Viscosity Simulation of Glass Microfiber and an Unusual Air Filter with High-Efficiency Antibacterial Functionality Enabled by ZnO/Graphene-Modified Glass Microfiber

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ABSTRACT: The current global pandemic of new coronary pneumonia clearly reveals the importance of developing highly efficient filtration and fast germicidal performance of multifunctional air filters. In this study, a novel air filter with a controllable morphology based on the rod-like to flower-like zinc oxide/graphene-based photocatalytic composite particles loaded on glass microfiber was prepared by one-step microwave rapid synthesis. The multifunctional air filter shows the following special functions: the 10 mg·L⁻¹ organic pollutant solution RhB was completely degraded within 2 h under a 500 W xenon lamp, and also 99% of *Escherichia coli* and *Staphylococcus aureus* were inactivated under a 60 W light-emitting diode lamp. Furthermore, after introducing the controllable morphology zinc oxide/graphene-based photocatalytic composite particles, the filtration efficiency of the multifunctional air filter was also kept at the same level (99.8%) as the one without any addition, indicating no loss of high-efficiency filtration while obtaining the rapid bactericidal function. The rapid antibacterial principle of the multifunctional air filter has also been proposed through the UV–vis spectroscopies, photoluminescence, and electron-spin resonance spectrum. The zinc oxide/graphene-based photocatalytic composite particles tightly coated on the glass microfiber surface could increase the active sites by changing the morphology of zinc oxide and, in the meantime, promote the separation of zinc oxide photo-generated electron–hole pairs to improve the rapid sterilization ability of the multifunctional air filters. In addition, an empirical formula to evaluate the relationship between the composition, viscosity, and viscosity modulus of glass microfiber was proposed by testing the viscosity of glass microfiber composed of 14 different compositions at 1300 and 1400 °C, which can be used as a criterion to evaluate the production technology of glass microfiber filters.

1. INTRODUCTION

At present, viral respiratory diseases have become one of the most serious threats to public health and safety, resulting from the pathogens of these diseases not only being widely spread through touching but also through aerosols. The current global outbreak of COVID-19 has clearly demonstrated the necessity for developing advanced personal and crowd protection equipment with multifunctional filtration and antibacterial functions. Usually, air disinfection is an effective method to prevent the respiratory virus from cross-infecting, especially in densely populated places, such as stations, offices, and shopping malls.

Currently, electrostatic adsorption elements are widely used in the field of air purification, which works on the principle that a strong inside electrostatic field could be excited by a high-voltage generator in order to adsorb the excited microorganisms with a positive charge in the air. However, under the current global pandemic of COVID-19, it is unsuitable for electrostatic adsorption elements to make a large-scale promotion due to a large amount of electricity consumption and strong radiation exposure to the human body. In addition, there are also nanofiber meshes with sterilizing particles prepared by the electrostatic spinning method to be used for air purification and sterilization. Although nanofiber has a larger specific surface area, energy, and tension to intercept airborne particulate and bacteria, the electrospun nanofiber as an air filter material has easily become a source of secondary pollution because nanofiber breaks down due to its poor mechanical property. Furthermore, there are also problems such as low dust
collection capacity and short service life resulting from the limited adsorption sites in the nanofiber membrane.\textsuperscript{14,15}

Glass microfiber air filter materials are widely used in ultraclean rooms for liquid crystal panel, chip production, and medical operations due to a larger dust holding capacity, higher filtration efficiency, and lower cost.\textsuperscript{16,17} However, the current widely used glass microfiber filter only has the function of air filtration by the physical interception and electrostatic adsorption of the microfiber three-dimensional network without any sterilization function.

The efficient air filtration and rapid antibacterial function could be achieved through physical interception of fibers and chemical disinfection of drugs, respectively. In view of this, the researchers found that nanophotocatalysis is a very environmentally friendly sterilization technology.\textsuperscript{18,19} As a food-grade photocatalytic antibacterial particle, ZnO has high-exciton binding energy (60 meV) and wide band gap energy (3.37 eV), leading to the generation of reactive oxygen species (ROS) under light irradiation to cause cell apoptosis by interacting with certain enzymes inside or on the surface of cell membranes.\textsuperscript{20,21} In addition, ZnO photocatalyst can completely decompose bacteria and microorganisms into H$_2$O and CO$_2$,\textsuperscript{22} avoiding secondary pollution during the sterilization process. However, the relatively low dispersion and high recombination rate of electron–hole pairs have greatly hindered the antibacterial application of ZnO nanoparticles.\textsuperscript{23}

Graphene has also become a new functional material for antibacterial applications due to its unique characteristics, such as high electrical conductivity, excellent solubility, biocompatibility, and relatively low cytotoxicity to mammalian cells,\textsuperscript{24–26} which could provide more active sites for ZnO nanoparticles in order to improve the photocatalytic antibacterial efficiency by effectively separating photo-generated electrons and holes. At the same time, graphene itself is able to show moderate antibacterial activity in the absence of light and shows stronger bacterial inactivation in the presence of light due to the photothermal effect.\textsuperscript{27,28} Moreover, the application of photocatalytic nanoparticles as a powder will also be difficult to recycle and cause secondary pollution problems.\textsuperscript{29,30} So far, there are relatively few reports about the combination of the excellent filtration ability of glass microfiber and the rapid antibacterial ability of zinc oxide/graphene photocatalytic composite particles in the synthesis of multifunctional air filters. When glass fibers are used for air filtration materials, the finer the diameter of the fibers, the better the air filtration effect.\textsuperscript{31,32} However, too fine a fiber will lead to a reduction in its strength, and thus, it is prone to breakage and damage, and the fiber will instead become a contaminant like the impurities originally planned to be filtered out. Viscosity is a key parameter in the production process to improve the composition and microstructure of the fibers so that they have higher strength at finer diameters. Viscosity needs to be regulated, but direct measurement of glass fiber melt viscosity is time-consuming, energy-intensive, and expensive, so a method is needed to establish a quantitative relationship between viscosity and the chemical composition.

Herein, a novel air filter that can meet the requirements of high-efficiency filtration and rapid antibacterial performance was prepared. The air filter adopts one-step microwave synthesis on glass microfiber of the air filter to support zinc oxide/graphene-based composite photocatalytic particles with controllable morphology that changes from rod-like to flower-like. The microstructural, photocatalytic ability and antibacterial performance of the multifunctional air filters were investigated. An evaluation system based on the relationship between the viscosity and composition of glass fibers is proposed, which is of guidance for the production of air filters. The multifunctional mechanism of microfiber filtration and photocatalytic sterilization of the developed air filter was proposed, which is a great potential technology for promoting air purification to meet the needs of clean, active, and healthy living.

2. MATERIALS AND METHODS

2.1. Materials. The zinc acetate dihydrate and ammonia solution (GR, 25–28%) were purchased from Chengdu Kelong Chemical Co., Ltd. Graphene and the glass microfiber (H13) were supplied by Zisun Technology Co., Ltd.

2.2. Synthesis of HGZ-R Air Filters. The specific synthesizing step for the novel multifunctional air filters is shown in Figure 1. Firstly, the graphene powder was dispersed in the absolute ethanol solution and ultrasonically oscillated for 10 min. The glass microfiber filter membrane was placed in the graphene dispersion and ultrasonically oscillated for 10 min. The purpose of the above step is to oscillate the graphene so that graphene nanoparticles could be uniformly loaded on the surface of glass microfiber to increase the growth sites of the subsequent ZnO. The zinc oxide precursor solution was prepared as follows: 8 g of Zn(CH$_3$COO)$_2$·2H$_2$O was added to 100 mL of distilled water, followed by several slow drops of 3 mL of ammonia to form a precipitate. A certain amount of the dried precipitate was taken to configure 1.6 mol/L of zinc oxide precursor solution, and the solvent was distilled water and ammonia in a volume ratio of 1:1. The graphene-loaded
H13 was immersed for 5 min to make it fully infiltrated. Then it was put into the microwave synthesizer, and the reaction temperature was set to 90 °C with a reaction time of 30 s and 1 min, respectively. According to the different morphologies of ZnO/graphene-based photocatalytic particles, the synthesized air filters were named HGZ (HGZ-F and HGZ-R). At the same time, air filters HG and HZ (HZ-F and HZ-R) without graphene were prepared under the same conditions in order to study the enhancement of the antibacterial effect of graphene on the composite air filter.

2.3. Characterization. The phase and composition of the as-prepared air filters were observed by using X-ray diffraction (XRD, Cu Kα radiation). The scanning electron microscope (Zeiss EVO LS10) at 3 kV and energy-dispersive X-ray spectroscopy (EDX) were used to obtain the morphology and element distribution on the surface of glass microfiber. The photoluminescence (PL) emission spectra were measured by using RF600 (Japan, SHIMADZU). The UV–vis diffuse reflectance spectra (DRS) were recorded on a UV3600 (Japan, SHIMADZU). The photocatalytic degradation performance of the as-prepared HGZ-R air filters was tested by UV–visible spectroscopy (Agilent Cary5000) in the wavelength range of 400–700 nm.

2.4. Evaluation of Photocatalytic Degradation. The photocatalytic properties of the as-prepared HGZ-R and HGZ-F air filters were studied by using a rhodamine B (RhB) solution.33 For this purpose, HGZ-F and HGZ-R air filters with a size of 4 × 4 cm were completely submerged in 50 mL of 10 mg L−1 of an aqueous RhB solution. The glass tubes were left in the dark for 30 min to allow the air filters and RhB solution to reach adsorption equilibrium. The RhB solution is degraded by irradiating with a xenon light with a power of 500 W within 2 h. 4 mL of liquid samples was taken out from the reactor regularly every 15 min. The degradation ratio (a) of RhB was calculated by the following equation

\[
a = \frac{C}{C_0} \times 100\%
\]

where \(C_0\) and \(C\) represent the initial dye concentration and the dye concentration after light radiation, respectively.

2.5. Antibacterial Assays. The antibacterial activities of the as-prepared HGZ-R air filters were evaluated by using the sterilization ability of *Escherichia coli* and *Staphylococcus aureus* under a 60 W light-emitting diode (LED) radiation lamp. An average colony counting method was adapted for the test.34,35 The two stains were first stored in Luria–Bertani (LB) medium and incubated for 18 h at 37 °C in an incubator.

Then, 50 μL of the bacterial culture was taken into 5 mL LB medium and incubated at 37 °C for 12 h, followed by setting the bacterial concentration to 10^5 CFU/mL. The unprocessed H13 and HGZ were cut into 1 × 1 cm square pieces and cleaned with 75% medical alcohol to remove microbial bacteria carried by the air filter itself. Afterward, 200 μL of the bacterial suspension was added to the sterilized air filter for bacterial growth under light and dark conditions at 37 °C, respectively. These pieces were then placed in a clean container with 10 mL of PBS to obtain a dispersion of bacteria. Finally, 100 μL of the diluted bacterial suspension was evenly spread on LB agar medium and propagated for 24 h at 37 °C. The above procedure was repeated three times and observed by microscopy to obtain the average number of colonies on LB agar plates. The fast photocatalytic antimicrobial performance of the air filter under visible light conditions was calculated by the following equation

\[
\text{growth inhibition rate (％)} = \frac{N_i - N_f}{N_i} \times 100\%
\]

where \(N_i\) and \(N_f\) are the average bacterial counts of H13 and air filters loaded with photocatalytic particles, respectively.

2.6. Air Filtration Efficiency Test. In this study, a charge-neutralized NaCl aerosol was used to measure the filter penetration level of the air filter using the TSI 3160 (USA TSI) automated filter tester. Two particle counts were used to measure the concentration of different particle sizes in the air before and after the filters. Also, the most penetrating particle size (MPPS) of the H13 and HGZ-R was measured using an excited monodisperse aerosol at a speed of 5.3 cm/s. The filtration capacity of multifunctional H13 air filters is characterized by MPPS and filtration efficiency.33

2.7. Viscosity Test. The viscosity of glass liquids of different compositions at different temperatures is tested by means of a rotating high-speed viscometer (1600, USA Orton). The melt is considered an equilibrium liquid, and the viscosity is measured by a fixed torque and a fixed shear according to the constant rotational speed method.36

\[
\eta = \tau / \gamma; \quad \tau = M / (2\pi R^2); \quad \gamma = 2\Omega / n(1 - k^{2/n}); \quad \Omega = 2\pi \omega / 60
\]

\(\eta\)—viscosity, \(\tau\)—shear stress, \(\gamma\)—shear rate, \(M\)—torque, \(R\)—radius of the rotor, \(l\)—the length of the rotor, \(\omega\)—rotational speed. In conclusion, the viscosity expression can be obtained.
\[ \eta = \frac{1}{4\pi l} \frac{M(1 - k^2)}{\pi R^2} \]

3. RESULTS AND DISCUSSION

3.1. XRD and Raman Analysis. The ZnO XRD patterns in Figure 2a showed nine important peaks at the 2θ values of \( \sim 31.8, 34.5, 36.3, 47.6, 56.6, 62.9, 66.4, 68, \) and 69.1° related to the crystal planes of (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes, respectively, of the hexagonal wurtzite crystal structure (JCPDS card no. 36-1451). The XRD patterns of the HGZ nanocomposites were similar to that of pure HZ, which indicated that the addition of Graphene to ZnO did not change the crystallinity of ZnO on H13 air filters. Raman spectroscopy was performed on the HGZ-R in order to further verify the presence of graphene/ZnO on the H13 air filter. The results are shown in Figure 2b. It can be observed in Figure 2b. For HGZ-R, the peak of the Raman spectrum at 334, 438, and 581 cm\(^{-1}\) could be corresponded to the \( \text{E}_2 \) (high)−\( \text{E}_2 \) (low), \( \text{E}_2 \) (high), and \( \text{E}_1 \)
(LO) vibration modes of ZnO, respectively. In graphene, the D band at 1348 cm\(^{-1}\) is caused by the defects and amorphous structure, while the G band at 1598 cm\(^{-1}\) is related to the active E\(_{2g}\) mode of the sp\(^2\) carbon atom. The above results further prove that the graphene/ZnO composite photocatalytic particles are loaded on H13 air filters to obtain a multifunctional antibacterial air purification filter.

3.2. Morphology and Structure. In order to characterize the morphology of the air filters, a field-emission electron microscope test was carried out, as shown in Figure 3. Figure 3a represents the original H13 air filter, and the interlacing of single superfine glass fiber filaments forms a multilayer mesh structure, which can prove the efficient physical interception effect of the H13 air filter, which explains well the excellent physical interception effect of the H13 air filter. Figure 3b–d shows that the flower-like ZnO grown in situ by microwave synthesis at different multiples is supported on the surface of a single glass microfiber. Figure 3e–f shows the further growth of flower-like ZnO particles to a rod-like structure at different magnifications. It was found that as the reaction time increased, ZnO gradually grew from a nanorod shape of about 600 nm to a micron-sized flower shape of about 5 \(\mu\)m. From this phenomenon, it can be inferred that the morphologically controllable ZnO/graphene-based composite particles grow on the glass microfiber to form a stable three-layer network structure. Without destroying the multilayer mesh structure of the original H13 glass microfiber, graphene provides zinc oxide with more active growth sites while effectively improving the dispersibility of zinc oxide. Based on the change in the morphology of ZnO photocatalytic particles, the flower-like ZnO exposes fewer catalytic action points than the rod-like ZnO, which may lead to changes in the antibacterial activity of the air filter.

In order to explore the element distribution on the surface of the filter, the EDX test of HGZ-R was characterized, and the test results are shown in Figure 3g. The distribution of Si, Zn, C, and O elements in the air filters can be obtained. The distribution of Si and C elements comes from SiO\(_2\) and the loaded graphene in the matrix glass microfiber, and the Zn element comes from ZnO synthesized. Furthermore, as shown in Figure 3g, obviously the existence of the O element is attributed to the ZnO rod and SiO\(_2\). From the above results, it can be concluded that C, Zn, and O elements are uniformly distributed in the glass microfiber filaments. This also further reveals the tight coating and good dispersibility of the ZnO/graphene-based composite particles with controllable morphology on the surface of HGZ-R glass microfibers.

3.3. Optical Properties. The UV–vis transmission spectra of the air filters are revealed in Figure 4a. Compared with pure HZ-R and HZ-F, it can be clearly seen that the HGZ-R and HGZ-F air filters exhibit stronger light absorption in both ultraviolet and visible light regions, which reveals that the introduction of graphene can enhance the photocatalytic performance of the air filters under the visible light conditions. Since ZnO is a direct band gap semiconductor, the optical band gap (E\(_g\)) of the air filter is calculated by the Tauc equation:

\[
(ahv)^2 = A(hv - E_g)
\]

where \(a\) explains the absorption coefficient, \(hv\) represents the photon energy, and \(A\) is a constant.

As shown in Figure 4b, the band gap energies of HZ-F, HZ-R, HGZ-F, and HGZ-R are found to be 3.15, 3.14, 3.12, and 3.10 eV, respectively. The band gap of HGZ-R air filters is narrower because graphene can act as an acceptor for the excited electrons of ZnO. At the same time, the DRS spectrum
of the air filters also shows stronger visible absorption as the change in morphology of ZnO-based particles from flower-like to rod-like. This means that the HGZ air filter can absorb more photogenerated electron energy, so the addition of graphene in HZ air filters and changes in the morphology of ZnO can greatly improve the photocatalytic degradation of air filters by increasing the light absorption in the visible wavelength range.

The photoluminescence spectra (excited at 325 nm) of the HZ-F, HZ-R, HGZ-F, and HGZ-R multifunctional antibacterial air filters are presented in Figure 4c. The PL peak at 386 nm is due to the recombination of electrons excited by light and holes in the valence band. The HGZ-R air filter has the weakest PL peak intensity, which further indicates that the introduction of graphene can improve the utilization of photogenerated electron pairs by reducing the recombination of electron pairs in the ZnO valence band because graphene is a good electron acceptor. At the same time, the peaks of HZ-F and HGZ-F are larger than HZ-R and HGZ-R, respectively, which also shows that the change of the morphology of ZnO from flower-like to rod-like can also promote the separation of photogenerated electron pairs. The above results prove that HGZ-R has better photocatalytic activity and antibacterial properties.

3.5. Photocatalytic Degradation Ability of the HGZ Filters. The degradation experiment of the RhB solution under light was investigated to prove the photocatalytic activity of the novel HGZ-F and HGZ-R air filter. Also, to verify the adsorption effect of graphene due to its large specific surface area, we performed a light-free adsorption test on HG, and the results are shown in Figure S2. The results demonstrate that the decrease in the concentration of the solution is not caused by the adsorption of graphene. The degradation rate of the RhB solution under dark conditions was 2%. From the decolorization of the RhB solution in Figure 5a, b, it can be seen that as the morphology of ZnO/graphene-based photocatalytic particles changes from flower-like to rod-like, the degradation of the RhB solution by the air filter has been enhanced within 2 h. The result of degradation efficiency is shown in Figure 5c, where the degradation efficiencies of HGZ-R and HGZ-F to RhB are 99.7 and 99%, respectively. It can be inferred that the reason for this result is that the smaller size will bring more active sites as the morphology of ZnO/graphene-based photocatalytic particles change from flower-like to rod-like, thereby improving the photocatalytic activity of the HGZ-R air filters for RhB solution compared to the HGZ-F filter. The above results are also consistent with the results displayed by the spectrum of DRS and PL.

3.6. Photocatalytic Antibacterial Performance. As shown in Figure 6, antibacterial activity was assessed to test the antibacterial efficiency of the multifunctional filters. Under the radiation of a 60 W LED lamp, the growth of E. coli and S. aureus in the H13 air filter without any load showed an increasing trend, and the number of colonies on the filter was 417 and 454 within 2 h, respectively. The antibacterial experiments on HG samples, as shown in Figure S1, showed that the inactivation rates of E. coli and S. aureus were 10 and 12% under dark conditions, and their inactivation rates could reach 30.2 and 35.4% when the light was irradiated, respectively. This result proves that graphene itself has moderate antibacterial activity. Compared with the air filter of zinc oxide produced by pyrolysis, the photocatalytic inactivation rates of E. coli and S. aureus were 76 and 74%, respectively. Furthermore, as the morphology of ZnO/graphene-based-photocatalytic particles changes from flower-like to rod-like, the inactivation rate of bacteria by the air filters reaches 99%. From these results, it can be seen that the ZnO/graphene-based photocatalytic particles base with controllable morphology acts a pivotal part in improving the rapid sterilization ability of the multifunctional filters. This observation is likely a result of the increase in the number of active sites where the air filters contact the bacterial surface. Thereby, the HGZ-R air filter has a very excellent ability to be a rapid bactericidal, which is suitable for use in the field of air purification in a complex environment.

3.7. MMPS and Filter Performance. The MMPS and filtration efficiency of the H13 and HGZ-R air filters is tested. As shown in Table 1 and Figure 7, the filtration efficiency of HGZ-R still remains at 99.8%, which was tested for five different diameters of NaCl particles. The filtration resistance of HGZ-R has been increased from 275.4 of H13 to 317.2 Pa, graphene-based-photocatalytic particles changes from flower-like to rod-like, the inactivation rate of bacteria by the air filters reaches 99%. From these results, it can be seen that the ZnO/graphene-based photocatalytic particles base with controllable morphology acts a pivotal part in improving the rapid sterilization ability of the multifunctional filters. This observation is likely a result of the increase in the number of active sites where the air filters contact the bacterial surface. Thereby, the HGZ-R air filter has a very excellent ability to be a rapid bactericidal, which is suitable for use in the field of air purification in a complex environment.

Table 1. MMPS and Filtration Efficiency of the HGZ-R and the H13 Filter without Any Addition

| name     | resistance (Pa) | penetration (%) | efficiency (%) | MPPS (µm) |
|----------|-----------------|-----------------|----------------|-----------|
| H13      | 275.4           | 0.1633          | 99.8366        | 0.136     |
| HGZ-R    | 317.2           | 0.1977          | 99.8022        | 0.133     |

Figure 6. Images of antibacterial activities of the air filters.

Figure 7. Air filtration testing of H13 and HGZ-R air filters.
which is the reason for the improvement of filtration efficiency. This result shows that the introduction of shape-controlled ZnO/graphene on H13 fibers by microwave synthesis does not lead to a reduction in the physical interception of airborne particles by the composite filter membrane. The MPPS of the standard H13 and the ZnO/graphene-loaded H13 is 0.136 and 0.133 μm, respectively, and the transmittance of micro/nanoparticles is 0.1633 and 0.1977%, respectively. The increase in filtration capacity may be due to the increase in ZnO/graphene-based photocatalytic particles on the smooth surface of the H13 air filter and the increase in roughness, which leads to an increase in the number of effective adsorption sites and the probability of filtration mechanisms, such as interception, Brownian diffusion, inertial impact, and inertial deposition improves. The above result indicates that the HGZ-R still maintains an efficient physical interception for aerosol particles.

3.8. Principle of Filtration and Antibacterial Capability. In order to study the photactive free radicals with strong oxidizing ability in the photocatalytic degradation of HGZ-R air filter, the electron-spin resonance (ESR) spectra are used to characterize the photogenerated free radicals on HGZ-R, as shown in Figure 8. The HGZ-R air filter has no obvious signal peaks for free radicals under dark conditions, which indicates that free radicals cannot be generated in the absence of light. On the contrary, the four characteristic peaks of DMPO−•OH (Figure 8a) and DMPO−•O2•− (Figure 8b) can be clearly found in the HGZ-R air filter after 4, 8, and 12 min of light irradiation. The corresponding peak intensity is also getting stronger and stronger, which means that the concentration of photocatalytic active groups increases with the prolonging of the illumination time. The mechanism of air filter membrane with high air filtration efficiency and fast antimicrobial capability is discussed as shown in Figure 9. The physical filtration mechanism of the glass microfiber filter is the result of various kinds of synthesized effects, such as the inertial effect, the diffusion effect, and the interception effect. The main filtering principle of the multifunctional HGZ-R air filters is the interfacing single glass microfiber filament to build a three-dimensional network structure, which can retain large particles and some harmful substances in the air. When the environment is suitable, bacteria will grow on the glass fiber filter membrane, and graphene itself has a moderate antibacterial ability to inactivate the bacteria. When the light shines on the air filter, the ZnO loaded on the surface begins to take effect. When the HGZ-R filter is excited by light radiation with a higher photon energy than the band gap of ZnO, photogenerated electrons (e−) excited from the valence band (VB) are transferred to the conduction band (CB) while retaining an equal number of photogenerated holes (h+) in the VB. However, due to the narrower CB of graphene than ZnO, the photoexcited electrons of ZnO are transferred to the CB of graphene (4.2 eV) to avoid direct complexation with the holes on the VB. This process greatly improves the separation of the photoexcited electron pairs of ZnO. Thus, electrons accumulated at the CB of ZnO or graphene can react with oxygen molecules on the surface of the filter to form radicals such as •O2•− and •OH. These free radicals can act as strong oxidizing agents to degrade RhB solutions and kill bacterial microorganisms. On the other hand, the possible antimicrobial mechanism of the HGZ-R air filter is to generate ROS to damage the cell membrane of bacteria, causing leakage of intracellular components and destruction of proteins/DNA, which eventually leads to bacterial death. Bacterial microorganisms are inactivated by active molecules to prevent the continued reproduction of bacterial microorganisms on the air filter, so as to achieve the purpose of air filtration, and at the same time, increase the life of the air filter, and reduce the environmental pollution caused by frequent replacement of the filter element.

3.9. Relationship between Viscosity (η) and Viscosity Modulus (Mη). The viscosities of 16 different composition ratios of H13 were measured at 1300 and 1400 °C, as shown in Table 2. To better describe the viscosity properties, the viscosity modulus (Mη) was introduced, which is defined as

![Figure 8](http://pubs.acs.org/journal/acsodf)

![Figure 9](http://pubs.acs.org/journal/acsodf)
\[ M_\eta = \frac{M_{SiO_2} + 2M_{Al_2O_3}}{2M_{Fe_2O_3} + M_{FeO} + M_{CaO} + M_{MgO} + M_{K_2O} + M_{Na_2O}} \]

where \( M_{SiO_2}, M_{Al_2O_3} \), and so forth represent the molar fractions of the corresponding oxides.

The viscosity modulus values \( (M_\eta) \) corresponding to each group of proportioned glass fibers were calculated from the compositions in Table 2 and related to the measured viscosity values \( (\eta) \) at the actual temperature to obtain a relationship between viscosity \( (\eta) \) and viscosity modulus \( (M_\eta) \) at 1300 °C (Figure 10a) and 1400 °C (Figure 10b). Based on the results in the figure and the available literature, two empirical formulas are proposed:

\[
\eta \text{(1300 °C)} = 120.09605 - 3.89453 \times 5.70111M_\eta
\]

\[
\eta \text{(1400 °C)} = 92.12202 - 1.5445 \times 1.89115M_\eta
\]

The actual viscosity at 1300 and 1400 °C is quantified based on the chemical composition of the glass fiber ratios, thus saving the time and resources consumed by direct viscosity measurements. Different chemical compositions affect the viscosity of the glass as it melts, and a highly viscous melt is difficult to control during the drawing process and can easily lead to fiber breakage and damage. In contrast, a low-viscosity melt can flow better and faster before solidifying into fibers. This also provides technical support for the production of finer glass fibers to ensure the strength of glass fibers as air filtration materials.

4. CONCLUSIONS

In summary, this study proposes a multifunctional air filter material. The relationship between the precursor composition and viscosity of H13 glass microfibers was established to provide a technical reference for obtaining finer and higher efficiency H13 air filters. A multifunctional air filter that meets the requirements of photocatalytic antibacterial and high-efficiency filtration is prepared by forming a three-layer tightly wrapped structure of glass microfiber/graphene/zinc oxide. At the same time, the morphology of ZnO/graphene-based composite photocatalytic particles is changed from flower-like to rod-like to improve the sterilization rate of the multifunctional filter. In addition to the synergistically enhanced photocatalytic and antibacterial effects of graphene on zinc oxide with changing morphology, the proposed composite HGZ-R air filter also shows excellent performance in intercepting particles of different sizes. The multifunctional composite air filter is able to achieve a 99.9% high-efficiency filtration, its MPPS can reach 0.133 μm, and it can achieve 98% degradation for RhB solution within 2 h. At the same time, the photocatalytic degradation mechanism and antibacterial mechanism of the HGZ-R air filter paper were discussed. The reason why the photocatalytic activity is improved is also verified by PL and DRS spectroscopy. From the above research results, the air filter material has a remarkable inactivation performance against \textit{E. coli} and \textit{S. aureus} due to ZnO/graphene-based photocatalytic particles, and 99% of the bacteria can be inactivated within 2 h of the 60 W led light irradiation. A quick and effective solution and cheap manufacturing cost can easily adapt to the application of the air filter in different scenarios. Owing to this antibacterial
property, the filter can be used for a longer time to achieve economic and environmental protection purposes.

**ASSOCIATED CONTENT**

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c00838.

Antimicrobial experiment of the HG air filter (Figure S1) and dark adsorption of the HGZ-F air filter (Figure S2) (PDF)

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Notes
The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This work was supported by the Ministry of Industry and Information Technology of the People’s Republic of China (no. 2019-09000-1-1), the Chongqing Municipal Education Commission (no. KJCX2020048), the research program of Chongqing University of Arts and Sciences (no. P2021CL07), the Chongqing Science and Technology Bureau (nos. T204040012; cstc2021jcyj-msxmX0775, cstc2019jcyj-qxq0021). This work was also funded by the Chongqing University Key Laboratory of Micro/Nano Materials Engineering and Technology (project no: KFJJ2011).

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