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Epoxy Resin Flame Retardancy: From Mechanisms to Advanced Modification Strategies

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Abstract. Epoxy resins (EP) represent a category of vital thermosetting materials distinguished by their exceptional comprehensive performance and find widespread utilization in electronic packaging, aerospace and numerous industrial fields. However, their inherent flammability limits their application in scenarios with strict safety requirements. By analyzing the combustion mechanisms of EP, this paper systematically explains the basic principles of flame-retardant modification of EP and summarizes two main modification strategies: physical blending and reactive modification. In view of the limitations and technical bottlenecks in current research, this paper further prospects the future development directions of flame-retardant EP technology, so as to provide a solid theoretical basis for subsequent research and engineering applications.

Keywords: Epoxy resins, Flame-retardant modification, Physical blending, Reactive modification, Combustion mechanism

1. Introduction

Epoxy resins (EP) are typical high-performance thermosetting polymers. Benefiting from outstanding mechanical properties, reliable insulating performance, strong anti-corrosion capacity and great interfacial affinity with fillers, they are widely utilized in electronic encapsulation, aerospace components and structural construction. Nevertheless, epoxy matrices feature intrinsic flammability. Mass heat and combustible volatiles will be released once ignited, speeding flame propagation and imposing severe limitations on their deployment in scenarios requiring stringent fire safety standards [1].

Consequently, ire-safety modification toward epoxy matrices remains a hot research topic across academia and manufacturing industries. Three primary pathways are responsible for their fire-suppression performance, namely gas-phase action, condensed-phase carbonization and multi-component synergism. In the gas-phase route, thermal decomposition of additives yields inert free radicals that capture high-energy H· and ·OH species, breaking the continuous combustion chain reaction [2]. Condensed-phase fire suppression suppresses polymer thermal degradation and accelerates dense char accumulation, which serves as a barrier to hinder heat and volatile mass exchange, creating a dense barrier layer on the material's surface [3]. Synergistic flame retardancy, leverages the complementary effects of multiple flame-retardant components to enhance fire-resistance efficiency [4]. Two mainstream approaches are adopted to endow EP with fire resistance:

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physical blending and reactive chemical modification. Physical blending has the advantages of a simple process and wide applicability, but additives are easy to migrate and separate out, which affects the long-term stability and mechanical properties of materials [5, 6]. In contrast, reactive modification introduces flame-retardant functional groups into the molecular chains of EP or curing agents via covalent bonds, thereby significantly improving flame-retardant durability and interfacial compatibility [5, 6].

Substantial breakthroughs have been attained in fire-resistant epoxy modification over recent decades, yet multiple obstacles persist. Conflicts exist among fire safety efficiency, mechanical robustness, eco-friendliness and the integration of diverse functional features [2, 7, 8]. This paper systematically elucidates the combustion mechanism of EP and the fundamental principles of flame retardancy, compares and analyzes the two approaches of blending and reactive modification, and identifies current bottlenecks and future development trends. This review delivers solid theoretical guidance and technical support to facilitate the design of high fire-safety epoxy composites.

2. Combustion characteristics and flame-retardant mechenism of EP

2.1. Combustion mechanism of EP

Epoxy combustion undergoes four sequential phases: preheating, thermal degradation, ignition and sustained burning [2]. External heat input enables the material to accumulate thermal energy, and polymer pyrolysis initiates once the temperature exceeds its decomposition critical point. The secondary hydroxyl (-CH(OH)-) groups in the epoxy backbone undergo dehydration reactions to form vinyl ethers (-CH=CH-O-). The C-O bonds within allyl ether structures exhibit weak thermal resistance, leading to dominant chain cleavage at allyl positions [1]. Thermal degradation at aliphatic chain terminals further releases allyl alcohol, acetone and multiple hydrocarbon products [1]. Once these flammable volatiles mix with atmospheric oxygen and reach the autoignition temperature, the material ignites. The ongoing gas-phase combustion chain reaction continuously releases substantial heat. At the same time, the thermal radiation from the flame accelerates the pyrolysis of the unburned material, releasing more combustible gases, which in turn serve as fuel for subsequent gas-phase reactions [2].

2.2. Flame-retardant mechanism

According to the "fire tetrahedron" theory, continuous combustion requires four elements: fuel, oxygen, heat, and chain reaction. Therefore, the core principle of flame-retardant technology is to break the balance of the fire tetrahedron by weakening one or more conditions, so as to stop the combustion process.

2.2.1. Gas-phase

Gas-phase retardancy typically occurs when flame retardants decompose and release inert radicals. Such low-active substances trap high-energy H· and ·OH radicals to suppress the progression of combustion chain reactions [2]. For example, halogen-containing flame retardants decompose to generate halogen radicals (·Cl, ·Br). Such radicals trap highly reactive ·OH and H· within flames, slowing down combustion and reducing the heat release rate. Furthermore, the halogenated hydrogen gases produced during these reactions dilute the oxygen concentration, further inhibiting flame spread [3]. As illustrated in Figure 1, phosphorus-containing flame retardants (e.g., DOPO and

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its derivatives) produce low-reactivity PO· and HPO· radicals during thermal decomposition. These radicals efficiently scavenge reactive H· and ·OH [9], which terminates the chain reactions of combustion [10].

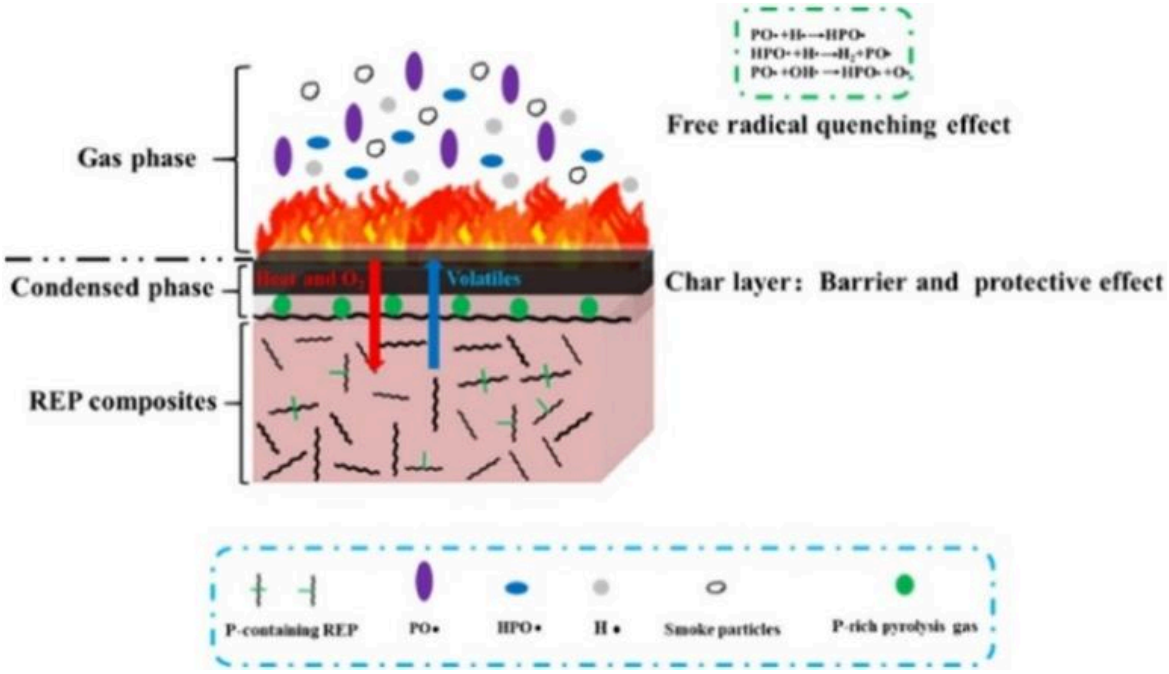

Figure 1. Schematic diagram of radical quenching effect [2]

2.2.2. Condensed-phase

In the condensed-phase mechanism, flame retardants restrain polymer pyrolysis and stimulate the growth of surface protective char. The formed char layer serves as a physical barrier, isolating the pristine polymer matrix, cutting off heat and volatile diffusion, and impeding continuous flame spread [3].

During combustion, phosphorus-based compounds thermally decompose into polyphosphoric acid, polyphosphonic acid, and other acidic products. These acidic substances adhere to the material surface and catalyze polymer dehydration and carbonization, ultimately constructing a thermally stable and heat-resistant char layer [11]. Similarly, silicate-based flame retardants form non-combustible residues such as silica after thermal decomposition, which stabilize the char layer, and reduce the heat release rate [3]. Zhai et al. developed D-POSS, a silicate-containing additive for EP fire-resistant modification. Their research verified that D-POSS is capable of promoting the generation of intumescent char layers during EP combustion. Fig. 2 reveals that thermally generated silica migrates to the char outer layer, improving structural stability and effectively inhibiting ongoing combustion [12].

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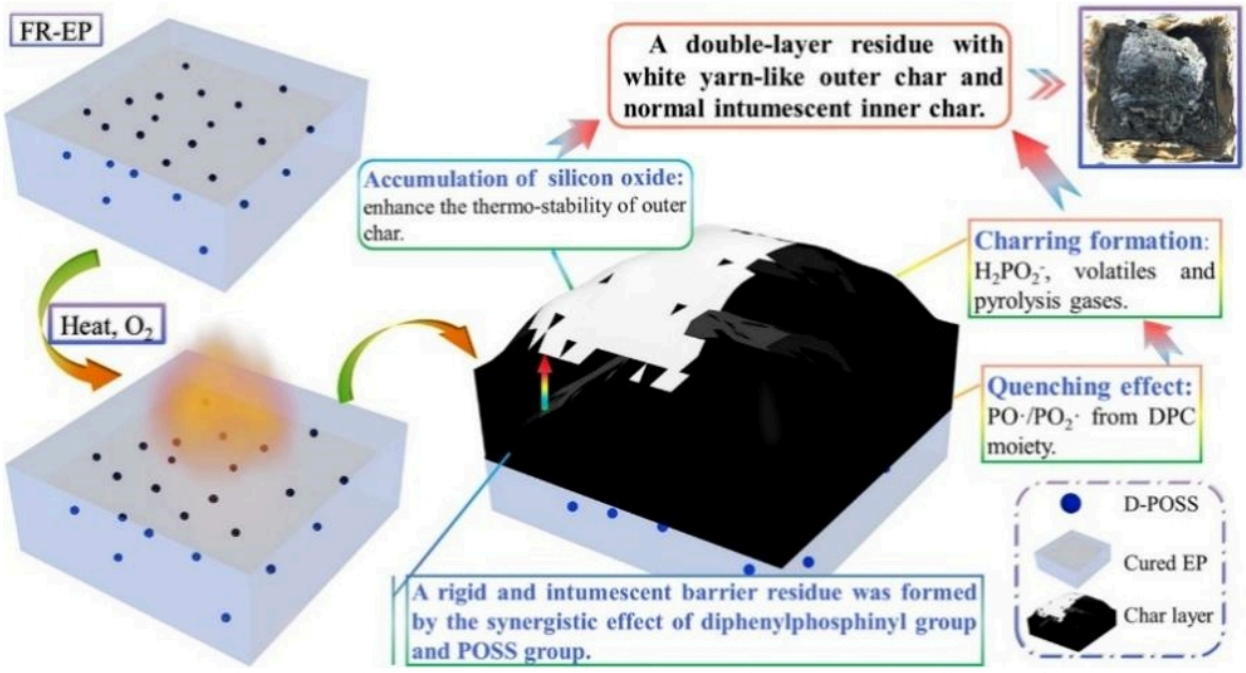

Figure 2. Illustration of the flame-retardant mechanism for the FR-EP [12]

2.2.3. Synergistic retardancy

Synergistic flame retardancy relies on the synergistic interaction of two or more additives in both the gas and the condensed phase, thereby producing a multiplicative effect. A typical intumescent flame retardant (IFR) system comprises three core components: an acid source, a carbon source, and a gas source [4]. Upon heating, the acid source decomposes and releases acidic species, which catalyze the dehydration of the carbon source to construct a char framework. Simultaneously, the gas source undergoes thermal decomposition to produce inert gases, prompting matrix expansion and the formation of porous swollen char [13]. Lim et al. further demonstrated that the combination of ammonium polyphosphate (APP) and melamine cyanurate (MC) in IFR systems delivers prominent synergistic flame-retardant performance [14].

2.2.4. Other effects

Some flame-retardant additives can generate non-flammable gaseous products upon degradation. Such inert gases reduce the content of flammable volatiles and oxygen in the combustion zone, achieving efficient fire suppression [15]. Other additives such as metal hydroxides decompose with a strong endothermic effect; the generated water vapor absorbs a lot of heat, significantly reduces the substrate temperature and slows down the heat transfer [16].

3. Flame-retardant modification methods for epoxy resins

3.1. Blending flame-retardant modification

Blending modification is to physically add non-reactive flame retardants into uncured EP prepolymer, and then prepare flame-retardant composites through conventional curing process [5].

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This method is characterized by simple operation and favorable compatibility, without complicated chemical modification on the resin molecular structure [6]. However, such flame-retardant systems possess intrinsic deficiencies. Without covalent linkage between fillers and the epoxy matrix, the additives tend to migrate and precipitate gradually, causing continuous degradation in mechanical integrity and fire resistance durability [5, 6]. Consequently, current research mainly focuses on developing additives with good interfacial compatibility, high efficiency, and environmental friendliness.

3.1.1. Inorganic additive flame retardants

Red phosphorus (RP) is a relatively stable and low-toxicity phosphorus allotrope [17]. RP has a significantly higher phosphorus content compared to other phosphorus-based flame retardants, enabling effective flame-retardant performance at low loading levels [18]. Its flame-retardant effect is particularly obvious when used together with metal hydroxides in EP matrix [17]. However, unmodified red phosphorus powder is flammable, which brings great risks in the pre-processing process. Moreover, after being added to polymers, it is easy to hydrolyze and degrade slowly, releasing trace toxic and odorous phosphine gas and corrosive phosphoric acid. Its deep red color is also difficult to cover with pigments [11]. To reduce operation and safety risks, commercial red phosphorus is mainly provided in the form of pre-dispersed masterbatches [19]. To address its hygroscopicity, RP is microencapsulated within an organic shell, forming core-shell microencapsulated RP (MRP) [18]. Cheng et al. fabricated microencapsulated red phosphorus (RP@CH/LS MRP) using chitosan/lignin sulfonate (CH/LS) composites as the shell layer, which was incorporated into epoxy matrices for flame-retardant modification [18] (Fig. 3.). Experimental results revealed that the EP composite filled with 7 wt% RP@CH/LS achieved a limiting oxygen index (LOI) of 30.6% and a UL-94 V-0 rating [18].

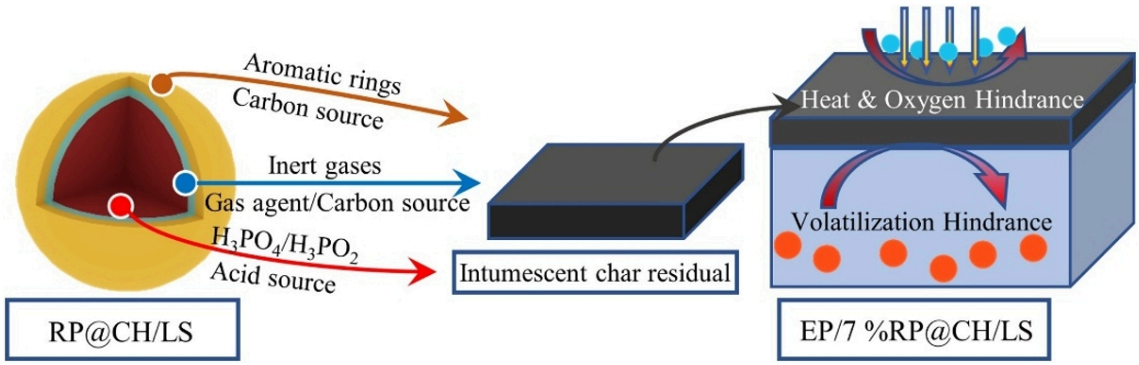

Figure 3. Flame retardance mechanism of RP@CH/LS in EP matrix [18]

3.1.2. Organic additive flame retardants

Widely recognized as eco-friendly halogen-free alternatives, organophosphorus flame retardants feature low smoke and low toxicity characteristics [20]. DOPO-based derivatives exhibit superior thermal stability, hydrolysis resistance and fire resistance. Even at a low dosage that endows EP with greatly improved flame retardancy, such fillers inevitably reduce the Tg value and impair the mechanical performance of epoxy matrices [21].

Yan and coworkers developed DiDOPO, a phenethyl-bridged DOPO derivative, and integrated it with organically modified montmorillonite to realize synergistic flame retardancy. The results

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demonstrate that the physical barrier function of montmorillonite cooperates with the chemical flame-retardant effect of DiDOPO, endowing the composites with outstanding comprehensive fire safety performance [22]. In order to solve the migration problem of small-molecule organophosphorus additives, hyperbranched phosphorus-containing polymers have attracted extensive attention. Battig et al. developed a series of hyperbranched phosphorus-based flame retardants (hb-FRs, Fig. 4) with excellent compatibility with epoxy matrices. At a dosage of 10 wt%, different hb-FR structures reduce the PHRR by 22% to 44% [23].

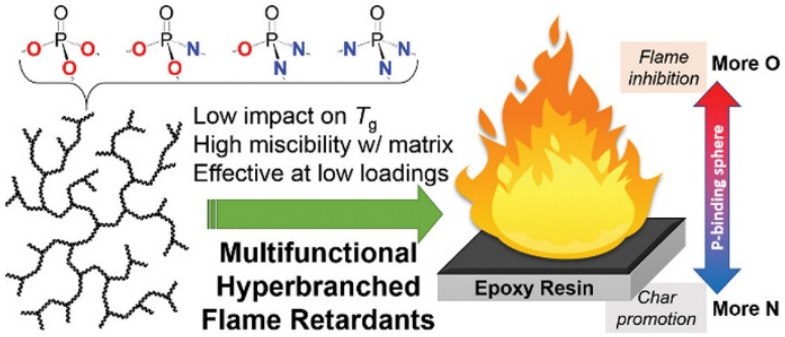

Figure 4. Novel hb-FRs provide multifunctional flame retardancy to EP [23]

3.2. Reactive modification

Reactive modification permanently introduces flame-retardant groups into the EP polymer network through covalent bonds, which fundamentally solves the migration problem existing in additive systems [5, 6]. This strategy primarily involves the copolymerization of functionalized monomers and the use of inherently flame-retardant curing agents [5, 6].

3.2.1. Copolymerization with flame-retardant monomers

Copolymerization chemically grafts functional groups (such as phosphonate, phosphate, nitrogen-containing heterocycles or siloxane) onto epoxy monomers before crosslinking. Reactive intermediates are usually synthesized by ring-opening, condensation or substitution reactions for active epoxy groups or hydroxyl groups. DOPO contains active P-H bonds that undergo chemical reactions with epoxy groups. This process yields phosphorus-containing monomers and firmly embeds the flame-retardant DOPO structure into the backbone of the cured polymer [5]. Liu et al. reacted DOPO with benzophenone derivatives to synthesize phosphorus-containing epoxy precursors (2DOPO-E1, 2DOPO-E2) with double DOPO side chains and a phosphorus-based diamine curing agent (2DOPO-A) to prepare a series of inherently flame-retardant EP [24]. The prepared polymer composites possess excellent thermal resistance, as evidenced by their 5% thermal decomposition temperature above 316 °C, along with high LOI levels that reflect outstanding flame retardancy. Similarly, Xu et al. used D-bp as a phosphorus source to react with glycidyl POSS (G-POSS) to prepare organic-inorganic POSS/D-bp/DGEBA hybrid composites (as shown in Fig. 5). The modified matrix passes the UL-94 V-0 test. It is worth noting that at an ultralow phosphorus content of 0.25 wt%, the composite achieves a 60.5% reduction in PHRR accompanied by improved mechanical properties [25].

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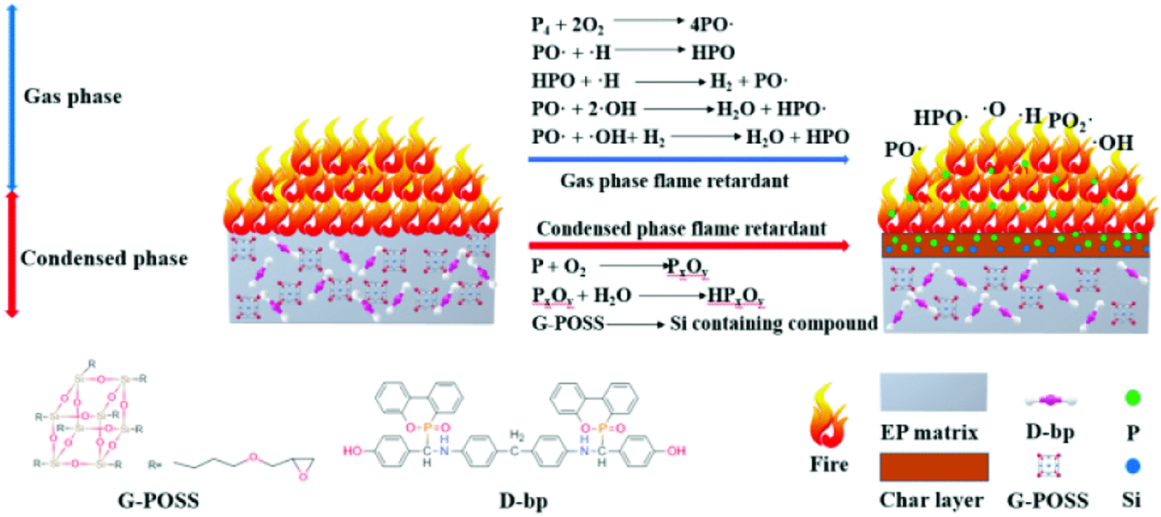

Figure 5. POSS/D-bp/DGEBA hybrid composites [25]

3.2.2. Introduction of flame-retardant curing agents

Curing agents are essential components of the crosslinked network. Integrating flame-retardant groups (such as phosphorus-nitrogen synergistic groups, melamine derivatives or boron-containing heterocycles) directly into their molecular structures can ensure permanent chemical combination during the curing process. This strategy has high process compatibility because it does not need to modify the base resin, only needs to replace conventional curing agents (such as ethylenediamine, diethylenetriamine).

Shao et al. grafted diphenylphosphinyl chloride (DPPC) onto polyethyleneimine (PEI) backbone to synthesize polymer curing agent DPPEI. When applied to EP, the obtained DPPEI-EP composite (Fig. 6) shows excellent flame resistance, reaching UL-94 V-0 rating without molten droplets, and delivers an LOI value of 29.8%. Cone calorimeter tests demonstrate that with 30 wt% DPPEI incorporation, the total heat release (THR) and total smoke release (TSR) of the composite are reduced by 69.5% and 78.3%, respectively, compared with the unmodified PEI-EP sample [26]. Huo et al. synthesized a novel DOPO-functionalized imidazole curing agent (DIB), which was adopted together with 4,4'-diaminodiphenyl sulfone (DDS) as a co-curing system for epoxy resin. The resulting EP/DDS/DIB thermosets exhibited prominent fire suppression performance, with peak heat release rate (pk-HRR), average heat release rate (av-HRR), and total heat release (THR) decreased by 57.2%, 37.9%, and 36.0%, respectively. Notably, an ultralow phosphorus loading of 0.75 wt% enabled the composite to achieve a UL-94 V-0 rating and a high LOI value of 38.5% [27]. A phosphorus-functionalized diamine monomer (PPXSPB) was fabricated by Li's group for epoxy resin modification. At a phosphorus loading of 3.24 wt%, the EP/PPXSPB composite attained V-0 flame retardancy and a high LOI of 31.4%. Notably, the introduction of PPXSPB did not compromise mechanical properties, with measured impact strength, tensile strength and bonding strength reaching 6.58 kJ/m², 47.10 MPa and 13.79 MPa, respectively [28].

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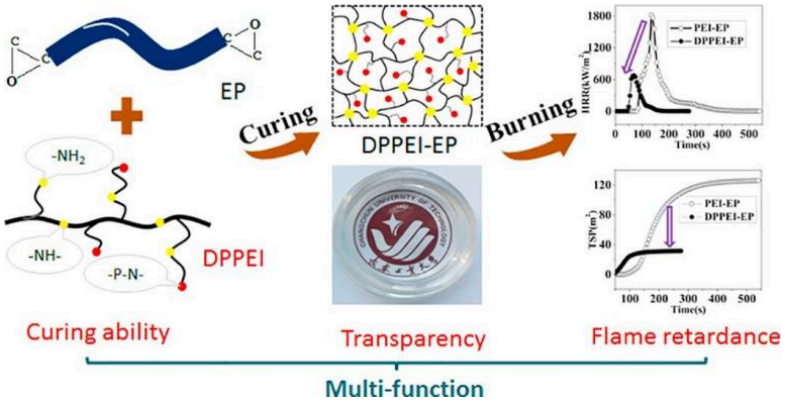

Figure 6. Synthesis and performance of multifunctional DPPEI-EP [26]

4. Challenges and future development trends

Although remarkable advances have been achieved in the flame-retardant modification of epoxy resins in recent years, the existing systems still have great limitations under the strict requirements of industrial applications and high-end technologies. Future research is gradually shifting from simply pursuing flame-retardant performance to balancing comprehensive performance, environment-friendly formula and advanced multi-functional integration.

4.1. Balancing flame retardancy and mechanical properties

It remains a common bottleneck that improving the fire resistance of epoxy resins inevitably compromises their mechanical properties. Traditional additive flame retardants have poor interfacial compatibility, which may lead to long-term migration and affect crosslinking density, resulting in the decrease of mechanical strength [8]. In contrast, although reactive modifiers (such as traditional DOPO derivatives) can alleviate migration problems, their introduction changes the polymer main chain and reduces crosslinking density, leading to the decrease of Tg and structural rigidity [16]. It is still a critical challenge to simultaneously improve the flame-retardant performance and maintain the original mechanical properties of epoxy resins [2].

High-aspect-ratio nanoparticles with large specific surface areas and superior physical barrier effects (such as layered silicates, carbon nanotubes, graphene, nano-silica) have been reported to simultaneously enhance the mechanical properties, thermal stability and flame retardancy of materials at low loading levels (2–5 wt%) [21]. Yu et al. fabricated phosphorus-nitrogen flame retardant-wrapped functionalized reduced graphene oxide (FRGO) via a facile one-pot synthetic strategy. After being covalently anchored into the epoxy network, the synergistic interaction between phosphorus-nitrogen flame-retardant moieties and graphene nanosheets prominently lowers the PHRR and THR of the composites, while well preserving the structural integrity of EP materials [29].

4.2. Environmental sustainability

With the increasing global attention to resource shortage and ecological environment deterioration, the concepts of carbon peaking and carbon neutrality are increasingly guiding the design of materials. Traditional halogenated flame retardants emit highly toxic and corrosive smoke once ignited, while more than 90% of conventional epoxy flame retardants rely on bisphenol A (BPA)

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from non-renewable petrochemical resources [2]. Accordingly, the development of high-efficiency, halogen-free, and bio-based sustainable flame-retardant systems has become an urgent research demand. Zhu et al. successfully synthesized a bio-based macromolecular ammonium phytate (PA-AEP) and applied it as a single-component curing agent for epoxy resins. At a loading of 20 wt%, the resulting bio-based epoxy composite achieved a UL-94 V-0 rating with an LOI value of 28.0%. The formation of a dense, phosphorus-enriched intumescent char layer in the condensed phase endows the material with outstanding halogen-free flame retardancy and efficient smoke suppression performance [30].

4.3. Multifunctional integrated flame-retardant modification

With the rapid advancement of 5G communication, aerospace engineering, and high-end electronic devices, sole flame-retardant capability can no longer satisfy the multifaceted application requirements of modern industries. Accordingly, the development of next-generation flame-retardant epoxy composites has shifted toward integrated multifunctional design, incorporating high thermal conductivity, electromagnetic interference (EMI) shielding, antibacterial activity, and hydrophobic performance. Guo et al. successfully pyrolyzed lignin-resorcinol-glyoxal prepolymer into carbon foam (CF), and then impregnated flame-retardant EP to prepare an advanced multi-functional composite material (FREP-CF). After continuous exposure to a 1000 °C flame for 10 min, the back-side temperature of the composite remains safely below 200 °C. Combined with its outstanding EMI shielding effectiveness of 33.5 dB and favorable electrical conductivity of 216 S/m, this multifunctional material offers a promising solution for next-generation aerospace and electronic components serving extreme service environments [7].

5. Conclusion

The intrinsic flammability of epoxy resins severely limits their practical applications in high-safety and high-end technical fields. This review systematically elaborates the gas-phase, condensed-phase, and synergistic flame-retardant mechanisms of modified EP, and comprehensively classifies current modification strategies into physical blending and reactive modification. Although reactive modification ensures good interfacial compatibility and long-term durability through covalent bonds, the field still faces the challenge of balancing mechanical properties and fire safety, as well as meeting increasingly stringent environmental protection requirements. Future research on flame-retardant EP will focus on the synergistic optimization of mechanical and flame-retardant performances, the exploitation of bio-based halogen-free flame-retardant systems, and the integration of multifunctional characteristics including thermal management and electromagnetic shielding. This review provides a valuable reference for the future innovation and engineering application of high-performance flame-retardant epoxy materials.

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