to withstand external forces that tend to pull apart and to what extent the material stretches before breaking. The composite samples were tested on Tinus Olsen H10KT at a constant speed. A total of five samples were tested for

each composite and the dimensions were considered as per ASTM D 3039 standards. The experimental setup used for the tensile test is shown in Figure 3 a. The tensile strength of composites was calculated by using the following relation.



Figure 2. Sisal and Caryota fiber hybrid composites.

$$\sigma_t = \frac{P}{A}\, MPa \tag{1}$$

where σ_t = tensile strength, P = maximum load, and A = cross sectional area.

The tensile modulus of a composite is the ratio of stress to elastic strain when the material is subjected to tension. The following relation was used to calculate the tensile modulus.

$$E = \frac{FL}{Ae}\, MPa \tag{2}$$

where E = tensile modulus, F = maximum load, A = cross sectional area, and e = change in dimension.



(**a**) (**b**)

Figure 3. Tensile test (**a**) Flexural test (Three-point bending test) (**b**) on Tinus Olsen H10KT.

2.2.2. Flexural Properties

The flexural tests were performed to find out the maximum stress and strain when the composites are subjected to external loading. To find out the flexural strength, composite samples are allowed for 3-point bending tests on Tinus Olsen at a constant strain rate of 0.10 mm/min and a crosshead speed of 20 mm/min. The dimensions of samples were considered as per ASTM D 790 standards. The experimental setup used for the flexural test is shown in Figure 3b. The flexural strength was calculated by using the following relation.

$$\sigma_f = \frac{3PL}{2bd^2}\ MPa \tag{3}$$

where σ_f = flexural strength, P = maximum load, L = Span length, b = Width, and d = thickness.

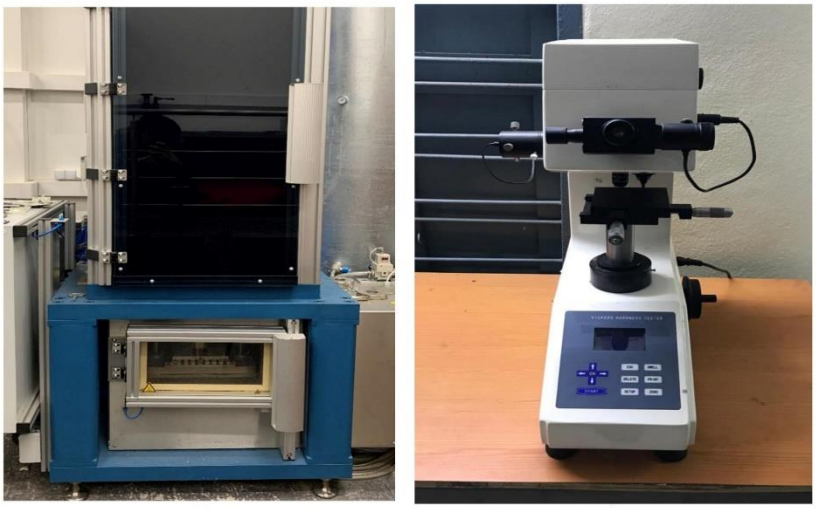
Flexural modulus or tangent modulus is defined as the capability of a composite sample to deform. It is calculated from the tangent of the stress-strain curve. The following equation was used to calculate the flexural modulus.

$$E_B = \frac{L^3 m}{4bd^3}\ MPa \tag{4}$$

where E_B = flexural modulus, L = span length, m = slope, b = width, and d = thickness.

2.2.3. Impact Strength Test

This test is used to define the ability of composite material to withstand the sudden shock loads. This test was carried on a Coesfeld Magnus impact testing machine. The machine consists of a high-speed drop tower with a maximum impactor speed of 40 m/s. The drop tower swung from a set height and which on releasing possess fixed kinetic energy. The test samples were placed like a simply supported beam on the resting position of the impact machine. The tests were conducted as per ASTM D 6110 standards. The experimental setup used for this test is shown in Figure 4a.



(**a**) (**b**)

Figure 4. Coesfeld Magnus Impact testing system (**a**) Micro Vickers hardness tester (**b**).

2.2.4. Vickers Hardness Test

This test is used to find the ability of composite material to resist indentation, scratch, or surface abrasion. In this work, the hardness test was carried out on the Micro Vickers hardness testing machine of model DH85 and Daksh made. It has a load range of 0.2–5 kgs and a measuring range of 5–3000 HV. The measuring magnification and observer magnification are given as 500X and 100X respectively. The experimental setup used for this test is shown in Figure 4b.

2.2.5. Moisture Absorption Test

A Moisture absorption test is used to find out the amount of water absorbed by a composite material when it is exposed to moisture. To find out the rate of absorption, composite samples were placed in a beaker full of distilled water. The sample dimensions were considered as per ASTM D 570 standards. The readings are noted for continuous-time intervals for one week. A total of five samples were tested for each composite material. The rate of absorption was calculated by using the following relation.

$$\% \, Weight \, gain = \frac{W_2 - W_1}{W_1} \times 100 \quad (5)$$

where W_2 = Final weight, W_1 = Initial weight.

2.2.6. Scanning Electron Microscopic Analysis.

The failure studies, surface morphology, and chemical composition of the composite samples were evaluated by using the scanning electron microscope (S-3400N from Hitachi) with an energy dispersive X-ray spectrometer (Quad 5040 from Bruker, Billercia, MA, USA).

3. Results

3.1. Tensile Properties

The analytical results for Caryota and sisal fiber hybrid composites obtained from tensile tests are mentioned in the following Table 4. All the composites specimens were tested as per the standards and for each composite, a total of five samples were tested to note down the average values. The tensile stress was applied in the longitudinal to the fibers direction and speed was maintained as 2mm/min. The tests were conducted at 21 °C temperature and relative humidity as 28%. The weight fractions of composites are 40C/0S, 25C/15S, 20C/20S, 15C/25S, 0C/40S.

Table 4. Tensile strength properties.

Composites	Tensile Strength (MPa)	Tensile Modulus (GPa)
40C/0S	22.2	4.20
25C/15S	28.1	5.92
20C/20S	35.4	6.19
15C/25S	38.2	6.44
0C/40S	25.7	5.66

Figure 5 shows the tensile strength behavior for Caryota and sisal fiber hybrid composites with varying weight fractions. From the results, the tensile strength values of 15C/25S hybrid composites are inferior as compared to the other composites. It showed the tensile strength as 38.2 MPa whereas 20C/20S showed 35.4 MPa. The single fiber composites showed the least tensile strength as 22.2 and 25.7 MPa. The hybridization effect is noticed in this case. It was observed that the tensile strength values among the hybrid composites increased with an increase in sisal fiber content in it. The reason for the above can be attributed in two ways. An increase in sisal fiber content increases the strength of the composite. However, higher individual fiber content leads to the agglomeration of fibers,

hence loss of strength. Poor bonding between matrix and reinforcement material at the interface might be another reason for lower the tensile strength value.

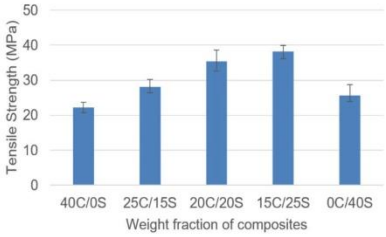


Figure 5. Tensile strength vs. weight fraction of C/S composites.

The tensile modulus of various weight fractions of composites is given in the following Figure 6. Among all the composites hybrid composites showed superior properties over single fiber composites and the values are close to each other. Single fiber Caryota composites showed the least tensile modulus among all the composites and 15C/25S composites showed the highest tensile modulus.

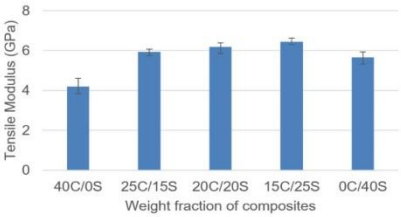


Figure 6. Tensile modulus vs. weight fraction of C/S composites.

3.2. Flexural Properties

The data presented in Table 5 demonstrates that the flexural properties for all the composites. Flexural tests were performed by using three-point bending tests and the dimensions of the test samples are taken from ASTM standards. The flexural test results include displacement and strength. The tests were conducted at 21 °C temperature and relative humidity as 28%. For each composite, a total of five samples were tested and the values are noted.

Table 5. Flexural strength properties.

Composites	Break Load (N)	Flexural Strength (MPa)	Flexural Modulus (GPa)
40C/0S	267	64.09	2.14
25C/15S	287.7	69.05	2.73
20C/20S	341.2	81.89	3.06
15C/25S	371.5	89.16	3.40
0C/40S	282.1	67.70	2.64

Figure 7 shows the relation between flexural strength and weight fraction of the fabricated composites. As it can be seen from the below figure that the hybrid composites showed superior flexural properties than single fiber composites. 15C/25S hybrid composites showed the highest flexural strength as 89.16 MPa and single fiber Caryota

composites showed the least strength as 64.09 MPa. Hybrid composites exhibited good flexural properties which indicate that the materials have brittle properties. It was observed that the flexural properties trend is similar to the tensile properties. The loss in strength of single fiber composites can be explained as (1) early failure of the composite upon external loading. (2) weak interfacial bonds between fiber and matrix material leads to the clustering of fibers.

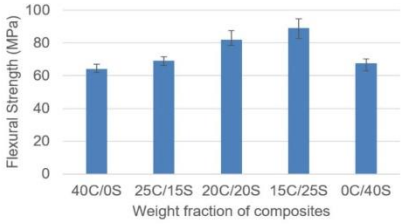


Figure 7. Flexural strength vs. weight fraction of C/S composites.

The results presented in Figure 8 show the evolution of flexural modulus according to the fiber weight fractions. It was observed from the results that single fiber Caryota and sisal fiber composites showed a lower flexural modulus than Caryota sisal hybrid composites. Single fiber composites showed 2.14 GPa and 2.64 GPa respectively.

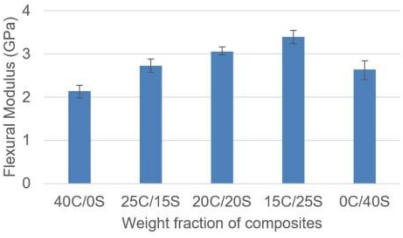


Figure 8. Flexural modulus vs. weight fraction of C/S composites.

The flexural strength is higher than the tensile strength. Indeed, the dimensions of the test samples are similar for both tests. Only at the center of the composite is stressed in the three-point bending test while the whole is in tension test, then fewer defects are involved in the 3-point bending test. Whereas tangent modulus or tensile modulus is more than the flexural modulus. The reason can be attributed to, in a tensile test, the maximum tensile stresses are experienced throughout the entire volume (and surface area) of composites whereas, in bending test, the maximum tensile stresses are conversely concentrated in a small region on the top surface.

3.3. Impact Test

The data presented in Figure 9 was obtained from the impact tests. The specifications of the samples were taken from ASTM standards and the tests were conducted at 21 °C temperature and relative humidity as 28%. It is well known that the impact strength of the composites mainly depends on the properties of reinforcement and matrix material presented in it. It was observed that the hybrid composites showed superior impact strength over single fiber composites. Single sisal fiber composites exhibited the least impact strength as 83.5 Joules.

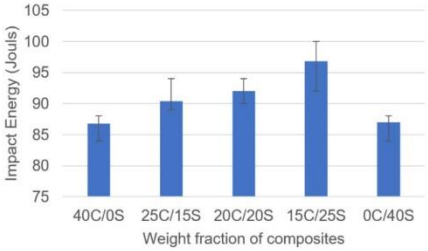


Figure 9. Impact strength vs. fiber weight fractions.

3.4. Vickers Hardness Test

The hardness values of various weight fractions of composites are given in Figure 10. It was observed from the results, single fiber composites showed the highest hardness values whereas hybrid composites showed the least hardness values. The hybridization impact is not noticed in this case. 20C/20S composite showed the least Vickers hardness values among all composites.

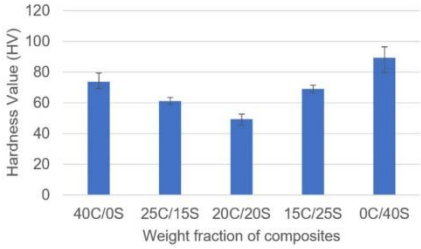


Figure 10. Hardness vs. weight fraction of C/S composites.

3.5. Moisture Analysis

The amount of moisture absorbed by the composites was calculated by using the moisture test. The samples were allowed for the heat treatment process (placed in an oven at 60 °C for 15 min time to eliminate the moisture in it) before placing them in the water container. The sample dimensions were taken from the ASTM standards and the test was conducted at 21 °C temperature and relative humidity as 28%.

Figure 11 shows the % weight gain by the composites with varying weight fractions to time. It was observed that all the composites samples responded to the moisture (water) when it is exposed. The amount of weight gain by the composites increased with time up to 120 h and remains constant. All hybrid composites showed intermediate results whereas single fiber composites showed the least and highest percentage of weight gain. The reason can be attributed to the presence of void content (porosity) in the composites and the chemical composition (cellulose content) of the fibers. Because the oxygen ($C_6H_{10}O_5$) in the cellulose reacts to the hydrogen in the moisture and forms the hydrogen-oxygen bonds with it. Sisal fibers contain more cellulose percentage than Caryota, hence it observed more % of water. The porosity in the composites specimens was observed by using SEM analysis and the results were mentioned in Figure 17. It was observed that the hybrid composites have an intermediate percentage of void places compared to the single fiber composites. The presence of void places attracts the water molecules hence the percentage gain is more.

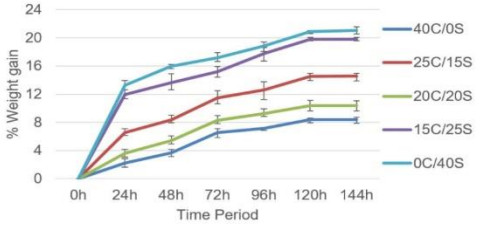
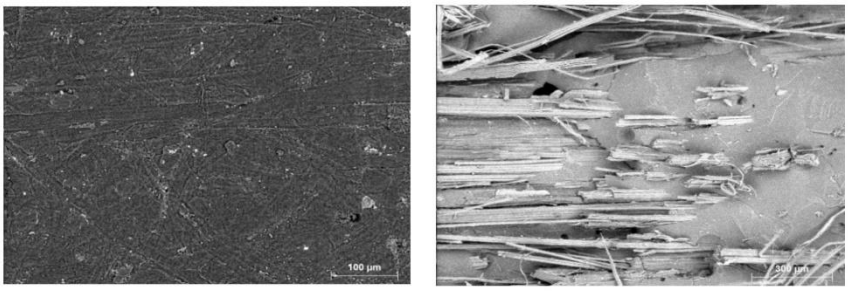


Figure 11. Weight gain in % by the composites vs. time period.

3.6. Scanning Electron Microscopic (SEM) Analysis

The SEM images of the flexural, tensile tests fractured composites and chemical composition at the surface and fractured specimens were taken to analyze the presence of voids, cracks, and the reinforcement and matrix adhesion (bonding) behavior.

The above Figure 12 shows the micrographic spectrum acquisition for single sisal fiber composite to analyze the chemical composition in it.



(**a**) (**b**)

Figure 12. SEM micrographs of single sisal fiber composite (0C/40S) (**a**) surface and (**b**) fractured position.

Based on the spectrum analysis of a specimen, single sisal fiber hybrid composites the presence of composition on the surface is shown in Figure 13. The data presented in the above graph is shown in the following Table 6. The structure is expressed by the atomic weight and number of each element presented in it. The weight % of carbon is more than the weight % of oxygen, this reflects that the matrix concentration is more at the surface of the composite than reinforcement. Due to this smooth surface finish will occur to the composite.

Table 6. Chemical structure of single sisal fiber hybrid composite at surface position.

Element Number	Element Symbol	Element Name	Series Name	Atomic Weight	Weight Conc. (%)	Atomic Conc. (%)
6	C	Carbon	K-series	12	68.68	74.66
8	O	Oxygen	K-series	16	30.75	25.09
11	Na	Sodium	K-series	23	0.17	0.10
17	Cl	Chlorine	K-series	35.5	0.40	0.15

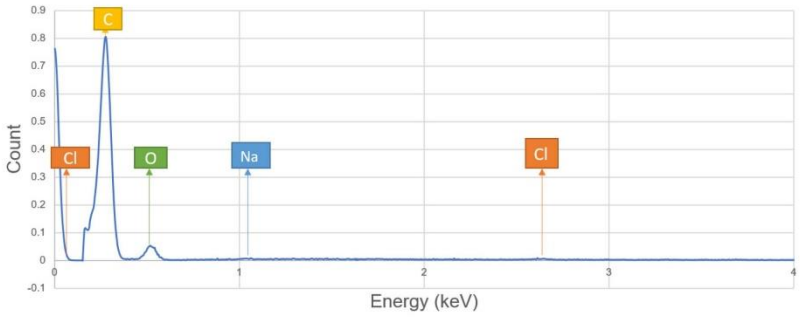


Figure 13. Spectrum analysis of the chemical structure of single sisal fiber hybrid composite surface position obtained at an intensity of 4 keV.

Based on the spectrum analysis of a specimen, single sisal fiber hybrid composites the presence of composition on the surface is shown in Figure 14. The data presented in the above graph is shown in the following Table 7. The structure is expressed by the atomic weight and number of each element presented in it. It was observed that the weight % of carbon is nearly equal to the weight % of oxygen, this reflects that the matrix concentration and reinforcements are in proper proportion inside the composite. This reflects strong bonding between matrix and reinforcement in the composite and also, improvement in the strength of the composite.

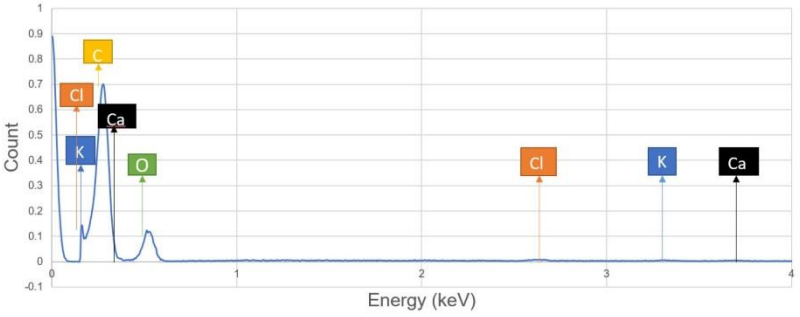


Figure 14. Spectrum analysis of the chemical structure of single sisal fiber hybrid composite fractured position obtained at an intensity of 4 keV.

Table 7. Chemical structure of single sisal fiber hybrid composite at fractured position.

Element Number	Element Symbol	Element Name	Series Name	Atomic Weight	Weight Conc. (%)	Atomic Conc. (%)
6	C	Carbon	K-series	12	52.56	59.98
8	O	Oxygen	K-series	16	46.18	39.57
17	Cl	Chlorine	K-series	35.5	0.46	0.18
19	K	Potassium	K-series	39.09	0.38	0.13
20	Ca	Calcium	K-series	40.08	0.42	0.14

After the analysis of chemical concentration presented at the surface and fractured position of single sisal fiber hybrid composite, we can conclude that the percentage of carbon is increased in the surface compared to the fractured (inside) position which denotes that epoxy concentration is more at the surface. Whereas oxygen percentage more at the fractured position which denotes that fiber concentration is more at the breakpoint.

Figure 15 shows the fracture surface of the flexural test composites as a function of hybridization. A brittle fracture of reinforcement and matrix material was observed from the results. The reason can be attributed to the strong interfacial bonds between fibers and epoxy resin. It can be seen from the above figures that the presence of voids and cracks leads to weakening the strength of the composites. It was observed that single fiber composites (Figure 15a,e)) have more cracks and air gaps in them when compared to the hybrid composites, this is due to lack of fiber/resin adhesion and poor bonding nature.

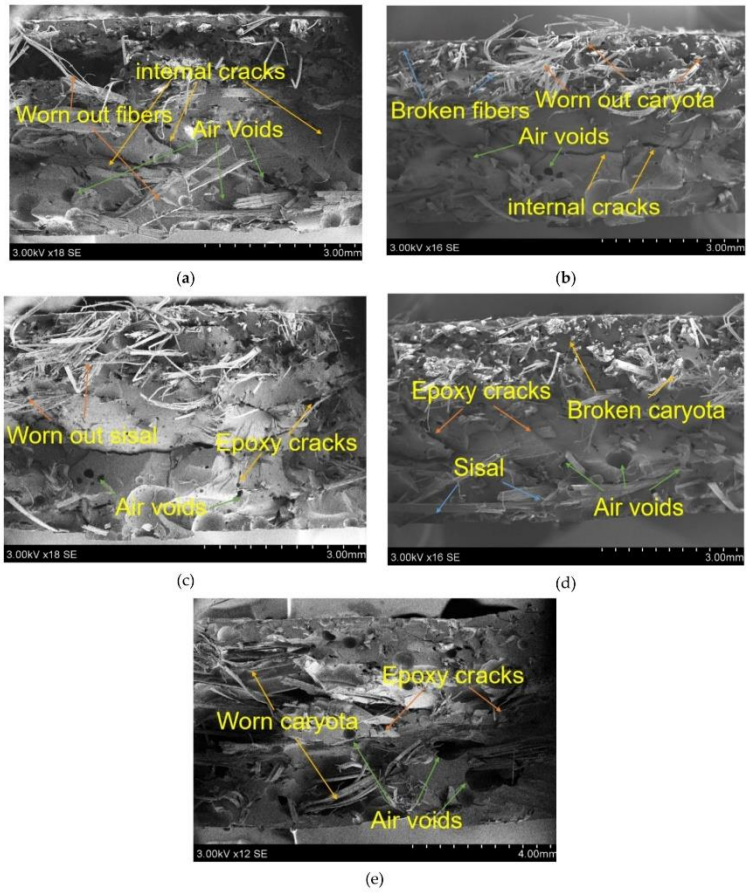


Figure 15. SEM images for fractured composites of flexure test specimens (**a**) 0C/40S, (**b**) 15C/25S, (**c**) 20C/20S, (**d**) 25C/15S, (**e**) 40C/0S.

The fracture properties of composites after the tensile test were inspected through SEM and it is given in Figure 16. it is observed that the cluster of matrix shards cling to the fiber reinforcements and enclosed it. This helps the composite to worn out when the external load acting on it when it reaches its maximum strength. The fiber pulled out needs extra strength compared to fiber fracture. it can be seen that in the case of single fiber composites (Figure 16a,e) the fibers are separated from the matrix due to poor adhesion. Wherein the case of hybrid composites some of the matrix material is presented at the top of fibers. This indicates the strong crosslinking reaction between reinforcement and matrix materials. That gives extra support to the composites to worn out. Moreover, it was observed that the presence of cracks and void places are less in hybrid composites compared to the single fiber composites.

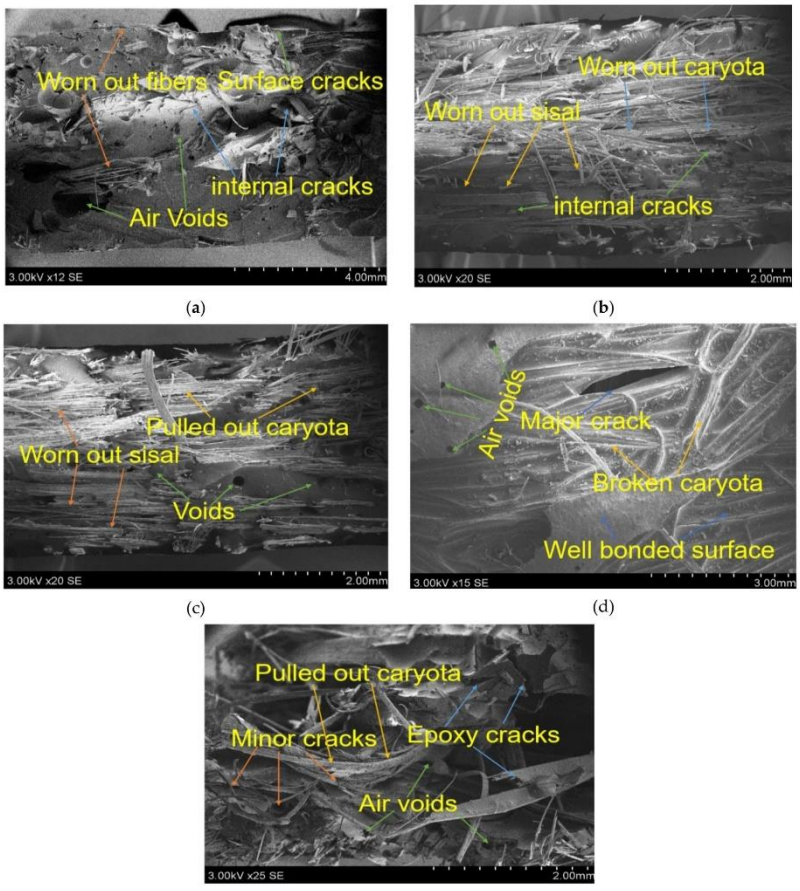


Figure 16. SEM images for fractured composites of tensile test specimens (**a**) 0C/40S, (**b**) 15C/25S, (**c**) 20C/20S, (**d**) 25C/15S, (**e**) 40C/0S.

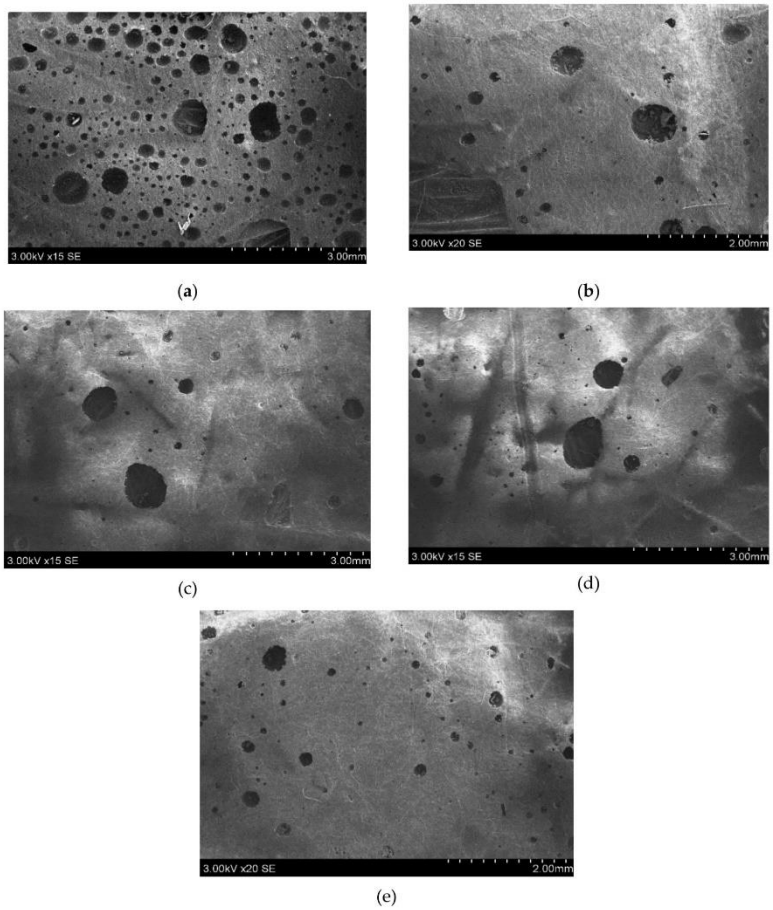


Figure 17. SEM images for porosity (void content) presented in the fabricated composites (**a**) 0C/40S, (**b**) 15C/25S, (**c**) 20C/20S, (**d**) 25C/15S, (**e**) 40C/0S.

The presence of air voids and gaps in the composite surfaces may lead to weakening the strength of the composite and also the composite may absorb more water when it faces the wet medium. Figure 17 shows the presence of porosity content in all the fabricated composites, it was observed that 0C/40S composite has the more void percentage whereas 40C/0S composite has the least percentage of void content. All the hybrid composites have intermediate percentages and close to each other. The reasons can attribute as (1) strong internal bonding between reinforcement and matrix in the hybrid composites. (2) fabrication errors. (3) atmospheric conditions such as temperature and humidity while fabricating the composites.

4. Discussion

Previous research was concentrated on the fabrication of banana, pineapple, hemp, flax, and sisal fibers-based hybrid composites for lightweight and biomedical applications [43–49]. The present research was concentrated on developing the hybrid composites with Caryota (derived from fishtail palm plants) and sisal fiber-based hybrid composites with varying weight fractions.

The Caryota and sisal fiber-reinforced epoxy resin-based hybrid composites were fabricated by using the hand layup technique. The developed hybrid composites have shown superior mechanical properties. In the previous research, banana/pineapple-based hybrid composites showed flexural properties ranging from 37 to 53MPa and hemp/flax hybrid composites showed that in between 45 to 63 MPa whereas Caryota and sisal-based hybrid composites showed in between 65 to 89 MPa which is far higher than other hybrid composites. Similarly, there is a considerable improvement in the tensile properties as well.

For future aspects, implementation of nanoparticles such as nano clay. ferrous oxide and silver particles in bio-based epoxy resin foreseen. Moreover, the orientation of fiber materials in the composite materials has to be considered because, previous researchers proved that, the orientation of reinforcement may increase the torsional strength and also crosslinking between matrix and fiber material. The bio-based composites could be used for biomedical and microfluidic applications.

5. Conclusions

Caryota and sisal fiber-based epoxy resin hybrid composites were developed by using the hand layup technique and the mechanical properties were investigated. Experimental results stated that hybrid composites showed superior properties. From the tensile test, it was found that 15C/25S hybrid composites have shown 38 MPa whereas single fiber composites (0C/40S, 40C/0S) showed 25 and 22 MPa respectively. From flexural properties, it was found that the hybrid composites showed improved flexural properties than single fiber composites. The 15C/25S hybrid composites showed flexural strength and flexural modulus as 89.16 MPa and 3.40 GPa. Single Caryota fiber composites showed the least strength and modulus as 64.09 MPa and 2.14 GPa. Coming to the impact strength 15C/25S composites showed as 97 joules and in single sisal fiber composites, it was observed that 83.5 joules. in all the tests hybridization effect was observed clearly except the hardness test. In the hardness test, single fiber composites showed superior properties to hybrid composites. The reason could be fabrication errors or poor bonding between matrix and reinforcement material.

Thus, SEM analysis showed that hybrid composites have fewer defects such as voids, minor and major cracks, poor adhesion between matrix and reinforcement material than single fiber composites.

Moisture absorption measurements stated that all the composites showed a constant increase up to 120 h and remain constant. Hybrid composites showed intermediate results whereas single fiber composites showed the highest and lowest rate of absorption. By considering all the results Caryota and sisal fiber hybrid composites showed superior mechanical properties than single fiber composites. And Caryota and sisal fibers are potential alternatives in fiber composite fabrication.

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Analysis of Mechanical and Wettability Properties of Natural Fiber-Reinforced Epoxy Hybrid Composites

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Abstract: Natural fibers have many advantages over synthetic fibers due to their lightness, low cost, biodegradability, and abundance in nature. The demand for natural fiber hybrid composites in various applications has increased recently, because of its promising mechanical properties. In this research work, the mechanical and wettability properties of reinforced natural fiber epoxy resin hybrid composites were investigated. The main aim of this research work is the fabrication of hybrid composites and exploit its importance over individual fiber composites. The composites were fabricated based on the rule of hybridization mixture (0.4 wf) of two fibers using sets of either hemp and flax or banana and pineapple, each set with 40 wt%, as well as four single fiber composites, 40 wt% each, as reinforcement and epoxy resin as matrix material. A total of two sets (hemp/flax and banana/pineapple) of hybrid composites were fabricated by using a hand layup technique. One set as 40H/0F, 25H/15F, 20H/20F, 15H/25F, 0H/40F, and the second one as 40B/0P, 25B/15P, 20B/20P, 15B/25P, 0B/40P weight fraction ratios. The fabricated composites were allowed for testing to examine its mechanical, wettability, and moisture properties. It has been observed that, in both cases, hybrid composites showed improved mechanical properties when compared to the individual fiber composites. The wettability test was carried out by using the contact angle measurement technique. All composites in both cases, hybrid or single showed contact angle less than 90°, which is associated with the composite hydrophilic surface properties. The moisture analysis stated that all the composites responded for moisture absorption up to 96 h and then remained constant in both cases. Hybrid composites absorbed less moisture than individual fiber composites.

Keywords: natural fibers; hemp and flax fibers; banana and pineapple fibers; hybrid composites; mechanical properties; contact angle; moisture absorption

1. Introduction

Recently, many researchers have become interested in working with natural fiber hybrid composites, because of its abundance, light weight, low cost, high specific properties, and the fact it does not pose any harm to the environment. Natural fibers are finding increasing use as reinforcement materials in polymer matrix composites, often replacing synthetic (manmade) fibers [1]. Hybrid fiber composites are materials that consist of two or more reinforcements and the same matrix material, and the procedure is called a hybridization process [2]. The matrix serves as binding material and gives the extra strength to the composite [3]. Composites are classified as per matrix and reinforcement materials. Based on the matrix material, composites are classified as metal matrix composites (MMCs),

ceramic based composites (CMCs), and organic matrix composites (OMCs), based on reinforcement material composites are classified as laminar composites, particulate composites, and fiber reinforced composites (FRCs) [4–6]. In the fiber reinforced composites, fiber material serves as reinforcement material and works as a stiffness agent. The matrix material does not only serve as a binding agent but also distributes the external forces (load) acting on the composite to the fibers. The majority of the external load is sustained by the reinforcement materials and a small portion is carried by the matrix material.

The fibers are classified as natural fibers and synthetic fibers. Abaca, bamboo, banana, cotton, coir, flax, hemp, jute, kenaf, pineapple, and sisal are some examples of natural fibers. Boron, carbon, ceramic, and glass are some examples of synthetic fibers. They are also called manmade fibers. Synthetic fiber composites have so many drawbacks when compared to natural fiber composites, such as rapid burning, not being good for the environment, not being skin-friendly and therefore uncomfortable for extensive wear [7]. Whereas natural fibers are biodegradable, low cost, environmentally friendly, light in weight, have great availability in nature, and are renewable. The biodegradability of natural fibers shows a major impact in balancing a healthy environment with inexpensiveness and high specific performance, which attract the economic interest of many industries. The physical and mechanical properties of natural fibers and their composites are still being explored.

Hybrid composites are fabricated to maintain the advantages of its constituents and overcome some of the drawbacks. The concept of hybridization is successful approach in plant fibers to address the deficiency of ductility of the composites [8]. Hybridization is defined as when the composites are fabricated with two or more reinforcements and are joined under the same matrix (resin) material with 0.4 wf ratio. As reported by many researchers, the hybridization of different fibers under varying weight fractions tends to a gradual increase in mechanical, physical properties, and also cost-effective composite materials. The main advantage is that the mixing of the fibers can be done with various methods, such as intermixed continuous fibers, intermixed discontinuous fibers, intermingled particulate fibers, aligned short fibers, and sandwich layers [9,10]. Due to the hybridization of natural fibers, composites showed greatly enhanced mechanical properties. A recent survey stated that the fabrication of natural fiber reinforced polymer composites has become more popular in commercial applications, such as in the construction, automobile, military goods, sports equipment, aerospace, and shipping industries. Fernanda Luz et al. [11] worked on composites with natural fibers and conventional materials applied in a hard armor. The results stated that the usage of natural fiber composites in armor plates to reduce the thickness of the hard armor plate. The experimental results stated that the natural fiber-reinforced composites have the potentiality to replace ceramic-based composites. Neuba et al. [12] investigated the mechanical, thermal, and ballistic properties of novel epoxy composites reinforced with Cyperus malaccensis sedge fiber. The results indicated an improvement in mechanical properties when compared to the neat epoxy composites. Yorseng et al. [13] worked on weathering studies of kenaf and sisal fiber reinforced hybrid composites for semi-structural applications. The mechanical and thermal properties of natural fiber hybrid composites have more enhanced properties than pure epoxy composites at both before and after weathering conditions. From the results, it is a considerable advantage to use bio-based composites in semi-structural applications.

In general, the chemical composition plays an important role in the natural fibers and it differs from fiber to fiber. The overall performance is mostly reliant on the aspect/ratio and cellulose crystallinity of the fiber materials. Cellulose plays a prominent role in the plant fibers, and it shows an adverse effect on moisture absorption. Hemicellulose and lignin are also major components, in addition to by cellulose. Cellulose occupies the major place in the plant fibers, and it comprises up to 55% of the total. Lignin serves as cementing material and its stimulus the surface morphology and properties [14]. Hybrid composites made from plant fibers and their by-products are the most widely used lignocellulosic materials, due to their enhanced mechanical properties. Banana and pineapple are the most cultivated plants, providing leaf fibers for composite applications in India. These leaf fiber composites and their by-products utilization increased in a wide range of commercial applications.

Bast fiber composites, such as leaf fibers, especially hemp and flax fiber composites, are also becoming popular. Hemp and flax fibers are mostly cultivated in European countries [15]. Researchers are showing much interest, due to its mechanical performance and lightweight applications. In agreement with the European Commission, the use of waste raw materials obtained from the processing of bast fibers, such as hemp and flax, is an ailment for the balanced use of plants [16]. Along with reinforcement material, matrix material also plays an important role in the hybrid composites. The structure and surface morphology properties are based on resin material. Based on dispensation techniques, matrix materials are classified as thermosetting and thermoplastic materials. A matrix material works as a load distributor in a composite when an external force acts on it [17]. Thermosets are widely popular for their exclusive property of forming 3D bonds after curing the composite. They are a contrast to plastic polymer materials, and have great mechanical performance, thermal stability, and resistance to creep [18]. Thermoset polymers are a more widely used resin material for composite fabrication than thermoplastic polymers. Thermosetting materials have superior mechanical properties, thermal stability, and ease of processing. Thermosets are easy to process and do not require additional heat treatment process for composite curing. Unlike thermoplastic material, these are not recyclable. Polystyrene, polyethylene, polyvinyl chloride, and natural rubber are some of the examples for thermoplastic resins. Epoxy, phenolic, polyester, vinyl ester, urea-formaldehyde, and unsaturated polyester are some examples of thermoset resins. Researchers are showing much interest in using epoxy resin, due to its exponential mechanical properties, low curing time, durability, availability, and low cost [19]. Epoxy resin—along with hardener used as a matrix material in the current research.

Many researchers found that reinforcing with natural fiber with epoxy resin matrix has given enhanced properties. Sapuan et al. [20] worked on the mechanical properties of banana fiber reinforced epoxy resin composites. The results stated that banana fiber composites showed improvement in flexural strength, durability, and stiffness. Al Rashid et al. [21] studied the utilization of banana fiber reinforced hybrid composites in the sports industry. Due to its extensive mechanical properties, banana fiber composites are a considerable replacement for glass fiber composites in the sports industry. Karimzadeh et al. [22] studied the effect of stacking sequence on the mechanical properties and moisture absorption characteristics of hybrid pineapple/glass fiber composites. The results indicated that hybrid composites have superior flexural strength, tensile strength, and tensile strength than pure composites. Pure pineapple absorbed more percentage of water than hybrid composites. Shivamurthy et al. [23] investigated the sliding wear, moisture absorption, flammability, and mechanical properties of banana short fiber/$Al(OH)_3$ epoxy composites. The mechanical properties, such as tensile strength and hardness, improved significantly due to the addition of natural fibers, and these properties are enhanced by increasing the fiber material to a certain extent. Neves et al. [24] worked on comparative mechanical properties between bio composites of epoxy and polyester matrices reinforced by hemp fiber. Hemp fiber composites with epoxy resin showed greater properties than polyester composites. Epoxy and polyester resin composites showed flexural strength as 66.7 and 49.1 MPa, and tensile strength as 50.5 and 25.4 MPa, respectively. Thiagamani et al. investigated the mechanical, moisture absorption, and swelling behavior of hemp/sisal fiber reinforced bio-epoxy hybrid composites: effects of stacking sequences [25]. The experimental results stated that hybrid composites displayed small variation in tensile strength when the stacking pattern was changed. Hybrid composites showed 40% higher mechanical properties than individual fiber composites. Moudood et al. [26] studied the mechanical properties of flax fiber reinforced composites at different relative humidifies: experimental, geometric, and displacement potential function approaches. The experimental results indicated that improved tensile strength and tensile strain in natural hybrid fiber composites. Mohammed Sh. Al-Otaibi et al. [27] worked on the characterization of date palm fiber-reinforced different polypropylene matrices. In this study, the authors investigated the mechanical, morphological, and thermal properties of date palm fiber composites. It was found that the reduction in tensile properties and improvement in modulus, when compared to the neat resin composites and date palm fiber composites, show advanced thermal and morphological properties. Junio et al. [28] worked on Copernicia prunifera leaf fiber epoxy

composites. The results showed a 40% increase in tensile properties and a 69% increase of the elastic modulus, when compared to the neat epoxy resin composites.

In this research work, two sets of hybrid composites (five different weight fractions) based on epoxy resin and reinforcement with four types of natural fibers, hemp (H), flax (F), banana (B), and pineapple (P), were fabricated by using hand lay-up technique. The fabricated composites were allowed for material properties testing, such as flexural strength, flexural modulus, interlaminar shear strength, contact angle measurement, and moisture absorption. The mechanical performance of the two sets of fabricated laminates was investigated and the results were compared.

2. Materials and Methods

2.1. Materials

The reinforcement materials used for the preparation of samples are banana, flax, hemp, and pineapple natural fibers and epoxy resin. The mechanical properties and chemical compositions of these fibers are given in Table 1. Banana and pineapple fibers are a type of leaf fiber that is readily available in nature. These are widely available in India. Whereas hemp and flax fibers are a type of bast fiber, which is extracted from the banks of the plants. Epoxy resin, along with hardener, was used as a matrix material and it was purchased from Composites 24, Riga, Latvia. The mold material was made from teak wood.

Table 1. The chemical and mechanical properties of natural fibers [29,30].

Properties	Banana	Pineapple	Hemp	Flax
Density (g/cm^3)	1.35	1.44	1.48	1.4
Young's modulus (GPa)	27–32	60.82	70	60–80
Elongation (%)	2.6–5.9	2.2	1.6–4.0	1.2–1.6
Cellulose (%)	63–83	12–60	70–74	64–42
Hemi cellulose (%)	6–19	19.5	21–24	16
Lignin (%)	5–10	4.6–11	3.7–5.7	2–2.2
Moisture (%)	10.71	8–11.8	6.2–12	8–12
Microfibrillar angle (°)	11–12	12–14	2–6	5–10

2.2. Preparation of Fibers

Fiber selection and extraction is one of the major parts of the research. All the obtained fibers were cleaned with water and then dried to eliminate the water in it. Then, the fibers were segregated slightly with hand sitting patiently. The fiber laminates were allowed for the hand retting process to separate individual fiber strands by using a mechanical combing process. After separating them, fibers were looked over with a cotton checking outline several times, to isolate the filaments. Then, fibers were allowed to dry at room temperature to eliminate any moisture. After that, all the fibers are measured for weight and length, as per the required dimensions. The fibers used for the fabrication purpose are shown in Figure 1.



Figure 1. Fibers used for the fabrication.

2.3. Weight Fraction of Reinforcement and Matrix Materials

The weight fraction of the constituents used for the fabrication process was considered based on the hybridization process. In the present work, total two sets of banana(B)/pineapple(P) and hemp(H)/flax(F) composites were fabricated by varying the weight fraction of fibers, such as 40B/0P (40% banana fibers), 25B/15P (25% banana and 15% pineapple), 20B/20P (20% banana and 20% pineapple), 15B/25P (15% banana and 25% pineapple), 0B/40P (40% pineapple fibers), 40H/0F (40% hemp fibers), 25H/15F (25% hemp and 15% flax), 20H/20F (20% hemp and 20% flax), 15H/25F (15% hemp and 25% flax), and 0H/40F (40% flax fibers).

2.4. Preparation of Matrix Material

Epoxy resin, along with hardener, was used as a matrix material. For the proper resin solution, the weight proportions of both resin and hardener were considered as 10:2 ratio as per the instructions. The epoxy resin and hardener were taken into a plastic container and mixed for 2–3 min at room temperature with a plastic stirrer, until the mixture was uniform in color. Then, the solution was stirred for another 30 s to scrape the side and bottom of the container. After sufficient mixing of the hardener and epoxy, the resin solution was applied to fibers. The mechanical properties of epoxy resin and hardener are in the following Table 2.

Table 2. Mechanical properties of epoxy resin and hardener [31].

Property	Epoxy Resin	Hardener
Density (g/cm^3)	1.15–1.18	0.97–0.99
Specific gravity	1.14 ± 0.1	1.02 ± 0.1
Color	Colorless	Clear or brown
Viscosity at 25 °C (MPa)	550 ± 50	600
Pot life (min) at 23 °C	30 ± 10	30 ± 10
Mixing proportion	10	2
Gel time (h) at 23 °C	24–36	24–36

2.5. Preparation of Mould

Mold material used in this work was made from teak wood, as shown in Figure 2a. The dimensions of the mold are 150 mm × 150 mm × 12 mm. In general, matrix material sticks to the surface of the mold material. To avoid sticky nature, the mold is covered with baking paper, because it does not show any impact on mechanical properties. After placing the fibers inside the mold resin material applied to it and a roller was used, as shown in Figure 2b, to get the uniformity.

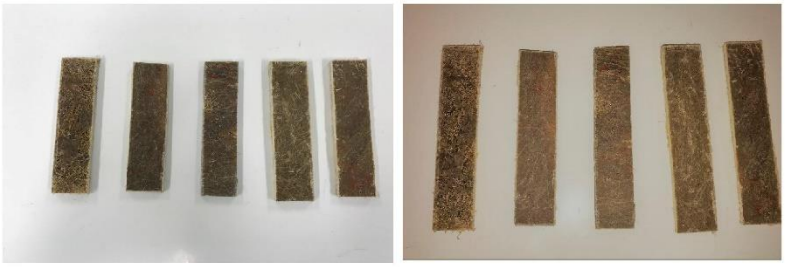


(a) (b)

Figure 2. Mold material for fabrication purpose (**a**); roller on the composite surface (**b**).

2.6. Composite Fabrication

Banana/pineapple, hemp/flax fiber hybrid composites were fabricated by using hand lay-up techniques. The mold materials were cleaned properly before placing the fibers in it. The mold was covered with baking paper and the fibers were placed in the form of beds, before applying the resin. After pouring epoxy resin in the mold material, it was compressed for a few minutes with a roller to spread the epoxy in all the corners of the mold. This avoids the gaps and formation of holes in the composite. A uniform load of 6KN was placed on the top of the composite surface and allowed for a curing time of 24–28 h. For each weight fraction, a total of five samples were prepared, to take the average values and ensure the specimens are cut as per ASTM standards. The fabricated samples of hemp/flax and banana/pineapple hybrid composites are shown in Figure 3a,b. The weight fractions of the composites are 40B/0P, 25B/15P, 20B/20P, 15B/25P, 0B/40P, 40H/0F, 25H/15F, 20H/20F, 15H/25F, and 0H/40F.



(**a**) (**b**)

Figure 3. Hemp and flax fiber hybrid composites (**a**); banana and pineapple fiber hybrid composites (**b**).

3. Testing of Composites

3.1. Flexural Strength Test

The flexural tests were performed to find the mechanical properties of the composite materials. This test is used to calculate the maximum stress and strain for the addition of external load. The specimens were tested on a Tinius Olsen H10K machine (Horsham, PA, USA) at a constant strain rate of 0.10 mm/min and speed as 20 mm/min. The force accuracy as 0.5% of applied load and 0.001 mm/min speed resolution. All the samples were tested at room temperature, and every time, five specimens were tested for each composite to get the average values. The specimens were tested as per ASTM D 790 standards. The dimensions of the samples are taken as 100 mm total length, 80 mm span length, 20 mm, and 5 mm thickness. The samples were allowed for three-point bending tests. The apparatus used for the flexural test is shown in Figure 4a. The strength of the composites was calculated by using the following relation.

$$\sigma_f = \frac{3PL}{2bd^2} \text{ MPa} \quad (1)$$

where σ_f is flexural strength, P is maximum load, L is length of the composites, b is width, and d is thickness.



(a) (b)

Figure 4. Three-point bending test for flexural properties on Tinius Olsen (**a**); interlaminar shear strength test on Tinus Olsen (**b**).

3.2. Flexural Modulus Test

The flexural modulus of composite material is defined as the ability for that composite to deform. It is calculated from the slope of the stress and displacement curve. It is also called a tangent modulus and modulus of elasticity. The following relation was used to find out the flexural modulus.

$$E_B = \frac{L^3 m}{4bd^3} \text{ MPa} \quad (2)$$

where E_B is flexural modulus, L is length, m is slope of stress-strain curve, b is width, and d is thickness.

3.3. Interlaminar Shear Strength

Shear tests of various types are widely used in the polymers industry to find the strength of the reinforcement to matrix materials. The interlaminar shear strength is the failure shear occurred in a composite material when the transverse load applied to it. The samples were taken for a notch cut near to the center on both sides of the specimen before going for the testing. All the specimens were allowed for testing on the Tinius Olsen H10K apparatus. The specimens were tested as per ASTM D-3846-02 standards. The dimensions of the samples are taken as 100 mm total length, 79.6 mm span length, 12.7 mm width, 5 mm thickness, 2.5 mm notch depth, and 6.4 mm distance between the notches. The following relation was used to calculate the shear strength of the composites. The apparatus used for this test is shown in Figure 4b.

$$\sigma_S = \frac{P}{A} \text{ MPa} \quad (3)$$

where P is maximum load and A is area.

3.4. Moisture Absorption Test

This test is used to find out the rate of water (moisture) absorbed by the composite when it is exposed to the wet medium. To evaluate the absorption rate, the composite sample is placed in a container full of distilled water. These samples were placed in an oven for 20 min at 60 °C for the heat

treatment process, before placing it in the container to eliminate the moisture in it. The specimens were tested as per ASTM D-570. The dimensions of the samples are taken as 76 mm length, 5 mm thickness, and 20 mm width. The % of weight gain by the composites were calculated in successive time intervals for five days. The following relation was used to calculate the moisture absorption of composite samples.

$$\% \, Weight \, gain = \frac{Final \, weight - Initial \, weight}{Initial \, weight} \times 100 \quad (4)$$

3.5. Contact Angle Measurement Test

The contact angle measurement technique is used to find out the wettability of the composite surfaces. The wetting property defines the ability of the liquid constituent to hold the interaction when it contacts the solid surface. It was found that the contact angle is an attractive method to observe the behavior of the liquid on a composite surface. It is used to find the hydrophobicity of the composite surfaces. The specimens were tested as per ASTM D-7334. The dimensions of the specimens are taken as 80 mm length, 20 mm width, and 5 mm thickness. A total of five samples were tested for each composite. In this method, a water droplet is placed on the composite surface with the help of a glass pipet. The volume of the water droplet kept constant for all the composites. The experimental setup used for contact angle measurement is shown in Figure 5, and it consists of an image processor, a moveable holder, camera, optical lenses, test sample holder, and a composite placed on a holder rest.

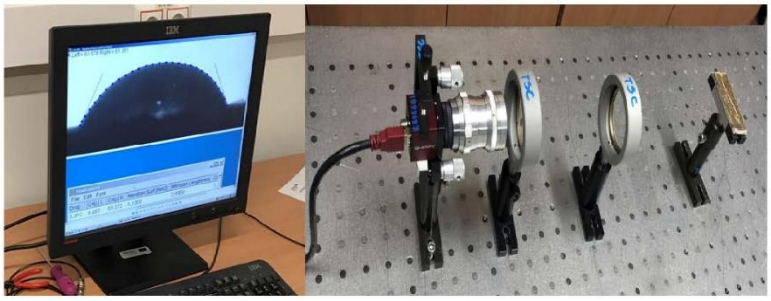


Figure 5. Contact angle measurement apparatus.

4. Results and Discussions

4.1. Flexural Properties

The analytical results for banana-pineapple and hemp-flax fabricated hybrid composites, obtained from flexural properties testing, are mentioned in the following Table 3.

Figure 6a,b show the relation between break load and weight fraction of various composites. In the first set of composites pure hemp (40H/0F) and pure flax (0H/40F) composites showed the least break load and hybrid composites showed improved break load. Additionally, 25H/15F showed the highest break load as 364.9 N at the breakpoint of the composite. In the second set of composites pure banana (40B/0P) and 20B/20P composites exhibited low break load as 135 N and 202.6 N, whereas 25B/15P showed the highest break load as 258.7 N. Hemp and flax fiber hybrid composites showed superior properties than banana and pineapple composites.

Table 3. Flexural strength properties.

Composites	Break Load (N)	Flexural Strength (MPa)	Flexural Modulus (GPa)
40H/0F	272.5	65.39	2.68
0H/40F	262.9	63.10	2.75
15H/25F	331.8	79.64	2.97
20H/20F	288.9	69.34	2.92
25H/15F	364.9	87.57	3.43
40B/0P	135.0	39.36	0.96
0B/40P	214.3	46.70	1.62
15B/25P	206.1	53.76	1.19
20B/20P	202.6	45.84	1.63
25B/15P	258.7	68.54	2.02

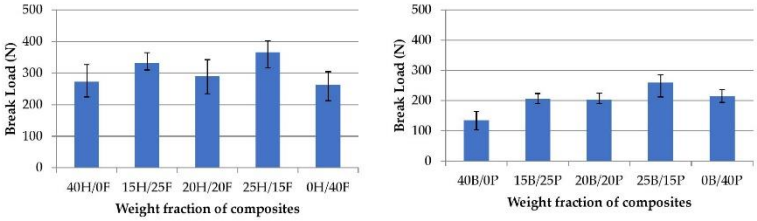


Figure 6. Break load vs. weight fraction of hemp/flax (H/)F composites (**a**) and banana/pineapple (B/P) composites (**b**).

The flexural properties of various weight fraction of composites are given in Figure 7a,b. Among all the composites, the 25H/15F hybrid composite has the highest flexural strength at 87.57 MPa, and pure banana (40B/0P) has the least strength at 39.36 MPa. Hemp and flax fiber composites exhibited a range between 63 and 88 MPa, whereas banana and pineapple composites showed in between 38 to 69 MPa. This is due to: (1) the hybridization impact for the fibers contributing superior flexural strength to the composites in both the cases; (2) the poor bonding between fiber reinforcement and matrix material in the pure composites leading to the agglomeration, hence, the loss, of strength, and fabrication errors might be the reason for lowering the strength in pure composites. The obtained results are proved that the hybridization of natural fibers gives superior results when compared to the individual fiber composites and natural fibers combined with synthetic fibers. It was proved from the previous results, giridharan worked on preparation and property evaluation of glass/ramie fibers reinforced epoxy hybrid composites [32]. The results were indicated that the glass and ramie hybrid composites have the flexural strength properties range of 50 and 65 MPa, which is less than hemp and flax hybrid composites. Singh et al. [33] investigated the tensile and flexural properties of hemp fiber reinforced virgin-recycled HDPE matrix composites. Hemp fiber composites were fabricated by varying the weight fraction of fiber and matrix material. The results showed that the hemp fiber composites have flexural properties range from 16 to 23 MPa, which is far less than the hemp/flax and banana/pineapple hybrid composites. Bazan et al. [34] investigated the mechanical, thermal, and ageing properties of bio-based polyethylene natural fiber composites. The results indicated that flexural properties were increased by increasing the fiber content and a similar trend followed up to 40% of fiber fraction and then decreased. It was noted that the properties are varied between 20 and 45 MPa. Sinha et al. [35] investigated the mechanical properties of natural fiber polymer composites. The results stated that coconut fiber epoxy composites showed flexural strength as 64.6 MPa. Raghuram et al. [36] worked on characteristics of treated natural

fiber epoxy composites with time-variant. The result stated that natural fiber epoxy-based composites showed flexural strength values between 33.35 and 70 MPa, which is better than the glass fiber blend results. Kumar [37] worked on a dataset on mechanical properties of natural fiber reinforced polyester composites for engineering applications. It was observed from the results that sisal, coconut coir fiber composites exhibited flexural strength ranging from 26.2 to 40.3 MPa. By considering the previous results, the hybridization of natural fibers has shown a significant impact on mechanical properties.

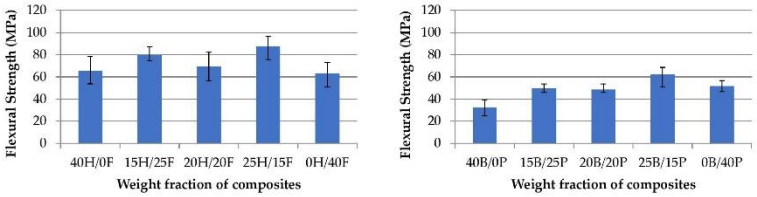


Figure 7. Flexural strength vs. weight fraction of H/F composites (**a**) and B/P composites (**b**).

Figure 8a,b show the trend of the flexural modulus of various composites. It was observed from the results that banana and pineapple composites showed lower flexural modulus than hemp and flax composites. Pure banana (40B/0P) has the least among all, and the 15H/25F composite has the highest flexural modulus at 2.97 GPa. All hybrid composites showed the flexural modulus close to each other in both the sets.

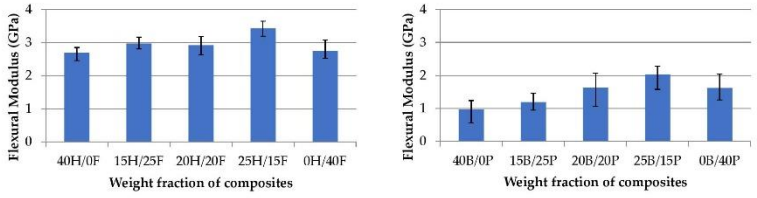


Figure 8. Flexural modulus vs. weight fraction of H/F composites (**a**) and B/P composites (**b**).

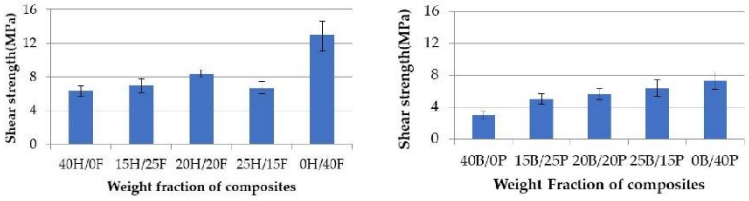
4.2. Interlaminar Shear Strength

The data presented in Table 4 show that the interlaminar shear strength properties for all the composites. The tests were performed to observe the failure shear of the composites. The results were considered when the failure shear occurred in between the notches.

Figure 9a,b show the interlaminar shear strength with respect to the weight fraction of all the composites. In both cases, pure composites showed the highest shear strength values and hybrid composites showed intermediate results. The pure hemp (40H/0F) composite showed the highest shear strength at 13.01 MPa, and the pure banana (40B/0P) composite showed the least shear strength at 3.03 MPa. The hybridization effect is negligible, because there is no such variation in results in both sets.

Table 4. Interlaminar shear strength properties.

Composites	Break Load (N)	Interlaminar Shear Strength (MPa)
40H/0F	513.29	6.32
0H/40F	1057.67	13.01
15H/25F	568.15	6.99
20H/20F	682.27	8.39
25H/15F	540.67	6.65
40B/0P	246.30	3.03
0B/40P	594.77	7.32
15B/25P	403.53	4.96
20B/20P	462.42	5.69
25B/15P	518.70	6.38



(a) (b)

Figure 9. Shear strength vs. weight fraction of H/F composites (**a**) and B/P composites (**b**).

4.3. Moisture Analysis

The amount of water absorbed by a composite material mainly depends on the fiber content, immersion temperature, area of contact to water, and void content. The results were calculated at regular intervals of time for five days.

Figures 10 and 11 show the percentage of weight gain in all fabricated composites. It was observed from the results that the percentage of weight gain in the composite increases with the increase of time. The increase rate remains constant after 96 h of time in both cases, which means that the composite reaches an equilibrium state after 96 h. Pure hemp and pure banana composites showed the highest and pure flax and pure pineapple the lowest values of absorption rate, whereas hybrid composites showed intermediate results. This can be attributed to: (1) the presence of hydroxyl groups in the cellulose structure, which attracts more water and binds the cells through hydrogen bonding; (2) the void content (porosity), which also attracts water uptake in the composite due to the fabrication errors.

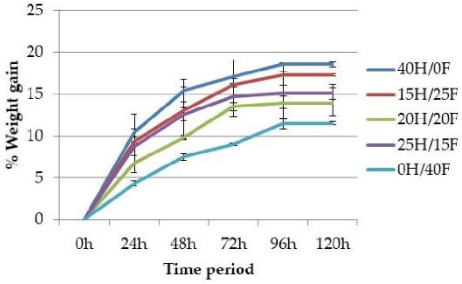


Figure 10. Percentage weight gain vs. time period for hemp and flax composites.

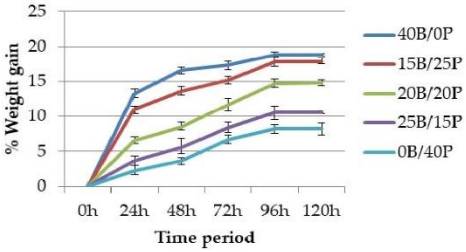


Figure 11. Percentage weight gain vs. time period for banana and pineapple composites.

4.4. Contact Angle Measurement

The wettability properties or hydrophilic nature of the composites are calculated by using the contact angle measure analysis. The contact angle was measured with the image processing apparatus as shown in Figure 5. All the values are measured (recorded) in a dark place and for each composite five samples were tested. The contact angle of the composites was measured and with deviation in each value presented in Figure 12a,b.

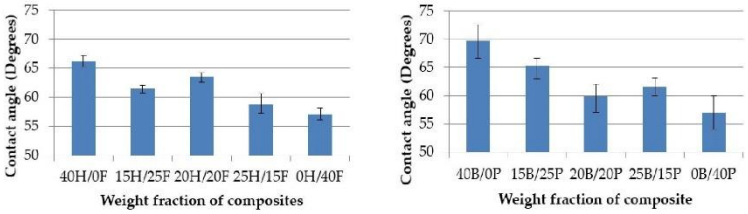


Figure 12. Contact angle vs. weight fraction of H/F composites (**a**) and B/P composites (**b**).

In general, the highest contact angle is recorded on rough composite surfaces, and the lowest is recorded on smooth composite surfaces. If the contact angle is less than 90°, then the composite has a hydrophilic nature. If it is more than 90°, then the composite has hydrophobic nature. It was found that all the fabricated composites showed a contact angle of lower than 90°, which means that composites have hydrophilic nature. Hemp and flax fiber hybrid composites exhibited a contact angle between 58° to 70°. Banana and pineapple composites showed between 55° and 75°. Hybrid composites showed an intermediate contact angle in both cases, and pure composites showed the highest contact angle and the lowest contact angle. The is due to both banana and pineapple fibers being cellulose rich fibers, and hybrid composites having a smooth surface finish. From the overall results, it was clear that the hemp and flax fiber composites have a smoother surface finish than the banana and pineapple composites.

5. Conclusions

Natural fiber reinforced epoxy resin hybrid fiber composites of varying weight fractions were fabricated and characterized. Two sets of composites (H/F and B/P) were fabricated based on the rule of hybridization mixtures, by using the hand lay-up technique for comparison purposes. Hybrid composites showed improved mechanical properties compared to pure composites in both cases. From the mechanical properties testing of composites in the first set (hemp and flax), it was found

that the hybrid composites showed higher flexural properties than pure hemp and pure flax composites. The 25H/15F composite showed a flexural strength and flexural modulus at 87.57 MPa and 3.43 Gpa, respectively. The pure flax composite showed the least flexural strength and modulus at 63.10 MPa and 2.75 GPa, respectively. In the second set (banana and pineapple) of composites, it was found that the 25B/15P composite showed the highest flexural strength and modulus at 68.54 MPa and 2.02 GPa, respectively. The 40B/0P composite showed the least flexural strength and modulus at 39.36 MPa and 0.96 GPa, respectively. In both cases, hybrid composites showed the intermediate interlaminar shear strength, and pure composites showed the highest and lowest values. From contact angle measurement analysis, all the composites showed a contact angle at less than 90°, which means composites are exhibiting hydrophilic surface properties. There is no such deviation in the contact angle results among all composites, but pure composites showed the highest contact angle in both cases, which means pure composites have a rough surface finish. From the moisture analysis results, it was found that all the composites absorbed water when it was placed in a water container. The percentage weight gain of the composites remained constant after 96 h in both cases, which means that the composites have reached an equilibrium position. Hybrid composites absorbed less water than pure composites in both cases. By considering the overall results, the hybrid composites showed improved properties compared to pure composites, and hemp and flax fiber composites have superior properties than banana and pineapple composites. Hemp and flax fibers are a potential replacement for reinforcements in the composites. Hemp and flax fibers can be used for structural applications.

Author Contributions: Conceptualization, A.A.; methodology, A.A. and A.P.; investigation, A.A. and M.S.; calculation and analysis, A.V. and M.S.; writing—original draft preparation, A.A.; writing—review and editing, G.J. and A.P.; visualization, A.A.; validation, G.J. All authors have read and agreed to the published version of the manuscript.

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Investigation of Hemp and Flax Fiber-Reinforced EcoPoxy Matrix Biocomposites: Morphological, Mechanical, and Hydrophilic Properties

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Abstract: Modern day industries are highly focused on the development of bio-inspired hybrid natural fiber composites for lightweight biosensor chips, automobile, and microfluidic applications. In the present research, the mechanical properties and morphological characteristics of alkaline (NaOH)-treated hemp, flax, noil hemp, and noil flax fiber-reinforced ecopoxy biocomposites were investigated. The samples were fabricated by employing the hand layup technique followed by the compression molding process. A total of two sets of composites with various weight fractions were fabricated. The samples were tested for mechanical properties such as flexural strength, interlaminar shear strength, moisture absorption, and contact angle measurement. The treated fibers were analyzed by using an optical microscope and Fourier transform infrared spectrometer (FTIR). The morphological characteristics, such as porosity and fracture mechanisms, were investigated by using scanning electron microscopy and SEM−EDX spectroscopy. The results revealed that the flexural properties of hybrid composites vary from 22.62 MPa to 30.04 MPa for hemp and flax fibers and 21.86 MPa to 24.70 MPa for noil fibers, whereas in individual fiber composites, the strength varies from 17.11 MPa to 21.54 MPa for hemp and flax fibers and 15.83 MPa to 18.79 MPa for noil fibers. A similar trend was observed in interlaminar shear properties in both cases. From moisture analysis, the rate of absorption is increased with time up to 144 h and remains constant in both cases. The moisture gain was observed more in individual composites than hybrid composites in both cases. Hence, the impact of hybridization was observed clearly in both cases. Also, hybrid composites showed improved properties compared to individual fiber composites.

Keywords: hemp and flax fibers; ecopoxy; mechanical properties; FTIR; morphology; SEM−EDX spectrometer

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1. Introduction

In recent years, multiple efforts have been made to produce new, creative, and environmentally friendly materials for diverse purposes due to increasing ecological and environmental consciousness [1]. Natural fibers are being investigated as reinforcements in polymer composites due to the expensive cost of synthetic fibers and their dangerous environmental impact. Natural fiber-based composites offer an environmental benefit by reducing reliance on petroleum-based resources in today's technologies [2,3]. Biocomposites are composites made up of natural fibers including hemp, flax, sisal, Carlota, banana, pineapple, palm, kenaf, bamboo, jute, coir, and sugarcane combined with biodegradable or non-biodegradable polymers [4,5]. Many researchers are paying attention to the natural fiber-reinforced composites due to their abundance, cost-effectiveness, eco-friendly nature, and lightweight applications. Further applications of natural fiber composites are automobile industries, marine, sports equipment, construction, aerospace, microfluidic [6–8], and energy harvesting applications [9].

Hemp and flax fibers are renewable resources with a promising mechanical property that can be used as a reinforcement in composites in a wide range of applications. Many research studies have been conducted to determine the effect of the fiber content and orientation of fibers on the mechanical properties of various natural fiber-reinforced polymer (thermoplastic and thermosetting) composites [10–14]. The importance of fabrication techniques on composite materials has also been studied [15]. In general, increasing the fiber content enhances the flexural strength, torsional strength, and tensile strength of natural fiber composites up to a point, after which they rapidly decline. However, due to weak interfacial adhesion between the reinforcement and matrix material, the composite mechanical properties may be significantly below their optimum values. Therefore, the researchers are finding ways to improve the bonding between the reinforcement and matrix material by employing various chemical treatment techniques [16,17]. The chemical treatments can modify the interface by interacting with both fiber and resin materials on the composite surface. Among the studied chemical treatment processes, alkaline treatment is the most commonly used coupling agent to boost the bonding between the fiber and matrix material [18].

The hybridization of composites influences the advancement of material properties and this has been proven in recent years. Many studies have been focusing on improving the material properties by creating new combinations such as joining two different natural fibers, joining natural fibers with synthetic fiber, and joining two different synthetic fibers in the same matrix material. These composites have been investigated in order to ensure the improvement of mechanical properties over single fiber composites [19,20]. There are several types of hybrid composites that exist by varying the size, filler material, and orientation of fibers. The hybridization may overcome the drawbacks of one component by including another type of fiber material. In general, the hybrid effect varies with the fiber's mechanical and chemical properties; thus, the effect is less when compared to the individual fiber composites [21,22]. Hybrid reinforcement with decent fiber selection is possible to provide striking properties and meet the present requirement for thermosetting matrix composites [23,24]. In the last few decades, many studies on the hybridization of synthetic fibers and plant (leaf, bast, seed) fibers have been investigated. However, natural fiber-reinforced hybrid composites are less investigated, and the recent studies mainly focused on the combination of natural fiber with artificial fiber. The best example of such composites includes the use of carbon, glass, and aramid in combination with hemp, flax, banana, and pineapple [25,26]. However, with the increasing use of synthetic polymer materials around the world, environmental problems such as waste treatment, waste transfer management, and burning contamination are becoming increasingly important. It shows the adverse effect on environmental and climate change issues [27].

Though there are many advantages to natural fiber hybrid composites, there are some drawbacks, such as the durability and moisture absorption behavior weakening the strength of the composite material. The endurance of the natural fiber composites at the structural lamination scale for their use in load-bearing applications is still unknown. While flax and jute composite columns have been shown in recent studies by M.R. Bambach [28] to have theoretically appropriate structural qualities for light, primary structural applications, such as residential and light commercial structures, flax and jute composite columns are not yet widely used. These tests showed that the start of compression buckling of the narrow cross-sectional elements and the post-buckling redistribution of stresses that result in the final limit state (material failure) dominate structural behavior. The presence of moisture in composite material leads to a weakening of the mechanical performance. Ramakrishnan et al. [29] studied the effect of moisture content on natural fiber-reinforced polymer composites. The experimental results stated that the rate of moisture absorption is less in hybrid composites than in individual fiber composites.

Many researchers were interested in working with natural fibers due to their abundance and ease of usage. Ramesh et al. [30] investigated the mechanical and thermal properties of banana−pineapple fiber-reinforced epoxy composites. The results stated that

the hybrid composites made up of banana and pineapple have shown improved mechanical properties and also stated that they can be extended to light load conditions for automotive and construction applications. The flame retardancy properties were analyzed by using thermogravimetric analysis but the results were not up to the mark, which can be achieved by using a proper chemical treatment process. Zalinawati et al. [31] studied the effect of fiber treatment on the mechanical properties of burl palm fiber epoxy composites. In their studies, they treated the burl palm fiber with sodium hydroxide solution and then conducted the fabrication process by using a hand layup technique. The results are stated that the treated fiber composites showed superior mechanical properties to the untreated fiber composites. It was observed that thickness swelling is greater in untreated fiber composites due to greater absorption of water when it is exposed to the wet medium. Torres-Arellano et al. [32] worked on the mechanical properties of natural fiber-reinforced bio-based epoxy resins. This work dealt with the natural fiber-reinforced bio-based epoxy resin composites. The composites were fabricated by using a vacuum-assisted resin infusion process. The results showed that the presence of bio-based epoxy resins in the composite material enhanced the mechanical properties and among all the fibers, the jute fibers showed superior properties to the others. Jamshaid et al. [33] investigated the mechanical and electrical properties of jute, sisal, coconut, and sugarcane natural fiber-reinforced bisphenol F epoxy-based biocomposites. The results stated that among all the natural fiber composites, the sisal/jute hybrid composites exhibited superior mechanical properties compared to the coconut/sugarcane composites. The results from the fabricated samples demonstrate that natural fiber biocomposites could open a new path for their application in electrical goods and as an environmentally friendly alternative. Rajak et al. [34] studied the flexural mechanical properties of natural fiber-reinforced polymer composites and their applications. In this overview, the authors tried to explain the advantage of natural fiber-reinforced composites and their various applications. Terwadkar et al. [35] investigated the mechanical properties of banana and kenaf fiber-reinforced epoxy hybrid composites. In this paper, the effect of hybridization was evaluated by comparing the treated fibers and untreated fibers. The alkaline treatment showed a significant impact on the mechanical properties and hybrid composites showed superior properties. Neves et al. [36] drew the comparison between the biocomposites of epoxy and polyester composites. An attempt has been made on hemp fiber composites by varying their weight percentages such as 10, 20, and 30%. It was observed that the increase in mechanical properties increased the fiber content to some extent. Epoxy-based biocomposites exhibited improved mechanical properties compared to polyester composites. Senthil et al. [37] investigated the mechanical properties of hemp and sisal fiber-based biocomposites. The experimental results specify that hybrid composites displayed a small variation in mechanical properties when the stacking sequence was altered. The hybrid composite with the intercalated arrangement showed the highest tensile modulus when compared with the other hybrid counterparts. Typical failure characteristics of the short beam test, such as inter-laminar shear cracks in the transverse direction, micro-buckling, and fiber rupture, were also observed. It clearly states the lack of chemical treatment of the reinforcements. Bazan et al. [38] worked on bio-based polyethylene composites with flax, basalt, and coconut fibers in the presence of wood flour particles. Results indicated the positive impact on biopolyethylene composites. Among these natural fibers, basalt fiber composites exhibited the greatest strengthening properties. By reviewing these literature studies, we can conclude that natural fibers, which are alkaline-treated, exhibited better mechanical properties by improving the adhesion between the reinforcement and matrix material. Though epoxy resin materials are easy to use (flexible) in composite fabrication, there are a few drawbacks to it such as non-biodegradability, chemical haggardness, and also not being skin-friendly. To overcome these problems, an attempt has been made in the present research based on the bio-inspired resin materials.

This research work presents the new composites based on ecopoxy (bio epoxy) with hemp, flax, noil hemp, and noil flax natural fibers. A total of two sets (Hemp/Flax, Noil

hemp/Noil flax) of hybrid composites with various weight fractions were developed based on the hybridization concept. The composites were fabricated by using the hand layup technique, followed by the compression molding technique. These composites were tested for material properties such as contact angle, moisture absorption, and mechanical properties. The failure mechanism was studied by using a scanning electron microscope. The surface properties were investigated by using the optical microscope. The mechanical properties were investigated and then compared. Also, we aimed to understand whether noil fibers (primary waste after processing) are able to serve as a replacement for the normal fibers. Thus, the obtained results showed the significance of appropriately constituted hybrid biocomposites with natural fibers in the presence of ecopoxy matrix material, holding good quality, decent mechanical, adhesion, and surface properties.

Research Significance

Due to the increased need for engineering applications of thermoplastic and thermoset polymer materials that outperform metals in a variety of applications, hybrid biocomposites with enhanced characteristics have emerged through the addition of diverse reinforcements. The significance of natural, inexpensive, and abundant materials is highlighted by rising environmental regulations and consumer awareness. Due to the tendency of reinforcement and matrix interactions to increase both the flexibility and rigidity of the natural fiber in one step, the hybridization of one natural fiber with another natural fiber is surprisingly favorable. The change in fiber characteristics and polymer composition in biocomposites reinforced with natural fibers showed the most encouraging results. Natural fiber hybrid biocomposites have garnered great attention since their discovery due to their wide variety of properties in food packaging, biomedical devices, automotive industries, and other consumer applications with better thermal, physical, mechanical, optical, and barrier properties. The presence of moisture and the poor surface properties of natural fiber severely affects the mechanical performance of composite material. Hence, the present article is devoted to modifying the hemp and flax natural fiber surface properties by using the alkaline treatment and heat treatment processes. An ecopoxy matrix, along with hardener material, was introduced to fabricate the hybrid composites. The ecopoxy matrix material contains 36% bioactive content (information from the manufacturer), whereas in traditional epoxy, the bioactive content is less than 8%, which is toxic to the human skin as well as the environment. The excess moisture presented in biocomposites was eliminated by using the post-curing heat treatment process. Also, to minimize the waste material during the fiber processing, the current research work planned to understand the material properties by using noil (primary waste) fibers as well as normal fibers. The fabricated samples allowed for mechanical property evaluation, after which the results were compared.

2. Materials and Methods

2.1. Materials

Hemp (H), flax (F), noil hemp (NH), and noil flax (NF) fibers were used as reinforcements in the fabrication of composites. These fibers (hemp and flax) were extracted manually from the plants. Noil fibers are processed fibers (primary wastage) from the normal fibers. NaOH solution were used for the purpose of chemical treatment, and this was purchased from Sigma Aldrich (St. Louis, MO, USA). The matrix material used here was ecopoxy (bio-inspired resin material having a bioactivity of 36%), along with hardener (acts as a cross-linking agent), which were purchased from Ecopoxy, (Tychy, Poland). The mechanical properties and chemical characteristics of the individual fibers are given in the following Table 1 and the mechanical properties of ecopoxy resin and hardener is given in Table 2.

Table 1. Hemp and flax fiber chemical composition and mechanical properties [39,40].

Properties	Hemp	Flax
Cellulose (%)	70–74	64–70
Hemicellulose (%)	21–24	16–18
Lignin (%)	3.7–5.7	2–2.2
Pectin (%)	7.3	2.3
Elongation (%)	1.6–4.0	1.2–1.6
Moisture (%)	6.2–12	8–12
Density (g/cm^3)	1.48	1.4
Young's modulus (GPa)	70	60–80
Microfibrillar angle (°)	2–6	5–8
Tensile strength (MPa)	550–900	800–1500
Specific strength (s/g)	393–643	570–1070

Table 2. Liquid ecopoxy resin and hardener material properties.

Properties	Ecopoxy	Hardener
Color	Viscous liquid	Clear
Viscosity at 25 °C	800 = 1000 cP	100–200 cP
Specific gravity	1.1	1.0
Bio content	36%	-
Mix ratio	2	1
Gel time at 25 °C	20–25 min	20–25 min
Full cure at 25 °C	72 h	72 h

2.2. Extraction and Chemical Treatment of Natural Fibers

One of the most important aspects of the study was the fiber selection and extraction, followed by the chemical treatment. Hemp fibers were extracted from the cannabis sativa plant. Flax fibers were extracted from the bast underneath the exterior of the stem of the flax plant. Noil fibers were obtained from the primary wasted after the processing of raw fibers. All the extracted fibers were cleaned with distilled water and dried to eliminate the moisture after being recovered from the resources. The fibers were then permitted to undergo an alkaline treatment process. This improves the bonding between the matrix and reinforcement. For the chemical treatment process, a 5% NaOH solution was prepared, added to the fibers separately and kept for the heat treatment process (80° temperature for 4 h). Once the heat treatment process was completed, the fibers were cleaned with water again to maintain the pH level in them and allow for drying. The dried fibers were separated using a cotton comb and a hand-sitting process. To maintain uniformity, all the fibers were cut into fine pieces (2–3 mm length).

2.3. Materials Weight Percentage

The weight fraction of both fiber and resin material was considered as per the hybridization concept. In the current research work, a total of two sets of hybrid biocomposites were fabricated based on hemp, flax, noil hemp, and noil flax natural fibers. The hybridization states that the composite material contains more than one reinforcement under the same matrix material with a 0.4 wf.%. The theoretical calculations of weight and volume fractions of both the reinforcement and matrix material are given in the following equations [41–43].

The weight fraction of the fiber (reinforcement) material is defined as

$$W_f = \frac{Weight\ of\ fibers}{Total\ weight} \quad (1)$$

Then, the weight fraction of the resin (matrix) material is defined as

$$W_m = \frac{Weight\ of\ matrix}{Total\ weight} \quad (2)$$

Similarly,

$$W_m + W_f = 1 \tag{3}$$

The volume fraction of the fiber (reinforcement) material is given as

$$V_f = \frac{Volume\ of\ fibers}{Total\ Volume} \tag{4}$$

Then, the volume fraction of the resin (matrix) material is defined as

$$V_m = \frac{Volume\ of\ resin}{Total\ Volume} \tag{5}$$

Similar to the previous equation,

$$V_m + V_f = 1 \tag{6}$$

It should be noted that one cannot change the weight fraction completely over to the volume fraction or the other way around. The densities of the fiber (ρ_f) and matrix (ρ_m) materials are the definite weight of the fiber and matrix individually. Then the equation can be written as follows:

$$V_f = \frac{\frac{W_f}{\rho_f}}{\frac{W_f}{\rho_f} + \frac{W_m}{\rho_m}} \tag{7}$$

From Equation (7), the weight fraction of fiber is written as

$$W_f = \frac{V_f \rho_f}{V_f \rho_f + V_m \rho_m} \tag{8}$$

The weight proportions are given in Table 3.

Table 3. Composite weight fractions.

Composites	Hemp Fiber (%)	Flax Fiber (%)	Total Fiber Vol. (%)	Total Resin Vol. (%)
40H/0F	40	0	40	60
25H/15F	25	15	40	60
20H/20F	20	20	40	60
15H/25F	15	25	40	60
0H/40F	0	40	40	60
	Noil Hemp Fiber (%)	Noil Flax Fiber (%)		
40NH/0NF	40	0	40	60
25NH/15NF	25	15	40	60
20NH/20NF	20	20	40	60
15NH/25NF	15	25	40	60
0NH/40NF	0	40	40	60

2.4. Fabrication of Composites

Hemp/flax and noil hemp/noil flax natural fiber-reinforced hybrid composites were fabricated by using the hand layup technique, followed by a compression molding process. The fiber materials were measured as per the weight fractions mentioned in the previous Table 3 and then placed in a mold material. The molds were covered with non-stick paper to avoid direct contact between the resin and mold material. After placing the fibers in the mold material, the ecopoxy resin solution was poured into it, a roller was applied on the fibers to get the uniform distribution, and then it was compressed with constant weight and left to undergo the curing process. The entire setup lasted for 72 h for the complete curing process. The biocomposite panels were allowed for the post-curing process after fabrication. For the post-curing process, composite panels were placed in an oven at 70 °C for 6 h. After heat treatment, the samples were cut from the panels as per ASTM standards.

2.5. Testing Methods

2.5.1. Interlaminar Shear Properties

The interlaminar shear strength of composite material is the failure shear that occurs when a transverse force is applied to it. To measure the shear strength, all the samples were allowed a notch cut 6.4 mm from the center and half of the thickness of the composite samples before testing. Tinus Olsen H10KT (Horsham, PA, USA) was used to test the samples. The dimensions of the tested samples are considered as per ASTM D-3846 standards. The interlaminar shear strength of the fabricated composites is calculated by using the following equation where P is the maximum load and A is the failure shear area.

$$\sigma_S = \frac{P}{A} \text{MPa} \tag{9}$$

2.5.2. Flexural Strength Properties

A three-point bending test was performed on Tinus Olsen H10K to find out the flexural strength of the composites. All samples were tested as per ASTM D-790 standards. The following relation is used to calculate the flexural strength values where P is the maximum load, L is the span length, b is width, and d is the thickness of the composite sample:

$$\sigma_f = \frac{3PL}{2bd^2} \text{ MPa} \tag{10}$$

The flexural modulus is computed by dividing the change in stress by the corresponding change in strain and is represented by the slope of the initial straight line segment of the stress−strain curve. As a result, the flexural modulus is defined as the ratio of stress to strain. The flexural modulus of composites is calculated by using the following relation where L is the length, m is the slope of the stress−strain curve, b is the width, and d is the thickness of the composite.

$$E_B = \frac{L^3 m}{4bd^3} \text{ MPa} \tag{11}$$

2.5.3. Hydrophilic Properties

The wettability of the composite surface is measured by using the contact angle measurement analysis. It is also possible to investigate the wetting of and dynamic contact angles on composite samples that are used as the test body. The main objective of this test is to determine the hydrophobicity of the composite material. The schematic representation of the contact angle on the composite surface is shown in Figure 1 and the test specimen standards were considered as per ASTM D-7334 standards.

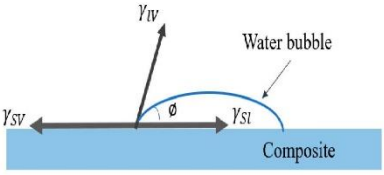


Figure 1. Contact angle formed on composite surface with interfacial tensions of the three phases.

2.5.4. Moisture Absorption

The rate of absorption of the composite samples was calculated by using the moisture analysis test. It is necessary to find out the rate of absorption when the composite sample is exposed to the moisture content. To evaluate the rate of absorption, all the samples were placed in a glass container full of distilled water. The rate is calculated by measuring the initial weight and final weight (after removal from the water) of the composite samples. The specimen standards were followed from the ASTM D-570. The amount of water absorbed

by the composites depends on the percentage of cellulose and the presence of porosity content. The following equation is used to calculate the absorption rate:

$$Rate\ of\ Absorption = \frac{Final\ weight - Initial\ weight}{Initil\ weight} \times 100 \quad (12)$$

2.5.5. Optical Microscopic Analysis

Fiber surface modification is often done by physical or chemical means to increase reinforcement−matrix adhesion. This results in a reduction in moisture gain as well as changes in the fiber surface. Changes in the fiber's surface morphology are critical to address its interaction with the polymer matrix in the composite materials. The effect of chemical treatment on the natural fibers is analyzed by using the optical microscope.

2.5.6. FTIR Analysis

The surface functional group analysis was performed using Fourier transform infrared spectrometer (FTIR, Bruker tensor 27, Borken, Germany). For the FTIR analysis, samples were first dried at 105 °C for 24 h according to the ISO 579 standard. After the preparation, the sample was placed on a horizontal ZnSe crystal plate with a 45° incidence angle and 10 internal reflections, pressed with a fixator to avoid any gap between the plate and sample. A background reference spectra was obtained before every experiment to eliminate the influence of the atmosphere. The scanning time of every wavelength is 32 times, while the wavelength interval is from 650 to 4000 cm^{-1}. The resolution of the detector is 4 cm^{-1}.

2.5.7. SEM and EDX Analysis

The morphological studies, biocomposite failure mechanisms, and chemical compositions of the biocomposites were investigated by using the SEM (scanning electron microscope) from Hitachi along with an EDX (energy dispersive X-ray) spectrometer from Bruker, Billerica, MA, USA.

3. Results and Discussion

3.1. Interlaminar Shear Strength Test

The interlaminar shear strength properties of ecopoxy-based natural fiber hybrid biocomposites were investigated and the results are presented in the following graphs. For each sample, a total of five composites were tested and the dimensions of a rectangular cross-section composite were taken as 79.6 × 12.7 × 4.6 mm in length, width, and thickness. Notch depth is taken as half of the specimen's thickness. All the samples were tested at room temperature and a constant relative humidity (26%) because it was observed from the literature that shear strength decreases when the humidity increases. Ali et al. [44] investigated the mechanical properties of bamboo fiber composites for structural applications. The results stated that the shear strength was decreased when the humidity increased from 60% to 90%.

Figure 2a,b shows the interlaminar shear strength properties of the hemp−flax, noil hemp−noil flax natural fiber hybrid biocomposites. As can be seen from the obtained results, the shear strength values of hybrid biocomposites are superior to the pure fiber (individual fiber) biocomposites in both cases. Natural hemp and flax fiber biocomposites showed better shear strength properties than noil fiber biocomposites. In the first set (H/F) of biocomposites, 25H/15F hybrid biocomposites showed the highest shear strength with 13.10 MPa, and pure hemp (40H/0F) showed the least with 5.67 MPa. To contrast, in the second set (NH/NF) of biocomposites, 25NH/15NF composites showed the highest shear strength with 9.03 MPa and pure noil flax (0NH/40NF) showed the least with 5.43 MPa. Overall, hemp fiber-rich hybrid biocomposites showed superior properties in both cases. The reasons for this can be attributed to the poor bonding between the fiber reinforcement and matrix material in pure composites, which causes an agglomeration, a loss of strength, and possible fabrication faults, whereas in hybrid composites the hybridization impacts the fibers, contributing superior flexural strength to the composites in both circumstances.

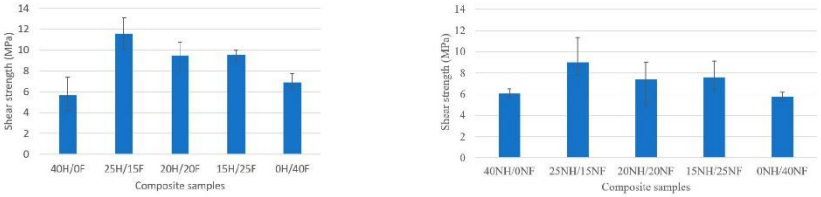


Figure 2. Interlaminar shear strength behavior of (**a**) H/F and (**b**) NH/NF natural fiber hybrid biocomposites.

3.2. Flexural Strength Test

The flexural strength properties of ecopoxy-based natural fiber hybrid biocomposites were investigated and the results are presented in the following graphs. A 3-point bending test was performed on Tinus Olsen at 21° and 28% relative humidity to calculate the bending strength of biocomposites. For each sample, a total of five composites were tested and the dimensions were taken as 80 × 20 × 4.6 mm in length, width, and thickness as per ASTM standards and the samples were taken in the form of a rectangular cross-section.

The flexural strength properties of natural fiber hybrid biocomposites are given in Figure 3a,b. In both cases, pure composites showed the least flexural strength values, whereas hybrid composites showed superior properties and minimal variation in the outcomes. Hemp/flax fiber hybrid biocomposites showed a range between 17 MPa and 30 MPa, whereas noil fiber biocomposites exhibited a range between 13 MPa and 24 MPa. This is due to the fiber's hybridization effect, which gives the biocomposites higher flexural strength in both circumstances. Poor bonding between the reinforcement and resin material in pure biocomposites results in the agglomeration and, hence, a loss of strength and mechanical performance could be the cause of pure biocomposites' reduced strength. Also, it is evident from the SEM analysis that the presence of internal defects such as matrix cracks, broken fibers, and voids may weaken the strength of a composite material. It was observed from the results that the presence of internal defects is reduced in hybrid composites compared to individual composites in both cases. The hybridization impact was observed clearly in both cases in terms of increasing the strength. A similar trend in the results was witnessed by various researchers investigating the flexural strength of natural fibers [45,46]. In most cases, the flexural strength increases with an increase in the fiber content up to 40%, after which it starts decreasing.

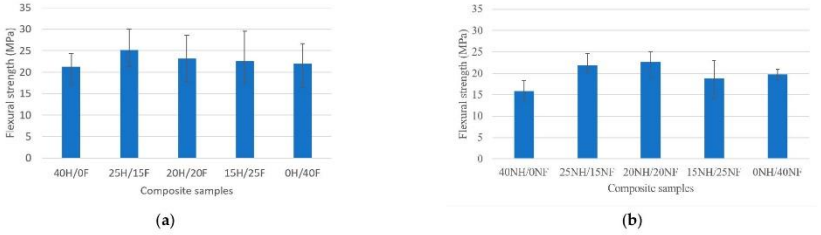


Figure 3. Flexural strength behavior of (**a**) H/F and (**b**) NH/NF natural fiber hybrid biocomposites.

Flexural Modulus

The results presented in Figure 4a,b show the assessment of the flexural modulus properties of various hybrid fiber biocomposites. The results are followed a similar trend to the previous results. The hemp and flax fiber hybrid biocomposites showed higher flexural modulus properties than noil hemp and noil flax fiber hybrid biocomposites. Pure noil

hemp fiber biocomposites showed the least flexural modulus with 0.261 GPa, whereas 25H/15F showed the highest with 0.825 GPa. There is not much variation in the hemp and flax hybrid composites as the values are close to each other, whereas in noil fiber biocomposites, the variation is quite observable.

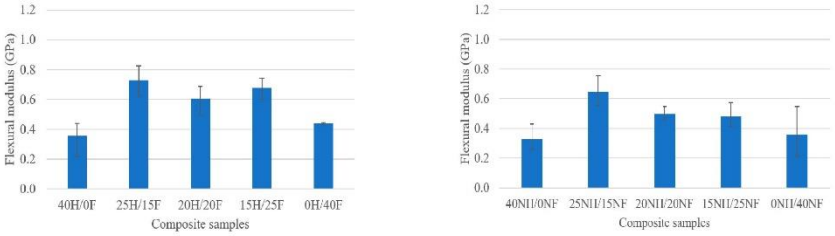


Figure 4. Flexural modulus of (**a**) H/F and (**b**) NH/NF natural fiber hybrid biocomposites.

3.3. Contact Angle Measurement Analysis

The contact angle measurement illustrations of the biocomposites are shown in Figure 5a,b. The lighting has been meticulously planned to eliminate any light reflections that may detract from the measurement. Precautions were also taken to keep the drips from becoming contaminated by air pollutants such as dust and particulates.

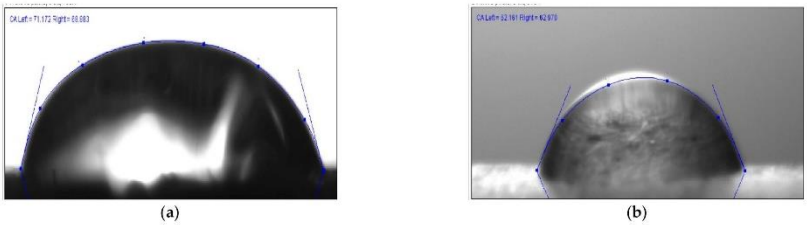


Figure 5. Contact angle image illustration for (**a**) H/F and (**b**) NH/NF biocomposites.

The graphical representation of measured contact angles for various natural fiber hybrid biocomposites is shown in Figure 6a,b. From the obtained results, the contact angle maximum (87.3°) was observed for the 20NH/20NF hybrid composite and the lowest (53°) was observed for the 40H/0F. Hemp and flax biocomposites showed the contact angle ranging from 53° to 66° and noil fiber composites exhibited angles between 61° and 87°. According to the literature [47,48], a material is well-thought-out as a hydrophilic material if the contact angle is <90° and if it is >90°, it is well-thought-out as a hydrophobic material. This means that all the composites showed hydrophilic surface properties, whereas the 20NH/20NF hybrid fiber composites almost fall in the range of hydrophobic material properties. Furthermore, the contact angle was investigated after 15 s, 30 s, and 45 s for all the biocomposites with distilled water to observe the spreading of water droplets on the biocomposites' surface. A study decrement was observed for the water contact angle for all biocomposites.

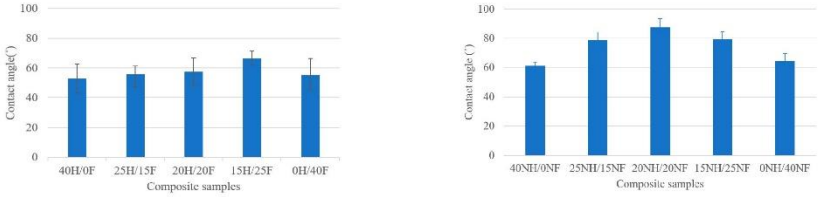


Figure 6. Contact angle measurement for (**a**) H/F and (**b**) NH/NF natural fiber biocomposites.

3.4. Moisture Analysis

The absorption rate for the fabricated composites was calculated by using the moisture absorption test. The absorption rate was calculated for 7 continuous days and the values were noted for 24 h intervals. All the biocomposites were placed in an oven for 30 min before conducting the test. This test was performed at 18° and 26% humidity in the environment.

Figures 7 and 8 below show the absorption rate for hemp/flax and noil hemp/flax fiber hybrid biocomposites. The results state that the absorption rate for the composition increased with time in both cases. A similar trend was observed in both cases. Pure composites showed the highest rate of absorption, whereas hybrid composites showed the lowest rate, with the values being close to each other. The reasons for this can be attributed to the presence of voids (porosity content) in the pure composites and the presence of hydroxyl groups in the pure composites, leading to the attraction of more water content. Overall, hemp and flax fiber biocomposites showed a lower rate of absorption than noil biocomposites.

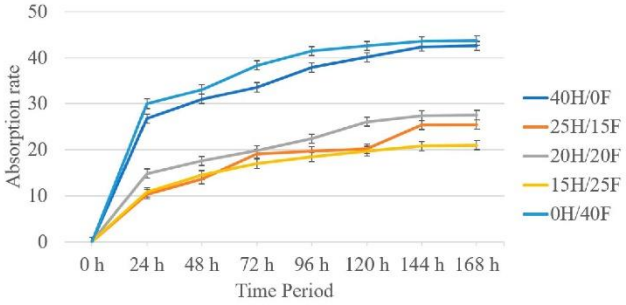


Figure 7. Time period vs. absorption rate for H/F hybrid biocomposites.

3.5. Fiber Morphology Studies

The fibers' surface morphology and the effect of chemical treatment on the natural fibers used in this research work are analyzed by using an optical microscope. The following figures differentiate the treated and untreated natural fibers.

The surface morphology of the fibers was studied using an optical microscope. The microscopic images of the fiber surfaces are presented in Figures 9 and 10. Figure 9a,b show the optical microscope images of untreated hemp and flax fibers, which have a smooth surface finish. This is due to the presence of adhesive polysaccharides on the surface of fibers. In contrast, Figure 10a,b show the images of treated hemp and flax fibers, which have a rough surface finish. This is due to the alkaline (NaOH) treatment of natural fibers. The polysaccharides were removed from the surface of the fibers and fibers were also

separated from each other. The roughness on the fibers improves the adhesion between the reinforcement and matrix material.

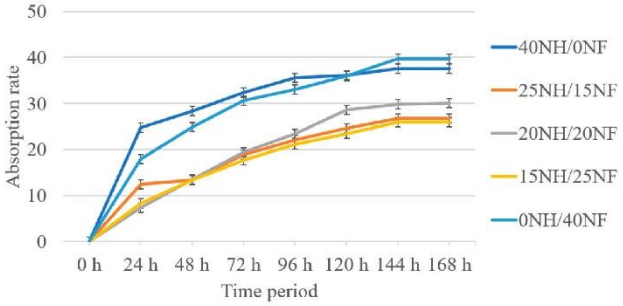
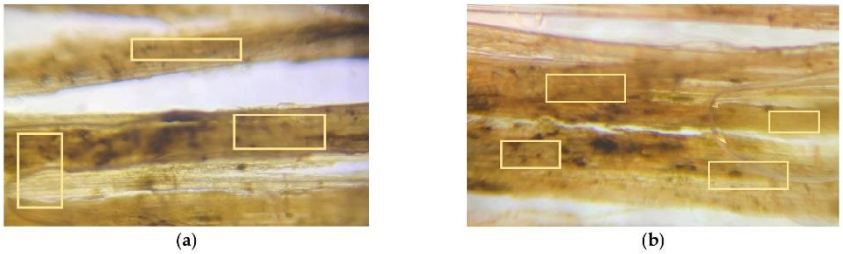
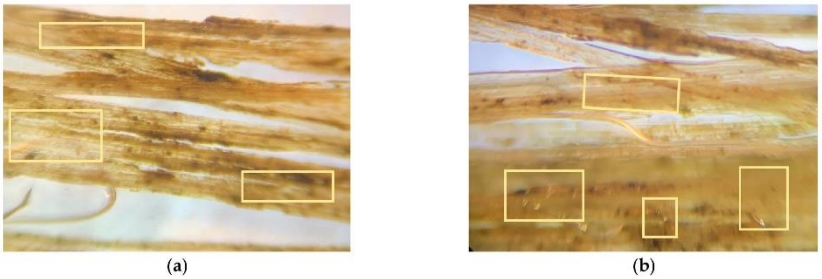


Figure 8. Time period vs. absorption rate for NH/NF hybrid biocomposites.



(a) (b)

Figure 9. Microscopic images for untreated (**a**) hemp and (**b**) flax fibers, with all having a smooth surface finish.



(a) (b)

Figure 10. Microscopic images for treated (**a**) hemp and (**b**) flax fibers, with all having a rough surface finish.

The microcracks and kink bands are two forms of flaws that are normally predicted to be found in the cell walls of natural fibers. Figure 11a,b shows the microcracks observed in the optical micrographs of noil hemp and noil flax fibers. However, Figure 11c,d shows the microscopic images for treated noil hemp and noil flax fibers without microcracks in the fiber cell wall. The kink bands were observed in untreated noil hemp fibers in Figure 11e. After chemical treatment, the kink bands were eliminated and shown in Figure 11f. The reinforcement materials act as a load carrier in a composite material. Therefore, the presence

of microcracks and kink bands affects the fiber properties and lowers the strength. We can say that to avoid these kinds of defects in natural fibers, a chemical treatment is one of the best solutions.

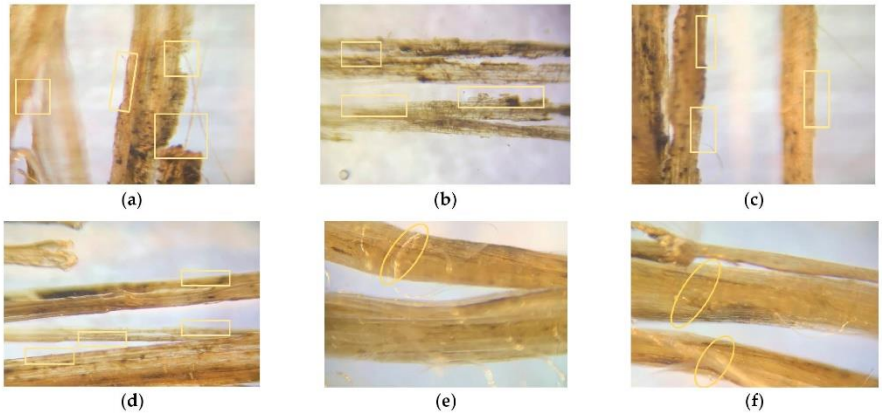
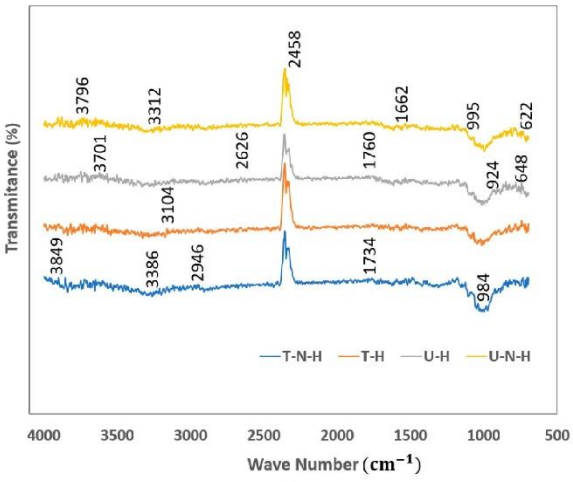


Figure 11. Microscopic images for untreated noil hemp (**a**), noil flax (**b**) fibers with microcracks, treated noil hemp (**c**), noil flax(**d**) fibers without microcracks, untreated noil hemp (**e**) with kink band-treated noil hemp, and (**f**) without kink bands.

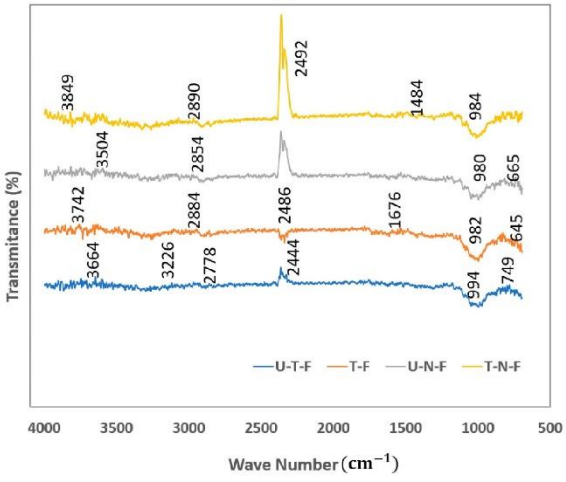
3.6. FTIR Analysis

The fibers' surface morphology and the influence of chemical treatment on natural fibers used in this research work are analyzed by using the FTIR analysis. The following figures illustrate the treated and untreated natural fibers.

The above Figure 12a,b show the influence of chemical treatment on natural fibers, analyzed by using the Fourier-transformed infrared spectroscopy analysis. The character peaks of treated hemp (T-H), treated noil hemp (T-N-H), untreated hemp (U-H), and untreated noil hemp (U-N-H) fibers are shown in Figure 12a. The character peaks of treated flax (T-F), treated noil flax (T-N-F), untreated flax (U-F), and untreated noil flax (U-N-F) fibers are shown in Figure 12b. These character peaks of fiber materials are attributed to the presence of cellulose, hemicellulose, and lignin content in the fiber material. In general, the IR spectra for natural fibers are presented in the 3250 to 2650 cm^{-1} range and the large bands indicate the deformation of the O-H groups. For the untreated hemp, flax, noil hemp, and noil flax fibers, this peak is found in the range between 3374 and 2846 cm^{-1}. The detected absorption band is connected to the O-H group's deformation. The second large peak, followed by the O-H group, is observed at 2672 cm^{-1}, which exhibits the C-O group of the hemicellulose in UH. The characteristic peak at 1584 cm^{-1} is characteristic of the N-H group of cellulose. The peak at 523 cm^{-1} is related to the irregular deformation of C-I of the cellulose and hemicellulose. Whereas in treated fibers, the highest peak is observed around 2780 cm^{-1}. The large peak represents the O-H groups in cellulose. The intensity of this peak is higher than the peak observed in the untreated fibers. It was observed from the figure that the obscene C-O band of the carboxyl group is present in the hemicellulose of natural fibers. Which indicates the removal of hemicellulose in the alkaline-treated natural fibers. Also, in the treated fibers, the lignin traces were not absorbed much. Therefore, due to the elimination of hemicellulose and lignin content in the natural fibers, the moisture intake of natural fibers can be reduced, which adversely affects the mechanical properties of the natural fiber composites.



(a)



(b)

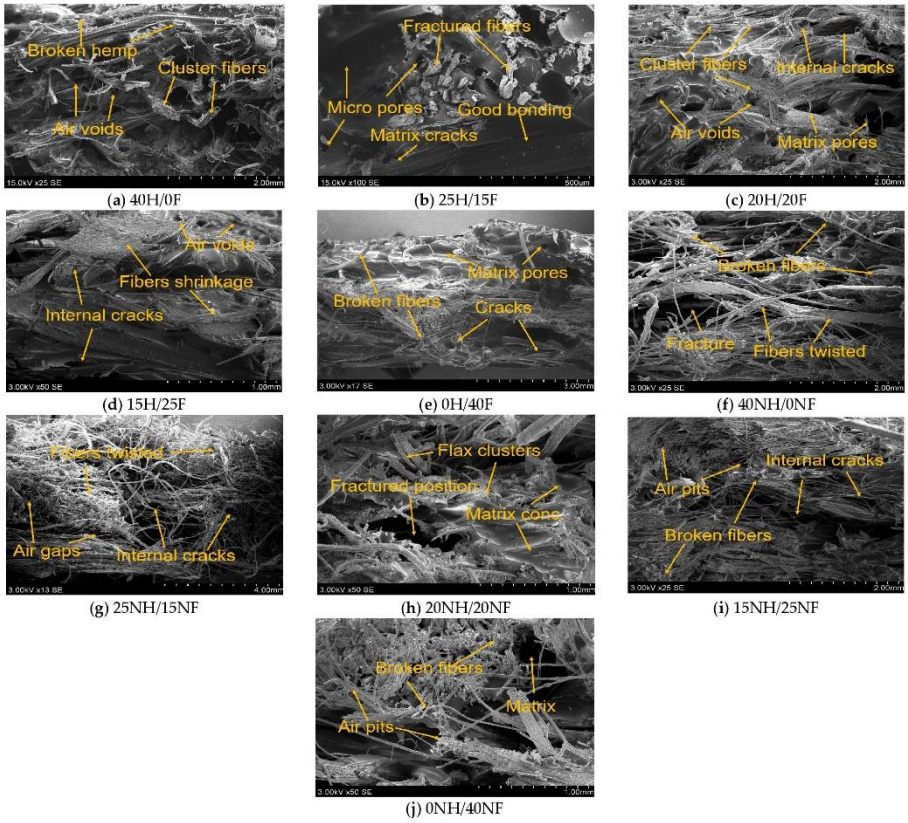
Figure 12. FTIR analysis of treated and untreated hemp and noil hemp fibers (**a**) and treated and untreated flax and noil flax fibers (**b**).

3.7. Scanning Electron Microscope (SEM) Analysis

The fracture mechanism of the composite samples was analyzed by using the scanning electron microscope. The SEM images of the hemp/flax and noil hemp/noil flax fiber hybrid biocomposite flexural test specimens were taken to analyze the defects in them.

Figure 13a–e above depicts the scanning electron micrographs taken for the fracture flexural test specimens of hemp and flax fiber hybrid biocomposites. From Figure 13a,e,

the breakage of fibers was observed in pure biocomposites. The breakage of the fibers lowers the strength of the composites because reinforcements are the load-carrying agent in a composite material when it is subjected to the external loading (force). Also, the presence of microcracks and air voids can be observed in hybrid composites (Figure 13b–d); the presence of defects in the composite material may lower the strength of the composites. The reasons for these defects could be fabrication error, residual stress from the curing process, or poor adhesion between the matrix and reinforcement. Overall, 40H/0F and 0H/40F biocomposites have more defects than hybrid biocomposites, leading to weaker strength. Figure 13f–i shows SEM micrographs of the shattered surfaces of the generated biocomposites (NH/NF) coming from samples that perform well in most of the attributes studied. It was observed from the results that the presence of fiber clusters and twisted fibers in the pure noil biocomposites (Figure 13f,j) may cause the agglomeration of the fibers in the composite material and lower the strength as well. The presence of air pits is observed in Figure 13g–i, which attracts the moisture easily. The reasons for the presence of internal defects are explained earlier.



(**a**) 40H/0F (**b**) 25H/15F (**c**) 20H/20F

(**d**) 15H/25F (**e**) 0H/40F (**f**) 40NH/0NF

(**g**) 25NH/15NF (**h**) 20NH/20NF (**i**) 15NH/25NF

(**j**) 0NH/40NF

Figure 13. SEM micrographs for hemp/flax fiber biocomposite flexural test fracture samples.

The rate of absorption for the composite material mainly depends on the presence of porosity and the fiber content. Figure 14a–e shows the presence of porosity (air voids) in hemp and flax fiber hybrid composites and f–j shows the noil hemp and noil flax fiber hybrid composites. It was observed that pure biocomposites have a greater void percentage, whereas hybrid composites have a low percentage. The porosity content in 25H/15F (Figure 14b) is minimal so it enhances the improvement in strength. Also, it was observed that 40NH/0NF composites have a greater void percentage and 25NH/15NF fiber composites have the least void content. In both cases, hybrid biocomposites have a lower void percentage, which reflects the improvement in strength, whereas pure biocomposites have more void content. Hence, the rate of absorption is higher in pure composites and lower in hybrid composites in both cases.

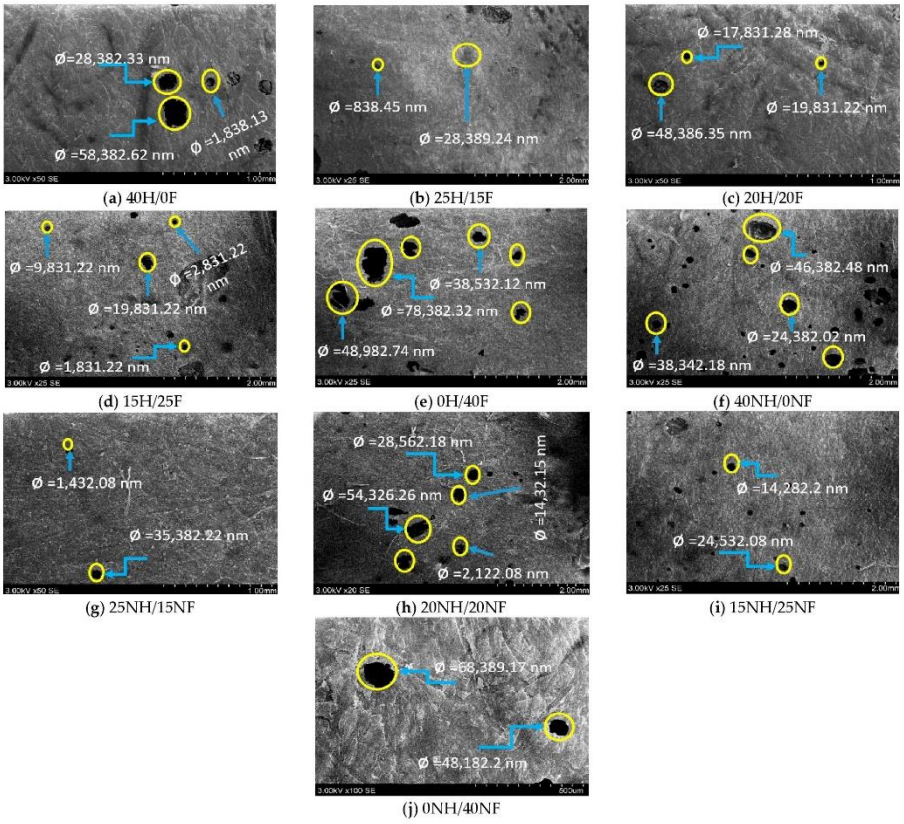


Figure 14. SEM micrographs for porosity content in hemp and flax fiber hybrid biocomposites.

3.8. SEM−EDX Spectrum Analysis

The chemical composition presented in the natural fiber biocomposites are analyzed with scanning electron microscopic energy dispersive X-ray spectroscopy and the results are shown in the following figures.

The chemical composition presented in the hemp and flax fiber hybrid biocomposites at surface position is studied by using the SEM−energy dispersive X-Ray spectrometer and the results are described in Figure 15a,b. The percentage of the weight concentration and atomic concentration of the individual elements at the surface position are given in Table 4. From the results, it was observed that there is no such variation in the biocomposites at the surface position. In both fibers, the percentage of carbon takes the top place, followed by oxygen. The presence of sodium and silicon is observed in hemp fiber biocomposites, though the percentage is minimal. This was not observed in the flax fiber composites. The presence of oxygen in the composite surface may form hydroxyl bonds with hydrogen present in the cellulose content, which leads it to attract greater moisture content and weaken the composite's strength.

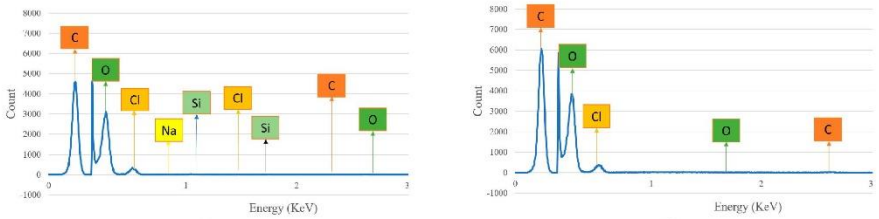


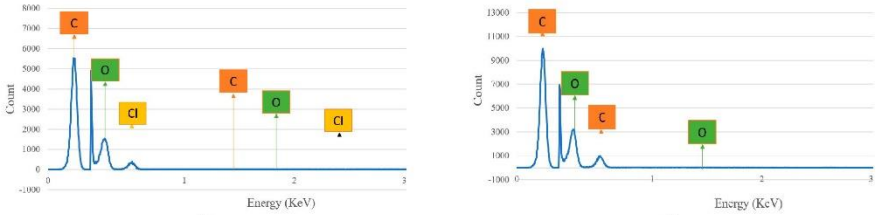
Figure 15. Spectrum analysis of chemical composition for (**a**) hemp and (**b**) flax fiber biocomposites at surface position.

Table 4. Spectrum analysis for chemical configuration of natural fiber hybrid biocomposites at the surface position.

Element Type	Element No.	Atomic Weight	Hemp Biocomposite		Flax Biocomposite	
			Weight (%)	Atomic Conc. (%)	Weight (%)	Atomic Conc. (%)
Carbon (C)	6	12	76.29	81.61	77.17	82.18
Oxygen(O)	8	16	22.04	17.70	21.85	17.47
Chlorine (Cl)	17	35.5	1.03	0.37	0.97	0.35
Sodium (Na)	11	23	0.28	0.16	–	–
Silicon (Si)	14	28	0.35	0.16	–	–

After the surface position, the spectrum analysis was carried out at the fractured position to understand the internal composition of the hemp and flax fiber biocomposites. The resultant micrographs are shown in following figures.

The chemical composition presented in the hemp and flax fiber hybrid biocomposites at fractured position was studied by using the SEM−EDX spectrometer and the results are described in Figure 16a,b. The percentage of the weight concentration and atomic concentration of the individual elements at the fractured position are given in Table 5. From the results, it was observed that there is some variation in the biocomposites at the fractured position, unlike at the surface. In both fibers, the percentage of carbon takes the top place, followed by oxygen. The presence of chlorine is observed in hemp fiber biocomposites, though the percentage is very small. This was not observed in the flax composites.



(a) (b)

Figure 16. Spectrum analysis of chemical structure for (**a**) hemp and (**b**) flax fiber biocomposites at fractured position.

Table 5. Spectrum analysis for chemical configuration of natural fiber hybrid biocomposites at the fractured position.

Element Type	Element No.	Atomic Weight	Hemp Biocomposite		Flax Biocomposite	
			Weight (%)	Atomic Conc. (%)	Weight (%)	Atomic Conc. (%)
Carbon (C)	6	12	63.73	70.26	68.82	74.62
Oxygen(O)	8	16	35.67	29.52	31.18	25.38
Chlorine (Cl)	17	35.5	0.60	0.22	–	–

After the spectrum acquisition for hemp and flax fiber biocomposites at the surface and fractured positions, we can deduce that the proportion of oxygen is greater at the fractured place than at the surface and it is the opposite for the carbon percentage. The presence of chlorine (Cl), sodium (Na), silicon (Si) along with oxygen (O), and carbon (C) was also observed, albeit in very low percentages. This reflects that the concentration of matrix (ecopoxy) material is greater on the surface than in the fractured position, which leads to a smooth surface finish at the surface of the composite.

4. Conclusions

The current research on hemp, flax, noil hemp, noil flax natural fiber-reinforced ecopoxy hybrid biocomposites was fabricated using the hand layup technique, followed by a compression molding process. The fibers were treated with a 5% NaOH solution before fabrication. The fabricated composites underwent mechanical and morphological testing and the results are as follows:

1. Hemp and flax fiber hybrid biocomposites showed superior mechanical properties to noil hemp and noil flax fiber biocomposites. In both cases, hybrid biocomposites showed improved properties compared to pure biocomposites. From the interlaminar shear test results, hemp and flax fiber biocomposites showed a range between 5.67 and 13.10 MPa, whereas noil fibers were from 4.9 to 11.3 MPa. From the flexural test results, the 25H/15F hybrid biocomposites exhibited the maximum flexural strength and modulus as 30.04 MPa and 0.825 GPa and pure noil hemp (40NH/0NF) fiber biocomposites exhibited the minimum as 13.31 MPa and 3.95 GPa.
2. All of the biocomposites had a contact angle of fewer than 90 degrees based on contact angle measurement, indicating that they have hydrophilic surface qualities. The maximum contact angle was observed for 20NH/20NF at 87.31° with the measurement error of ±1.56° and the smallest contact angle was observed for 40H/0F at 53.01° with the measurement error of ±1.94°. From the rate of absorption analysis, all the biocomposites responded to the moisture and the rate increased up to 144 h, remaining constant. In both cases, pure biocomposites absorbed more water than hybrid biocomposites.

3. The fiber morphology studies of treated and untreated fibers were investigated by using the optical microscope. The results were presented in a way that contrasted the treated and untreated fibers in terms of defects. The fracture mechanisms and the porosity content in the biocomposites were investigated by using scanning electron microscopy. The spectra acquisitions for the chemical composition presented in hemp and flax fiber biocomposites were observed by using SEM−EDX spectrographs.

Overall, hemp and flax fiber hybrid biocomposites showed superior properties to noil fiber biocomposites so they can be efficiently used as reinforcements in lightweight biosensor chips, flexible electronics, microfluidics, and biocomposite applications. Although these hybridization techniques and chemical treatment processes achieved promising results, more investigations are essential to improve the porosity content and moisture properties, which may be possible with the addition of filler materials, additive materials, and heat treatment processes.

Author Contributions: Conceptualization, A.A.; methodology, A.A. and A.P.; investigation, A.A. and J.E.; calculation and analysis, A.A. and J.E.; writing—original draft preparation, A.A.; writing—review and editing, G.J. and A.P.; visualization, A.A.; validation, J.E. and G.J. All authors have read and agreed to the published version of the manuscript.

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Numerical and Experimental Analysis of Mechanical Properties of Natural-Fiber-Reinforced Hybrid Polymer Composites and the Effect on Matrix Material

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Abstract: The impact of matrix material on the mechanical properties of natural-fiber-reinforced hybrid composites was studied by comparing their experimental, and numerical analysis results. In the present work hemp and flax fibers were used as reinforcement and epoxy resin and ecopoxy resin along with hardener were used as matrix materials. To study the influence of the matrix material, two sets of hybrid composites were fabricated by varying the matrix material. The composite samples were fabricated by using the compression-molding technique followed by a hand layup process. A total of five different composites were fabricated by varying the weight fraction of fiber material in each set based on the rule of the hybridization process. After fabrication, the mechanical properties of the composite samples were tested and morphological studies were analyzed by using SEM-EDX analysis. The flexural-test fractured specimens were analyzed by using a scanning electron microscope (SEM). In addition, theoretical analysis of the elastic properties of hybrid composites was carried out by using the Halpin–Tsai approach. The results showed that the hybrid composites had superior properties to individual fiber composites. Overall, epoxy resin matrix composites exhibited superior properties to ecopoxy matrix composites.

Keywords: natural fibers; matrix material; mechanical properties; SEM-EDX; Halpin–Tsai model

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1. Introduction

Natural fiber composites have gained much popularity in recent times due to concerns about the environmental impact of synthetic fiber composites [1,2]. Composites of natural fibers in thermosetting or thermoplastic polymer matrices have attracted a significant amount of interest from researchers due to their cost-effectiveness, ease of availability, environmental friendliness, and ability to compete with synthetic composites [3,4]. Natural-fiber-reinforced polymer composites have been employed in a variety of industries, including in automobile parts, aerospace components, sporting products, marine applications, military goods, and in the biomedical, microfluidics, and construction industries [5–7]. Low self-weight, high specific strength, free formability, and strong corrosion and fatigue resistance of natural-fiber-reinforced composites are some of the additional reasons for their increased popularity. On the other hand, natural composites have been limited in their usage due to their poorer mechanical and thermo-physical qualities when compared to synthetic composites and traditional structural materials [8,9]. Plant-based natural fibers such as banana, pineapple, abaca, jute, okra, sisal, hemp, flax, caryota, kenaf, and date palm fibers have proved to be potential reinforcement in the thermosetting and thermoplastic polymer composites and are being used in a wide range of applications [10,11].

In a composite material both the reinforcement and matrix materials play a crucial role. The reinforcement material acts as the load-carrier whereas the matrix material acts as the load-distributer when an external load is applied to the composite [12,13]. The matrix

materials are classified as thermoset polymers and thermoplastic polymers. Thermoplastic materials can be reused even after the curing process, but need proper heat treatment to enable their reuse. Polyester, polypropylene, nylon, and Teflon are some examples of thermoplastic polymers. Unlike thermoplastic polymers, thermoset polymers cannot be used after the curing process. When the liquid thermoset resin is condensed (solidified) at a necessary temperature, it cannot recycle. The most common thermoset resins are epoxies, polyamides, and vinyl esters. In recent years, thermoset polymers have become more popular than thermoplastic polymers due to their wide range of applications in various sectors [14–16]. Among all polymer composites, thermoset polymers based on epoxy resin composites have had the most attention due to their good adhesive properties, low contraction, curing (liquid-state to solid-state) time, and ease of use [17,18].

The static and dynamic mechanical properties of the polymer composites reinforced with natural fibers are mostly the result of the quality and quantity of fiber materials [19,20], in addition to the interfacial strength and adhesion properties between the constituents of polymer composite [21]. Hybridization of composite materials is an alternative way to improve their mechanical performance. Hybridization occurs when a composite is produced by the combination of two or more reinforcements under the same matrix material, and the composites are called hybrid composites [22,23]. Several researchers have analyzed the mechanical properties of hybrid composites and shown the advancement in their properties. The main advantage of hybridization is that the mixing of the reinforcements can be done by various approaches such as blended continuous fibers, short fibers, discontinuous fibers, and particulate-matter fibers [24–26]. By far most of the hybrid compositions are focused on natural fibers with synthetic fibers such as glass and carbon fibers and they offered a range of properties superior to individual fiber composites. However, natural fiber hybrid composites have become more popular than synthetic fiber composites due to environmental concerns [27].

The chemical composition of natural fibers plays a prominent role in plant-based fiber composites, but environmental conditions such as climate dissimilarity and geographical changes influence the composition of natural fibers [28]. Hence, in most of the cases, their composition is expressed as maximum and minimum values [29]. In general plant-based fibers mainly consist of cellulose, hemicellulose, and lignin. Cellulose content and the aspect/ratio of natural fibers play a key role in mechanical performance [30,31]. In the absence of these chemical compositions in artificial (synthetic) fibers, their mechanical properties do not change much. On the other hand, natural fibers grow in open environments with the assistance of air, light, water, soil, etc. These conditions vary from time to time, which affects the properties of the fibers [32,33]. Some research studies have shown that fiber composites reinforced with cellulose-rich plant-based natural fibers such as hemp, flax, and banana have superior properties to composites reinforced with natural fibers with lower cellulose content [34]. Hence, in the current research hemp and flax fibers were used as a reinforcement to study the effect of matrix material on mechanical performance.

Recently, several researchers have shown an interest in working with natural fiber composites in the presence of thermoset polymers. Although there are many advantages with natural fiber composites the applications are still limited due to moisture uptake and flame-retardancy properties. Yihui et al. [35] developed a micromechanical model to understand the effect of moisture on the degradation of composite material. The mechanical degradation and swelling properties of natural fiber composites were analyzed by the Mori–Tanaka model with the inclusion of different shapes. The results showed that a proper matrix material can avoid mechanical degradation as well as reduce the moisture uptake by the natural fiber composite. Neves et al. [36] investigated the mechanical properties of hemp fiber composites with various matrix materials. They drew a comparison between epoxy and polyester resins. They showed that hemp fiber composites based on epoxy resin polymer have superior properties to polyester resin composites and stated that these composites have a potential uses in armed applications. Sapuan et al. [37] investigated the mechanical properties of woven banana-fiber-reinforced epoxy composites. A theoretical

model based on the ANOVA technique was developed to compare the experimental results and it was observed that there was a slight variation in the theoretical and experimental results. Several researchers have offered useful and comprehensive review articles on natural-fiber-reinforced hybrid composites and the importance in terms of mechanical performance [10,38,39]. Summerscales et al. [40] reviewed various statistical models which have been useful for natural fiber composites. This paper also presents the various harvesting techniques and growing conditions of natural fibers. Alhazmi et al. [41] worked on tribological and mechanical properties of hybrid composites based on epoxy resin. The results showed that the presence of nanofiller materials cannot affect any sort of improvement in terms of wear resistance of the composite material. Singh et al. [42] investigated the mechanical properties of hemp fiber composites. To analyze the mechanical properties, tensile and flexural tests were conducted. The results showed that the mechanical properties were increased with an increase in fiber content. These properties increased up to 40% and then decreased. Moudood et al. [43] studied the mechanical properties of flax fibers under various environmental conditions. The mechanical properties that were measured experimentally agreed strongly with the statistical results.

Zhong et al. [44] reviewed the challenges in numerical simulations and modeling of natural fiber composites. Microstructures of natural fiber composites were presented by featuring their properties that should be viewed before displaying. The authors also discussed and summarized the previous numerical works. Unlike artificial fibers, natural fiber composites certainly stand out concerning numerical approaches [45]. Existing specialized difficulties in this subject are summed up to give possible options for future research. Jose da Silva et al. [46] worked on numerical and experimental analysis of natural-fiber composites. This work primarily emphasized sisal and banana natural-fiber-reinforced epoxy-resin polymer composites. Tensile tests were performed to validate the numerical data. To verify the experimental results both 3D and 2D models were developed by considering the elastic properties of each constitution of the composite material. Two-dimensional analysis was conducted based on the rule of mixtures (ROM) and three-dimensional analysis was conducted on ANSYS software. The results stated that the variation of Poisson's ratio values did not affect the finite element analysis results. Sathishkumar et al. [47] studied the experimental and numerical analysis of filler material on sisal-fiber-reinforced polymer composites. The mechanical properties were tested by using flexural and tensile tests. The 2D analysis of polymer composites was conducted by using the ROM method and Halpin–Tsai approach. The 3D analysis was conducted by using the ANSYS workbench by considering the elastic properties of the materials used for fabrication purposes. The results showed the improvement in mechanical properties due to the addition of filler material to the composite materials. Prasanthi et al. [48] investigated the waviness effect on sisal fiber composites through numerical and experimental methods. The idea of the work was to understand the effect of waviness on fiber composites and compare the results in both analytical and experimental methods. The analytical works were carried out by using ANSYS model and the results stated that there was a significant effect of waviness on the fiber composites. Balasubramanian et al. [49] analyzed the mechanical properties of natural fibers by using experimental and FEA analysis. Hybrid composites were fabricated by using flax, sisal, and aloe vera fibers as reinforcement and epoxy resin as a matrix material. The mechanical properties were tested by using tensile, flexural, and impact tests. The finite element analysis was carried out to find the stress, deformation, and displacement of the composite materials, and then the results were compared.

From the above literature, two significant points are perceived. One is that the mechanical properties of natural fibers can be improved by using a hybridization process and the second is that the numerical and theoretical analysis of natural fibers can be done by assuming definite properties through finite element analysis and micromechanics. With this this information, the main objective of our research on the effect of matrix materials on natural fiber hybrid composites was defined. This objective was to understand the impact of matrix material on the mechanical properties of the natural-fiber-reinforced

epoxy and ecopoxy hybrid composites. For this study, hemp and flax fibers were used as reinforcements, and to draw the contrast between matrix materials, epoxy and ecopoxy materials were used as matrix materials. The results are assessed by using experimental and numerical methods. The mechanical properties of hybrid composites were tested by using flexural and interlaminar shear properties. The wettability properties are tested by using contact angle measurement and the morphological studies were carried out with SEM-EDX spectrum analysis. Theoretical analysis of composite materials was estimated by using Halpin–Tsai method and the effect of matrix material on mechanical properties of natural fibers hybrid composites was addressed.

2. Materials and Methods

2.1. Materials

To carry out the current research work, hemp and flax fibers were procured from Naturalus Pluostas UAB, Vilnius, Lithuania. The epoxy resin and its hardener were procured from Composite 24, Riga, Latvia and the ecopoxy (the bio-based resin contained 36% of bioactivity) resin along with hardener was procured from EcoPoxy EU. Warsaw, Poland. Teak wood was used for the preparation of mold material.

2.2. Preparation of Reinforcement and Matrix Materials

The reinforcement materials were available in the form of a bundle. Each fiber was carefully chosen by eliminating the unwanted materials, such as the thick stems and small pellets by using the hand sitting process. Once the waste materials had been separated from the fibers, water treatment was carried out and the fibers were dried until the moisture was eliminated. However, the fibers were still not ready for fabrication due to their smooth surface finish and poor adhesive properties. Therefore the separated fibers underwent a chemical treatment process. The fibers were treated with a 5% NaOH solution and then cleaned with distilled water. After alkaline treatment, the fibers were dried in an oven ready for the fabrication process.

The matrix materials used for the fabrication process were epoxy and ecopoxy resin along with hardener material. The weight portion of both the matrices was considered a 2:1 ratio as per the material description provided by the company from which they were procured. The required amount of resin and hardener were put into a plastic container and stirred with a plastic stirrer for 4 minutes and the walls of the container were scratched properly to get the proper mix and left for two minutes to allow the clear solution to settle. The process was repeated for both matrix materials. Once the mixed solution had settled it was taken for the fabrication process.

2.3. Composite Sample Preparation

The weight fractions of the reinforcement and matrix materials were considered as per the rule of the hybridization process. This states that a composite sample is fabricated by using two or more fiber laminates under the same resin material with a 0.4 Wf.% [50,51]. In the present research work, hemp (H) and flax (F) fiber hybrid composites were fabricated in the presence of epoxy resin and ecopoxy resin. The composites were fabricated by using the compression molding technique followed by the hand-layup technique. The required weight proportions of fibers were placed uniformly in the mold material and compressed to acquire the mold shape and then the epoxy resin was applied to it. The fibers were placed in uniform direction ("0" degrees) for neat hemp and flax composites whereas in the hybrid case the fibers were placed in uniform-direction layer-by-layer mode and for testing the orientation of fibers was not considered. The schematic representation of the ply sequence for ecopoxy composites is given in the Figure 1 and a similar sequence is followed for the epoxy-matrix-based hybrid composites. The mold material was covered with a backing paper and then compressed with a uniform load for 72 hours. Once the curing process was completed the composites were subjected to the heat treatment process. A total of five different weight fractions of composites were fabricated (40H/0F, 25H/15F,

20H/20F,15H/25F, and 0H/40F). In all cases the fibers were laid down horizontally with a 0 degree angle. The sample weight fractions are given in Table 1. All the samples were fabricated as per ASTM testing standards. The same process was repeated for ecopoxy matrix composites.

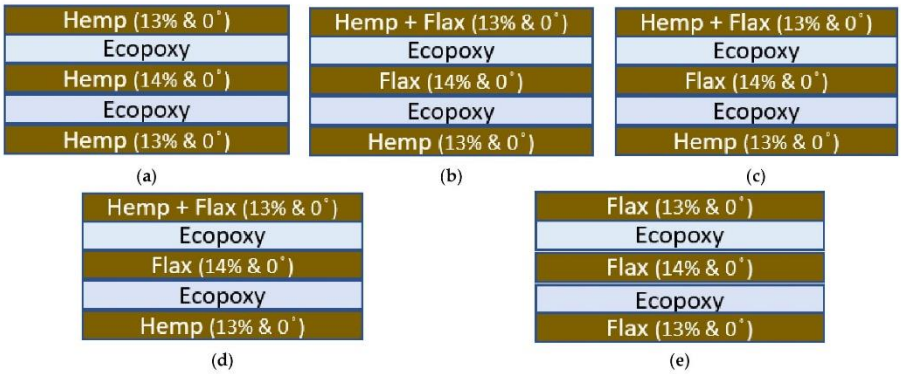


Figure 1. Schematic layout of the ply sequence for ecopoxy-matrix-based hybrid composites. (**a**) 40H/0F, (**b**) 25H/15F, (**c**) 20H/20F, (**d**) 15H/25F, and (**e**) 0H/40F.

Table 1. The weight fraction of composite samples for epoxy/ecopoxy matrices.

Composites	Hemp (%)	Flax (%)	Total Reinforcement Vol. (%)	Total Matrix Vol. (%)
40H/0F	40	0	40	60
25H/15F	25	15	40	60
20H/20F	20	20	40	60
15H/25F	15	25	40	60
0H/40F	0	40	40	60

2.4. Characterization Methods

2.4.1. Flexural Test

To measure the flexural properties such as flexural strength and modulus of the composite samples, a universal testing machine (Tinus Olsen H10K) was used. To test the samples, the 3-point bending test was preferred over the 4-point bending test because the external load acts at the center of the composite so load distribution was uniform [52]. The test was conducted at a constant crosshead speed of 10 mm/min and the strain rate was 0.10 mm/min for all specimens. All the samples were tested as per standard ASTM D-790 [53]. For each sample, a total of five composites was tested to take average values. The flexural modulus of a composite sample was calculated by drawing a tangent line for the stress–strain curve; hence it is also called the tangent modulus. In general, the flexural modulus is defined as the capacity of a composite sample to deform when an external load acts on it. The flexural test samples were analyzed by using the scanning electron microscope.

2.4.2. Interlaminar Shear Test

To calculate the interlaminar shear strength of composite material, a universal testing machine with a full load capacity of 10 KN was used. To test the samples, the composite was fixed on both ends in between the jaws of the testing machine, and the load was

applied in an axial direction from one end and the lower end was fixed. The samples were tested as per standard ASTM D-3846 [54]. The rectangular samples had a notch cut on both sides, 6.4 mm apart. The notch depth was considered half of the thickness of the sample on each side of the composites specimen. The shear strength was measured only when failure occurred between the notches. The load acting on the composite was gradually increased until the composite broke.

2.4.3. Wettability Test

The wettability properties of composite material were tested by using the contact angle measurement process. This provides quantitative information on interfacial energy between a fluid drop and composite surface [55]. A variety of fluids, such as spirit, water, and glycerin are used to measure the contact angle of a solid surface, of which, water is the most popular. Wettability indicates the examination of contact angles that regulate the wetting degree between a solid surface and a liquid drop. If the contact angle is less than 90 degrees it means the solid surface has a hydrophilic nature and if it is more than 90 degrees it has a hydrophobic nature. There are several factors that can affect the contact angle such as temperature, atmospheric conditions, type of liquid, the viscosity of the liquid, and time to take the measurement [56,57]. In the present work, all the samples were tested in a dark room to avoid light, and dimensions were considered as per ASTM testing standards. The composite surfaces were cleaned properly before forming the water bubble. The contact angle values were measured at various positions on the composite surface.

2.4.4. SEM Energy Dispersive X-ray Spectrum Analysis

A scanning electron microscope (SEM) from Bruker, Billerica, MA, USA was used to investigate the morphology of both the composite surface and the fractured sample. The element composition the surface of each composite sample and the facture position were evaluated by using SEM-EDX spectroscopy. The error % connected with the elemental composition analysis was evaluated.

2.5. Numerical Analysis

Numerical analysis of natural fiber composites was carried out by the using micromechanical method i.e., the Halpin–Tsai approach. Although there are many methods available to predict theoretical analysis, these are easy to apply to natural fiber composites. To calculate the elastic properties the basic assumptions must be considered as the composite sample was isotropic and homogeneous, as well as being linear elastic in nature [58,59]. We modified the equations to analyze the hybrid fiber composite elastic properties. The modified equations to calculate the elastic properties are given below:

Halpin–Tsai method [60–62]

$$E = \frac{E_m\left(1 + \zeta\eta V_f\right)}{1 - \eta V_f} \tag{1}$$

where η is

$$\eta = \frac{E_f - E_m}{E_f + \zeta E_m} \tag{2}$$

By substituting the η value in Equation (3) the final equation will be as follows:

$$E = \frac{E_m + \zeta V_f E_m\left(E_f - E_m\right)}{E_f + \zeta E_m - V_f\left(E_f - E_m\right)} \tag{3}$$

where E_f and E_m are the Young's modulus of fiber and matrix material, respectively. V_f and V_m are volume fractions of fiber and matrix material, respectively. ζ is an arbitrary

variable, also called an empirical factor considered based on the assumptions. The ζ values are taken as 0, 1, 2, and so on. The modified Halpin's equation is as follows:

$$E = \frac{E_m + \zeta E_m (V_{f1} + V_{f2})\left(E_{f1} + E_{f2} - E_m\right)}{E_{f1} + E_{f2} + \zeta E_m - (V_{f1} + V_{f2})\left(E_{f1} + E_{f2} - E_m\right)} \quad (4)$$

E_{f1}, E_{f2}, and E_m are Young's modulus of hemp fiber and flax fiber, and the matrix material, respectively. V_{f1}, V_{f2}, and V_m are the volume fractions of hemp fiber, flax fiber, and matrix material, respectively. In the present work, an attempt has been made to check the extent of the modified Halpin's equation to the natural fiber hybrid composites. The test was conducted by assigning properties such as Poisson's ratio, young's modulus, density, bulk modulus, and shear modulus to the material. The values are given in Table 2. The mechanical properties such as elastic modulus and displacement were determined and the results were compared with the theoretical and experimental results.

Table 2. Mechanical properties of the materials.

Material	Density (g/cm^3)	Poisson's Ratio	Young's Modulus (GPa)	Shear Modulus (GPa)	Bulk Modulus (GPa)
Epoxy resin	1.10	0.29	3.35	2.10	1.408
Flax	1.4–1.5	0.41	60	21.27	1.11
Hemp	1.48	0.39	70	25.18	1.06
Ecopoxy	1.13	0.23	2.28	0.98	1.26

3. Results

3.1. Flexural Properties

The flexural strength and flexural modulus of hemp and flax hybrid composites with various weight fractions in the presence of epoxy resin matrix and ecopoxy resin matrix are shown in Figure 2. In both cases, hybrid composites showed superior flexural properties to those of individual fiber composites. This reflects the impact of hybridization on composite materials. It was observed that the epoxy-matrix-based composites exhibited greater flexural strength and flexural modulus than the ecopoxy matrix composites. This can be attributed to poor bonding and adhesion properties between the ecopoxy matrix and the reinforcement material. The epoxy matrix composites 25H/15F exhibited the highest flexural strength and modulus and the lowest was observed in the 0h/40F composites, whereas in ecopoxy matrix composites, 25H/15F hybrid composites showed the highest values and 40H/0F composites showed the lowest values. These results proved that the matrix material plays a crucial role in the mechanical performance of the composite material.

Scanning electron microscope analysis of flexural test specimens of epoxy matrix composites is illustrated in Figure 2. The presence of broken of fibers and fiber shrinkage in individual fiber composites can be observed (Figure 3a,e). This lowers the strength of the composite, whereas in hybrid composites (Figure 3b–d) strong bonding between reinforcement and matrix material was observed. This strong adhesion property of composites improves the stress distribution inside the composite material and improves the strength of a composite. In addition, minor cracks were observed in hybrid composites.

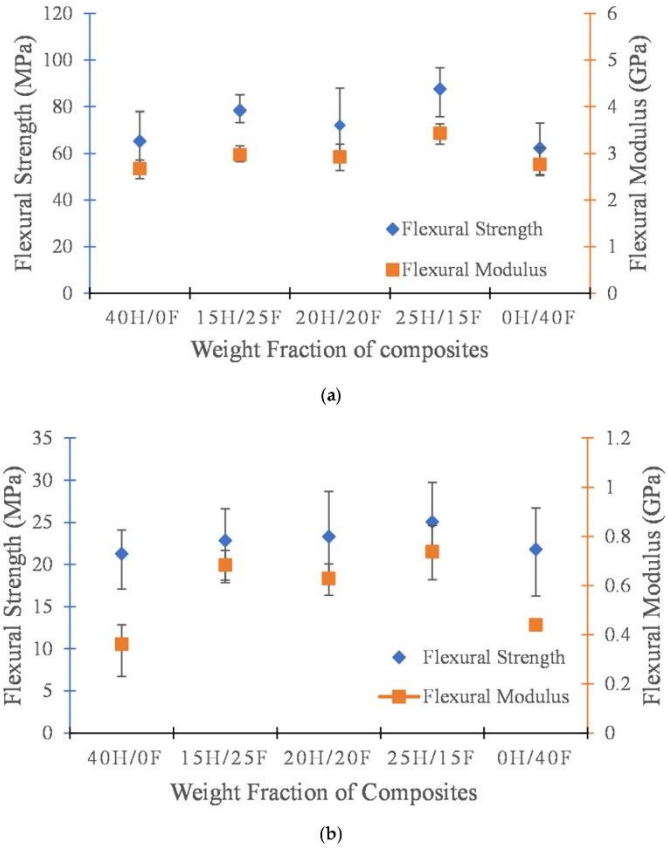


Figure 2. Flexural strength and modulus of H/F hybrid composites with epoxy (**a**) and Ecopoxy (**b**) matrices.

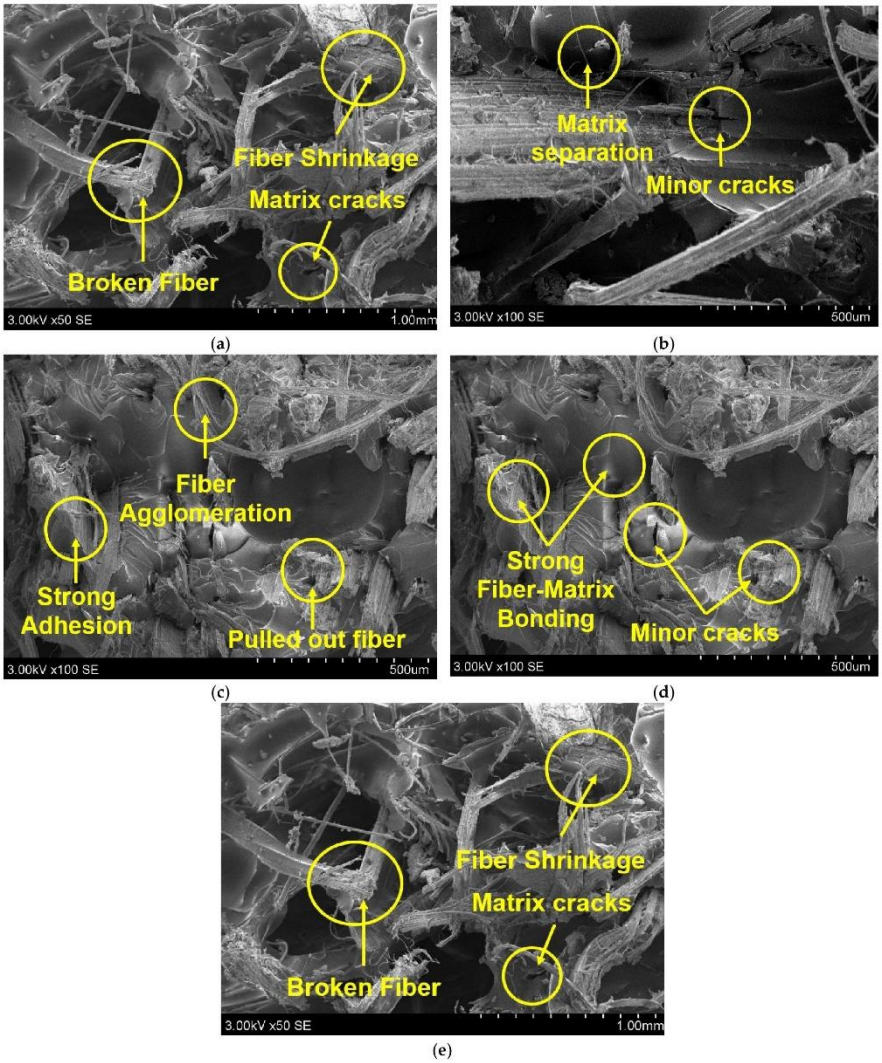


Figure 3. SEM analysis of flexural tested hemp- and flax-fiber-reinforced epoxy matrix composites. (**a**) 40H/0F, (**b**) 15H/25F, (**c**) 20H/20F, (**d**) 25H/15F, and (**e**) 0H/40F.

3.2. Interlaminar Shear Properties

The experimental results of epoxy and ecopoxy-based hybrid composites obtained from interlaminar shear tests are presented in Figure 4. The shear strength of a composite

is one of the crucial parameters in defining the capability of a composite to withstand delamination damage. From the results, it was clear that the interlaminar shear strength properties of hybrid composites were superior to those of the pure fiber (40H/0F and 0H/40F) composites in both cases. This indicates the strong bonding properties between the layers of the hybrid composite compared to the pure composite. The epoxy-based 25H/15F hybrid composite showed the highest shear strength at 13.01 MPa and the ecopoxy-based 40H/0F composite exhibited the lowest at 5.67 MPa.

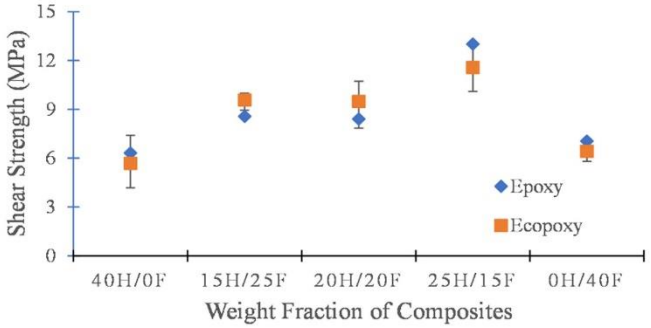


Figure 4. Interlaminar shear strength of epoxy and ecopoxy composites with various weight fractions.

3.3. Wettability Analysis

To analyze the wettability properties of a composite material a contact angle measurement was conducted on the hemp and flax fiber composites and the results are illustrated in Figure 5. To understand the surface property of composite material, it is necessary to find the wetting nature of a composite [63]. It is easy to determine whether a composite is exhibiting a hydrophilic or a hydrophobic surface property by conducting the contact angle measurement analysis. It was found that both epoxy and ecopoxy composites showed contact angles of less than 90 degrees which meant that all composites exhibited hydrophilic surface nature. In both cases, hybrid composites showed a similar contact angle whereas pure composites exhibited a slight variation in it.

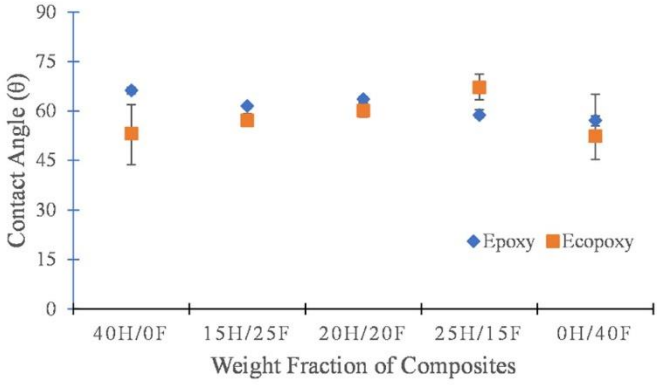
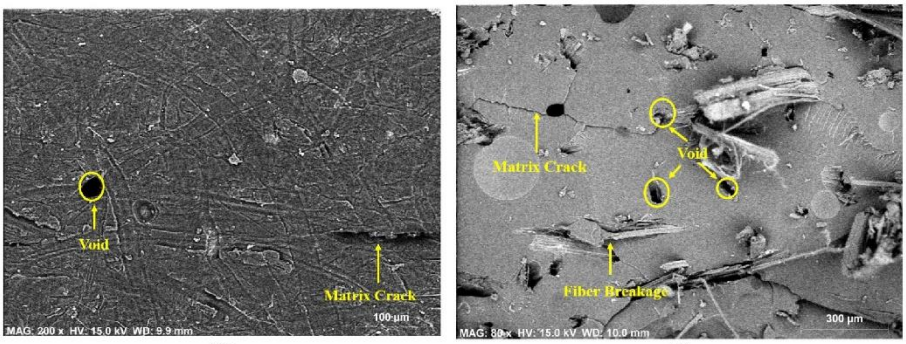


Figure 5. Contact angle measurement of epoxy and ecopoxy composites with various weight fractions.

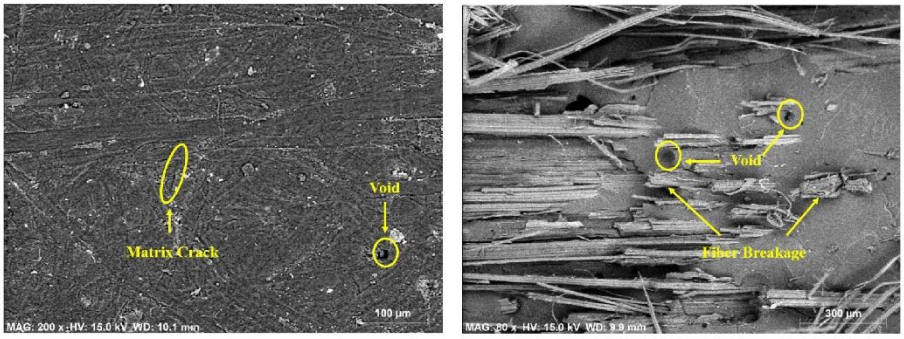
3.4. SEM-EDX Analysis

Morphological studies on individual hemp- and individual flax-fiber-reinforced epoxy composites at the surface position and the fracture position were carried out by using the scanning electron microscope energy dispersive X-ray spectroscopy and the results are shown in Figures 6 and 7. From SEM results it was observed that the presence of defects such as void content, fiber breakage, and matrix cracks were greater at the fracture position than at the surface position. The reasons for this could be poor reinforcement–matrix interactions and agglomeration of the fibers in the composite material.



(**a**) (**b**)

Figure 6. SEM EDX analysis on hemp-fiber-reinforced epoxy composite at surface (**a**) and fracture (**b**) positions.



(**a**) (**b**)

Figure 7. SEM EDX analysis of flax fiber reinforced epoxy composite at surface (**a**) and fracture (**b**) positions.

To determine the chemical composition and element analysis of the composite material, EDX spectrum analysis is usually preferred. The chemical composition and error % of the hemp and flax fiber epoxy composites are given in Table 3. In both fiber composites, carbon had the highest percentage followed by oxygen and the percentage of carbon was greater at the surface than at the fracture position. However, the atomic% of oxygen was greater at the fracture position than at the surface position. Only carbon, oxygen, chlorine, and a small

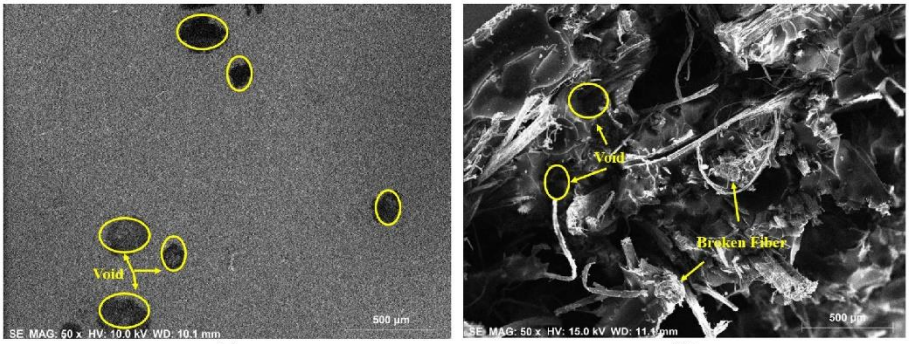
amount of sodium were observed at the surface position, but at fracture position, calcium, aluminum, and potassium were observed in addition to those chemical compositions present at the surface of the epoxy-based composite material.

Table 3. Chemical composition of hemp and flax fiber epoxy composites.

Element	Hemp Surface		Hemp Fracture		Flax Surface		Flax Fracture	
	Atom. C (%)	Error (%)	Atom. C (%)	Error (%)	Atom. C (%)	Error (%)	Atom. C (%)	Error (%)
Carbon	73.74	21.6	69.97	20.3	74.66	21.9	59.98	21.8
Oxygen	26.15	10.4	29.76	11.4	25.09	10.0	39.57	14.7
Chlorine	0.11	0.0	0.10	0.0	0.15	0.0	0.18	0.0
Calcium	–	–	0.09	0.0	–	–	0.14	0.0
Aluminum	–	–	0.08	0.0	–	–	–	–
Sodium	–	–	–	–	0.10	0.0	–	–
Potassium	–	–	–	–	–	–	0.13	0.0

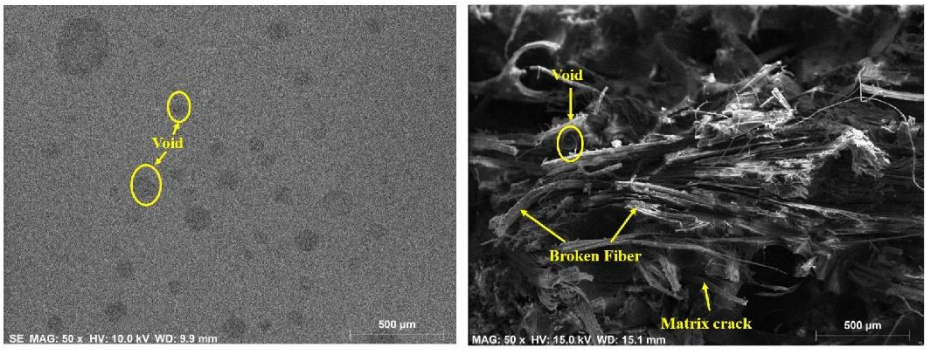
The morphological studies on individual hemp- and individual flax-fiber-reinforced ecopoxy matrix composites at the surface position and fracture position were carried out by using SEM-EDX spectroscopy and the results are shown in Figures 8 and 9. From SEM results it was observed that, similarly to epoxy composites, the presence of defects such as void content, fiber breakage, and matrix cracks was greater at the fracture position than at the surface position. There were, however, more defects in the ecopoxy composites than in the epoxy composites. The presence of defects leads to the weakening of the composite strength. It ultimately affects the mechanical performance of the composite material.

The chemical composition and error % of the hemp and flax fiber ecopoxy composites are given in Table 4. In both fiber composites, oxygen had the highest percentage followed by carbon, The percentage of oxygen was higher at the fracture than at the surface position, whereas the atomic% of carbon was more at the surface position than the fracture position. Only carbon, oxygen, chlorine, and a small amount of silicon were observed at the surface position, but at the fracture position, carbon, oxygen, chlorine, silicon, and calcium were observed.



(a) (b)

Figure 8. SEM EDX analysis on hemp fiber reinforced ecopoxy composite at surface (**a**) and fracture (**b**) positions.



(a) (b)

Figure 9. SEM EDX analysis on flax fiber reinforced ecopoxy composite at surface (**a**) and fracture (**b**) positions.

Table 4. Chemical composition of hemp and flax fiber ecopoxy composites.

Element	Hemp Surface		Hemp Fracture		Flax Surface		Flax Fracture	
	Atom. C (%)	Error (%)	Atom. C (%)	Error (%)	Atom. C (%)	Error (%)	Atom. C (%)	Error (%)
Carbon	48.11	23.6	40.76	16.8	48.76	25.2	37.42	35.0
Oxygen	51.30	20.6	58.97	20.3	51.07	19.6	62.12	22.7
Chlorine	0.39	0.1	0.17	0.0	0.17	0.1	0.27	0.1
Silicon	0.20	0.1	–	–	–	–	–	–
Calcium	–	–	0.10	0.0	–	–	0.19	0.1

Overall, the defect percentage was greater in the ecopoxy composites than in the epoxy composites; hence, the strength of the composite was decreased in the ecopoxy case. Due to the biological nature of the ecopoxy resin material the percentage of oxygen was greater in ecopoxy composites than in the epoxy composites.

3.5. Numerical Analysis

Numerical analysis was carried out by using the modified Halpin's method. The values are presented in Figures 10 and 11. In both cases, experimental values and numerical values were not close to each other and a large variation was observed when using Halpin's method. Halpin's method gave the higher data for all the hybrid composites investigated. This means that the modified Halpin's equations for calculation of elastic properties of hybrid-fiber-reinforced epoxy and ecopoxy composites gave higher bound results. Individual fiber composites showed the lowest elastic properties in both cases.

The data presented in Tables 5 and 6 shows the % error calculated for the epoxy and ecopoxy-matrix-based hybrid fiber composite's elastic properties. The error % was calculated by taking the ratio of the difference between theoretical values and experimental results upon the experimental results. It was observed that the error % calculated by using Halpin's method was far higher than the other methods. This indicates that the modified Halpin–Tsai theoretical calculations cannot be applied to natural fiber hybrid composites. It was observed that the elastic properties obtained from the numerical models were slightly higher than the experimental results in both epoxy and ecopoxy cases. This can be attributed to the fabrication errors and environmental conditions during the experimentation process whereas in theoretical predication, the outside factors are not considered.

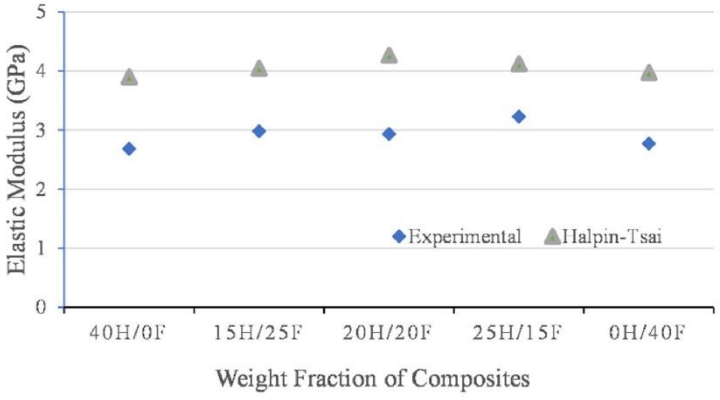


Figure 10. Elastic properties of epoxy-based hybrid composites by various methods.

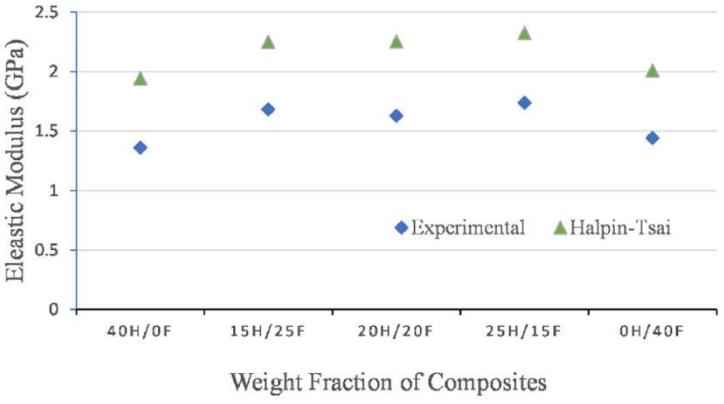


Figure 11. Elastic properties of ecopoxy-based hybrid composites by various methods.

Table 5. Comparison of elastic properties of epoxy composites.

Epoxy Composites	Experimental	Halpin–Tsai Method	Error (%)
40H/0F	2.683	3.896	45.21
15H/25F	2.981	4.042	35.59
20H/20F	2.934	4.264	45.33
25H/15F	3.226	4.118	27.65
0H/40F	2.772	3.968	43.15

Table 6. Comparison of elastic properties of ecopoxy composites.

Ecopoxy Composites	Experimental	Halpin–Tsai Method	Error (%)
40H/0F	1.361	1.942	42.69
15H/25F	1.683	2.248	33.57
20H/20F	1.628	2.254	38.45
25H/15F	1.737	2.324	33.79
0H/40F	1.442	2.008	39.25

The percentage error was calculated for epoxy and ecopoxy hybrid composites. The error was calculated by taking the ratio of the difference between numerical values and experimental results. The maximum error % of elastic properties was observed in pure composites such as 0H/40F and 40H/0F in both epoxy and ecopoxy hybrid composites and the lowest was observed in 25H/15F epoxy hybrid composites. In the case of hybrid composites, the error % was similar whereas in pure composites it was different.

4. Discussion

Most of the previous research work has have been carried out on natural fiber reinforced thermosetting or thermoplastic hybrid composites. Natural fibers such as banana, okra, jute, bamboo, and pineapple were used as a constituent of composite material [64–66]. The present research was mainly focused on the impact of matrix material on the mechanical properties of a composite material. To draw a contrast between matrix materials, epoxy and ecopoxy (bio-based resin) were used as a matrix material.

The hemp- and flax-fiber-reinforced epoxy composites showed superior properties to the ecopoxy hybrid composites. In addition, hybrid composites exhibited superior properties to individual fiber composites. The results showed that epoxy-based hybrid composites had flexural strength properties ranging from 58 MPa to 80 MPa whereas in the previous research this was between 45 MPa to 64 MPa. Similarly, there was also a considerable development in the interlaminar shear properties. From contact angle measurement analysis, previous results showed that all the natural fiber composites exhibited hydrophilic surface properties (contact angle less than 70 degrees). However, in the current research, ecopoxy hybrid composites showed a contact angle close to 90 degrees which means that composites tend to show hydrophobic surface properties.

5. Conclusions

In the present research work, the effect of matrix material on the mechanical properties of composite material was investigated by drawing a comparison between the epoxy-resin-matrix natural fiber composites and ecopoxy (bio-resin)-resin-matrix natural hybrid fiber composites. The results are given below.

Epoxy-matrix-based hybrid composites showed superior mechanical properties to ecopoxy-matrix-based hybrid composites. It was observed that the mechanical properties of epoxy-resin hybrid composites were almost two times greater than ecopoxy composites. In both cases, pure composites showed the poorest mechanical properties such as flexural strength and interlaminar shear strength and hybrid composites exhibited the maximum strength and shear properties. In the case of epoxy composites, the maximum flexural and shear strength were observed as 78 MPa and 13MPa, respectively, whereas in the case of ecopoxy composites, values were 30MPa and 10 MPa, respectively. The wettability properties of natural fiber hybrid composites were investigated by conducting contact angle measurement. The results showed that all the composites showed hydrophilic ($\theta < 90°$) surface properties. The morphological studies and chemical composition presented in the composites were investigated by using SEM-RDX spectrum analysis.

To validate this research work, a numerical analysis was carried out including the experimental analysis. Theoretical studies were carried out to determine the elastic properties based on the modified Halpin–Tsai method. It was observed that the error % was less with the numerical methods but in the case of Halpin's method, it was more.

Hence Halpin's method is not suitable for predicting the elastic properties of natural fiber polymer composites.

Overall epoxy-based natural hybrid composites showed improved mechanical properties compared to ecopoxy-based hybrid composites. Hence the hemp and flax fiber reinforced epoxy matrix composites could be a potential replacement for synthetic fiber composites in various industries such as the automobile, construction, microfluidics, and biomedical industries.

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Influence of Filler Materials on Wettability and Mechanical Properties of Basalt/E-Glass Woven Fabric–Reinforced Composites for Microfluidics

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Abstract: This paper presents the development of novel hybrid composites in the presence of filler particles and manufactured using a proposed new fabrication technique. The hybrid composites were fabricated using a basalt and E-glass woven fabric–reinforced epoxy resin matrix combined with graphite powder nanoparticles. Six sets of samples were fabricated using the vacuum-assisted free lamination compression molding technique. After the fabrication, wettability, mechanical properties (tensile, flexural and impact properties) and moisture properties were evaluated. Surface morphology and chemical composition of the composite samples were examined using a scanning electron microscope (SEM) and spectroscopy. The obtained results showed that the use of filler materials in hybrid composites improves the properties of hybrid composites. Basalt/E-glass hybrid composites with 10% graphite material exhibited superior mechanical properties over the other composites, with high-quality, improved adhesion and surface morphology. Thus, novel composites with the combination of exceptional properties may be integrated in the design of flexible electronics and microfluidics devices as a structural layer of the system. High flexibility and good surface tension of the designed composites makes them attractive for using the thermal imprint technique for microfluidics channel design.

Keywords: basalt; E-glass; epoxy resin; graphite particles; wettability; mechanical properties

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1. Introduction

The importance of natural fibers in the development of fiber-reinforced hybrid polymer composites is increasing day by day in the modern era because it is directly associated with environmental concerns as well as with a circular economy [1,2]. The combination of natural fibers with synthetic fibers in reinforced hybrid polymer composites offers several attractive features such as light weight, condensed lifecycle costs, high adhesion properties, flexibility and superior mechanical properties [3]. Nature is a decent source for the creation of composite materials using hemp, flax, sisal, palm [4], okra, banana, wood, bamboo, jute, sugarcane, or cowhide. Thus, all these natural materials can be a portion of the regular fiber composites, providing exceptional properties and expanding the application areas in various fields [5].

In recent years, synthetic fiber-based composites have become the leading progressive composite materials for automotive, construction, marine, sporting goods, biomedical, microfluidics and other engineering applications due to their ease of availability, high strength and modulus and also good corrosion and fatigue resistance behavior [6]. Despite the advantages to the presence of natural fibers in composite materials, there are some disadvantages, such as fiber shrinkage, poor adhesion and moisture, which may have a high effect on the mechanical properties. At the same time, most synthetic fibers are

brittle in nature, which may cause a catastrophic mode of failure in the composite material [7]. Synthetic fibers such as carbon, glass, mica and nylon also play a prominent role in the composite industry. Fiber-reinforced composite materials have a better strength and modulus than numerous conventional metallic materials. These composites exhibit low explicit gravity, high strength-to-weight ratio and higher modulus–weight proportions [8,9]. Moreover, natural fibers are lower in weight compared with metallic fibers and may be applied in many weight-basic applications such as those in the aviation, civil industry and automotive areas [10].

Many researchers found interest in developing hybrid composites. The reason can be attributed to the adverse effects of polymer composites on nature (environment), their high cost and other unfavorable mechanical properties [11]. The hybridization of polymer composites is known for the improvement of mechanical properties over individual fiber composites. It is obtained by joining two or more reinforcement materials under the same matrix (thermoset or thermoplastic) material. The possible combinations are natural fiber–natural fiber, natural fiber–synthetic fiber and synthetic fiber–synthetic fiber hybrid composites [12,13]. These hybrid composites have been established to overcome the drawbacks of individual fiber composites. Thus, properly configured hybrid composites produce high-quality composites and offer good mechanical properties such as strength, stiffness, flame retardance and corrosive resistance properties [14].

Among all the prominent methods for understanding the wettability properties of composite material, various droplet based techniques are used [15], such as gravitational [16,17], acoustic [18] and magnetic [19] driving methods. Among these droplet driving methods, contact angle measurement is widely popular due to its accuracy, lower operational cost and low complex procedure. The wettability of composite material is used to understand the material's surface properties, such as its hydrophilic or hydrophobic nature, and is also used to understand the surface tension of driven droplets. Thus, the mechanical properties of thermoset and thermoplastic resin-based hybrid polymer composites with synthetic fibers, such as carbon, nylon, glass, rayon, spandex, acrylic fibers and natural fibers, such as hemp, flax, sisal, jute, bamboo and palm fibers, have been broadly investigated by many researchers. Various researchers stated that hybrid composites have superior properties over individual fiber composites. Static and dynamic mechanical properties of basalt fiber–reinforced polymer composites showed their sensitivity to the strain rate. The mechanical properties such as tensile strength, elastic modulus and failure strain were increasing quickly with strain rate when the strain rate was over 120 s^{-1} [20]. Thus, replacing glass fiber with basalt fiber in the composites enhances mechanical properties by 35–42%, i.e., compressive strength, impact energy and Young's modulus [21], and replacing glass fiber with E-glass fiber composites gives superior tensile strength [21,22]. Moreover, when applying the plasma treatment to basalt fibers during the fabrication, the adhesion properties and surface roughness may be improved compared to those of commercial fibers [22]. The influence of waste basalt powder on rigid polyurethane foams had some influence, too. It was observed that the addition of basalt powder triggered an increase in the reactivity of the polyurethane system during the foaming process, but it caused a reduction of mechanical properties [23]. Additionally, the basalt powder can be used as an eco-friendly modifier for the epoxy resin, improving the thermo-mechanical properties for an increase in filler content [24]. The mechanical properties of composites reinforced with fabrics having the same areal density confirmed that basalt fiber composites possess an elastic modulus higher than the corresponding glass fiber composites, while their tensile strength approaches that of equivalent carbon fiber composites, and the results of fatigue behavior confirmed the better performance of basalt fiber composites than the corresponding glass fiber composites, with higher stiffness retention at low fatigue loads and better damping properties [25]. Thus, basalt and hybrid laminates with an added configuration exhibited higher impact energy absorption capacity than glass fiber composites applying hybridization with reinforcement material [26–32]. However, the lack of filler material in the composite fabrication may improve adhesion but significantly reduces its mechanical properties. To overcome these problems, an attempt was made in the present paper.

In this paper, basalt-fiber (BF) and E-glass-fiber (EF)-reinforced epoxy polymer composites including graphite filler material with various weight fractions (5%, 10% and 15%) were fabricated using the vacuum-assisted free lamination compression molding technique. After, the samples were allowed the heat treatment process and then tested for mechanical properties. The mechanical characterization was carried out by conducting tensile, flexural, impact, hardness and moisture analyses. The contact angle measurement technique was used to evaluate the wettability of composite materials and to understand the material's surface properties, such as its hydrophilic or hydrophobic nature and the surface tension of driven droplets. Surface morphology and chemical composition were examined using scanning electron microscopy (SEM) and spectroscopy methods. Thus, the obtained results proved the relevance of properly configured hybrid composites possessing high quality and good mechanical and adhesion properties together with qualitative surface morphology. The obtained results proved the relevance to use novel composites in the design of microelectromechanical devices, flexible electronics and microfluidics.

2. Materials and Methods

2.1. Materials and Fabrication

Basalt (B) and E-glass (E) woven fabrics were used as a reinforcement, and graphite (G) powder nanoparticles were used as a filler material. Epoxy resin, along with hardener, was used as a matrix material. Both basalt and E-glass fabrics have the same areal density of 198 g/m^2, and the fabrics were taken in a square shape (300 mm × 300 mm) to fabricate the composites. The mechanical properties and chemical characteristics of the materials were considered before the experimentation in order to understand the nature and behavior during the process (Table 1).

Table 1. The mechanical properties of materials [33–36].

Property	Basalt (B)	E-Glass (E)	Epoxy Resin
Density (g/cm^3)	2.40–2.72	2.48–2.62	1.15–1.18
Diameter (μm)	15	6	-
Tensile Strength (MPa)	4800	3200	72
Young's Modulus (GPa)	90.0	70.0	2.9
Elongation (%)	3.15	4.00	5.30
Maximum Temp. (°C)	645	450	-
Color	Brown	White	Pale

A total of six sets of hybrid composites were fabricated using the vacuum-assisted free lamination compression molding technique. In each set of composites, a total of six sheets were taken to fabricate 3 mm thickness composite plates. To make the hybrid composites with filler material, the graphite powder nanoparticles with varying weight fractions (5%, 10% and 15%) were mixed in epoxy resin for 20 min and then applied to the composite panels. The ply sequence and orientation for each composite are described in Table 2.

Table 2. Ply sequence and orientation of basalt and E-glass hybrid composites.

Composite	No. of Plys		% Of Graphite Filler Material	Ply Sequence	Orientation (°)
	B	E			
B	6	0	0	B-B-B-B-B-B	0-90-0-90-0-90
E	0	6	0	E-E-E-E-E-E	0-90-0-90-0-90
B/E	3	3	0	B-E-B-E-B-E	0-90-0-90-0-90
B/E/G5	3	3	5	B-E-B-E-B-E	0-90-0-90-0-90
B/E/G10	3	3	10	B-E-B-B-E-E	0-90-0-90-0-90
B/E/G15	3	3	15	B-E-B-E-B-E	0-90-0-90-0-90

After degassing, a nominal pressure of 1.5 MPa was applied, and the composite panels were cured under the same pressure and vacuum for 8 h at room temperature. Then, the composite panels were taken for a heat treatment process for eliminating the moisture content, which also serves as a post-curing process for composites. The heat treatment process was carried out for 8 h at 90 °C. Total of six sets and in each five samples were developed using different materials and filler concentrations (in total 30 samples manufactured). Thus, the composite notations are considered as pure basalt (B), pure E-glass (E), basalt and E-glass (B/E), basalt and E-glass with 5% filler (B/E/G5), basalt and E-glass with 10% filler (B/E/G10) and basalt and E-glass with 15% filler material (B/E/G15) (Table 2).

2.2. Analytical Equipment

After the fabrication process, composites were allowed wettability testing and mechanical characterization by conducting various tests. In each set of composite groups, samples were tested according to ASTM standards (30 measurements in total). Thus, performed mechanical tests were: the flexural test, tensile test, impact test and moisture analysis. The morphological studies were carried using the scanning electron microscope (SEM) and the chemical compositions of the composite samples were tested using the SEM-EDX spectrometer.

Since fabricated hybrid composites contain the filler material throughout the surface, and in future research are intended for use in flexible electronics and microfluidics devices, the wettability property of the fabricated composite surface plays a crucial role. The same test was conducted on composites without filler material for comparison purposes as well. The wettability properties of novel composite materials were measured using the contact angle measurement technique (Figure 1).

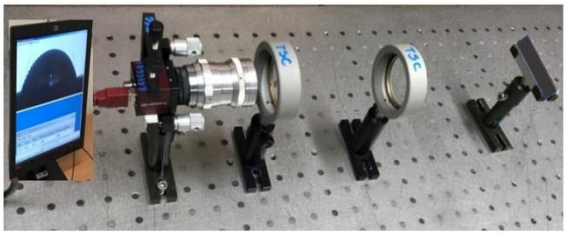


Figure 1. A contact angle measurement setup.

The experiment was carried out in a dim laboratory space with ambient lighting. It was vital for the measurement accuracy and image analysis that the liquid drop look black, and thus the light settings were set up accordingly. The experiment was set up to eliminate any light reflections that would have tampered with the measurement (Figure 1). Additionally, precautions were taken to keep airborne contaminants such as dust and particulates from contaminating the drips. Additionally, the critical distances between the parts were kept constant during the measurements (Figure 2).

Further, flexural tests were performed on fabricated samples to analyze the flexural strength and modulus, i.e., three-point bowing tests were performed on each set of composites according to ASTM D-790M-86 testing standards (total of 30 measurements). The dimensions of the samples were considered as 100 mm × 25 mm × 3 mm (length, width, thickness). The three-point bowing tests were conducted on an electronic tensiometer (METM 2000 ER-1 model (Plate II-18), provided by M/S Microtech Pune, Maharashtra, India), with outside rollers in diameter of 70 mm. Thus, composites were loaded at a strain rate of 0.2 mm/min (Figure 3).

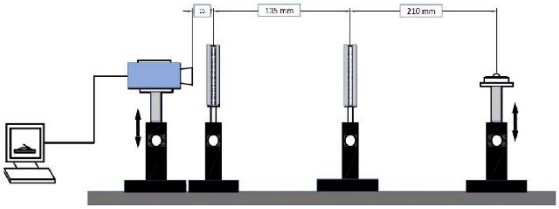


Figure 2. Critical distances between contact angle testing device.



Figure 3. Electronic tensiometer for flexural and tensile properties testing.

Tensile tests were performed on the same electronic tensiometer which was used for flexural tests, at a crosshead speed of 0.2 mm/min. An electronic micrometer was utilized to quantify the necessary thickness and width of composite examples. The check length, width and thickness were estimated with a 0.001 mm negligible count mechanized micrometer. This electronic tensiometer was fixed with weight and expansion pointers, which had an insignificant count of 0.01 kg and 0.01 mm, independently. The tensile tests were performed according to ASTM-D3039 testing standards. The specimen dimensions were considered 165 mm × 20 mm × 3 mm (length, width, thickness). In each set of composites, a total of five samples were tested (total 30 measurements).

Impact tests were performed on a falling dart impact testing machine (Impact analyzer M/S International equipment, Mumbai, India). The composite samples were tested as per ASTM D256-97 testing standards, and the specimen dimensions were 63.5 mm long, 12.36 mm wide and 3 mm in thickness. Designed composites were fixed in the vertical position and a pendulum was used to break the composite. The pendulum sled was delivered from the locking position, which is at a state of 1500 mm and hits the sample with a striking rate of 2.46 m/s. The sample was stripped and energy was exhibited in joules by the pointer on the specific scale.

The amount of moisture absorbed by the composite samples when exposed to moisture was analyzed using the moisture analysis test. The tests were performed according to ASTM-D 570 testing standards. The sample specifications were considered as 60 mm long, 20 mm wide and 3 mm in thickness. The samples were placed in a container full of water at room temperature and the amount of moisture gained by the composites was measured for six weeks' duration (readings taken after every 7 days).

Finally, morphological studies such as surface morphology, porosity content and fracture mechanism were investigated using SEM and the chemical composition of the composite samples was analyzed by using SEM-EDX spectroscopy.

3. Results and Discussion

The basalt and E-glass fiber hybrid composites (B, E, B/E, B/E/G5, B/E/G10 and B/E/G15) in the presence of graphite filler (with varying weight fractions 5%, 10% and 15%) were tested to evaluate their mechanical characteristics, wettability and surface morphology.

3.1. Wettability Analysis

The wettability properties of composite materials were investigated using the contact angle measurement technique imaging water droplet images on the composite surface (Figure 4). The volume of the water droplet was kept constant and the values are noted from the left lower endpoint to the right lower endpoint.

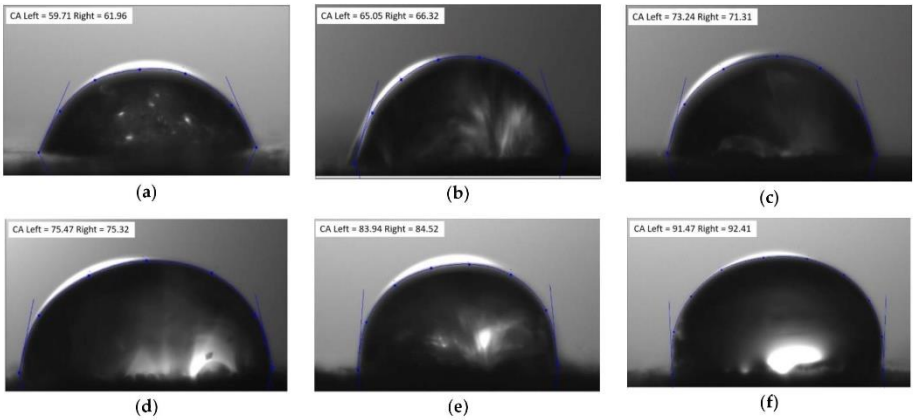


Figure 4. The droplet profile's polynomial fit was achieved by using knots that go from the left to the right while accounting for the contact angle (**a**) B, (**b**) E, (**c**) B/E, (**d**) B/E/G5, (**e**) B/E/G10 and (**f**) B/E/G15.

Results (Figure 5) showed the assessment of contact angle measurement of basalt and E-glass woven-fabric hybrid composites and that all sample surfaces exhibited hydrophilic properties. The measured contact angle formed by the water droplet on the B/E/G15 hybrid composite was 86.45° with an error of ±2.36°, being the maximum among all, whereas for individual basalt (B) composites, the least contact angle of 61.76° with an error of ±1.84° was exhibited. Thus, if the contact angle is less than 90°, then it is considered hydrophilic, and if it is more than 90°, then it is hydrophobic [37–39]. Moreover, hybrid composites with graphite filler material tended to exhibit hydrophobic surface properties. To extend the analysis, the same tests were conducted varying the measurements in time, and the results showed that the contact angle decreased with an increase in time (at the continuous intervals of time such as 10 s, 20 s, 30 s) in all the cases due to the seeping of a water droplet on the composite surface. The hydrophilic surface properties of the composite material play a prominent role during the formation of micro channels on solid surfaces (composite material).

3.2. Flexural Properties

The flexural properties, such as the flexural strength and flexural modulus, of basalt and E-glass woven-fabric reinforced with epoxy matrix composites, including filler material (graphite powder particles) were investigated (Figure 6). Results showed that, the strength and modulus were superior in hybrid composites compared to pure composites. The flexural properties increased with filler material and the impact of hybridization was observed, i.e., there was a slight increment in both strength and flexural modulus in basalt and E-glass hybrid composites over pure basalt and pure E-glass woven-fabric composites. The epoxy composites compared to pure basalt (B) composites exhibited the least flexural strength and modulus at 27.82 MPa and 2.06 GPa, respectively. Thus, basalt and E-glass

(B/E/G15) hybrid composite with 15% graphite filler material showed the highest values of 43.81 MPa flexural strength and 3.57 GPa flexural modulus (Figure 7). Many researchers found that the incorporation of filler materials into the composite material increased both flexural strength and flexural modulus [40,41]. The reason is attributed to the uniform distribution of filler material and the strong adhesion properties between polymer matrix and filler material. Hence, the composites with graphite filler material exhibited superior flexural strength and modulus properties.

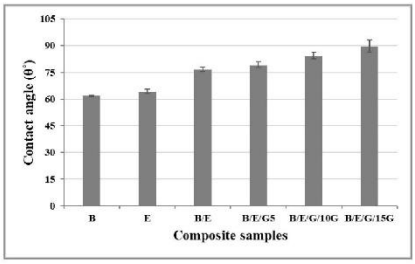
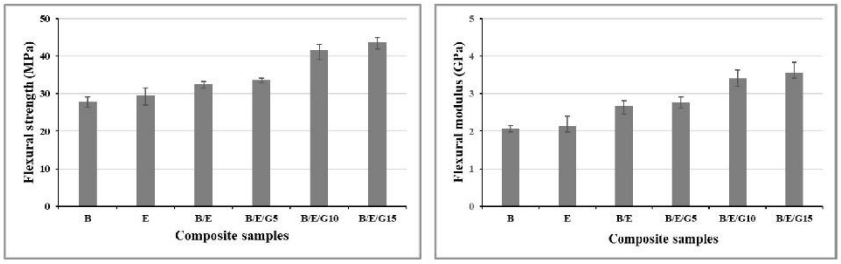
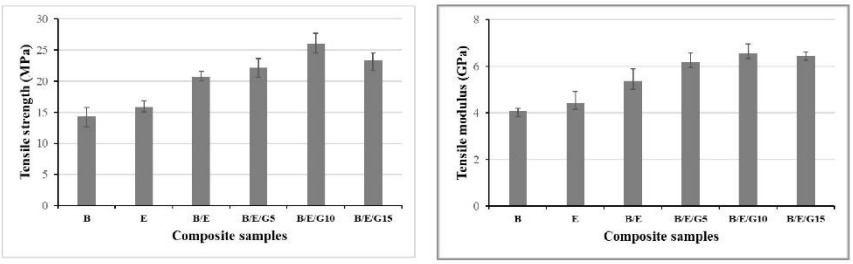


Figure 5. Contact angle measurement for composite samples.



(**a**) (**b**)

Figure 6. Flexural strength (**a**) and flexural modulus (**b**) of basalt and E-glass composite samples.



(**a**) (**b**)

Figure 7. Tensile strength (**a**) and tensile modulus (**b**) of basalt and E-glass composite samples.

Experimental investigations showed that incorporation of the filler in to different matrices played a crucial role in improving the mechanical performance of the designed novel composite materials.

3.3. Tensile Properties

The experiments with basalt and E-glass woven-fabric composites were conducted by performing tensile testing (Figure 7). Thus, the obtained results showed, that the impact of filler material in the hybrid composites was quite noticeable and effective. Unlike the flexural properties, the tensile strength and tensile modulus were increased up to 10% with a graphite filler material. Results for basalt and E-glass (B/E) hybrid composites and basalt and E-glass hybrid composites with 5% graphite particles (B/E/G5) were almost identical. The epoxy-based basalt fiber composite showed the least tensile strength and modulus of 14.68 MPa and 4.07 GPa, respectively (Figure 7a,b). Thus, epoxy-based basalt and E-glass hybrid composites with 10% filler material showed the highest results, i.e., tensile strength of 27.68 MPa (Figure 7a) and tensile modulus of 6.25 GPa (Figure 7b). Hence, the composition in B/E/G10 could act as a very good reinforcing filler in the hybrid composites.

The presence of a filler material in the composites may improve the adhesion properties and also reduce the porosity content. The hybrid composites in the presence of graphite filler material possessed great strength and modulus over the individual fiber composites. Hence, these exceptional properties lead to a potential replacement of other conventional materials used for microfluidics, in the formation of various dimension groves and channels or in absorbent-coated microchannels.

3.4. Impact Properties

Further, investigations were performed on the impact energy absorbed by the composite samples (Figure 8). It was defined that the amount of energy was absorbed by an object when an external load (impact load or sudden load) was applied to it. Tests were conducted using a Charpy impact tester at room temperature with a relative humidity of 30%. The results followed previously obtained results, i.e., the impact energy increased with an increase in filler material. The pure basalt composites showed the least impact energy at 3.1 kJ/m^2, and B/E/G15 hybrid composites showed the highest at 4.9 kJ/m^2. Furthermore, the B/E hybrid composites and B/E/G5 hybrid composites had almost the same impact energy. Composite B/E/G15 exhibited the highest impact energy, and the pure composites showed the lowest impact energy values (Figure 9). This rise in hybrid composites is the result of energy-dissipating fiber-related phenomena such as fiber debonding, pull-out, bridging and fracture, which cause the resin matrix to flex plastically prior to failure. A set of glass fibers with a length greater than the crucial value for effective reinforcement is likely to result in bridging and fiber fractures, while a set of glass fibers with a length less than the critical value is likely to result in debonding and fiber pull-out. Similar to these results, many researchers reported that the hybridization of composites improves the impact strength [42,43]. The same was observed in this research, stating that the hybridization of composites improved the impact strength by two times.

3.5. Moisture Analysis

To measure the amount of moisture absorbed by the composites, a moisture analysis test was conducted (Figure 9). After fabrication, the composite samples were placed in an electric oven to eliminate the moisture in them. To analyze the moisture properties, the composite samples were placed in a jar containing distilled water. The readings were noted by differentiating the initial weight and the final weight (after moisture gain) of the composite samples for 6 weeks. After 6 weeks, results showed that the presence of the filler material in a composite sample absorbed less moisture (Figure 9) because the filler material occupied the porosity content in a composite. Thus, pure samples absorbed more moisture than hybrid composites, meaning that the hybrid composites with filler particles

have the property of repelling the water when it comes into contact with it. This occurrence was due to unbalanced molecular forces in the water and the solid interface bringing out surface tension. It was observed that the increase in moisture increased over time in all the composites, and after about 5 weeks it reached a steady state.

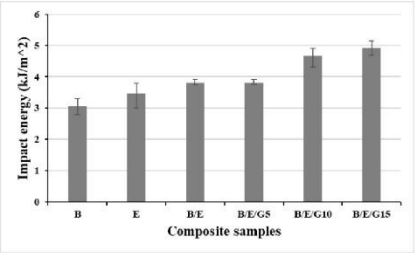


Figure 8. Impact energy of basalt and E-glass composite samples.

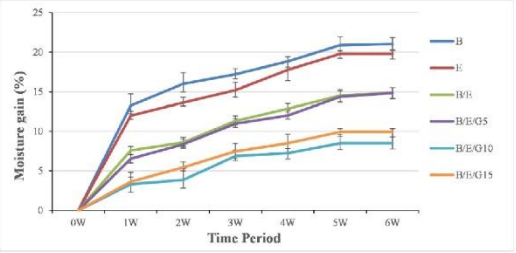


Figure 9. Moisture absorption of basalt and E-glass hybrid composites.

3.6. Morphological Studies

The surface morphology and porosity formation in basalt and E-glass woven-fiber hybrid composites were studied using the SEM. Thus, the presence of pores in a composite may affect the mechanical performance of a composite material. Previous researchers focused on evacuating the air trapped in voids by keeping the volatiles in solution and resin intrusion of the evacuated pore spaces. However, these methods can drive additional problems such as shrinkage and are not suitable for all composites [44,45]. Hence, the graphite powder particles were introduced into the composite material to avoid the microbial effect in it. The SEM results showed that the presence of pores in the form of spherical, cylindrical and helical shapes were observed in the hybrid composites without filler particles (Figure 10a–c). The percentage of porosity in hybrid composites with graphite filler material was smaller when compared to the pure composites (Figure 10d–f). Most of the pores were replaced with nanoparticles in the B/E/G hybrid composites and that can be attributed to improving the mechanical performance and adhesion properties.

The morphological studies were conducted on fractured hybrid samples to understand the chemical composition of the composites. It was observed from SEM-EDX results that hybrid composites exhibited good adhesion properties as well as the surface roughness on composite materials (Figure 11). The map data of chemical composition allowed to evaluate the dominant materials in designed composites: the color notations are red for carbon (C), green for oxygen (O), blue for chlorine (Ci), yellow for calcium (Ca), light blue for sodium (Na) and orange for aluminum (Al). The dominance of carbon in the B/E/G10 hybrid composite was observed clearly, i.e., it was more at fracture over the surface position. In

the B/E hybrid case, the presence of oxygen on the surface position was observed. Thus, a detailed description of atomic and weight concentrations is given in the Table 3.

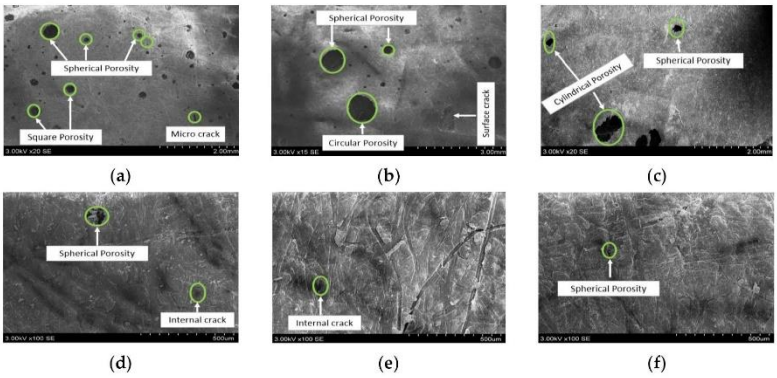


Figure 10. Porosity content in basalt and E-glass woven-fiber hybrid composites (**a**) B, (**b**) E, (**c**) B/E, (**d**) B/E/G5, (**e**) B/E/G10 and (**f**) B/E/G15.

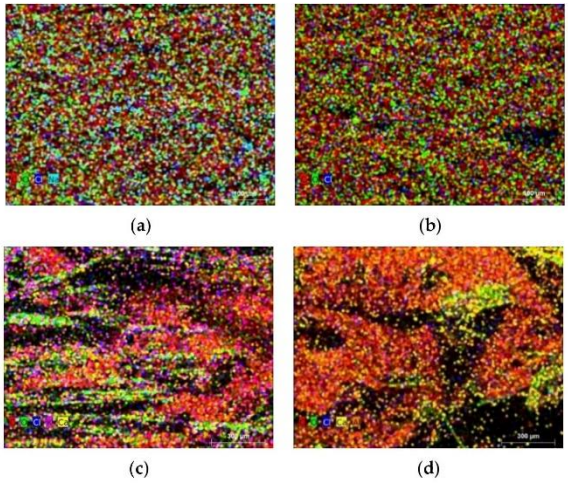


Figure 11. SEM images for map data of hybrid composites (**a**) B/E at surface, (**b**) B/E/G10 at surface, (**c**) B/E at fracture and (**d**) B/E/G10 at fracture.

The chemical composition of basalt and E-glass woven-fabric hybrid composites with and without filler materials was analyzed by using SEM-EDX spectroscopy, too. The chemical composition of a hybrid composite at surface and fracture position was observed for comparison purposes. The highest peak was observed for carbon followed by oxygen. The results stated that the carbon percentage was higher at a fractured place than the surface position (Figure 12).

Table 3. The chemical composition of basalt and E-glass hybrid composites at surface and fracture position.

Element	B/E Surface		B/E Fracture		B/E/G10 Surface		B/E/G10 Fracture	
	Weight (%)	Atom. (%)	Weight (%)	Atom. (%)	Weight (%)	Atom. (%)	Weight (%)	Atom. (%)
Carbon (C)	39.85	48.31	53.56	58.98	67.62	73.24	78.09	81.86
Oxygen (O)	58.77	51.10	45.18	40.57	31.89	26.51	10.85	16.74
Calcium (Ca)	0.00	0.00	0.42	0.14	0.24	0.11	0.08	0.10
Chlorine (Cl)	0.94	0.39	0.00	0.00	0.17	0.08	1.92	1.28
Sodium (Na)	0.44	0.20	0.28	0.13	0.08	0.06	0.00	0.00
Silicon (Si)	0.00	0.00	0.56	0.18	0.00	0.00	0.06	0.02

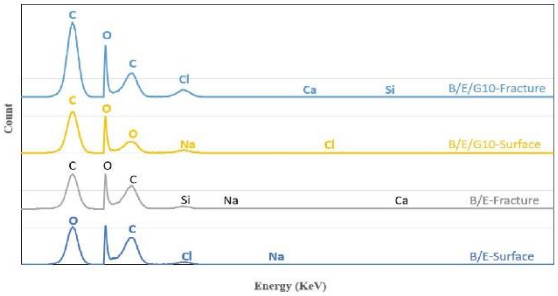


Figure 12. SEM-EDX spectrum analysis on hybrid composites at surface and fracture position.

Due to the presence of graphite particles in B/E/10G, the percentage of carbon reached 78.09% at the fracture position, whereas at the surface position it was 67.72% (Table 3).

Overall, the results proved that the mechanical performance of basalt and E-glass woven-fabric hybrid composites increased with the addition of filler material. The effect of the hybridization concept on woven-fiber laminates was quite impressive. Thus, the designed novel hybrid composites in the presence of filler material can be useful for various engineering applications such as automobiles, construction, biomedical and microfluidics applications. The deeper research into microchannel formation was left for future research.

4. Discussion

Novel composite materials with graphite filler were designed for the future design of composite microfluidics chips. Since microfluidics channels are usually made by layering, these channels need to be perfectly aligned on top of each layer. Moreover, the behaviors of fluidic flow mainly dominate because of the interfaces between the fluids and the microchannels' surface walls, playing critical roles in the regulation of the microflow in the channel. The pressure in microfluidics channel is an important factor, too. Too much pressure in the channel could cause delamination or fracture, depending on the strength of the bonding. Thus, evaluation of surface morphology, wettability and mechanical properties, chemical bonding, etc. are crucial in the design of composites used as the base material for the formation of channels in microfluidics.

Implementation of novel composite materials opens the opportunity to integrate high-complexity physical and chemical surface modification strategies in the design of microchannels in microfluidics devices. The future work will be focused on the formation of such microchannels using designed composite materials as the basic material, evaluating these materials from the perspective of interfacial interactions between the inner surface and the fluid and determining the behavior of the microflow.

5. Conclusions

Basalt and E-glass woven-fiber-based epoxy hybrid composites in the presence of graphite filler material were fabricated using a vacuum-assisted free lamination compression molding fabrication technique.

Novel hybrid composites showed superior mechanical properties over individual basalt and E-glass fiber composites. The addition of filler material to the composite material improved its mechanical performance. It was observed that all the composites exhibited hydrophilic surface properties with a contact angle of less than 90°, and the contact angle increased with an increase in filler material in the composite. From the flexural results, the individual basalt fiber composites exhibited the least flexural strength of 27.82 MPa and flexural modulus of 2.06 GPa. Thus, the B/E/G15 composite showed the highest flexural strength of 43.81 MPa and flexural modulus of 3.57 GPa. From the tensile results, the epoxy-based B/E/10G composites exhibited the highest tensile properties, and epoxy-based basalt composites showed the lower tensile properties. The impact energy values observed in individual basalt and E-glass were 3.1 KJ/m^2 and 3.5 KJ/m^2, respectively. The amount of moisture gained by the composites was investigated using moisture analysis. It was observed that all the composites followed the same trend for up to 5 weeks and remained the same even after a 6-week period.

Moreover, the porosity content in hybrid composites with filler material was less than that of composites without filler material. The presence of filler material in hybrid composites showed a sudden peak in carbon weight concentration, proving that incorporation of filler material and the application of the hybridization concept gave superior mechanical performance. Thus, the obtained exceptional composite properties enable future research in the application of microfluidics and microchannels.

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