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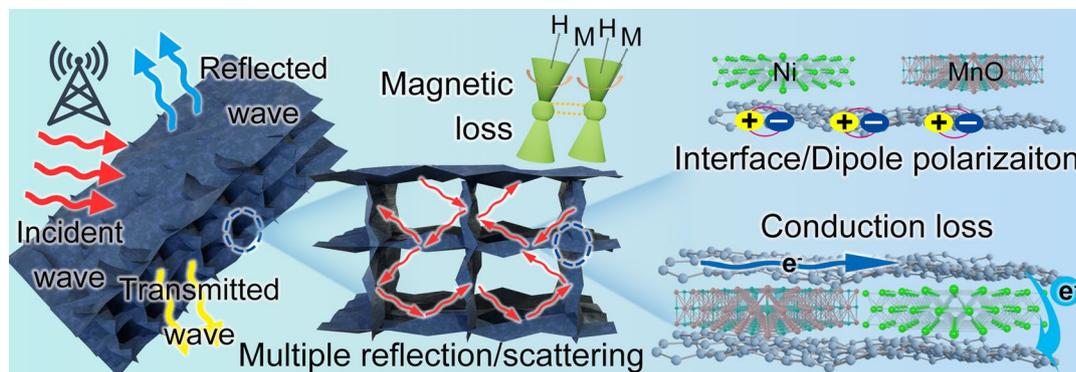
# Nitrogen-Doped Magnetic-Dielectric-Carbon Aerogel for High-Efficiency Electromagnetic Wave Absorption

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## HIGHLIGHTS

- An ingenious design achieved magnetic-dielectric-carbon coupling.
- Nickel and manganese oxide particles were in situ reduced and grew on the carbon aerogels.
- The aerogels demonstrated radar stealth, infrared stealth and thermal management capability.

**ABSTRACT** Carbon-based aerogels derived from biomass chitosan are encountering a flourishing moment in electromagnetic protection on account of lightweight, controllable fabrication and versatility. Nevertheless, developing a facile construction method of component design with carbon-based aerogels for high-efficiency electromagnetic wave absorption (EWA) materials with a broad effective absorption bandwidth (EAB) and strong absorption yet hits some snags. Herein, the nitrogen-doped magnetic-dielectric-carbon aerogel was obtained via ice template method followed by carbonization treatment, homogeneous and abundant nickel (Ni) and manganese oxide (MnO) particles in situ grew on the carbon aerogels. Thanks to the optimization of impedance matching of dielectric/magnetic components to carbon aerogels, the nitrogen-doped magnetic-dielectric-carbon aerogel (Ni/MnO-CA) suggests a praiseworthy EWA performance, with an ultra-wide EAB of 7.36 GHz and a minimum reflection loss ( $RL_{\min}$ ) of  $-64.09$  dB, while achieving a specific reflection loss of  $-253.32$  dB  $\text{mm}^{-1}$ . Furthermore, the aerogel reveals excellent radar stealth, infrared stealth, and thermal management capabilities. Hence, the high-performance, easy fabricated and multifunctional nickel/manganese oxide/carbon aerogels have broad application aspects for electromagnetic protection, electronic devices and aerospace.



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**KEYWORDS** Electromagnetic wave absorption; Wide bandwidth; Dielectric-magnetic synergy; Multifunction

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## 1 Introduction

Gigahertz band wireless communication technology has brought great convenience to human life, but the impact of electromagnetic pollution on human health and environment cannot be ignored [1, 2]. Electronic equipment used in aerospace, radar and computer system is closely related to daily life of human beings, so electromagnetic wave absorption (EWA) materials with high performance, facile fabrication and environmentally friendly need to be developed to solve the matters of electromagnetic pollution [3, 4].

EWA materials, by absorbing the incident electromagnetic waves and converting electromagnetic energy into Joule heat and releasing it into the surrounding environment, so as to achieve the purpose of attenuating electromagnetic waves [5, 6]. As an EWA material with excellent performance, minimum reflection loss ( $RL_{\min}$ ) and effective absorption bandwidth (EAB) are the basic criteria for evaluating its characteristic [7]. With the evolution of miniature and lightweight electronic equipment, especially in the fields of aviation and aerospace, low density and thin thickness have become two critical requirements [8, 9]. Not only that, considering the complex practical application environment of the material, the absorber is usually coated on the surface of the equipment, so the thermal insulation ability of the material needs to be further improved in order to reduce the temperature changes on the performance of the equipment [10, 11]. Obviously, conventional powder materials cannot cope with the above requirements. With the deepening of research, the high specific surface area and rich porosity of foams, sponges and aerogels themselves have given the materials the ability to be lightweight and thermal insulating, which makes them potential candidates among absorbing materials.

In recent years, biomass-derived carbon aerogels have developed vigorously in the field of electromagnetic compatibility and protection due to wide range of sources, diversity of morphology and structure, and facile component combination [12, 13]. Due to their high porosity, large specific surface and abundant functional groups, carbon aerogels provide a variety of interfaces and polarization sites for the effective attenuation of electromagnetic waves. However, the impedance mismatch caused by excessive electrical conductivity and single dielectric loss make carbon aerogels still unsatisfactory [14, 15]. Using the adjustable morphological

structure, the magnetic/dielectric components are combined with carbon aerogels, which may give the aerogels materials suitable impedance matching and synergetic magnetic loss, so as to realize the preparation of broadband EWA materials [16, 17]. Nickel, as a typical soft magnetic material, exhibits a higher saturation magnetization strength compared to most ferrite materials, which is beneficial in enhancing magnetic loss. Ni is conducive to raising the Snoek's limit at high frequency and enhancing the magnetic loss ability of materials due to high permeability and magnetization strength [5, 18]. As a cost-effective dielectric material, manganese oxide (MnO) has excellent stability and unique physical and chemical properties. Its low dielectric constant can be used to adjust the impedance matching of carbon aerogels, inducing more incident electromagnetic waves to enter the aerogel interior. MnO with low conductivity can adjust the overall conductivity of composite materials while suppressing eddy current loss of magnetic materials [19]. Chitin is a biological resource whose content is second only to cellulose in nature, and it has rich nitrogen content [20, 21]. The introduction of nitrogen atoms can cause various defects in the carbon skeleton, such as dislocation and lamellar bending. The difference in electronegativity between nitrogen and carbon atoms changes the distribution of electron clouds and leads to the formation of electric dipoles in the electromagnetic field, inducing dipole polarization and contributing to the dissipation of electromagnetic waves [14, 22]. Chitosan, deacetylation of chitin, can be dissolved into acidic solution to form stable suspension and then prepared into aerogel by freeze drying. Therefore, chitosan has received extensive attention and as a biomass resource for the preparation of three-dimensional porous carbon aerogels [23, 24]. Chitosan-derived carbon aerogels have received widespread attention in the electromagnetic field due to their wide availability, environmentally-friendly nature, and controllable preparation. However, the electromagnetic wave absorption properties of aerogels previously studied are not outstanding, including narrow band width and weak absorption capacity [14, 25]. Limited by the acidic treatment conditions of chitosan, the typical combination method of impregnation is used, but the method can lead to weak binding and uneven distribution between components and carbon aerogels. At present, the implementation of the above attributes still faces huge challenges.

In this work, we have fabricated a nitrogen-doped multi-component carbon aerogel (Ni/MnO-CA) by facile

freeze-drying and carbonization, and realized in situ composite of magnetic/dielectric components with carbon aerogel. The Ni and MnO modulated the impedance matching of the carbon aerogel, and the synergistic effect of dielectric and magnetic loss allowed Ni/MnO-CA to obtain an impressive EWA performance with  $RL_{\min}$  of  $-64.09$  dB and EAB of  $7.36$  GHz. The three-dimensional cell structure and the synergy of multiple components made aerogels versatile, such as considerable radar stealth, infrared stealth, and thermal management capabilities. Therefore, compared with the same type of absorbers, the magnetic-dielectric-carbon aerogel of this work has significant advantages in the field of electromagnetic protection and electronic device applications.

## 2 Experimental Section

### 2.1 Materials

Chitosan ( $(C_6H_{11}NO_4)_n$ ,  $<200$  mPa s) and glacial acetic acid were bought from Shanghai Aladdin Biochemical Technology Co., Ltd. Nickel (II) chloride hexahydrate ( $NiCl_2 \cdot 6H_2O$ ) and manganese (II) chloride tetrahydrate ( $MnCl_2 \cdot 4H_2O$ ) were purchased from Sinopharm Chemical Reagent Co., Ltd. All the chemicals were analytically pure without any other treatment.

### 2.2 Fabrication of Magnetic-Dielectric-Carbon Aerogels

The fabrication process of aerogels included directed freezing, freeze drying and carbonization. In the first step,  $0.75$  g of chitosan powder was dissolved in  $30$  mL of deionized water and magnetically stirred for  $20$  min. Subsequently,  $1.05$  mmol  $NiCl_2 \cdot 6H_2O$  and  $1.05$  mmol  $MnCl_2 \cdot 4H_2O$  were added to the above solution and magnetically stirred for  $10$  min. After that,  $0.51$  mL of glacial acetic acid was quickly dripped into the solution to form a suspension. Then, the suspension was slowly poured into a homemade Teflon mold with copper base ( $4 \times 4 \times 4$  cm<sup>3</sup>) and placed together in liquid nitrogen ( $-196$  °C) for directed freezing. After  $24$  h of freeze-drying, the xerogel was carbonized under nitrogen atmosphere ( $700$  °C) to obtain magnetic-dielectric-carbon

aerogel (Ni/MnO-CA). For comparison, we also prepared the Ni-CA and MnO-CA via the similar procedure, without introducing Mn or Ni sources in the precursor, where the total content of the precursor metal salts was maintained as  $2.1$  mmol. In addition, in order to explore the influence of magnetic/dielectric components on electromagnetic parameters, the total content of precursor metal salts (the content of  $NiCl_2 \cdot 6H_2O$  and  $MnCl_2 \cdot 4H_2O$  was 1:1) was increased to  $3.15$  and  $4.2$  mmol, and the final aerogels were named after Ni/MnO-CA-1.5 and Ni/MnO-CA-2.0, respectively.

### 2.3 Characterizations

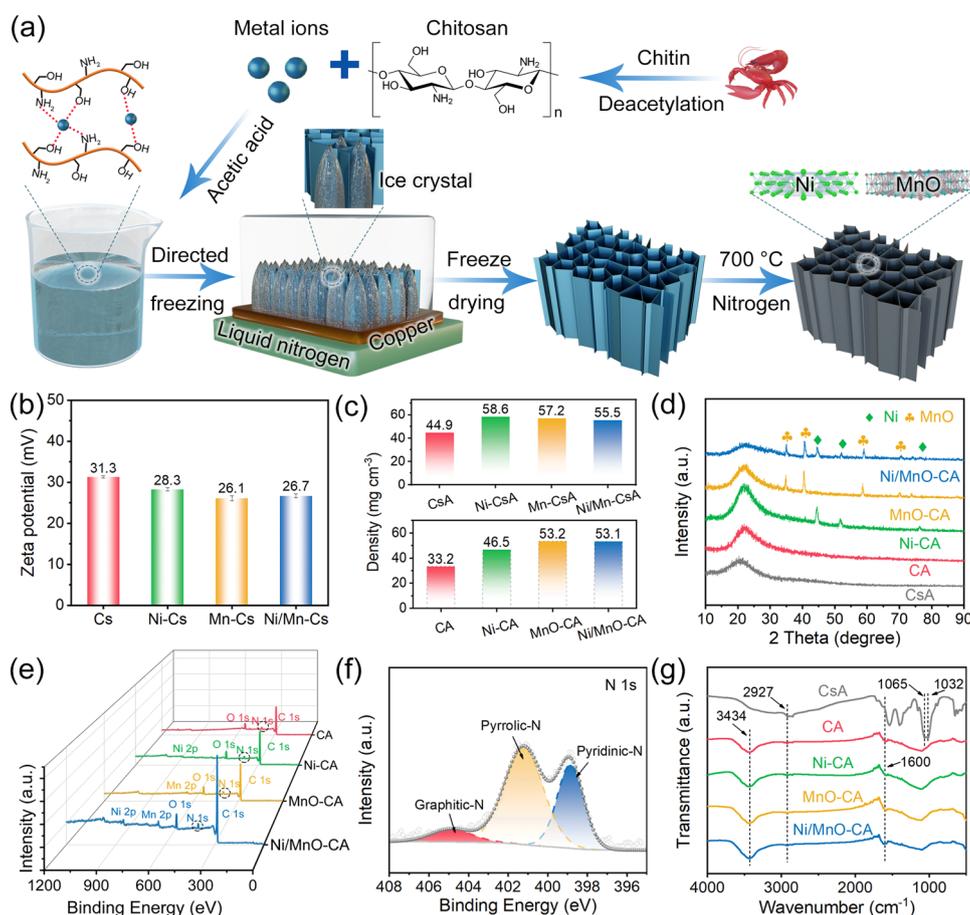
The morphology and microstructure were recorded by field emission scanning electron microscopy (FE-SEM; Hitachi, Model SU-70) and high resolution electron microscope (HRTEM, JEM 2100F). The X-ray photoelectron spectroscopy (XPS) were tested by photoelectron spectrometer (Thermo ESCALAB 250Xi). The X-ray diffraction (XRD) spectra were recorded by powder X-ray diffractometer (DMAX-2500PC, Rigaku). Fourier transform infrared spectra (FT-IR) were recorded on Nicolet iS 5 FT-IR spectroscopic analyzer. Thermogravimetric analysis (TGA, HCT-1) was used to assess the content of carbon. Nitrogen adsorption-desorption isotherms were recorded by specific surface area and porosity analyzer (ASAP2460). The magnetic loops at room temperature were recorded by a vibration sample magnetometer (VSM, LakeShore 7404). The zeta potential was determined by using a potentiometric analyzer (Malvern, UK). The Raman spectra were through by a Raman spectrometer (LabRam HR Evolution). Thermal conductivity at room temperature was obtained by LFA 467. Infrared emissivity values in  $3-5$  and  $8-14$   $\mu$ m wavelengths were recorded by China-Wanyi-IR2 type dual-band emissivity measuring instrument. Thermal images of the aerogels were obtained from a FLIR E86 infrared thermal camera and FLIR Tools + software was used to deal with the data. The electromagnetic parameters were measured by a vector network analyzer (VNA; Agilent PNA N5244A). Here, a coaxial ring method was applied, and an annulus sample with  $10$  wt% aerogel fragment and  $90$  wt% paraffin mixing was made by a tailored metallic mold.

### 3 Results and Discussion

#### 3.1 Composition and Structure

The fabrication process of nitrogen-doped magnetic-dielectric-carbon aerogel (Ni/MnO-CA) is depicted in the figure (Fig. 1a), including freeze-drying and subsequent carbonization process. Initially,  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$  and chitosan powder were dissolved in an aqueous solution of glacial acetic acid at room temperature to form a homogeneous and stable suspension (Ni/Mn-Cs, Fig. S1, Tyndall effect), which demonstrated the good stability and dispersion. Due to the protonation of a large number of free amino groups in the chitosan molecular chain, long-chain polymer chitosan could be dissolved in glacial acetic acid solution to form a homogeneous suspension [26]. The introduction of Ni and

Mn ions reduced the zeta potential of the suspension slightly, but the suspension still maintained good stability (Figs. 1b and S2). In particular, the transition metal ions would react with the amino and hydroxyl groups on the chitosan molecular chain, which could promote the cross-linking of molecular and maintain the stability of the suspension [27]. Subsequently, the suspension was freeze-dried and the xerogel was obtained to test the FT-IR (Fig. S3). For pure chitosan xerogels (CsA), the peaks at 1533 and 1396  $\text{cm}^{-1}$  corresponded to the stretching vibration peaks of amide  $-\text{NH}_2$  and C-N bonds [15, 20]. Analysis of the curves of xerogels with Ni and Mn ions revealed that the above two peaks underwent blue shift, which proved that transition metal ions induced complexation of chitosan chains. During directed freezing, a large number of ice crystals grew vertically because of the temperature gradient, and the suspension located at the



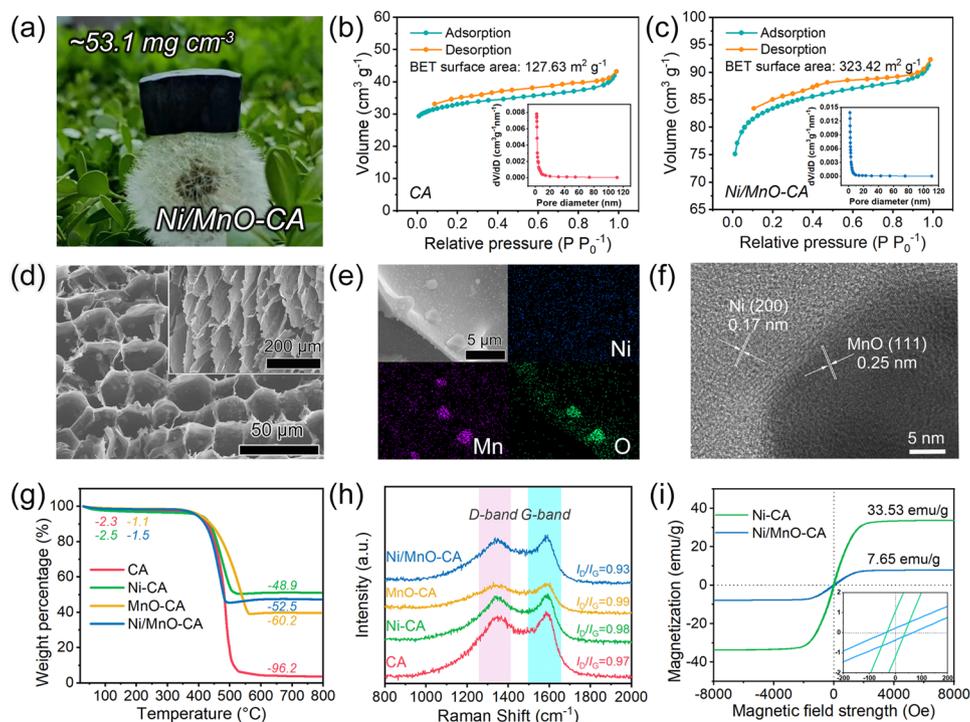
**Fig. 1** a Schematic illustration of fabricating Ni/MnO-CA. b Zeta potential of chitosan suspension before and after Ni/Mn ion addition. c Density of aerogels before and after carbonization. d XRD patterns of CsA, CA, Ni-CA, MnO-CA, and Ni/MnO-CA. XPS spectra of e CA, Ni-CA, MnO-CA, and Ni/MnO-CA and f N 1s region of Ni/MnO-CA. g FT-IR

front of the ice crystals solidified to form a continuous cell wall [28, 29]. After freeze-drying, ice crystals sublimated and escaped to form xerogel (Ni/Mn-CsA, Fig. S4). Finally, under the protective atmosphere of nitrogen, chitosan was pyrolyzed into amorphous carbon and reducing gas [30], and nickel and manganese ions were reduced to metal nickel and manganese oxide uniformly distributed on the carbon skeleton via carbothermal reaction. Herein, anisotropic N-doped magnetic-dielectric-carbon aerogel (Ni/MnO-CA) was successfully prepared. The density of aerogels before and after nitrogen treatment is shown in Fig. 1c, and XRD further analyzed the component transformation in Fig. 1d. After CsA was carbonized, the wide peak of the carbon aerogel (CA) at  $22.6^\circ$  corresponded to the (002) crystal plane of graphite [31, 32]. In addition, the characteristic peaks of  $44.5^\circ$ ,  $51.8^\circ$ , and  $76.4^\circ$  corresponded to (111), (200), and (220) planes of Ni (No. 87–0712), the peaks at  $35.0^\circ$ ,  $40.7^\circ$ ,  $58.9^\circ$ , and  $70.4^\circ$  were assigned to (111), (200), (200), (220), and (311) planes of MnO (No. 75–0626), respectively, which confirmed the successful in situ reduction in metal nickel and manganese oxide [33, 34].

The surface chemistry of magnetic-dielectric-carbon aerogels was meticulously analyzed by XPS. Since chitosan was rich in amino functional groups, nitrogen doping was obtained after carbonization, and the spectrum of XPS further confirmed the existence of nitrogen (Fig. 1e). In the N 1s orbit (Fig. 1f), doped nitrogen atoms existed in the form of pyridinic N (398.9 eV), pyrrolic N (401.2 eV) and graphitic N (404.8 eV) [22], which favored the conductivity loss of carbon materials because nitrogen doping could effectively reduce the intrinsic resistance and promote electron transport. In particular, pyridinic N provided unpaired electrons to form  $\pi$ -conjugated rings with  $sp^2$  hybrid carbon atoms, which was conducive to enhancing electron transport and graphitic N replaced carbon atoms in the graphite layer and introduced delocalized  $\pi$  electrons, which reduced the intrinsic resistance of carbon materials. Moreover, based on the C 1s orbit (Fig. S5a), the deconvolution peaks of 284.6, 285.6, and 288.8 eV were consistent with C–C, C–N, and C=O bonds, respectively, where oxygen-containing groups were derived from remnants of chitosan pyrolysis [35, 36]. After pyrolysis, most hydrophilic groups in chitosan were reduced, such as –COC, –COH, and –CH<sub>2</sub>OH in the range of 1200–1000  $\text{cm}^{-1}$ . And due to the acid treatment moved from 3434 to 2927  $\text{cm}^{-1}$ , the –OH vibration peak was also narrowed and the group was reduced. However, the peak

of the C=O bond of the amide at 1600  $\text{cm}^{-1}$  was more pronounced (Fig. 1g). The oxygen groups would induce dipole polarizations to promote electromagnetic wave attenuation. In addition, the XPS spectra of O 1s, Ni 2p, and Mn 2p are depicted in Fig. S5b–d, which once again revealed the presence of metal nickel and manganese oxide.

The N-doped magnetic-dielectric-carbon aerogel prepared in situ had a lower density ( $\sim 53.1 \text{ mg cm}^{-3}$ ), and lightweight properties allow it to stand on a dandelion (Fig. 2a). Compared with carbon aerogel, in situ grown particles increased the specific surface area of the aerogel (Figs. 2b, c and S6a, b), and the nitrogen absorption and desorption curves confirmed that Ni/MnO-CA had a high BET specific surface area ( $323.42 \text{ m}^2 \text{ g}^{-1}$ ) and mesoporous presence [37]. In addition, the macroscopic pore size and microstructure of Ni/MnO-CA were confirmed by scanning electron microscopy (SEM). All aerogels exhibited three-dimensional porous structures (Figs. 2d and S7), which provided favorable channels for multiple reflection and scattering of electromagnetic waves [38, 39]. Further, the lamellar structures were arranged parallel to the direction of ice crystal growth, with pores of about 25  $\mu\text{m}$  perpendicular to the direction of ice crystal growth, and Ni and MnO particles were evenly distributed on them. Based on transmission electron microscopy (TEM) analysis, the interplanar spacing of 0.17 and 0.25 nm belonged to Ni (200) and MnO (111) lattice plane, respectively. The close bonding of Ni and MnO particles with carbon could improve the interface quality and facilitate polarization. Analyzing the TGA curves (Fig. 2g), the decline in the first stage (less than 150  $^\circ\text{C}$ ) belonged to the evaporation of the absorbed water, and the second stage came from the decomposition of the carbon support [15, 40]. Ignoring the adsorbed water, we determined the amount of carbon in the aerogel composites with the help of formulas (Eqs. S1–S3). The carbon content in Ni-CA, MnO-CA, and Ni/MnO-CA was 59.85, 62.98, and 57.51 wt%, respectively, indicating that the carbon content in aerogel composites was similar. For carbon-based absorbing materials, the degree of graphitization of carbon affected the conductivity of the material, which in turn affected the EWA performance of the material [41, 42]. The  $I_D/I_G$  ratio in the Raman curve was used to assess the degree of graphitization of carbon [43], in which the D band (an active  $A_{1g}$  mode of crystallite boundaries) at around 1340  $\text{cm}^{-1}$  was thought to be the existence of disorders and defects, and the G band (from an active  $E_{2g}$  mode of infinite



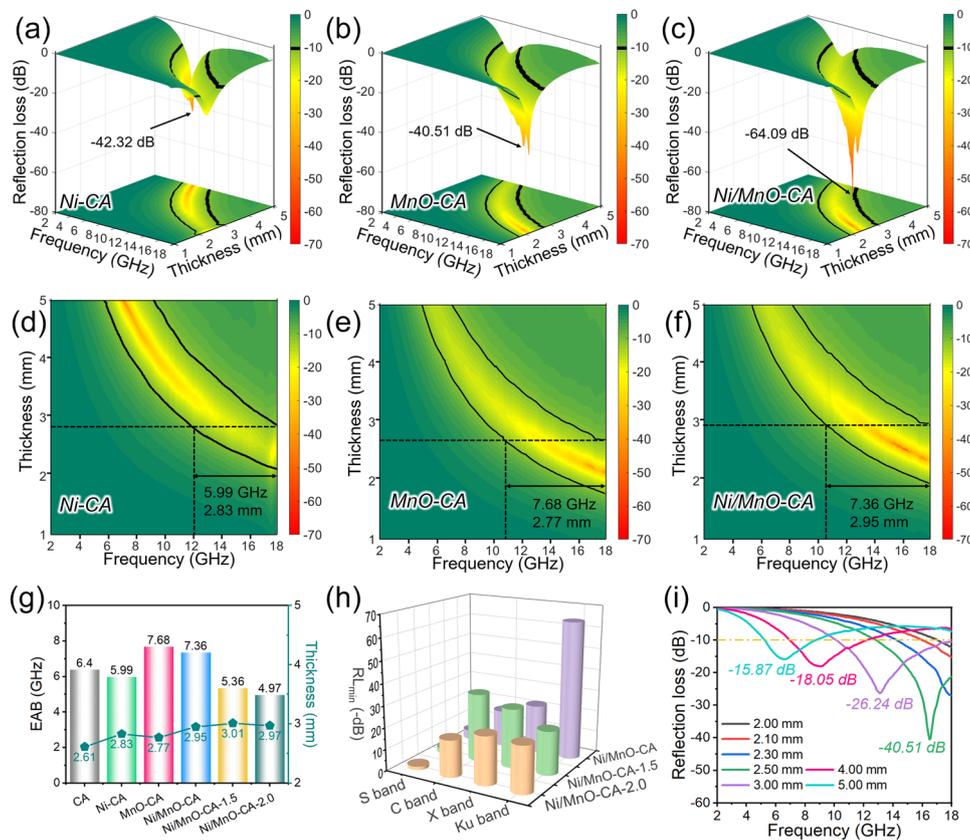
**Fig. 2** a Digital photograph of Ni/MnO-CA standing on a dandelion. BET surface area and pore size distribution of b CA and c Ni/MnO-CA. d Field emission scanning electron microscope (FE-SEM) of Ni/MnO-CA, the inset was parallel to the direction of ice crystal. Ni/MnO-CA: e EDS mappings and f High-resolution TEM image. g TGA curves, h Raman spectra and i Magnetic hysteresis loops

crystals) at  $1590\text{ cm}^{-1}$  corresponded to the perfect graphite lattice regions [44, 45]. The four carbon aerogel composites had a comparable degree of graphitization (Fig. 2h), revealing that the introduction of Ni or MnO components did not destroy the degree of graphitization of the carbon matrix [46, 47]. The introduction of magnetic Ni notably affected the magnetism of aerogel composites, as shown in Fig. 2i. Ni-CA possessed a high saturation magnetization of  $33.53\text{ emu g}^{-1}$  and the value of Ni/MnO-CA was  $7.65\text{ emu g}^{-1}$ . In addition, the coercivity of the two was 33.2 and 42.1 Oe, respectively, which were higher than that of bulk nickel ( $\sim 0.7\text{ Oe}$ ) [10]. The large coercivity would lead to increased magnetic anisotropy, which caused the natural resonance to move towards higher frequency and improved the magnetic loss performance in GHz frequency range.

### 3.2 Electromagnetic Wave Absorption Properties

According to the transmission line theory, the electromagnetic wave absorption performance of magnetic-dielectric-carbon aerogels was expressed by the reflection

loss [22, 48] (Eqs. S4–S5). The absorption capacity of CA for electromagnetic wave was not commendable, featuring an  $RL_{\min}$  of  $-18.31\text{ dB}$  and an EAB of 6.4 GHz (Fig. S8). Typically, a reflection loss value of less than  $-10\text{ dB}$  meant that 90% of the incident electromagnetic waves were attenuated and dissipated in the form of heat [11, 25]. However, the loss capability of CA was insufficient for the absorbers with excellent performance. Obviously, the in situ introduction of magnetic Ni and transition metal oxide MnO improved the loss capability of aerogel composites for electromagnetic waves (Fig. 3a–f). In detail, Ni-CA had an  $RL_{\min}$  value of  $-42.32\text{ dB}$  while maintaining an EAB of 5.99 GHz, and MnO-CA had a slightly reduced  $RL_{\min}$  value of  $-40.51\text{ dB}$ , but the EAB reached 7.68 GHz. Interestingly, the coexistence of Ni and MnO made Ni/MnO-CA obtain the outstanding EWA performance, in which  $RL_{\min}$  was  $-64.09\text{ dB}$  and a wide EAB of 7.36 GHz. Further, we increased the ratio of Ni and MnO in aerogel composites, and found that their ability to lose electromagnetic waves decreased, and the frequency range with reflection losses below  $-10\text{ dB}$  was narrowed (Fig. S9). More intuitively, the EAB and

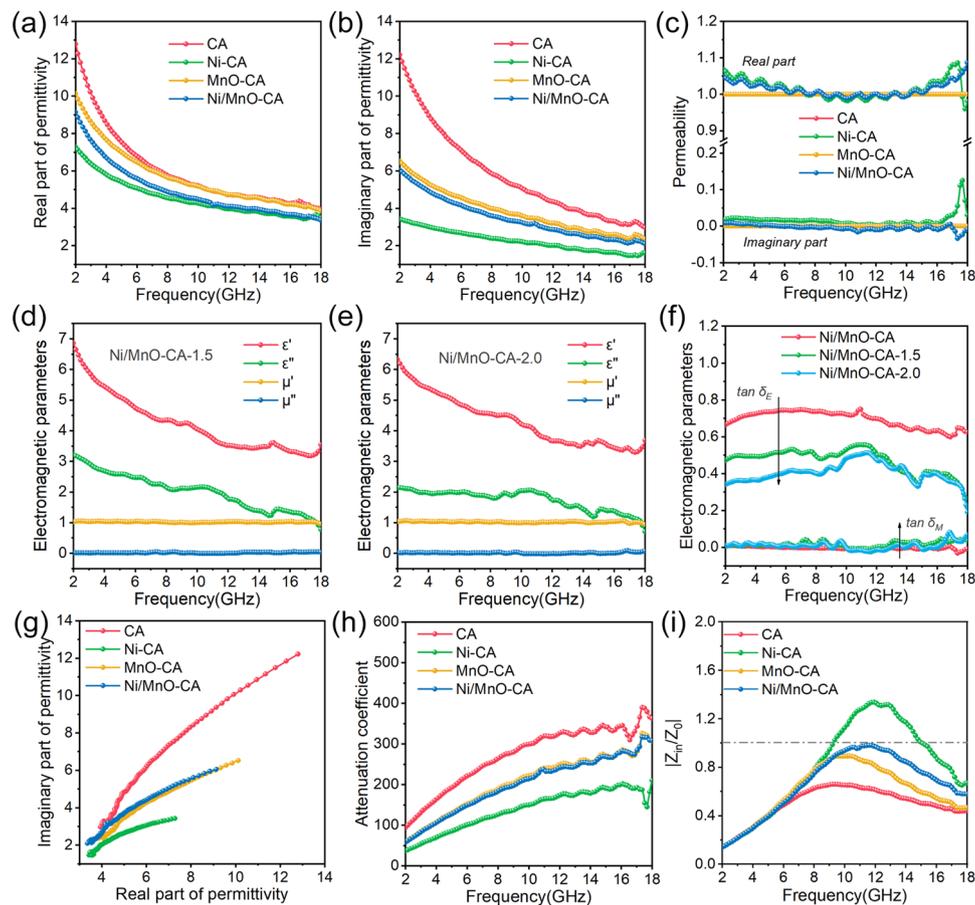


**Fig. 3** 3D RL plots of **a** Ni-CA, **b** MnO-CA and **c** Ni/MnO-CA. 2D contours of RL values versus frequency and thickness of **d** Ni-CA, **e** MnO-CA and **f** Ni/MnO-CA. **g** EAB and corresponding thickness for the aerogels. **h**  $RL_{min}$  values of the aerogels at different frequency bands. **i** 1D curves of RL values versus frequency at different thicknesses for Ni/MnO-CA

corresponding matching thickness of the above aerogel composites were compared (Fig. 3g). Despite the slight advantages and disadvantages of the loss capability of electromagnetic waves, aerogel materials exhibited a wide EAB and a thin matching thickness, which was good for absorbers. Subsequently, for aerogel composites with different Ni/MnO contents, we compared the minimum reflection loss in different frequency bands, and it could be found that with the increase in component content, the strongest absorption of electromagnetic waves by aerogels generally moved towards the low-frequency region (Fig. 3h). In addition, Ni/MnO-CA absorbed more than 90% of electromagnetic waves at different thicknesses, demonstrating its excellent absorption performance (Fig. 3i).

### 3.3 Electromagnetic Parameters

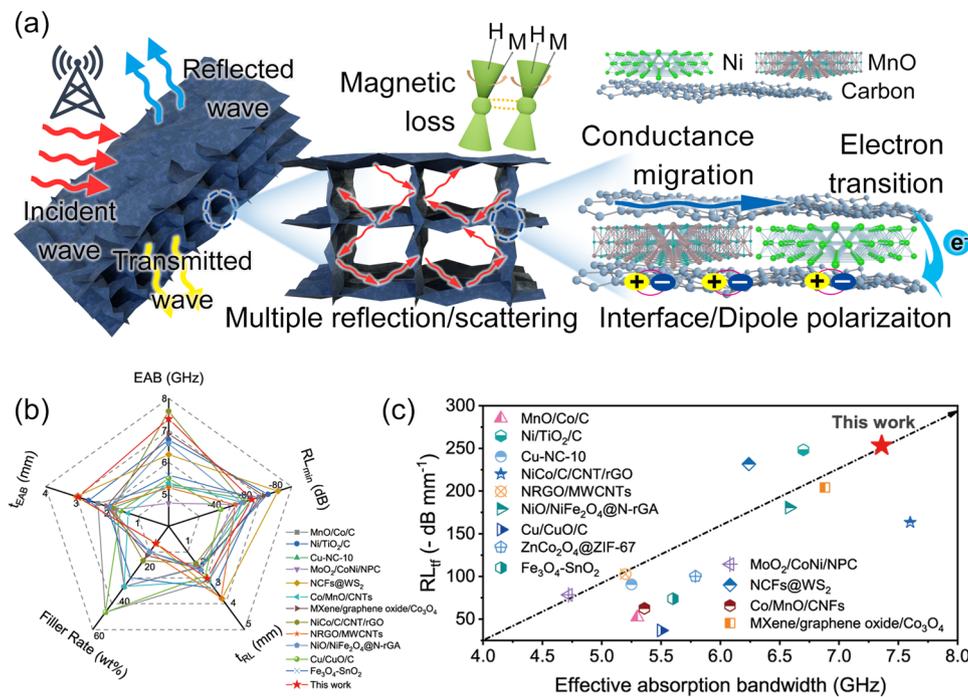
Aiming at deeply exploring the mechanism of the above aerogels, it was crucial to measure the electromagnetic parameters, which include complex permittivity ( $\epsilon_r = \epsilon' - j\epsilon''$ ) and complex permeability ( $\mu_r = \mu' - j\mu''$ ). In general, the real parts of permittivity ( $\epsilon'$ ) and permeability ( $\mu'$ ) represented the ability to store electrical and magnetic energy, and correspondingly, the imaginary parts ( $\epsilon''$ ,  $\mu''$ ) indicated the ability to dissipate energy [49]. The permittivity of aerogels showed a decreasing trend with increasing frequency (Fig. 4a), which was consistent with the frequency dispersion behaviors of carbon materials [50, 51]. The  $\epsilon'$  and  $\epsilon''$  value of CA decreased from 12.8 to 4.0 and from 12.2 to 3.0 (Fig. 4b), respectively, indicating the dominant contribution of conductive losses. According to the free electron theory ( $\epsilon'' = \sigma / (2\pi\epsilon_0 f)$ ), the imaginary part of permittivity was related to the conductivity of materials [52]. For CA



**Fig. 4** **a** Real part of permittivity. **b** Imaginary part of permittivity and **c** permeability. Electromagnetic parameters of **d** Ni/MnO-CA-1.5 and **e** Ni/MnO-CA-2.0. **f** The loss tangent. **g** The Cole–Cole plot. **h** Attenuation coefficient and **i** impedance matching values

obtained by carbonized chitosan under  $N_2$  atmosphere, its imaginary part of permittivity was too high, which was due to excessive conductivity. Too high a permittivity was not conducive to impedance matching, resulting in strong reflection of electromagnetic waves on the surface of the absorbers, so the EWA performance of CA was insufficient. In more detail, the  $\epsilon''$  of Ni-CA and MnO-CA was from 3.4 to 1.6 and 6.5 to 2.4. The in-situ introduction of the components Ni and MnO inhibited the electrical conductivity of the composite aerogel thereby reducing the imaginary part of permittivity. Magnetic nickel particles gave aerogels the ability to store and dissipate magnetic energy, which was manifested by the change of permeability with frequency (Fig. 4c). The curve in the imaginary part had obvious peaks at high frequency, indicating the presence of magnetic resonance loss, which could be understood as exchange resonances. In addition, we analyzed the contribution of eddy

current loss to magnetic loss by plotting the  $C_0$  curve (Fig. S10a). Typically, if eddy current loss was present in magnetic loss, the  $C_0$  was constant [53]. In the test frequency range, the  $C_0$  values decreased with increasing frequency, showing that the eddy current loss in magnetic materials was suppressed and magnetic resonance dominated magnetic loss [54]. By plotting the tangent curves of dielectric loss and magnetic loss (Fig. S10b–c), the dielectric and magnetic loss capabilities were evaluated. By comparison, the dielectric loss contributed greatly to the attenuation of electromagnetic waves. When the content of Ni/MnO increased, the dielectric loss capacity of aerogels decreased, and the magnetic loss increased (Fig. 4d–f), which ultimately led to a decrease in EWA performance. Therefore, dielectric loss in the loss mechanism had a greater impact on performance [55, 56].

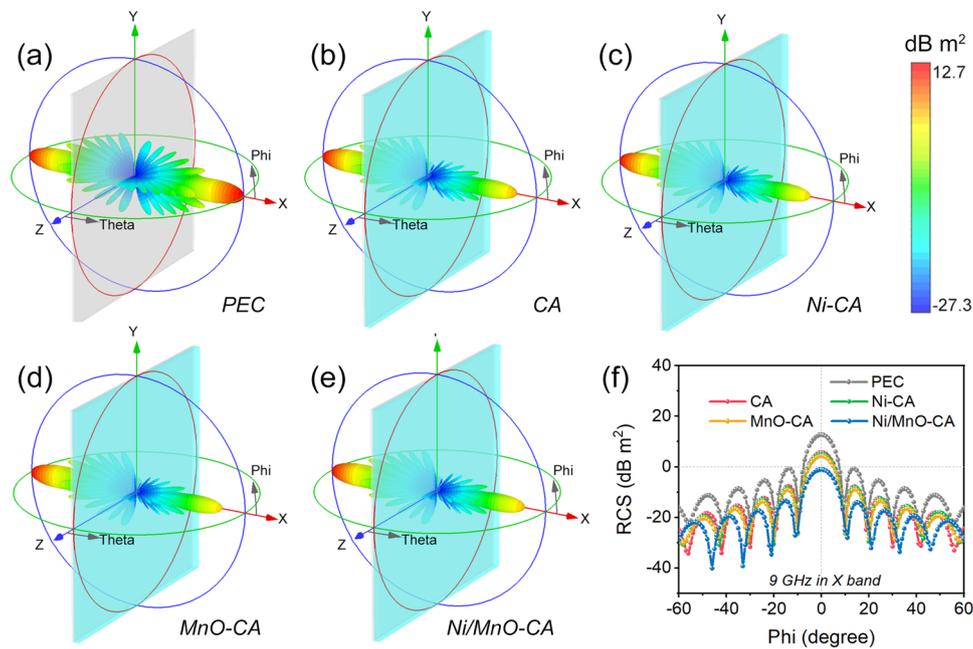


**Fig. 5** **a** Schematic diagram of EWA mechanisms for Ni/MnO-CA. **b** Radar chart of the properties. **c** Comparison of  $RL_{eff}$  values and EAB with previous works

When considering the attenuation characteristics of materials, dielectric loss included conductivity loss and dielectric relaxation loss [57, 58]. In the Cole–Cole plot (Fig. 4g, Eqs. S6–S8), the long and tall tail indicated the conductivity loss of the four aerogels, and the CA was significantly higher than the other three. A few semicircles at the curve indicated that the material had dipole polarization and interfacial polarization [4]. Specifically, the C=O functional groups and C–N bonds derived from chitosan contributed to the dipole polarization. The interface between particles and carbon constituted the interfacial polarization in the aerogels. In order to further evaluate the comprehensive attenuation ability of aerogels to electromagnetic waves, we discussed the attenuation coefficient ( $\alpha$ , Eq. S9). The introduction of Ni or MnO reduced the attenuation ability of carbon aerogels to electromagnetic waves, in which Ni-CA had the weakest ability, and both MnO-CA and Ni/MnO-CA had the similar capabilities (Fig. 4h). However, Ni/MnO-CA possessed the best EWA performance, so we further analyzed the impedance matching coefficient ( $Z$ , Fig. 4i). When  $Z$  value was closer to 1, the excellent impedance matching could be achieved, so

the reflection at the interface of absorber was minimized. Although the loss capability of aerogels to electromagnetic waves was a key factor affecting performance, it could not be ignored that electromagnetic waves could effectively enter the absorber and then be dissipated when the absorber was adapted to the impedance of space. CA has the strongest attenuation capability, but its performance was not the best due to its impedance mismatch, resulting in too many electromagnetic waves being reflected on the surface of absorbers. The in-situ recombination of Ni or MnO with carbon aerogels improved the impedance matching of materials, as shown in the figure (Fig. S11). It could be noted that the EWA performance of Ni/MnO-CA was more prominent, because the introduction of magnetic nickel further improved the impedance matching between the aerogels and space, and the magnetic loss brought by it achieved the best EWA performance.

Based on the above discussion, the possible interaction mechanism of magnetic-dielectric-carbon aerogels with electromagnetic waves was shown (Fig. 5a). Firstly, the porous structure of aerogels provided a transmission channel, which was conducive to multiple reflections and



**Fig. 6** 3D far-field response of RCS simulations at 9 GHz. **a** PEC, **b** CA, **c** Ni-CA, **d** MnO-CA and **e** Ni/MnO-CA. **f** 1D plot of RCS simulated values

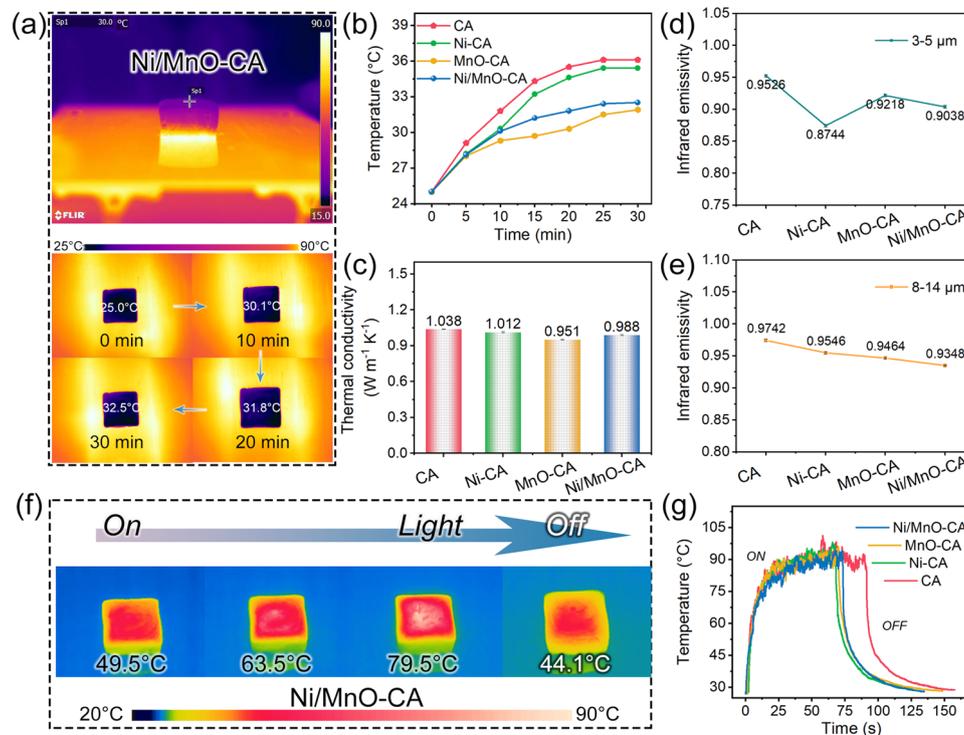
scattering of electromagnetic waves in the internal space structure of the aerogels to achieve the attenuation of electromagnetic waves [59]. Secondly, the combination of Ni/MnO with carbon aerogels regulated the impedance matching of aerogels and free space, so that the electromagnetic waves could enter into aerogels more effectively and interact with them. Based on electron migration in graphite crystallite and electron transitions between crystallite, conductive loss in aerogel contributed to the attenuation of electromagnetic waves. Finally, Ni/MnO-CA achieved excellent EWA performance due to the synergistic effect of dielectric and magnetic loss, conduction loss by electron migration of carriers in the conductive carbon layer, dipole polarization and interface polarization of different components, and resonance in magnetic loss [60].

In comparison with the EWA materials in the previous literature (Table S1), Ni/MnO-CA demonstrated the wide EAB of 7.36 GHz, strong absorption of  $-64.09$  dB and low filler rate (10 wt%), as shown in radar chart (Fig. 5b). Moreover,  $RL_{tf}$  was used to explain the  $RL_{min}$  vs thickness and filler rate, further highlight the lightness, thinness, width, and strength of excellent EWA materials [16, 18]. While maintaining a wide EAB, the  $RL_{tf}$  of this work

reached  $-253.32$  dB  $mm^{-1}$ , which had a significant advantage over the same type of absorbers (Fig. 5c).

### 3.4 Electromagnetic Wave Absorption by CST Simulation

To further verify the stealth performance of aerogels in practical applications, we simulated the far-field response in radar cross section (RCS) by *CST microwave studio*. At the 9 GHz monitoring frequency in X band, the 3D far-field simulation plots of perfect electric conductor and four aerogels are shown in Fig. 6a–e, and we also gave the results at the monitoring frequency in other bands (Figs. S13–S14). RCS values of materials stood for the physical quantity of the echo intensity under radar detection, therefore the smaller the values, the better the stealth performance [61]. The maximum RCS values for the PEC model, CA, Ni-CA, MnO-CA, and Ni/MnO-CA composites were 12.7, 5.62, 5.11, 4.31, and  $-1.06$  dB  $m^2$ , respectively. These simulated results were consistent with the EWA performance of each aerogel material. Compared with PEC and three other aerogels, Ni/MnO-CA demonstrated its fantastic radar stealth performance in practical applications (Fig. 6f).



**Fig. 7** **a** Infrared thermal images of Ni/MnO-CA on a constant temperature heating plate of 90 °C. **b** Temperature–time curves of aerogels surface. **c** Thermal conductivity of CA, Ni-CA, MnO-CA and Ni/MnO-CA. Infrared emissivity of **d** 3–5 μm and **e** 8–14 μm. **f** Infrared images of solar-thermal of Ni/MnO-CA. **g** Photothermal curves of aerogels, and the simulated sunlight used was 180 mW cm<sup>-2</sup>

### 3.5 Infrared Stealth Performance and Thermal Management Capability

In order to cope with complex and changeable application environments, the versatility of EWA materials was worth exploring. Therein, infrared stealth performance was vital in practical applications, and excellent stealth performance could reduce the risk of being detected by infrared detectors [62]. Hence, we tested the thermal insulation performance of aerogels and infrared emissivity of different wavelength bands. First, the temperature change of the aerogel surface was recorded with an infrared camera by placing the aerogel (2.45 × 2.45 × 0.95 cm<sup>3</sup>) on a 90 °C heating plate (Figs. 7a and S15–S17). After about 30 min, all aerogels showed excellent thermal insulation properties, where the surface temperature of CA was only 36.1 °C, while the thermal insulation ability of the other three aerogels was further improved (Fig. 7b). The thermal conductivity of the above aerogels was shown in the figure (Fig. 7c), the outstanding thermal insulation ability was due to the air inside the aerogel

and the pores of the skeleton hindering heat conduction. Second, we tested the infrared emissivity of aerogels in the two atmospheric window regions of 3–5 and 8–14 μm in infrared detection, as shown in the figure (Fig. 7d–e). The overall infrared emissivity of aerogels at 3–5 μm was lower than that of 8–14 μm, and the lower emissivity confirmed the brilliant infrared stealth performance of aerogels. In addition, Ni/MnO-CA showed good photothermal conversion ability (Fig. 7f), which could reach 85 °C with 35 s under simulated sunlight of 180 mW cm<sup>-2</sup> (Fig. 7g). The photothermal conversion ability made aerogel materials excellent thermal management ability, which depicted that it had great potential in the field of electronic devices.

## 4 Conclusion

In summary, the nitrogen-doped magnetic-dielectric-carbon aerogel (Ni/MnO-CA) was facilely fabricated by ice template method and carbonization treatment.

The reasonable component design and pore structure endowed the aerogel efficient EWA performance, and the radar, infrared stealth and thermal management capabilities made it suitable for more complex and changeable environments. The in situ introduction of nickel and manganese oxide components adjusted the impedance matching of the carbon aerogel, so Ni/MnO-CA obtained an  $RL_{\min}$  of  $-64.09$  dB and a specific absorption intensity of  $-253.32$  dB  $\text{mm}^{-1}$ , while maintaining an ultra-wide EAB of 7.36 GHz at a matching thickness of 2.95 mm. Further, RCS far-field simulation revealed its considerable radar stealth performance, while its outstanding thermal insulation ability and low infrared emissivity confirmed its excellent infrared stealth performance. In addition, the rapid photothermal conversion further demonstrated good thermal management capabilities. High-performance, easily fabricated and multifunctional Ni/MnO-CA possessed broad application prospects for electromagnetic compatibility protection, electronics and aerospace.

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#### Declarations

**Conflict of interest** The authors declare no interest conflict. They have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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