



Review

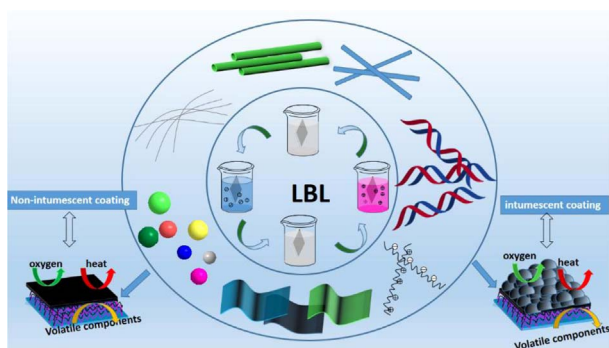
Flame retardant coatings prepared using layer by layer assembly: A review



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GRAPHICAL ABSTRACT



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ABSTRACT

Many polymers, especially polyurethane foam and fiber fabric, are particularly easy to burn and generate a large number of droplets leading to the rapid spread of the fire. Therefore, high-performance flame retardants or flame retardant technologies so is important to ensure the safety and reliability of polymer-based composites. In this respect, the flame retardant coatings prepared by simple and environmentally friendly layer by layer assembly could be especially important because they can improve the flame retardant performance of polymers while the intrinsic properties of the polymers remain unchanged. The present review summarizes the research progress and future development trends of flame retardant coatings, including intumescent coatings, non-intumescent coatings and multifunctional coatings prepared using layer by layer assembly; in particular, this review highlights the flame-retardant mechanisms of the assembled materials with different dimensions.

1. Introduction

Polymeric materials are ubiquitous and permeated in all aspects of human life and play a huge role in our daily life [1–3], due to their good mechanical properties, heat resistance, durability and corrosion resistance, among other favorable properties. However, some polymers, especially porous polymers with a large specific surface area, such as polyurethane foam [4–6], cotton [7,8], and poly(ethylene terephthalate) (PET) fabrics [9,10], burn easily, releasing a large amount

of heat, flame and smoke in association with the generation of a large amount of droplets during the burning process, thereby threatening people's lives and property. Thus improvements in the flame retardant properties of polymers are urgently needed.

A common approach to improving the flame-retardant properties of polymers is to add flame retardants. For example, halogen-containing flame retardants, such as tris(2-chloroethyl) phosphate, tris(1,3-dichloroisopropyl) phosphate, pentabromobenzyl acrylate and tris(1-chloro-2-propyl) phosphate, have been widely applied as the early stage

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flame retardants to commercial products such as furniture, mattresses, electronics [11,12]. However, these halogenated compounds produce dioxin, furan, and other carcinogenic substances in the combustion process, which highly limits their applications in engineering [13,14].

To avoid the aforementioned shortcomings of halogen-containing flame retardants, researchers have been actively pursuing halogen-free flame retardant technology which mainly involves phosphorus, nitrogen, silicone, boron, zinc, iron, and aluminum-containing flame retardants. Among them, phosphorus-containing flame retardants, one category of the most promising halogen-free flame retardants, exhibit favorable flame retardant efficiency. The reason lies in that phosphorus-containing compounds can undergo dehydration and carbonization to form protective carbon layers thereby effectively reducing the flammability of the polymers [15–18]. Nitrogen-containing flame retardants can absorb heat and produce noncombustible gases to dilute the concentration of combustibles during the decomposition process of polymers. In the meantime, although nitrogen-containing flame retardants are less competitive than phosphorus-containing flame retardants in terms of the flame-retardant efficiency, the nitrogen-containing flame retardants are relatively environmentally friendly and release only a small amount of smoke particles and toxic gases in the combustion process [19–21]. Silicone-containing flame retardants can often form an insulating layer on the polymer surface upon burning, thereby effectively impeding the transmission of oxygen, heat and mass and reducing the flammability of polymers [22–25]. Moreover, compounds containing boron [26,27], zinc [28,29], iron [30–33], aluminum [34–37] and so on can suppress smoke and retard flame to some extents, but their application in engineering is limited [38]. However, there are still some problems in the use of these flame retardant additives: increasing viscosity and modulus of the final polymeric material which will make industrial processing difficult, high loading and their potential toxicity to human health and environment [11,39–44]. This is why highly effective and environmentally benign flame retardant techniques are attracting attention increasingly. Particularly, fire retardant coatings prepared via layer by layer (LbL) assembly could be used as a potential substitute for additive flame retardants, due to their high flame-retardant efficiency as well as desired environmental acceptance and low impact on the intrinsic properties of polymers [45].

As a versatile and cost-effective method to fabricate multifunctional films, LbL assembly involves the alternate immersion of a charged substrate into positively and negatively charged species by electrostatic interaction, although other interactions (e.g. donor/acceptor [46,47], hydrogen bonding [48,49], or covalent bond formation [50,51]) can also be employed, and it is applicable to assembling a variety of materials such as polyelectrolytes, nanoparticles, or biomolecules. Fig. 1 depicts the process of LbL assembly with a wide range of assembly materials. This novel method has been widely used for containing gas barrier [52–54], antimicrobial [55,56], biosensing [57,58], charge storage [59,60], antireflection [61,62] and drug delivery [63–65]. In addition, it is only in recently years that this technique has been

considered to design flame retardant coatings.

Compared with the traditional flame retardant method, the LbL method offers several advantages. On the one hand, the flame retardant multilayer films are constructed between a substrate and the exterior which can directly interfere with the combustion process and eliminate the challenges associated with processing or adversely modifying mechanical behavior when incorporating flame retardants into the substrate itself [27,66]. On the other hand, if the positively and negatively charged functional polyelectrolytes or nanoparticles are properly selected, the LbL assembly method, which is simple and versatile, can be readily adopted to fabricate multilayer films with controllable thickness, composition, and function. In addition, the LbL assembly technique relies on mild experimental conditions such as room temperature, atmospheric pressure and a low concentration of to-be-assembled materials (below 1 wt%), which often means it is a cost-effective route for fabricating coatings [67,68].

This paper summarizes recent research progress and future development trends of the flame retardant coating through LbL assembly. It also highlights the evolution of various assembly methods and the unique flame retardant mechanisms of non-intumescent coatings, intumescent coatings and multifunctional coatings.

2. Non-intumescent flame retardant coatings

Generally non-intumescent coatings mainly consist of inorganic nanoparticles which can be divided into zero-dimensional assembly, one-dimensional assembly and two-dimensional assembly according to their different sizes. The obviously improved fire safety of polymers is attributed to the fact that non-intumescent coating can provide an inorganic ceramic barrier (thermal shielding effect) during the pyrolysis of the polymer, which can effectively hinder the transmission of heat, oxygen, mass and volatile products from the underlying polymer to the atmosphere [69].

2.1. Zero-dimensional assembly materials and their composites

Zero-dimensional material assembly refers to the three dimensions of materials all at the nano-scale. These materials typically contain inorganic alumina and aluminum hydroxide, as well as non-metallic silica. Alumina and silicon accumulate on the surface of the burner during the combustion of the polymer which can form a dense ceramic physical barrier to protect the bottom of the undegraded polymer. The metal hydroxide can produce water vapor during the thermal degradation process, which can cool the matrix and dilute the concentration of oxygen and volatile combustible components; in addition, the metal oxide produced after thermal degradation will cover the surface of the polymer to form a physical barrier.

Alumina coated silica as a cationic polymer coupled with silica nanoparticles with different sizes (10 or 30 nm) was designed and deposited on PET fabric via LbL assembly [70]. When containing the smallest nanoparticles, the system exhibited the best results, increasing the time to ignition (TTI) by 99 s (45%) and reducing the peak heat release rate (PHRR) by 20% with 5 bilayer compared with the uncoated fabric. The same group also explored the effects of different assembly types of silica/silica and silica/polyethylene imine (PEI) and different sizes of negatively charged silica particles (8 or 27 nm) on the morphology and flame retardant properties of coated cotton fibers [71]. The PEI/silica films were thicker than silica/silica films because the silica particles in this system were better dispersed. When the diameter of the negatively charged silica was 8 nm, the cotton fibers coated with 10 bilayers PEI/silica exhibited the best flame retardant effect because the bilayers were composed of smaller colloidal particles with a higher practical packing density around the cotton fibers. The smaller inorganic solid particles more favor to the deposition of coating. In addition, silica/silica bilayers deposited on polycarbonate (PC) films improved the flame retardancy of the PC films [72]; and their flame-

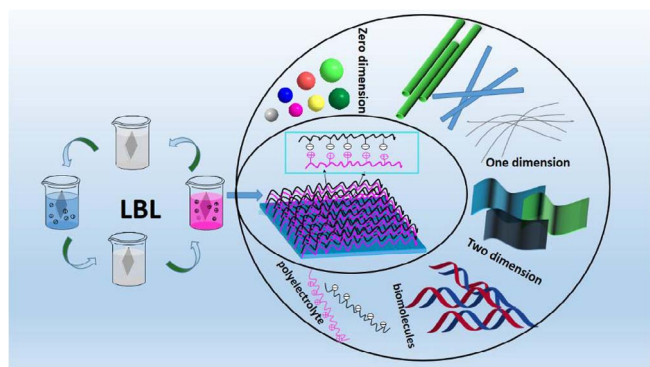


Fig. 1. General scheme of the LbL process with a wide variety of assembly materials.

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