Hexachlorocyclotriphosphazene Functionalized Graphene Oxide as a Highly Efficient Flame Retardant

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ABSTRACT: A flame-retardant composite was synthesized through a simple graphene oxide functionalization route with hexachlorocyclotriphosphazene and p-phenylenediamine. Flame experiments conducted on the synthesized composite proved its importance as tremendously resistant to fire. The thermogravimetric analysis (TGA) shows clearly that the functionalized graphene oxide (FGO) exhibits an enhanced thermal stability and better temperature resistance. A thermostet epoxy resin was prepared by incorporating different percentages (2, 5, and 10%) of FGO to diglycidyl ether of bisphenol A (DGEBA). The flame-retardant properties, thermal degradation behavior, and combustion of the DGEBA thermosets cured by m-phenylenediamine were investigated using a Bunsen burner flame approaching the flame temperature of a fire and TGA. The chemical structure of FGO was characterized with spectroscopic and imaging techniques including Fourier transform infrared spectroscopy, Raman spectroscopy, X-ray diffraction, TGA, scanning electron microscopy, energy-dispersive X-ray spectroscopy elemental mapping, and X-ray photoelectron spectroscopy. Due to its high flame-retardant capabilities, such a composite could promise potential applications in the manufacture of inflammable materials for different uses.

INTRODUCTION

Synthesis and applications of graphene and graphene oxide (GO) have been the subjects of numerous studies in recent years. Graphene exhibits inert properties toward chemical reactions, which limit their use as potential reagents for further chemical syntheses and applications. Despite graphene, graphene oxide (GO), however, is a very promising material for various applications in chemical synthesis. The presence of many oxygen atoms in the form of hydroxyl and epoxide functionalities on the basal plane of graphene sheets, as well as carbonyl and carboxyl moieties on the edge of the sheets, renders this material as a potential reagent for various chemical applications. These applications included use of GO as catalysts in Friedel–Crafts reactions, oxidation, esterification and Knoevenagel condensation reactions, and Diels–Alder reaction. Kaviyarasu et al. studied the effect of a GO/ZnO material in photocatalytic applications, such as reduction of Cr(IV) compounds. Further applications of GO materials were reported by Maaza and co-workers; such applications include irradiation of Cu2+ ions on Co3O4-NiO-ZnO/GO nanowires and its effect on different electrochemical properties of the material. Analytical studies were also conducted by Bajjou et al. regarding charge carrier dynamics and the effect of pH on optical properties of porphyrin-graphene oxide composites. Numerous studies were conducted on the functionalization of GO with different positional isomers of phenylenediamine (PDA), pertaining to the plethora of applications for the materials obtained, including but not limited to anticorrosive agents, removal of heavy metals and other contaminants from water, and preparation of superhydrophobic surfaces. Studies of the electrochemical properties of PDA functionalized GO (GO-PDA) have received the highest interest. For instance, Dasari et al. reported a facile functionalization of GO with p-phenylenediamine (p-PDA) to produce a material (GO-p-PDA) that showed a sharp increase in electrical conductivity compared to pure GO. Incorporation of the obtained GO-p-PDA material into electrically insulating polypropylene produced a new electrically conducting composite. Wong and co-workers reported a detailed study on the functionalization of GO with o-PDA, m-PDA, and p-PDA, showing that m-PDA and p-PDA enlarged the graphene interlayer spacing by 1.3 and 1.41 nm, respectively, and showed considerable improvement of electrical capacitance. Yuan and co-workers showed that grafting of the HCCP. p-PDA to GO improved the flame retardancy of polypropylene. Though Yuan’s FGO showed...
good flame retardancy, the use of Ni(OH)$_2$ was necessary to further improve flame retardancy, and the maximum residual char yield that they obtained was only 8.5% when 2% of their FGO/Ni(OH)$_2$ was incorporated on polypropylene.$^{22}$ The phosphazene materials are polymers with transition nitrogen and phosphorus atoms known for their high thermal and chemical stabilities. Cyclic polyphosphazene ([PCl$_2$N]$_3$) obtained during phosphazene ring synthesis$^{23}$ shows exceptional properties, which impose it as one of the most interesting subjects of thermal and chemical stability studies.$^{24,25}$ In fact, its connection with other materials allows increased properties as they are thermally stable and flame retardants.$^{26,27}$ Herein, we investigate the properties of a polyphosphazene/p-PDA/GO (HCCP-p-PDA functionalized GO) composite as a flame retardant. To obtain the FGO product as an efficient flame retardant, the graphene oxide (GO) was successfully functionalized with hexachlorocyclotriphosphazene (HCCP) and p-phenylenediamine (p-PDA). Then, different percentages of FGO were incorporated into the epoxy resin DGEBA by in situ curing polymerization to enhance the thermal stability of composites. The typical preparation procedure for FGO is illustrated in Scheme 1.

### RESULTS AND DISCUSSION

Characterization of FGO. The functional groups of the obtained products GO, HCCP, and FGO were characterized by FTIR spectroscopy. Figure 1a shows the FTIR spectrum of GO with different peaks of the characteristic functional groups due to oxidation. The GO has a broad band characteristic of the hydroxyl functions at 3315 cm$^{-1}$ of the C−OH and C=O stretching vibrations and at 1735 cm$^{-1}$ frequencies of the −COOH. The peak at 1635 cm$^{-1}$ belongs to the aromatic C=C vibrations. The peaks at around 1225 and 1061 cm$^{-1}$ correspond to phenolic C−OH and epoxy C−O−C groups, respectively, similar to the previous literature.$^{22,28}$ Some new absorption peaks in the spectrum of FGO appear at around 1170 and 834 cm$^{-1}$ corresponding to the groups P=NC and P−N in the starting material (hexachlorocyclotriphosphazene),$^{29}$ which indicated that the phosphazene ring structure is not destroyed during the reaction. Meanwhile, another important peak appearing at 938 cm$^{-1}$ was assigned to the stretching P−NH−Ph,$^{30}$ which indicated the occurrence of copolymerization between HCCP and p-PDA. The peaks at 3330, 1511, and 1386 cm$^{-1}$ are assigned to N−H stretching, C−N stretching, and N−H bending,$^{22}$ respectively. The P−Cl peak at 660 cm$^{-1}$of HCCP$^{32}$ disappeared in the FGO, indicating that a nucleophilic substitution of HCCP by p-PDA had taken place. Hence, FTIR analysis confirms the functionalization of graphene oxide by HCCP. In parallel, Raman spectroscopy was used to investigate the quality of the carbon nanomaterials of GO and functionalized FGO. As shown in Figure 1c, the Raman spectrum of GO shows two prominent peaks at 1345 and 1584 cm$^{-1}$, corresponding to the D and G bands, respectively. The D band relates to the mode of radial vibration of the aromatic cycle and is related directly to the defects in the structure, while the G band comes from the mode of vibration associated with stretching of the C=C sp$^2$ links in the plane for the aromatic and aliphatic cycles.$^{13}$ Compared to the Raman spectrum of GO, the Raman spectrum of FGO also contains both D and G bands at 1383 and 1556 cm$^{-1}$, respectively. It is clear that the G band was broadened and shifted with higher intensity, while the D band around 1383 cm$^{-1}$ can be deconvoluted into two Lorentzian components, which can be used to assess the degree of the defect that could
be attributed to the Raman mode activated by the distortions.\textsuperscript{31} We agree that the D band is related to sp\textsuperscript{3} C atoms. However, we believe that the preparation route of FGO does not increase the amount of the sp\textsuperscript{3} carbon species. The addition between cyclophosphazene and p-phenylenediamine (p-PDA) involves the carboxylic moieties instead of the ethylenic double bond. In parallel, the ratio between D (D\textsubscript{1}+D\textsubscript{2}) peak and G peak intensities (I\textsubscript{D}/I\textsubscript{G}) leads to the amount quantification of the defect in the material. Measurements of ratios (I\textsubscript{D}/I\textsubscript{G}) for GO and FGO materials were collected. The (I\textsubscript{D}/I\textsubscript{G}) of FGO was 0.43, and the (I\textsubscript{D}/I\textsubscript{G}) of GO was 0.83. These changes could be assigned to the grafting of HCCP and p-PDA molecules into the hexagonal structure of carbon atoms, which corresponds to an increase of structural disorder (sp\textsuperscript{3} defects).\textsuperscript{32} This supposition will be confirmed by XRD analysis.

Solid-state NMR was used to clarify the molecular structure of FGO. The solid-state \textsuperscript{13}C magic angle spinning (MAS) NMR spectra of GO and FGO are depicted in Figure 1b; the NMR spectrum of GO reveals several peaks at 61 ppm (C−O−C), 70 ppm (C−OH), 101 ppm (O−C−O), 100−140 ppm (C≡C sp\textsuperscript{2}), 164 ppm (C==O carboxylic acid), and 190 ppm (C==O), and all are in good agreement with previously reported work.\textsuperscript{33} However, the typical GO resonances were also observed in the solid-state \textsuperscript{13}C NMR spectrum of the FGO, with the appearance of new peaks at 0−50 ppm, which correspond to the amine bonds (C−NH−R\textsubscript{2}). Moreover, two strong signals between 100 and 140 ppm associated to sp\textsuperscript{3} carbon atoms are observed, suggesting that they are more graphite-like.\textsuperscript{34} These results confirm that p-PDA is indeed incorporated on the surface of GO. Solid-state \textsuperscript{31}P NMR was used to identify the chemical environments of phosphorus. As shown in Figure 1d, a single peak at 2.5 ppm confirms the total substitution of chlorines by amines. A very small peak appearing at 18 ppm corresponds to unreacted P−Cl, as phosphorous would have a different chemical environment. The peaks indicated with asterisks are sidebands.\textsuperscript{35,36}

XPS was employed to detect the surface composition and chemical components of GO and FGO; the results are tabulated in Table S1 and shown in Figure S2 (see the Supporting Information). The survey XPS spectrum of FGO shows three new peaks attributing to N 1s, P 2p, and P 2s. Meanwhile, no Cl peaks are observed (Figure S2a), implying that the Cl atoms in HCCP have been completely substituted with phenyl amine groups of p-PDA.\textsuperscript{30} High-resolution XPS spectra are shown in Figure S2b−d. The C 1s spectrum of GO (Figure S2b) presents four types of carbons: C−C (285.7 eV), C−OH (287.9 eV), C−O−C (288.6 eV), and C==O (289.7 eV), which is in good agreement with earlier work results.\textsuperscript{22,37} The C 1s spectrum of FGO (Figure S2c) also indicates the appearance of C−O−P/C−N groups (285.9 eV),\textsuperscript{22} which confirms the successful modification based on GO. Further-
more, the intensity of oxygen functional groups in FGO decreases, which can be ascribed to the functionalization with polyphosphazenes containing phosphorus and nitrogen. The high-resolution spectra of N is for FGO (Figure S2d) show that there exist three chemical species of nitrogen: P=\( \text{N} \) (397.8 eV), P–NH–Ph (399.5 eV), and Ph–NH\(_2\) (400.1 eV), suggesting that the HCCP and p-PDA were grafted onto the surface of the GO nanosheets.

The X-ray diffraction pattern was used to study the crystal structure and interlayer distance of nanosheets in GO and FGO. As shown in Figure S3, the XRD pattern of pristine GO has a sharp diffraction peak located at \( 2\theta = 10.09^\circ \), indicating the crystalline structure of GO with the interlayer distance of 8.5 Å, which corresponds to the (002) plane due to the formation of oxygen functional groups (hydroxyl, carboxyl, and epoxy). A smaller peak appeared at \( 2\theta = 42.48^\circ \) (\( d = 2.9 \) Å), corresponding to the (100) plane, which is in good agreement with previously reported results. However, in the XRD pattern of FGO, the (002) plane diffraction peak shifts to a higher-angle position at \( 2\theta = 15.62^\circ \) leading to a decrease in the interlayer distance \( d = 5.7 \) Å by calculation with the Bragg equation, which indicates the conversion of GO into FGO. The peak corresponds to the (100) plane shifted to \( 2\theta = 47.25^\circ \). A new weak peak was observed at \( 2\theta = 22^\circ \), indicating that the \( \pi–\pi \) stacking of the GO was functionalized. Moreover, the presence of a broad diffraction peak on the spectra at around \( 2\theta = 22–34^\circ \) suggests that the structure of FGO is amorphous, which could be confirmed by selected-area electron diffraction (SAED) patterns (Figure S4).

The SEM image in Figure 2a presents the morphology of the obtained GO in the form of wrinkled sheets separated from each other and closely linked together, which confirms the accuracy of our synthesis. Compared to GO, the SEM of FGO (Figure 2c) shows a compact morphology, which indicates that the functionalization has been achieved. With a higher magnification (Figure 2b), the TEM image shows that the GO nanosheets are almost transparent and have no structural defect. However, the TEM image of the FGO (Figure 2d) demonstrates a different structure with dark spots incorporated and distributed on the GO nanosheets. The high-resolution transmission electron microscopy image (HRTEM) of GO clearly shows the nature of nanosheets as well as the thickness of the GO (Figure 2e). The spacing between two adjacent monolayers is 0.290 nm, which is consistent with the value of GO interlayer spacing of the (100) plane also obtained from SAED measurements (Figure S4A). Representative HRTEM and SAED images of the FGO are also shown in Figure S4B, implying a low degree of crystallization. Energy-dispersive X-ray spectroscopy (EDS) was used to follow the surface elements in FGO. The presence of N and P atoms on the GO nanosheet is confirmed by signals in the EDX spectrum (Figure 2f). Elementary mapping was performed to reveal the uniform distribution of atoms on the surface of the FGO: phosphorus (blue), oxygen and chlorine (black), nitrogen (red), and carbon (green) (Figure 2g).

TGA was used to follow the FGO thermal stability and the variation in mass loss for GO and functionalized GO with HCCP as a function of temperature under a nitrogen atmosphere (Figure 3). As a control experiment, compared to GO, the TGA thermogram of HCCP clearly shows a sharp mass loss from 60 to 180 °C, which is attributed to the sublimation of HCCP. For freshly synthesized GO, the TGA thermogram shows three stages of mass loss. The first mass loss below 100 °C was attributed to the removal of residual moisture. The second stage between 190 and 220 °C corresponds to the decomposition of the various labile oxygen functional groups, such as the hydroxyl group (~36 wt %). However, the last stage between 680 and 900 °C corresponds to the total mass loss with about ~24 wt % char residues. Therefore, the GO is thermally unstable, indicating that it could not be used individually as an efficient flame retardant for polymers. After functionalization, FGO has a mass loss around ~9 wt % at a 190–220 °C range compared to that of the GO (~36 wt %), indicating that the thermal stability is greatly improved. The mass loss above 300 °C in the TGA curve of FGO principally belongs to the decomposition of HCCP. Importantly, the char residues of FGO between 680 and 900 °C are increased to ~39 wt % compared to that of the GO (~24 wt %). Consequently, the good thermal stability for the FGO is explained by the elimination of the various oxygen-containing groups present on the GO, which were used as links with the HCCP.

**Thermal Stability and Thermal Decomposition.** The thermal stability of EP and its composites EP@GO and EP@FGO is investigated by TGA under a nitrogen atmosphere. TGA and differential thermogravimetric (DTG) curves are displayed in Figure 4a, and the detailed data are tabulated in Table S2. \( T_{10} \) is defined as the temperature at which the mass loss is 10%. \( T_{\text{max}} \) is the temperature when the mass loss of the sample reaches its maximum. The degradation of pure EP and its composites has two mass loss stages. The first stage is at 250–400 °C; it can be seen that the EP composites display a first maximum degradation temperature lower than that of the pure EP, which shows that the P–N-containing FRs decompose preferably at a lower temperature in order to catalyze carbonization of the EP matrix. The second stage occurs between 400 and 500 °C, indicating that EP@FGO composites exhibit higher thermal stability than the pure EP. Therefore, EP@FGO composites decompose slower at high temperatures due to the strong interfacial interactions between the FGO layers and the EP matrix, which can increase the thermal degradation activation energy of the composites. As shown in Table S2, after pyrolysis at 800 °C, the char yield ratio increases remarkably with increasing phosphorus and nitrogen content. The char yield of EP@FGO2% is...
progressively increased to 18.3%, higher than those of pure EP or EP@GO2%. Furthermore, the char yield of the EP@FGO composites is increased to 23% in EP@FGO10%, which demonstrates that FGO can retard the decomposition of the
composite. The main reason behind this phenomenon for this retarded decomposition is that FGO sheets are well-distributed in the EP matrix and form linkages with EP segments to effectively increase the intermolecular cross-linkage density.\textsuperscript{45} On the other hand, FGO catalyzes the char formation of the polymer matrix EP due to HCCP and therefore lowers the rate of maximum mass loss to produce an additional protective residue rich in phosphorus and nitrogen. As shown in the DTG curves (Figure 4b), it is clear that maximum mass loss rates of the composites attenuated by FGO are about 45% (EP@FGO10%) because of the enhancement in the char formation and the excellent physical barrier effect provided by the FGO sheets, which can accelerate the heat transfer.\textsuperscript{43}

**Flammability Tests.** In order to understand the different inflammability characteristics between EP, EP@GO, and EP@FGO, a flame test was performed to confirm the flame-retardant effect. For this purpose, the different polymers have been modulated in the form of pellets for practical application (as shown in Scheme 2).

The sample pellets were exposed vertically to a Bunsen burner flame approaching the flame temperature of a fire then recorded by a digital camera at different stages of the combustion experiments and are shown in Figure 5.

It is clearly observed from these photographs that the pellets of pure epoxy EP and its composite EP@GO immediately caught fire, releasing a large flame whose propagation was total, while the epoxy functionalized with a flame retardant (EP/FGO) only exhibited slight combustion and almost non-existent propagation. After applying a flame for 8 s, the pure EP still burned with severe flaming drips, indicating a highly combustible behavior. However, in the case of the EP@FGO, one of the fascinating characteristics of the combustion is that it just burned lightly with a little flame and extinguished by itself within 10 s while keeping its initial form. At the end of the combustion experiment, it is interesting to observe that part of the surface of the epoxy composite was covered with a network of expanded carbon, indicating that this epoxy composite formed an effective carbon that was able to impede heat transfer and the spread of flame during combustion. These results demonstrate the flammability of epoxy thermosts linked to phosphazene due to the presence of a unique combination of phosphorus and nitrogen. Finally, we have found that the retarding effect has been produced with the incorporation of only 2% by weight of FGO in the epoxy resin EP.

**CONCLUSION**

A new method for the preparation of functionalized graphene oxide (FGO) is revealed in this study. The obtained FGO prepared via reaction of hexachlorocyclotriphosphazene (HCCP) and \( p \)-phenylenediamine (\( p \)-PDA), hexachlorocyclotriphosphazene (HCCP), potassium carbonate (\( K_2CO_3 \)), concentrated sulfuric acid (\( H_2SO_4 \), 98%), potassium permanganate (\( KMnO_4 \)), hydrogen peroxide aqueous solution (\( H_2O_2 \), 30%), phosphoric acid (\( H_3PO_4 \), 88%), hydrochloric acid (\( HCl \)), toluene, acetone, methanol, bisphenol A diglycidyl ether (DGEBA), and \( m \)-phenylenediamine (\( m \)-PDA) were supplied from Sigma-Aldrich (Oakville, ON, Canada). Deionized (DI) water was produced by a water purification machine (Milli-Q Reference). Toluene was dried by 4 Å molecular sieves before being used.

**EXPERIMENTAL SECTION**

**Experimental Methods.** Graphite powder (mesh size, \( \leq 100 \) \( \mu \)m), \( p \)-phenylenediamine (\( p \)-PDA), hexachlorocyclotriphosphazene (HCCP), potassium carbonate (\( K_2CO_3 \)), concentrated sulfuric acid (\( H_2SO_4 \), 98%), potassium permanganate (\( KMnO_4 \)), hydrogen peroxide aqueous solution (\( H_2O_2 \), 30%), phosphoric acid (\( H_3PO_4 \), 88%), hydrochloric acid (\( HCl \)), toluene, acetone, methanol, bisphenol A diglycidyl ether (DGEBA), and \( m \)-phenylenediamine (\( m \)-PDA) were used.

**Characterization Methods.** Fourier transform infrared (FTIR) spectroscopy was performed on a Nicolet Smart iTR 7600 spectrometer in the wavenumber range of 4000–500 cm\(^{-1}\) (Thermo-Nicolet, Madison, WI). Data were analyzed using OMNIC software (Thermo-Nicolet, Madison, WI). X-ray diffraction (XRD) was performed using a diffractometer (Bruker D8 Advance) with Cu Kα radiation (\( \lambda = 1.54182 \) Å) in an angular interval from 20 to 100° at room temperature. X-ray photoelectron spectroscopy (XPS) data for elemental composition and valence information were collected with an XPS PHI 5600-ci (Physical Electronics, Eden Prairie, MN, USA). The morphology of materials was obtained with a scanning electron microscope (SEM) by utilizing a JEOL JSM7600F operated at a 10 kV accelerating voltage and a working distance of 14 mm using secondary electron imaging mode. Transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDS) were performed on a Jeol JEM-2100F. A thermal gravimetric analysis (TGA Q500/Discovery MS) was used to measure the thermal stability under nitrogen (\( N_2 \)) with a heat ramp of 5 °C/min between 30 and 1000 °C. Raman measurements were acquired on a Renishaw inVia microspectrometer using a 50X objective and a 514 nm laser wavelength. \( ^{13}C \) and \( ^{31}P \) MAS NMR spectra were recorded using a 400 MHz solid-state Bruker Avance III HD spectrometer (Milton, ON, Canada), operating at frequencies of 150.874 MHz for \( ^{13}C \) and 162 MHz for \( ^{31}P \). Cross-polarization with the magic angle spinning (CP/MAS) technique was adopted.

**Synthesis of the GO by the Modified Hummers Method.** Graphene oxide was obtained according to the modified Hummers method.\textsuperscript{46} Briefly, 3 g of graphite flakes was mixed with 360 mL of concentrated acid \( H_2SO_4 \) and 40 mL of 85% \( H_3PO_4 \). Then, 18 g of \( KMnO_4 \) was added gradually for 10 min. The reaction mixture was heated at 50 °C for 4 h with continuous stirring. To promote exfoliation of graphite oxide, short periods of ultrasonication (4×) were applied every hour (15 min) during the reaction (4 h). When the reaction was over, the suspension was neutralized with a mixture of ice water containing 30% \( H_2O_2 \) (10 mL) with a strong release of gas. The suspension was centrifuged and washed several times with deionized water (250 mL), 10% HCl solution (250 mL), and anhydrous ethanol (2 × 250 mL). The obtained light brown material was then precipitated in anhydrous ether (1000 mL) and dried in a vacuum oven for 24 h at 40 °C.
yielding 4.9 g of a crude mixture of graphite/graphene oxide. Finally, the crude product (4.9 g) containing a mixture of graphite and graphene oxide was diluted in NanoPure water (1 L) with stirring and sonication until complete homogenization to have a concentration of 4.9 g/L, and the solution was stored at room temperature.

Preparation of Functionalized Graphene Oxide by Hexachlorocyclotriphosphazene. Graphene oxide (100 mg) was dissolved in anhydrous toluene (10 mL), sonicated for 30 min at ambient temperature, and then stirred with a mechanical stirrer to form a brownish black slurry. K2CO3 (1.40 g) was added to the GO suspension, and then, HCCP (348 mg, 1 mmol) dissolved in toluene was added dropwise to the mixture with continuous stirring under N2 protection. Then, 325 mg (3 mmol) of p-PDA dissolved in toluene was slowly added to the mixture. Afterward, the reaction was refluxed and maintained at 60 °C for 72 h. Finally, the obtained FGO was filtered and washed copiously with deionized water and methanol and then dried under vacuum at 70 °C for 24 h to yield a light violet-brownish solid.

Preparation of Polymer Composites EP@GO and EP@FGO. A flammable epoxy resin (EP) was mixed with GO and FGO (see the Supporting Information). The EP was prepared by mixing an epoxy precursor bisphenol A diglycidyl ether (DGEBA) and m-phenylenediamine (m-PDA) as a hardener (Figure S1). The main monomer of this type of resin has two very electrophilic epoxy functions, which will react with different nucleophilic functions of graphene oxide (COOH and OH). In parallel, the hardener could react with epoxy functions as well as with halogenated reactive sites present on phosphazene. For flame retardancy testing, various composites were prepared, such as EP (100%), EP loaded with graphene oxide (EP@GO) comprising 2% by mass ratio of GO, and EP loaded with the functionalized graphene oxide (EP@FGO), 2, 5, and 10% as mass ratios of FGO in total mass. Prior to the composite preparation, GO and FGO were dissolved in acetone, which we added to the prepared EP. The reaction mixture was stirred at 40 °C in a water bath to evaporate acetone. Finally, the mixture was kept in a vacuum oven at 80 °C for 45 min until the polymers (foams) become dry, see Scheme 2 in the Flammability Tests section.

■ ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c05815.

Illustration of synthesized thermostet epoxy resin, survey XPS spectra of GO and FGO and high XPS resolution for C 1s and N 1s for FGO, XRD spectra of GO and FGO, high-resolution transmission electron microscopy (HRTEM) image with the corresponding SAED pattern (top inset) of (A) (GO) and (B) (FGO), table for element content of GO and FGO measured from XPS analysis, and table for detailed TGA data for EP and its composites under a N2 atmosphere (PDF)

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K.R., A.S.EL, and M.S. performed the conceptualization; K.R., S.C., and A.S.EL performed the methodology; K.R., S.C., and A.S.EL performed the validation; K.R. performed formal analysis; M.S. contributed resources; K.R., S.C., and A.S.EL performed data curation; K.R. performed writing—original draft preparation; K.R., S.C., A.S.EL, and M.S. performed writing—review and editing; M.S. conducted supervision; M.S. performed funding acquisition; all authors have read and agreed to the published version of the manuscript.

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