Flame retardants, their beginning, types, and environmental impact: a review

Ali I. AL-MOSAWI

PhD in polymers Engineering at Institute of Ceramic and Polymer Engineering, Faculty of Materials Science and Engineering, University of Miskolc, Hungary. Research Interests: Polymers, Composite Materials, Rubber Technology, Flame Retardants, Materials Testing, Materials Processing.

Ali I. AL-MOSAWI ▪ Institute of Ceramic and Polymer Engineering, University of Miskolc, Hungary
▪ aliibrahim76@yahoo.com, alialmosawi76@gmail.com

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Abstract

Flame retardants have made the world safer by significantly reducing fire risks and reducing human and material losses. At the same time, however, environmental problems have been created and their impact on health, as the compounds that make up the flame retardant are heavy metals. Therefore, in order to make these materials safer, efforts must be focused on using environmentally friendly ones. Furthermore, this green approach must be in line with the laws and legislation developed by many countries due to the recommendations of environmental institutions that warned of the dangers of continuing to use flame retardants containing heavy metals. Not only that but all government and private institutions and civil facilities must be obliged to use flame retardants as a primary building material. This review will focus on the most critical developments that flame retardants have experienced and their impact on the environment and health through their types and mechanism of action.

Keywords: flame retardants, historical review, environmental considerations

Kulcsszavak: égésgátlók, történeti áttekintés, környezetvédelmi megfontolások

1. Historical overview of flame retardants

Flame retardants are defined as chemical agents that can withstand direct flame by stopping flame entry into the material, controlling its spread, and even extinguishing utterly. These materials can be added during or after the products' fabrication for burning protection. The development of flame retardants allowed the safe usage of fabrics that cause flammable behaviour by reducing flammability and reduce the rate of burning. The compounds of phosphorus, nitrogen, chlorine, bromine, boron, and antimony are ones of the most widely used for flame retarding [1-9]. Many materials with flame retardant properties have been known for many centuries. The earliest use of flame retardants was by the eastern civilizations, specifically by the Egyptians and Chinese. Where about 3000 years ago, the ancient Egyptians were soaking grass and reed in seawater before they used it for roofing, so when these grass and reed dry, the mineral salts will crystallize and act as a fire retardant. The Egyptians and Chinese also used alum and vinegar to paint timbers in temples to protect them from the fire [10-13]. The first attempts to use flame retardants by Western civilizations were by the army of the Roman Empire when they besieged the city of Piraeus about 87-86 B.C. The Roman army painted siege towers made of wood with a mixture of alum, vinegar, and clay to protect it from burning by the fire of city defenders. The Roman flame retardant mixture was developed from what the ancient Egyptians used.

Although the ancient civilizations did not have the necessary equipment to analyze these materials' components, they pay attention to the nature of these substances acting as retardants. The discovery of the flame retardancy properties of these materials was through coincidence. Still, they inevitably conducted some practical experiments, even if primitive, to prove these materials' effectiveness in fire protection,

in addition to searching for other materials with the same properties. Indeed, this led to the development of testing devices to detect these materials [14].

Material	Used by	Date
Potassium Aluminium Sulphate (Alum)	The Egyptians have used Alum and Vinegar to reduce wood flammability	450 B.C.
Vinegar	The Egyptians and Chinese used painted timbers by vinegar to increase their resistance to burning	360 B.C.
Alum, Vinegar, and clay	The Roman army painted the wooden siege towers with alum, vinegar, and clay to protect them from burning when they besieged Piraeus	87-86 B.C.
Clay-Gypsum Mixture	Had been used by Nicola Sabbatini in France for reducing painted canvas inflammation at Parisian theatres	1638
Alum- Ferrous Sulfate-Borax Mixture	It was discovered by Obadiah Wyld and used in Britain for preventing paper, linen, canvas, & c., from flaming or retaining fire, & c. Wyld received the first patent for fire retardants No.551	1735
Potassium Aluminium Sulphate (Alum)	Used by the French brothers Montgolfier to reduce flammability of hot air balloons	1783
(NH₄)₃PO₄ - NH₄Cl - Borax Mixture	Used by the French chemist Gay-Lussac for linen and hemp fabrics	1821

Table 1 A historical overview for early use of flame retardants [11-20]
1. táblázat Történeti áttekintés az égésgátlók korai használatáról [11-20]

With the development of human lifestyle, the needs to discover new materials that reduce the risk of fires have increased. This calls for relying on a scientific and practical basis and not to depend on chance only. So the first practical experiments to reduce fire risk began to appear. As a scientific

procedure recorded, the first fire testing experiments can be traced to London's 1790s done by the Associated Architects. There was Quantitative work in Germany beginning in the 1880s, and in the U.S. and England in the 1890s. A summary of the timeline for the use of flame retardants is included in *Table 1*. The growing interest in these tests led to establishing unified test standards in the early twentieth century. Where the first standard about fire testing methods was ASTM C19 (later called E 199) [14-18].

2. Categorization of flame retardants

Flame retardants are typically categorized into four major categories: Inorganic FRs; Phosphorus-containing FRs; Nitrogen-containing FRs; and Halogenated organic.

2.1 Inorganic flame retardants

The most significant types of this category are antimony trioxide containing halogen, magnesium hydroxide, zinc borate, aluminium hydroxide, and zinc sulfide. If this category of flame retardant is exposed to heat, it does not evaporate. Instead, it decomposes and released non-flammable gases, such as water vapour, carbon dioxide, sulfur dioxide, hydrogen chloride, and other gases. Most of these compounds produce endothermic reactions. These flame retardants' mechanism of action depends on its disintegration at high temperatures and causing the release of non-flammable gases, which minimize the mixture of flammable gases. Then, it separates the plastic's surface by forming a glassy protective layer on the surface of the plastic that prevents oxygen and heat access [21-27].

2.2 Phosphorus containing flame retardants

This category typically includes phosphate esters, red phosphorus, ammonium polyphosphates, and ammonium orthophosphates. Its principle of action is based on its oxidation during combustion to phosphorus oxide, which converted into phosphoric acid when interacting with water. This acid induces the release of water and removes it from the substrate layer of the thermal decomposing material, which contributes to its decomposition and thereby increases the formation of carbonaceous waste and decreases the emission of combustible gases [28-31].

2.3 Nitrogen containing flame retardants

Nitrogen-containing flame retardants: Often referred to as organic flame retardants. Melamine and its derivatives are the essential compounds in this category. These retardants' principle of action depends on the formation of an insulating surface layer by gases released from these retardants during combustion cause the material to swell [32-37].

2.4 Halogenated organic flame retardants

This category contains primary bromine and chlorine in particular. Its principle working action depends on chemical interference with the root chain mechanism, which occurs in the gaseous phase during combustion. Halogen flame retardants eliminate the high-energy hydrogen and hydroxide

generated during the combustion process by combining them, so the flame is poisoned by the halogen radicals released during combustion [21,38-41].

3. Classification of flame retardants

Flame retardants can be classified into three primary groups, depending on how they are applied to the polymer, and these two groups are: Additive FRs; Reactive FRs.; and Additive-Reactive FRs

3.1 Additive flame retardants

This type is used for thermoplastics in particular. Additive flame retardants can be added to plastics in three ways: (1) before polymerization, (2) during the process, or (3) after polymerization, and it is the most common way to add them. In addition to its direct action as flame retardants, it can be a plasticizer if they are compatible with the plastic or as fillers if they aren't compatible with it. The main disadvantage of these retardants, is their instability, where they sometimes can be volatile or even bled from the plastic, especially from the layers near the surface, causing plastic to lose its ability to resist burning, which is very dangerous because this state can only be realized when combustion occurs [42-44].

3.2 Reactive flame retardants

This type of flame retardant is binding to the polymer molecule with the other components. This chemical bonding prevents it from evaporating and bleeding from the polymer over time. It happens with additive flame retardants, enabling the polymer to maintain flame retardant properties as long as possible. Although they are chemically binding to the polymer molecules, they have no plasticizing effect and are non-affecting the polymer's thermal stability. The practical and widespread use of reactive flame retardants is with thermosets [41-44].

3.3 Additive- Reactive flame retardants

This type of flame retardants is referred to also as synergism. Several materials are not classified as stand-alone flame retardants, which will often work as fillers if used alone. Still, they have a special synergistic effect if they are added to other retardants, where the effectiveness and efficiency of these retardants increase to resist fire. An example of such materials is antimony compounds, especially antimony trioxide. The main purpose of this combination is to reduce the cost and, as mentioned, increase the effectiveness of the primary flame retardant [21, 42, 43, 42, 45, 46].

4. Working mechanism of flame retardants

The method of slowing or even stopping the flame depends on the nature of the flame retardant, as the flame retardant can act chemically or physically in the solid, liquid, and gas state, where the flame retardant is interpenetrating with the combustion process during the various stages of this process, i.e., heating, decomposition, ignition, or flame propagation [21,47,48]. In general, there are two standard methods of flame retardation. The first method is based on preventing oxygen

from accessing the flame area by generating non-combustible gases, where these gases can poison and extinguish the flame with free radicals. The second method is based on the thermal flame theory, which states that flame retardants require thermal energy for decomposition, leading to a decrease in the material's surface temperature to a temperature lower than its burning point, and the combustion breaks down [49].

4.1. Physical mode

This working mode can be achieved in three ways [47,50,51]:

4.1.1 Cooling

Additives cause endothermic processes that cool the substrate of the material to a temperature lower than the level of combustion. An example of flame retardants behaving like this is aluminum hydroxide. The cooling mechanism is shown in Fig 1.

Fig. 1 Cooling mechanism

1. ábra Hűtési mechanizmus

4.1.2 Formation of insulation layer

The insulating semi-glass layer is created by a flame retardant that expels the oxygen needed to begin the combustion process, preventing heat transfer. Inorganic phosphorous compounds and boron are behaving like this mode.

4.1.3 Dilution

The incorporation of inert materials (fillers) and additives that release inert gases after their thermal decomposition dilute the fuel in both the solid and gaseous states in such a way that the minimum ignition limits for the gas mixture are not exceeded. Phosphorous and boron compounds work this way.

4.2 Chemical mode

Can be achieved in two ways:

4.2.1 Gaseous phase reaction

The free radical mechanism is stopped by the flame-retardant materials, causing eliminating the exothermic processes and, so the system will cool down. The percentage of flammable gases is reduced and even wholly prohibited as shown in Fig 2. The flame retardant which exhibits this behavior is halogen compounds [21,51].

Fig. 2 Gaseous phase reaction

2. ábra Gáz-halmazállapotú fázisreakció

4.2.2 Solid Phase Reaction

A carbon film is formed on the surface of the polymer by the dehydrating action of flame retardant, forming double bonds in the polymer (see Fig 3). These double bonds will create a carbon film by cross-linking. An example of these retardants is phosphorous compounds. In the case of phosphorous compounds addition, their mode of action depends on substituting the hydroxyl and hydrogen radicals in the combustion cycle by low-potency radicals, eliminating their damage as shown in the equations below, which represents the combustion reaction (Chain reaction equations), and the hinderation reaction (Chain hinderation equations). This mechanism is similar to that of the halogen compounds. The change in the combustion cycle's radical composition in the gaseous phase leads to flame suppression and reduced heat production, which cools the combustion zone. The combustion process activated several sequential reactions, which include: chemical chain-branching, chain-propagating, and chain-breaking reactions. These chemical processes help preserve the flame by changing the quantity, form, and mole ratio of the radicals present in the gas phase. Therefore, in order to change this state, Lower energy radicals are required to remove the unstable radicals of OH and H. Replacing the unstable OH and H radicals with less reactive and more stable radicals works to inhibit the combustion chain reaction and lead to cut this reaction. Thus the self-extinguishing process of the system will occur [21,52-54].

Fig. 3 Solid phase reaction
3. ábra Szilárd fázis reakció

Chain reaction equations [54]:

Chain hinderation equations [54]:

Polymer	Flash-ignition Temperature, °C	Self-ignition temperature, °C
Poly(methyl methacrylate)	300	430
Polypropylene	320	350
Polyethylene	340	350
Polystyrene	350	490
Rigid Poly(vinyl chloride)	390	450
Plasticized Poly(vinyl chloride) (Insulation)	330	385
Plasticized Poly(vinyl chloride) (Fire resistant, low acid emission)	400	410
Poly(acrylonitrile)	480	560
Polyamide 6'6	490	530
Poly(tetrafluoroethylene)	560	580

Table 2 Flash and self ignition temperatures for various polymers [58,59]
2. táblázat Vaku- és öngyulladási hőmérséklet különböző polimerekhez [58,59]

5. Flammability and combustibility

Many people think that flammability or ignitability and combustibility is similar in denoting the same characteristic. Still, the truth is that they are different, so I will explain what these two terms refer to in this paragraph. The flammability or ignitability is defined as a material's ability to catch fire (ignite),

causing a fire, combustion, or even an explosion if the material is unstable. Whereas combustibility is defined as how easily a material burns to cause a fire or combustion [55]. The main criterion by which materials are classified as flammable or combustible is the flash point, where flammable materials have a lower flash point than combustible materials. The flash point of flammable materials is less than 37.8 °C, while in the case of combustible materials; its value is higher than 37.8 °C and less than 93.3 °C [55-58]. Table 2 represents the flash-ignition and self-ignition temperatures for various polymers.

6. Flame retardants and the environment

Most of conventional flame retardants and stabilizers for plastics contain heavy metals or other ingredients e.g., bromine being potentially harmful to the environment or health. For example, in European countries, concentrations of flame retardants have been found in human milk and the bodies of birth cohorts (but their concentrations were lower than in the United States) and household dust. Concentrations of flame retardants were also found in the bodies of birds and their eggs [60-69]. Therefore, in light of these environmental challenges and responsibilities, most of these flame retardants must be reconsidered in terms of use and finding an environmentally friendly and sustainable alternative. Especially since countries have already started issuing laws prohibiting the use of certain types of flame retardants due to their severe damage to the environment after research has proven this [70-72]. Many of flame retardants currently in use are included in European Union regulation (EC) No. 1272/2008 and its amendments for materials classification, which have been classified as dangerous materials. So it has become imperative to search for safe alternatives to these materials [73]. There are heavy metal-free flame retardants (earth metal hydroxides), but it must be used in high concentrations (40- 60 phr), which in turn will reduce the mechanical properties and wear resistance of the plastic [60].

Therefore, there must be a precise harmony between environmentally friendly flame retardants and preserving the materials' properties. This matter requires more time and research for a complete shift from traditional flame retardants to those environmentally friendly [7, 60, 73-75]. Certain recent flame retardants are now available to comply with the successful flammability tests regulations. Also, the interference of retardants with flame reaction chains will restrict the oxidation of the hydrocarbon. This interference will prevent the process of converting carbon monoxide to carbon dioxide, which causes highly volatile, very smoky fire effluents and rich in incomplete combustion products [76]. With the tendency to replace traditional flame retardants with environmentally friendly ones, the environmental conditions surrounding the product containing this type of retardants must be taken into account and the applications for which it is used. One study revealed that environmentally friendly retardants could be harmful when breaking down by heat and ultraviolet rays [77]. Therefore, it is necessary to use these retardants carefully. Obtaining the ideal flame retardant requires great research efforts that may not take as long as research in the past due

to scientific research development. Whereas the chemical stability of environmentally friendly flame retardants in different conditions is of critical importance to increase safety level when using these retardants.

7. Conclusions

Every year, flame retardants save thousands of lives around the world. Still, unfortunately, many also lose their lives because many of these retardants are made from harmful components to both health and the environment. Many legislations and laws have restricted the use of flame retardants containing heavy metals, which will make the heavy metals-free ones (green flame retardants) commonly used. To obtain the maximum effectiveness of flame retardants, each type must be used with the appropriate material intended to improve its flame resistance.

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