
NATURAL FIBRE POLYMER COMPOSITES, THEIR RESPONSE TO FLAME AND APPLICATIONS: A SURVEY

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ABSTRACT

Natural fibre reinforced polymer composites are fast dominating the synthetic ones in the composite world. Light weights, non-susceptibility to corrosion, and eco-friendly benefits of natural fibre composites have attracted the attention of many researchers. Natural fibres consist of three major natural constituents namely; cellulose, lignin and hemicelluloses that are extremely responsive to flames and sequel to this, their uses are limited particularly to fire-prone areas. The addition of flame retardants during fabrication of natural fibre reinforced composites helps to enhance their flame properties and thermal stability. In this work, recent status of available natural fibres in various polymeric composites, their reactions to fire have been surveyed. From the studies carried out, we got to know that there are still gap in knowledge on this topic, therefore, a need for more research and publications in this promising area.

Keywords: Natural fibre, polymer composites, flame retardants, flammability properties, cone calorimeter, Underwriters Laboratory (UL-94)

1. INTRODUCTION

Natural fibres being environmentally friendly and renewable in nature attract lot of scientists, researchers and engineers for their use in polymer composites. This study aims at providing a thorough review on reaction to fire of the commonly used natural fibre polymer composites (NFPCs) and their uses in various fields. It equally covers the summary of various flame-retardant additives and their effects on various properties of polymeric composites reinforced with natural fibres. The type of fibre, fibre orientation,

interfacial bonding and fibre aspect ratio can bring about variation in the properties of NFPCs. Some of the shortcomings of NFPCs are poor flame resistance, high water absorption and lower mechanical properties. These shortcomings restrict their applications in various fields. NFPCs find applications in construction industry, automobile industry, and textile and for structural goals. To obtain good mechanical properties, chemical treatment of natural fibres is done. The treatment helps to improve the adhesion between the fibre surface and the polymer matrix [1- 3]. It is pertinent to note that high cellulose in natural fibres is what causes high flammability while high lignin content reduces the flammability by forming char [3, 4]. The char formed during the burning of the composite creates a barrier for heat transfer. Flame retardants are usually added to help delay, prevent fire or its propagation by interrupting or impeding the combustion process and thereby causing a delay in the spread of fire or in some cases discontinuance depending on the type and contents of the flame retardants used [3]. Many flame retardant additives have been studied, and their major effect is to reduce the flammability of the composite.

2. NATURAL FIBRES

Natural fibres are many in nature and vary from region to region across the globe. According to Kiran [5], fibre is described as entity that is elementary and linear, has a characteristic cross-sectional and longitudinal shape and made up of a primary chemical substance. Fibres are mainly cellular in form and structure with a degree of inherent cell walls. The cell walls contain mainly sugar based polymers (cellulose, hemicelluloses) usually on dry basis [6]. Natural fibres are classified based on their origin, either as plants, animals or minerals fibres. All animal fibres contain protein, while plant fibres contain mainly cellulose [7]. Plant fibres are also called ligno-cellulosic fibres because they contain cellulose and other non-cellulose materials such as pectin, lignin and hemi-cellulose. Ligno-cellulosic fibres are the most abundant and renewable bio-based materials source in nature [8]. The bast fibres (those extracted from stem) are known to be the best fibres for reinforcement of polymeric composites as a result of their superb mechanical properties [9].

Table 1: Physicochemical Properties of selected Natural Fibres, Sources [3, 10 – 19]

Type of fibre	Origin	Bulk Density (kg/m ³)	Cellulose (%)	Lignin (%)	Hemicellulose (%)	Pectin (%)
Oil palm	fruit	1300	65	29	22.1	-
Jute	stem	1300	58 - 63	12 - 14	21 - 24	-
Hemp	stem	1480	57 - 77	3.7 - 13	14 - 22.4	0.9
Wood	stem	650	40 - 45	20 - 30	22	2 - 4
Flax	stem	1500	64.1	2.0	16.7	-

Bagasse	stem	1250	81	25.3	9.45	-
Sisal	leaf	1370	65	9.9	12	10
Coir	fruit	1200	32 - 34	40 - 45	0.15 – 0.25	-
kenaf	stem	1500	31 - 57	15 - 17	21.5 - 23	-
Ramie	bast	1510	68.6 - 91	0.6 – 0.7	5 – 16.7	1.9
Banana	stem	1350	50 - 56	21 - 31	25 - 30	-
Henquen	leaf	1300	77.6	131	3 - 8	-

3. FLAME RETARDANT COMPOUNDS

Polymers and natural fibres are organic in nature and are very sensitive to almost any properties if flame or fire is introduced to them. Therefore, to extend their use in industries and ensure safety in today's society, flame retardants are very important in order to accomplish safety measures while developing natural fibre composites. Flame-retardant systems are proposed to delay or to inhibit combustion and there exist a large amount of flame retardants on the globe [3, 20, 21]. Depending on their nature, flame retardants can act in the condensed or in the gas phase through a physical or chemical process [20 – 22].

- **Char-forming flame retardants:** Usually, ammonium polyphosphate (APP), pentaerythritol and melamine are the main ingredients of an IFR.
- **Endothermic flame retardants:** They undergo an endothermic decomposition in the range of temperatures at which combustion takes place. This endothermic reaction helps to withdraw heat from the substrate. The metal oxide formed during the decomposition of a metal hydroxide form an insulating protective coating on the condensed phase.

As a matter of fact, many flame retardant compounds can be formulated to delay or retard a flame. Flame retardants are categorized as reported by Atkinson et al [23] into two key categories, namely halogen free flame retardants (HFFRs) and halogen based flame retardants (HBFRs).

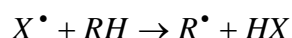
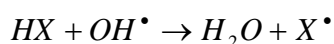
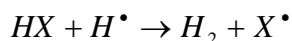
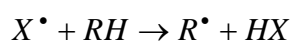
3.1 Halogen Free Flame Retardants (HFFRs)

The substances are utilized at levels below about 20% of the polymer resin in order to enhance efficiency. HFFRs are very essential materials preferable in the condensed phase of the burning process [24, 25]. The mode of action of HFFRs blended in composites is that they act as heat sink by releasing water vapour. The blanket coating formed eliminates the formed heat and cools the substrate to a temperature below that which is needed to maintain combustion. Their relative low cost, easy to handle, non-toxicity and good

anticorrosion properties makes them very attractive over other class of flame retardants. Besides, their drawbacks such as poor thermal stability, low efficiency for loading up to 50% from the mass of the material and a decrease in the strength properties in sustaining their applications has raised serious concern. However, studies by Hirschler [26], Babrauskas et al [27], Harris et al [28], have shown that HFFR provides formulations that meet suitable standard test for many uses.

3.2 Halogen Based Flame Retardants (HBFRs)

The halogen based flame retardants show their effectiveness in the increasing order as Fluorine < Chlorine < Bromine < Iodine. Chlorine and bromine-based Flame retardants are commonly used; whereas the fluorine and iodine based flame retardants are not used. The HBFRs removes the radicals. The flame retardant breaks down as shown below where X is either chlorine or bromine [29]



By reaction with HX the high energy H^{\bullet} and OH^{\bullet} radicals are removed and replaced with low energy X radicals. Thus, the hydrogen halide, HX produces the actual flame retardant effect. By reaction with hydrocarbon the HX consumed is regenerated. Thus, the hydrogen halide acts as a catalyst as shown above [29]. There are theoretically four classes of chemical compounds that can be used as HBFRs: those containing fluorine, chlorine, bromine, or iodine. The very low thermal stability coupled with the difficult in processing fluorine and iodine compounds with commercial polymers makes brominated and chlorinated halogen flame retardant a better choice and most diversified class of retardants for flame inhibition [30]. Bromine and chlorine incorporated into a polymer chain by copolymerization [31] forms a flame retardant additive that controls the flame temperature of a fire. Their effectiveness depends on the release of halogen in the form of radical or halogen halide at the same temperature range or below the decomposition temperature of the polymer. The major problem with halogen based compounds is the release of smoke, containing corrosive, acidic and toxic gases that are serious health and environmental threats [30, 31].

3.3 Intumescent Flame Retardants (IFRs)

The French verb *tumere* means “to swell”. The mode of action of intumescent flame retardants is the formation of a voluminous, insulating protective layer through carbonization and simultaneous foaming [32]. The basic components are:

1. Acid donors (e.g. ammonium polyphosphate).
2. Carbon donor (e.g. polyacohols such as pentaerythritol, starch).

3. Spumific compounds (e.g. melamine).

A typical IFR system is a combination of ammonium polyphosphate/pentaerythritol/melamine (APP/PER/MEL) mixture [33]. As APP acts, both as the acid source and blowing agent during combustion, MEL may be optional. The optimal weight ratio of APP/PER has been investigated by several researchers; the best flame retardant performance was observed when the APP to PER ratio was 2:1 in the system [34]. Nevertheless, there are shortcomings associated with the IFR systems with polymers, such as; poor compatibility, thermal stability, moisture resistance and poor flame retardant efficiency at low IFR concentrations consequently leading to considerable decrease in mechanical properties. These shortcomings need to be evaluated.

4. Laboratory Test Methods Used in Characterising Flammability Properties

There are several laboratory test methods used for characterising the flammability properties of composite materials and they are well established in these references [35- 38]. In fact, the 1998 edition of the collection of fire tests by the American Society for Testing and Materials (ASTM) enumerated some 77 tests [39].

4.1 Underwriters Laboratories standard UL-94 test

However, the most commonly used test method in the UL-94 standard is the 20 mm vertical burning test [V-0, V-1 or V-2]. The standard specifies this bench scale test method to determine the acceptability of plastic materials for use in appliances or other devices with respect to flammability under controlled laboratory conditions [66]. Rating can be obtained by testing five conditioned sample specimens and averaged according to the ASTM D 3801 standard test procedure. This categorization is intended to serve as a preliminary indication of their acceptability with respect to flammability for a particular application.

Table 2: Standard ratings for UL-94 vertical burning test [39, 40].

Rating	Description
UL-94 V-0	Flame must be out in 10 seconds or less.
UL-94 V-1	Flame must be out in 30 seconds or less. No glow beyond 60 seconds and no burning material can fall.

4.2 Cone Calorimetric Test (CCT)

The CC test is at present the most advanced method for assessing the reaction to fire. The test gives the possibility to evaluate ignitability, combustibility, smoke production and production of toxic gases. The testing procedure and measurement observations are described in both the ISO standard (ISO DIS 5660) and ASTM standards (ASTM E1354). The test is carried out by applying a constant external heat flux to

100 x 100 mm specimen under forced flaming condition. If specimen thickness is less than 50 mm additional layers of insulation used to fill out the depth and permit to hold the specimen securely in the holder [41]. This will allow the full surface of the test specimen to be exposed to a constant level of heat irradiance, within the range, 0-100 kW/m², from a conical heater. At the early stage of the test, the surface temperature will increase rapidly and level off to a steady state [40]. Heat transformation from the sample plate to the surrounding is equal to the input at its surface [42, 43].

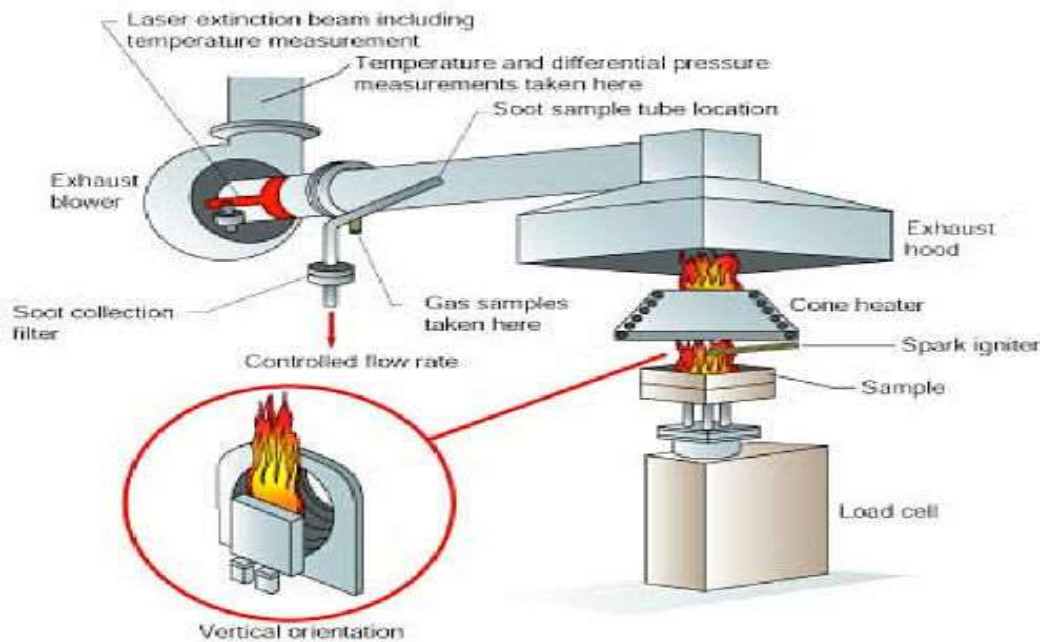


Figure 4 Schematic view of Cone calorimeter used for analyzing flammability properties [3]

4.3 Reaction to Flame Studies on Natural Fibre Composites in UL-94 and Cone Calorimeter

It is pertinent to note that research on reaction to flame/ fire of natural fibres started many decades ago when the Romans discovered flame retardant treatment of wood used in building ships and other structures, and it became extremely important to restrict the ignitability of the wood. Nevertheless, **Hill [44]** in 1887 carried out the first quantitative study on the ignition temperature of wood. After this, focus has been on the flammability of natural fibres, especially wood and other textile materials used in industries. Recently, researchers, scientists and engineers are delving into finding ways of improving the flame retardancy of natural fibre composites by adding flame retardants which will in effect expand their relevance. **Lucasz et al [45]** prepared and characterised an eco-friendly Mg(OH)₂ / lignin hybrid material used as a functional filler for poly vinyl chloride. A 10% increase in the oxygen index of Polyvinyl chloride composites upon addition of 10 wt % hybrid fillers has also been observed, which contributes to an extended range. **Shartel et al [46]** combined flax and polypropylene (PP) using Ammonium Polyphosphate (APP) and expandable graphene (EG) as retardants. They observed that APP based systems reduces the fire duration at light heat fluxes. Fire retardancy is higher with 25wt% EG over APP with 30wt. % flax. **Chen, et.al [47]** used aluminum salt of hypo phosphorous acid (AP) as flame retardant for

glass-fibre-reinforced poly (butylene terephthalate) via melt compounding and tested their flame retardance and combustion behavior using LOI, vertical burning UL-94 test and cone calorimetric test. They found AP as an effective flame retardant for the above composite, which showed V₀ classification, increased LOI of 32.5% and the char indication from XPS and FT-IR analysis. **Hsiao et al [48]** compounded rice husk/polypropylene composite filled with grapheme nanoplatelets and two kinds of metal hydroxide flame retardants, aluminum hydroxide (ATH) and magnesium hydroxide (MH) using a Brasbender Plasticoder. The flammability properties of natural fiber composites of different formulations were evaluated. The horizontal burning test results showed that plain 50 wt% PP/rice husk composites demonstrated a horizontal burning rate of 36.08 mm/min. On the other hand, a synergetic effect was observed when grapheme nanoplatelets were used in conjunction with aluminum hydroxide (ATH) or magnesium hydroxide (MH). Horizontal burning rates were significantly reduced. **Kim et al [49]** combined wool/ PP using APP as flame retardant. 20wt. % APP, 30wt. % wool composite achieved 53.7% pk-HRR reduction by indicating significant improvement in the fire retardancy. **Qiu et al. [50]** proposed the use of magnesium hydroxide in the form of Mg (OH)₂/vinyl acetate nanocomposite as a material with advantageous non-combustible properties. Based on the results, an increase in the oxygen number from 24.0 to 38.3 was noted for the material with nanometric refinement.

Table 3: Applications of Some Flame Retarded Natural Fibre Composites [51-52]

fibre	uses
Bagasse fibre	Window frame, panels, decking, railing systems, and fencing.
Stalk fibre	Building panel, furniture panel, bricks and constructing drains and pipelines.
Wood fibre	Window frame, door shutters, decking, panels, fencing, railing system.
Hemp fibre	Textiles, cordage, construction products, furniture, paper and packaging.
Oil palm fibre	Building materials such as door frames, windows, roofing, fencing, insulated panel building systems and other building materials
Ramie fibre	Industrial sewing, thread, fishing net, packing materials, filter cloths, upholstery, canvas.
Cotton fibre	Textile, furniture industry, cordage
Kenaf fibre	Mobile cases, packaging materials, bags, insulation, clothing grade –cloth, materials that absorb oil and liquid
Sisal fibre	In construction industry such as doors, panels, roofing and shutting plates, manufacturing of pulp and paper

5. CONCLUSIONS

The present review explores the flammability and thermal properties of some natural fibre reinforced polymer composites and their uses in industries. Natural fibre polymer composites have found wide applications in industries as a result of their intrinsic properties such as biodegradability, light weight and eco-friendliness. Irrespective of these good properties, the high vulnerability of natural fibres and polymers to flames has restricted their uses mainly under conditions where fire menace is impending. Current studies have shown that the incorporation of flame retardant additives in polymeric materials can lower the flammability properties and hinder the propagation of fire in the composite materials. Flame retardant fillers equally act as heat barrier, limiting fuel or flame and therefore guard the composite surface from heat and air. It was equally indicated that the heat release rate is the single most important property used in classifying a composite fire risk. Other flammability properties such as; time to ignition (TTI), total heat release (THR), specific extinction area (SEA), etc are correlated to the HRR. The review also pointed out that reduction of the materials to nano scale helps to improve the overall properties of the composite. We look forward to using bio-based retardants to know the synergistic effect on the composites.

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