REVIEW ARTICLE

A Review on Phenol-Formaldehyde Biocomposites

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Abstract: Due to the greater thermal stability, chemical resistance, and dimensional stability of Phenol Formaldehyde (PF) resin, it occupies a very special position in the resin field. Nowadays, natural fiber reinforced PF composite materials are widely used. The objective of this study is to discuss the property improvements of natural fiber reinforced PF biocomposites. This review paper discusses thermal, electrical, diffusion, viscoelastic, tribological, morphological, and mechanical and biodegradability properties. Biocomposites will be a substitute for plastics which provides properties of both natural and synthetic ones. The greater the pollution magnitude, the more devastating the impacts on people's health, the environment, and economic well-being. The main sources of pollution contributing to it are vehicle exhaust, open waste burning, lighting, heating and the combustion of various fuels for cooking. When compared with plastic materials, PF biocomposites are partially biodegradable, hence limiting the amount of pollution rate. Moreover, it has a wide range of applications, such as packaging, construction, automobiles, and household purposes. In short, this review aims to provide detailed information regarding PF biocomposites.

Keywords: Phenol formaldehyde (PF), resin, natural fiber, biocomposites, pollution, biodegradability.

1. INTRODUCTION

Most plastics are made from a base material like natural gas or oil. In addition, it is not fully degradable in nature. The introduction of composites is one of the major breakthroughs in the field of polymer science [1]. Biocomposites can be replaced with up to 60% of plastic material with natural fibers without losing the benefits of plastics. Biocomposites give an opportunity to combine the effects of plastics and fiber. Composite materials are designed to satisfy different requirements without compromising the integrity of the substances. Biodegradable composite offers a very much better solution to waste disposal problems associated with oil-derived plastics, and it is also economically friendly too in the market. The mechanical properties of biocomposites are comparable to glass-fiber reinforced plastics, such as tubes and sandwich plates [2-6]. Low density, appropriate stiffness, greater mechanical and disposition, and renewability are the main characteristics of natural fibers. Bio fibers are the principal component of the bio-composite. Biocomposite use is rapidly expanding in industrial applications, packaging, construction, aerospace, military, and fundamentals [7-12] fibers were used many years ago. Egyptians used linen cloth impregnated with resins and honey to wrap mummies as far back as history (5000-3300 BC) proving that natural fiber reinforced PF composites are better than glass fiber composites [13]. The effect of environmental harmfulness can be reduced by the use of natural fiber reinforced PF composites. It has wider acceptance at the international level [14, 15].

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2. PHENOL FORMALDEHYDE RESIN

Phenol formaldehyde resin is a synthesized polymer. These are synthesized by a condensation reaction. The first phenolic resin was prepared by polycondensation of phenol and aldehyde in 1860. Von Bayer first reported the reaction between phenol and aldehyde. Blümmer used the industrial condensation reaction for the first time in 1902 to produce novolac. They can be widely used for water resistance, thermal stability, chemical resistance, electrical insulation, dimensional stability, and low cost [16-20]. Phenolic resin is a heat-cured plastic formed from a carbon-based alcohol and a chemical called aldehyde. Formaldehyde is the raw material for this type of resin. Resin is a hard, heat-resistant material that can be mixed with a variety of other materials to be used in a variety of applications. This has a wide range of industrial applications in adhesives, casting and moulding and structural parts [21-25].

There are two types of PF resin, namely resoles and novolac. An excess of formaldehyde is made to react with phenol in the presence of a base catalyst in water solution to produce a low-molecular-weight prepolymer called a resole (Fig. **1**). Here, the resole frequently found in liquid form or

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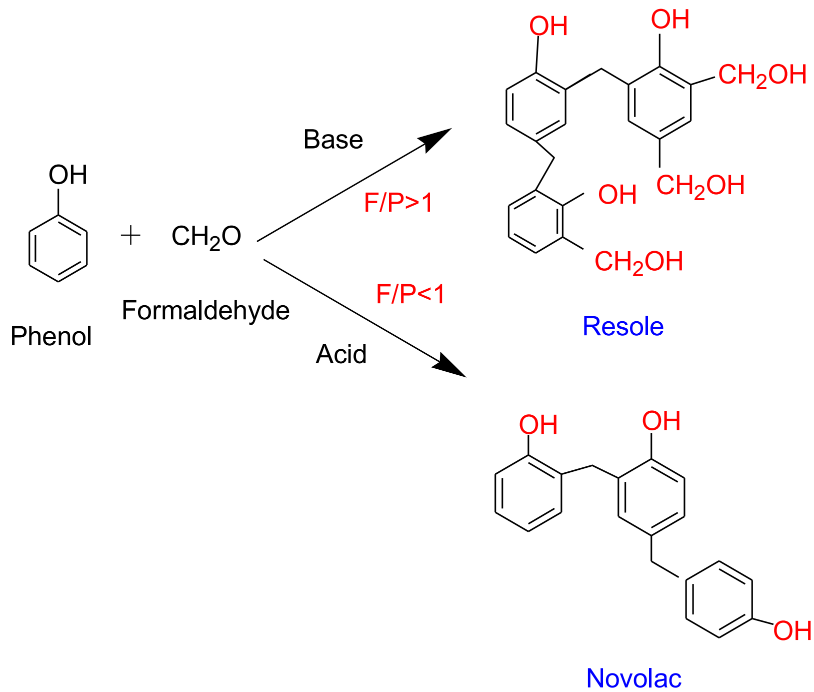

Fig. (1). Preparation of resole and novolac.

solution, is cured to a solid thermosetting network polymer [26-32]. For instance, compressing it between layers of wood veneer, and therefore, heating this assembly under pressure to form plywood. Another method involves the reaction of formaldehyde with an excess of phenol, in the presence of an acid catalyst. This process produces a solid prepolymer known as novolac. Novolac resembles the final polymer; lower molecular weight and thermoplastic too. Doesn't need to undergo chemical decomposition for softening. Only heating is required for softening.

3. PROPERTIES AND APPLICATIONS OF PHENOLIC RESIN

Phenolic resins are brown in color. Being green and red on occasion, phenolic moulding compounds typically appear as powder as opposed to pellets commonly seen in thermoplastic moldings; resin laminates usually appear as rods, tubes, or sheets. Pale phenolic resin is coloured immediately after production during storage or processing. The coloration is less intense only in the case of para-alkyl-substituted phenols. The viscosity of phenolic resins or their solutions is measured at a high concentration, *e.g.*, in a 30-80% solution. Cross-linked phenolic resins are hard substances that only have a small fracture strain and cannot be melted. Phenolic resins can be plasticized. Their compatibility with plasticizers can be adjusted by the introduction of hydrophilic or hydrophobic groups. Phenolic resins that are not cross-linked are commercially available as solids or solutions. Phenolic resins are reactive with both organic and halogenated solvents. But they have very poor compatibility with inorganic bases. Phenolic resins can be easily moulded into desired shapes and dimensions. Novolac is more dimensionally stable due to its dimensional accuracy and stability under normal conditions. But the resole-based compounds give off water. The water molecule is larger than the ammonia molecule. Resole-based resins are more water resistant. Only 0.003 to 11.75% of their water is absorbed. With these qualities, phenolic resins have low smoke emissions, so they can be used in architectural businesses [33-42]. Phenolic resins can withstand the intense conditions that are present in harsh chemical environments. The fundamental makeup of phenolic resin makes it capable of acting as an impermeable barrier, which shields a wide range of substrates from the corrosive effects of various substances. During extensive testing in laboratories, it has been determined that many compounds degrade very little after lengthy contact, frequently at high temperatures. Gasoline, alcohol, oil, glycol, brake fluid, a variety of hydrocarbons, and mild acids and bases are examples of common irritants that people are exposed to. Phenolic resins are frequently utilised to make the protective linings that are installed inside tankers that are used for the bulk transfer of acids and other corrosive chemical goods. Phenolic resin is extensively used for electrical applications like circuit board preparation and insulation for electrical equipment. WBP (weather and boil proof) plywood is exterior plywood. Phenolic resin is used for the preparation of exterior plywood. They have no melting point at all. They act as very good adhesives, anti-corrosive agents, and protective. Tasteless, odourless phenolic coating resins are good electrical insulators [43-47]. Phenolic resin was first used in combination with paper and fabrics as insulation for electrical equipment. Phenolic resin is used as a binder in cloth-based loudspeaker driver suspension components. These are widely used in the manufacturing of oriented standard boards. Due to its high brittleness, it must include fillers to bolster the resin integrity. Based on the nature of the filler, phenolic resin has different aspects. For example, glass fillers, which improve the heat resistance of resin, and mineral fillers affect the stiffness of resin. Thermoset polymers take longer processing times than thermoplastics. Due to its light weight and low cost, it is used to replace light bulb adhesives, smoker articles, and coil insulation [48-50]. Phenolic resins are widely used in technical applications and aerospace because they show high tensile and flexural properties. Binders made of phenolic resin are able to withstand the challenges posed by high temperature conditions, making them ideal for use in demanding applications such as refractory, friction, foundry, and aerospace products. Natural gas valves, automobile brake pistons and pulleys, hydraulic and water pump housings and seals are some examples of applications that take advantage of the dimensional stability offered by phenolic moulding compounds. Other examples include applications such as natural gas valves.

4. SCOPE OF REVIEW

Natural fibers are environmentally friendly, cheaper, and biodegradable. Natural fiber-reinforced biocomposites have good potential in the present scenario. This review is a compilation of the most notable research related to various types of natural fibers and their property improvements when reinforced with phenol formaldehyde resin. Mechanical properties, thermal, viscous-elastic, tribological, diffusion studies, bio-degradability, electrical, and morphological studies were analyzed. Natural plant fibers such as sisal, jute, coir, *etc*. are available abundantly in developing countries. Ecological concerns and the large amounts required for the synthesis of materials and products led to the discovery of natural fiber-reinforced phenol formaldehyde composites. Because it is more cost-helpful and less harmful to the environment than synthetic fiber reinforced polymer composites. Natural fibers such as cotton, flax, hemp, bananas, *etc*. can be recycled. Based on the origin of natural fibers, they are classified into bast, leaf, seed, core, grass, reed, wood, and roots [51, 52]. Lignocellulose is the major constituent in natural fiber. Lignocellulose is the most abundant material, with a primary production rate of 21011 tons. Compared to synthetic polymers, it is very high. So, this review aims at the effective usage of natural fiber reinforced PF composite, which can be used as a substitute for synthetic composites [53-55]. Each of the sections consists of the introduction of natural fiber reinforced composites and their uses.

5. TYPES OF NATURAL FIBER

In this last decade, increasing awareness of environmental protection makes concerns about protecting nature by replacing synthetic fiber and also looking for sustainable materials. Resultant studies lead to the use of natural fiber, instead of synthetic ones. These are environmentally friendly and cost effective [56] hair-like raw material, directly obtained from vegetable, animal and mineral sources.

Based on the botanical type classification of natural fibers, there are 6 types of fibers are present. That are,

1. Bast fibers *e.g.*: jute, flax, hemp, ramie and kenaf
2. Leaf fibers *e.g.*: sisal, banana, pineapple and agave
3. Seed fibers *e.g.*: coir, cotton
4. Grass and reed *e.g.*: rice,wheat, corn
5. Other types *e.g.*: woods and roots

5.1. Bast Fibers

Bast fibers are the fibers that lay under thin bark. They provide strength to stem and are originated from the phloem of a dicotyledonous plant. Ultimate fibers are the smaller units of strands of fibers. Ultimate fibers of Jute, kenaf, ramie, hemp, flaxis are very long [57].

5.1.1. Jute

India is the world's major exporter of jute clothes and materials. It is also grown in Burma, China, Brazil, and Bangladesh. Two plants that are grown on the Indian subcontinent are corchorus capsularis and corchorus olitorius. They are hygroscopic in nature. When plants are 8-15 cm tall, weeding and thinning are carried out. After plantation, the plants are grown as a single stalk. After being fully grown, the plants get mature and their colour changes from green to yellow. After fully maturing, a small budge of yellow flowers appears with a particularly sweet smell. From the flower, seeds are formed to be used for the next crop. The growing cycle for jute is 120-150 days. The average yield of jute bast fibers is 1700 kg/ha. Retting is the technique that is used to separate jute fibers from the stalk. These are the retting techniques used for jute fibers: dew retting, stagnant water retting, running water retting, and chemical retting. After retting, bundles of stalks must be dried to make the fibers stronger. Dried jute stalks are usually broken by passing through a fluted roller. That is called crushing. In this process, the roller breaks and reduces the woody particles into small pieces. This particle is removed from the fiber by a process known as scotching. Moreover, clean jute fiber can be extracted from fiber bundles by the hackling process. It may be manual or mechanical. Jute has a tensile strength of 5-8 cm/density, a density of 1.48 gm/cc, a moisture recovery of 13%, and ultimate fibers with an average length of 2 mm and a width of 20 m.

5.1.2. Flax

Among all natural fibers, flax is the oldest fiber crop in the world. Flax fiber comes from the stem of a plant called "Linum Usitatissimum". Plant fiber layers lie beneath the bark of the plant. The inner bark of this plant is composed of long, slender, thick-walled cells of which the fiber strands are composed. Flax fiber was the first bast fiber used by man for making textiles. Flax fiber stems have a diameter of 1.6 to 3.2mm. Flax plant cultivation in March or April has a very short growing period (100 days), producing fewer branches and longer finer fiber. When plants are flowered and seeds are ripened, the crop is pulled by root. Harvesting can be done by manual as well as mechanical methods. Flax fiber has been used to make linen for over 5000 years. It has a degree of polymerization of about 18,000. This means the flax polymer is about 18000nm long and about 0.8 nm thick. The polymer system of flax is more crystalline because of its longer polymers, which is spiral around each other at approximately the fiber axis, thereby contributing towards the tenacity and durability of the fiber. When the stem of the plant turns yellow and the seeds turn green to pale brown, the plants are pulled out by the roots. These are tied into bunches. During the 17th century, linen manufacture became established as a domestic industry in western Europe. The chemical composition of flax fiber is as follows: Cellulose: 75%, Hemicellulose, Pectin, Aromatics: 5%, Lignin: 4%, Fat/wax: 3%, Ash: 0.5% and Water: 12.5%.

5.1.3. Hemp

It is considered the oldest cultivated fiber plant in the world. Hemp is typically found in the northern hemisphere. It is a cannabis sativa plant species, grown specifically for the industrial uses of its derived products. Hemp is one of the fastest-growing plants. It is refined into a variety of commercial items, including clothing, biodegradable plastics, paint, insulation, and animal feed. Hemp is usually planted between March and May in the northern hemisphere and between September and November in the southern hemisphere. It matures in about 3-4 months. A total of 26 varieties of hemp with low levels of THC are certified by the EU. They have, unlike other types, very high fiber content of 30-40% hemp in cellulosic fiber. Chemical composition is about 77.5%, Cellulose, Hemicellulose: 10%, lignin: 6.8%, pectin: 2.9% and fat/wax: 0.9%. These plants are cut at 2 to 3cm above soil and left on the ground to dry. Mechanical harvesting is common for hemp. Once hemp is harvested, it goes through retting. This may take up to 5 weeks and produce a coarse fiber with light brown color. Water retting occurs when stems are bundled and then submerged in water, so bacteria can breakdown pectin. This takes 7-10 days and produces a better quality of fiber. Plant grows up to 4.5m (1.2-5m) in height, and having diameter 4-20mm.The last centuries show that hemp is fast growing and producing strong bast fibers. The average hemp bast has fiber length of 25mm& width of 25µm.

5.1.4. Ramie

Ramie is a flowering plant native to eastern Asia that belongs to the Urticaceae. It has smaller heart-shaped leaves which are green on the underside, and it appears to be better suited to tropical conditions. It is one of the oldest fiber crops, having been used for at least 6000 years and is also known as "China grass". Ramie requires chemical processing to de-gum the fiber. It is a fine, absorbent, quick-drying fiber that is slightly stiff and possesses a high natural luster.

The plant grows to a height of 2.5m and has an 8-fold greater strength than cotton. It is a very tough, durable fiber that is being used since ancient times. The Egyptians made fabric from ramie fibers. Ramie fiber is very similar to linen fiber, and it has been grown in China for many centuries, with farmers in ancient China using fiber to weave clothing. It is used to produce an open-weave fabric called mechera, which is used for shirts and dressing gowns suitable for warm climates. It is not as durable as other fibers, and so it is usually used as a blend with other fibers such as cotton or wool. It can reach a height of 1-2.5m.

These fibers are located in the cortex layer of the stem. Due to the gummy, pectinous nature, the separation of fiber includes scrapping, pounding, heating, washing, and chemical action. Fiber length averages 120mm and fiber width is 50m. Japan introduced an instrument for scrapping stalks using a pulping method, which can be isolated. It is also found as a blend with cotton in knit sweaters. That is also used in fish nets, canvas, strawnats, and fire hose [58, 59].

5.1.5. Kenaf

The genus of kenaf is Hibiscus. This is a plant in the Malvaceae family. It is grown mainly in China, India, Indonesia, Malaysia, and Bangladesh. It is sown from May to October. Growing quickly, reaching a height of 5-6 m in just 6-8 months. The temperature needs to reach 20 to 27 °C. Kenaf fiber can be separated from the stalk mechanically by retting. Jute and kenaf have almost the same retting process. Plant stems are immersed in water tanks for 7-14 days. Then it is washed, dried, and separated. The growing kenaf fiber has a length of 2.6 to 4 mm and a diameter of 17 to 21.9 mm and the cycle is of 150-180 days. Kenaf stem is composed of bast fibers, the large central area of stick fibers, outer bark, used to making ropes, paper, engineered wood, soil-less plotting mixes, animal bedding and packing materials. Kenaf fiber contains chemical composition as cellulose: 44 to 55%, Hemicellulose: 21%, pectin: 2% and lignin: 15 to 19% [60-63].

5.2. Leaf Fiber

5.2.1. Pineapple Fiber

Pineapple fiber is a Brazilian native and Anastas comosus is its genus name. Since fiber is obtained from leaves, the leaves have to be cut first from the plant. Then the fiber is pulled or split away from the leaf. Pineapple fiber is intensive, as each step is done mostly by hand. The leaves are 30-100 cm long [64]. Ultimate fiber has a 20m width and a length of 61.7mm. The fiber is lignocellulose in nature. The chemical constituents of various pineapple fiber constituents are -cellulose, pentosans, lignin, fat, nitrogenous matter, pectin, wax, and ash. Standard methods are used to determine the degree of polymerization and crystallinity of cellulose.

5.2.2. Sisal Fiber

Sisal fiber is obtained from the outer leaf skin by removing the inner pulp. It is exceptionally durable and low-maintenance with minimal wear and tear and is recyclable too and is about 1.5-2m tall, with sword-shaped leaves. The average life span of fiber is 7-10 years old. Sisal fibers are recyclable and anti-static, meaning they don't attract dust particles and absorb moisture [65]. Traditionally, sisal was the material used for agricultural twine. But now it has been overtaken by polypropylene. The ultimate fibers of sisal are 3mm long and 20m wide. Each leaf of sisal fiber is approximately 10-15 cm wide, 1-2 m long and 6 mm thick. Overall, it contains 1000 fibers. Sisal is commonly used for making spa products, rugs, slippers, clothes and in the shipping industry for lashing and handling cargo.

5.2.3. Agave Fiber

Agave belongs to the agavaceae family. This is similar to the sisal plant. Each hectare of agave produces 52 tones of cellulose per year. Agave Americana, anattenuata, and blue agave are the most commonly grown agave species. The leaf is 1-2m long, 10-15cm wide, and 6mm thick. Fiber length is less than 5 mm long and 25 wide. For the preparation of tequila, the hearts of blue agave are used. Hearts are placed in an oven. After milling, we get liquid and fibers. Using that liquid, tequila can be prepared.

5.3. Seed Fiber

5.3.1. Cotton

Cotton fiber is referred to as the king of fiber. the world's most important textile fiber. Cotton is a vegetable fiber that surrounds the seeds of the cotton plant. It has been cultivated for more than 5000 years and belongs to the family GOSSYPIM. Cotton fiber is made up of countless cellulose molecules. Cotton is removed mechanically from the seed balls by cotton ginning. The ginned cotton is then pressed into bales and sent to the factories to be spun into yarns. This plant grows to approximately a 0.5-1.5m height.

Major producers of cotton are China, US, Russia and India. Commercially cotton can be classified based on their staple length.

1. Very short staple cotton (less than and equal to 21mm)
2. Short staple cotton (between 22-25mm)
3. Medium staple (between 26-28mm)
4. Ordinary long staple (between 29-34mm)
5. Extra-long staple (equal and greater than 34mm)

As a product of cotton, the raw cotton passes through several cleaning processes before it is halled [66]. As a result, the grower obtains valuable by-products that amount to approximately one-sixth of the entire income derived from the cotton plant. Cotton linters are short hair fibers used in making regenerated fiber hulls: the outside portion of the seed is rich in nitrogen and is used as a fertilizer; the seed inside the hull gives cotton seed oil, which is used in cooking and making soaps.

5.3.2. Coir

Coir fiber is a natural fiber extracted from the husk of coconut and used in products such as floor mats, door mats, brushes, and mattresses. Coir is a fibrous material found between the hard internal shell and the outer coat of a coconut. It is used in a variety of ways worldwide, being especially popular for rope and matting, and there are a number of sources for coir and coir products. There are 2 types of coir fiber; white fiber and brown fiber. White coir fiber is also known as yarn fiber, mat fiber, or retted fiber, and is a golden yellow colour. This coir fiber is made by soaking fresh, green, immature coconut husks in saline water. It is 10cm to 25cm in length, white in color, moisture 12 to 15%, maximum impurities 1%, maximum packing 120kgs. Brown fiber, or mattress fiber is extracted from the husk of coconut and used in products such as floor mats, door mats, brushes, and mattresses. Other uses of brown coir are in upholstery pudding, sacking, and horticulture. White coir, harvested from unripe coconuts, is used for making finer brushes, string, rope, and fishing nets [67]. The major producers of coir fiber are India and Sri Lanka. Brown coir is finer and more flexible than white coir. The ultimate fiber length of the coir is 1 mm and 10-20 m in width.

5.4. Core Fibers

Coir fiber is a natural product produced from coconut husk. The strongest and most durable of all commercial natural fibers is coir. The ability to make durable items with a low disintegration rate is a key benefit. Coir fiber ropes from the early 1800s have been discovered. For ages, the exceptional strength of coir fiber has been the primary cause of rope production. Coir fiber is divided into two types: brown fiber from mature coconuts and finer white fiber from immature green coconuts after soaking for up to ten months. One of the most lignin-rich natural fibers is coir. Core fibers are very short fibers averaging less than 1mm in length.

5.5. Grass and Reed Fibers

5.5.1. Sugarcane

Sugarcane is a natural plant fiber, collected from sugar cane plants. Sugarcane fiber is called "bagasse". It is widely grown in Brazil and the Pacific islands. Saccharum is the genus name of sugarcane. Bagasse is a fibrous material that remains after sugarcane is crushed to extract its juice. It is made up of dry pulp residue left after the extraction of juice from sugarcane. The soft core part of the pith is removed from the bagasse manually to get the outer hard ring. The samples are then dried in the sun. Then, these samples undergo chemical treatment. After chemical treatment, the fibers are separated [68, 69]. Sugar cane fiber contains 45-55% cellulose, 20-25% hemicelluloses, 18-24% lignin, 1-4% ash, and 1% wax. It is usually propagated by cutting. The ultimate fiber length of bagasse is 1.7 mm and the width is 20 m.

5.5.2. Bamboo

Bamboo is a regenerated cellulosic fiber produced from bamboo. This fiber is made from the starchy pulp of bamboo plants. It is the world’s most sustainable resource. The first patents for bamboo paper occurred in 1864 and 1869. The tensile strength of the fiber is very low as it has to give 350-410 twists per meter. Fiber is sensitive to acid or alkali, so the volume should be maintained carefully. Reactive dyestuffs are suitable as they react with bamboo fiber in a mild alkaline medium. It is very smooth, soft, luxurious, with good abrasion ability, temperature adaptability, and antibacterial properties. One of the fastest growing plants, as much as 120 cm/day, the diameter of the stem is 5-15 cm. After years of growth, the bamboo plant has reached maturity [70, 71]. The total fiber length is 2.7 mm and the width is 14 m. Bamboo fiber is used in bathroom textiles, and medical and hygienic clothing.

5.5.3. Johnson Grass

Johnson grass belongs to the sorghum family. This grass is found in almost every state in the U.S. Rice straw fiber has an ultimate fiber length of 1.4 mm and a width of 8 m. Corn straw has an ultimate fiber length of 1.5 mm and a width of 18 m and the fiber length and width vary with specius.

5.6. Other Fibers

5.6.1. Wood

Wood is a porous and fibrous structural tissue found in the stems and roots of trees and other woody plants. It has been used for thousands of years for both fuels and as a construction material. It is an organic material, a natural composite of cellulose fibers embedded in a matrix of lignin that resists compression. Basically, wood is characterized as either soft wood or hard wood. Soft wood is non-porous and vessel-free with fiber lengths of 4.1 mm and 2.5 m. They grow in a pyramid shape and have scales like evergreen leaves and gymnosperms too. In hard wood, water transport takes place through vessel elements. These are angiosperms with fiber lengths of 1.2 mm and 3 m [72].

Table 1. Natural fibers' chemical composition, according to publications.

Feedstock	Cellulose%	Hemicellulose%	Lignin%	Moisture%	Other	Refs.
Jute	61-71	13.6-20.4	12-13	~12.6	~0.5-2.0	[73]
Flax	65.2-72.2	10.1-17.1	2.3-7.7	-	-	[74]
Ramie	68.6-76.2	13.1-16.7	0.6-0.7	8		[75]
Kenaf	45–57	21.5	8-13	-		[75]
Pineapple	70–82	18.8	12.7	-		[75]
Sisal	67–78	10-14.2	8-11	-	2	[75]
Agave	68–80	15	5-17	8		[76]
Cotton	82.7	5.7	-	-	-	[75]
Coir	36-43	0.15-0.25	41–45	-	3-4	[75]
Sugarcane	46	27	23	-	-	[77]
Bamboo	45-50	20-25	20-30	-	-	[78]
Wood	35-50	20-30	25-30	-	-	[79]

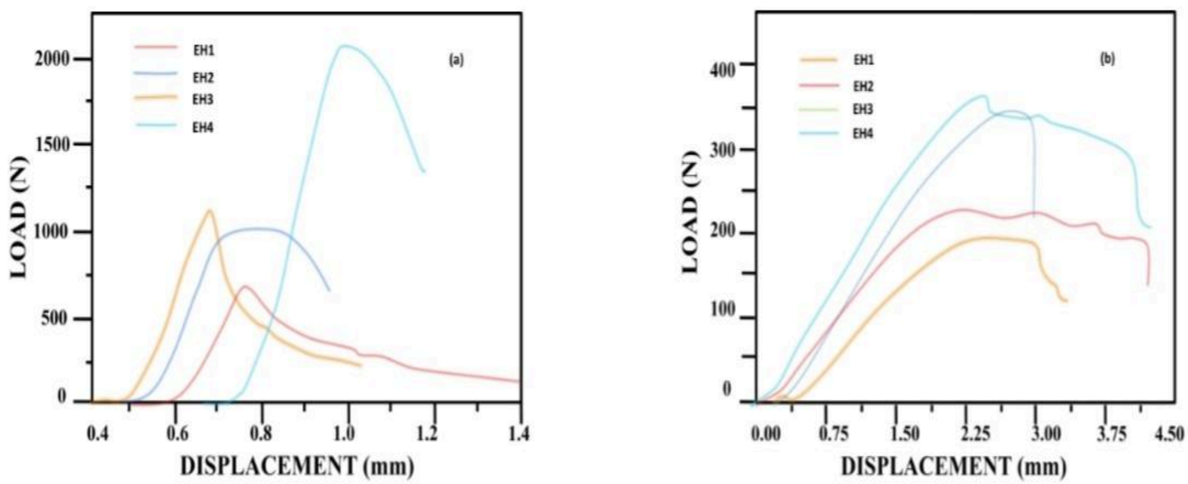

Fig. (2). Load versus displacement curves for energy reed and rice straw fibers reinforced PF composites: an Impact bending strength and b flexural properties. Adapted from figure from Cellulose, Hasan KMF, Horváth PG, Bak M, Le DHA, Mucsi ZM, Alpár T. Rice straw and energy reed fibers reinforced phenol formaldehyde resin polymeric biocomposites. 28(12): 2021; 7859-75. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

5.6.2. Animal fibers

Long ago, human beings used leaves, barks of trees, and skins of animals to cover their bodies. Then they learned the art of making cloth. These include wool and silk. Wool is obtained from the fleece of sheep and also from the goat, yak, camel, and silk produced by silk worms. Cashmere is an extremely soft, resilient, and easy-to-dye fiber, collected from Cashmere goats. Yak wool is popular in Tibet and Ladakh. Table **1** shows the chemical composition of natural fibers from different literature.

6. PROPERTY IMPROVEMENTS OF NATURAL FIBERS REINFORCED PHENOL FORMALDEHYDE COMPOSITES

Natural fibers such as jute, flax, hemp, areca, oil palm, *etc* are reinforced with phenol formaldehyde resin to produce composites. These composites have a wide range of applications. Compared to synthetic composites, natural composites are environmentally friendly and low cost.

6.1. Mechanical Properties

Hasan *et al.* [80] constructed hybrid biocomposites using PF resin in various proportions of energy reed fibers and rice straw fibers to determine the most convenient ratio of energy reeds and rice straw fibers for biocomposites' manufacturing. With increasing energy reeds loading in the composite systems, the obtained results reveal that the mechanical characteristics and moisture resistance of the systems improve. When compared to rice straw, the mechanical capabilities of the biocomposite made from 100 percent energy reeds were significantly higher than those of rice straw. It is discovered that when compared to rice straw reinforced polymeric biocomposite panels, the 100 percent energy reed fibers reinforced biocomposites with PF resin demand the largest load for breaking/bending the samples (Fig. **2**).

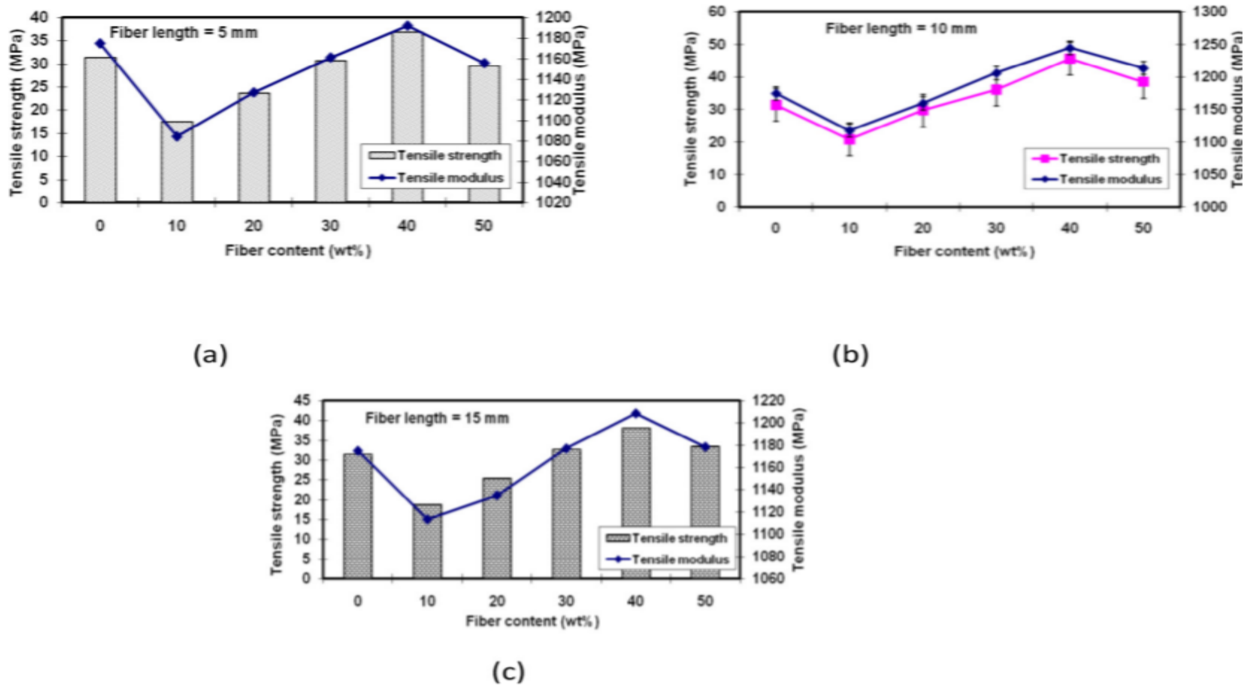

Fig. (3). Variation of tensile properties of SF/PF composites: (**a**) fiber length = 5 mm, (**b**) fiber length = 10 mm, and (**c**) fiber length = 15 mm [82]. Adapted from renewable resources, Maya MG, George SC, Sreekala MS, Jose T. Mechanical properties of sisal fiber reinforced phenol formaldehyde eco friendly composites. 2017; 8(1), 28-42. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

The mechanical characteristics of phenol formaldehyde (PF) composite reinforced with Areca Fine Fiber (AFF) and Sisal Fiber (SF) were studied as a function of fiber lengths and contents [81]. Hand lay-up techniques were used to create composites with varying fiber lengths (5, 10, and 15 mm) and content (10, 20, 30, 40, and 50 wt.%). The results demonstrated that as the fiber content and length rise, the mechanical property values of both composites increase (Fig. **3**). The highest mechanical property values were achieved when 40 wt.% and 10 mm were combined, and the mechanical property values declined as the addition was increased. As a result, for the optimal combination of mechanical characteristics with both composites, the ideal fiber length and content are 49 wt.% and 10 mm, respectively.

Maya *et al.* [82] studied the mechanical properties of sisal fiber-reinforced PF composites. Figs. (**4** and **5**) explain the stress-strain effects of sisal fiber-reinforced PF composites with varying lengths of fiber. From the graph, it is clear that the neat sample is brittle at first, then it becomes stiffer. Whatever the first composite shows, there is a deviation from the linearity and it leads to a non-linearity condition. The longer the length of sisal fiber that we took, the more reinforced it is with phenol formaldehyde, which has greater stress transferability. There is a strong bond between the fiber matrix and the fiber surface, which ensures an efficient transfer of stress from the matrix to the fiber component. Here, the critical fiber length is 50mm. Stress-transfer is not possible if the fiber has a length higher than the critical fiber length. The fiber-matrix bond breaks at a point below the critical fiber length. In a similar way, by varying fiber content, sisal-reinforced PF composites show that the strength of the composite increases with fiber loading. Up to 54 wt.% fiber loading increases along with tensile strength. Then, it decreases due to the agglomeration of fibers. Normally, the tensile strength of composites decreases with increases in fiber loading, with exceptions. From Fig. (**4**), maximum tensile strength is observed for 54wt% fiber loading. Compared with other natural fibers, sisal fiber has better reinforcement with a phenol formaldeyde matrix.

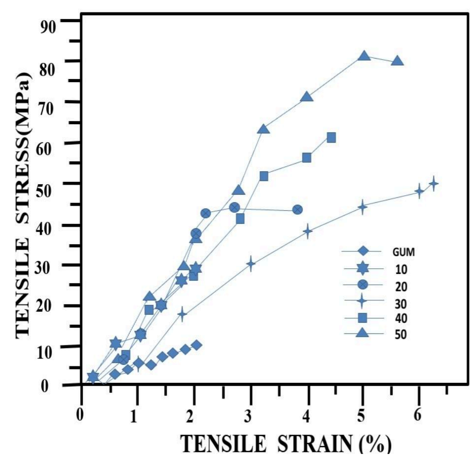

Fig. (4). Stress-strain of sisal fiber reinforced phenol formaldehyde composites, by varying fiber length. Adapted from renewable resources, Maya MG, George SC, Sreekala MS, Jose T. Mechanical properties of sisal fiber reinforced phenol formaldehyde eco friendly composites. 2017; 8(1), 28-42. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

Similarly, Azim *et al.*'s [83] studies reveal that pineapple leaf fiber and kenaf fiber phenolic composites enhance the mechanical properties of pineapple leaf fiber and kenaf fiber, respectively. It was found that fiber content and also fiber length affect the mechanical properties of composites. Jawaid *et al.* [84] studied the effects of alkali treatments on the physical and mechanical strength of pineapple leaf fibers. According to their study, the hydrophilic nature of natural fibers makes them difficult to integrate with the matrix. But when treated with an alkali like NaOH, it enhances the compatibility with the matrix. This increases the mechanical properties of the composites.

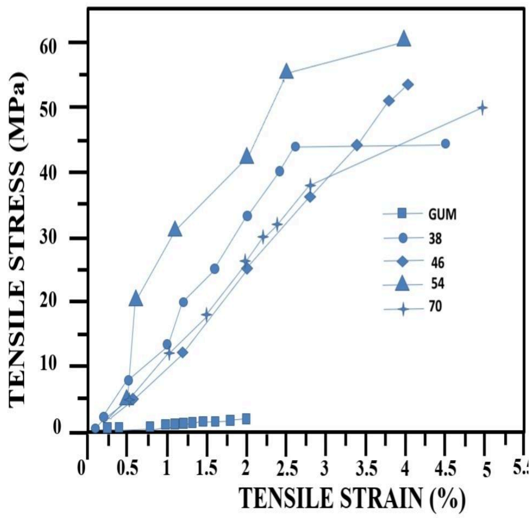

Fig. (5). Stress-strain of sisal fiber reinforced phenol formaldehyde composite by fiber loading. Adapted from renewable resources, Maya MG, George SC, Sreekala MS, Jose T. Mechanical properties of sisal fiber reinforced phenol formaldehyde eco friendly composites. 2017; 8(1), 28-42. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

Agnivesh Kumar Singh and colleagues [85] investigated abaca fiber-reinforced polymer composites. According to the study, the high specific strength of abaca fiber reinforced polymer composites is used for car exterior production. Feng *et al.* [86] studied the tensile and fatigue responses of cellulosic fiber reinforced composites. According to their study, natural fiber reinforced composites show the qualities of natural as well as synthetic ones. Ashvinder *et al.* [87] studied the mechanical properties of cellulosic grewia optiva fiber polymer composites. Because of the unique mechanical properties of fiber, it is widely used in our daily life.

6.2. Thermal Properties

Thermogravimetric analysis is a percentage of weight loss as a function of temperature, which is used to determine a material's thermal stability and its fraction of volatile components by monitoring the weight change that occurs as a sample is heated at a constant rate. Cellulose, hemicellulose, and lignin are the major components of banana fiber and have strong and fast moisture absorption capacity. Compared with other natural fibers, banana fiber-reinforced ones have better thermal stability. Natural fibers are hydrophilic in nature.An increase in water absorption capacity with fiber loading leads to heat resistance, good spinning and tensile strength. Sabu *et al.* [88] developed banana fiber-phenol formaldehyde composite. In addition, they developed a composite with more enriched thermal performance. Thermogravimetric analysis is a way to show improved thermal stability. When we check the Tg of banana fiber reinforced composite, it is evident that alkali-treated banana fiber has better thermal stability than alkali-untreated banana fiber. The peak before 100 °C implies the elimination of moisture content. Cellulose, lignin, and hemicelluloses are the main components of banana fiber. The peak temperatures of 305 and 410 °C indicate cellulose fiber destruction. At 544 °C, it shows degradation of the PF resin matrix.. For the good performance of composites, silane and amino silane is normally used. They work as coupling agents and enhance interfacial adhesion between fibers and matrix. Silane and aminosilane coupling enhance the strength of banana fiber-reinforced phenol formaldehyde composites and also affect their surface modification. It is clear that the percentage of weight loss has decreased but the percentage of residue has increased. Thereafter, the thermal stability of composites increased. Resin penetration levels are at their best, which leads to a strong fiber matrix and enhanced thermal stability of banana fiber-reinforced phenol formaldehyde composite. The thermal stability of composites made of banana fiber-reinforced phenol formaldehyde is found to be significantly higher than that of fiber, but it is found to be lower than that of unmodified PF resin. Untreated fiber/PF composites were found to have worse thermal stability compared to NaOH-treated fiber composites with increased fiber/matrix adhesion. These composites were found to have greater stability overall. Silane treatments were found to be efficient in improving the composite's stability; nevertheless, it was discovered that vinyl silane-treated fiber composites were more successful than aminosilane treated fiber. It has been discovered that fiber-reinforced composites that have been cyanoethylated or treated with latex have higher thermal stability. Despite the fact that the hydrophobicity of the fiber reduces the fiber matrix adherence, these composites have been found to have higher thermal stability. According to the findings of this investigation, composites with very good fiber/matrix adhesion demonstrated better heat stability.

Burhan *et al.* [89] discussed the thermal characteristics of natural fiber reinforced PF composites. The research study explains the effect of fiber content and alkali on thermal performance. Barath *et al.* [90] studied coconut leaf sheath reinforced phenol formaldehyde composite thermal properties. Naresh *et al.* [91] studied the importance of natural fiber by replacing synthetic fibers. In kenaf fiber reinforced polymer composites, the thermal properties of kenaf fiber composites decreased as fiber content increased. Asim *et al.* work deals with date palm fiber effects on the PF composites. The tan delta of these has a lower value with loading when Tg increased. It possesses superior thermal properties [92]. Azim *et al.* [93] studied the fiber loading effect and how it influenced the pineapple leaf fiber and kenaf fiber reinforced phenolic composites along with its thermal properties.

To create hybrid MDF using basalt fiber and phenol-formaldehyde resin hybridization, coconut coir was employed in conjunction with phenol-formaldehyde resin hybridization. The panel was constructed using a variety of coir and basalt fiber combinations while maintaining a constant resin proportion. According to the analysis, there were three distinct stages of degradation. The temperature ranged from 35 ℃ to 203 ℃ in the first stage, and the weight loss was caused by water absorption on the panel's surface and in the panel's interior. The evaporation of formaldehyde from the resin, water, and organic molecules caused the second stage of weight loss, which occurred at temperatures ranging from 203 ℃ to 310 ℃. The basalt fiber, which is present in the third stage of thermal disintegration, contributes to the panel's stability and resilience [94].

6.3. Electrical Properties

The dielectric constant of natural fiber-reinforced phenol formaldehyde composites was studied with a dielectric constant. It is the ratio of the electric permeability of the material to the electric permeability of the vacuum. The quantity of cellulose content determines the changes in the dielectric properties of the composites. In short, composites having cellulose content will have higher dielectric constants. But compared with phenol formaldehyde resin, natural fibers have a lower dielectric constant.

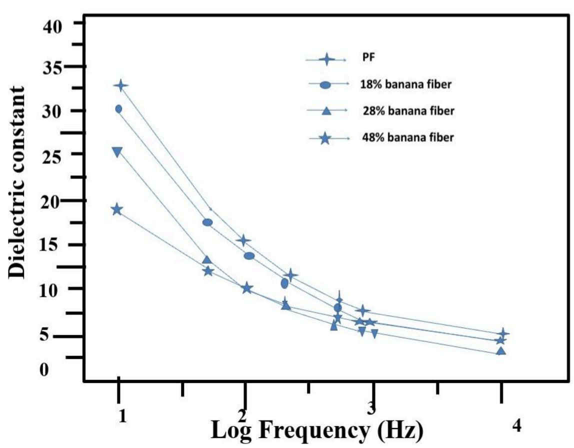

Fig. (6). Banana fiber reinforced PF composites. Adapted from journal of applied polymer science,Joseph S, Thomas S. Electrical properties of banana fiber reinforced phenol formaldehyde composites. 2008; 109(1), 256-63. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

Fig. (**6**) shows the dielectric constant and frequency as a function of the dielectric constant. From this graph, it is clear that the greater the fiber loading, the lesser the dielectric constant will be. Also, a neat PF sample has the highest value of dielectric constant. A dielectric constant measures the amount of material that can store the electrical energy when placed in an electrical field. In terms of interfacial polarization, it can be explained that PF resins contain a polar group that is why E tends to be high. However, since the composite is heterogeneous there is interfacial polarization. Interfacial polarization supports low-frequency E. Chemical treatments such as acetylation and cyanoethylation affect the electrical properties of natural fiber reinforced PF composites. Studies reveal that with increased chemical treatments and fiber loading, dielectric value decreases (Fig. **6**) [95].

Gupta *et al.* [96] studied the dielectric properties of bio-fiber reinforced PF resin. Studies reveal that the dielectric constant measures electrical energy. Thereby, electrical property improvements of bio-fiber reinforced PF resin can be easily analyzed. Shanbhag *et al.* [97] studied the coir composites-based electronics. According to the study, they introduced a low-cost micro strip antenna and transmission line. It is expensive and can be used widely to charge vehicles, as the high power handling capacity of micro capacity can reduce environmental concerns.

6.4. Visco-Elastic Properties

Visco-elastic properties of banana fiber reinforced PF composites have been evaluated as a function of fiber content, frequency and temperature. The storage modulus and temperature graphs were analyzed using dynamic mechanical analysis, which allowed for the extraction of significant information on the degree of cross-linking, stiffness, and fiber/matrix interfacial bonding of the materials. The storage modulus of untreated and treated hybrid composites with varied PALF/KF fiber ratios was investigated at various temperatures (Fig. **7**) [98]. The storage modulus dropped as the temperature increased. At low temperatures, the storage modulus of untreated and treated hybrid composites showed a significant difference in their characteristics; treated hybrid composites had a higher storage modulus than their untreated counterparts, but T-3P7K had a lower storage modulus than untreated 3P7K hybrid composites. As the temperature rises, material components become more mobile and lose their close packing structure; nonetheless, the rubbery region's modulus does not change significantly. 7P3K hybrid composites had the lowest storage modulus value, while T-7P3K hybrid composites had the highest ones. 7P3K hybrid composites demonstrated reduced modulus and rubbery phase compared to all untreated and treated hybrid composites, resulting in material flexibility and low stiffness (Fig. **8**).

The dynamic mechanical properties of banana fiber reinforced PF composites manufactured by RTM and CM processes were examined as a function of fiber content, fiber treatment, temperature, and frequency. These variables include storage modulus, loss modulus, and tan delta [99]. RTM (resin transfer molding) and CM (chemical molding) is the fabrication techniques used for analysis. Fig. (**9**) shows the effect of the storage modulus of untreated banana fiber reinforced/PF composites on varying fiber content. From Fig. (**10**), it is clear that the neat PF storage modulus is low. Storage modulus increases with an increase in fiber loading. The maximum storage modulus is up to 40% of fiber content, and then it decreases. The greater the storage modulus, the greater the transfer of stress from matrix to fiber. At 50 wt.%, both fabrication techniques show a decrease in storage modulus. This is due to insufficient wetting, void formation, and agglomeration formation in the composite.

Careful examination shows that at 40 wt.%, RTM composites have a higher storage modulus than CM due to the reduction of intermolecular forces at higher temperatures, which causes E to decrease with increasing temperature (Fig. **10**). This resulted in a reduction in modulus between 130 and 160 °C CM composites.

Tg values are lowest for composites (Fig. **10**). Whatever else, as a result of fiber loading, the extent of crosslinking in

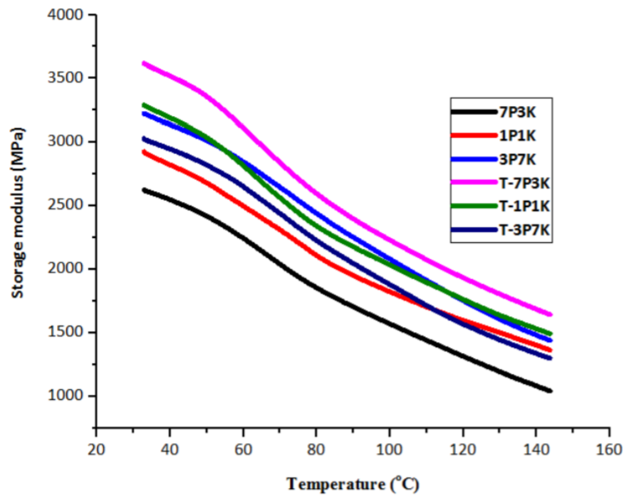

Fig. (7). Storage modulus of untreated and treated PALF/KF/phenolic hybrid composites. Adapted from Fibers and Polymers, Asim M, Jawaid M, Abdan K, Ishak MR, Hammami H. Effect of pineapple leaf fibre and kenaf fibre treatment on mechanical performance of phenolic hybrid composites. 2017; 18(5), 940-7. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

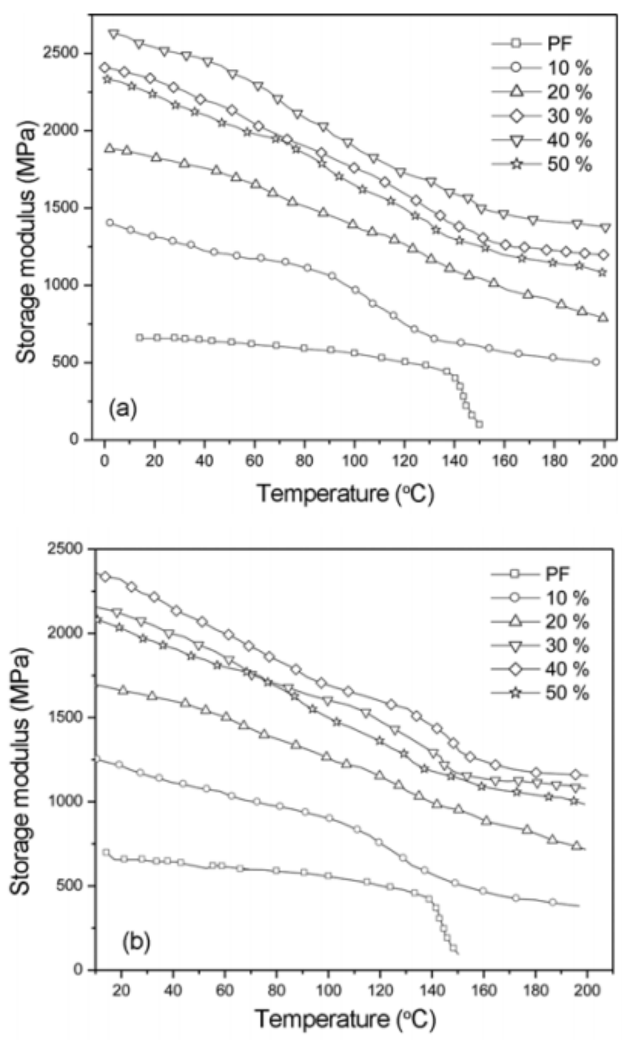

Fig. (8). Untreated banana fiber/PF composites with varying fiber content. **a**) RTM at 0.1 Hz **b**) CM at 0.1 HZ (15(1), 91-100) [99]. Adapted from Fibers Polym, Indira KN. Viscoelastic behavior of untreated and chemically treated banana fiber reinforced phenol formaldehyde composites. 2014; 15(1), 91-100. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

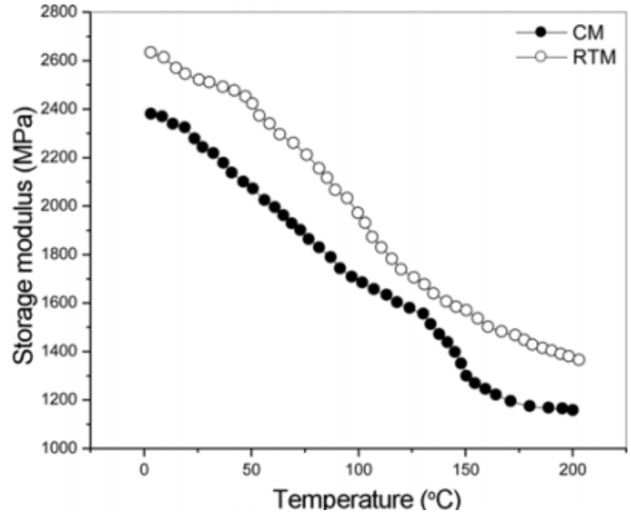

Fig. (9). Untreated banana fiber/PF composites (40wt %) by CM and RTM at 0.1 Hz [99]. Adapted from Fibers Polym, Indira KN. Viscoelastic behavior of untreated and chemically treated banana fiber reinforced phenol formaldehyde composites. 2014; 15(1), 91-100. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

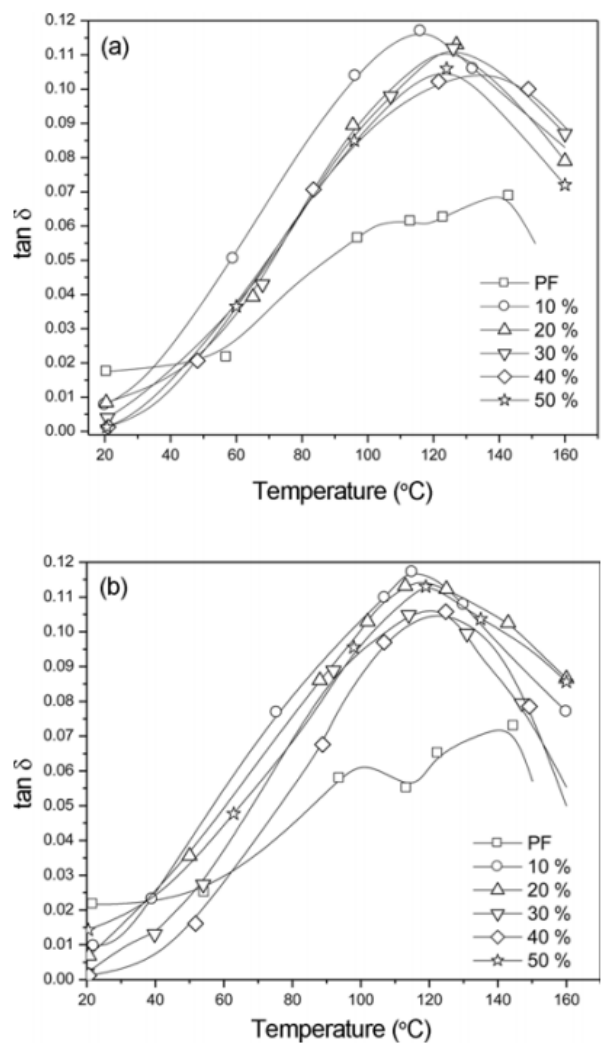

Fig. (10). Tan delta *vs* temperature curves of untreated banana fiber/PF composites fabricated by a) RTM at 0.1 Hz and b) CM at 0.1 Hz [99]. Adapted from Fibers Polym, Indira KN. Viscoelastic behavior of untreated and chemically treated banana fiber reinforced phenol formaldehyde composites. 2014; 15(1), 91-100. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

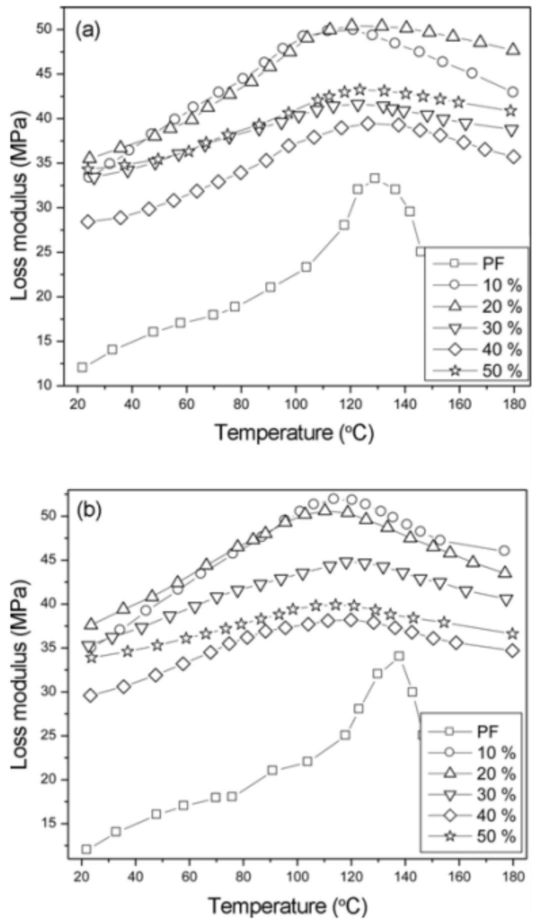

Fig. (11). Varying banana fiber loading on loss modulus of the composites with temperature **a**) RTM **b**) CM [99]. Adapted from Fibers Polym, Indira KN. Viscoelastic behavior of untreated and chemically treated banana fiber reinforced phenol formaldehyde composites. 2014; 15(1), 91-100. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

the cured PF decreases. Peak temperature rises as the day progresses and about 40 wt.% has the highest storage modulus. Compared to the damping values of banana/PF and neat PF, the banana fiber-reinforced PF composites have greater damping values. The highest fiber content has the highest storage modulus.

PF has a lower loss modulus compared to composites (Fig. **11**). This is due to the greater fiber content loss when the modulus increases. From the table, it is clear that the neat PF has greater Tg. With the addition of fiber, the extent of cross-linking in the cured PF decreases. Due to the resistance against DMA deformation of the PF matrix with increasing fiber weight, the composite loss modulus also increases.

Above, Fig. (**12**) gives a very clear idea about storage modulus and loss modulus. The visco-elastic properties depend upon frequency and temperature. Figs. (**12a** and **b**) shows the variation of storage modulus with temperature. If material is subjected to constant stress, in order to reduce stress, molecular rearrangements occur. Figs. (**12c** and **d**) shows the loss modulus shift towards higher temperatures. They have concluded that there is a consistent pattern that can be seen in the composites' viscoelastic properties, regardless of the fabrication method that was used. The findings of the DMA showed that the addition of fiber to the PF matrix leads to an increase in the storage modulus, and the highest value was found for composites with a fiber content of 40wt.%. When compared to the pure PF matrix, the loss modulus curves became broader. The decreased tan value can be attributable to the improved fiber/matrix interaction, which occurred as the fiber content grew and increased over time. The alkali-treated composites achieved the highest storage modulus and glass transition temperature out of all the chemically treated composites.

Sreekala *et al.* [100] evaluated the effect that fiber content and hybrid fiber ratio had on the dynamic mechanical characteristics of oil palm fiber reinforced phenol formaldehyde (PF) composites and oil palm/glass hybrid fiber reinforced PF composites. In the case of oil, palm fiber reinforced PF composite variations of tan delta and storage modulus is given in Fig. (**13**).

It is clear that damping increases with fiber loading. Composites with 50 mm have two damping peaks due to agglomeration within the fiber and matrix, and fiber-matrix has a larger interfacial area (Fig. **14**). Frequency decreases as damping increases. Composites containing 30% by weight have a higher modulus. At higher fiber loading, agglomeration and voids increase, thereby decreasing the modulus of the composite. About 50 wt.% composites have higher

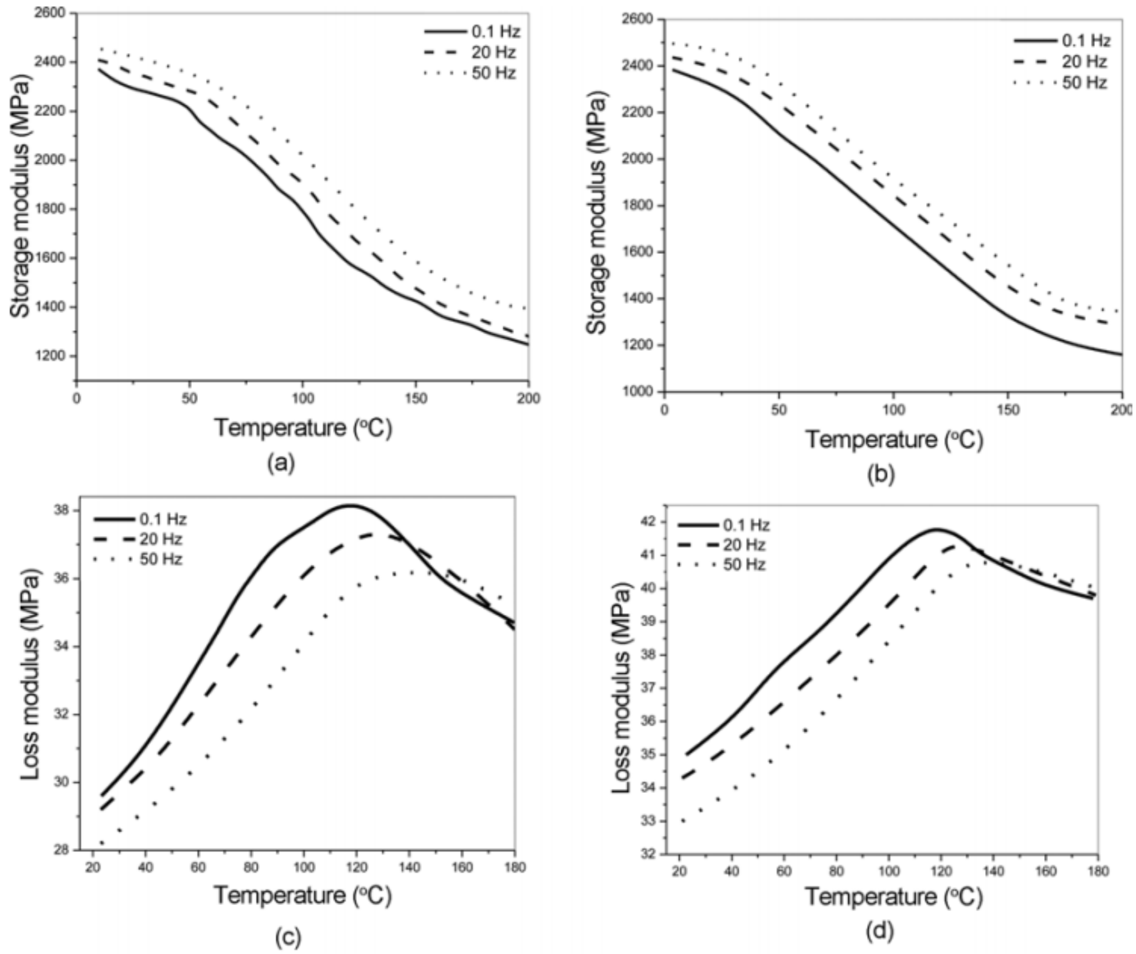

Fig. (12). a) Frequency effect on E' of RTM fabricated by banana fiber/PF composites (40 wt.%). **b**) Effect of frequency on E' of CM fabricated banana fiber/PF composites (40 wt.%). **c**) Effect of frequency on E" of banana fiber/PF composites (40 wt.%) fabricated by RTM. **d**) effect of frequency on E" of banana fiber/PF composites (40 wt.%) fabricated by CM [99]. Adapted from Fibers Polym, Indira KN. Viscoelastic behavior of untreated and chemically treated banana fiber reinforced phenol formaldehyde composites. 2014; 15(1), 91-100. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

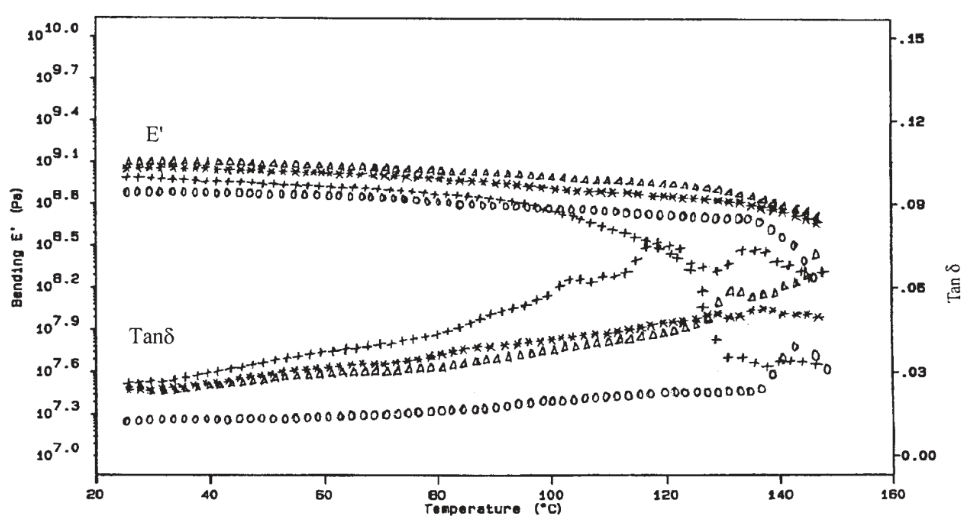

Fig. (13). Variation in tan delta and storage modulus of oil palm fiber/PF composites fiber loading having 40mm fiber length at 10Hz frequency [100]. Adapted from Polym Compos, Sreekala MS. Dynamic mechanical properties of oil palm fiber/PF and Oil palm fiber/glass hybrid phenol formaldehyde composites. 2005; 26(3), 388-400. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

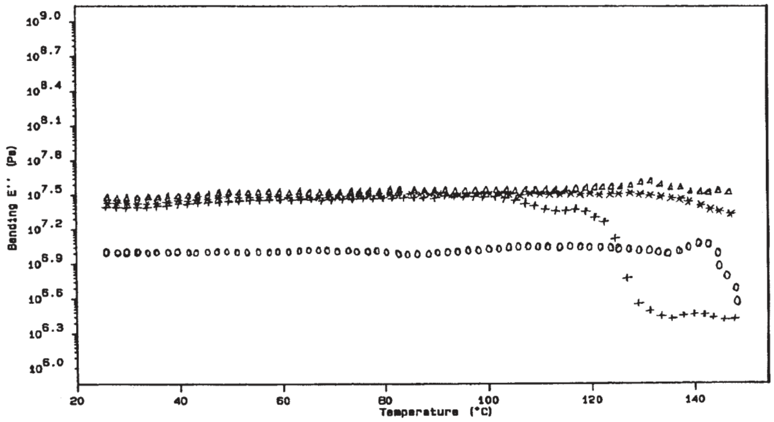

Fig. (14). Loss modulus of oil palm /PF composites at 10 Hz frequency [100]. Adapted from Polym Compos, Sreekala MS. Dynamic mechanical properties of oil palm fiber/PF and Oil palm fiber/glass hybrid phenol formaldehyde composites. 2005; 26(3), 388-400. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

modulus. With decreasing frequency, loss modulus increases and by increasing fiber loading loss, the modulus decreases. Compared to the damping peak, the loss modulus peak shifted to low-temperature region.

From the results, they concluded that the addition of fibers had a positive impact on the dynamic mechanical properties of the PF composites. The inclusion of fiber improves tan delta and storage modulus values significantly. The value goes up as the amount of fiber in the composite increases. The loss modulus demonstrates a reversal of trend as the fiber loading level is increased. The glass transition temperature is shifted toward lower values when oil palm fiber is incorporated into the material. The temperature at which the hybrid composites undergo glass transition is lower than the temperature at which it occurs in the unhybridized composites. Hybrid composites have been found to have the highest values of both mechanical and thermal damping. The storage modulus of hybrid composites is lower than that of a composite made of oil palm fiber and PF that has not been hybridised. The addition of fibrous reinforcement results in an increase in activation energy.

6.5. Tribological Properties

Tribology is the study of an interacting surface in relative motion. It includes the study of friction, wear and lubrication. The figure shows the graphical representation of the friction coefficient *vs*. temperature. It is possible to investigate the fade and recovery responses of composites from this. Here the responses of pineapple fiber composites can be seen (FFR-first fade run, FRR-first run recovery, SFR-second fade run, SRR-second run recovery [101]. Figs. (**15-17**) give an idea of the changes in coefficient of friction with temperature and changes during FFR, SFR, and SRR.

In the case of pineapple fiber during FFR, the initial temperature of 93 °C decreases the coefficient of friction. Greater pineapple fiber loading tends to decrease. PF-4 composites have a fiber content of 20 wt.% and show a decreasing value above 233 °C, which begins to fall during FFR. However, with the introduction of SFR, prices began to rise. In the case of PF-1, increases. The PF-2/PF-3/PF-4 value is between 0.3 and 0.4. During FFR, the value is above 0.5 for PF-1/PF-2/PF-3 composites. PF-4 composites have a 0.4 value. Which shows a decreasing coefficient of friction. SRR showed increases in value for PF-1 composites. Above 250 °C PF-2/PF-3/PF-4 composites showed a decrease in values. In the case of synthetic fibers like Kevlar, the beginning of the SFR value increases to 0.6. Above 300 °C, the value decreases to 0.57. KF-1 composites showed good values during FFR. Above 150 ℃, the KF-2 value starts to reach 0.52.

With increasing fiber content, μ_p(μ-performance), μ_f(μ-fade), μ_R(μ-recovery) values were found to decrease. The above graph shows the response ofμ_p, μ_f, μ_R values by the addition of pineapple fiber and also Kevlar fiber (Fig. **17**) μ_p is 0.548 when pineapple fiber content is 5 wt % and it decreases 2% with the further addition of 10-15 wt.% fiber content. Moreover, it decreases 5% by the addition of 20% fiber. In the same way, μf value is decreased by fiber loading and μR showed (0.585 + 0.005) responses. P and μ_f decrease values by fiber loading. This is due to organic ingredient degradation with temperature due to which friction performance decreases. The adhesive component created by PF matrix led to the formation of contact film. As a result, performance of friction decreases. But in the Kevlar fibers, there is an increase in the performance of friction. Increasing fiber loading creates increased wear performance. Whatever greater fiber loading led to agglomeration and their detachment becomes easier. All these factors are responsible for an increased wear rate.

6.6. Biodegradability

Biodegradation is the process in which microorganisms can consume an entire material and emit water and carbon dioxide as by-products. For the degradation of polymer materials, active microorganisms are required. Temperature effects vary according to organisms. Natural fiber reinforced PF composites are more eco-friendly than synthetic fiber reinforced polymer composites [102, 103]. Compared to nonrenewable petroleum-based composites, natural fiber reinforced PF composites show effective degradability. Due to the low cost and comfortability of plastics, they have so many applications in our day-to-day lives. However, it has serious consequences on our environment. Only 1% of plastics in the market are bio-based. Increased use causes greenhouse gas emissions and is also harmful to the living organisms [104]. In order to reduce this toxicity, the invention of

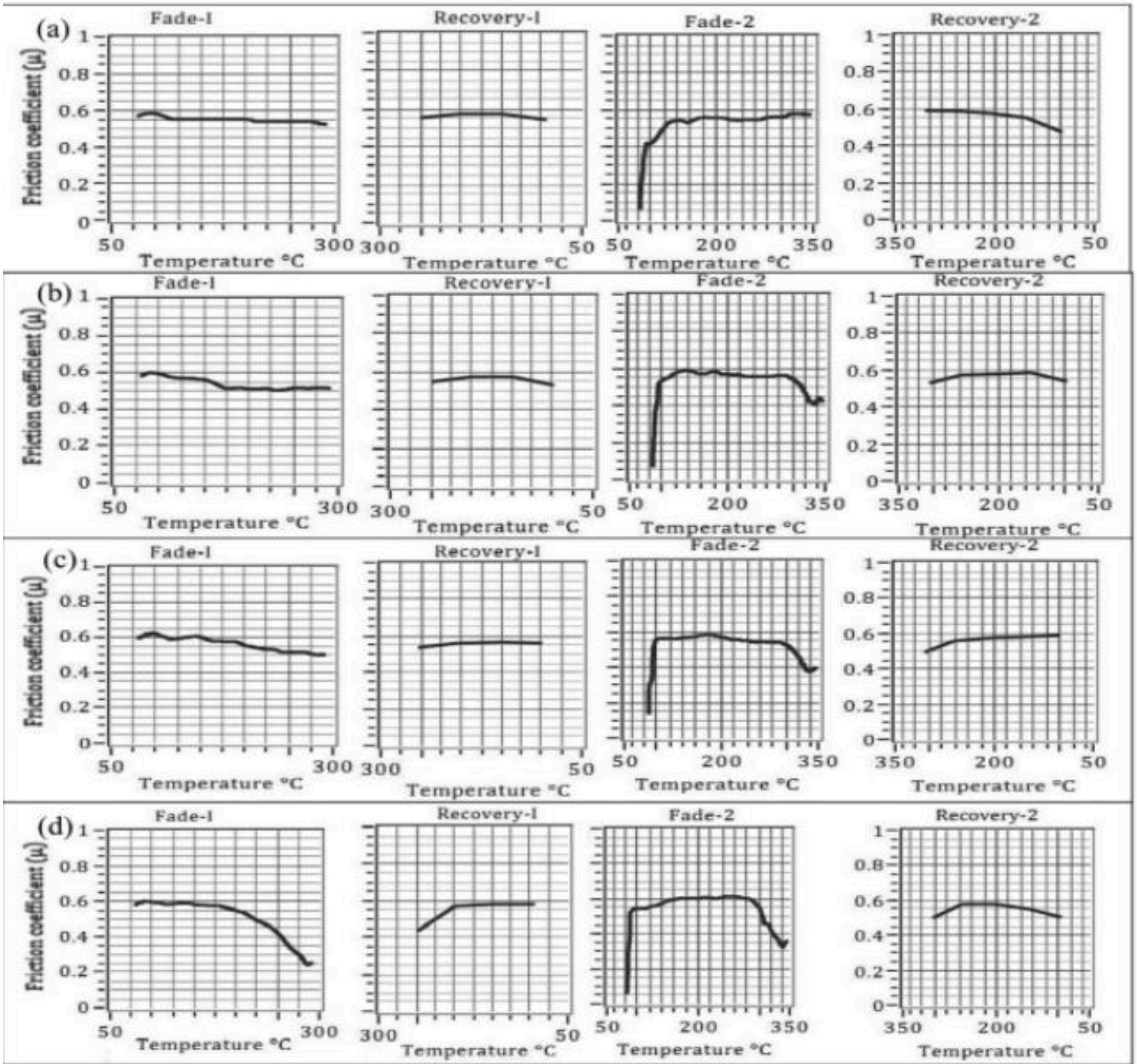

Fig. (15). Friction coefficient *vs* temperature of PF composite [101]. Adapted from J Mater Res Technol, Singh Tej, Pruncu CI, Gangil B, Singh GV. Comparative performance assessment of pineapple fibers based friction composites. 2020; 9(2), 1491-9. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

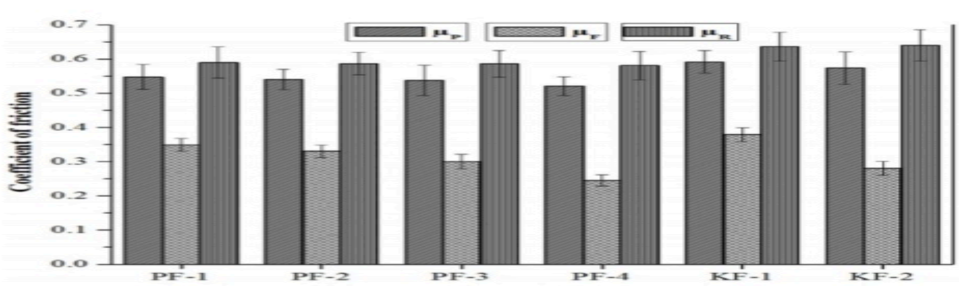

Fig. (16). Coefficient of friction *vs* composition of PF composite [101]. Adapted from J Mater Res Technol, Singh Tej, Pruncu CI, Gangil B, Singh GV. Comparative performance assessment of pineapple fibers based friction composites. 2020; 9(2), 1491-9. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

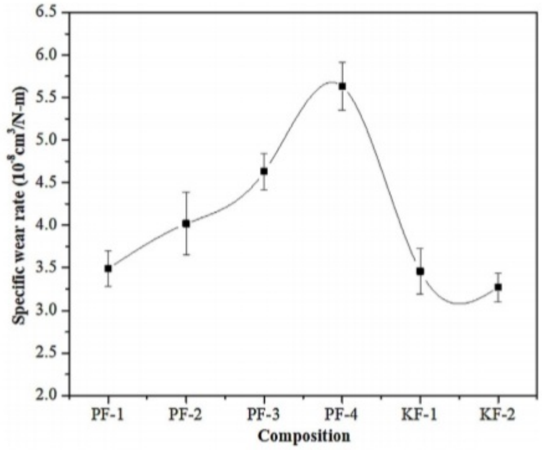

Fig. (17). Wear rate *vs* Composition of PF composite [101]. Adapted from J Mater Res Technol, Singh Tej, Pruncu CI, Gangil B, Singh GV. Comparative performance assessment of pineapple fibers based friction composites. 2020; 9(2), 1491-9. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

natural fiber reinforced phenol formaldehyde PF composites is helpful. It gives both plastic and natural qualities.

6.7. Diffusion Studies

Diffusion mechanism takes place in the interfacial region, not in the cross-sectional region. The cross section region favours capillary mechanism. Fiber-matrix extension is an unavoidable factor to understand the sorption nature of composite. Phenolic composites have very low gap width. This is due to the hydrophilic nature of resin and fiber. Water sorption leads to filling up of these small gaps [105]. By hand lay-up technique, oil palm empty fruit bunch (OPEFB) and sugarcane bagasse (SCB) fibers were employed as fillers in varied ratios to build hybrid composites with a total fiber loading of 50 wt.% [106]. In comparison to pure fiber composites, hybridised OPEFB/SCB fiber composites have improved performance and characteristics, according to this study. When compared to pure composites, 7OPEFB: 3SCB hybrid composites had the highest tensile strength and modulus, 5.56 MPa and 661.MPa, respectively, with reduced porous and voids area. After 24 hours of testing, 3OPEFB: 7SCB hybrid composites show less water absorption and thickness swelling (Fig. **18**).

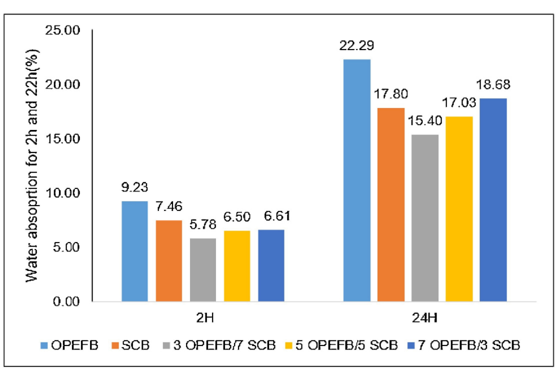

Fig. (18). Water absorption against immersion time of composites [106]. Adapted from J Mater Res Technol, Ramlee NA, Jawaid M, Zainudin ES, Yamani SAK. Tensile, physical and morphological properties of oil palm empty fruit bunch/sugarcane bagasse fibre reinforced phenolic hybrid composites. 2019; 8(4), 3466-74. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

To learn more about the impact of water absorption on the mechanical properties of hybrid phenol formaldehyde (PF) composite made from Areca Fine Fibers (AFFs) and Calotropis Gigantea Fiber (CGF), hand layup was used to make hybrid CGF/AFF/PF composites with varying weight percentages of fiber reinforcement (25, 35, and 45%) [107]. When compared to other hybrid composites, the 35 wt.% hybrid composite had improved mechanical properties (tensile strength of ca. 59 MPa, flexural strength of ca. 73 MPa, and impact strength of 1.43 kJ/m^2) in both wet and dry circumstances. The incorporation of the fibers improved the mechanical characteristics of neat PF in general. The water absorption increased as the fiber content rose, but after 120 hours of immersion, all of the composites reached equilibrium. The degree of water penetration increased linearly for all hybrid composites exposed to distilled water and reached saturation after 10 days of immersion. The lowest and maximum percentages of water absorption were found at 25 wt.% hybrid composite and 45 wt.% hybrid composite, respectively, according to the water absorption data. Because the amount of matrix is higher at the lower fiber weight percentage, 25 wt. percent of the composites demonstrated reduced water absorption when compared to 35 wt.% and 45 wt.% of the composites. The hydrophobic nature of the matrix material prevents water molecules from interacting with the fiber content (Fig. **19**).

6.8. Morphological Properties

SEM analysis is helpful in identifying the morphology of fibers. It is a technique in which an incident electron beam scans across the surface and creates an image of the surface. Electrons interact with atoms in the fiber, producing various signals that contain information about the surface topography and composition of the sample. It is used to generate high-resolution images of the shapes of objects and to show spatial variations in chemical compositions. Surface morphology plays a vital role in the aerospace, medical, textile, and building sectors. Tensile fracture of treated and untreated composites: From the picture (Figs. **20**-**23**), it can be observed that fiber-matrix adhesion is better for alkali-treated composites than for alkali-untreated composites [108]. Stronger interaction between fibers and matrix increases the composite interfacial compatibility, leading to efficient load transfer from the matrix to neighboring fibers and it produces a positive effect on composite mechanical properties. Evaluation of interfacial performance between PF resin and oil palm fiber composites clearly shows that alkali treated composites have better fiber-matrix adhesion than untreated ones.

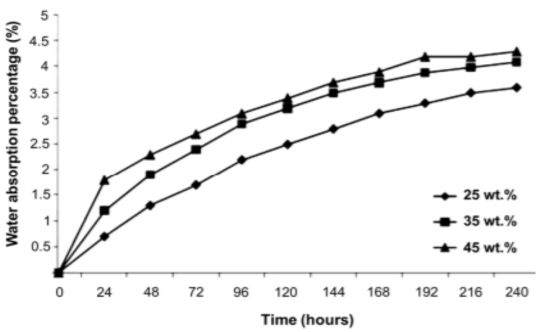

Fig. (19). The water absorption percentage *versus* immersion time curves of CGF/AFF/PF hybrid composites [107]. Adapted from Sci Rep,Sanjeevi S, Shanmugam V, Kumar S, et al. Effects of water absorption on the mechanical properties of hybrid natural fibre/phenol formaldehyde composites.2021; 11(1), 13385. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

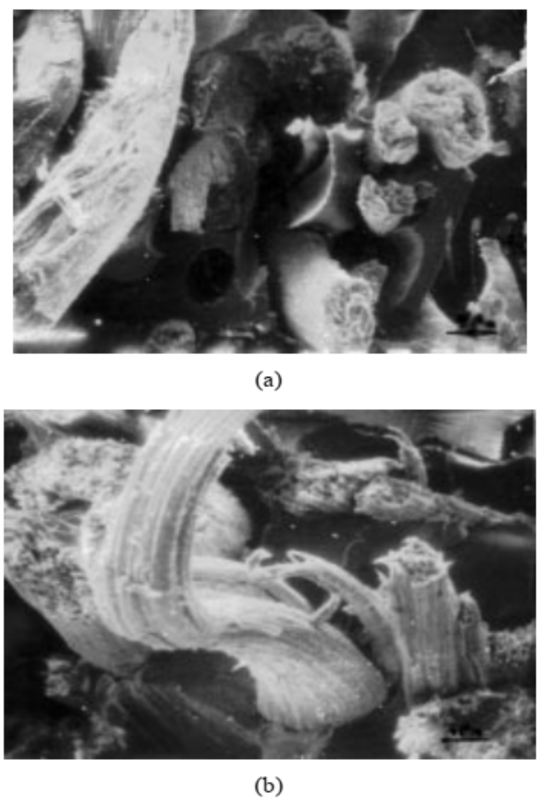

Fig. (20). SEM of oil palm fiber/PF. **a**) untreated **b**) mercerized [108]. Adapted from Appl Compos Mater, Sreekala MS, Kumaran MG. Seena Joseph and Maya Jacob. Oil Palm Fiber Reinforced Phenol Formaldehyde Composites: Influence of Fiber Surface Modifications on the Mechanical Performance. 2000; 7(5/6), 295-329. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

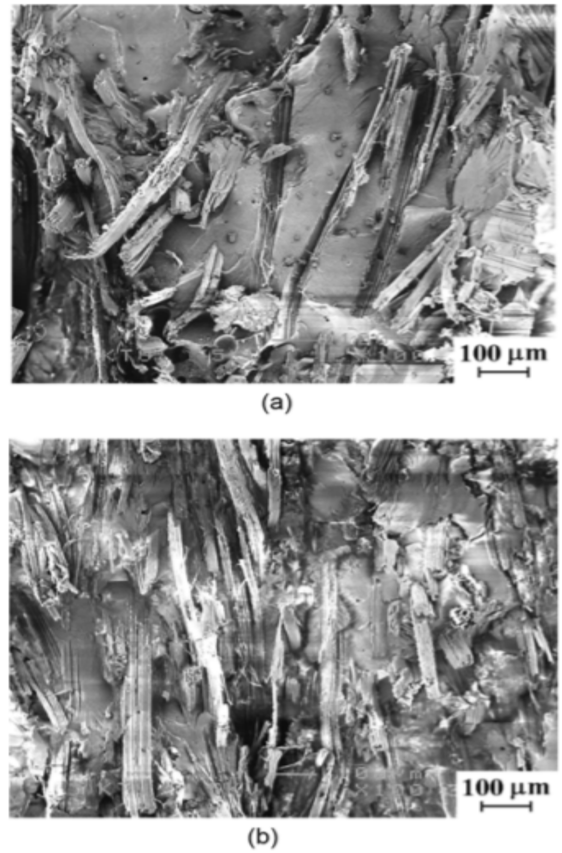

Fig. (21). SEM micrographs of tensile fractured surfaces of jute/PFcomposites; Adapted from Fibers Polym, Ozturk B, Hybrid effect in the mechanical properties of jute/Rockwood hybrid fibers reinforced PF composites. 2010; 11(3): 464-73. (**a**) fiber loading 16 vol.% and (**b**) fiber loading 42 vol.% [109]. (*A higher resolution / colour version of this figure is available in the electronic copy of the article*).

Öztürk *et al.* [109] jute/PF, rockwool/PF and jute/rockwool hybrid PF composites were developed and the morphology of fracture surface was analyzed. Fig. (**22**) (**a**) shows SEM images of the tensile fractured surfaces of jute/PF composites having 16 and 42 vol.% fiber loadings, respectively (**b**). Since the hydrophilic nature of cellulose and PF resin causes a significant contact between fibers and phenolic resin in natural fiber reinforced composites, fiber breaking is the most common failure mechanism. As a result, the debonding of fiber from the matrix is more difficult and fiber pullout is lower in natural fiber/PF systems. During the fracture process, jute fibers are broken without being completely pulled out, and a large amount of PF matrix coats the fibers as a result of the increased fiber loading. Furthermore, tearing failures of the fibers are identified; however, no complete interfacial breakdown is seen, showing excellent adhesion in the jute/PF combination.

There were equal amounts of jute and banana fibers with four different fiber orientations incorporated in the phenol-formaldehyde resin used to manufacture hybrid laminated products using the hot press process. Fiber and matrix failure were evaluated using SEM to examine the debonding of fibers, voids, and fiber pullouts that occurred during the

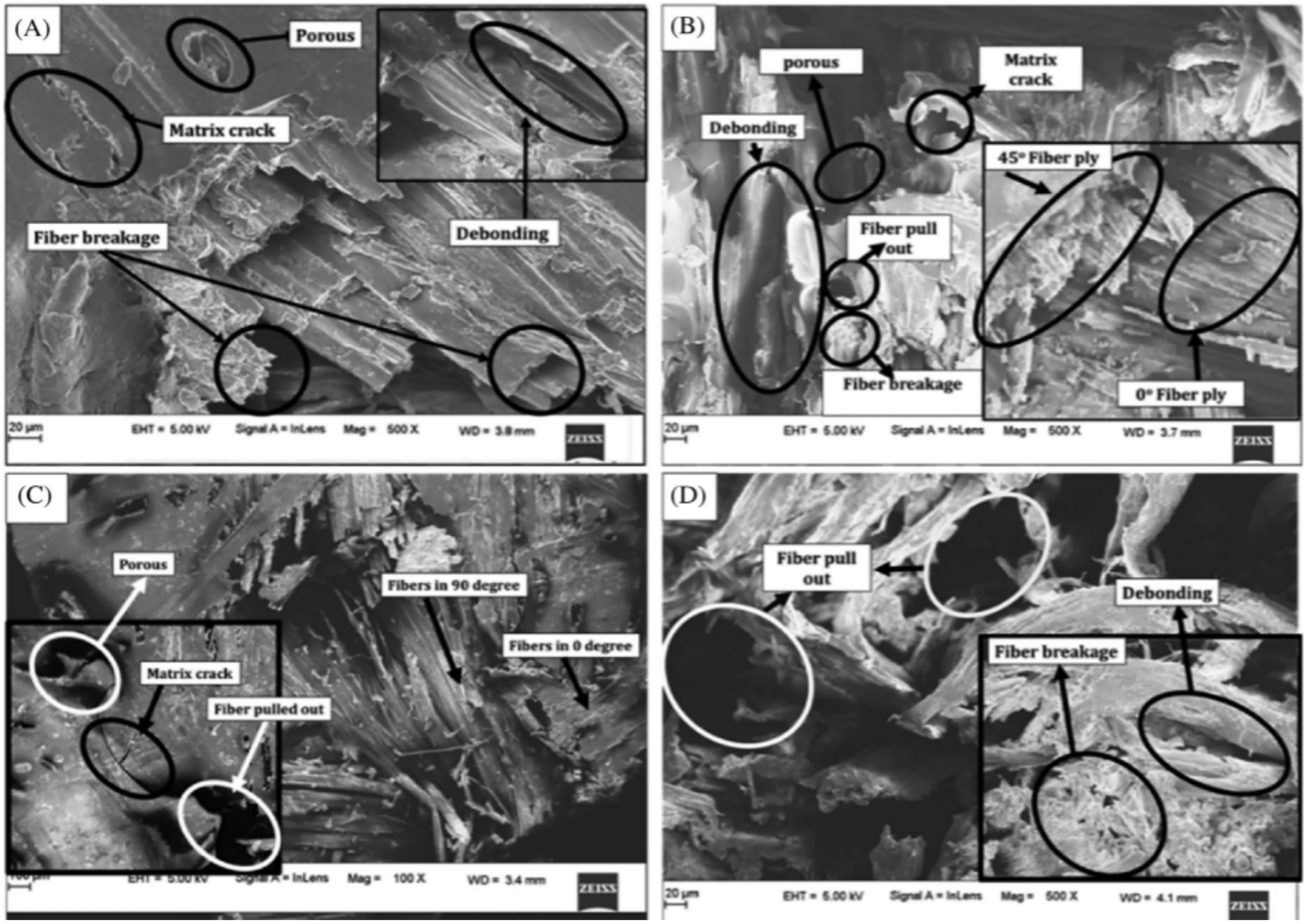

Fig. (22). SEM photomicrographs showing tensile test fractured surfaces of jute-banana PF NFHCs for different fiber orientations. (**A**) JB-1, (**B**) JB-2, (**C**) JB-3, and (**D**) JB-4 composites. NFHCs, natural fiber hybrid composites; PF, phenol formaldehyde; SEM, scanning electron microscopy [109]. Adapted from Fibers Polym, Ozturk B, Hybrid effect in the mechanical properties of jute/Rockwood hybrid fibers reinforced PF composites. 2010; 11(3), 464-73. *(A higher resolution / colour version of this figure is available in the electronic copy of the article*).

fracture of jute-banana NFHCs. As can be seen in there are very few fiber pullout places. Fiber matrix interfacial adhesion is good in the JB-1 composite, which results in better tensile strength and modulus. This is a good indicator. The JB-2, JB-3, and JB-4 composites had increased matrix cracking and fiber pullouts, thus resulting in weaker interfacial bonding in inferior tensile characteristics.

7. APPLICATIONS OF NATURAL FIBERS REINFORCED PHENOL FORMALDEHYDE COMPOSITES

Uses of natural fiber such as jute, kenaf, flax, *etc* reinforced phenol formaldehyde composites are wide. These biocomposites are applid in the fields of aerospace, building blocks and automotive. Ecological concerns regarding synthetic composites have made the development of biocomposites necessary. Areca fiber reinforced phenol formaldehyde composites give 40% better structural packing results than conventional wood-type packing systems. They have been widely used for the production of billiard balls, laboratory counterparts, and as coatings and adhesives [110, 111]. Due to their excellent thermal stability and nonflammability nature, they can be used in the marine transportation system and automotive. Natural fibers can be used instead of harmful materials like asbestos. Because of their good mechanical and physical properties, pineapple fiber reinforced PF composites are used in brake friction material design. This one is more economical and environmentally friendly than the synthetic one. Research studies show that fruitful applications of biocomposites in furniture, railways, and irrigation systems by the combined effect of mechanical and chemical treatment of wood fiber led to several changes to the wood [112-116].

In terms of vehicle parts, natural fiber-reinforced phenolic composites can outperform steel and aluminium in terms of tensile strength distribution. Phenolic compounds reinforced with natural fibers have the potential to replace metals in new and larger applications as resin materials. The orientation of the fibers and the technology used to fabricate composites have a significant impact on their mechanical strength. Parts for aviation and industrial machines, including automobiles, could benefit from the use of long fiber-reinforced phenolic composites. PF-natural fiber composites are widely recognized in the aviation, transportation and construction industries because of their resistance to ignite, low smoke generation, high-temperature resistance, and strength.

CONCLUSION AND FUTURE SCOPE

This review paper makes an attempt to discuss the work in natural fiber-reinforced PF composites. Studied composites influence mechanical, electrical, morphological, tribological, diffusion, visco-elastic, and thermal properties. Natural fiber reinforced /PF has a wide scope in the future. Ecological concern makes us use natural fiber instead of synthetic ones. For the development of high-performance engineering materials with fire-resistant capacity, it appears that fiber-reinforced phenolic composites are attracting increased attention from academics and industry.

For a variety of reasons, natural fiber polymer composites have recently attracted increased attention. The search for more fuel-efficient automobiles, less expensive and better building materials and a growing public concern for environmental preservation are a few examples. Because of its simplicity of manufacturing, increased productivity, decreased density and weight, and use of renewable resources, natural fiber reinforced composites (NFRCs) have become increasingly popular in recent years. Aside from these drawbacks, bio-based composite materials have a number of advantages over traditional materials, such as dimensional stability and processability issues. Creating a good interface (or interphase) for stress to be transferred between two different materials is essential in any good composite. The automotive sector has been approached to overcome these issues by using NFRCs in a number of exterior and interior panel applications because of the large weight reduction and inexpensive cost of the raw constituent elements. However, further research needs to address significant material and production obstacles before commercially available natural fiber reinforced PF composite can be widely used in the industries. The development of new phenolic and biophenolic FRP composites and their curing properties is the next challenging aspect. The physical, mechanical, and thermal properties of green composite materials using phenolic resin will be improved for structural and non-structural applications.

LIST OF ABBREVIATIONS

AFF	=	Areca Fine Fiber
CM	=	Chemical Molding
NFRCs	=	Natural Fiber Reinforced Composites
PF	=	Phenol Formaldehyde
RTM	=	Resin Transfer Molding
SF	=	Sisal Fiber

CONSENT FOR PUBLICATION

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CONFLICT OF INTEREST

The authors declare no conflict of interest, financial or otherwise.

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