

Review Article

Composition-engineered graphene oxide-based fire warning research: A systematic review and new insight

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ABSTRACT

Fire-warning systems (FWSs) attract increasing attention due to their potential for highly-efficient fire management, which have made some significant progress recently. Graphene oxide (GO) is one of the prime candidates on account of the insulate-to-conductive transformation under high temperature attack, it is precisely the electrical transformation can be captured for serving as a warning response. In this review, polymer-supported composition-engineered GO-based FWSs are reasonably sorted out and comprehensive analyzed, which constitutes a significant segment of FWSs research. Specifically, the evaluation covers the diversity of fabrication methods, variety of composition-engineered GO-based FWSs, the core fire-warning performance, as well as the relevant working mechanisms, etc. Furthermore, given the state-of-the-art in composition-engineered GO-based FWSs, the present challenge and the emerging perspectives are proposed, providing a guidance for tackling the main issues and promising developing direction of the next-generation of composition-engineered GO-based FWSs.

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1. Introduction

Fire statistic data about the change in number of fires per 1000 inhabitants (Fig. 1A), indicates a remarkable increased fire hazards recently, such as an Ideal MEGA vehicle fire in Shanghai, October 2025, Hong Kong residential area fire in November 2025. In addition, as presented in Fig. 1B, fire hazards are widely distributed in various scenarios, mainly concentrated in the building fire incidents compared to other fire types with approximate value of 25.5 %. These cases not only provide specific warnings but also imply the systemic risks resulted from fire hazards [1]. Fire hazards are ubiquitous in society, which seriously threaten human survival and sustainable development [2–5]. Fire hazards damage

is comprehensive, comprising of materials destruction, and release of smoke, toxicant, and other byproducts integrated during burning. Although most disasters cannot be avoided thoroughly, various efficient fire-safety management are taken to mitigate or reduce the fatality [6–9].

Broadly, these fire-control strategies are generally divided into passive and active fire-control approaches based on the working mechanism. The addition of flame retardant, as a passive method, can suppress the fire propagation or even make material self-extinguish during the whole combustion processing by reducing heat release and toxic products to mitigate fire hazards [7,10–13]. However, fires will sweep through all the areas quickly once materials are burned, leading to an uncontrollable risk [14]. Namely, introducing flame retardants is essentially to delay the spread of fires rather than to eliminate combustion completely, indicating limitations in fire management. Comparatively, more human power is put into active fire-control measures. Fire-warning strategy mainly working in the early stage of combustion process is gaining rising attention because of its proactivity of prevention,

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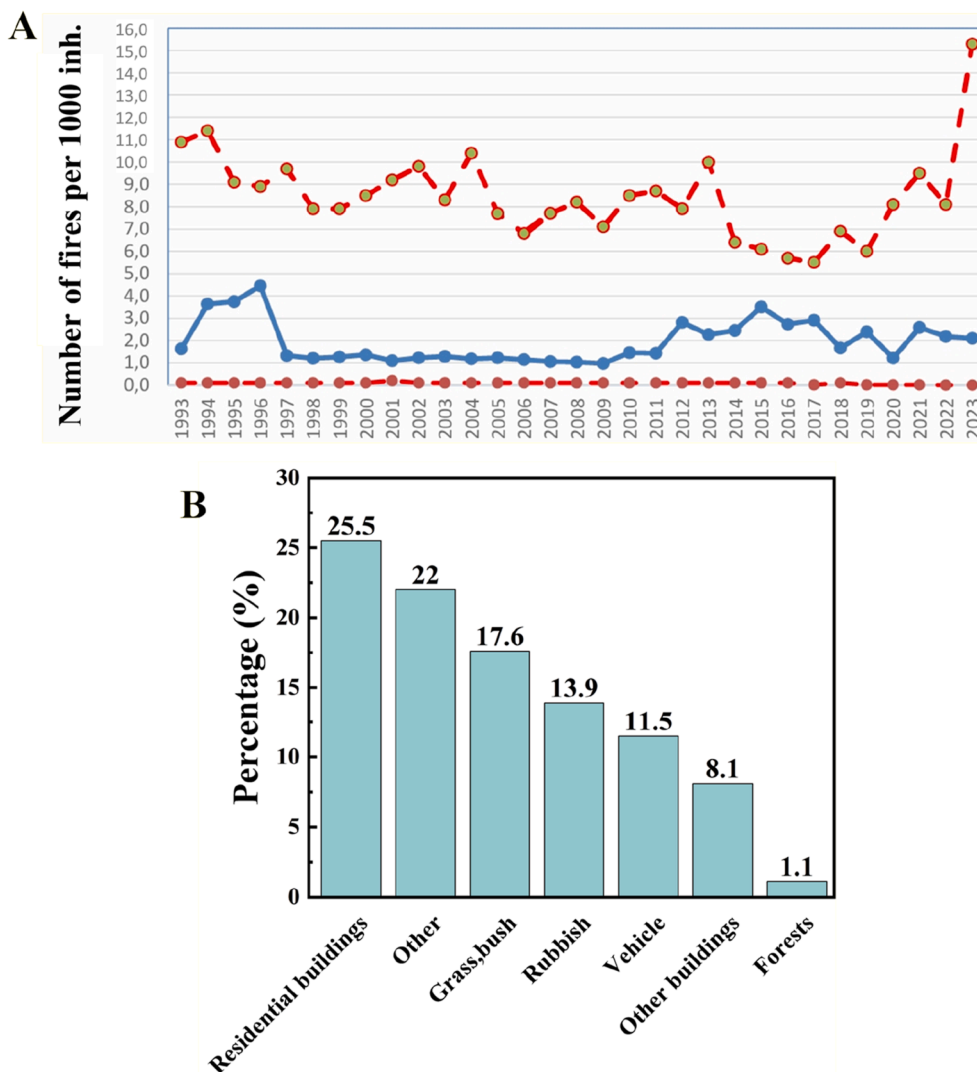
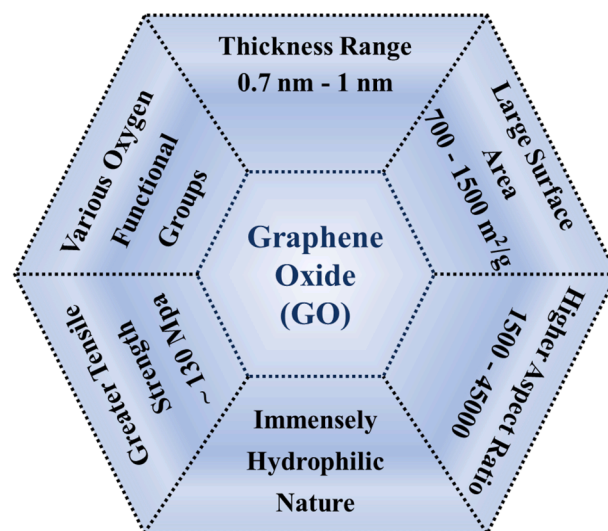


Fig. 1. **A.** Trends in number of fires per 1000 inhabitants. from CTIF Center for World Fire Statistics. **B.** Distribution of fires types in the world (2023) [1].

suppression effectiveness, and intelligent tendency, providing a large critical window for fire management.

Traditional fire sensors based on gas and smoke concentrations have been predominantly employed [15–19]. While mature and reliable, these fire sensors are nevertheless constrained by inherent response delays, especially during the initial smoldering phase of fires or in clean-burning scenarios where combustion byproducts are minimal [17,20,21]. In response to the above limitations, the research focus gradually shifts to thermal sensing in recent year. Temperature, as one of most direct physical parameters during combustion, can reflect the phenomena of energy release and heat accumulation at the earlier stage of combustion process, offering more incipient warning signals than gases and smoke. Therefore, thermal fire-warning sensor exhibits strong promising in enhancing the real-time performance, reliability of fire warning [7,10,22–29]. Two-dimensional (2D) GO is endowed with the economic efficiency, unique structure and tunable performance, containing large surface [30], higher aspect ratio, various oxygen functional groups (epoxide, hydroxyl group on the basal plane of GO, and carbonyl group on the edge of GO [31–33]), super mechanical strength, amphiphilic properties, etc. [31,32,34–36] (Scheme 1). More inspiring, GO presents typical



Scheme 1. Schematic illustration for representing GO properties.

thermal resistance change characteristic that transfer insulating feature to reduced graphene oxide (RGO) [36–40], which can qualify it for superior candidate in fire-warning study. Since the first report of GO-based FWS, remarkable progress has been made, which involves in not only the optimization of response sensitivity, but also a variety of investigation on composition engineered GO-based FWSs.

Herein, this review primarily concentrates on GO-based FWSs for systematic summarization and reasonably sorting particularly based on composite-engineering strategies, filling a critical void in the dedicated attention to composition-engineered GO-based FWSs studies in review literature. As depicted in Scheme 2, in consideration of the various treatment via composition engineering, the state-of-the-art GO-based FWSs prepared through the combination between GO and different polymeric substrates are comprehensive reviewed. Moreover, these GO-based FWSs are systematically evaluated with regard to fabrication approaches, composition-engineered variety, fire-warning performances, and the underlying mechanisms, *etc.* Furthermore, according to the current development, the ongoing challenges and prospectives of composite-engineered GO-based FWSs are proposed. This review is hope to offer an in-depth understanding of GO-based fire warning study for readers, with the expectation of inspiring researchers to tackle the key issues and facilitating further exploration about the next-generation of composition-engineered GO-based FWSs.

2. Fabrication approaches of composition-engineered GO-based FWSs

Early fire warning matrix materials are diverse, comprising of polymers, metal oxides, *etc.* Considering the significantly different physical and chemical properties caused by matrix materials, distinct fabrication techniques are utilized in a targeted manner to optimize the performance of fire-warning sensors.

2.1. Layer-by-layer self-assembly (LBL) technology

As presented in Scheme 3, LBL mainly relies on intermolecular interactions such as electrostatic interactions, hydrogen bonds, or covalent bonds to alternately adsorb molecules or nanomaterials

with opposite charges to fabricate functional materials layer by layer [41–43]. Specifically, LBL technology has the following advantages: (1) Controllable functional film construction, like precise tuning of composition, thickness, *etc.*; (2) Universality of matrix materials, which is compatible with different matrix materials; (3) Mild preparation conditions, possibly maintaining the structural integrity of active ingredients [10]; (4) The synergistic effect of multifunctional integration makes it easy to control the assembly sequence to impart multiple characteristics [42].

Given to the above-mentioned advantage, LBL technology can enable to develop the advanced composition-engineered GO-based FWSs that integrated exceptional thermal responsiveness and enhanced multifunctional performance [44–54], where the physical barrier effect of GO nanosheets synergizes with introduced additives to improve fire resistance [55,56], while the interfacial coupling between GO and polymer matrix enhances mechanical strength through precisely engineered layered architectures [55,57–60]. Beyond these fundamental properties, modulation of the assembly chemistry further endows the system with programmable humidity sensing [61], acid-alkali resistance [56,59], strain sensing ability [62], or emerging self-healing function [63], transparent feature [64], *etc.* making them promising candidates for sophisticated warning systems. Nevertheless, if the desired architecture were constructed, the inherently time-intensive deposition processing requires numerous cycles, leading to the time consumption and the constraint on the scalable manufacturing. While resisting mechanical delamination and maintaining structural integrity under prolonged environmental stressors provides the high demand of interfacial adhesion requirements. This presents critical challenges of composition-engineered materials that could be systematically tackled through coordinated optimization of deposition protocols and interfacial chemistry to bridge the gap between the laboratory innovation and the industrial implementation.

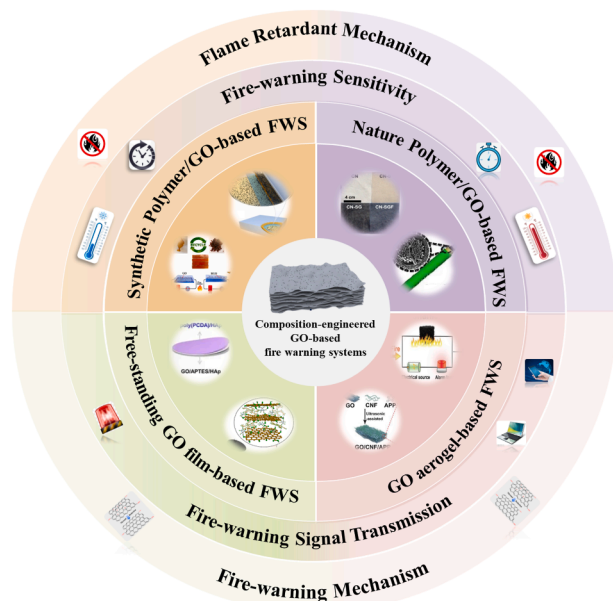
2.2. 3D printing

3D printing, as an additive manufacturing technology, has shown significant advantages in different fields, especially in the sensing area [65]. For instance, a functional three-dimensional aerogel with special porous structures was constructed by 3D printing way, demonstrating early fire warning performance and improved yield stress and elastic modulus [66]. Chen et al. also employs 3D printing technology to fabricate GO-based hydrogel modified by sodium alginate (SA) and carbon nanotubes (CNT), which can respond to a fire, accompanied by flame retardancy, adhesion, tensile resistance and self-healing ability [67]. Collectively, 3D printing technology is a vital approach to modulate the microstructure of materials as well, leading to robust fire-warning performance.

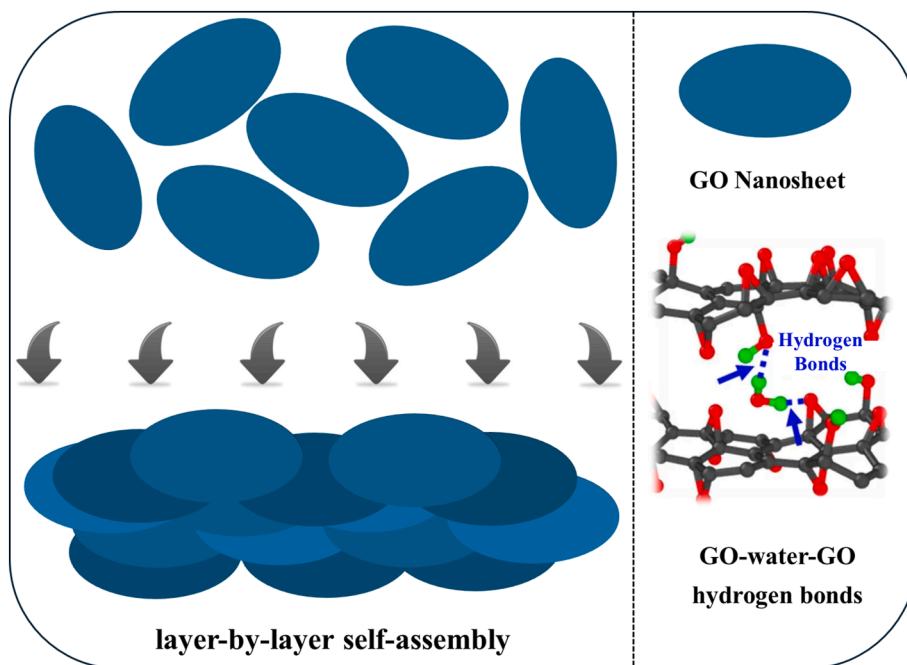
3D printing technology can break through the limitation of traditional manufacturing on geometric shapes. It displays the exceptional design flexibility without any molds, which is helpful for the custom structural design of personalized products. Moreover, 3D printing technology is highly economic efficient, showing in terms of a high utilization rate and low material waste. Nonetheless, there are still some primary limitations for the further development, specifically including narrow printable materials availability, constraints in material properties, the compromise of printability after functionalization, inherent process limitations, challenges in scaling up, *etc.*

2.3. Spray-coating

Spray-coating technology offers competitiveness, containing excellent adaptability to complex substrate geometries and



Scheme 2. Schematic diagram for composition-engineered GO-based FWSs.



Scheme 3. Schematic illustration of LBL technology.

precise process controllability, making it suitable for surface modification across diverse materials. Utilizing these advantages, spray-coating technology was employed to coat one kind of fire-warning solution onto the surface of polypropylene and wood substrate, with the controllable thickness of coating through optimized spraying parameters [68]. Nevertheless, this method faces restriction in material utilization efficiency and might raise environmental concerns due to overspray and emissions.

3. Composition-engineered GO-based FWSS

How to efficiently integrate GO into various substrates to build the matching GO-based FWSS is the key to accomplish the composite-engineered applications. Herein, we divide GO-based early fire-warning sensors into three categories comprised of one-dimensional (1D), 2D, three-dimensional (3D) GO-based FWSSs based on the spatial dimension of sensing network and the functional integration strategy. For each type of GO-based FWSSs, details are given in the following paragraphs.

3.1. 1D GO-based FWSSs

1D GO-based FWSSs are mainly manufactured by integrating GO into linear or point-like units, such as fibers, wires, etc. These units enable signal conversion via their linear electrical response to abnormal temperatures, providing early fire warning through a directional 1D transmission path. The mode presents characteristics of structural simplicity, directional response, as well as easy integration. The following sections elaborate on a systematic discussion centered on the 1D GO-based FWSSs.

As shown in Fig. 2A, the core-sheath functional fax yarn treated by GO, chitosan (CS) molecules, and polydopamine was prepared as a fire-warning sensor, achieving a low-temperature response within 3 s [69]. The implementation of the fire-warning modulation is precisely achieved through strategic structural modifications of cotton fabrics, allowing the preparation of a large-scale size. Moreover, A type of GO-based loop-fiber incorporated a fiber

Bragg grating was proposed, showing the temperature sensing and humidity sensing at the same time [70] (Fig. 2B).

3.2. 2D GO-based FWSSs

2D GO-based FWSSs aim to achieve the encompassing temperature sensing network, which mainly rely on the construction of the continuous and integral GO coatings onto the surface of various substrates, or free-standing films, etc. In comparison, the 2D distribution of temperature sensing monitoring is beneficial for the visual detection of abnormal temperatures and analysis of temperature gradients. Thus, this section mainly centers around 2D GO-based FWSSs and their fire-warning performance in 2D space to conduct a systematic discussion and evaluation.

As a typical natural cellulose-based material, cotton fabrics present softness, breathability, and biodegradability, broad applicability [71,72]. At the molecular scale, the polar groups from primary cellulose not only confer exceptional hydrophilicity but also serve as versatile active sites for chemical functionalization. Structurally, cotton fibers exhibit a naturally occurring helical lumen and surface nanopores, forming a hierarchical capillary network with a special surface area. When woven into cotton fabrics, the inter-yarned macropores further established 2D pathways. The multi-scale synergistic integration of the molecular and structural features can provide an applicable carrier platform for designing the thermal sensitive material with its unique chemical structures and multi-level structural characteristics, showcasing the potential candidate for fire warning applications. Capitalizing on these advantages, diverse cotton fabrics enabled thermal alert systems to be developed. For instance, Kan et al. had coated the GO, polydimethylaminoethyl methacrylate, and hexagonal boron nitride layers on the surface of cotton fabric sequentially to assemble a functional sensor with short fire-warning time, shown in Fig. 3A [73]. Lyocell fabrics treated through electrostatic assembly of GO, polyethyleneimine, and phytic acid (PA) was utilized for the creation of FWSSs, which presented 3 s alarm trigger and durable signal transmission of

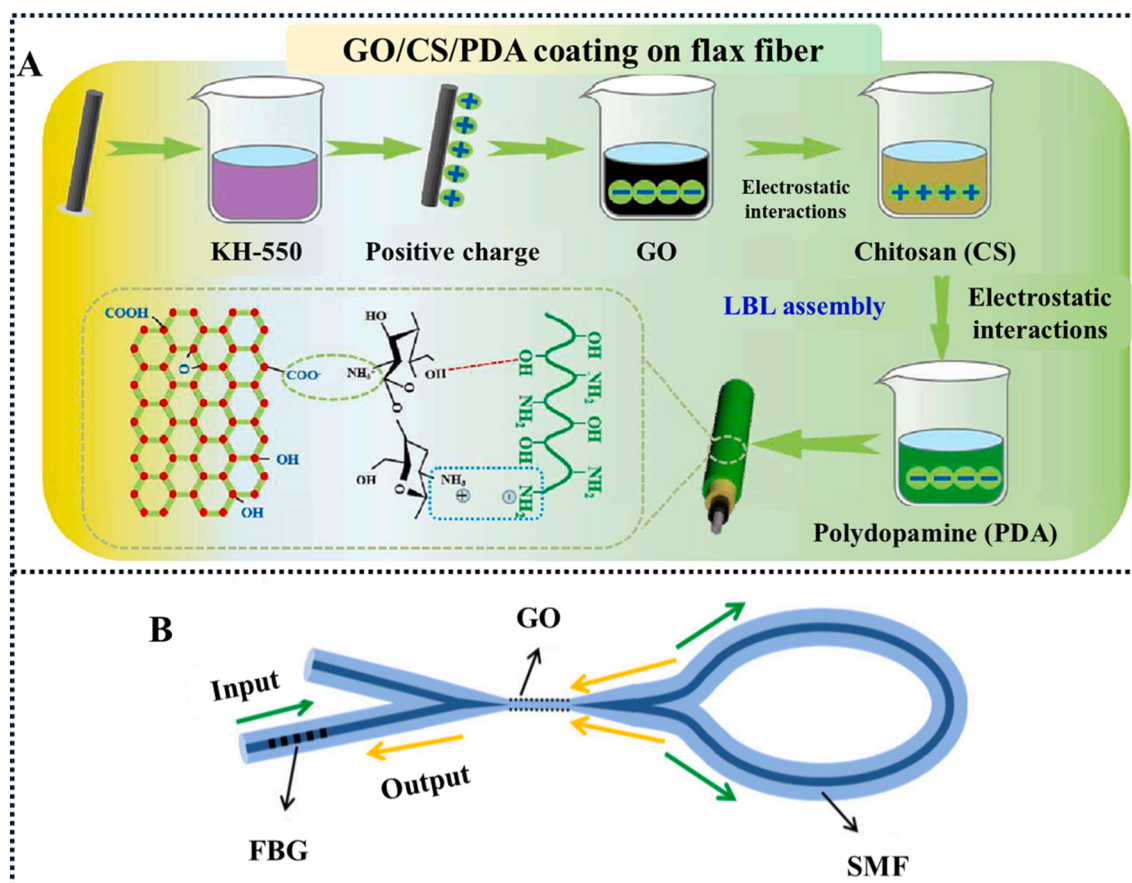


Fig. 2. A: Schematic illustration of the preparation of flax yarn-based fire-warning sensors. Reproduced with permission from Ref. [69]. Copyright 2023 Elsevier. B: Schematic loop-fiber sensing structure. Reproduced with permission from Ref. [70]. Copyright 2022 Elsevier.

>2600 s [74]. Wang et al. employed composite coating consisting of PA, CS, CNT, and GO to alter polyester/cotton blend fabrics, shortening the fire alarm trigger time to 1.5 s [53]. Moreover, a kind of hierarchical coatings were deposited on cotton fabrics, comprised of a silicone layer, GO layer, and hydrophobic layer in sequence [75]. Fang et al. prepared the composite coating composed of PA, CS, and carbon-based nanomaterials for fire-warning cotton fabrics, indicating a trigger time of 1 s and long-term alarm of 60 s [54] (Fig. 3B).

Cellulose paper is endowed with porous network that drives through hydrogen bond between cellulose fibers, rendering the high advantages for thermal sensing applications [76]. The hierarchical pore structure, arising from the interwoven cellulose fibers, endows material with both a high specific surface area and exceptional air permeability, while simultaneously serving as an optimal substrate for manufacturing the multifunctional thermo-sensitive materials. Using mature wet papermaking techniques, cellulose paper can be produced at scale, and its mechanical processability enables precise device miniaturization, which are the critical requirement for developing high-performance flexible thermal sensors. Furthermore, the abundant surface hydroxyl groups not only facilitate the grafting modification of thermo-sensitive polymers but also enhance the dispersion stability via interfacial interactions. These synergistic properties result in adequate 2D cellulose paper-based thermal sensors, driving their widespread adoption in a wide variety of sensing fields. Considering indoor fire monitoring, a MXene/GO modified fire-warning cellulose paper was provided, showing a fast response time of

2 s at 250 °C and warning message of “FIRE DANGER” to the remote computer monitor immediately [77]. To mitigate the detrimental effects of chemical modification on the flexibility of cellulose paper, poly (ethylene oxide) was incorporated to precisely modulate the flexibility. Concurrently, the integration of GO imparts fire-warning capabilities, rendering particularly suitable for house decorations as smart wallpaper for indoor fire monitoring (Fig. 3C) [26]. Apart from cellulose paper, the wood pulp paper-based fire warning study also was reported, which was modulated through phenoxycyclophosphazene-functionalized GO and chitosan-functionalized CNTs [55]. In addition, the wood pulp paper was alternately immersed into SA@phenoxycyclophosphazene modified GO suspension and the positively charged CNTs/CS suspension for a FWS with a short response time of ~0.63 s and a continuous long fire-warning time of 11,000 s [78].

The above-mentioned cotton fabric and cellulose paper, as natural substrates, were exploited for composite-engineered GO-based FWSs through leveraging the intrinsic structural properties of biocompatibility, sustainability, as well ingeniously functionality. However, there are still several challenges to further enhance their nanoscale engineering. Normally, the functional modification processing of natural polymers leads to the sacrifice of mechanical properties of materials, confining its long-term reliability in flexible applications. Additionally, the inherent thermal stability, especially at high temperatures, is not strong enough to afford durable warning. Furthermore, the large-scale production process is still not mature enough, restricting practical applications.

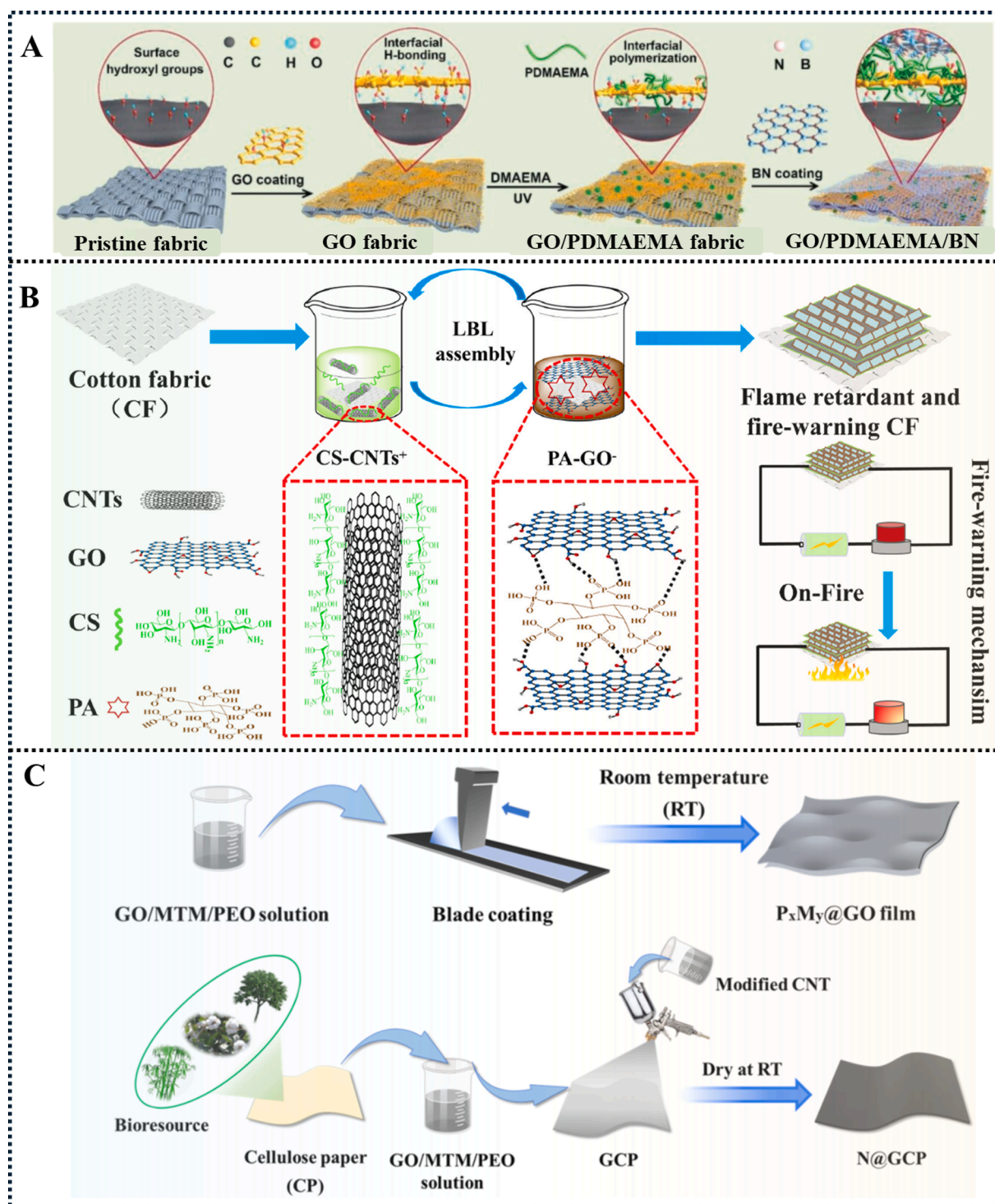


Fig. 3. **A.** The preparation of cotton fabric-based early fire-warning sensors. Reproduced with permission from Ref. [73]. Copyright 2021 Wiley-Blackwell. **B.** The graphical illustration of the preparation of fire-warning cotton fabric and its working mechanism. Reproduced with permission from Ref. [54]. Copyright 2024 Elsevier. **C.** cellulose paper-based GO type fire-warning sensor. Reproduced with permission from Ref. [26]. Copyright 2023 Elsevier.

The 2D architecture of GO nanosheets confers on inherent hydrophilicity and a negative surface charge [79–81]. Inter-sheet interactions containing electrostatic attractions, hydrogen bonding, and van der Waals forces can serve as the primary bonding mechanisms, facilitating the ordered assembly nanosheets and ultimately resulting in robust self-supporting architectures [79]. Leveraging these properties, Free-standing GO-

based FWSs show substantial potential for realizing the large-scale and flexible fire-warning application with demonstrated efficacy in advanced FWSs. As shown in Fig. 4A, He et al. provided a hydroxyapatite nanowire paper-based FWS that was modified by single-stranded deoxyribonucleic acid-modified GO, which responded to a low temperature of 159 °C, implying an advance in the low response temperature [82]. He et al. developed an

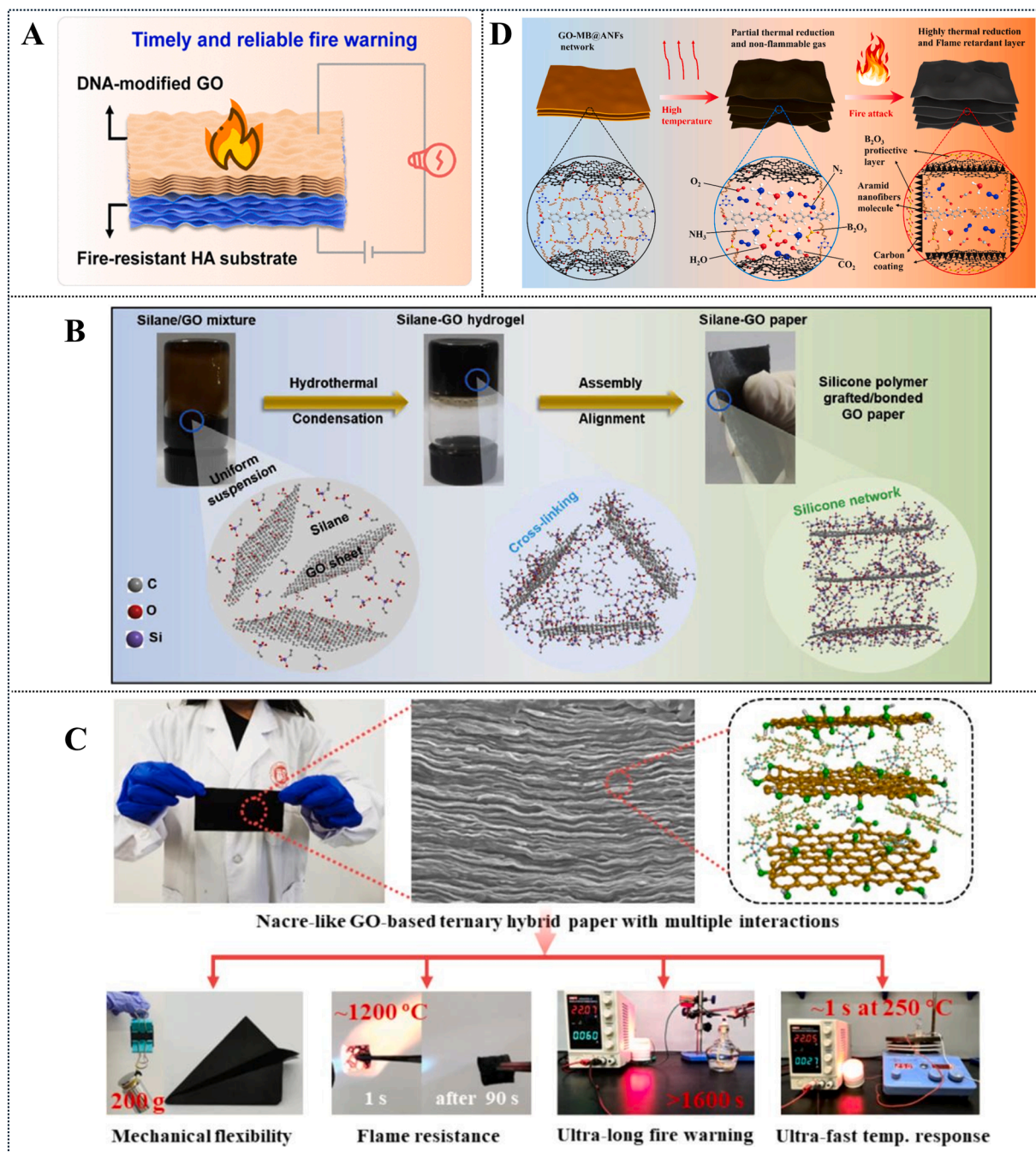


Fig. 4. **A.** The fire-warning process of hydroxyapatite nanowire paper-based sensor. Reproduced with permission from Ref. [82] Copyright 2021 American Chemical Society. **B.** Scheme demonstration of preparation for silane-GO paper and its corresponding electric resistance measurement. Reproduced with permission from Ref. [56] Copyright 2019 Elsevier. **C.** Schematic illustration of a nacre-like ternary hybrid paper (TA-GO/HHACP) based fire-warning sensor. Reproduced with permission from Ref. [57] Copyright 2024 Elsevier. **D.** Schematic representation of nano-paper based fire warning sensor including melamine diporate modified GO and ANFs. Reproduced with permission from Ref. [87] Copyright 2024 Elsevier.

asymmetric composite paper based on GO and hydroxyapatite nanowires, achieving dual-mode response with color transmission at 55 °C and fire warning at 159 °C, along with a satisfactory response time of ~1 s [83]. In addition, Yu et al. utilized water-based GO nanoribbon, montmorillonite, and polyethylene glycol to manufacture the hybrid network, which could present a rapid response time of ~2 s [46]. A kind of GO/BP-MoS₂ film modified by black phosphorus and molybdenum disulfide prepared by LBL technology showcased a response time of ~1 s under fire attack [84]. The GO-aramid nanofiber films were fabricated which

enabled the rapid fire detection of ~1 s [85]. A facile β -cyclodextrin-immobilized GO film showed ~0.15 s of fire-warning response time under the contribution of GO reduction and stable char layer through LBL technology [86]. Moreover, multifunctional acid modification is an efficient strategy for synergistically enhancing the flame retardancy of GO-based composites, such as boric acid and PA. Specifically, boric acid can enhance the thermal stability of GO nanosheets through B–O–C covalent cross-linking, forming a dense protective layer that isolates heat and hydrogen diffusion while preserving a three-dimensional conductive network.

Consequently, boric acid modified GO based fire-warning paper achieved a rapid flame response within ~ 0.8 s [44]. In contrast, PA modification further reduces the trigger time to ~ 0.5 s under the alcohol attack due to its catalytic charring [45]. In parallel, GO based hybrid papers were constructed by combining melamine diborate or co-function of 3-methacryloxypropyltrimethoxysilane and L-ascorbic acid or synthesized multi-amino molecules, respectively indicating ~ 0.72 s and the ultralong alarming time > 1200 s under a fire [59], and ~ 7 s at 300°C [47], 3 s at 300°C and 1 s at 350°C [58]. Additionally, three types of GO with tunable oxidation degrees were modified separately by polyethylene glycol and montmorillonite could respond to 300°C within ~ 3 s, 18 s, and 33 s [49]. After multi-amino modification, free-standing GO-based composite film presented an ultra-sensitive response within ~ 0.6 s, accompanied by ~ 2.3 and ~ 5.7 times of tensile strength and toughness compared to pure GO film [58]. As depicted in Fig. 4B, Huang et al. fabricated silane-GO papers with rapid response time of ~ 1.6 s under a flame condition [56]. A composite film incorporating vermiculite, gourd, and GO showed a rapid flame detection, responding to an alcohol lamp within 0.2 s while concurrently undergoing humidity-triggered angular deflection [61]. Moreover, a hybrid network fabricated by GO, phosphorylated-cellulose nanofibrils, and tannic acid (TA) molecules presented warning time < 1 s under fire, 45 s at 200°C (3 s at 350°C , 27 s at 250°C), improved tensile strength and Young's modulus as well [60]. A kind of 3-mercaptopropyltrimethoxysilane (MPTS) modified GO based MPTS-GO-10 wt% paper showed a rapid response of 1 s under a flame and 232 s at 200°C [48]. Furthermore, hexachlorophosphazene-based multi-hydroxyl molecule (HHACP) and TA were chosen to modify GO nanosheets for fire-warning composite films (Fig. 4C), exhibiting both of tensile strength of 103.7 MPa and toughness of 1.51 MJ/m^2 , response properties of ~ 1 s at 250°C and ~ 19 s at 200°C , and a durable response time of ~ 1600 s under a butane flame attack [57]. Melamine diborate was also used to modify GO, incorporated into aramid nanofibers (ANFs) for the preparation of GO-MB@ANFs nano-paper, demonstrating a rapid warning time of 0.73 s under 300°C and lasted for 1260 s, while also exhibiting the improved mechanical property and enhanced acid and alkali resistance [87]. To further shift the fire-warning temperature to a lower one, Zhu et al. regulated the GO microstructure by synthesized at low temperatures, combined with nano-graphite to fabricate LGO/G nano-paper, showcasing a response time of ~ 148 s at 150°C , significantly expanding the warning temperature range to low temperatures of 150°C [50]. Moreover, modulating the surface microstructure of GO can provide an efficient approach to lowering the fire-warning response temperatures. For instance, RGO wrapped GO based fire-warning film strip (RGOS) responded a fire with ~ 3 s for 150°C and ~ 0.5 s for a flame [88] (Fig. 4D). Polydopamine surface modified GO nanosheets combined with hydroxyapatite nanowires to prepare functional paper could further reduce the fire-warning temperature, achieving a rapid temperature at 126.9°C within 2 s and a warning signal for 5 min [89].

As an alternative strategy, functional GO-based fire-warning coatings offer promising for the fire-warning application via the surface treatment for diverse matrix materials. The functional coating consist of silicone-modified GO was one typical example to be applied onto the surface of polypropylene resin, implying the feasibility to constitute a FWS through designing fire-warning nano-coating layer [75]. Xie et al. sprayed three nanocoating on the surface of PP, which are silver nanowires/polyvinyl butyral, GO/functional cellulose, silver nanowires/fluoride polyvinyl butyral, indicating ultra-fast response to the alcohol flame within 0.83 s [68].

3.3. 3D GO-based FWSs

3D GO-based FWSs refer to utilizing GO into the interior of various 3D macroscopic network structures, like aerogels, foams, etc. through uniform dispersion and directional assembly strategies for the establishment of a continuous, interconnected and 3D temperature sensing network permeated the entire materials matrix. 3D GO-based FWSs demonstrate an advance in the real-time monitoring of 3D temperature field, especially compared with the limitations of traditional surface monitoring. Therefore, the comprehensive revise of 3D GO-based FWSs will clarify in detail in the following section.

3D sponge foam with the porous and lightweight intrinsic structures are attractive candidates for the construction of efficient thermal sensitive 3D GO-based network. The design methodology and performance evolution of different form matrix, such as melamine foam (MF), polyurethane foam (PU), aerogels, etc. in 3D GO-based FWSs, will be systematically analyzed. MF, as a representative thermosetting porous material, expresses distinctive structural characteristics owing to its special architecture. A high cross-linked 3D network formed through melamine resin foaming, displaying exceptional poriness, which is beneficial for homogeneous dispersion of functional materials. Moreover, abundant surface functional groups make the effective multifarious modification available. Notley, MF has a high nitrogen content, helping to form a protective carbon layer insulating heating and oxygen after combustion, which can guarantee the durable warning. These synergistic characteristics establish MF as an optimal substrate for advanced thermal sensitive systems. Some investigations have demonstrated that rational engineering of composite architecture can effectively utilize the inherent advantages of MF. For example, researchers have precisely adjusted the microstructure of MF by incorporating functionalized GO nanosheets, such as flame-retardant silica tailored GO (Fig. 5A) [90], aluminum oxide hydroxide (AlOOH) decorated GO (Fig. 5B) [91], different GO derivatives [92,93], CS treated GO nanosheets [52,94], attapulgite doped GO [51], etc., to optimize its architecture for efficient fire-warning applications. This multi-level modification strategy not only achieves fire-warning performance but also introduces additional multifunctional properties. The intrinsic flame retardancy of MF can synergize with adding functional coatings to enhance fire resistance, prolonging warning duration, for instance the reported continuous alarm time of 1280 s [66] and $> 1330\text{ s}^{51}$). However, there are still some issues that need to be paid attention to: 1). surface pretreatment requirements - the inert MF surface lacks active functional groups, necessitating ethanol cleaning or polyvinyleneimine activation to improve coating adhesion. 2). environmental stability - performance degradation may occur under complex conditions. 3). scalability limitations - most of current methods face difficulties in ensuring coating uniformity during large-scale production.

PU has excellent tensile strength, impact resistance, light weight, and thermal insulation effect, widely applied in diverse applications [95]. The porous structure of PU allows them to achieve different modifications. Some early fire-warning sensors based on synthetic PU materials have been employed. As shown in Fig. 5C, Chen et al. immersed PU into functional GO solution and CS-modified CNTs to prepare an early fire-warning sensor, which presented the sensitive fire-warning property [96]. Another sandwich-like PU based FWSs was modified by PA, flame retardant copolymer, GO, CNTs, and boron nitride. This sensor responded to abnormal high temperature within ~ 8 s, indicating its high thermos-sensitivity capability [97]. There reported one kind of biological carbon-nitrogen-phosphorous multifunctional PU

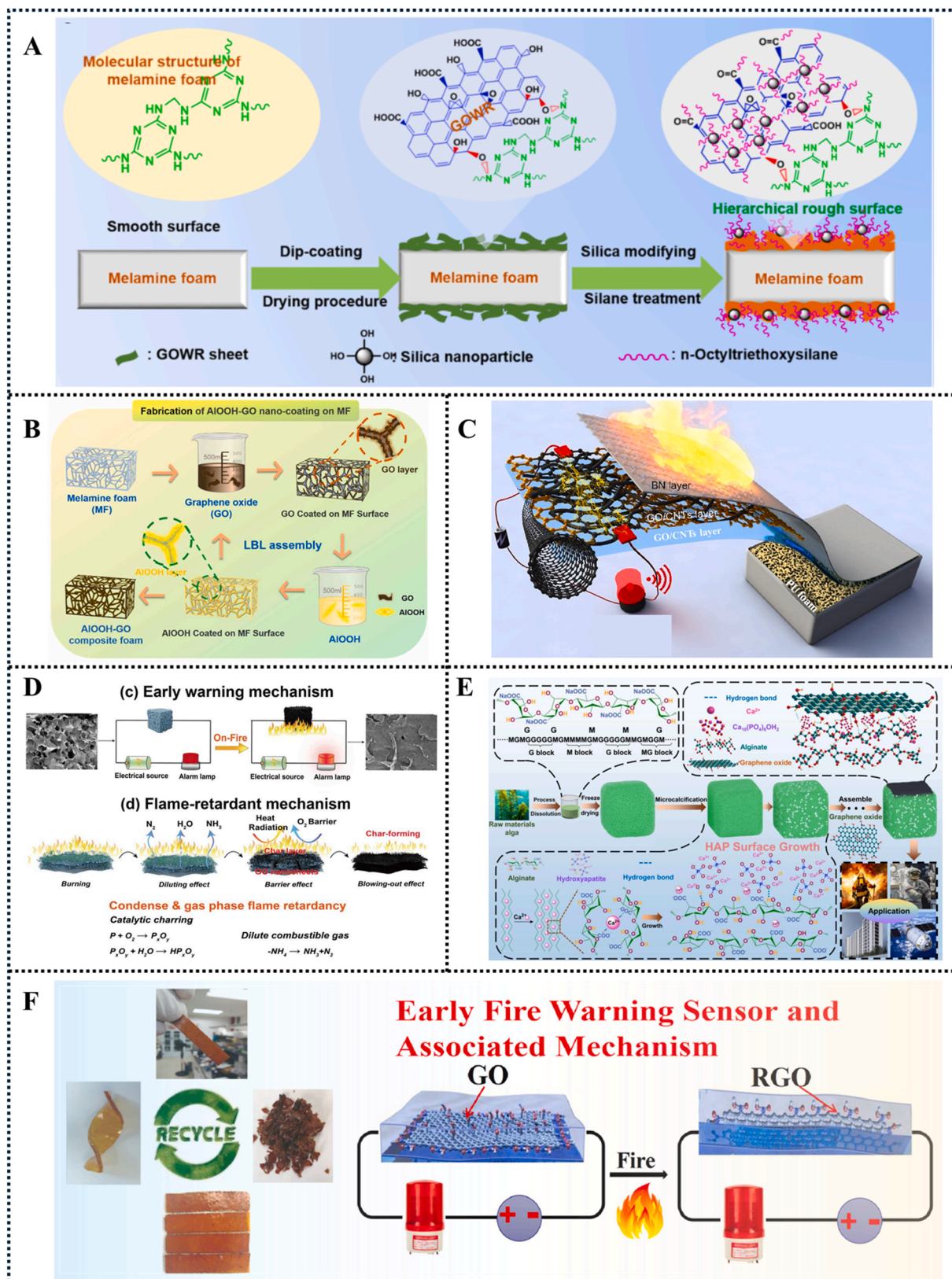


Fig. 5. A. Graphical scheme for the fabrication of the functionalized silica/GOWR coated MF (M-GOWR@MF) sponge. Reproduced with permission from Ref. [90] Copyright 2021 Elsevier. B. Fabrication of AlOOH-GO layer wrapped MF. Reproduced with permission from Ref. [91] Copyright 2022 Elsevier. C. Sandwich-like PU foam-based fire-warning foam. Reproduced with permission from Ref. [106] Copyright 2022 Elsevier. D. Graphical scheme for a porous structure containing CNF, APP, and GO as fire-warning sensor. Reproduced with permission from Ref. [66] Copyright 2024 Elsevier. E. Schematic diagram for the SA/hydroxyapatite/GO-based aerogel as a fire-warning sensor. Reproduced with permission from Ref. [101] Copyright 2024 Elsevier. F. Recyclable bio-based epoxy resin/GO type early fire-warning sensors. Reproduced with permission from Ref. [105]. Copyright 2024 Elsevier.

through the modification of fire-safety adhesive and cyclodextrin modulated GO, which exhibited response time of 5.3 s and long fire-warning durability of >3600 s [98]. Besides, the ammonium polyphosphate (APP)/GO hybrid layer was coated on the PU surface for modified PU-based sensor, sending warning within ~2 s when a fire occurred or ~11.2 s when the temperature of sensor reached to 300 °C [99]. In addition, GO/silicone layered structure was also introduced to modify PU matrix for a fire-warning sensor, demonstrating the monitored alarm to the public [75]. These above-mentioned PU based sensors activated the lamps as transmitted signals to alter people. In the informative era, wireless conversion was also operated to send detected warning messages. For instance, the detected dangerous information was transferred to remote computer monitor in the phosphorus/silicon-modified GO layer wrapped PU-based fire-warning sensor [100]. The cooperation with smart wireless signal transmission facilitates conveying detected information in ultra-short time, improving the fire-warning sensitivity. Zeng's research group manufactured a kind of fire-warning mixture solution, which could be coated on PU for PU-GO/FC-40 fire-warning sensor, showing a rapid response time of ~3.4 s. Besides, this functional solution could also be coated on the surface of wood and polypropylene to make wood-GO/FC-40 and PP-GO/FC-40 fire-warning sensors, exhibiting a response time of ~2.9 s and 3.2 s [63].

With the exception of MF and PU used as 3D carriers, aerogels formed by the assembly of GO itself represents an intrinsic structural-functional integrity in 3D network construction. This type of aerogels composed of the inherent GO itself not merely inherits the porous and lightweight characteristics but achieves the continuous and uniform distribution of core component in 3D space as well, which can facilitate the deep exploitation of the structure-property relationship of 3D GO-based FWSs. As shown in Fig. 5D, Zhou et al. employed cellulose nanofiber (CNF), APP and GO to prepared fire-warning aerogel, showcasing the response time of ~1.32 s and the sustaining response time of ~170 s [66]. By using in-situ growth and directional assembly technology, SA/hydroxyapatite/GO composite aerogel with an intelligent “bionic armor” was established (Fig. 5E), presenting multifunctionality including a fast warning of ~1.95 s, long water resistance, and thermal insulation [101]. Yuan et al. fabricated a biomass-based aerogel comprised of GO and montmorillonite modified CNF, which was triggered in ~1.9 s and continue warning time of 137 s [102]. In addition, another biomass GO-based composite aerogel integrated GO with sodium carboxymethyl cellulose (CMC), SA, polyaniline (PANI) for functional fire-warning aerogel indicated rapid fire response within 1.3 s, in combination with thermal insulation and mechanical robustness [103]. He et al. provided a kind of composite aerogel composed of GO, MXene and CS via freeze-drying, which presented an ultra-fast fire warning response within 1 s and a sustained alarm lasting over 120 s, as well as their self-powered fire warning functionality [104].

Unlike the static porous structure of aerogel, the dynamic cross-linked network of hydrogels exhibits active thermal response capabilities. An instance of GO-based hydrogel was prepared by 3D printing technology, which showed a rapid response to a fire [67]. Meanwhile, Jia et al. introduced GO into the recyclable bio-based epoxy resin to form 3D fire-warning network, which was triggered within ~10 s under ignition treatment. It kept fire warning even after the removal of fire because a conductive pathway was already formed under the fire attack (Fig. 5F) [105].

In a nutshell, the typical characteristics of temperature perception in different dimensions was comprehensively analyzed by sorting out the relevant research on 1D, 2D, and 3D GO-based FWSs, thereby deeply reveal the essential differences in perception ability among different dimensions. To this end, it is necessary

to simultaneously optimize the flame-retardant functionalization of matrix and interfacial engineering between functional nano-coatings and polymeric substrates to accomplish target fire-warning performance. In particular, the operational temperature range accuracy and fire-warning endurance, as pivotal performance metrics, will subsequently undergo detailed analysis to help to establish structure-property correlations in the following section.

4. Comparison of fire-warning performance

4.1. Fire-warning sensitivity

Fire-warning sensitivity is a critical performance metric for thermal responsive materials, directly governing the ability of a system to detect early-stage fires or abnormal thermal excursions. High sensitivity enables not only precise recognition of minor temperature fluctuations but also minimizes false alarms, significantly improving the reliability of FWSs. As above-mentioned, researchers typically engineer material microstructures or modify functional compositions to achieve optimal fire-warning sensitivity of FWSs. Specifically, the modification is intrinsically linked to the modification of three key parameters, like shortening fire-warning time, lowering fire-warning temperature, and enhancing fire-warning durability, which systematically optimizes the fire-warning performance.

Fire-warning time are the interval time quantified from when the ambient temperature exceeds the thermal anomaly threshold to when the warning sensor unit of FWSs generates an effective alarm signal. Its core value lies in obtaining a key intervention time window for emergency response, the analysis of which mainly employed to evaluate the warning timeliness and early intervention of potential fire hazards, that is, whether the system can trigger the early warning and initiate intervention measures before the fire risk reaches the critical point based on the synergistic effect of thermal sensing property and wireless signal transmission. For the GO-based fire-warning study, their fire warning time is mainly regulated by the thermal response sensitivity of GO and signal transmission efficiency. As the above-mentioned composition-engineered GO-based thermal FWSs, they can achieve a rapid warning time within seconds under different thermal field conditions, such as ignition guns, alcohol lamps, ovens, etc. From the perspective of materials, the thermal reduction kinetics characteristics of GO directly affect the response speed. Accelerating the thermal reduction process of GO can significantly shorten the fire-warning time, thereby broadening the available intervention time window. A typical strategy is to precisely control the thermal reduction dynamics by tuning the carbon-oxygen ratio to gain a thorough understanding of the structural evolution of GO during the thermal attack, resulting in FWSs with adjustable response characteristics directionally [50,88]. In terms of wireless signal transmission, the rate of signal conversion is strongly related to accuracy of warning and real-time feedback of monitoring information. In sum, accelerating the thermal response sensitivity of GO and optimization of wireless signal transmission efficiency are directly accolated with the efficiency of personnel evacuation and potential fire suppression.

Fire warning temperature specifically represents the critical thermal threshold for alarm activation. This parameter must accurately reflect how ambient thermal conditions influence both the ignition probability of combustible materials and subsequent fire propagation rates. From the viewpoint of materials science, fire-warning temperature, as the core judgment parameter of the thermal warning system, is intrinsically linked to the thermodynamic response characteristics of GO, specifically its thermal

reduction temperature range. In the study of GO-based fire warning sensors, the thermal response behavior of GO mainly depends on the thermal stability and spatial distribution characteristics of its oxygen-containing functional groups (e.g., hydroxyl, epoxy and carboxyl groups). Thus, the reasonable regulation of synthetic optimization and post-processing of GO become two core strategies to design high-performance FWSs [107]. For example, GO with tunable decreased oxygen-containing functional groups were synthesized under low temperature, which indicated reduced initial thermal response threshold and improved low-temperature response performance because of the low-temperature thermal decomposition [50]. While a kind of GO/MXene/CS aerogel presented an accelerated response time of 1 s because of the optimized thermal conductive pathway of GO contributed from the reducibility and conductivity of MXene nanosheets [104]. Moreover, studies have confirmed that the critical temperature threshold for GO typically ranges between 120 and 200 °C [108], which is accompanied by the gradual dissociation of oxygen-containing functional groups and the restoration of sp^2 carbon network. This structural evolution results in an order of magnitude conductivity enhancement, thereby establishing a fundamental physical basis for defining the fire-warning temperature threshold. Theoretical analysis establishes that GO exhibits an intrinsic fire warning temperature range of 120–200 °C. However, most of current GO-based FWSs consistently employ activation thresholds exceeding 200 °C, presenting the lowest fire-warning temperature of 126 °C (Fig. 6). Even optimized low-temperature configurations reported fail to achieve an operation lower 120 °C. This systematic threshold elevation stems from the complex practical constraints, such as thermal stability requirements (slow reduction kinetics below 200 °C degrade signal-to-noise ratios), environmental interference suppression, ignition matching of combustible materials. More inspiring, in the forest fire monitoring, the integration electrical resistance change and temperature response with a specific machine learning model efficiently improved the recognition accuracy and reliability of fire warning, offering a clear direction for subsequent research [109].

Fire-warning durability quantifies the sustained duration of activated alarm signals, in essence reflecting the structural stability of materials under thermal exposure. This stability manifests

specifically through the maintenance of conductive pathways at triggered temperatures. Crucially, fire warning persistence exhibits synergistic correlation with flame retardancy and the degradation kinetics of materials. When polymer substrates with nano-fillers undergo chain scission or oxidative decomposition under thermal radiation, the resulting disintegration of electron transport networks terminates warning signals. In state-of-the-art composition-engineered GO-based FWSs, flame-retardant modification is a core approach to enhance the fire-warning durability through the key mechanism of substrates protection and regulation of thermal reduction kinetics. To illustrate, the supermolecule melamine diborate was integrated into GO to improve fire resistance of GO-based FWS because of the formation of protective layer under fire exposure, accompanied by the positive role in fire-warning optimization [59]. Another case in point is the flame-retardant treatment of GO aerogel by carboxylated cellulose nanofibers (CCNF) and sodium-based montmorillonite (Na-MMT), which significantly increased fire-warning durability to 300 s due to the catalytic carbonization and barrier effects [110]. Currently, operational durability of composite-engineered GO-based FWSs maintains functional integrity for 1600 s, demonstrating exceptional thermos-structural stability - an essential property enabling continuous surveillance of multi-stage fire evolution dynamics (Fig. 7). Notably, thermal energy density influences structural stability of materials: high energy fluxes can accelerate structural collapse, whereas low energy fluxes prolong functional integrity by retarding decomposition processing. It is worth emphasizing that the reported fire-warning durability cannot be directly comparable due to the different test conditions and the lack of standardization assessment criteria. Therefore, any cross-system comparison should be cautiously made, and the potential biases caused by different experimental setups and performance measurement approaches also needs to be fully taken into account.

Comprehensive analysis of composition-engineered GO-based FWSs that comprised of fire warning time, fire warning temperature, and fire warning durability are clarified. The fire warning time at the second level is regulated by the synergistic interaction between the thermal reduction kinetics of GO and signal transmission efficiency. Concurrently, fire warning temperature is constrained by competing thermodynamic factors. Furthermore, fire-warning durability affects by the exceptional structural

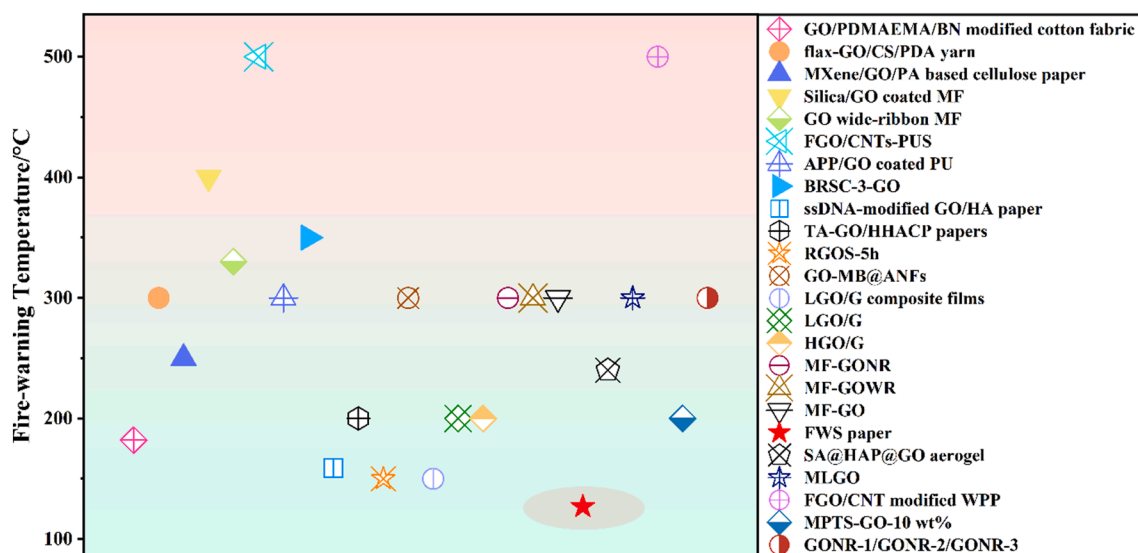


Fig. 6. Summarized fire-warning temperatures of composition-engineered GO-based FWSs.

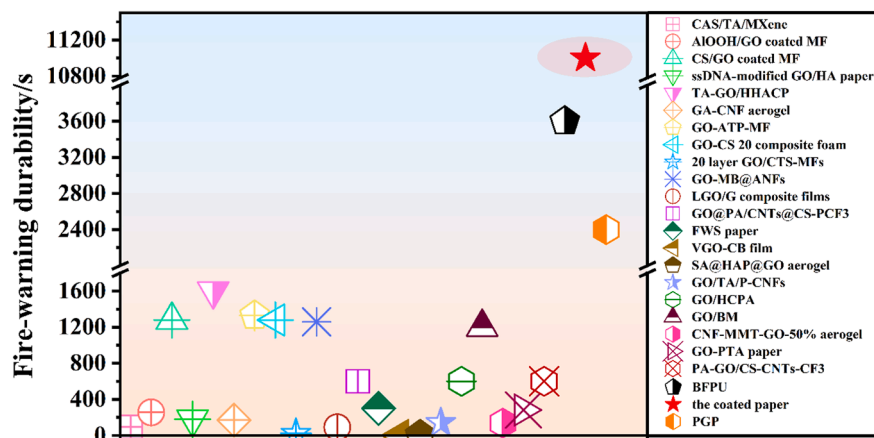


Fig. 7. Summarized fire-warning durability of composition-engineered GO-based FWSS.

integrity under thermal radiation. These not only establish the superiority of GO in fire monitoring applications but also implies the importance of the flame retardancy. Consequently, the following section dissects the flame-retardant mechanism.

4.2. Fire-warning signal transmission

Ultra-fast fire-warning mainly depends on the precise perception of abnormal thermal state and issuing timely and reliable alerts when the present danger threshold is reached. These two are closely related to the strong thermal sensitivity of materials themselves and the efficient analysis capabilities and transmission of the captured signal, respectively. In terms of intelligent signal sensing, intelligent sensing units can not only highly convert the physical quantity of temperatures into quantifiable and easily transmissible electrical signals, such as electrical signals, optical signals or other quantifiable signals, *etc.* Notley, they integrate cutting-edge technologies such as micro-nano processing and intelligent algorithms endowing with sensing system with “intelligent” characteristics. Therefore, the development level of intelligent signal sensing technology directly determines the fire-warning sensitivity, response speed and reliability. It can serve as a key bridge connecting the physical response of materials and actual output of warning signals, and core engine driving abnormal temperature to shift from passive perception to active, intelligent and precise early warning.

In the research of GO-based fire warning investigation, intelligent signal conversion plays an indispensable and increasing core role as well. Thus, it is necessary to integrate the transducer (thermos-sensitive materials), electronics, and communication parts into a sensor unit. For sensing, resistance changes are typically calibrated via four wire method to quantify conductivity shifts, as reported by literatures [93,111]. In this setup [111], a power supply applies current through two drive electrodes positioned at a fixed distance across the materials. Two separate sensing electrodes measure the resultant voltage drop. When temperature increasing, conductivity rises, enabling current flow along the inter-electrode pathway. This electrical current activates the communication system connected to the sensing electrodes. The communication system employs an XLPCF20 transmitter to send an activation signal to an R080A receiver. This receiver powers three components: (1) a sound alarm, (2) an Arduino UNO driving a “FIRE DANGER” display interface, and (3) an MKRWAN1300 LoRa emitter. The LoRa emitter transmits warnings to the internet via a RAK concentrator (connected through Wi-Fi/

Ethernet). Transmission distances between the fire source and LoRa receivers reach up to 2 km in urban areas and 20 km in rural, interference-free environments under line-of-sight conditions. While other research system employing the four-wire resistance way support local alarms but exclude remote notification functionality [50,93,112].

4.2.1. Local fire-warning signal conversion

Local signal transmission encompasses the technique process wherein raw temperature signals are processed in real-time and used for generating early fire warnings, all within the sensor nodes or their directly interfaced embedded units. The core features of this mode are the localization and decentralization of its data processing capabilities. This architecture utilizes micro-processing units (MCUs/MPUs), which are integrated directly into the sensor modules or deployed in their immediate vicinity. Critically, these units are equipped with standalone capabilities to perform signal conditioning, analog-to-digital conversion (ADC), initial data analysis, and threshold judgment. The monitoring process is initiated by the acquisition of physical temperature signals via sensing elements (GO-based thermosensitive materials [46,50,100]), which transduce the thermal input into analog electrical signals. Subsequently, these raw signals undergo local processing including amplification and filtering to improve the signal-to-noise ratio prior to further analysis. On this basis, the embedded processor performs extraction and analysis of key parameters, such as instantaneous values, change rates, and average, autonomously initiates local warning actions, containing audible or visual alarms, in accordance with predefined safety threshold logic. Local signal conversion demonstrates the exceptional low latency and high operational reliability, leading to the eliminating the need for external network connectivity. This approach effectively mitigates potential issues related to width constraints, transmission interruptions, and network delays associated with remote data communication. Consequently, the system guarantees instantaneous response capability even under critical conditions, making it particularly suited for applications with stringent real-time requirements - such as industrial safety interlocking systems and the protection of critical equipment.

4.2.2. Remote fire-warning signal conversion

Wireless signal transmission is a key link in building a remote centralized monitoring and wide-area early FWSS. Its main function is to reliably transmit the characteristic data or alarm information preprocessed locally by edge nodes to the remote gateway

or cloud platform. This process is usually implemented relying on low-power wide-area (LPWA) wireless communication technologies (such as LoRaWAN, NB-IoT) or cellular networks (such as 4G/5G) [85]. Compared with local signal transmission limited to certain areas, wireless transmission demonstrates significant advantages in achieving remote real-time monitoring and large-scale networking, and has broader practical application value. The practicality and effectiveness of wireless transmission technology have been verified in multiple studies. For instance, a typical work built a fire early warning prototype system based on microcontroller and Bluetooth communication technology: when the sensing resistance exceeded the preset threshold, the embedded Arduino microcontroller would trigger a fire signal and send it wirelessly to a remote smart device [51]. This system has achieved a reliable long-distance connection between the fire scene and the monitoring terminal, fully demonstrating the significant role of wireless transmission in real-time monitoring and immediate early warning.

Nonetheless, wireless signal transmission faces numerous challenges in complex physical environments. The core issue lies in how to achieve coordinated optimization among transmission efficiency, energy consumption and reliability. Due to the inherent openness of wireless channels, signals are vulnerable to factors such as multipath fading, obstruction by obstacles, and co-frequency interference. For this reason, it is often necessary to introduce mechanisms such as forward error Correction (FEC) and Automatic Retransmission Request (ARQ) to ensure the integrity of data and the reliability of transmission. Meanwhile, to meet the demand for the long-term stable deployment of fire monitoring terminals, the communication protocol stack needs to be deeply optimized, and power-saving technologies such as PSM (Power Saving Mode) and eDRX (extended Discontinuous Reception) should be adopted to greatly reduce the power consumption of the equipment.

To recapitulate, study about composition-engineered GO-based FWSs have been made some remarkable progress in key performance parameters through various modification strategies. As summarized in Table 1, these advancements are mainly reflected in the short fire warning time, progressively lower trigger temperatures, prolong alarm durability, and intelligent signal conversion, etc. Some fire warning systems also present the potential multi-functional integration. Nevertheless, the current investigation is still confronted with obstacles on the developing road ahead. Clarifying the underlying flame-retardant behavior (carbonization process, the thermal reduction kinetics of GO, etc.) and fire-warning mechanism (the evolution of the electronic conduction pathway between GO nanosheets, or at the interface, etc.) can provide a theoretical foundation for performance optimization of fire warning materials.

5. Underlying mechanisms of composite-engineered GO-based FWSs

5.1. Synergistic flame-retardant mechanisms

The desirable flame retardancy of FWSs is a critical prerequisite for the establishment of the stable conductive pathway to realize alert in composition-engineered FWSs. Unfortunately, the intrinsic flame retardancy of pure GO is difficult to independently satisfy the need of keeping structural integrity and functional stability of FWSs in practical applications. Therefore, most of current researches generally employ a synergistic flame-retardant strategy to optimize fire resistance behavior. To systematically reveal the essence of the synergistic influence between GO and the compounded flame retardants, the classification including synergistic

solid-phase and gas-phase flame-retardant mechanisms is carried out for the in-depth understanding, which is detailed elaborated as follows.

5.1.1. Synergistic solid-phase flame-retardant mechanism

Upon heat exposure, GO nanosheets in composite-engineered GO-based FWSs experience the thermal decomposition to construct a protective char layer with the graphitized structure through the deoxidation and carbonization reactions [57,58,60]. The formed char layer can serve as the physical barrier to impede the transfer of heat, oxygen, and flammable volatiles, thereby disrupting the fundamental fire triangle (heat, fuel, oxygen) to suppress combustion [90]. However, as result of the limited pyrolysis process of GO nanosheets, the char layer normally is not porous, insufficient continue, low-density enough to support satisfactory sufficient mechanical strength and efficient isolation effect to accomplishment durable and effective substrate protection. Encouragingly, the synergistic solid-phase flame retardancy is one highly effective strategy to enhance fire resistance, which mainly systematically regulate and strengthen the structure of char layer through multi-functional modification of GO involving the enhancement mechanism of char layer construction, heat conduction regulation mechanism, and catalytic synergistic char formation mechanism, as shown in Fig. 8. Specifically, the introduction of materials such as APP [66,99], Si-containing molecules [48,75,83,90,100], can catalyze the dehydration of matrix to form char during pyrolysis for the establishment of a more stable and high cross-linking char structure to significantly improve the density and integrity of the char layer [74,115]. Moreover, the treatment of nano-materials like CNT, BN, etc. cannot only help to build a three-dimensional network structure, but also effectively delays heat conduction to reduce heat transfer efficiency, thereby enhancing physical barrier efficacy [73,96]. Additionally, the incorporation of nitrogen (N)-containing [74], phosphorous (P)-containing [57,78,84], oxygen-containing molecules [87] (like TA, DNA [82], etc.) into FWSs make GO and the matrix undergo catalytic synergistic reactions during pyrolysis, promoting the transformation from a loose and porous char layer to a continuous, dense, and more graphitized “catalytically enhanced physical barrier” [52,94]. The better physical barrier with superior antioxidant capacity and thermal stability can more effectively prevent the escape of flammable gases and penetration of external thermal oxygen, which significantly enhance the flame-retardant performance.

In short, the synergistic solid-phase mechanism of composite-engineered GO-based FWSs is advantageous to the transformation from imperfect physical barrier to multiple physical barriers protection by the directional modification and multi-component design, providing solid theoretical guidance for the development of highly efficient GO-based FWSs.

5.1.2. Synergistic gas-phase flame-retardant mechanism

The integration of gas-phase flame-retardant components into GO is another available strategy, which can enhance flame-retardant performance of FWSs comprehensively across multiple scales and mechanistic levels. In brief, the core of this strategy primarily depends on the addition of gas-phase flame-retardant units comprised of N, P, halogens elements into GO through the covalent or non-covalent bonds, which go through the decomposition to directionally release highly-active free radical scavengers such as $\text{PO}\cdot$ and $\text{PO}_2\cdot$, etc. [57,66,78,84], as well as non-flammable gases [57,66,69,77,82,98,100,115], like NH_3 [87,93], sulfur dioxide [106], CO_2 [99,115], H_2O [51,91], etc. when subjected to thermal shock (Fig. 9). Theoretically, free radical scavengers remarkably improve the capture efficiency of $\text{H}\cdot$ and $\text{HO}\cdot$ free radicals in the

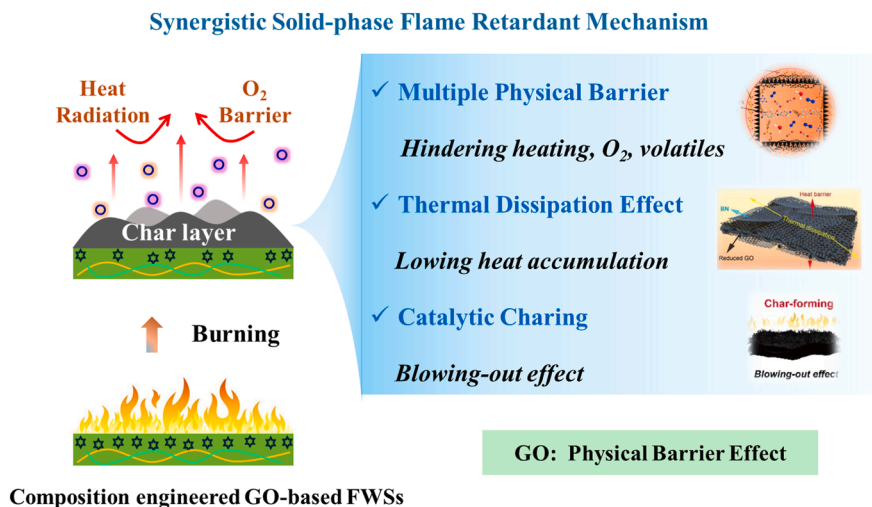
Table 1
Detail summary of composition engineered GO-based FWs.

Samples	Triggering temperature	Response Time	Duration time	Signal conversion type	Multi-functions	Ref.
GO/PDMAEMA/BN modified cotton fabric	182 °C	<3 s	–	Warning lamp	Flexibility, large-scale fabrication, washability	[73]
Flax-GO/CS/PDA yarn	<300 °C	~3 s at 250 °C	–	Alarm lamp and wireless remote warning	–	[69]
MXene/GO/PA modified cellulose paper	250 °C	2 s	–	Wireless local and remote warning	–	[77]
GO/silicone wrapped	Fire	2–3 s	–	Warning lamp	–	[75]
CAS/TA/MXene mixture	Fire	2–3 s	93 s	Warning lamp	Good mechanical flexibility; repeated flame-triggered conversion	[113]
Silica/GO coated MF	400 °C	5 s	–	Warning lamp	Flexibility, hydrophobicity	[90]
AlOOH/GO coated MF	Flame	2.5 s	256 s	Warning lamp	Hydrophobicity	[91]
CS/GO coated MF	Flame	3 s	1280 s	Warning lamp	–	[94]
GO wide-ribbon MF sponges	330 °C	33 s	–	Warning lamp	Lightweight, good hydrophobicity, reversible compressibility	[93]
FGO/CNTs-PUS	500 °C	2 s	–	Warning lamp	–	[96]
PA/PVH/GO/CNTs/BN coated PU	Fire	~8 s	–	Warning lamp	–	[106]
APP/GO coated PU	300 °C	11.2 s	–	Warning lamp	Flexibility, super-hydrophobicity	[99]
P/Si-decorated GO coated PU	Fire	~3 s	–	Warning lamp and wireless signal conversion	–	[100]
BRSC-3-GO	350 °C	~20 s	–	Warning lamp	–	[105]
ssDNA-modified GO/HA paper	159 °C	–	180 s	Warning lamp	–	[82]
TA-GO/HHACP papers	200 °C/250 °C	19 s/1 s	~1600 s	Warning lamp	Tensile strength	[57]
GA-CNF aerogel	Alcohol flame	1.32 s	170 s	Warning lamp	Superior rheological properties	[66]
GO-ATP-MF composite	Alcohol flame	<1 s	>1330 s	Warning lamp	Bluetooth wireless transmission	[51]
GO-CS 20 composite foam	Alcohol flame	3 s	1280 s	Warning lamp	–	[94]
RGOS-5h	Flame/150 °C	0.5 s/3 s	–	Warning lamp	–	[88]
20 layer GO/CTS-MFs	Flame	~4 s	22 s	Warning lamp	Self-power, nanogenerator	[52]
GO-MB@ANFs nanopaper	Alcohol flame/300 °C	0.73 s/-	120 s/1260 s	Warning lamp	Improved mechanical property, solvent resistance	[87]
LGO/G composite films	150 °C/flame	148 ± 45 s/1 s	~90 s	Warning lamp	–	[50]
LGO/G composite films	200 °C	74 ± 3 s	–	Warning lamp	–	[50]
HGO/G composite films	200 °C	385 ± 30 s	–	Warning lamp	–	[50]
MF-GONR	Alcohol flame/300 °C	2.1 s/20 s	–	Warning lamp	Hydrophobicity, enhanced maximum compressive stress	[92]
MF-GOWR	Alcohol flame/300 °C	2.1 s/34 s	–	Warning lamp	Hydrophobicity, enhanced maximum compressive stress	[92]
MF-GO	Alcohol flame/300 °C	2.4 s/40 s	–	Warning lamp	Hydrophobicity, enhanced maximum compressive stress	[92]
GO@PA/CNTs@CS-PCF3	Alcohol flame	1.5 s	600 s	Warning lamp	Flame retardancy	[53]
FAW paper	126.9 °C	2 s	300 s	Alarm lamp, alarm buzzer	Dyeability, foldability	[89]
VGO-CB film	Alcohol flame	0.2 s	1 s	Warning lamp	Solvent response, self-driven and smart switching	[61]
SCAA yarn	–	–	–	–	Stretchability, flame retardancy	[24]
SA@HAP@GO aerogel	Alcohol flame/240 °C	1.95 s/3.14 s	6.95 s/40 s	Warning lamp	High strength, water resistance, thermal insulation	[101]
GO/TA/P-CNFs hybrid network	Fire/350 °C/250 °C/200 °C	<1 s/3 s/27 s/45 s	140 s under fires	Warning lamp	Improved tensile strength and Young's modules	[60]
MPMS-GO film	Flame	~1.6 s	–	An alarm light	Mechanical flexibility, strong acidic/alkaline tolerance, flame resistance	[56]
GO/HCPA network	Alcohol flame	~0.6 s	>600 s	Warning lamp	Increased tensile strength and toughness	[58]
GO-BA paper	Flame	0.8 s	–	Warning lamp	–	[44]
PMG paper	Alcohol flame	2 s	–	Warning lamp	Improved flame retardancy	[46]
GO/BM hybrid paper	Flame	0.72 s	>1200 s	Warning lamp	Enhanced mechanical property, acidic/alkaline tolerance	[59]
CNF-MMT-GO-50 % aerogel	Fire	1.9 s	137 s	Warning lamp	–	[102]
MLGO film	300 °C	7 s	–	Warning lamp	–	[47]
GO-PTA paper	Fire	~0.5 s	283 s	Warning lamp	–	[45]
Wood-GO/FC-40	Alcohol flame	2.9 s	–	Warning lamp	Self-healing	[63]
PP-GO/FC-40	Alcohol flame	3.2 s	–	Warning lamp	Self-healing	[63]
PU-GO/FC-40	Alcohol flame	3.4 s	–	Warning lamp	Self-healing	[63]
PA-GO/CS-CNTs-CF3	Alcohol flame	1 s	600 s	Warning lamp	Flame retardancy	[54]
FGO/CNT modified WPP paper	Fire/500 °C	5 s/7 s	–	Warning lamp	Flame retardancy	[55]
SFR coated PP	Alcohol flame	0.83 s	–	Warning lamp	Flame retardancy, self-cleaning, water resistance	[68]
BP-MoS ₂ /GO composite films	Flame	~1 s	–	Warning lamp	Flame retardancy	[84]
MPTS-GO-10 wt% paper	Flame/200 °C	~1 s/232 s	–	Warning lamp	Flame retardancy	[48]
GONR-1/GONR-2/GONR-3	300 °C	3 s/18 s/33 s	–	Warning lamp	Flame retardancy	[49]
BFPU	Alcohol flame	5.3 s	>3600 s	Warning lamp	Flame retardancy, mechanical property	[98]
The coated paper	Alcohol flame	0.63 s	11,000 s	Warning lamp	High tensile strength, electrical conductivity	[78]
GO-ANF	Alcohol flame	<1 s	–	Warning lamp	Wireless alerts	[85]

Table 1 (continued)

Samples	Triggering temperature	Response Time	Duration time	Signal conversion type	Multi-functions	Ref.
PGP	Fire	2 s	2400 s	Warning lamp	Enhanced tensile strength toughness	[114]
GO/PEI/HAP paper	Alcohol flame	8 s	26 s	Warning lamp	Flame retardancy	[115]
CF8	Alcohol flame	3 s	>2600 s	Warning lamp	Flame retardancy	[74]
CSGP aerogel	Alcohol flame	1.3 s	–	Warning lamp	Thermal insulation, mechanical robustness	[103]
GO/MXene/CS aerogel	Alcohol flame	1 s	120 s	Warning lamp	Self-power	[104]
CIGO	Alcohol flame	0.15 s	–	Warning lamp	–	[86]

PDMAEMA: Polydimethylaminoethyl methacrylate; BN: hexagonal boron nitride; CS: Chitosan; PDA: Polydopamine; PA: Phytic acid; CAS: casein; TA: Natural tannic acid; MF: melamine sponge; AlOOH: Aluminum hydroxide; FGO: Flame retardant-modified GO; CNTs: chitosan-functionalized carbon nanotubes; PU: polyurethane foam; PVH: flame retardant copolymer; BN: boron nitride; BRSC: flame-retardant epoxy resin; DNA: The single-stranded deoxyribonucleic acid; HA: hydro-yapatite nanowire paper; HHACP: hexachlorophosphazene-based multi-hydroxyl molecule; GA: GO modified by APP; CNF: cellulose nanofiber; ATP: attapulgit; MF: melamine foam; RGOS-5h: a surface reduced graphene oxide strip; GO/CTS-MFs: graphene oxide/chitosan-modified melamine foams; MB: melamine diborate; ANFs: aramid nanofibers; LGO: low-temperature synthesized graphene oxide; G: nano-graphite paste; GONR: graphene oxide nano-ribbon; GOWR: graphene oxide wide-ribbon; PA: phytic acid; CS: chitosan; CNTs: carbon nanotubes; PCF: polyester/cotton blend fabrics; FAW paper: ultralong hydroxyapatite nanowire/glass fiber fire-resistant inorganic paper; VGO-CB film: an layer nano-material developed by vermiculite with cucurbituril and GO; SCAA yarn: spandex/CNT@Aramid/Aramid yarn; SA@HAP@GO aerogel: sodium alginate/hydroxyapatite/graphene oxide aerogel; TA: tannic acid molecules; P-CNFs: phosphorylated-cellulose nanofibrils; MPMS: 3-methacryloxypropyltrimethoxysilane; HCPA: the multi-amino molecule; BA: boric acid; PMG: GO nanoribbon/montmorillonite/polyethylene glycol hybrid networks; BM: melamine diborate; CNF: cellulose nanofiber; MMT: montmorillonite; MLGO film: GO-based paper modified by 3-methacryloxypropyltrimethoxysilane and L-ascorbic acid (LAA); PTA: phytic acid; FC: functional cellulose; PP: polypropylene; PA-GO: phytic acid-graphene oxide dispersion; CS-CNTs: chitosan-carbon nanotube; CF3: cotton fabric; FGO: Phenoxycyclophosphazene-functionalized GO; WPP: wood pulp paper; SFR coating: three layers with silver nanowires/polyvinyl butyral as the bottom layer, GO/functional cellulose as the middle layer, silver nanowires/fluoride polyvinyl butyral as top layer; BP: black phosphorus; MoS₂: Molybdenum disulfide; MPTS: 3-mercaptopropyltrimethoxysilane; GONR-1, GONR-2, GONR-3 paper: polyethylene glycol/GO/montmorillonite nanocomposite papers with three oxidation; BFPU: carbon-nitrogen-phosphorous multifunctional PU; the coated paper: wood pulp paper modified by sodium alginate@phenoxycyclophosphazene modified GO suspension and the positively charged CNTs/chitosan suspension; GO-ANF: GO-aramid nanofiber films; PGP: composition PGP with GO and bio-extracted phytic acid (PA) introduced into PVA; PEI: polyethyleneimine; HAP: hydroxyapatite; CF8: GO, PA, and Polyethyleneimine modulated lyocell fabrics; CSGP: aerogel composed of GO, sodium carboxymethyl cellulose, sodium alginate, polyaniline; CIGO: β -cyclodextrin-immobilized GO film.

**Fig. 8.** Schematic presentation of synergistic solid-phase flame-retardant mechanism.

combustion process for the effective interruption of the chain combustion reaction [57,60]. Moreover, the produced non-flammable gases can dilute the concentration of flammable gases to suppress the flame spread at the gas phase level [58]. Therefore, the above-mentioned gas-phase influence provides a synergistic effect with the physical barrier from GO in the solid phase. These two mechanisms jointly establish a better flame-retardant mechanism to delay the fire spread and suppressing heat release, thereby achieving a systematic improvement of flame-retardant efficiency.

To summarize, the flame-retardant mechanism of composite-engineered GO-based FWSs is essentially the systematic and co-ordinated flame-retardant process, integrating the basic char framework of the pyrolysis of GO and introduced solid-phase mechanism through physical barrier, thermal conduction regulation and catalytic carbonization and/or gas-phase flame-retardant

mechanisms through free radical quenching effect and the dilution effect of flammable gases. They do not operate in isolation but synergize, collectively forming the multi-level protective barrier that significantly enhances overall flame-retardant efficiency. Thus, the in-depth understanding of the multi-scale collaborative mechanism can provide a crucial theoretical foundation for the rational design and optimization of composite-engineered GO-based FWSs.

5.2. Fire warning mechanism

The working principle of GO-based fire-warning materials principally stems from its unique thermal reduction characteristics, realizing conductivity conversion under thermal driven through multi-scale physical and chemical synergistic effects. In essence, this is a thermodynamically driven GO reduction reaction,

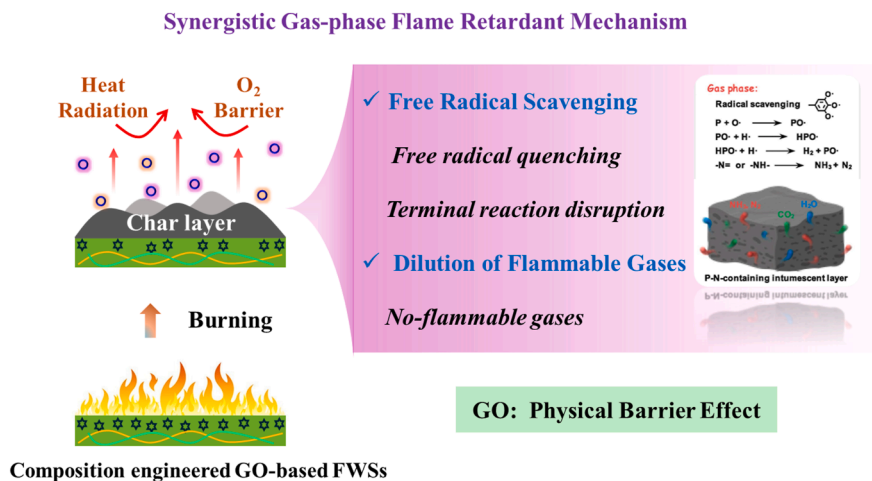


Fig. 9. Scheme diagram of synergistic gas-phase flame-retardant mechanism.

involving the gradual thermal decomposition of oxygen-containing functional groups and the dynamic reconstruction of the carbon skeleton structure of GO, which finally obtains conductive RGO from insulating GO (Fig. 10).

In terms of microscopic reaction mechanism, the abundant oxygen-containing groups of GO nanosheets undergo periodically pyrolysis reactions under high temperatures conditions, which is the initial stage and basic driving force of the reduction processing [116,117]. While undergoing the thermal processing, the epoxy groups (C–O–C) and hydroxyl groups (C–OH) on the surface of GO are preferentially thermally degraded, releasing gases of CO, CO₂, and H₂O [118]. With the temperature rising, carboxy groups begin to decompose and produce CO₂. As the temperature further increases, the residual oxygen-containing groups continue to be removed. The higher the temperature, the more removal of oxygen-containing groups during the thermodynamics thermal decomposition. More critically, the repair and graphitization of the carbon skeleton structure of GO are the core transformations of the reduction process. The carbon skeleton of GO undergoes significant structural reorganization accompanying occurs under high temperatures, leading to sp³ hybridized defective carbon (oxygen-containing group attachment sites) to reform sp² hybridized conjugated carbon network (graphene intrinsic structure), which can restore the conductivity

of the material [116]. Additionally, structural evolution happens as well under high temperatures simultaneously, involving changes in the interlayer distance and graphitization of GO. The gradual reduction of GO can cause a decrease in interlayer distance, approaching the initial distance to that of graphene, and resulting in a more compact stacking arrangement. When the temperature exceeds 500 °C, carbon atoms undergo migration and structural rearrangement, effectively healing holes and edge defects in GO. This restoration process is evidenced by the reduced D-to-G peak intensity ratio in raman spectroscopy, indicating enhanced structural ordering and increased graphitization degree of the material.

From a macroscopic perspective, the above-mentioned micro-structural changes directly determine the thermal sensitivity of GO-based fire-warning materials. During thermal exposure, gaseous products can cause the volume expansion and the formation of microcracks within the materials. Simultaneously, the reconstruction of the carbon skeleton reorganizes the conductive network. These two competing mechanisms (gas evolution induced expansion and carbon network reorganization) collectively regulate the electric resistance change of materials. When a specific temperature threshold is reached, these processes induce a significant electric resistance mutation, enabling the materials to function as a fire-warning sensor.

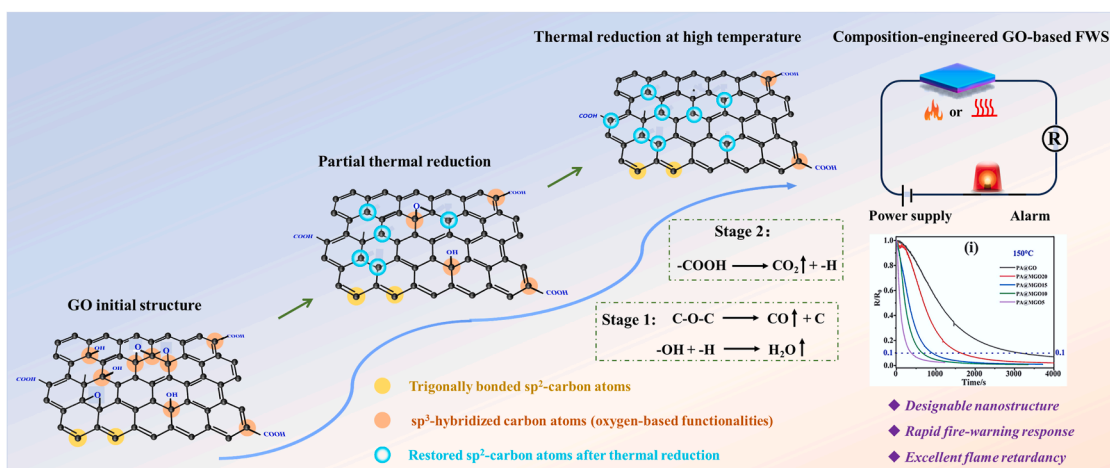


Fig. 10. Schematic illustration of fire-warning mechanism of GO-based FWSs.

It is worth noting that the reduction process of GO is highly dependent on temperature, as well as environmental factors, such as atmosphere and duration of heating. By carefully controlling the oxidation degree and distribution of functional groups on GO, the warning temperature can be precisely tailored. As the mentioned analysis, literature reports indicate that with optimized structural design, the fire-warning temperature can be bellowed to as low as 126 °C.

6. Challenges and perspectives

Inspired by its unique thermal sensitivity, a lot of efforts have been made in the scientific community of composition-engineered GO-based FWSs in recent year. Nonetheless, the challenges of this type of sensors are still ongoing, especially the performance and reliability need to be further optimized in response to the increasingly requirements for complex and diverse application scenarios. Against this backdrop, the main challenges and potential opportunities in the future about composition-engineered GO-based FWSs are proposed.

6.1. Challenges

6.1.1. Narrow response temperatures

According to the fire-warning mechanism, the response temperature window of GO-based FWSs directly hinges on the thermal reduction temperature range for GO or treated GO inherently. As the reports shown, GO experiences varying degrees of reduction under different temperatures attack. Thermal reduction of GO occurs at a moderate temperature of 100–250 °C, which is within the transitional stage from GO to RGO [108,119]. When reduction temperature reaches to 250 °C, the representative characteristics of RGO are manifested. With temperatures further increasing, the thermal reduction of GO is accelerated and a more complete restoration of the graphitic structure is promoted. To put it another way, the response temperatures higher 250 °C can enable alert, as demonstrated in most above-mentioned GO-based study, particularly at temperatures exceeding 300 °C. Although thermal reduction of GO can occur in the moderate temperature ranges, the sufficiency of the thermal reduction degree for enabling fire warning remain unclear, which can be improved by the structural regulation, leading to the low response temperature threshold shifting to 126 °C. Currently, the response temperature window for GO-based FWSs is 126–350 °C, showcasing the limitation of low-temperature warning applications. This also indicates the narrow response temperature, which attributed to the insensitivity of couple mechanism between the thermal reduction of GO nanosheets and its electrical response.

6.1.2. Undesirable fire-warning time

In GO-based fire-warning research, the shorter the fire-warning time, the better it is for fire management. Even though the response time of the currently reported systems has been shortened to the level of several seconds, the fire-warning sensitivity still fails to meet the requirements of practical applications. Most of current research is carried out in a well-controlled laboratory environment, we mainly focus on the interval time from key material undergoing a thermal reduction reaction until a warning signal is triggered, defining which as the response time. Nevertheless, owing to significant differences in experimental conditions and detection methods among various laboratories, it is difficult to make a direct comparison between these response time, especially under the lack of unified standards. In addition, in the practical application processing, the response time is still far from the satisfactory. The actual application environment is highly

complex, such as the large-scale requirements, interference and attenuation in wireless signal transmission, etc. which plays a negative role in fire-warning sensitivity. Moreover, the current laboratory conditions fail to fully replicate the complexity and unpredictability of real fire scenarios, meaning that the experimental findings offer limited guidance for real practical applications. Thus, how to further optimize the response sensitivity of early fire warning in the complex and real environments remains the core challenge faced in composition-engineered GO-based FWSs.

6.1.3. Limited fire-warning durability

In current reports, researchers overly focus on the timeliness of fire warning, while the discussion about fire-warning durability are relatively scarce. In fact, the durable fire warning is vital for implementing effective prevention and mitigation efforts, ensuring the early warning information can be received timely by ministry of emergency management or the public continuously, rather than transient notifications. The core challenge in prolong durability is preserving the integrity of the conductive pathway under elevated thermal conditions, which is strongly associated to structural integrity under continuous fire attack, containing microscopic stability, functional retention etc.

6.1.4. Single-use of fire-warning

GO-based FWSs mainly rely on the conversion of GO to RGO under high-temperature treatment or flame attack, leading to the structural alteration of nanosheets. After thermal reduction, the oxygen-containing groups on the edge or surface of GO are removed to restore the regular sp^2 network to become conductive material. Unfortunately, the structural change from sp^3 network of GO to sp^2 network of RGO is irreversible even though the heat sources are removed. The RGO can rarely be converted back to insulating GO, leading to the lack of reusability of GO-based fire-warning sensors.

6.1.5. Poor fire-warning accuracy under harsh environment

Most current fire-warning investigations are assumed to be applied in normal situations without considering the impact from special environmental conditions, such as rainy days, foggy days, snowy days, blizzards etc. These external factors seriously affect the fire-warning sensitivity or even make the sensors lose their fire-warning capability. Traditional single-parameter warning systems are difficult to cope with complicated and changeable environmental and disaster scenarios, particularly where multiple interference sources and multiple risk are coupled, leading to the obvious limitations of reliability and adaptability. Therefore, how to withstand the negative effects from uncomfortable circumstances is an open question.

6.1.6. Scale manufacturing and practical applications

Integration and scalability challenges are two points prevent FWSs moving from the laboratory to practical applications. In practical implementation, the structural stability, signal fidelity, as well the integration compatibility of FWSs with existing monitoring systems under industrial high temperatures, especially the influence of corrosive atmospheres or long-term hygrothermal environment have not fully verified. In addition, at the manufacturing level, the laboratory-scale preparation processing of GO-based FWSs pose a remarkable barrier to direct scale-up, which can be tackled through the scalable manufactory techniques including praying, printing, and 3D printing with fundamental balance in cost, uniformity, adhesion, and throughput. A systematic analysis of the process windows and cost-benefit

profiles for each manufacturing way is a critical prerequisite for enabling the successful engineering application of this technology.

6.2. Perspectives

Responding to the above unsettled issues during development of composition-engineered GO-based FWSs, the following research direction might foster ongoing development.

6.2.1. Target regulation of nanosheet structure – artificial intelligence (AI)

Different practical applications provide specific requirements for performance characteristics of materials. While the conventional structure-property relationship offers general guidance, the efficiency of directional structural design is constrained due to the complicated relationship between structural variables like layer numbers, functional group distribution, stacking mode and the performance. Introducing artificial intelligence (AI) to deeply explore and quantify the complex nonlinear relationship between the microstructure and macroscopic properties is becoming one key strategy to tackle this issue. In specific, the AI-driven reverse prediction can determine the optimal structural parameters of GO nanosheets based on the target performance. In addition, all the available modulation strategies can be quickly evaluated to choose one most promising one by using AI. Furthermore, AI can drive the exploration of innovative structure-property relationships, providing a guidance for the development of new FWSs. In brief, AI-driven structural directional regulation can directly specific requirements into precise, executable proposal. Due to the clear objective, targeted regulation of nanosheet structure aided through AI can directly tackle the specific bottleneck, which can predictably significantly enhance performance, but also show strong potential for overcoming limitations of materials.

6.2.2. Multi-functional integration notion

The integration of multi-functionality constitutes a pivotal research frontier for advancing the performance and adaptability of FWSs. Specifically, these multiple functions can be extended to self-power supply, self-healing, self-adaption, hydrophobicity, except for the basic properties of flame retardancy and fire warning, which could promote the transformation of early fire warning systems from a “passive response” to an “intelligent active defense” mode. Thus, intelligent fire-warning system with multi-function integration is not only inherent demand of technological evolution, but also provides insight into the new-generation of composition-engineered GO-based FWSs.

6.2.3. Multi-parameter cooperative sensing

Apart from the optimization of materials, systematically exploring GO-based composites with multi-responsive capability is another advanced developing direction, such as temperatures, smoke, gases, etc. The development of an intelligent GO-based fire warning systems possessing a multi-parameter sensing platform that integrated multi-source information, identified characteristic fire patterns, and accurately discriminates risk levels, is of importance for significantly reducing false alarm rates and enhancing the reliability of fire warnings. Multi-parameter cooperative sensing not only expands the practical applications scenarios, but also promotes the development of fire safety towards intelligence, precision and multi-functionality.

6.2.4. The establishment of standardization and evaluation systems

The establishment of standardization and evaluation systems plays a crucial role in ensuring measurement consistency, comparability among different results, and system reliability.

Unified standards not only provide an objective basis for the evaluation, but effective comparison between different investigations of their thermal response property of FWSs as well, which thereby can promote the technological transformation from the laboratory to practical applications.

The potential solutions are presented below: **i). the creation of traceable calibration benchmarks.** This entails constructing an environmental simulation chamber capable of precise control over parameters, like temperature, humidity, air pressure, which could be utilized to calibrate sensors deployed across various test situations against national standard metrological devices. The normalized output could be obtained through the correction model to eliminate the performance difference inherent to the sensors themselves. **ii). simulation testing based on physical models.** By setting up and running a series of standardized thermal environment test scenarios, a series of standardized thermal scenarios are constructed within a virtual testbed to offer completely consistent input conditions for various fire-warning algorithms. This enables the infinite reproduction and absolute controllability of all test scenarios. **iii). a dynamic process-oriented performance evaluation system.** This method mainly centers on dynamic behavior during the thermal response process instead of absolute accuracy, comprised of consistency, fire-warning timeliness, and monitoring performance. Long-term synchronous comparative tests in diverse real environments could be quantified comprehensively of the reliability and timeliness of each system in dynamic risk management. **iv). AI-driven evaluation system.** More encouragingly, AI can offer a breakthrough route to establish standardization and evaluation systems through data driving. AI technology can integrate all the influencing factors to elucidate the deep correlations among features through machine learning models, thereby constructing the multi-dimensional and dynamically adjustable evaluation system. Moreover, with the help of AI technology, efficient intelligent detection models can be built, helping to mitigate the false alarm of fire warning and improving the fire-warning accuracy. Furthermore, AI technology can also achieve advanced prediction and dynamic assessment of potential fire risks. In short, the introduction of AI technology can support intelligent fire warning in complex environments, while simultaneously foster the development towards standardization, systematization, and engineering essential for its scalable deployment.

CRedit authorship contribution statement

Xiaolu Li: Writing – review & editing, Writing – original draft, Validation, Investigation, Data curation, Conceptualization. **Chuanghui Chen:** Methodology. **José Sánchez del Río Sáez:** Writing – original draft. **Guoying Wei:** Methodology. **De-Yi Wang:** Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no competing financial interests or personal relationships that could have appeared to influence this work.

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