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# Recent development of metal-organic frameworks as a novel flame-retardant in polymeric applications

A Elashker<sup>1</sup>, A M Eldakhakhny<sup>2</sup>, and M Mokhtar<sup>1</sup>

<sup>1</sup> Chemical Engineering Department, Military Technical College, Cairo, Egypt

<sup>2</sup> School of Chemical Engineering, Military Technical College, Cairo, Egypt

[ahmedmohamed98769876@gmail.com](mailto:ahmedmohamed98769876@gmail.com)

**Abstract.** The increasing concern regarding hazardous fires related to polymeric materials has prompted numerous research into eco-friendly flame retardants. In recent years metal-organic Frameworks (MOFs) have emerged as promising flame-retardant materials, characterized by metal-containing units linked to organic linkers to create strong and porous crystalline frameworks. The organic ligands make MOFs possess significant surface area and adsorption properties desirable for different applications including flame retardancy. This paper gathers many of the recently published to discuss the use performance of polymer composites with MOFs for flame retardancy in comparison with polymeric composite materials without MOFs. Besides, a comparison based on mechanical, physical, and thermal properties shows the effect of MOF on the total effectiveness of the polymeric composite materials. This paper is of great interest to both the readers involved in the sphere of heat-resistance materials, encompassing contemporary advanced composite materials used for dampening hazardous fires. Furthermore, prospective trends, including the integration of innovative fillers such as MOFs in rubber composites such as NBR (Nitrile Butadiene Rubber), EPDM (Ethylene Propylene Diene Monomer), and silicone rubber, are deliberated upon.

**Keywords:** Polypropylene, Polystyrene, Rigid polyurethane foam, Epoxy, Flexible polyurethane foam, Metal-organic frameworks, MOF, NBR, EPDM, and TPU.

## 1. Introduction

Recently, people have been paying more attention to the fire risks linked with certain plastic materials. Because of this concern, there has been a rise in the development of environmentally friendly flame retardants to address this issue [1]. MOF has become more popular over the last decade, especially in the field of fire safety. They are being seen as new and promising flame retardant materials, gaining attention from fire safety associations in recent years [2]. These fascinating substances are created through the linking of metal clusters or ions to organic linkers. They form strong bonds to fabricate open crystalline matrices featuring enduring holes or gaps [3]. Thanks to the organic linkers, MOFs have a unique porous framework, (MOFs) that owns a substantial surface area, excellent crystallinity, low density, structural versatility, and precisely defined geometric arrangement. These attributes, along with their large surface area, high structural flexibility, and robustness, render MOFs highly suitable for diverse applications. These applications include gas separation and storage, organic molecule



adsorption, luminescence, nanomaterial carriers, polymerization, heterogeneous catalysis, chemical sensing, membrane fabrication, and fire-retardant additives [4].

Additionally, the organic component of MOF improves the compatibility properties between polymers and additives without needing additional organic alterations. This method is efficient and saves time in comparison to alternative methods involving inorganic flame retardants. Typically, the absence of compatibility between polymers and inorganic a flame retardant can adversely affect the mechanical efficiency of the composite material. For example, poor compatibility may lead to weak bonding within the polymer structure and the filler, resulting in decreased strength, stiffness, or toughness of the composite. Additionally, it can cause areas of stress concentration or uneven distribution of the additives inside the polymer structure, leading to defects and ultimately reducing the general mechanical performance of the composite material. Therefore, the organic linker of MOFs facilitates favorable compatibility between the filler and the polymer structure is essential for maintaining and improving mechanical performance in composite materials [5].

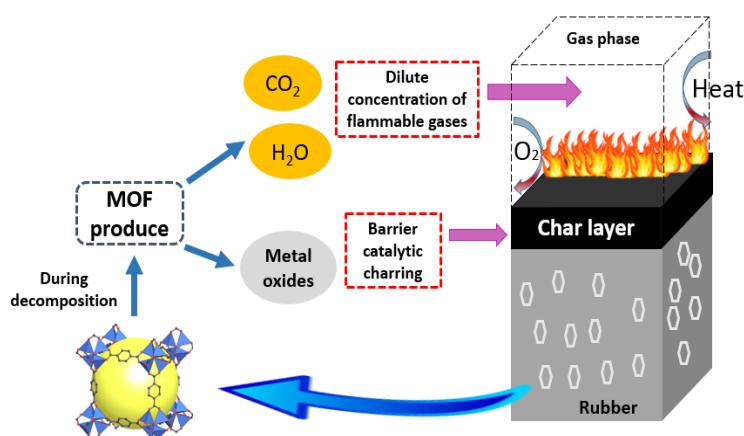
In this regard, the performance of MOFs does not only depend on the mechanical properties but both components of MOFs can serve as flame retardants. The metal compounds of MOFs serve as the inorganic assistants and their catalytic activity can reduce smoke emission and facilitate the formation of char layers. Meanwhile, the ligands in MOFs will act as the organic part and provide great compatibility and also contain flame-retardant elements or functional groups, such as nitrogen, aromatic derivatives, and phosphorus. The approach in which the organic and inorganic components are combined in the manufacturing of hybrid flame retardants has paved the way for the development of hybrid nanocomposites. Here, nanocomposites utilize the advantages of both constituents and at the same time, they reduce their weaknesses. MOFs fulfill this role of hybrid nanocomposites incorporating properties that are superior to those of traditional flame retardants [4].

MOFs can be classified into three generations. First-generation coordination polymers using  $d^{10}$  configuration metal ions like copper(I), faced issues such as non-porous dense structures and collapse upon guest molecule removal due to weak metal-ligand bonds. To address this, second-generation porous coordination polymers (PCPs) were developed with stronger metal-ligand interactions, using carboxylate-based coordination chemistry and aromatic organic linkers. These second-generation PCPs have stable porous frameworks with higher porosity and specific surface area suitable for guest molecule adsorption and desorption. Third-generation PCPs were later introduced, distinguished by their ability to undergo reversible structural changes upon guest exchange or removal. These Third-generation PCPs feature cooperative, reversible crystal-to-crystal transitions that greatly affect their porosity. Known as MOFs and classified as soft porous crystals, they offer structural flexibility and consistent porosity, enhancing their functional properties. These advancements in MOFs offer significant potential for advancing current fire safety measures as a flame retardant [6].

This article explains how MOFs work as flame retardants when incorporated with polymeric materials. Besides, evaluating the effectiveness of polymers combined with MOFs, and recent studies on improving polymeric material performance. It also explains a new trend in which MOFs are used in conjunction with different polymeric materials such as silicone rubber, EPDM, and NBR.

## 2. Mechanism of MOFs as a flame-retardant

The mechanism of MOFs as a flame retardant is demonstrated in figure 1. Initially, MOFs were observed as releasing, not flammable gases such as water vapor ( $H_2O$ ), and carbon dioxide ( $CO_2$ ). The non-flammable gasses mix with flammable gas making its concentration low around it, thus the possibility of ignition or combustion is reduced. Then MOFs could form char to act as a barrier, blocking the spread of flames, flames, and combustion products during a fire. finally, the metallic species serve as catalysts inside MOFs for both the production of char and the decomposition of flammable gases. This catalytic action promotes the stabilization of the char layer. Also, the presence of metal oxides within MOFs enhances the absorption of heat, resulting in a reduction in flame temperature and the temperature of the surrounding environment, thereby effectively limiting the propagation of fire [7].



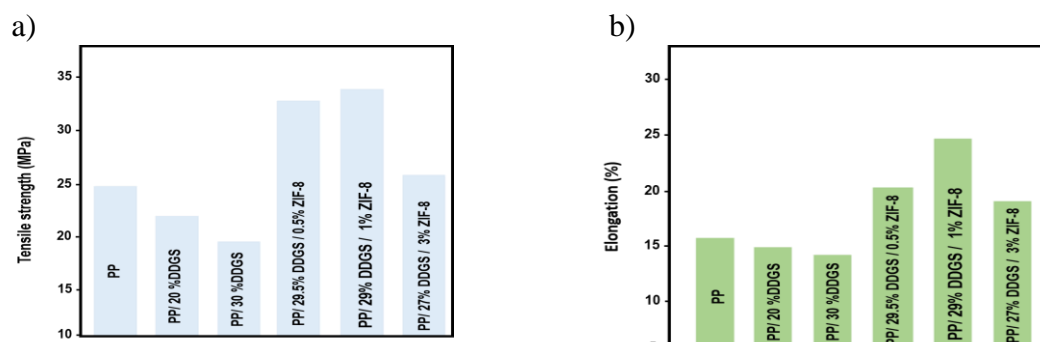
**Figure 1.** A schematic representation of the flame-retardant mechanisms employed by MOFs.

### 3. Polymeric materials utilized in flame retardant polymer–MOF composites

The researchers conducted extensive research on polymeric materials for a variety of applications, including polypropylene (PP), polystyrene (PS), rigid polyurethane foam (RPUF), epoxy (EP), flexible polyurethane foam (FPUF), and thermoplastic polyurethane (TPU). However, these materials have demonstrated a relatively high flammability, which causes a substantial amount of smoke to be produced during combustion. The incorporation of MOFs has been tested as a means of reducing the burning ability of these polymer materials.

#### 3.1. Polypropylene-MOF composites

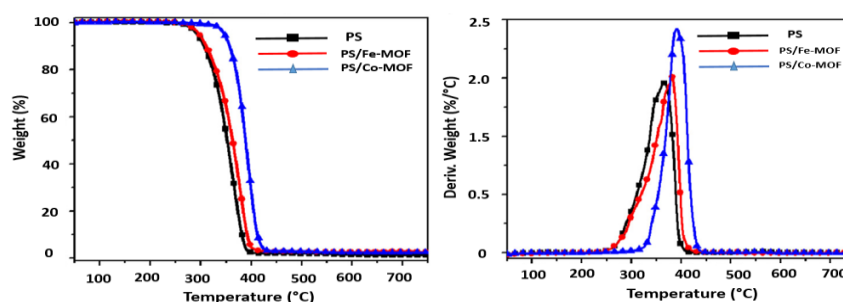
Polypropylene (PP) is widely used in everyday life because it is both durable and affordable. However, its structure makes it easy to catch fire. By adding MOF, we can make PP much better at resisting flames. For instance, Xie et al. [8] tried a method where they combined dried distiller grains with solubles (DDGS) with nano ZIF-8 to make polypropylene (PP) more flame retardant. Without any additives, PP burns easily, with 17.5% from a low Limiting Oxygen Index (LOI). But when they mixed in DDGS, the PP/DDGS mixture showed some resistance to burning because DDGS forms a protective layer of char. By keeping the flame-retardant mix at 30% of the total weight, they found that replacing some of the DDGS with nano ZIF-8 decreased how fast the fire spread horizontally and increased the LOI. Specifically, PP mixtures with 0.5%, 1.0%, and 3.0% ZIF-8 had LOI values of 20.0%, 22.5%, and 25.0%, and burned horizontally at speeds of 25.3 mm/min, 19.9 mm/min, and 14.5 mm/min, respectively. Moreover, the integration of ZIF-8 contributed to an enhancement of the mechanical properties of Polypropylene composite materials, surpassing those of PP pure and composites containing DDGS alone, as shown in figure 2.



**Figure 2.** The mechanical properties of PP compositions: tensile strength (a), elongation (b) [8].

### 3.2. Polystyrene-MOF composites

Polystyrene (PS), a widely used thermoplastic polymer renowned for its mechanical properties, low density, thermal resistance, and processability, faces limitations due to its high flammability, resulting in significant smoke and noxious gas emissions. Hou et al.[9] produced nickel-based MOF supported by graphene oxide (MOF-74(Ni)/GO) and investigated the burning properties of composites (PS/ MOF-74(Ni)/GO) with it as the modifier. After only 1.0 wt% of additives was implemented, there were significant effects such as a 33.0% decrease of PHRR and a decrease in total CO by 52.3% and smoke by 21% compared to neat PS. In a different investigation, Hou et al. [10] produced cobalt and iron-based MOFs, and combined them with PS. Thermogravimetric analysis (TGA) revealed that the thermal stability of Polystyrene enhanced on MOF loading with an impressive char yield in PS/MOF composites as shown in figure 3. MIL-88(Fe)/PS and ZIF-67(Co)/PS composites that have 2.0 wt% MIL-88(Fe) and ZIF-67(Co) incorporation exhibited significant reductions in total heat release (THR) values (4.0% and (17.6%) less than neat PS and the peak of heat release rate (PHRR) (14.4% and 28.0 %, respectively). Additionally, total CO production decreased by approximately 30% and 40% in MIL-88(Fe)/PS and ZIF-67(Co)/PS composites, attributed to both absorption and catalytic effects, enhancing safety in emergency situations.



**Figure 3.** TGA and the derivative thermogravimetric analysis (DTG) curves for PS with MOF compositions [10].

### 3.3. Polyurethane foams-MOF composites

Rigid polyurethane foam (RPUF), prized for its porous structure and exceptional thermal insulation properties, finds extensive use in packaging and insulation. However, its elevated porosity and aliphatic constituents render it significantly flammable, leading to rapid ignition and substantial heat release. To address this issue, Xu et al.[11] developed a tripartite hybrid, the diatomite-modified composite denoted as ZIF-8@melamine (ZMD), integrated into RPUF to improve its fire resistance. Compared to pure RPUF, the composite materials that included 10 wt% (ZIF-8@melamine) and (ZIF-8) showed reductions in PHRR by 23.8% and 16.6%, respectively. The modification of (ZIF-8@melamine) with diatomite further increased the PHRR reduced to 50.1%, surpassing the heterogeneous admixture of (ZIF-8), melamine, and diatomite. Cheng et al. [12] investigated the effect of (ZIF-11), (ZIF-8), and (ZIF-7) on RPUF fire hazard, revealing significant reductions in PHRR values for composites containing 12 wt% of each MOF. The superior flame retardant effect of (ZIF-8) was attributed to earlier insulating barrier formation by zinc oxide resulting from its degradation.

Flexible polyurethane foam (FPUF), has garnered extensive commercial utilization in bedding, carpet, furniture, packaging, support materials, and motor vehicles sectors. Due to FPUF containing a high concentration of oxygen, carbon, and hydrogen, it is significantly susceptible to combustion and emits hazardous gases and smoke. Hou et al.[13] introduced a novel approach involving treatment FPUF with (3-mercaptopropyl) trimethoxysilane (MTS) followed by accumulation of (MOFs-NH<sub>2</sub>) on the frameworks FPUF via in situ synthesis. In contrast to FPUF pure, (FPUF/MTS/MOFs-NH<sub>2</sub>) exhibited remarkable reductions in PHRR and THR, alongside the suppressed release of toxic carbon monoxide. Additionally, (FPUF/MTS/MOFs-NH<sub>2</sub>) demonstrated strong superhydrophobic properties rendering it well-suited for efficient oil absorption. Another investigation coated FPUF with (ZIF-8) through an in

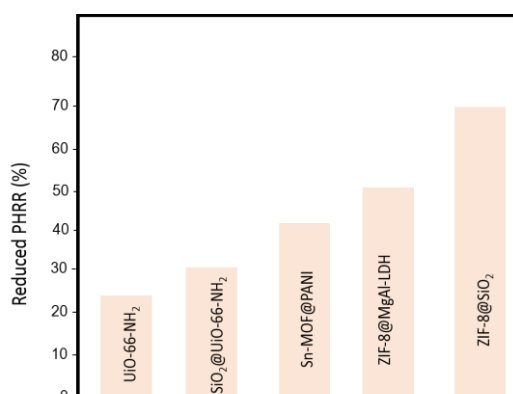


situ self-assembly method, resulting in enhanced flame retardant traits and reduced PHRR and smoke production rate (SPR) [14].

### 3.4. Epoxy-MOF composites

EP finds extensive applications in petrochemicals, electronics, transportation, and construction due to its thermosetting nature and favorable mechanical properties. However, its high flammability poses a significant challenge. Investigations into flame-retardant EPs have predominantly focused on ZIF-based MOFs. Xu et al. [15] functionalized reduced graphene oxide (RGO) with (ZIF-67) with borate ions adsorbed onto zeolitic imidazolate framework-67 (ZIF-67/RGO-B), resulting in a notable reduction of 65.1% and 41.1% in (PHRR) and (THR), respectively, with 2 wt% incorporation. The (LOI) value also increased from (21.4%) to (26.4%). Similar advancements were observed by incorporating ZIF-8@SiO<sub>2</sub> hybrids, leading to increased char yield and improved flame-retardant performance in terms of LOI, vertical combustion test rating, and reductions in THR, PHRR, and smoke production rate (SPR). Li et al. [16] investigated ZIF-8, MgAl-layered double hydroxide (LDH), and hybrids thereof, ZIF-8@MgAl-LDH in EP applications, observing substantial reductions in PHRR (51%) and THR (30%) with 2 wt% of (ZIF-8@MgAl-LDH). The charring role of (ZIF-8@MgAl-LDH) contributed to enhanced flame retardancy compared to individual MgAl-LDH.

Ma et al. [17] produced a network hybrid of MOF and polyaniline, denoted as Sn-MOF@PANI, achieving a 42% reduction in PHRR and a 32% reduction in THR with 2 wt% incorporation. The hybrid also enhanced storage modulus and impact strength. Zheng et al. [18] conducted a comparative study on different MOFs, observing significant reductions in THR, PHRR, and SPR values with 2 wt% incorporation of MIL-88(Fe), ZIF-8(Zn), and ZIF-67(Co). ZIF-67(Co) exhibited superior smoke suppression and flame retardancy due to increased absorption and catalytic oxidation. Notably, ZIF-8 exhibits superior flame-retardant efficiency over (UiO-66-NH<sub>2</sub>) within the epoxy system, and hybrids based-MOF, particularly those involving phosphorous and borate, demonstrate promise as flame-retardant synergists. SiO<sub>2</sub>-associated MOFs are particularly efficient, especially in smoke suppression applications within the EP system. Figure 4 illustrates the decrease in (PHRR) observed in flame-retardant EP composites with the incorporation of MOFs in various recent studies.



**Figure 4.** MOFs have been found to lower the PHRR result of flame-resistant EP composite materials in various recent studies [4].

### 3.5. Thermoplastic polyurethane-MOF composites

Thermoplastic polyurethane (TPU), renowned for its versatility, is widely applied in traffic equipment, wastewater filtration, tissue engineering scaffolds, 3D printing cable jacketing, textiles, surgical meshes, and aerospace owing to its excellent processability, resilience, abrasion resistance, chemical resistance, and superior mechanical properties. However, applications in these contexts necessitate elevated fire-resistant capabilities. Zhang et al. [5] endeavored to enhance the flame retardant of TPU by combining cobalt-MOF produced from new organic ligand (H<sub>2</sub>L) consisting of salicylaldehyde (SA) and 4,4-Diaminodiphenyl methane (DDM) with ammonium polyphosphate (APP). Untreated TPU exhibited

susceptibility to ignition accompanied by significant molten droplets and had an (LOI) of only 21.7%, failing to achieve any UL-94 classification in vertical flame spread tests. The integration of 1.5 wt% Cobalt-MOF and 4.5 wt% APP elevated the LOI to 28.2%, successfully attaining a UL-94 V-0 grade without dripping. Moreover, the (PHRR) of the composite material that included 1.5 wt% Cobalt-MOF with 4.5 wt% APP demonstrated a substantial 81% reduction compared to pure TPU, indicating the promising potential of combining MOFs with traditional additives flame-retardant to enhance TPUs fire safety characteristics.

**Table 1:** Represent the composition of a polymeric material such as NBR, EPDM, and silicone rubber Without using metal-organic frameworks [20].

	Compositions	Ablation rate (mm/s)	Thermal conductivity (W/mK)	Tensile strength (MPa)	Elongation (%)	Density (kg/m <sup>3</sup> )
Nitrile rubber matrix	0% PR/NBR-50%PR/NBR	0.235/0.095	0.07-0.185	-	-	1.03-1.14
	NBR/15phr Silica aerogel	0.108	0.172	8.2±0.2	1040±23	1.190
	NBR/6phr KP – NBR/8 phr MPI	0.137-0.05	-	15.4-18.2	666.6-768.7	-
	Carbon + SiC/NBR	-	0.52–1.61	85-125	0.43-0.47	-
	NBR/6% SCF	0.06	0.08	4.75	22	-
	NBR/50phr silica	0.15	0.259	255	715	1.19
	25 phr CF /25 phr KP /EPDM	-	0.198	11	7.4	1.16
	30phr KP/EPDM	0.015	0.171	9.35	11.7	1.185
EPDM matrix	20% Wollastonite/EPDM	-	-	3.8±0.4	195.3±19.7	1.06
	EPDM/10phr KP/5phr Al	0.01	0.21	8.5	16	1.2
	10 phr KP /5phr MWCNT /EPDM	0.092±0.007	-	6.28±0.40	385±25	1.120±0.02
	EPDM/10 phr Kynol	-	-	5.5	185	0.97±0.02
	EPDM/20phrSiC	0.21	-	12.09	716	1.081
	Silicon rubber	40 phr ZrC/silicon / 40 phr ZrO <sub>2</sub>	0.045/0.02	-	4.46/4.38	83.3/78.7
20 phr CF/silicon		.05	-	5.63	-	1.12

Abbreviations: PR: phenolic resin CF: carbon fiber. KP: Kevlar pulp. SCF: short carbon fiber. MWCNT: multiwalled carbon nanotubes. (-): not analyzed. phr: parts per hundred resin. FS: Fumed silica. MPI: modified polyimide. SiC: silicon carbide.

#### 4. New Approaches to Flame-Retardant Polymeric Materials

In this study, MOFs were not integrated with polymeric materials such as NBR, EPDM, and silicone rubber. Such polymers possess many benefits, including low density, low thermal conductivity, high mechanical properties, and low ablation rate. Despite their favorable characteristics, their inherent flammability poses a significant challenge. Therefore, addressing this flammability issue is crucial to unlocking the full potential of these materials in various industries and applications. Strategies for enhancing fire resistance, such as incorporating flame-retardant additives are essential for broadening their usage and ensuring safety in diverse environments. Table 1 presents various research studies on

thermal insulation compositions incorporating different types of fillers [19]. It also offers a comprehensive overview of the filler types utilized, including silica nanoparticles, Kevlar pulp, and aerogels their corresponding impacts on thermal insulation performance. These fillers serve to augment the materials thermal insulation properties by mechanisms such as diminishing thermal conductivity or enhancing heat resistance and ablation rate. Hence, looking into the effect of the MOFs on these polymers could lead to the generation of useful information for the improvement of these polymers flame-retardant properties. Studying the influence of MOFs on NBR, EPDM rubbers, and silicone rubbers, researchers can estimate synergistic effects that play an important role in improving their fire safety performance. The alignment of findings tests in table 1 enables the researchers to assess the performance of various fillers regarding the achievement of thermal insulation goals and to identify prospective tracks of further studies, e.g. using the MOFs as flame retardants in rubber compositions, owing to the diversity of MOF types as a flame retardant utilized in rubber compositions. This research approach can be a major step towards developing a new generation of flame retardant solutions that will be highly specific to particular polymeric matrices and will enable progress in engineering fire safety and materials science.

## 5. Conclusion

MOFs have proven their efficacy as innovative flame retardants for polymeric materials in recent years. ZIFs, UiOs, MILs, or recently designed MOFs demonstrate outstanding compatibility with polymer matrix, and the catalytic effect of metal nodes, in conjunction with multifunctional ligands provide possibilities for acting as highly efficient flame retardants. The incorporation of MOFs has shown significant reductions in (PHRR) and (THR), as well as improvements in (LOI) values, indicating enhanced flame retardancy. However, there is a need for more research to fully investigate the potential use of MOFs in addressing polymer fire risks and to improve their performance in various applications. Presently, a multitude of ligands for MOFs are synthesized across the globe, with MOFs-based flame retardants currently in the early stages of fundamental research. Going forward, the main challenges will be to reduce the cost of ligand synthesis, improve the functionality of MOF ligands, find a way to functionalize MOFs and apply MOFs or their derivatives in a useful way. These steps will be essential for developing future MOF-based flame retardants with high performance aligning with the ongoing trends in this field. Looking ahead, there is a noticeable trend toward employing MOFs as replacements for halogen-based flame retardants. This transition is motivated by the ease of processing MOFs and their compatibility with the base matrix in composite materials. Furthermore, there's a growing interest in studying the effects of incorporating MOFs into polymeric materials such as NBR, EPDM, and silicon rubber to understand how they interact and influence the properties of these polymers. Also, the next assessments will focus particularly on evaluating the effectiveness of MOF incorporated with polymeric materials in scenarios involving oxyacetylene flames.

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