Hybrid Fiber/Filler Reinforced Vegetable Oil-Based Composites: A Short Review

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Abstract. The development of filler- and fibre-reinforced vegetable oil composites has received considerable attention in recent years due to their environmental friendliness and potential to replace synthetic composites. Vegetable oil composites can be reinforced with either natural or synthetic fillers/fibres, resulting in partially or fully green composites. However, synthetic fibers have the disadvantage of being non-renewable and unsustainable due to their production from petroleum-based sources. To address this limitation, there is a growing trend toward hybridizing two or more types of filler/fibers as a hybrid reinforcement system, which can improve the supporting properties of the composites. Therefore, this review article specifically focuses on the use of hybrid filler/fibers in vegetable oil-based composites, including the various types of hybrid fiber/fibers, vegetable oils used, and their potential applications. Additionally, the mechanical and thermal properties of these composites are also being reviewed.

Keywords: Hybrid · Composites · Vegetable Oil · Mechanical Properties · Thermal Properties

1 Introduction

In recent years, plant oils have gained attention as a potential source for creating resins and pre-polymers that could replace petroleum-based resources. Vegetable oils such as soybean, linseed, cottonseed, and castor oils have shown promise in the development of composite materials due to their abundance, low cost, and renewable nature [1]. These oils can be functionalized through the addition of crosslinking agents to create crosslinked functionalized oils, which can then be reinforced with natural fibers to enhance their mechanical and thermophysical characteristics.

The utilization of natural or synthetic fillers/fibers in vegetable oil-based composites can make them partly or fully eco-friendly. There are two categories of fillers/fiber reinforcements: natural and synthetic. Natural fibers comprise cotton, flax, hemp, coconut, flaxseed, kenaf, and jute, while synthetic fibers include glass, carbon, nylon, rayon,

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acrylic, and Lycra [2]. Figure 1 demonstrates the classification of fibers, while Table 1 provides annual statistics on natural fiber production. Industrial applications typically use glass fibers because of their strength and stiffness compared to natural fibers [3].

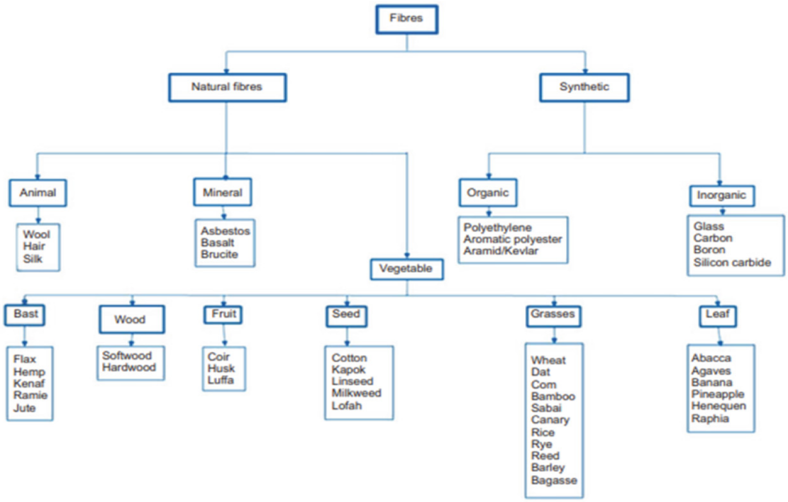

Fig. 1. Classification of the fibers [4]. Open access.

However, synthetic fibers have certain drawbacks such as poor renewability, high production cost, poor recyclability, high energy consumption, machine abrasion, and health risks [4, 5].

In recent times, natural fibers have emerged as a promising alternative to petroleum-based fiber products owing to their eco-friendliness and renewability, providing an edge over synthetic materials [4]. Despite these advantages, natural fiber composites have some disadvantages, including poor chemical and fire resistance, low liquefaction point, weak interaction between the matrix and the fibers, and low moisture absorption [4, 6]. As a result, plant fibers must undergo surface treatment using chemical or physical methods before being used in composites. Surface properties should be modified to reduce flammability and water absorption for best results [7]. Alkali and silane are commonly used chemical treatments [8].

Hybridization is an active area of research because combining two or more fillers/fibres to produce composites can create sustainable materials with improved properties. Hybrid fillers/fibres can compensate for the disadvantages of one type of reinforcement with another, resulting in composites with higher stiffness and strength than single reinforced polymer composites [5]. The three types of fillers/fibers used in hybridization are fiber-fiber, filler-fiber, and filler-filler. Hybrid composites have gained significant attention compared to single reinforced fillers/fibers composites, such as natural fiber composites or glass fiber compounds. The concept of hybridization is prevalent in various fields of study, including mechanics, polymer and chemical chemistry, metalworking, physics, science and technology, and energy sources, with the main goal

Table 1. The yearly manufacture of natural fiber [9].

Natural fiber	Origin	World production (x 10^3 Tons)
Coir	Fruit	100
Banana	Stem	200
Bamboo	Stem	10.000
Jute	Stem	2500
Hemp	Stem	215
Flax	Stem	810
Abaca	Leaf	70
Kenaf	Stem	770
Roselle	Stem	250
Ramie	Stem	100
Sisal	Leaf	380
Sun hemp	Stem	70
Cotton lint	Fruit	18.500
Wood	Stem	1.750.000
Broom	Stem	Abundant
Elephant Grass	Stem	Abundant
Linseed	Fruit	Abundant
Oil Palm Fruit	Fruit	Abundant
Rice Husk	Fruit/grain	Abundant

of mixing three or more elements to produce superior performance or qualities for the intended purposes.

In this context, the present study aims to provide an overview of hybrid filler- and fibre- reinforced vegetable oil-based composites and their mechanical and thermal properties. By understanding the properties and potential applications of these materials, we hope to pave the way for their wider use in various industries while reducing the environmental impact of the manufacturing processes.

2 Hybrid Fiber-Fiber Reinforced Vegetable Oil Composites

Hybrid fiber-reinforced thermoset composites offer many possibilities, and natural fibers, organic-synthetic fibers, and synthetic-synthetic fibers can be used, as shown in Fig. 2. In various applications, including medical devices, furnishings, structural elements, and vehicle parts, vegetable oil reinforced with hybrid natural fiber can replace synthetic fibers. However, creating these hybrid composites can be challenging, as their varied characteristics and cross-sectional adhesion must be considered [10]. Different processing techniques, such as simple mixing, lattice structures, and segmented sandwich laminates, can be used [11]. Natural fibers can be combined with other natural fibers or incorporated into a polymer matrix to balance cost while improving functionality and quality [10].

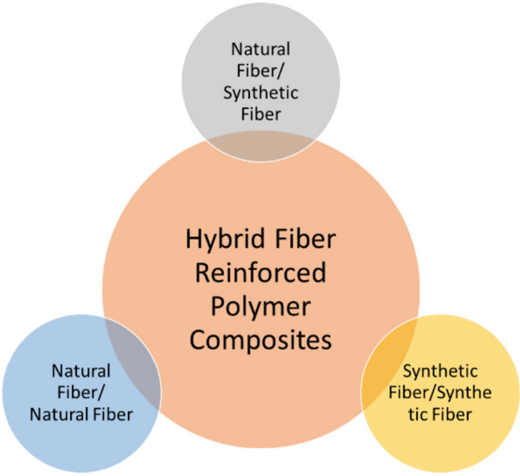

Fig. 2. An overview of natural fiber hybridization.

While research on natural-natural fiber reinforced hybrid composites as alternatives to synthetic fibers is ongoing, limitations exist when hybridizing natural-natural fibers due to factors such as fiber arrangement, matrix selection, interfaces tension, and permeability, which can affect natural fiber performance [4, 5].

For instance, in an experiment conducted by Hanan et al. [12], the tensile and flexural properties of oil palm empty fruit bunch (EFB) composites improved significantly when kenaf fiber content was increased, while pure EFB composites had better impact characteristics than hybrid composites. When natural-natural fiber reinforced hybrids are subjected to high strain, their tensile strength is maximized.

Furthermore, Uriquiza et al. [13] found that the hybridization of Henequen and ixtle yarn fibers as a reinforcement for bio-laminates improved viscoelasticity and increased fabric compaction compared to henequen laminate alone. The post-curing process of all the laminates investigated in this study was aided by an increase in the glass transition temperature.

Queiroz et al. [14] investigated the differences in reinforcement between interlaminar jute/glass fiber hybrids and pure jute and discovered that the mechanical properties of jute fiber-based composites were significantly improved by macroscopic hybridization of the interaction. The improvements in flexural strength were attributed to the higher interlaminar shear strength of the outer plastic layers.

Finally, Darshan et al. [15] conducted a study on silk fiber reinforced epoxy composites and basalt and hybrid fiber reinforced epoxy composites. Hybridization of silk/basalt fibers outperformed silk fiber-reinforced epoxy composites because the two reinforcing fibers act synergistically, resulting in an epoxy matrix with exceptional hardness, strength, modulus, and toughness.

In recent years, several researchers have conducted research on hybrid fiber reinforced vegetable oil composites. The development of vegetable oil composites using hybrid fibers as reinforcement in vegetable oil-based resins are still in progress owing to their potential as a sustainable and eco-friendly alternative to conventional thermoset resins/composites. There is limited literature available regarding hybrid fiber reinforced composites that utilize vegetable oil as a base. Few researchers have reported on this topic.

For example, a study conducted by Lascano et al. [16] carried out a comparative study on the flexural and impact strength of hybrid basalt-flax fiber reinforced vegetable oil-based epoxy composites made from basalt/basalt, basalt/flax, and flax/flax. According to their findings, the composite materials that utilized hybrid basalt/basalt exhibited superior properties compared to other composites. However, when one of the basalt fabrics in the face sheets was replaced with a flax fabric, it had a significant impact on the overall properties, particularly the flexural strength and modulus, while the impact strength remained largely unaffected.

On the other hand, Mustapha et al. [17] investigate the effects of hybrid kenaf and glass fiber reinforced acrylated epoxidized palm oil (AEPO)/epoxy filled nanoclay composites on water absorption properties. They found that using glass fibers as the outer layer of the composites (GKKG, G- glass, K- kenaf) and alternated layer KGKG exhibited the lowest water uptake than other layering sequences.

Motoc et al. [18], investigated the dynamic mechanical and thermal decomposition properties of flax/basalt hybrid laminates, which were made from an epoxidized linseed oil-polymer composite. The researchers examined the impact of maleinized linseed oil (MLO) and glutaric anhydride (GA) as crosslinking agents, analyzed the effects of stacking order as the primary factor, and assessed the dynamic mechanical and thermal decomposition properties of the composites. The findings showed that incorporating glutaric anhydride (GA) led to the production of relatively brittle flax and flax/basalt laminates with high hardness. The loss moduli decreased as the number of basalt layers increased. In addition, the glass transition temperatures (Tg) shifted from 70 °C to 59 °C and 56 °C with increasing MLO content in the GA:MLO curing agent system, resulting in lower brittleness of the crosslinked resin.

In the study by Santhosh and Rao [19], the mechanical and thermal behavior of cotton/nylon fiber hybrid composites were compared with and without the addition of castor oil. The results showed that the composites with castor oil exhibited higher tensile, flexural and impact strength than the composites without castor oil. The thermal properties of the composites were analyzed by DMA, which indicated an improvement in the damping properties of the composites with castor oil. In addition, the decrease in glass transition temperature for the composites with castor oil indicated improved damping performance at higher temperatures.

3 Hybrid Fiber-Filler Reinforced Vegetable Oil Composites

Another type of hybrid composites are filler-and fiber-reinforced vegetable oil composites, which are made by mixing several types of fillers and fibers with resins derived from vegetable oil. These composites are versatile and can be used in a variety of industries, including automotive, aerospace, construction, and consumer goods. The fillers used in these composites serve to improve their mechanical properties and can be either organic or inorganic. Some commonly used organic fillers include cellulose, lignin, and starch, while inorganic fillers can be made from materials like silica, clay, or calcium carbonate. Fibers are incorporated into the composites to reinforce and strengthen them. Natural fibers such as jute, hemp and flax are commonly used, but synthetic fibers such as glass and carbon are also used.

In a study by Arun Prakash and Viswanthan [20], researchers investigated the mechanical and thermal properties of a neem oil-blended epoxy composite reinforced with sea urchin tips and kenaf tissue fibers with surface modifications. The addition of neem oil to the epoxy resin degraded the mechanical and thermal properties, but transformed the epoxy composite into a bioform. Incorporation of surface-modified kenaf fibers into the epoxy-nem bio-mixture resulted in improved mechanical properties, while the use of surface-modified sea urchin particles further improved both mechanical and thermal properties compared to the starting material. In addition, water uptake results showed that the designations of the surface-modified composites remained unchanged after immersion.

Yang et al. [21] studied the tensile test, water resistance, and thermal analysis of bioplastic composites reinforced with epoxidized oils based on hybrid starch and empty fruit grapes. The researchers found that low epoxidized oil content (EO) resulted in smoother composite surfaces, while high EO content resulted in voids and discontinuities due to phase separation. The addition of epoxidized soybean oil (ESO) increased the thermal stability of the bioplastics due to its strong interaction with starch/EFB. In addition, a small amount of EO acted as a compatibilizer and improved the mechanical properties of the bioplastic composites, while a higher content of EO had a negative effect on the mechanical properties. The introduction of EO resulted in a moderate decrease in water absorption and solubility, but increased water vapour permeability.

Khandelwal et al. [22] conducted a study to evaluate the electrical, morphological, thermal, and mechanical properties of epoxy linseed oil (ELO) composites. These composites were composed of biobased materials and were filled with carbon nanotubes (CNTs) and polyaniline (PANI). Through morphological analyses, it was confirmed that both CNTs and PANI were distributed uniformly within the ELO matrix. Results showed that the tensile strength and elastic modulus of the composite increased considerably upon incorporating CNTs, indicating that the composite was reinforced. Furthermore, the toughness of the ELO/PANI5 composite improved when 0.1% CNTs were added, demonstrating an increase in the composite's impact strength.

4 Hybrid Filler-Filler Reinforced Vegetable Oil Composites

One of the options for hybrid composites is the hybrid filler-fiber reinforced vegetable oil-based composites. These composites are made by combining two different types of fillers with resins based on vegetable oil. The primary objective of using two types of fillers is to achieve a synergistic effect that enhances the mechanical properties compared to using a single type of filler.

The fillers used in these composites can be organic or inorganic, and common organic fillers include cellulose, lignin, and starch. Meanwhile, inorganic fillers can be made from materials like silica, clay, or calcium carbonate. Using two different types of fillers can enable the composite to benefit from the unique properties of each filler, which can result in a combination of properties that cannot be achieved using only one type of filler.

Kaatubi et al. [23] investigated an environmentally friendly method to improve the mechanical properties of soybean oil-based composites reinforced with flaxseed and aluminium tris hydroxide (ATH), as well as with an ethylene-methyl acrylate (EMA)-based polyamide nanocomposite. The inclusion of ATH particles into the organic polymer enabled the composites to maintain stiffness without sacrificing durability, while also improving barrier and material characteristics. The researchers conducted mechanical tests, such as tensile and impact strength, using the conventional ASTM test plan. The study's results highlight the possibility of achieving best-practice designs that maximize constituent interactions, thereby broadening the spectrum of microbiologically polymer nanocomposites.

Dutta et al. [24] investigated the potential of epoxidized soybean oil (ESO) as a compatibilizer and plasticizer in a nanocomposite of polyvinyl chloride (PVC)/rice husk ashand modified montmorillonite (OMMT). The team fabricated several composites, keeping the optimized ESO constant and varying the OMMT content. The composites were characterized and their properties studied using sophisticated instrumentation. The results showed that the addition of 1 phr OMMT significantly improved the tensile, flexural and hardness properties by 97%, 90% and 91%, respectively. In addition, the addition of OMMT increased the hydrophobicity of the composites, making them suitable for outdoor applications.

5 Disadvantages of Hybrid Fiber/Fiber Reinforced Vegetable Oil Composites

Most natural fiber, including kenaf fibers, must undergo chemical modification because they are highly polar, which makes them incompatible with non-polar polymers and leads to lower composite performance. Alkali treatments are a common method of modifying and treating cellulose fibers such as kenaf fiber. This is because alkali treatments are an inexpensive and relatively effective method of removing lignin and other soluble substances from the cellulose surface, increasing the roughness of the fiber surface and improving adhesion at the fiber-matrix interface [25]. The reaction for alkali treatment using sodium hydroxide (NaOH) is shown below:

$$\text{Fiber} - \text{OH} + \text{NaOH} — \text{Fiber} - \text{O} - \text{Na+} + \text{H}_2\text{O} \tag{1}$$

Although NFRPs have the potential to replace synthetic fibers, they have two major issues that limit their widespread application. First, they have a low processing temperature. Second, natural fibers have a poor mechanical strength that cannot rival the strength of synthetic fibers.

Therefore, recent studies have focused on developing hybrid composites with two or more types of reinforcing filler/fiber in a single matrix. Hybrid reinforced polymer composites are very attractive materials for high performance applications because they have the ability to develop products with better mechanical properties, environmental performance, and cost effective. There is a growing interest in enhancing polymer properties by employing hybrid natural-synthetic filler/fibers to replace existing natural fiber or glass fiber composites. Hybrid composites have properties that could not attained by mono-fiber composites. They offer more advanced composites with reduced cost, acceptable corrosion resistance, good mechanical properties i.e. flexural modulus, good strength properties, and good thermal stability.

Natural fiber/filler reinforced vegetable oil-based composites also are susceptible to water absorption, which can lead to swelling and degradation over time. This can be a major issue in applications where the composite is exposed to water or high humidity levels, as it can result in a loss of mechanical properties and reduced lifespan.

To mitigate this issue, researchers have been investigating different strategies to improve the water resistance of these composites. These include modifying the vegetable oil-based resins to enhance their water resistance, applying water-resistant coatings or treatments to the natural fibers and fillers, and blending natural fibers with hydrophobic synthetic fibers or fillers to create composites that are more resistant to water absorption. While some progress has been made in improving the water resistance of vegetable oil-based composites reinforced with natural fibers/fillers, there remains a need for further optimization to enhance their suitability for various applications.

6 Current Application on Hybrid Fiber-Reinforced Vegetable Oil Composites

The extensive mechanical properties exhibited by NFRP composites render them well-suited for various structural applications. These properties include impact strength, tensile strength, flexural strength, compressive strength, creep resistance, and fatigue resistance. Hybrid NFRPs are particularly attractive due to their low production cost, high strength-to-weight ratio, and ease of fabrication [26]. However, natural fibers have certain drawbacks, such as inconsistent raw material and property characteristics, poor water barrier capabilities, and unfavorable bonding behavior with the matrix. The variability in characteristics due to factors such as the plant section, climate, and plant age makes it difficult to utilize natural fibers in industrial applications, which has led to the development of hybrid biocomposites. Despite the common use of hybrid fiber as a reinforcer, little research has been conducted on composites made of vegetable oil.

Recently, researchers have been exploring the use of plant oils, such as soybean, linseed, cottonseed, and castor oils, as a replacement for petroleum-based resources in the creation of resins and pre-polymers. However, crosslinked functionalized oils have inferior mechanical and thermophysical characteristics, which can be improved by reinforcing them with renewable bio-fibers. Although fully biocomposites (matrix and filler of natural origin) are still in the research stage, hybrid materials already have a wide range of uses. For instance, Kamarudin et al. [27] discovered that composite materials made by epoxidizing fiber and resins derived from soybean oil are employed in the walls, floors, and roofs of residential and low-rise commercial buildings. These panels were produced using a resin vacuum infusion method or vacuum-assisted resin transfer moulding.

O'Donnell et al. [28] also produced panels from acrylated epoxidized soybean oils (AESO) and natural fiber mats (flax, cellulose, pulp and recycled paper, hemp) for indoor uses such as housing, building components, furniture, and vehicle parts. These materials exhibited good mechanical properties compared to woven AESO composites reinforced with E-glass fibers, although the flexural modulus values varied depending on the type of fibers used, although the latter exhibited higher strength. Guna et al. [29] mentioned that hybrid composites are used in the automotive, construction, sports, and medical industries. Many hybrid biocomposites, such as tanks, piping, and vessels, are being investigated as alternatives to glass- and fiber-reinforced composites.

7 Conclusion

In summary, the field of hybrid filler/fiber reinforced vegetable oil composites is relatively new, with ongoing studies exploring the use of multiple fillers and fibers with vegetable oil-based resins. There is still much to be learned about the benefits and limitations of these materials. As the demand for environmentally friendly materials in applications such as automotive and housing construction continues to rise, there is a need to develop compatible composites that are sustainable and eco-friendly. The properties of these composites depend on the types of resin, vegetable oil, fiber/filler, as well as the chemical and physical modifications used. Developing fully green composites is a challenging task, but further research can lead to innovative ways of creating high-performance composites with commercial viability and increased bio-based content. Therefore, future studies should focus on developing composites with higher mechanical performance and bio-based contents to meet the demand for eco-friendly materials.

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