

Tailoring dielectric relaxation time in ultralight graphene aerogels for ultra-broadband microwave absorption and multifunctional applications

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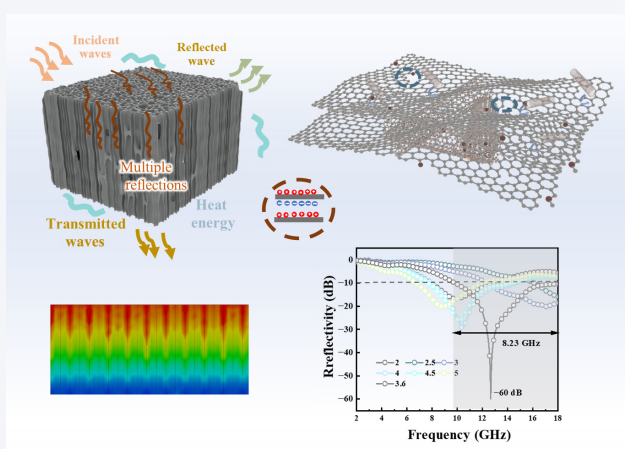
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ABSTRACT: Composite aerogels composed of reduced graphene oxide (RGO), single-walled carbon nanotubes (SWNTs), and polyimide (PI) with an oriented porous structure exhibit flexible, high-performance microwave absorption and a tunable porosity. The polarization relaxation time is varied by adjusting the heat treatment temperature to achieve ultra-broadband microwave absorption at lower thicknesses. SWNTs contribute to the control of aerogel morphology and significantly improve its mechanical toughness. The prepared aerogel has a bulk density of 9.13 mg/cm³ and excellent absorption properties with a minimum absorption peak of -64.5 dB and an effective absorption bandwidth (EAB) of 9.14 GHz. Through experimental comparison and time-domain finite integration calculation, the effect of oriented porous structure on the electromagnetic loss mechanism and performance was elucidated, which can be used for porous wave-absorbing materials. At the same time, the light and flexible PGS composite aerogel fabric is also remarkably flexible, and its excellent mechanical and thermal insulation properties are attributed to its unique orientated microchannel structure. This low-cost, simple and high-performance approach will greatly simplify the application of multifunctional microwave-absorbing materials in wearable and other extreme conditions, including high-temperature environments.



KEYWORDS: graphene oxide (GO), broadband wave, thermal isolation, flexographic, Ku-band

1 Introduction

The proliferation of 5G communication networks and radar systems has made electromagnetic wave-absorbing materials play an increasingly important role in civil and military applications. Electromagnetic wave-absorbing materials play a critical role in mitigating electromagnetic interference (EMI) by effectively attenuating undesired electromagnetic waves through energy dissipation mechanisms such as destructive interference, dielectric/magnetic losses, and thermal conversion. These materials

are indispensable for safeguarding sensitive electronic devices from EMI-induced malfunctions and minimizing potential health risks associated with prolonged electromagnetic exposure in humans [1–14]. Meanwhile, excellent wave-absorbing properties, wide effective absorption bandwidths (EAB), and lightweight characteristics are essential for more convenient applications in industrial production and daily life. In the past decades, the research on conventional microwave absorbers (including ferrites [15], metals [16], conductive polymers [17], carbon materials [18], etc.) has achieved remarkable results. All of these materials have filler rates higher than 20%, and those with filler rates below 20% need to be compounded with carbon materials [19, 20]. According to previous studies, graphene has been widely used in the field of electromagnetic wave absorption due to its ultra-high specific surface area, light weight and high carrier mobility. However, during the reduction of graphene oxide (GO) to reduced graphene

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oxide (RGO), individual graphene sheets tend to restack, which significantly hinders electromagnetic wave penetration and degrades impedance matching. To address this limitation, RGO is typically composited with magnetic or dielectric nanoparticles (e.g., RGO/Fe₃O₄ and ZnO/RGO). These nanoparticles, when assembled on both surfaces of the graphene sheets, not only suppress sheet restacking but also enhance impedance matching through either improved dielectric/magnetic loss characteristics or optimized wave impedance properties. However, graphene may clump together, lose its ability to withstand corrosion, and—most significantly—lose its lightweight characteristics when nanoparticles are added. Impedance matching also can be improved by integrating low-dimensional nanomaterials like carbon nanotubes [21], MXene [22], and nanowires [23] into three-dimensional (3D) cellular architectures.

In practice, these wave-absorbing materials are usually used as absorbers, and in the process of use they usually need to be compounded with other materials, which will greatly sacrifice the lightweight properties of the material. At the same time, the addition of polymers such as paraffin wax, epoxy resins and other polymeric materials will destroy the original porous structure of the absorber, and these materials can not be fully considered as electromagnetically transparent, and will also interact with the absorber [24]. Although three-dimensional porous structures can somewhat mitigate the adverse effects of additives, matrices have been used in tests and applications to boost strength because of the limited mechanical properties of porous structures. However, this destroys the original three-dimensional porous structure, and the unique features may also interact with the added matrix to alter the material's original properties. This mostly restricts the use of lightweight 3D honeycomb structures in industries like aerospace that are sensitive to weight. Recently, significant research progress has been made on wave-absorbing aerogels based on graphene [25, 26], graphene/multi-walled carbon nanotube (MWCNT) [21], Ni@graphene [27], and MXene/conducting polymers (CPs) [28] with free-standing honeycomb structures. Solvothermal-produced aerogels have asymmetrical shapes, their diameter is constrained by the reactor, and the majority of them have substandard mechanical properties that make them unsuitable for use in both production and daily life. The inability to prevent additional cutting and assembly raises the workload and expense, and the low mechanical properties of basic graphene composite aerogels make them easy to distort and restrict their use. Therefore, it has proved to be quite a challenge to design aerogels that are ultralight (less than 10 mg/cm³) flexible, thin, cost-effective, and have broadband MA properties [29].

A critical challenge in developing fully dielectric aerogels is achieving broadband microwave absorption at reduced thicknesses. Thinner wave-absorbing aerogels offer better compatibility with flexible electronics and wearable devices. According to quarter-wavelength theory (Eq. (1)), aerogels require higher dielectric constants for effective broadband absorption at small thicknesses. However, excessively high dielectric constants cause impedance mismatch. This phenomenon is governed by the frequency-dependent dielectric response, as described by dielectric dispersion theory. The underlying mechanism involves dipole and charge relaxation/polarization processes. When the frequency of oscillation of the electric field exceeds the relaxation time of the dipoles, their response becomes inefficient, leading to a rapid decrease in the real part of the dielectric constant and a simultaneous peak in the imaginary part.

$$t_m = \frac{n}{4} \lambda = \frac{nc}{4f_m \sqrt{\epsilon_r \mu_r}} \quad (n = 1, 3, 5, \dots) \quad (1)$$

This means that excellent broadband microwave absorption can be obtained by adjusting the polarization relaxation time even at very low thicknesses, whereas in the case of GO aerogel materials, the heat treatment temperature can effectively modulate the surface active groups of GO, and the interfacial and dipole polarization produced by these active groups can effectively modulate the polarization relaxation time [30, 31].

This study demonstrates the successful fabrication of polyimide/RGO/single-walled carbon nanotubes (SWNTs) composite aerogels (PGS) through a facile directional freezing approach. Polyimides with electromagnetic transparency enhance the mechanical properties of the materials while also improving thermal stability, which enriches multi-scenario applications while enhancing energy-absorbing properties. We changed the polarization relaxation time by adjusting the heat treatment temperature, and by prolonging the relaxation time the aerogel can be made to obtain broader bandwidth wave-absorbing properties at lower thickness. The single-walled carbon nanotubes distributed on graphene sheets not only improve the conductivity and polarization effect of the material, but also enable PGS to achieve better mechanical properties and flexibility. The prepared aerogel exhibits bulk density of 9.13 mg/cm³ and has excellent wave-absorbing properties, with a minimum absorption peak of −64.5 dB and an EAB of 9.14 GHz, covering half of the X band and the whole Ku band. The impact of the oriented pore-like structure on the electromagnetic loss mechanism and performance has been clarified for the use of porous wave-absorbing materials by experimental comparisons and time-domain finite-integral calculations. Furthermore, the PGS composite aerogel exhibits notable flexibility alongside its electromagnetic wave-absorbing properties. The material's exceptional mechanical strength and superior thermal insulation characteristics stem from its well-aligned microchannel architecture, which was achieved through the directional freezing process. This cost-effective and facile fabrication approach enables the practical implementation of multifunctional microwave-absorbing materials, even under demanding operational conditions including elevated temperatures.

2 Experimental section

2.1 Materials

Potassium permanganate (KMnO₄) was purchased from Aladdin. Single-walled carbon nanotubes and graphite (1000 mesh) (SWNTs, outer diameter: < 2 nm, length: 5–30 μm) were purchased from Shenzhen Guoheng Qihang Technology Co., Ltd. Sodium nitrate (NaNO₃), nitric acid, sulfuric acid, homophthalic anhydride (PMDA), 4,4'-diaminodiphenyl ether (ODA), triethylamine (TEA), and N,N-dimethylacetamide were obtained from Sinopharm Chemical Reagent Co., Ltd. All the chemical reagents were of analytical grade without further purification. The deionized water was produced by our laboratory with a resistivity of about 18.5 megohms.

2.2 Synthesis of PGS

As shown in Fig. 1, 500 mg of PAA (see synthesis method in the Electronic Supplementary Material (ESM)) was dissolved in 50 mL of deionised water at room temperature with the aid of

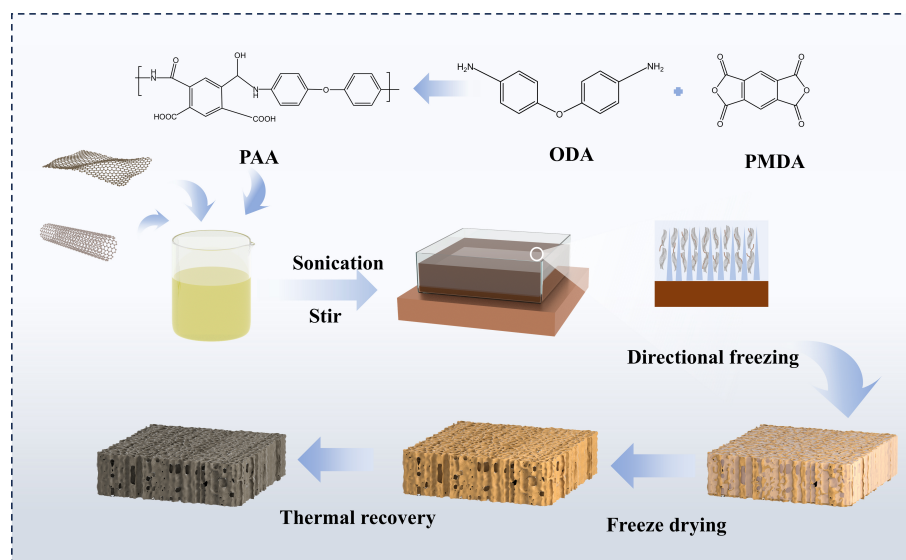


Figure 1 Flowchart for the preparation of PGS aerogel.

triethylamine to obtain a water-soluble polyimide precursor solution. Then, 500 mg of prepared graphene oxide and carboxylated single-walled carbon nanotubes were added to the water-soluble polyimide solution. Subsequently, the suspensions were placed in quartz molds attached to pre-cooled copper cube substrates and frozen at $-80\text{ }^{\circ}\text{C}$, followed by freeze-drying for two days ($-60\text{ }^{\circ}\text{C}$ and $< 10\text{ Pa}$), resulting in the formation of PAA/GO/SWNTs hybrid aerogels. Finally, the fabricated PAA/GO/SWNTs aerogels were annealed at high temperature in a nitrogen atmosphere to achieve thermal imidization of PAA and thermal reduction of GO. PGS composite aerogels with different final bulk densities and thicknesses could be fabricated by simply varying the initial concentration and volume of the mixed suspension. Samples of complexes without added SWNTs were named PG (ratio of PAA content to GO content). Composite aerogels with different dielectric constants were obtained by varying the concentration of SWNTs to 5, 15, 25, and 50 mg, and the temperatures of heat treatment to 200, 300, 400, and $500\text{ }^{\circ}\text{C}$, and were named as PGS concentration-temperature. The obtained aerogels were cured with melted paraffin wax and prepared into coaxial rings with an inner diameter of 3 mm and an outer diameter of 7 mm to test the electromagnetic parameters of the samples.

3 Results and discussions

3.1 Morphology and structure

As illustrated in Fig. 1, the addition of TEA enables complete dissolution of PAA in aqueous solution. Both GO lamellae and carboxylated SWNTs possess abundant surface functional groups that promote stable suspension in the aqueous medium through electrostatic repulsion effects, thereby facilitating subsequent aerogel formation. During the directional freezing process, when the mold containing the PAA/GO/carboxylated SWNTs solution was placed on pre-cooled copper substrates, vertically aligned ice crystals nucleated and grew unidirectionally along the temperature gradient. This configuration restricted heat transfer primarily to the axial direction perpendicular to the copper plate surface. Additionally, during the unidirectional freezing process, the

growing ice crystals induced the alignment of suspended GO sheets and SWCNTs along their advancing solid-liquid interfaces. This resulted in the formation of a hierarchical tubular architecture, which after freeze-drying produced an aerogel with well-aligned, anisotropic pore channels. The simplicity of both the apparatus design and assembly process enables precise control over the aerogel's structural parameters, including density, geometry, and thickness—critical features for developing modular absorption components in wireless communication systems such as base stations and portable electronic devices. Figure 2 displays the microscopic morphology of the aerogels at varying concentrations of GO, SWNTs, and PAA. A copper plate at $-80\text{ }^{\circ}\text{C}$ was placed directly on the bottom of the mold during the aerogel's creation, causing the water in the dispersion to solidify from the bottom and create tiny ice crystals. Small ice crystals are eliminated during the freeze-drying process, leaving behind a dense porous structure resembling a honeycomb and a channel structure that resembles a slit. These features aid in creating a connected conductive network, which causes numerous electromagnetic wave scattering and reflection. The microscopic morphology of various graphene to PAA ratios is shown in Figs. 2(b) and 2(c), and Fig. S1(a) in the ESM. It is evident that as the graphene content increases, the slit-like channel shape and thick honeycomb porous structure eventually turn perpendicular from wavy, while the vertical slit-like channel structure tubular channel becomes more regular and straight with the addition of SWNTs (Figs. 2(d) and 2(e), and Fig. S1(b) in the ESM). Without the addition of SWNTs, the oriented void structure is only supported by lamellar GO, which is not enough to make the oriented tubular structure straight, and when the GO content is low, the oriented tubular structure is more difficult to maintain. In contrast, with the addition of SWNTs, due to the reinforcing effect of the fibers, the SWNTs appear as a mesh on the surface, and it is difficult for lamellar GO to provide sufficient support, whereas the fibers can effectively withstand tensile stresses and prevent the bending and elongation. The theoretical Young's modulus of SWNTs is about 1 TPa, and the tensile strength is about 50 GPa. The monolayer structure can be uniformly dispersed and form strong interfacial interactions (e.g., π - π stacking, covalent bonding), which can lead to a more straight and oriented void structure. Comparatively speaking, van der Waals' forces between the layers of MWNTs is weaker, which is

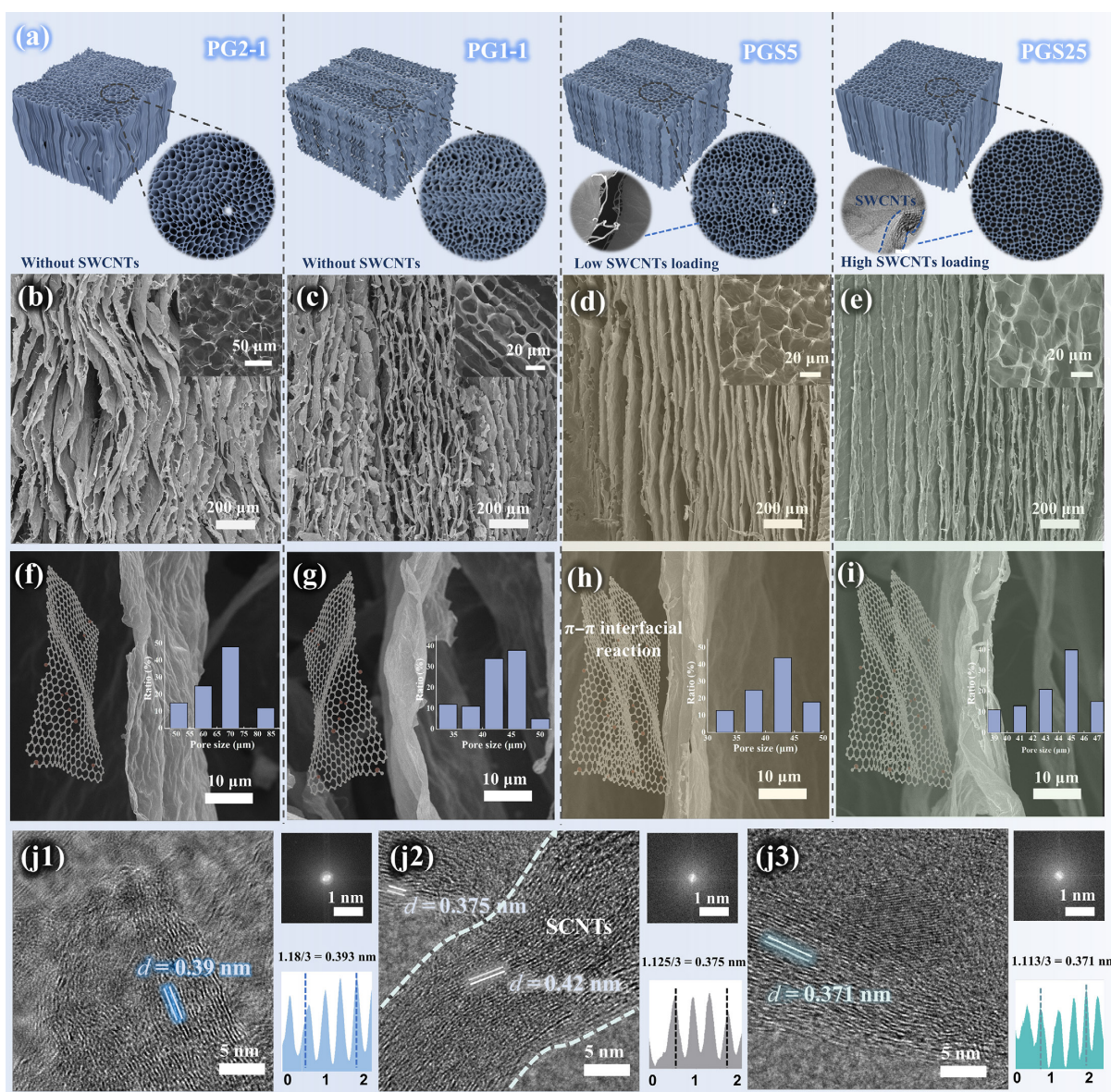


Figure 2 (a) Morphology of aerogels with different compositions. SEM images of ((b) and (f)) PG2-1, ((c) and (g)) PG1-1, ((d) and (h)) PGS5-300, and ((e) and (i)) PGS25-300. TEM images of ((j1) PGS5-300, (j2) PGS5-400, and (j3) PGS5-500).

prone to interlayer slip, and the actual strength is lower than the theoretical value [32]. Figure 3(c) shows the most available porosity and void fraction of PI/SWNTs, PI/GO and PI/GO/SWNTs aerogels. Clearly, the addition of GO can act as a backbone supporter for the aerogel, with an increase in void fraction from 90.91% to 98.56%. Moreover, relative to the PI/SWNTs, the porosity of the PI/GO and PI/GO/SWNTs aerogels shows more concentrated pore size distribution, and the average pore size decreased from 16.9 to 6.03 μm . The test data demonstrate that the addition of SWNTs could help the pore size of the aerogel to be more homogeneous and concentrated, and the average pore size was reduced to 4.9 μm , while obtaining a higher void fraction of 99.61% and a larger specific surface area of 6.07 m^2/g (Table S1 in the ESM). The diameter size of the honeycomb holes decreased with increasing graphene (71 to 45 μm). Notably, without SWNTs, the supports of the slit-like channels are mainly monolithic graphene, whereas with the addition of SWNTs, the monolithic graphene is interconnected due to the π - π interfacial interaction

between them, which enhances the toughness of the aerogel and improves the electrical conductivity. Figures S1(f) and S1(g) in the ESM) show the TEM images of the PGS, and it can be seen that the presence of SWNTs effectively prevents the re-stacking of graphene sheets, and the SWNTs are inserted into neighboring nanosheets to form a dense conductive network. Meanwhile, large graphene nanosheets and SWNTs are easily found, leading to numerous heterostructures and intricate interactions. Figure 2(j) shows the change in layer spacing in PGS composite aerogels with increasing temperature, which corresponds to the (002) crystallographic plane of carbon. The corresponding layer spacing for PGS is wider than that of graphite (0.34 nm) due to the formation of oxidized and carboxylated functional groups between the GO layers [33]. As the pyrolysis temperature rises, the oxidized functional groups of graphene oxide gradually diminish, the graphene's layer spacing gradually shrinks, and the (001) crystallographic plane gradually changes into the (002) crystallographic plane, suggesting that the PGS's carbon undergoes graphitization.

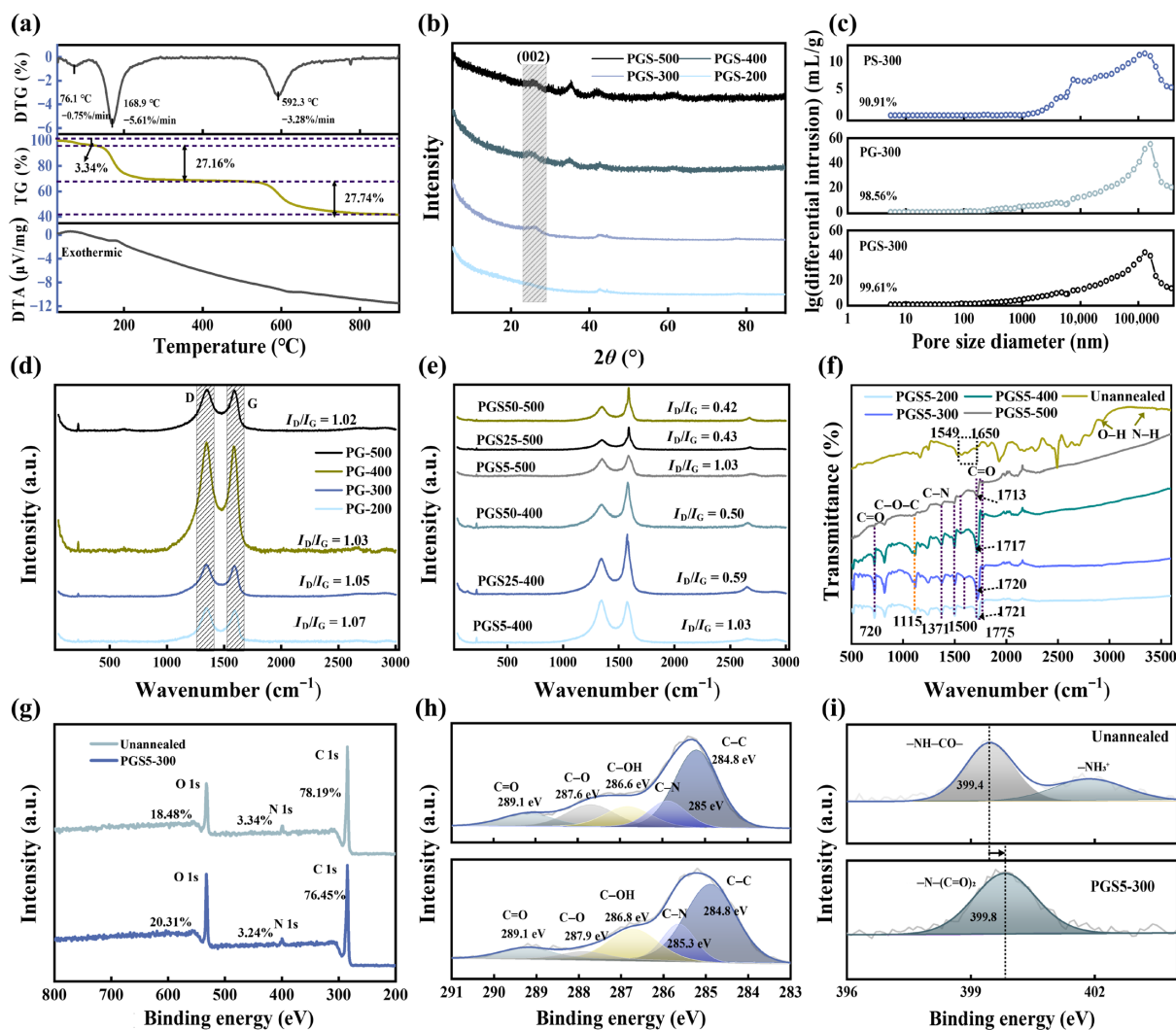


Figure 3 (a) The TGA of PGS5. (b) XRD patterns, (c) porosity and fewest pores, ((d) and (e)) Raman spectra, (f) FT-IR spectra, and (g)–(i) XPS comparison of PGS before and after heat treatment.

Thermogravimetric analysis of the PAA/GO/SWNTs aerogel reveals a multi-stage degradation process under nitrogen. The initial weight loss up to 100 °C is due to moisture evaporation, followed by a major step between 100–200 °C (peak at 168.9 °C) from polyamide acid imidization and GO reduction. A plateau region from 200–500 °C indicates the formation of a stable polyimide and reduced GO structure, beyond which significant weight loss resumes from 500 to 900 °C, attributable to the carbonization of the polyimide matrix. Comparatively, the samples pre-heated to 300, 400, and 500 °C exhibit enhanced thermal stability with shifted decomposition profiles and reduced total weight loss, confirming that the pre-treatment effectively advances crosslinking and removes labile groups, yielding a more robust material. Additionally, as shown in the figure, the characteristic peaks corresponding to the (002) crystal planes become increasingly prominent with rising heat treatment temperatures, indicating progressive restacking of graphene lamellae. The characteristic peaks around 25° in the XRD spectra of Fig. 3(b) are from the (002) crystal planes of graphene, reflecting the degree of graphitization of graphene. The drop in the interplanar spacing of (002) from 0.39 to 0.37 nm in Fig. 2(k) further supports this. Observing Fig. S2 in the ESM, it is clear that the increase in temperature shifts the

(002) crystal plane peak from 25° to 26°, whereas the addition of single-walled carbon nanotubes attenuates the movement of the characteristic peaks and appropriately inhibits graphitization. Meanwhile, the characteristic peak at 13° is observed, which comes from the exfoliated graphene (001) crystal plane, implying that SWNTs effectively prevented the stacking of graphene lamellae. Raman I_D/I_G (the sp^2 structural domain's chaotic vibrations and structural defects are reflected in the D-band peak at 1340 cm^{-1} . The graphitic carbon layer is reflected in the G-band peak at $\sim 1600\text{ }cm^{-1}$) was used to assess the degree of graphitization of carbon in the PG (Fig. 3(d)). As the annealing temperature rises from 200 to 500 °C, the I_D/I_G ratio decreases from 1.07 to 1.02, suggesting a reduction in defect density. The 2D characteristic peaks at 2655 cm^{-1} appearing in the PGS due to the incorporation of SWNTs become increasingly pronounced, which is indicative of the enhanced electrical conductivity of the material, and this is reinforced by the change in the I_D/I_G ratio from 0.59 to 0.42 (Fig. 3(e)).

Attenuated total reflectance fourier transform infrared spectroscopy (ATR-FTIR) was used to characterize the chemical structures of the samples with varying compositions (Fig. 3(f)). It can be clearly seen that the unannealed PAA/GO/SWNTs are

distinctly different from the other annealed samples, with 1650 cm^{-1} coming from the amide I band ($\text{C}=\text{O}$ stretching vibration, main chain $-\text{CO}-\text{NH}-$), 1549 cm^{-1} from the amide II band ($\text{N}-\text{H}$ bending vibration + $\text{C}-\text{N}$ stretching vibration). The $\text{C}=\text{O}$ stretching vibrations of free carboxylic acids can be observed at 1710 cm^{-1} . The characteristic peaks at 2954 and 3300 cm^{-1} arise from $\text{O}-\text{H}$ and $\text{N}-\text{H}$ stretching vibrations (hydrogen bonding in carboxylic acids and amides) [34]. After heat treatment and annealing, PAA is dehydrated to close the ring and form an imide five-membered ring, and both the original amide nitrogen ($-\text{NH}-\text{CO}-$) and protonated nitrogen ($-\text{NH}_3^+$) are converted to symmetrical imide nitrogen, which is clearly visible. The amide I band at 1650 cm^{-1} and the amide II band at 1549 cm^{-1} have a clear tendency to disappear.

The asymmetric and symmetric $\text{C}=\text{O}$ stretching vibrations of the imide band are responsible for the characteristic imide ring absorption peaks at 1775 and 1711 cm^{-1} , respectively, as illustrated in Fig. 3(f). As the heat treatment temperature is increased, the original peak at 1720 cm^{-1} shifts to the lower frequency of 1711 cm^{-1} due to the charge transfer of the π -electron cloud of RGO to the $\text{C}=\text{O}$ of the imide ring. The enhanced benzene ring vibration at 1500 cm^{-1} is suggestive of π - π stacking of PI with RGO. The imine group's $\text{C}-\text{N}$ stretching and $\text{C}=\text{O}$ bending modes are responsible for the other two new peaks, which are located at 1371 and 720 cm^{-1} [35]. It is evident from these new peaks that PAA-to-PI conversion was successful. All foams showed maxima at 1562 and 1053 cm^{-1} , which could be the result of $\text{N}-\text{H}$ deformation vibration [23]. The $\text{C}-\text{O}-\text{C}$ peak (1115 cm^{-1}) is derived from the $\text{C}-\text{H}$ stretching vibration, and it is noteworthy that the $\text{C}-\text{O}-\text{C}$ peaks are enhanced at 300 and 400°C , demonstrating the formation of an ether-bonded interfacial coupling between the PI and the RGO. The peak at 1172 cm^{-1} can be attributed to the $-\text{CO}-\text{NH}-$ covalent bonding in the PI, which overlaps with the $-\text{CO}-\text{NH}-$ covalent bonding between the PI and graphene [36, 37]. The $\text{C}=\text{C}$ peak (1600 cm^{-1}) in the aromatic ring in RGO is more pronounced at heat treatment temperatures of 200 – 300°C , whereas the new peak at 1560 cm^{-1} is attributed to the $\text{N}-\text{H}$ stretching vibration in the $\text{C}-\text{NH}-\text{C}$ band, suggesting a successful reaction between the amine and epoxy groups.

XPS of PGS before and after annealing in Figs. 3(g)–3(i) was used to characterize the changes in chemical structure before and after annealing. The full spectrum XPS shows characteristic peaks centered at 545.46 eV ($\text{O } 1s$), 400.39 eV ($\text{N } 1s$), and 284.8 eV ($\text{C } 1s$), which indicate that there is a large change in O and N before and after the reaction. The $\text{C } 1s$ spectra show peaks at 284.8 eV ($\text{C}-\text{C}$), 285 eV ($\text{C}-\text{N}$), 286.1 eV ($\text{C}-\text{OH}$), 287.6 eV ($\text{C}-\text{O}-\text{C}$), and 289.1 eV ($\text{C}=\text{O}$), where $\text{C}-\text{OH}$ is the characteristic peak of RGO and $\text{C}-\text{N}/\text{C}-\text{O}$ peaks originate from unclosed-loop PAA [38]. The characteristic peaks of $\text{C}-\text{N}/\text{C}-\text{O}$ were significantly weakened after heat treatment, while $\text{C}=\text{O}$ (imide) in $-(\text{C}=\text{O})-\text{N}-$ (five-membered ring) was significantly enhanced. This result is also seen in $\text{N } 1s$, where $-\text{NH}-\text{CO}-$ (unclosed ring amide nitrogen) located at 399.4 eV forms $-\text{N}-(\text{C}=\text{O})_2$ (five-membered cycloimide nitrogen) after thermal treatment [37, 39].

3.2 Absorption properties

By examining the electromagnetic characteristics of aerogels, the wave-absorbing properties of PGS are evaluated, and the impacts of composition and structure on electromagnetism are investigated to determine the relationship between structural

composition and wave-absorbing properties. The permeability and dielectric constant's real (ϵ' , μ') and imaginary (ϵ'' , μ'') components, respectively, represent the capacity to store and lose electromagnetic energy [40, 41]. Figure 4 presents the dielectric properties of aerogels containing varying SWNT concentrations after thermal treatment at different temperatures. The data reveal a consistent positive correlation between treatment temperature and dielectric constants. Specifically, PGS5-200 exhibited ϵ' ranging from 4.68 to 2.67 and ϵ'' from 3.27 to 0.98 , while PGS5-500 showed significantly enhanced values ($\epsilon' = 11.54$ – 3.92 , $\epsilon'' = 11.26$ – 3.54), as shown in Figs. 4(a) and 4(b). This enhancement stems from two concurrent processes: (1) progressive graphitization of graphene oxide at higher reduction temperatures and (2) PI carbonization in inert atmosphere generating amorphous carbon that repairs RGO defects and strengthens the conductive network, as confirmed by XRD and Raman spectroscopy [42]. The observed frequency dispersion effect, where both ϵ' and ϵ'' decrease with rising frequency, results from dielectric polarization hysteresis under alternating EMW electric fields [43]. This fluctuation in dielectric constants enhances electromagnetic wave loss susceptibility. It is worth noting that the increase of SWNT content in the PGS-300 samples produces a similar trend of dielectric enhancement. The dielectric constant of PG-300 without the addition of SWNTs as shown in Figs. 4(d) and 4(e) is very low, with the imaginary part of the dielectric constant being less than 1, and even close to 0 at high frequencies, which indicates that the electromagnetic loss capability of PG-300 is very low, whereas the increase in the dielectric constant with the addition of SWNTs is evident as PGS50-300 reaches $\epsilon' = 14.38$ – 3.8 and $\epsilon'' = 13.8$ – 4 (Figs. 4(d) and 4(e)), suggesting a substantial improvement in energy storage and dissipation capability. However, SWNT incorporation exhibits a dual effect: while enhancing conductive loss and interfacial polarization, excessive SWNT loading (beyond 25–300 ratio) creates an overly dense conductive network that impedes electromagnetic wave penetration. Notably, all PGS samples exhibit similar dielectric behavior across the 2–18 GHz range, characterized by a gradual decrease from 2–8 GHz followed by multiple relaxation processes between 8–18 GHz, where three distinct resonance peaks appear. This behavior confirms significant polarization loss contributions. The dielectric loss capability, evaluated through $\tan \delta_\epsilon = \frac{\epsilon''}{\epsilon'}$, shows temperature-dependent enhancement (Fig. 4(c)). Both PGS5-200 and PGS5-300 display comparable decreasing trends, with PGS5-300 demonstrating particularly strong loss performance ($\tan \delta_\epsilon$ decreasing from 1.1 to 0.57) along with multiple resonance peaks in the 8–18 GHz range. This improvement stems from the thermal imidization of PAA to PI at 300°C , which generates numerous microcapacitor structures with RGO/SWNTs serving as conductive electrodes and PI as the dielectric layer, thereby significantly boosting interfacial polarization loss through ϵ'' enhancement. For samples treated at 400 – 500°C , the dielectric loss capacity stabilizes around $\tan \delta_\epsilon \approx 0.9$, suggesting that both graphene oxide reduction and PAA imidization processes reach completion by 400°C . In contrast, the samples with higher SWNT content (PGS15-300, PGS25-300) show an increasing trend similar to the increase in annealing temperature, while a significant decreasing trend starts to appear when the concentration of SWNTs reaches 50, and this decrease may be attributed to the skin-converging effect in the overdense conductive network that hinders the penetration of electromagnetic waves [41], while according to free electron theory [44]

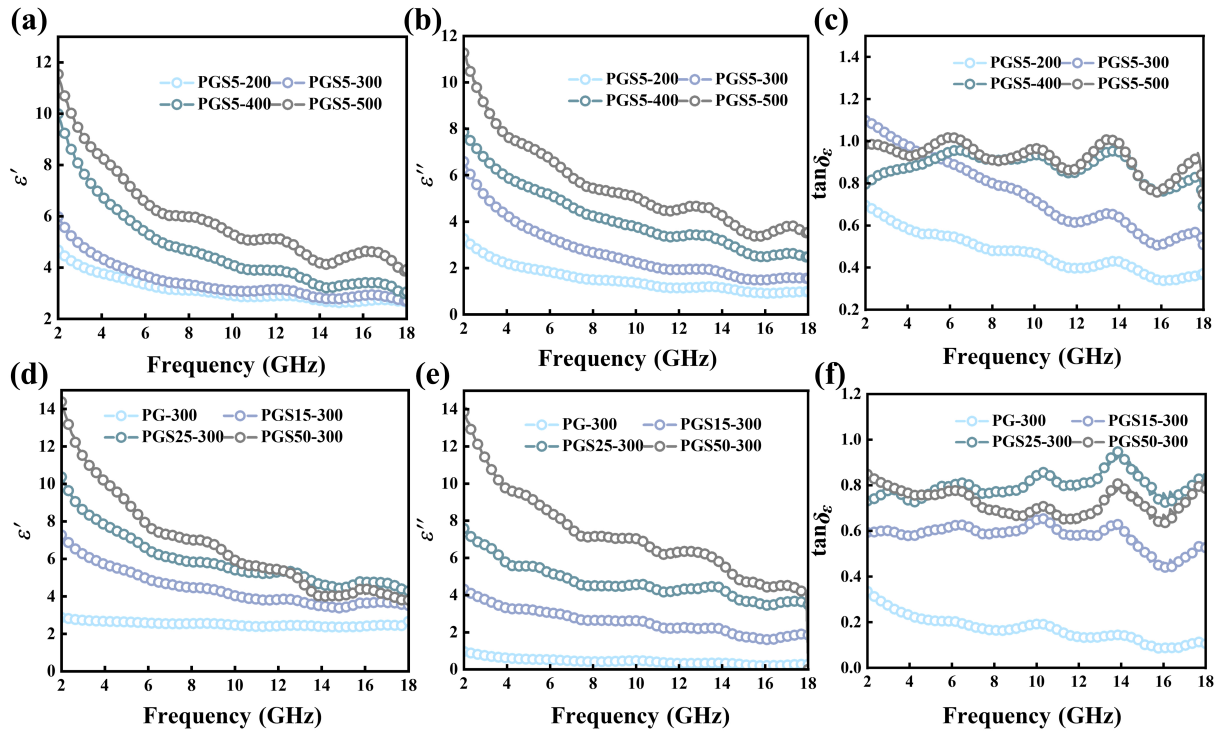


Figure 4 (a) Real part of the dielectric constant of PGS5 at different temperatures. (b) Imaginary part of the dielectric constant of PGS5 at different temperatures. (c) Tangent of the dielectric loss of PGS5 at different temperatures. (d) Real part of the dielectric constant of PGS-300 with different SWNTs contents. (e) Imaginary part of the dielectric constant of PGS-300 with different SWNTs contents. (f) Tangent of the dielectric loss of PGS-300 at different temperatures.

$$\varepsilon'' = \frac{1}{2\pi\rho f\varepsilon_0} \quad (2)$$

where ρ and ε_0 denote the resistivity and vacuum dielectric constant, the conductivity directly determines the dielectric loss capability of the material, so the loss capability can be designed and regulated by the conductivity. Since conductivity directly affects a material's dielectric loss capability, it can be used to design and control the loss capability. This is where resistivity and vacuum dielectric constant are shown. It is observed that μ' of PGS aerogel at different temperatures and carbon nanotube concentrations is close to 1, μ'' is close to 0, and $\tan\delta_\mu$ is also close to 0 (Fig. S3 in the ESM), and $\tan\delta_\varepsilon$ is much larger than $\tan\delta_\mu$. As a result, dielectric loss predominates in PGS composite aerogel and magnetic loss is essentially absent.

The ideal dielectric constant shows a sharp decrease with increasing frequency in the low-to-mid band range, while thinner materials exhibit a more pronounced reduction. As illustrated in Figs. S4 and S5 in the ESM, the dielectric constant and 2D reflection loss plots for the ideal case (1–5 mm thickness) demonstrate that full-band absorption (2–18 GHz) remains achievable even at reduced thicknesses. A comparison between the actual and ideal dielectric constants is presented in Figs. S6 and S7 in the ESM. In practice, the dielectric properties of microwave-absorbing materials typically follow the Debye relaxation model [45], where dielectric loss originates from the hysteresis of polarization under an external electromagnetic field. When the polarization rate matches the field oscillation frequency, strong polarization occurs, generating a distinct peak in the imaginary part of the dielectric constant (Figs. 4(b) and 4(e)). This significantly enhances the overall dielectric response. However, at higher frequencies, the polarization process fails to synchronize with the rapidly oscillating field,

resulting in diminished polarization and a consequent decline in the dielectric constant. This frequency-dependent behavior is quantitatively characterized by Eqs. (3) and (4) [46, 47]

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2} \quad (3)$$

$$\varepsilon'' = \omega\tau \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2} \quad (4)$$

The angular frequency is represented by $\omega = 2\pi f$, with ε_∞ , ε_s , and τ denoting the relative permittivity, static permittivity, and polarization relaxation time, respectively. Since microwave absorption primarily occurs through dielectric relaxation processes, the relaxation time τ serves as a key parameter influencing the absorption characteristics. Zhang et al. established that longer relaxation times result in dielectric constants approaching ideal values, which enhances broadband microwave absorption through dielectric constant normalization [45]. A comprehensive understanding of the microwave attenuation mechanism in PGS aerogels requires detailed examination of dielectric loss components. According to Debye's theory, ε'' can be decomposed into conductive loss and polarization loss contributions, as expressed in Eq. (6) where the first term represents polarization loss and the second term corresponds to conductive loss. In the low frequency regime, non-magnetic materials exhibit predominantly conductive loss behavior, enabling Eq. (5) to be simplified to Eq. (6). This formulation reveals a linear $\varepsilon''-1/f$ relationship that permits calculation of material conductivity in the GHz range through slope analysis. The derived conductivities for various PGS aerogels (PG-300, PGS5-200, PGS5-300, PGS5-400, PGS5-500, PGS15-300, PGS25-300, and PGS50-300) in the 2–5 GHz range are 0.09, 0.246, 0.546, 0.552, 0.782, 0.251, 0.497, and 0.939 S/m

respectively, as shown in Figs. 5(a) and 5(d)). This is in general agreement with the actual conductivity of the aerogel (Fig. S17 in the ESM). Analysis of low-frequency dielectric data (Figs. 5(b), 5(c), 5(e), and 5(f)) demonstrates that polarization loss constitutes the principal dissipation mechanism, with conductive loss contributions being quantitatively determined through this approach.

$$\varepsilon'' = \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2 \tau^2} \omega \tau = \varepsilon_p'' + \varepsilon_c'' = \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2 \tau^2} + \frac{\sigma}{\omega \varepsilon_0} \quad (5)$$

$$\varepsilon'' = \frac{\sigma}{2\pi f \varepsilon_0} \quad (6)$$

The dielectric loss (ε'') is determined by a combination of conductivity loss (ε_c'') and polarization relaxation loss (ε_p''). Figures 5(b) and 5(e) show a general decreasing trend in the conductivity of the PGS aerogel samples, with the conductivity loss concentrated in the low frequency region. As the temperature increases the overall conductivity increases, due to the thermal reduction compensating for the defects on the graphene surface, It

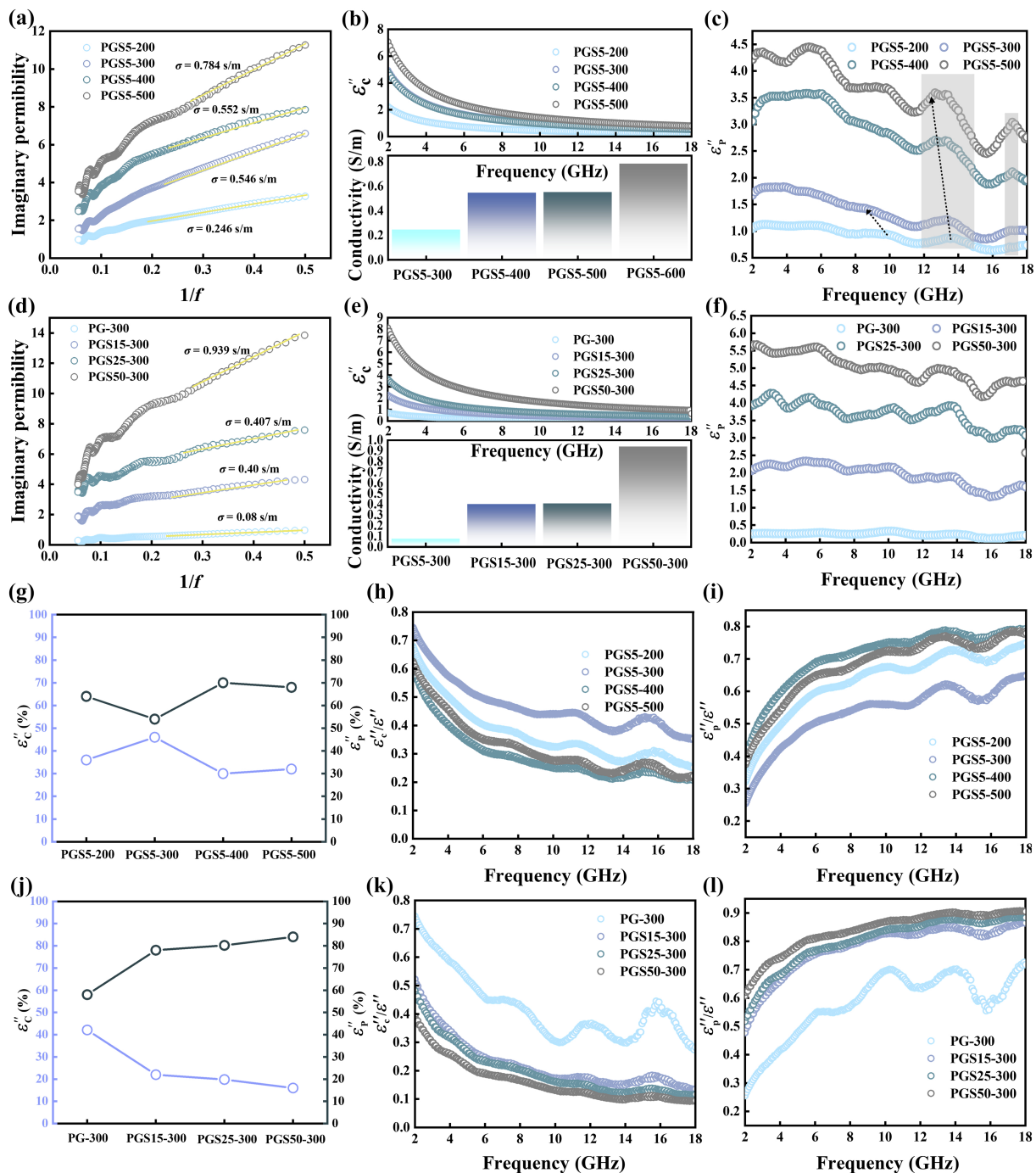


Figure 5 (a) and (d) The ε'' – $1/f$ curves of PGS aerogels. (b) and (e) Conductivity losses. (c) and (f) Polarization loss. (g) Average conductivity loss vs. polarization loss at different temperatures. (h) Conductivity loss at 2 to 18 GHz for PGS5 at different temperatures. (i) Polarization loss at 2 to 18 GHz for PGS5 at different temperatures. (j) Average conductivity loss vs. polarization loss at different SWNTs contents. (k) Conductivity loss at 2 to 18 GHz for PGS-300 at different SWNTs contents. (l) Polarization loss of PGS-300 at different SWNTs contents.

is worth noting that the very significant increase in overall conductivity from 200 to 300 °C heat treatment is due to the fact that thermal imidization is essentially complete at 300 °C, whereas the increase in overall conductivity from 400 to 500 °C comes from the deeper reduction of GO and the carbonization of PI. At the same time, there is a steady increase in the overall conductivity of PGS at the same temperature as the content of SWNTs increases. The conductivity has two abrupt changes here, one is the increase in the incorporation of SWNTs from 0.09 to 0.40, which can also be seen from the change in the dielectric constant, and the other one is the increase in the amount of SWNTs to 50 mg and the increase in the conductivity to 0.939 s/m, which may be induced by the amount of SWNTs reaching a threshold value. Therefore, the conductive network inside the material shows a positive correlation with the conductivity loss. However, while excessive conductivity can generate more induced currents and thus stronger conductive losses, the resulting conductive network turns the material itself into an electromagnetic shield, preventing more electromagnetic waves from entering the material, which can result in impedance mismatch.

Figure 5(g) presents the relative contributions of conductivity loss and polarization loss at varying temperatures, clearly demonstrating that dielectric loss primarily originates from

polarization relaxation mechanisms. Frequency-dependent analysis across 2–18 GHz reveals distinct loss behavior in PGS aerogels: conductivity loss dominates in the lower frequency range (2–8 GHz), while polarization relaxation becomes the predominant loss mechanism throughout the entire measured spectrum (8–18 GHz), as evidenced in Figs. 6(h) and 6(i). The introduction of SWNTs significantly influences these loss characteristics, as shown in Fig. 6(j), where their incorporation creates an extensive conductive network that enhances both interfacial effects and defect-mediated polarization. This network development leads to progressively stronger polarization loss with increasing SWNTs content, exhibiting more pronounced impact on conductive loss compared to temperature variations. These observations align with established research confirming that microwave attenuation in PGS composite aerogels results from the combined effects of polarization and conduction losses, each dominating specific frequency regimes. The polarization loss principally arises from interfacial polarization at RGO-SWNT-PI junctions and dipole polarization induced by inherent defects in graphene oxide.

Observing Figs. 5(c) and 5(f) polarization loss part of the PGS aerogel we can find the existence of peaks around 17 and 13 GHz, different annealing temperatures and different additions of SWNTs affect the enhancement of his peaks, and it is noteworthy that the

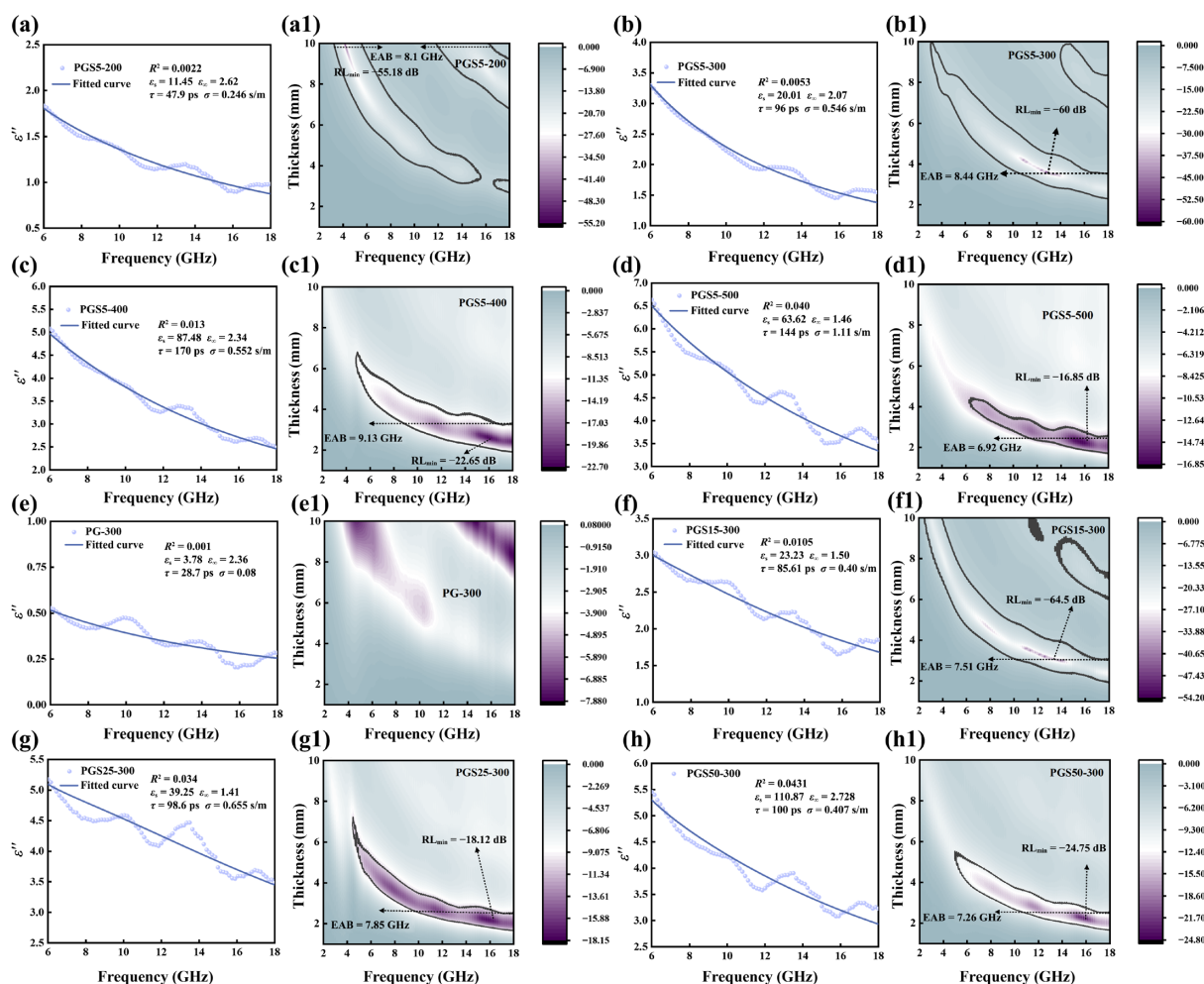


Figure 6 Imaginary part curves of dielectric constant fitted using the Debye model: (a) PGS5-200, (b) PGS5-300, (c) PGS5-400, (d) PGS5-500, (e) PG-300, (f) PGS15-300, (g) PGS25-300, and (h) PGS50-300. 2D reflection loss mapping of (a1) PGS5-200, (b1) PGS5-300, (c1) PGS5-400, (d1) PGS5-500, (e1) PG-300, (f1) PGS15-300, (g1) PGS25-300, and (h1) PGS50-300.

peaks are shifted towards the low frequency region as the annealing temperature is increased from 200 to 500 °C. Theoretically, a peak represents the polarization relaxation process, where the polarization relaxation time (τ) can be determined using Eq. (7) as follows

$$\tau = \frac{1}{2\pi f_m} \quad (7)$$

The frequency (f_m) corresponding to peak polarization maximum reveals distinct dielectric relaxation characteristics. For PGS5-200 and PGS5-300, three relaxation processes occur at 17 GHz ($\tau = 9.3$ ps), 13.5 GHz ($\tau = 11.7$ ps), and 10 GHz ($\tau = 15.9$ ps). Higher-temperature treated samples (PGS5-400 and PGS5-500) exhibit four relaxation processes at 17 GHz ($\tau = 9.3$ ps), 12.5 GHz ($\tau = 12.2$ ps), 9.5 GHz ($\tau = 16.8$ ps), and 5.3 GHz ($\tau = 30$ ps). Notably, the 13.55 GHz peak shifts to 12.5 GHz with increasing temperature, accompanied by a relaxation time increase from 11.8 ps to 12.58 ps (Fig. S15 in the ESM). In contrast, SWNT content variation affects only polarization peak intensity while maintaining constant peak positions. These findings demonstrate that annealing temperature can effectively modulate dielectric relaxation times. Good impedance matching and excellent loss capability is the key to achieve efficient microwave absorption capability, and it is difficult to achieve efficient loss for materials with excellent impedance matching capability, so we usually modulate ϵ' and ϵ'' , which have strong microwave loss capability, by ϵ' and ϵ'' , and according to the Derbyshire relaxation model τ determines the dispersion (frequency response) positions of ϵ' and ϵ'' : ϵ'' peaks when $2\pi f_m \tau = 1$ and ϵ' undergoes its sharpest drop (from ϵ_s to ϵ_∞) around this frequency. As the heat treatment temperature increases, the ϵ'' -polarization peak of f_m has a clear tendency to move to the left, while ϵ' all maintains a continuous decreasing trend, which is a result of the increase in the polarization relaxation time due to the heat treatment which in turn affects the wave-absorbing properties.

Further we fitted the imaginary part of the dielectric constant with the Debye model and evaluated the dielectric loss capability of the PGS aerogel, which correlates with the theoretical dielectric constant in the 8–18 GHz frequency range (Figs. S6 and S7 in the ESM). The electromagnetic wave (EMW) absorption performance of a material is typically characterized by two key parameters: the reflection loss (RL) and the EAB. The EAB is defined as the frequency range where the reflection loss reaches ≤ -10 dB, indicating 90% wave absorption. These parameters can be derived from the material's complex permittivity ($\epsilon = \epsilon' - j\epsilon''$) and permeability ($\mu = \mu' - j\mu''$) through the following transmission line theory equations [48]

$$RL \text{ (dB)} = 20 \lg \left| \frac{Z_{in} - 1}{Z_{in} + 1} \right| \quad (8)$$

$$Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\epsilon_r}} \tan \left[j \left(\frac{2\pi f d}{c} \right) \sqrt{\epsilon_r \mu_r} \right] \quad (9)$$

where Z_0 represents the characteristic impedance in free space (377 Ω), and Z_{in} , d , f , and c represent the input impedance, material thickness, respectively, frequency, and propagation speed of the EMW in vacuum. The dielectric behavior of PGS aerogels is effectively characterized by the Debye model, as evidenced by the high coefficients of determination ($R^2 \approx 1$) shown in Figs. 6(a)–6(h),

indicating strong agreement between experimental and theoretical data. Analysis of microwave absorption properties (Figs. 6(a1)–6(h1)) reveals two key trends: first, elevated temperatures significantly reduce the required absorber thickness while improving performance—the EAB is maintained from 9.3 mm (PGS5-200) to just 2.5 mm (PGS5-500), with a concurrent bandwidth expansion from 8.1 to 9.13 GHz as relaxation times increase. Second, SWNT incorporation produces similar EAB enhancement but through different mechanisms: while temperature primarily affects relaxation times (Fig. 6), increased SWNT loading amplifies interfacial polarization through additional phase boundaries, without altering the characteristic polarization peak positions (Fig. 5(f)). Based on the 2D reflection losses shown, the wave absorbing properties of PGS25-300 and PGS50-300 are degraded due to the formation of a conductive network with a high content of SWNTs, which improves the loss capability but also blocks the entry of electromagnetic waves into the interior of the material. This consistency between the fixed polarization peaks and fitted relaxation times confirms the reliability of our dielectric analysis.

Further. We provide an understanding of the relaxation process from the dielectric loss point of view using Cole–Cole curves. The following equation provides the link between ϵ' and ϵ''

$$\left(\epsilon' - \frac{\epsilon_s + \epsilon_\infty}{2} \right)^2 + (\epsilon'')^2 = \left(\frac{\epsilon_s - \epsilon_\infty}{2} \right)^2 \quad (10)$$

A form of dipole and interfacial polarization is represented by each downward semicircular curve in the Cole–Cole diagram, where ϵ' is the horizontal coordinate and ϵ'' is the vertical coordinate. From Figs. 7(c) and 7(d), and Fig. S8 in the ESM, it can be seen that the PGS aerogel produces multiple polarization relaxation mechanisms in the 2–18 GHz band, which may be attributed to the dielectric relaxation induced by interfacial polarization and dipole polarization relaxation caused by the abundant SWNTs/RGO/PI heterojunctions and carbon-paraffinic matrices in the PGS composite aerogel material, attenuating the incident electromagnetic wave. It is noteworthy that the interfacial polarization shown in the figure is mainly distributed in 10–18 GHz. For PGS5-300, the interfaces include RGO/SWNT, RGO/PI, SWNT/PI and the interface between the sample and the paraffin, all of which lead to the interfacial polarization. PGS5-400 and PGS5-500 have high permittivity due to the high thermal reduction temperature and the gradual carbonization of some PIs, which in turn leads to high permittivity, and PGS25-300 and PGS50-300 have a dense conductive network due to the excess of SWNTs and the skinning effect that affects the entry of electromagnetic waves. The low permittivity of PGS5-400 has the effect of a large amount of interfacial polarization loss and relatively low permittivity, which leads to a wider frequency range and a higher frequency of range microwave absorption. The conductivity loss of the material is shown by an approximate straight line in the upper right corner of the Cole–Cole diagram for all PGS composite aerogels. This suggests that conduction loss is the primary factor causing EMW attenuation at low frequencies. The conductivity and the line's slope have a positive correlation, which is advantageous for the wave-absorbing properties. This is consistent with the discussion of the Debye model above.

Figure 9 summarizes the PI/rGO/MnFe₂O₄ composite aerogel's electromagnetic wave absorption process based on the prior discussion. PMG aerogel has a directed pore structure that lets more electromagnetic waves into its interior than other wave-absorbing materials. This is useful for optimizing impedance

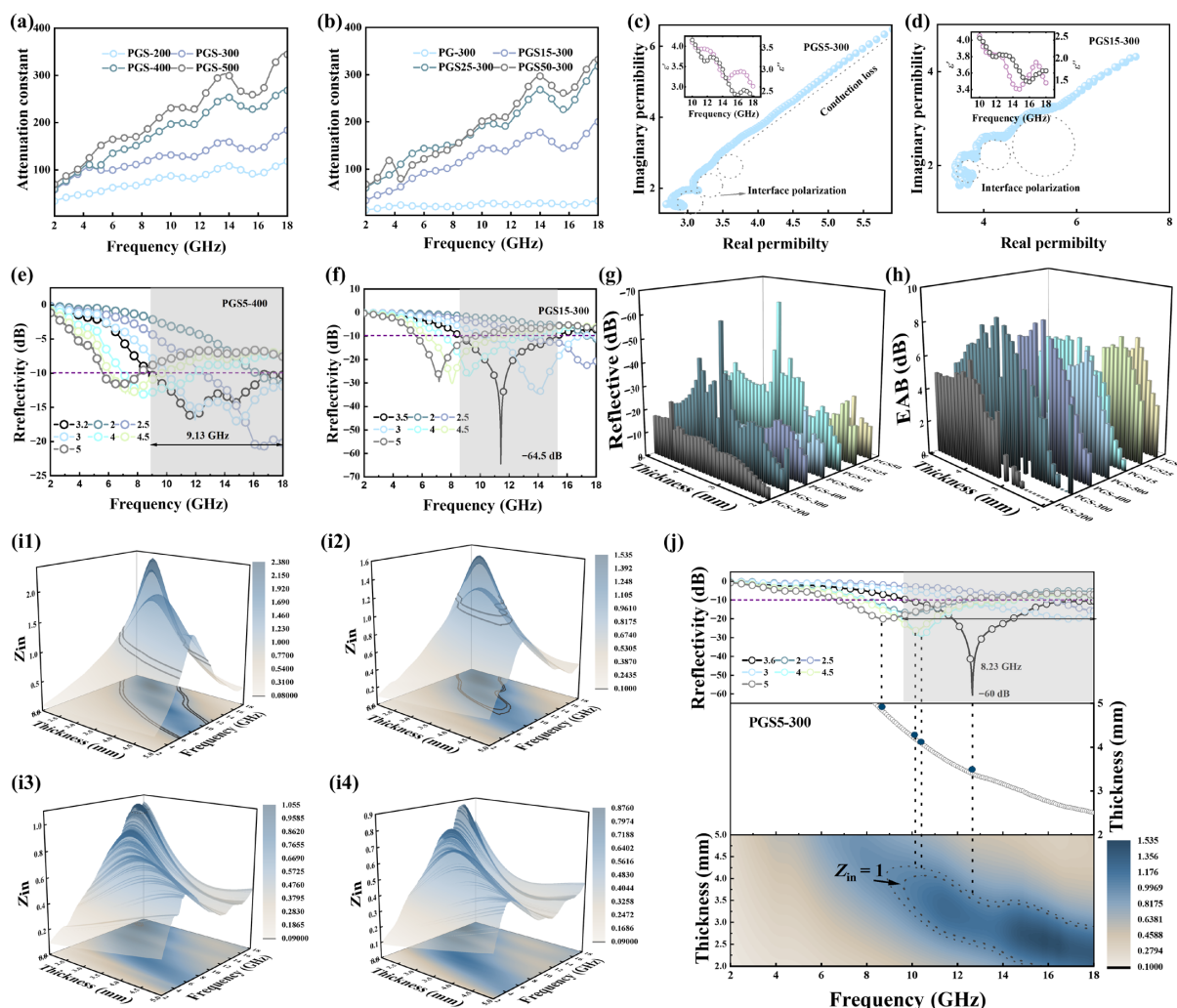


Figure 7 (a) Attenuation coefficient of PGS5. (b) Attenuation coefficient of PGS-300. Cole-Cole semicircle of PGS composite aerogel materials in the frequency range of 2 to 18 GHz: (c) PGS5-300, (d) PGS15-300, and (e) PGS5-400. (f) PGS15-300 absorption performance chart. (g) Minimum absorption of PGS composite aerogel at different thicknesses of small. (h) EABs of PGS composite aerogel at different thicknesses of small. (i) Impedance matching diagram of PGS-5 at different heat treatment temperatures. (j) Comparison of microwave reflection loss, quarter-wavelength matching theory of PGS5-300.

The microwave absorption and dielectric properties of PGS composite aerogels are significantly influenced by the SWNT concentration and GO reduction temperature. The reduction temperature controls the conductivity of graphene while preserving its flake morphology, which facilitates multiple internal reflections of electromagnetic waves. Incorporated SWNTs serve dual roles by preventing graphene restacking and creating conductive networks that enhance both conductivity and interfacial polarization. A uniform SWNT distribution on GO lamellae improves dipole polarization, increasing the dielectric constant and loss factor to boost absorption efficiency. At low SWNT concentrations and reduction temperatures, discontinuous conductive networks limit dielectric loss. However, exceeding a critical SWNT threshold induces nonlinear dielectric enhancement through rapid pathway formation, even with partially reduced graphene. The aerogel's oriented pore structure further enhances performance by aligning conductive channels with the electric field vector. Optimal microwave absorption thus requires balanced SWNT loading and reduction temperature to achieve sufficient conductivity without excessive reflection while maximizing interfacial polarization and maintaining structural anisotropy. These parameters enable precise

tuning of the aerogel's dielectric properties and absorption characteristics. The reflection loss analysis of temperature-treated PGS aerogels reveals distinct microwave absorption characteristics across different frequency and thickness ranges (2.0–18.0 GHz, 2.0–5.0 mm) (Fig. S9 in the ESM). Initial testing shows PGS5-200 exhibited limited absorption ($RL_{\min} = -11.82$ dB at 11.66 GHz, 4.5 mm), while higher temperature treatments significantly improved performance. PGS5-300 achieved optimal absorption ($RL_{\min} = -60$ dB at 12.66 GHz, 3.63 mm) (Fig. 7(j)), with PGS5-400 and PGS5-500 showing $RL_{\min}$ values of -22.59 dB (15.92 GHz, 2.6 mm) and -25.16 dB (16.54 GHz, 2.4 mm) respectively. Notably, PGS5-300 demonstrated superior broadband absorption with an 8.23 GHz EAB (9.77–18 GHz) at 3.63 mm, while PGS5-400 achieved the widest bandwidth (9.12 GHz, 8.88–18 GHz) (Fig. 7(e)) despite its moderate $RL_{\min}$. SWNT concentration variations similarly influenced absorption properties. PGS15-300 showed exceptional $RL_{\min}$ (-64.37 dB at 11.42 GHz, 3.5 mm) (Fig. 7(f)) but reduced EAB (6.7 GHz). Higher SWNT loadings decreased both $RL_{\min}$ (-24.75 dB at 16.1 GHz and -18.12 dB at 15.82 GHz) and required thickness (2.2 mm), while maintaining reasonable EABs (7.26 and 6.4 GHz). These results indicate that the heat treatment

temperature can modulate the polarization relaxation time and thus enable the samples to obtain a wider effective absorption bandwidth at lower thicknesses, and that the addition of SWNTs leads to a better dielectric loss capability for PGS aerogels. Excessive SWNTs create a richer conductive network, which raises the conductive loss of the network while also somewhat raising the dielectric constant [49]. However, it also has an impact on how electromagnetic waves enter the material's interior, which results in an impedance mismatch. In addition, the directed pore structure lengthens the electromagnetic radiation's transmission channel, which improves the polarization loss [50] and multiple scattering loss [51] of the radiation between the carbon nanotubes and the pore walls. It is important to note that PGS aerogels have broadband wave-absorbing properties because of their special oriented pore structure (Fig. 7(h)). The depletion capacity increased with increasing heat treatment temperature and concentration of SWNTs, while the displayed RL_{min} showed an enhancement followed by a decrease (Fig. 7(g)). This is because the rich conductive network leads to an impedance mismatch, which prevents the effective entry of electromagnetic radiation. Two requirements must be met for excellent wave-absorbing properties: First, the electromagnetic waves must be able to enter the aerogel's interior with the proper impedance matching; second, the ideal wave-absorbing material is one in which the electromagnetic waves fully penetrate the interior, meaning that the material's impedance equals that of the free space [52]. The other is the ability to adequately disperse the EMW that enters the aerogel's interior. The ability of a material to attenuate EMW is typically expressed by its attenuation coefficient. The wave-absorbing material uses a variety of loss mechanisms, such as conduction loss, polarization relaxation loss, eddy current loss, etc., to dissipate the EMW that enters the material as other forms of energy, such as heat. By increasing attenuation coefficients and impedance matching, absorbent materials can improve their wave-absorbing capabilities. An absorber's attenuation and impedance matching constants are written as [53]

$$Z = \left| \frac{Z_{in}}{Z_0} \right| = \sqrt{\frac{\mu_r}{\epsilon_r} \tanh \left(j \frac{2\pi f d}{c} \sqrt{\mu_r \epsilon_r} \right)} \quad (11)$$

$$\alpha = \frac{2\pi f}{c} \times \sqrt{(\mu''\epsilon'' - \mu'\epsilon') + \sqrt{(\mu''\epsilon'' - \mu'\epsilon')^2 + (\mu'\epsilon'' - \mu''\epsilon')^2}} \quad (12)$$

The design of the wave-absorbing material can have the impedance matching coefficient converge to 1 in a wider frequency range when $Z = 1$, which means that more electromagnetic waves can enter the interior of the aerogel because the input impedance is equal to the air impedance. This can increase the absorption efficiency of the wave-absorbing material in a wider range of electromagnetic wave frequencies [54]. Z values for all samples with varying thicknesses at 2–18 GHz are displayed in Fig. 7(i) and Fig. S10 in the ESM, and it is evident that PGS5-400, PGS5-500, and PGS15-300, which have lower heat treatment temperatures and lower SWNTs concentrations, have more regions with Z close to 1 (the area between the two grey straight lines). As the temperature of reduction increases, PGS5-400 and PGS5-500 go from a point to a region where there is no Z close to 1. The same situation occurs with the increase in the concentration of SWNTs at the same temperature, which is due to the increasing graphitization and conductivity of the graphite as a result of the increase in the

temperature of reduction and the increase in the concentration of SWNTs, which affects the entry of electromagnetic waves into the interior of the aerogel. As shown in Figs. 7(a) and 7(b), The α value has a positive correlation with PGS aerogels' capacity to attenuate EMW. The increase in graphitization brought on by the rise in reduction temperature and the rise in SWNTs concentration to create a more homogeneous conductive network may be the reason why the α -value of PGS composite aerogels rises at the same frequency as the concentration of SWNTs and the reduction temperature. All of the samples' attenuation constants exhibited an increasing trend as frequency increased. The samples exhibits superior attenuation constants when the temperature and concentration of SWNTs grow. This is because polarization loss is enhanced in addition to the conductive loss capabilities increasing. Meanwhile, when the absorber approaches the matching thickness, which satisfies the quarter wavelength, the experimental results are in good agreement with the theory (tm) to further examine the electromagnetic impedance matching of the materials (Fig. 7(j)). We fabricated 180 mm × 180 mm × 3.6 mm aerogel of PGS5-300 and tested its actual wave-absorbing properties, and the results were close to those of our simulations (Fig. S14 in the ESM). The reflected wave at the interface, which is out-of-phase at the composite interface of the PGS composite aerogel, may partially cancel the incident wave. The phase shift causes incident microwave attenuation, and tm meets the quarter-wavelength matching theory, as seen in Fig. 7(j). The PGS-300's RL shifts from the Ku-band to the X-band, which has a relatively low frequency, as tm grows. The optimal impedance match, which is in line with the experimental thickness Tm, is the optimum reflection loss when combined with the RL curves. Tm EXP is indicated by the blue circle, which coincides with the majority of the $\lambda/4$ curve. Therefore, it is valuable to design the thickness of PGS composites based on the quarter-wavelength matching theory.

To evaluate the practical applicability of PGS aerogels for electromagnetic wave absorption, radar cross-section (RCS) simulations were performed. The RCS serves as a quantitative measure of a target's electromagnetic scattering characteristics, where a reduced RCS value directly correlates with enhanced absorption capability. These simulations analyze the material's echo signal intensity when illuminated by incident electromagnetic waves, providing critical performance metrics for potential real-world applications [55]. The model utilized in the RCS simulation is displayed in Fig. 8(e). The simulation is modeled as a 180 mm square with two layers: a PGS aerogel layer (the simulated thickness is the material's ideal absorption thickness) and a 0.5 mm perfect electrical conductor (PEC). The incident wave is incident along the negative Z-axis. The PEC, PGS5-300, PGS15-300, and PGS5-400 3D radar wave scattering signals are displayed in Figs. 8(a)–8(d), respectively. It is evident that the PEC is perfectly reflected on both the front and rear sides. At aerogel thicknesses of 3.63 and 3.5 mm, PGS5-300 and PGS15-300 are completely absorbed at the frontal surface, however, PGS5-400 shows a noticeable scattering signal even at the ideal absorber thickness of 3.2 mm. It is obvious that the PGS has a far lesser scattering signal than the PEC absorber, which has the greatest scattering signal. In particular, the vertical reflection intensity of PGS5-300 is as high as −42.05 dB·m².

The absorber's 2D RCS values for incidence angles ranging from −90° to 90° are displayed in Fig. 8(g). We can see that the RCS values of both the PGS5-300 and PGS15-300 composite aerogels are less than −20 dB·m². In order to study the effect of the PGS aerogel

on the RCS more intuitively, the RCS reduction can be calculated by subtracting the RCS value of the PEC from the RCS value of the PGS aerogel (Fig. 8(f)). The results showed that all PGS aerogels were able to reduce RCS, with only PGS5-300 and PGS15-300 absorbers being more capable. According to the simulation results, PGS composite aerogel materials are expected to be an excellent EMW absorbing material because they have both RL performance and EMW attenuation performance. This oriented pore structure is more favourable for EMW entry and more called favourable for impedance matching of the material. Compared with other carbon materials, PGS composite aerogels have superior wave absorption

performance and effective bandwidth [26, 56–61] (Fig. 8(h)). The power loss models for various PGS composite aerogel structures are displayed in Fig. 8(i). Under comparable test conditions, the composite aerogel with oriented microchannels has a richer power loss density than the composite aerogel with a homogeneous pore structure, which is also consistent with our earlier real-world experiments [21].

Figure 9 illustrates the electromagnetic wave absorption mechanism of the PGS composite aerogel. The oriented pore structure demonstrates two key advantages: (1) enhanced wave penetration into the aerogel's interior due to reduced surface

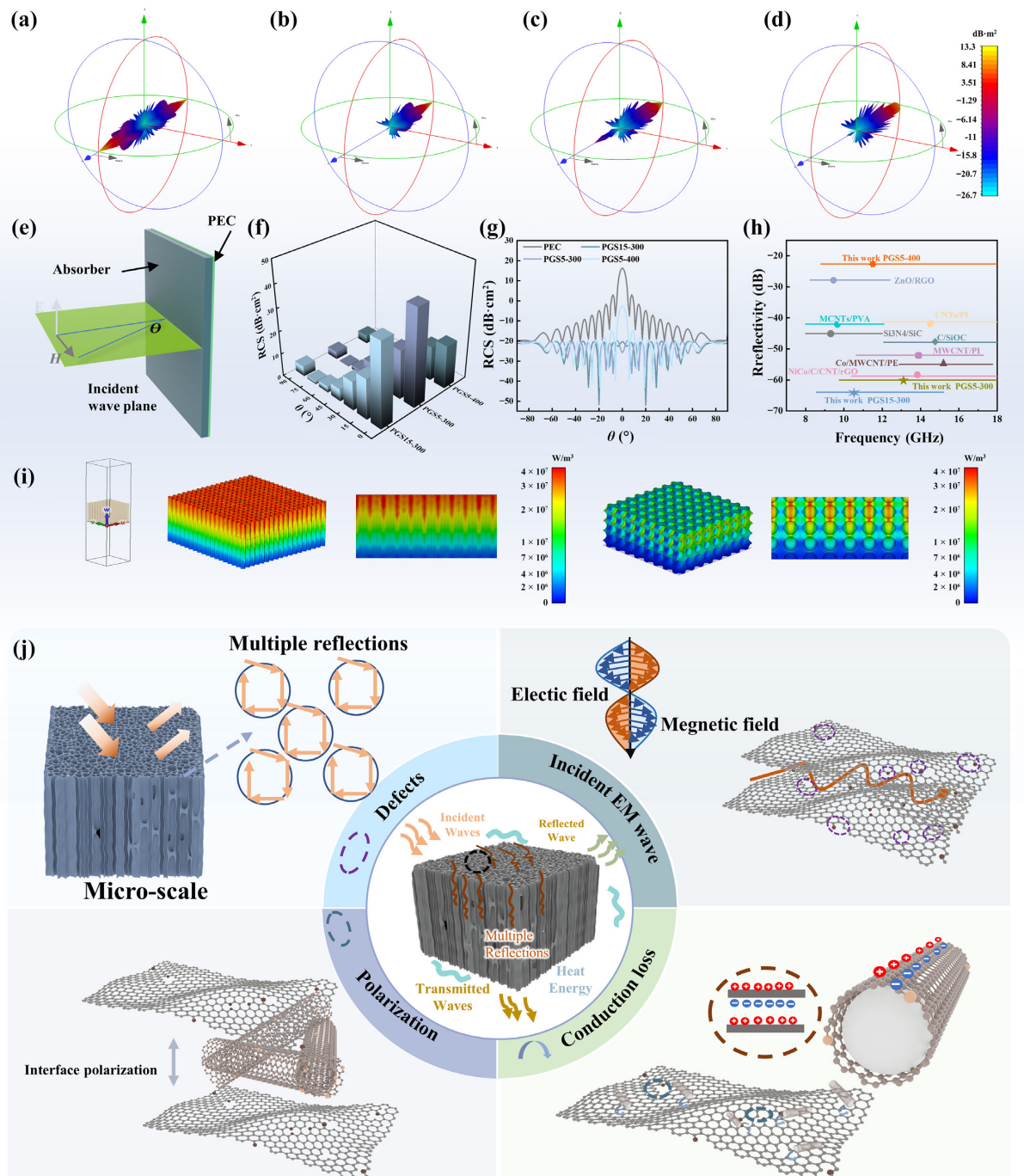


Figure 8 CST simulation results: (a) PEC, (b) PGS5-300, (c) PGS5-400, and (d) PGS15-300. (e) RCS simulation model. (f) RCS reduction of PGS. (g) RCS values of the samples with different angles. (h) Comparison of the wave absorption performance with other aerogels with different carbon systems. (i) Power composite aerogels with different pore structures loss of composite aerogels with different pore structures. (j) Wave absorption mechanism diagram.

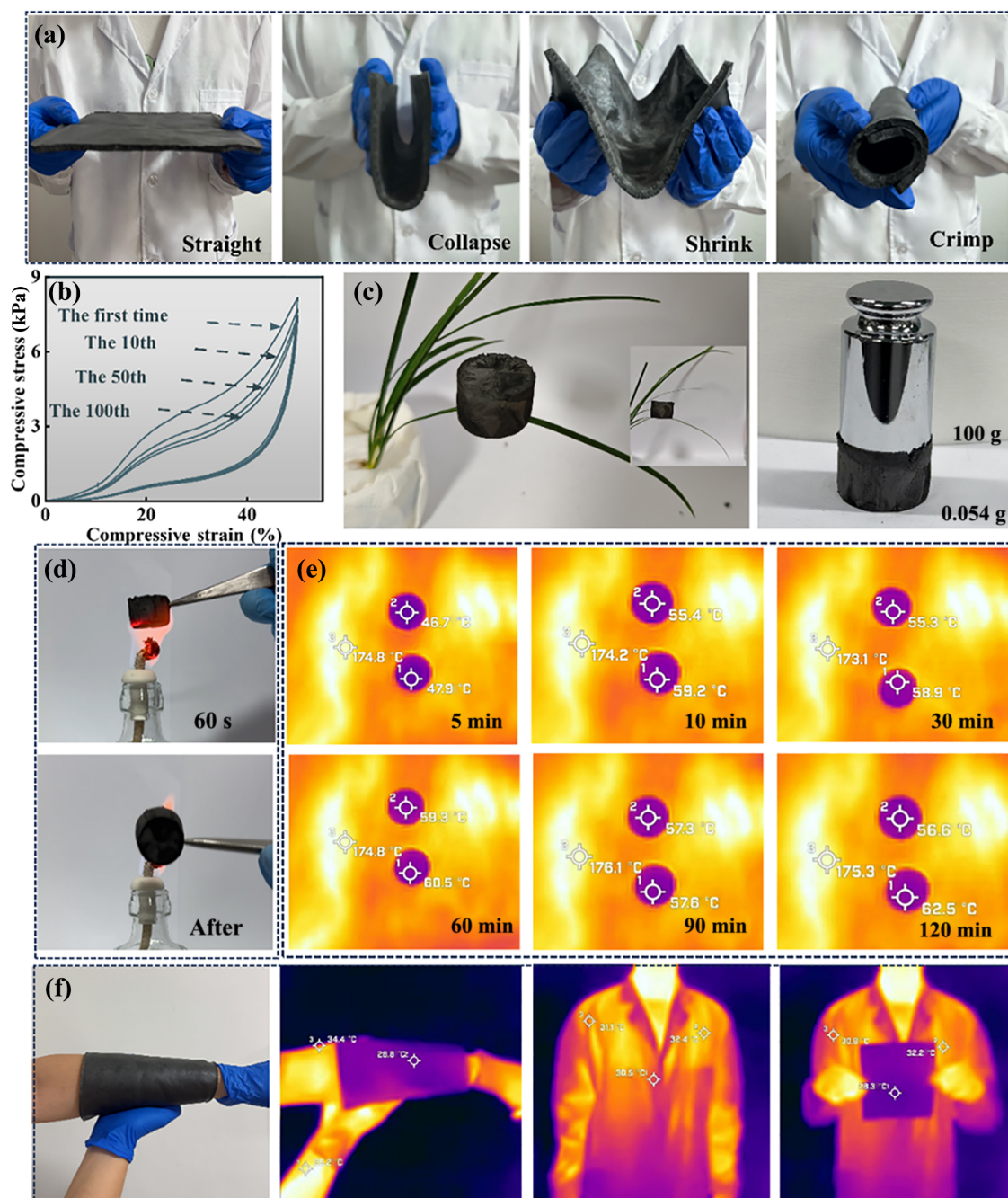


Figure 9 (a) Digital image of PGS aerogel material after folding, kneading and curling. (b) Cyclic compression test of PGS composite aerogel. (c) Digital photographs of ultralight aerogel assisted on a single blade of grass and 0.054 g sample weighing 100 g. (d) Digital photographs of PGS composite aerogel before and after burning for 60 s on an alcohol burner flame. (e) Thermal insulation performance graph of PGS composite aerogel. (f) PGS aerogel fabric for infrared stealth.

reflection, and (2) improved impedance matching characteristics compared to conventional wave absorbers and isotropic aerogel structures. This unique architecture facilitates more efficient electromagnetic energy dissipation through multiple scattering and absorption events within the three-dimensional conductive network. The incident EMW's path length is increased and the absorption efficiency is raised by multiple reflections and scattering within the porous structure. The energy is gradually attenuated by repeated EMW scattering in this microstructure, increasing the absorption capacity. The reflection loss linked to EMW is successfully reduced by this multiple reflection and scattering

phenomenon. Furthermore, the polarization effect is further enhanced by the PGS composite aerogel's unique oriented pore structure, which aligns the conductive channels with the direction of the electric field vectors. Second, with a sufficient reduction temperature, graphene, a dielectric-tunable material, can create a continuous conducting network in the composite aerogel. In the meantime, the addition of SWNTs not only creates a clear conductive network that generates considerable dielectric and conduction losses when exposed to EMW, but it also successfully stops graphene layers from re-stacking during reduction, which is more advantageous for the effects of electron migration, hopping,

and tunneling. A conductive loss process that transforms electromagnetic radiation into thermal energy mediates these occurrences [62]. Notably, GO, SWNTs, PI, and air produce a significant number of heterogeneous interfaces that result in interfacial polarization and charge accumulation [63]. The polarization effect that results from the EMW passing through these surfaces causes further losses that efficiently absorb and weaken the EMW's energy. Interfacial polarization, the primary loss process in PGS aerogels, is a crucial factor for enhancing absorption efficacy. Furthermore, when exposed to electromagnetic fields, residual oxidized carboxylated groups in graphene oxide and carboxylated carbon nanotubes cause dipole polarization, which further increases the material's polarization loss [64]. In summary, the combined impact of these loss processes greatly increased the PGS composite aerogels' ability to absorb waves.

3.3 Mechanical and thermal insulation properties

The performance of wave-absorbing materials is typically evaluated based on four key criteria: thickness (thin), weight (light), absorption bandwidth (wide), and attenuation capability (strong). Beyond these electromagnetic characteristics, practical applications additionally require adequate mechanical properties—particularly for flexible absorbers, whose enhanced conformability and durability significantly facilitate real-world implementation in consumer electronics and other daily applications. As shown in Fig. 9(a), the prepared PGS aerogel has excellent flexibility and is able to recover its original shape after folding, crumpling, and curling with no damage, and at the same time, the PGS composite aerogel can be easily adhered to the blade, and the aerogel is very light in weight, with a density of only 7.9–9.8 mg/cm³, and the oriented pore size structure is also able to withstand 1850 times of its own weight without deformation (Fig. 9(c)). Figure 9(b) shows the compressive stress–strain curve of PGS5-300 at 50% strain over 100 cycles. With increasing compression cycles, the aerogel shows only minimal stress reduction, maintaining ~8 kPa after unloading, demonstrating durable elastic recovery. Notably, non-heat-treated PGS aerogel exhibits inferior mechanical properties, with a compressive strength of only 16 kPa. After heat treatment, PGS-300 achieves 22.6 kPa compressive strength, while PGS15-300 (with SWNTs) reaches 25.3 kPa (Fig. S12 in the ESM). Apart from the above mentioned intriguing characteristics, PGS composite aerogels also demonstrate exceptional heat resistance. As shown in Fig. 9(d), the PGS composite aerogel remained intact after being burned under an alcohol lamp (external flame temperature 500 °C) for 60 s or more. To illustrate the material's flame retardant properties, samples that were 1 cm thick were subjected to the heating platform for varying durations, as illustrated in Fig. 9(e). The aerogel surface temperature grew steadily to 46.7 and 47.9 °C after 5 min, and the heating platform temperature reached 174.8 °C. After 10 min, the PGS5-300 and PGS15-300 surface temperatures reached 55.4 and 59.2 °C, respectively. After 30, 60, 90, and 120 min, the surface temperatures of PGS5-300 and PGS15-300 did not significantly rise, and the heating table's temperature stayed at 175 °C, demonstrating the superior thermal insulation qualities of the PGS aerogel. The thermal conductivity of PGS5-300 is only 0.027 W/(m·K) (Fig. S16 in the ESM). According to these results, PGS composite aerogels show promise for use in lightweight insulation. Furthermore, following six hours of exposure to a heated plate at 200 °C, the PGS composite aerogel's complex dielectric constant and RL were determined. After being exposed to high temperatures, the material

retained its exceptional microwave absorption qualities, as seen in Fig. S13 in the ESM, which makes it appropriate for long-term use as a wave-absorbing material under harsh conditions. Figure 9(f) shows that PGS as aerogel fabric can effectively insulate the heat generated by human body and can be used as an infrared stealth material. These properties enable PGS aerogel materials to be used as a multifunctional aerogel in more demanding environments, which is more favourable to military and production life applications than previous single-function aerogels.

4 Conclusions

In this study, we fabricated lightweight polyimide/RGO/SWNTs composite aerogels by a facile unidirectional freeze-casting method. A wider effective wave-absorbing bandwidth was obtained at a lower thickness by varying the annealing temperature and thus modulating the polarization relaxation time, and the increase of SWNTs strengthened the polarization loss capability. The prepared aerogels have an ultra-low bulk density of 9.13 mg/cm³ and excellent broadband microwave absorption with an EAB (RL ≤ −10 dB) up to 9.14 GHz, and the optimized aerogels have a minimum reflection loss of −64.5 dB, which demonstrates their excellent attenuation of electromagnetic waves. In addition to its excellent microwave absorption properties, the aerogel has excellent mechanical flexibility and robust structural integrity. These characteristics, coupled with their excellent thermal insulation performance, are attributed to the hierarchically ordered microchannel architecture formed during the directional freezing process. This unique anisotropic pore structure not only enhances mechanical resilience but also facilitates efficient electromagnetic dissipation through multiple scattering and interfacial polarization mechanisms. Given their exceptional thermal stability, mechanical resilience, and broadband absorption properties, these materials are well-suited for deployment in high-temperature environments and other demanding operational conditions. This combination of processing simplicity and superior performance significantly enhances the feasibility of implementing such materials in real-world applications, including aerospace, defense, and next-generation electronic systems.

Electronic Supplementary Material: Supplementary material (XRD, SEM, TEM, electrical conductivity and eddy current criteria C_0 data) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908359>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Z. Z. D.: Data collation, project management, validation, manuscript drafting, and experimental design. C. J., Z. X. Z., Y. T. Z., S. L. H., Y. L. C., Q. C. Z., M. W. Z., and D. Z.: Data collation, manuscript review. All authors have reviewed and approved the final manuscript.

Use of AI statement

None.

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