# Applications of High Melting Point Natural Fiber and Green Composite Materials: A Review

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## Abstract

Natural fibers and green composites may compete with conventional materials in the structural and transportation sectors. We currently know and utilize natural fiber-reinforced polymer matrix composites (NPMCs). These composites have been studied extensively in the literature, and new findings are being made. They pose an unwelcome threat of flammability in the majority of applications. Fire-resistant natural fiber and green composite materials have newly gained attention from researchers. This article focuses on natural fiber and green composite flammability, which provides an overview of recent developments. The mechanism for fire retardancy has been described. All factors that affect fire retardancy and how to measure it have been entirely covered. Fire retardants have been shown to affect composite's other properties.

**Keywords:** Fiber, fiber-reinforced polymer, Green Composite Materials, Flammability, Fire retardants

#### 1. Introduction

Renewable, cost-effective, and environmentally friendly materials have emerged in response to environmental concerns and the increasing cost and diminishing supply of conventional synthetic materials. Most non-biodegradable and toxic inorganic waste materials are inorganic wastes[1]. On the other hand, biodegradable materials include natural fibers, green polymers, and fillers, all of which decompose[2]. Natural fiber-reinforced composite materials are widely used in automobiles and construction because of their lightweight and low cost[3]. On the other hand, composites have several drawbacks, such as their tendency to burn quickly [4]. Research on the flammability of natural fibers and green composites is few at this time. They may be used in the automotive, aerospace, marine, construction, and electronics sectors if more research is done[5]. An in-depth look at current developments in the flammability of natural fibers and green composites is provided in this article. Fire retardant methods, fillers, and other factors that affect the flammability of composites are summarized in this paper.

Engineered techniques, less flammable polymers, and fire retardant (FR) additives are utilized to boost the fire resistance of polymer composites[6]. Despite being the cheapest, engineering is not considered safe. A durable polymer made of low-flammability polymers costs a lot. Compounds containing FR additives are the most widely used and well-accepted technique. Nano or micro powder fillers are often used in FR additives, which work in various ways to inhibit the spread of fire[7].

Wood flour/polypropylene composites containing zinc borate (ZB), montmorillonite, manganese dioxide (MnO2), and stannic oxide (SnO2) were developed[8]. FR additions had a positive synergistic effect on the composites' fire resistance. According to the researchers, Flame retardant epoxy resin composites using hemp fibers are being developed[9]. FR additives based on phosphorus were used to enhance the flammability of hemp/epoxy composites. Fire retardant, thermal, and mechanical properties of sisal/polypropylene composites may be improved by adding ammonium polyphosphate, magnesium hydroxide, and ZB[10]. Shah et al. improved a polypropylene matrix's fire resistance and thermal properties by adding oyster shell powder as an organic filler[11]. APP, MPP, and aluminum hydroxide (AlHx) were used by Umemura et al. to reduce the flammability of wood-plastic composites[12]. In the recent literature, further study on natural fiber reinforced composites (NFRC) is included in Table 1. In order to include FR materials into composites, the production process has to

be altered. The most crucial factor is to ensure that the additive particles are well incorporated into the other components of the composite. Improper distribution of flame retardant (FR) additives leads to unsatisfactory mechanical and water absorption properties[12]. There are several techniques to mitigate the negative consequences of FR additions[13].

# 2. Characteristics of Polymers

Composites rely on polymeric matrices, which look good and serve an important function. The polymeric matrix and manufacturing procedure greatly influence composite properties. It is possible to see the most common polymers and their physical characteristics in Table 2. Our everyday lives revolve around polymeric materials because of their unique properties. As a result, both natural and artificial polymers are highly flammable because of carbon's fundamental component. Polymer matrix (polymer) protects reinforcements in composites against fire propagation and heat loads. However, this is not always the case[14]. Several papers have dealt with polymers and polymer composites[15]. Polymer flammability may be described using characteristics such as LOI and HRR. Table 2 provides information on the flammability of some of the most commonly used polymers.

# **3.** Characteristics of Natural Fibers

Figure 1 illustrates the classification of natural fibers based on how they were taken from plants, animals, and minerals. For example, natural fibers are flammable because of their chemical composition. Plant fibers include cellulose, hemicellulose, and lignin, while animal fibers are mainly composed of protein[16]. Plant fibers have long been used as significant reinforcing fibers over the globe, although animal and mineral fibers are less well-known[17]. The flammability of plant-based natural fibers is the primary focus of this review paper. Seed, leaf, bast, fruit, and stalk are all sources of plant-derived natural fibers. The lignin, pectin, and wax combination give these fibers their structure. Natural fibers are flammable because of their chemical composition. Table 03 explains the role of fiber components in a product's qualities, as shown. The cellulose content of natural fibers determines their mechanical properties[18]. Hemicellulose was aids in the absorption of water and microbial and thermal degradation[19]. Hemicellulose is the flammable element of natural fibers, whereas lignin is the char-forming part[20].

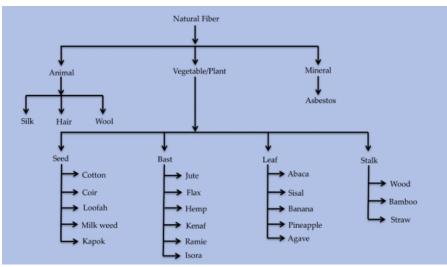


Figure 1: Natural fiber classification according to their source of extraction **Table 1:** List of newly published work on natural fibers or polymer matrix composites with FR additives

Natural fibers	FR additives	fibers/polymer matrix	References	
Rice husk &	Sodium metasilicate &	High-density	[21]	
sawdust	zinc borate	polyethylene		
Cellulose, Hemp, Ammonium		Poly butylene succinate	[22]	
Flax, Sugar cane,	polyphosphate			
and bamboo				
wood flour	Zinc borate	Polyvinyl chloride	[23]	
Jute	Halloysite nanotubes	Soy protein concentrate	[24]	
Coconut & jute	Di-Ammonium	Polylactic-Acid &	[20]	
	phosphates	Polypropylene		
Flax	Phosphorous	Poly butylene succinate	[25]	
	compounds			
Banana	Nano-Clays	Polypropylene	[26]	

Polymer	Density (g/cm3)	Tensile strength (MPa)	Tensile modulus (GPa)	Tm ( °C)	Thermal conductivity (w/m ° k)	Total HR (kJ/g)	LOI (%)
PE	0.925	15	0.08	105-115	0.33-0.51	41.6	18
PP	0.920	40	1.9	130	0.1-0.22	41.4	17
PAN	1.184	57	2.7	300	1.0	13.3	27
PC	1.2	70	2.6	157	0.19	20.3	26
PS	1.04	40	3	240	0.03	38.8	18
PMMA	1.18	47	2.2	130	0.20	24.3	17
PVC	1.4	51	2.4	160	0.19	11.3	45
PVA	1.19	40	1.7	200	0.31	21.6	20.5
PLA	1.2-1.4	50	3.5	150-160	1.13	14.2	21.7
РЕТ	1.38	55	2.7	260	0.15	15.3	21

Table 3: Shows the different types of fibers and their characteristics

Components	Properties
Cellulose	Mechanical properties
Hemicellulose	Moisture absorption, Thermal stability,
	Biodegradation, Flammability
Lignin	Char formation

# 4. Composites Flammability

The matrix of polymer rapidly degrades [28]. The thermal stability of polymer composites may be improved by adding fiber reinforcement at high temperatures. Natural fibers are much more susceptible than synthetic fibers[29]. As illustrated in Figure 2, the thermal breakdown of polymer composites in the presence of combustion is a multistage process. A composite is broken down into various gases and char during a fire. In the presence of oxygen, flammable gases may be more easily ignited. Gases released as a result of combustion include CO2, CO2, and smoke. The breakdown process is aided by the heat generated during combustion, and the burning cycle is self-sustaining. The flammability of polymer composites may be affected by tampering with the combustion process at any point. Burning char layers are often used to stop heat transfer in contemporary fire retardant methods[30].

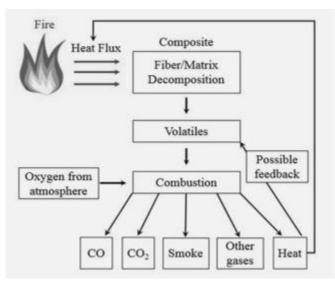


Figure 2: Polymer composite thermal degradation

# 5. NFRC Properties Affected by FR Additives

There is a focus on FR additives-based fire retardant technologies in this review paper. Additives for fire retardancy (FR) may increase the fire resistance of composites, but it is essential to know how they impact other properties. The mechanical, water absorption, and thermal properties of NFRC are examined in this work due to the addition of FR.

# **5.1 Mechanical Properties Effects**

A strong interfacial connection improves mechanical properties when the FR additions are chemically compatible with the matrix and natural fibers. In contrast, most FR additions do not interact with composite components chemically and instead function physically. Fire-retardant additives to NFRC may increase fire resistance while decreasing NFRC's mechanical properties. Mechanical strength is reduced by deboning, stress concentration, and the buildup of filler powder in composites. As a result of the high rigidity of the composite materials with FR additives, the elastic modulus may increase.

Polypropylene (PP)/wood-fiber composites were tested for the effects of ammonium polyphosphate (APP) and silica on their mechanical properties and flammability[4].

The mechanical properties of PP/wood composites were reduced with fillers. Tensile strength increased slightly for smaller silica loadings, but this effect disappeared as silica loadings increased. Experts say experts reduced mechanical properties due to incompatibilities between FR additives and the matrix. Aluminum trihydroxide (ATH) was employed as an FR additive in Flax/PP[31]. Grades 32, 40, and 60 CD of Apyral were used in the experimentation. Tensile and impact strength were decreased, despite a gain in fire resistance and thermal stability.

Composites' stiffness was measured by the elastic modulus, which rose. It was found that the APP filler had a significant impact on the biocomposites made from ramie fibers and polylactic acid (PLA)[32]. At lower filler loadings, the APP increased the fire resistance of ramie/PLA biocomposites without compromising their mechanical properties. However, the mechanical properties of the composites were observed to decrease with higher filler loadings substantially. The polymer/filler contact was hindered by the presence of APP, resulting in a decrease in mechanical properties. Flax fiber-reinforced PLA/TPS biocomposites were studied by Bocz et al., and one of the FR additions they used was APP, which has a wide range of applications[33]. APP decreased the HRR rate of the composites in order to increase fire retardancy. However, this came at the sacrifice of tensile and

flexural strength. As an inorganic filler, Shah et al. employed OSP (oyster shell powder) with PP at various doses (FR additives)[11].

The inclusion of OSP led to an improvement in PP's fire resistance but a decrease in its tensile strength. With a decreased tensile strength, burning time is increasing. The poor interfacial bonding between the FR additives and matrix resulted from the incompatibility between OSP and PP. FR additives and the matrix may sometimes be made to work better together via a compatibilizer. Filler dispersion and chemical bonding between filler and matrix may increase mechanical properties. There are a few reported cases of FR additives being used to increase the fire resistance and mechanical properties of NFRC. According to Biswal et al., the nano-clay/banana fiber/PP fire retardant composite demonstrated better tensile and flexural capabilities[26]. The scientists used maleic anhydride grafted polypropylene (MAPP) as a compatibilizer, which helped bind the nano-clay to the PP matrix.

Polymer chains were inserted into silicate clay layers to enhance the surface area of contact between silicate clay layers and the polymer matrix to improve mechanical properties. Using Cloister 30B (C30B) nanoclays as FR additives, Paluvai et al. studied the properties of unsaturated polyester (UP) toughened epoxy nanocomposites reinforced with sisal fibers in their study[34]. Epoxy/tensile, UP's flexural, and impact properties improved with the addition of C30B- and alkali-silane-treated sisal fibers. Chemical bonding between alkali silane-treated sisal fiber and the epoxy/UP system was claimed to improve the material's mechanical properties.

## 5.2 Water Absorption Effects

FR additions boost the NFRC's water absorption capability since natural fibers are often hydrophilic. Since the filler particles have uneven shapes and poor interfacial interactions with the matrix and natural fibers, some micro-gaps and fractures occur at the composite interfaces. These flaws allow water to seep into the structure. The impact of FR additions on NFRC's water absorption has been studied in the literature.

Composites made of wood flour and expanded polystyrene waste were tested for diammonium phosphate (DAP)[35]. Fire resistance was improved while the DAP modification increased water absorption. Hydrogen interaction between water molecules and free OH groups in the DAP structure and possible diffusion of water molecules into the composites were cited by scientists as the mechanism for water absorption. CaCO3- and DAP-filled PP/kenaf composites were studied for their properties[36]. The PP/kenaf composites' water absorption improved by adding FR additives. Calcium carbonate, according to experts, absorbs water, causing the composite to grow in weight as a result.

The water resistance of nanocomposites containing FR additives may be improved by taking specific measures. FR additives boron and phosphate compounds were used to study wood/PP composite materials[26]. The composites employed MAPP as a coupling agent. The water absorption parameters of MAPP and without MAPP were compared. A team of researchers found that FR additions to composites without utilizing MAPP lowered their water resistance, but that after applying MAPP, the water resistance increased. Coupling agents improve interfaces by sealing and reducing water infiltration into the composites' interior flaws and fractures. Nano-clay-filled cellulose fiber/vinyl ester nanocomposites were tested for water absorption[37]. A reduction in water absorption was seen after adding nanoclay to the composite materials. In the water molecules pathway, the high aspect ratio of nano-clay platelets is to blame.

#### **5.3 Thermal Stability Effects**

The FR additives often form a char deposit when burned at high temperatures. The char layer acts as a heat-resistant medium on the composite surfaces. It is also worth noting that, as opposed to the

FR additions in the polymer matrix, FR additions are more prone to crystallization and melting. During burning, the char layer protects the fibers since the breakdown temperature of cellulosic fibers is very low. According to the literature, the thermal stability of composites may be improved by including FR compounds. Simal fiber/PP composites were tested for the impacts of APP, magnesium hydroxide, and ZB on their performance[10]. When heated to 500 degrees Celsius, polypropylene completely degrades, losing all of its weight. The decomposition temperature of sisal fiber is lower than that of polypropylene (PP).

Despite the author's FR additions, char residues were formed in the sisal/PP composites to increase their heat stability. Thermal barrier properties of PBS/BF biocomposites may be improved by employing microencapsulated ammonium polyphosphate (MCAPP) as an FR addition, and the char residue serves as a barrier at high temperatures [38]. When PBS was reinforced with BF, it lost some of its thermal stability; however, adding MCAPP to the PBS/BF composites restored that stability. Composites made with MCPAP are more thermally stable than those made with non-MCAPP components. Wood/PP composites containing melamine, ZB, APP, ATH, natural flake graphite, and expandable graphite were studied for their reactivity to fire[39]. When the composite specimen was exposed to high temperatures, the authors calculated the mass-loss rates of the composite specimen. To their surprise, they found that char residues in the wood/PP composites with FR additions are higher than those in the typical wood/PP specimen. In terms of mass loss, flame retardant-based wood/PP composites had a lower rate than non-flame retardant-based wood/PP composites. The wood/PP composite filled with APP and expandable graphite was determined to be the most stable in terms of thermal stability. Nano silicon dioxide (APP) and wood fiber/polyethylene composites were studied[40]. Thermogravimetric analysis (TGA) and derivative thermogravimetry (DTG) were employed to measure the weight loss of composites at high temperatures (DTG). The composite's thermal degradation was minimized when APP and nano-SiO2 were added.

Both FRs resulted in a char deposit on the surface and increased heat resistance. Spartium junceum L. short fibers reinforced PLA biocomposites were tested for the effects of montmorillonite nanoclay[41]. According to the researchers, the thermal stability of nano clay-treated biocomposites was superior to that of untreated composites and pure PLA. Using nano clay as a "charring agent" reduced the rate of weight loss at higher temperatures. There was a paper on the properties of OSP-filled PP composites[11]. An FR additive, OSP, was used to enhance the heat stability of composites. When OSP is added, the thermal stability of pure PP is believed to increase significantly. Airborne TGA thermograms of composites made of OSP and PP (as a medium). Findings show that PP declined in a single step, whereas OSP/PP composites degraded in two phases.

# 6. Flame Retardant (FR) Additives Types

The use of additives for flame retardant coatings, plastics, thermoplastics, thermosets, rubbers, and textiles. These FR additives may be used to prevent, reduce, suppress, or halt the combustion process of materials. Polymer flames may be extinguished or reduced in various ways using these devices. In 2012, a market study found that [42], Four years ago, FR retardant usage was at its lowest level ever.

#### 6.1 FR Additives with Halogens

This class of FR additives is mainly used in producing everyday items that we all rely on. Depending on the ratio of halogen to carbon and the bond energy of the carbon-halogen bond, they are excellent in capturing free radicals and releasing them. Consequently, they may be able to stop or slow the spread of fire[43]. Halogenated FR additives typically employ a gas phase approach to extinguish fires. These FR additives reduce flammability in the environment by interacting with ambient oxygen and hydrogen, which lowers both oxygen and fuel concentrations in the air. Because aromatic halogen FR additions have lower binding energy, they are less effective than aliphatic and

alicyclic FR additives. FR additives can only be utilized with fluorine, which has a high electronegativity, and iodine, which has a low one. At greater temperatures, however, bromine creates more free radicles than chlorine. On the other hand, Flame retardant chlorine has a higher level of efficacy. The decomposition of halogenated FRs is shown in Equation 1.

 $R-X \longrightarrow R^*, HX+H^* \longrightarrow H_2+X^*$ 

 $HX+OH^* \rightarrow H_2O+X^*$ .....(1)

Reaction mechanism of halogenated FRs (R-X=FR; X\*=Cl, Br)

#### 6.1.1 Bromine FR Additives are a kind of bromine

Because of their low cost and high-performance efficacy, bromine FR additives are often used in commercial and domestic items such as construction materials and electrical equipment to protect against property damage and harm. These flame retardant chemicals degrade at a lower temperature to achieve the desired results, and their use is constant across all compounds[44]. The most prevalent brominated FR additives are brominated bisphenols, diphenyl ethers, cyclododecane, phenols, and phthalic acid derivatives.

#### 6.1.2 FR Additives with Chlorine

Additives for polyalkene compounds, such as chlorinated FRs, are common. In order to be effective, they must be used in more significant amounts because of their poor stability and plasticization at higher temperatures compared to the brominated FR additions. Compared to the chloride FR additives, a combination of metal chloride may enhance flame retardancy even further[45]. Paraffin and alkyl phosphate are the most frequent kinds for most chlorinated fire retardants.

Hydrochloric compounds are released in the gaseous phase; in addition, these hydrochloric compounds break down and generate chlorine-free radicals that oxidize volatile products during the thermal breakdown of polymers.

# 6.2 FR Additives with Phosphorus

As a result of environmental concerns, the phosphorous FR additions are used as halogen-free FR additives with the exact flame retardant mechanism as halogenated FR additives [46]. Metal phosphates may have a gas phase action and a condensed phase effect (char formers and enhancers). These FR additives are critical with low concentration levels and compelling action against fire. Combined with the polymer's high oxygen content, they are active and restfully on the matrix structure.

Some of the most well-known phosphorus-containing FR additives include phosphate esters, phosphonates, and phosphinates. Some examples are Triphenyl phosphate, Triphenylphosphine oxide, Tris-(2-chloroethyl) phosphate, Tri (chloroprene) phosphate, Resorcinol-bil (diphenyl) phosphate, Red phosphorous, Ammonium polyphosphate, Melamine phosphorus.

Polymeric materials' solid phase may benefit from the inclusion of phosphorus FR. These phosphoric FR additions are converted into their respective acids, then changed into polyacids by the thermal breakdown. Unsutured char compounds are formed when acids esterify and dehydrate the pyrolyzing polymer. Polyphosphoric acids prevent the carbonaceous layer from further pyrolyzing at high temperatures, safeguarding the polymer's constituent parts from exposure to oxygen and radiant heat.

#### 6.3 FR Additives for Nitrogen

They are environmentally friendly because of their low toxicity, less smoke during fire evaluation, and easy recycling. These compounds remain inert and do not break down even at high temperatures.

As a result, they can halt the decomposition of materials at greater temperatures. A greater level of flame retardancy is achieved by nitrogen-containing additives (nitrogen-phosphorous) due to the synergistic impact between nitrogen and phosphorous. It is also a weak acid with minimal corrosion potential, making it perfect for producing electrical installation components from the combustion of hydrogen cyanide.[47]. Nitrogen FR additives such as melamine polyphosphates, melamine poly (zinc/ammonium) phosphates, melamine-based hindered amine light stabilizers, melem, and melon are the most commonly used melamine and melamine compounds. Also, ammonium polyphosphate (as described in the text) is used as a component of olefin intumescent compounds, including melamine cyanurate. Methylamine Pyrophosphates (MPPs) and other nitrogen-phosphate compounds are used in amides. Reactive compounds are urea, isocyanurates, triazines, guanidine, and cyanuric acid derivatives.

In addition to gaseous and condensed phase methods, nitrogen FR additions act several ways. Stable nitrogen-based molecules may be released in the gaseous phase to limit fire propagation, and complex nitrogen compounds can be created in the condensed phase to generate char to protect polymer materials from the breakdown in the fire. Burning may be hindered by releasing nitrogen-mixed gases from these materials.

## 6.4 Additives to Silicon FR

Several silicon compounds, including silicones, silica, organosilanes, silsesquioxane, and silicates, have been studied as suitable flame retardants for polymeric materials.

#### 6.4.1 Silicons

It has long been accepted that silicones and silicon co-additive FR additives are functional and ecologically friendly. It is possible to utilize fewer amounts of these compounds because they dilute more organic components and offer a barrier on the surface of the materials to a growing flame. Flame retardant properties may be improved during deterioration because of their low surface energy characteristics. Mechanics, significantly impact resistance might benefit significantly from these.

# 6.4.2 Silica

As a filler, silica combines silicon and inorganic components that may also be used as an FR additive. Several commercial polymers have also been made using this material. Multi-coordinate organic silicone compounds are synthesized when silica and organic alcohols are chemically combined with a catalyst[48],[49].

# 6.4.3 Silanes

There are a variety of chemical structures and functional groups in silanes, which are silicon-based compounds. Polysilanes, poly carbosilanes, and poly silazanes are among the most crucial silane compounds. Silanes may bind fillers and polymer matrices utilizing simple synthetic processes as coupling agents. Char formation and the improvement of physical and dynamic properties are also enhanced by the presence of these compounds[50].

These include polyhedral Silsesquioxanes, such as dimethyldichlorosilane, methyltriethoxysilane, methyltriacetoxysilane, and other polyhedral Silsesquioxanes.

#### 6.4.4 Silicates

Some polymers may benefit from silicates, which include anionic silicon in oxides (clay). This nanofiller is inexpensive, environmentally benign, and has unique mechanical properties. They are incorporating less silicate into the polymer matrix results in a significant reduction in inflammability. By reducing oxygen permeability, they mostly succeed in raising thermo-oxidative stability.

Mechanically enhanced char, combined with silicates, lowered dripping and, hence, decreased flammability in these flame-retardant additives' principal mode of action[51].

Silicon oxomonophosphate  $(Si_5O (PO_4)_6)$ , talc  $(3MgO.4SiO_2.H_2O)$ , Montmorillonite (phyllosilicate). The condensed and the vapor phases of silicon FR additives are essential in their performance. To arrest the degradation process, dilute organic gases, trap active radicals in the flame zone, and prevent heat and mass transfer in the condensed phase, silica may migrate to the surface at high temperatures in the condensed phase. Only when coupled with other fire-retardant additives, can the qualities of these be used to their full potential.

#### 6.5 FR Carbon Additives

## 6.5.1 Carbon Nanotubes

Single, double, or multi-walled graphite sheets and a polymer matrix as an addition make up carbon nanotubes. On the other hand, carbon nanotubes are far more costly than carbon fibers/fabrics or fillers, but they have a unique virtue of flame retardancy at low concentrations. The mechanical properties are also improved, and they are more efficient than nanoclays. In addition, the trapping of free radicals may reduce the pace at which the polymer matrix breaks down, hence increasing its thermal stability. Carbon nanotubes with hydrocarbon polymers are superior to hydrophilic additions in terms of practicality. They may be included directly into the polymer matrix without the need for any organic treatment or compatibilizer. Organophilic[52].

## 6.5.2 Hydroxycarbonates

Hydroxy carbonate FR additives are an environmentally beneficial alternative to metal hydroxide flame retardants. Fire retardant additives, including non-toxic substances like water and carbon dioxide, release water in the gas phase, reducing flame and creating 20 times less smoke than halogenated FR additions. These pieces are both gaseous and condensed. Breaking down and oxidizing carbon-containing compounds may help create an insulative barrier layer on the matrix polymer. The most often used hydroxy carbonate FR additions are magnesium carbonates (natural (magnesite) and synthetic (hydromagnesite)) and calcium carbonates. An increased flame retardancy and mechanical properties may be noted with other FR additives, such as clay nanoparticulates[53].

Thermal stability and flame retardant properties are influenced by carbon nanotubes, polymer matrix, and nanotube dispersion in the polymer matrix. As a result of condensed phase mechanisms, the melting viscosity of the polymer matrix is increased by using carbon nanotubes in threedimensional cross-link network designs. The polymer matrix is protected from flame propagation by its network structure. A multiple char layer is formed over the polymer matrix when the nanotubes are combined with other flame retardant chemicals. This improves the polymer's heat stability and flame retardancy.

## 6.6 Hydroxides and Metallic Oxides

When metals react with oxygen in the air, they produce metallic oxides. In addition to the strong interactions between the polymer surface and the nanoparticle surface, these FR additives have proven effective additives for improving thermal stability and morphological structure of char residues and the FR of polymers because their degradation temperature is higher than polymer processing temperature[54]. It is possible to make titanium dioxide by heating titanium (TiO2) to a high enough temperature to form a titanium oxide (TiO2).

Metal hydroxides are mineral FR additives that include a mixture of hydroxides and carbonates. Inexpensive-smoking FR additives typically employ them because of their low cost and wide availability. Using an endothermic process, these flame retardants generate one-third of the total metal hydroxide and an inorganic fire-retardant residue. Using these metal oxides, flame propagation is reduced. Polymer flammability is seldom improved by combining them with other flame retardants, although they are rarely used as synergists[55]. Aluminum tri-hydroxide (Al(OH)3), magnesium di-hydroxide (Mg(OH)2), antimony trioxide (Sb2O3), expandable graphite, and others are examples.

## 6.7 Borate FR Additives

Commercial inorganic family members include fillers with minimal mammalian toxicity and mild volatility safe for the environment and mammals. Additives like halogenated and mineral fillers are typically utilized as a synergist because of their poor charring performance when used on their own. Fire retardants like crystal water, boric acid, and boron oxide (endothermic decomposition (503 KJ/g) between 290 and 4500 C) are formed as a result of this endothermic reaction in the combustion process, which results in the formation of a glassy shield layer that blocks fire propagation and stops polymer chain oxidation[56]. ZnO, ammonium pentaborate, melamine borate (C3H9N6O3B), and barium metaborate are some of the most often used forms of the mineral borates, which are also known as borates.

#### 6.8 Nano-Clays

These fine-grained minerals (phyllosilicates) are found in rocks, sediments, and soils as nano-clays and are a kind of natural inorganic material. More smoke and carbon dioxide can be produced using nano clays because of their superior thermal stability. After burning, a thick silicate layer is formed on the polymer matrix, which functions as a fire barrier and inhibits the degradation of the composite. In addition, when burned, these materials may produce a carbonaceous layer that prevents oxygen permeability while leaving the flame unquenched. At high temperatures, nano clays lower polymer viscosity, preventing polymer composite leakage and increasing char formation. When combined with other FR additives, nano clays function even better as flame retardants. These techniques may also improve polymer composites' mechanical properties, scratch resistance, glass transition temperature, and stiffness [57].

#### 6.9 Natural FR Additives

# 6.9.1 Chitosan (CS)

Deacetylated chitin (the exoskeleton of living animals such as fungi (cell walls), crustaceans (shrimps and crabs), and insect cuticles) is used to produce chitosan a non-toxic, highly biocompatible organic polymer. When it comes to carbohydrates, the most common one to use is Chitosan, which is an amino polysaccharide that may be carbonized and has many hydroxyl groups. Thus, chitosan might help to produce char during the burning process. Because of this, chitosan is used in polymer composites as an intumescent flame retardant and a natural FR additive. Burning chitosan may produce a blackened layer on polymer materials' surfaces that acts as an effective heat insulator or thermal shield to contain fire[58].

## 6.9.2 Oyster Shell Powder (OSP)

A mineral filler made from oyster shells that are biodegradable, non-toxic, and does not accumulate in the environment may be found on the beaches of many nations. Calcite and aragonite crystals (pH 9.8) make up around 96 percent of their composition. Above 8000 degrees Celsius, an oyster shell decomposes to produce CaO and CO2 as a byproduct. Carbon dioxide, a byproduct of combustion, may put out a fire by blocking oxygen from reaching the flames. Carbon dioxide is the only source of energy for the fire-retardant mechanism. Oyster shell powder is used in plastics and polymer-based composites as an FR additive for ecologically acceptable fire retardant polymers. Additionally, oyster shell powder enhances the plastic's rigidity, flexural, and dielectric properties[11].

6.9.3 Egg Shell

Recyclable, biological, and calcium-rich eggshell is common in city dustbins and rural farmyards. It comes from various sources, including the food industry, the chicken industry, and even the average household. According to a report from China, Chinese egg output in 2013 was estimated to be 484.6 billion eggs. CaCl2 (calcium and magnesium carbonate) is 96% of the egg shell'S chemical composition; the rest is composed of X collagen, Sulfated polysaccharides, and various proteins (including Gly and Arg). When heated, the eggshell's calcium carbonate decomposes into CaO and CO2, and CO2 is a suitable fire extinguisher, preventing the egg from catching fire. More CaCO3 in the polymer matrix results in a composite with a decreased risk of catching fire due to the eggshell's abundance and particular composition and structure. Polymer nanocomposites may benefit from their use as a bio-filler and an FR additive[59].

#### 6.9.4 Lignin

Polyphenolic and structurally amorphous, lignin contributes 20 to 30 percent of the plant's total weight. When heated to 4500 C in a nitrogen environment, it breaks down into stable, dense cross-linked char, increasing density as temperatures rise. Flame retardant chemicals are well-known for forming char during thermal degradation, which slows the combustion rate of polymeric materials by preventing oxygen from entering the combustion zone. Due to the cross-linked structure, including phenolic groups, lignin may be employed to improve the thermal properties of polymeric materials since it provides a large char production during decomposition[60]. Additional advances in thermal and flame retardant properties may be achieved by changing the chemical structure of lignin with phosphorus and nitrogen components[61].

## 6.9.5 Biochar

Charcoal formed from biomass is known as biochar. Nitrogen, hydrogen, oxygen, and ash are all present in the microporous carbon-rich material. It results from the thermochemical treatment of organic waste and subsequent pyrolysis at high temperatures. Carbonaceous material keeps its physical form and honeycomb structure with distinct functional groups on its outer surface. Polymer firms and research institutions are interested in its important position in many purification systems as remedial pollution assistance and a long-term carbon detachment technique because of these characteristics. In addition to its high porosity, antibacterial and antifungal qualities, breathability, and temperature management, biochar may also be used to reduce and absorb odors, making it an ideal material for odor control. Biochar's components inhibit reactive chemicals during the heating process and limit weight loss in composites. The thermogravimetric and differential scanning calorimetric studies found that the thermal stability of the biochar-filled composite was enhanced in the literature[62].

#### 6.9.6 Human Hair

An alpha-helix structure of amino acid units connected by a peptide (-CO–NH–) bond is found in the protein keratin, which accounts for roughly 91% of hair. In hair, amino acids such as cytosine, threonine, glutamine, glycine, leucine, and valine are present. On average, hair has 45.68 percent carbon, 27.9 percent oxygen, 6.6 percent hydrogen, 15.72 percent nitrogen, and 5.03 percent sulfur in it. Because it is convenient, inexpensively, abundantly, and collectible, this is the most convenient biomaterial to transport and store. It has long been used as a biomarker for human exposure to medicines and heavy metals. In addition, since hair contains nitrogen and hydroxyl groups that may be implicated, a few studies have looked at hair to quantify human exposure to persistent organic pollutants, including flame retardants[63]. Researchers Chai et al. have concluded their investigation into the thermal properties of human hair. Researchers are currently trying to assess bioderived composite materials' flammability by utilizing human hair as a fiber in their experiments[64].

# 7. Surface Treatments' Effect on Flammability

Additionally, surface treatments directly impact the fiber's mechanical and thermal properties. By removing as much amorphous material from the fabric as possible during the surface treatment process, a higher proportion of crystallinity is achieved in the fiber. The fabric's surface area is increased when the threads' connections become more organized, increasing surface area. Compared to natural fiber composites that have not undergone this treatment, the wettability and bonding between the material and the resin are improved, resulting in a more thermally stable composite[65].

#### 8. Flame Retardancy Mechanism

FR additives suppress or diminish the flammability of polymer matrix composites by a variety of methods, including physical dilution, chemical interaction, inert gas dilution, thermal quenching, and protective (char formation) coatings.

#### 8.1 Physical Dilution

The FR additives themselves serve as inert substances and have thermal capacity against flame to reduce the fuel concentration below the lower flammability limit. It is also impeded by cooling, barrier development, and changes in heat capacity, thermal conductivity, and viscosity throughout the physical dilution process. One or more physical mechanisms are always involved in chemical processes, such as the dissociation of fuel in the presence of heat. Nano clays and glass filler compounds operate when used as FR additives.

## **8.2 Chemical Interaction**

Free radicals produced by the combustion of polymers are disrupted and captured by FR additives or their byproducts in this breakthrough process. Neutralizing free radicals allows for cooling and a reduction of inflammability. Most halogenated (brominated) flame retardant (FR) additives act this way. To restrict flame growth, OH\* and H\* free radicals formed from polymers react with halogen-free radicals in the gas phase.

#### 8.3 Inert Gas

Evaporation and condensation reactions of FR additives produce large volumes of non-flammable gases or inert gases in the gas phase. These inert gases contaminate the air around them, reducing the concentration of combustible fuel. Chemicals like metal hydroxides and carbonates may produce nitrogen in this way.

#### 8.4 Thermal Quenching

Endothermic chemical reactions release water molecules from FR additives during burning at high temperatures and use thermal quenching (sudden cooling) to dilute the combustion process (polymer matrix). Halogen-free flame retardant additives, metal hydroxides, and carbonates are all examples of products that use this technology.

## 8.5 Char-Formation

A carbonaceous char coating is formed on the polymer matrix, shielding it from further oxidation, preventing further flame activity. Thermal insulation is improved, combustible gases are blocked, and the layer of carbonaceous char increases thermal capacity. This approach is often used for halogen-free flame retardant chemicals, such as phosphorus and nitrogen.

# 9. Conclusion

Composites made of natural fiber-reinforced polymers and flame retardant additives were examined in this research. Polymer matrices with a broad range of natural fibers might exacerbate the problem of the flammability of natural fiber-reinforced composites. Flame-resistant composites may be created with the help of proper FR additives, which widens their uses. With polymers, even small amounts of additives may have a positive impact. In matrices containing oxygen or nitrogen backbones, phosphorus and nitrogen-based chemicals have shown to be highly efficient nonhalogenated FR additives, while silicon-based compounds also seem to be helpful. Even yet, it is tough to boost the flammability of natural flame-reinforced composites. However, flame-resistant natural fiber-reinforced polymers may be produced by carefully selecting and applying the best FR additives.

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