



## Review

## Bio-based flame retardants: When nature meets fire protection

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## ABSTRACT

Fire retardancy of polymeric materials is a subject of major preoccupation due to the need to minimize fire risk and meet fire safety requirements. Numerous efficient conventional flame retardants based on halogen, mineral, and other compounds have been developed. However, some of these compounds, particularly halogen flame retardants, are harmful to our health and the environment, and their use has been restricted. In addition, the increasing concern about the reduction of the ecological footprint of materials, has encouraged the development of new plastics and additives made from renewable resources. As a result, renewed interest is emerging for the development of sustainable solutions for flame retardants for polymeric materials. This paper focuses on the identification of biomass compounds that have a potential as flame retardants for polymers due to their high availability and inherent properties. The last section of this paper explores the recent progress in flame retardant systems, based on the use of renewable products, which constitutes a promising approach to provide materials with improved fire resistance.

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**Abbreviations:** ABS, acrylonitrile butadiene styrene; Al-Phyt, aluminum phytate; APP, ammonium polyphosphate; B.C., before Christ; CH or CS, chitosan; CO, carbon monoxide; CO<sub>2</sub>, carbon dioxide; COT, cotton; CP, cellulose propionate; CuSO<sub>4</sub>, copper sulfate; DMF, dimethylformamide; DNA, deoxyribonucleic acid; Et<sub>3</sub>N, trimethylamine; EVA, ethylene vinyl acetate; Fe(NO<sub>3</sub>)<sub>3</sub>, iron nitrate; Fe-Phyt, iron phytate; GDP, gross domestic product; GPCS, glycidyl phosphorous chitosan; HCHO, formaldehyde; H<sub>2</sub>O, water; HSO<sub>3</sub><sup>-</sup>, hydrogen sulfite ion; IFR, intumescent flame retardant; KCl, potassium chloride; K<sub>2</sub>CO<sub>3</sub>, potassium carbonate; La-Phyt, lanthanum phytate; LbL, layer by layer; LDH, layered double hydroxide; LOI, limit oxygen index; MCC, microcrystalline cellulose; MCC-P, phosphorylated microcrystalline cellulose; MgCl<sub>2</sub>, magnesium chloride; MMT, montmorillonite; NaCl, sodium chloride; NaOH, sodium hydroxide; Na-Phyt, sodium phytate; Na<sub>2</sub>CO<sub>3</sub>, sodium carbonate; Na<sub>2</sub>S, sodium sulfide; NCC, nanocrystalline cellulose; NH<sub>4</sub>OH, ammonium hydroxide; NiCl<sub>2</sub>, nickel chloride; PA, phytic acid; PCL, phosphorylated cellulose; PEC, polyelectrolyte; PEI, polyethylenimine; PER, pentaerythritol; PET, polyethylene terephthalate; pHRR, peak of heat release rate; PLA, polylactide; PNFC, POSS-modified nanofibrillated cellulose; POCl<sub>3</sub>, phosphoryl chloride; POSS, polyoligosilsesquioxane; PP, polypropylene; PT, phosphorylated chitin; PU, polyurethane Tg glass transition temperature; TG-DTG, thermogravimetric-derivative thermogravimetric; THR, total heat released; TNT, titanate nanotubes; ZnCl<sub>2</sub>, zinc chloride.

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## 1. Introduction

Fire hazard is a constant risk in everyday life because of the massive use of combustible materials, such as fabrics, polymeric materials and wood. The increased use of systems to prevent fires or to stop them, such as smoke alarms, sprinklers and emergency exit plans, has enabled a significant reduction in the number of deaths by fire since the late 1900s. Even with these improvements, fire is still an important issue, which costs approximately 1% of global gross domestic product (GDP) per year, and causes several thousand deaths in Europe alone [1]. Moreover, in the second half of the 20th century, polymeric materials, which are easily combustible, became the most universally used materials. These multipurpose materials, which are now essential to modern life [2], have been important to the global economy for many years, and the plastic industry experienced yearly growth of 8.7% between 1950 and 2012. In order to minimize fire risk and meet fire safety requirements, solutions to prevent the ignition of the materials, or to lower the heat released during their combustion, have been developed and they act by interfering with the processes involved in polymer combustion using various physical or chemical strategies. The main categories of compounds for improving fire behavior are mineral, halogenated, phosphorus-containing, nitrogen-containing, silicon-containing, and nanometric compounds [3]. Mineral flame retardants, such as aluminum trihydroxide, are very efficient in decreasing fire hazard and they are widely used. However, a large amount must be incorporated to achieve the expected improved fire properties and these additives can cause material defects. Halogenated flame retardants, which have been used since the 1930s, represent a cost-effective technology, acting in the vapor phase by scavenging reactive free radicals by releasing halogen radicals that inhibit combustion. However, due to environmental and health concerns, the use of halogenated flame retardants has been increasingly avoided and restricted [4]. Some of these products have been proven to be linked to cancer, reproductive problems and impaired fetal brain development. Moreover, during combustion, they induce an increase in smoke and corrosive gases, and they are persistent environment pollutants if they migrate out of the polymer matrix. Three halogenated products are currently forbidden: penta- and octa-bromodiphenyl ether and hexabromocyclododecane. To address these problems, considerable attention must be devoted to the development of halogen-free flame retardant additives. Flame retardant systems that promote the formation of an insulating char layer on the surface of the burning sample are among the most promising environmentally friendly strategies for replacing halogenated flame retardants. They have indeed proven to be efficient in improving the fire behavior of many polymer matrices, while reducing the amount of smoke released as a result of their condensed phase action. Today, new solutions of such flame retardants, based on renewable resources, are attracting great interest, due to their availability and the growing awareness of environmental issues linked to the increased use of fossil feedstock. Furthermore, the development of flame retardant systems from renewable resources is suitable for supporting the increasingly use of bio-based polymers in many technical fields, maintaining thus their sustainability and preserving their good environmental impact.

Moreover, the world demand in energy for the transport sector was approximately 84 million barrels per day in 2010 and it was estimated to increase to about 116 million barrels per day by 2030 [5]. The Intergovernmental Panel on Climate Change has highlighted in a report that this growing demand is leading to a rapid increase in greenhouse gas emissions [6]. To reduce CO<sub>2</sub> gas emissions while meeting the increased energy demand, the utilization of renewable instead of fossil resources seems to be a feasible option. Renewable carbon sources enable industrial processes to be nearly CO<sub>2</sub> neutral since bio-based products release no more CO<sub>2</sub> at the end of their life than was originally metabolized in the biological production of the raw material. During photosynthesis, plants use sunlight to convert water and carbon dioxide from the atmosphere into carbohydrates [7]. While in the case of fossil-based products, the CO<sub>2</sub> released at the end of their life was geologically sequestered for millennia, which has been demonstrated to have an impact on the environment [8].

For several decades, the biorefinery has appeared to be an efficient approach for the transformation of biomass components into biofuels, bio-energy (heat and power) and bio-based chemical products and materials, in the same way as a petroleum refinery does for fossil based resources. The difference between the petrochemical industry and a biorefinery lies in the highly oxygen-functionalized bio-based feedstock in comparison to the hydro-carbon based fossil resources [9]. Biorefineries are facilities that convert biomass, i.e. biological materials from living or recently living organisms, into bio-based products. A biorefinery can use a wide range of different feedstocks as input and various process technologies to convert them. The input products of the first generation of biorefineries are mainly edible biomass products, such as sugar-rich, starch-rich and oily plants, whose can be controversial because of the need to use these resources for food. However, the second-generation of biorefineries overcome this problem by treating residual non-food parts of crops and other non-food sources, such as grasses and algae. The inedible lignocellulosic materials which are found in plants and composed of cellulose, lignin and hemicelluloses represent a promising feedstock to convert into useful bio-based products in biorefineries. The wide development of these biorefineries made biomass components more and more easily available for many applications. Furthermore, the economic viability of biorefineries requires the use of some of their production for high value applications such as flame retardant additives [8].

This paper, which aims to present recent developments in the valorization of biomass as flame retardant additives for polymeric materials, provides a presentation of the main biomass components able to generate char during their thermal decomposition and the recent developments of bio-based flame retardant additives.

## 2. Toward the development of bio-based flame retardants

Fire retardant additives have high-value applications for which some biomass constituents are predisposed. In nature, certain lignocellulosic plants have developed defense behaviors against fire aggression. This is the case of cork oak that is more fire-resistant than other trees owing to the slow combustion of cork, containing mainly suberin (waxy substance composed of

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