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Spatial Segmentation and Controllable Connection Enabled Hollow Microsphere-Carbon Distributed RLC Networks for Multifunctional EM/Thermal Regulation

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Abstract

Lightweight hierarchical porous composites for electromagnetic (EM) wave attenuation and thermal management face a common trade-off: conductive networks needed for dissipation often ruin impedance matching. Here we resolve this by an ice-templated heterogeneous nucleation strategy. Magnetic dual-shell hollow microspheres (DSHM) serve as ice-nucleating agents, directing nitrogen-doped carbon nanosheets (N-CNSs) to bridge the isolated microspheres without forming continuous conductive shells. The resulting architecture is a distributed resistive-inductive-capacitive (RLC) network. The optimized aerogel achieves a reflection loss of -54.86 dB and an effective absorption bandwidth of 6.56 GHz at only 8 wt% filler, together with a thermal conductivity of $0.0605 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ and $>95\%$ infrared absorptivity. Nitrogen doping (pyridinic/pyrrolic N) and nickel-induced graphitization of amorphous carbon enhance polarization and conduction losses, respectively. Resin infiltration further raises the compressive strength to 45.70 MPa without harming EM performance. This ice-templated heterogeneous nucleation paradigm provides a generalizable route to program filler connectivity in carbon nanosheet networks for lightweight EM/thermal management.

1.Introduction

The rapid advancement of high-frequency electronics, aerospace systems, and telecommunications has escalated the concurrent demand for efficient electromagnetic (EM) wave attenuation and thermal management. Developing lightweight, multifunctional non-metallic or composite aerogels has thus become a pivotal research frontier[1-3]. However, hierarchical porous composites intended for these dual purposes face a fundamental and ubiquitous trade-off: establishing robust, continuous conductive networks—which are highly favored for EM energy dissipation—inevitably ruins impedance matching, causing severe surface EM reflection[4-9].

Along with this trend, two primary material architectures have been explored: hetero-double-shelled hollow microspheres and multi-component doped aerogels. The former,

typically comprising an inner dielectric shell and an outer magnetic shell, leverages internal voids for lightweight and thermal insulation, while the magnetic shell induces EM loss[10-14]. However, a fundamental trade-off exists: excessive contact between conductive shells forms detrimental thermal/electrical pathways, whereas excessive dispersion disrupts the formation of effective EM loss networks. Conversely, aerogels with hierarchical pores offer excellent thermal insulation via air entrapment and phonon scattering, and their open structures benefit EM impedance matching[15]. Yet, for a better EM energy attenuation ability, the integration of functional units (e.g., magnetic nanoparticles) into aerogels is necessary, which relies traditionally on indiscriminate doping and lacks precise spatial control over the distribution and connectivity of these functional units. This randomness limits the synergistic optimization of EM and thermal properties[16-23].

Therefore, although it can be expected that the rational combination of hollow microspheres and porous aerogel structure may hold great potential in lightweight yet robust simultaneous EM wave regulation and thermal management, its key challenge lies in achieving precise spatial orchestration of functional units within a porous matrix—simultaneously ensuring microscale aggregation for effective loss and macroscale isolation to prevent adverse coupling. Herein, we propose a novel ice-templating-guided hierarchical assembly strategy to tackle this challenge. Our approach uniquely employs pre-synthesized EM-loss hollow microspheres as ice-nucleating agents to direct the growth of ice crystals and, consequently, the bridging architecture of a carbon nanosheet network[24, 25]. Concurrently, via a seeding-assisted in situ chemical reduction process, magnetic nickel nanoparticles are controllably assembled onto the microsphere surfaces[26]. This dual-level control enables the intentional creation of concentrated loss units at the microscale while maintaining their spatial isolation within a three-dimensional macroscopic framework.

By virtue of this strategy, we constructed a composite carbon-nickel-glass aerogel (CNGA) with hierarchical porosity, which comprises chitosan-derived nitrogen-doped carbon nanosheets (N-CNSs) serving as conductive bridges and spacers, and nickel-

decorated glass dual-shell hollow microspheres (DSHMs) acting as concentrated loss units. The composition and microstructure were meticulously tailored. The optimized CNGA achieves an exceptional minimum reflection loss ($RL_{\min}$) of -54.86 dB and a maximum effective absorption bandwidth ($EAB_{\max}$) of 6.56 GHz at an ultralow filler loading of 8 wt%, while maintaining excellent thermal insulation. Finite element simulations elucidate the EM loss mechanisms enhanced by the controlled microsphere distribution[27]. Furthermore, by partially impregnating the aerogel with a resin to form a hybrid matrix of closed and open cavities, we demonstrate the feasibility of integrating mechanical robustness without significantly compromising EM performance[28]. This study not only presents a high-performance material but also, more importantly, introduces a generalizable strategy for the spatially controlled integration of functional units into porous scaffolds. Furthermore, new insights and a versatile platform are proposed for designing advanced composites with precisely controllable internal architecture, multifunctionality, low density, structural robustness and beyond.

2.Experimental section

2.1 Raw materials

Hollow microspheres with silicate glass shell (HGM) were produced by spray drying and subsequent high-temperature melting process as reported previously.[29] γ -Aminopropyltriethoxysilane (APTS) was purchased from Shanghai yuanye Bio-Technology. $NiSO_4 \cdot 6H_2O$, $(NH_4)_2SO_4$, $KNaC_4H_4O_6 \cdot 4H_2O$, $NaH_2PO_2 \cdot H_2O$, $SnCl_2 \cdot 2H_2O$, Hydrochloric acid, Chitosan, methylstyrene, benzoyl peroxide, TED85 and MHHPA were obtained from Sinopharm Group. $NH_3 \cdot H_2O$ solutions (25–28%), Acetate (99.5%) and $PdCl_2$ were got from Aladdin. All chemical reagents were used as received without further purification.

2.2 Preparation of DSHM

The synthesis of double-shelled DSHM was accomplished via the process of autocatalytic electroless plating, wherein metallic nickel building blocks were assembled onto glass microspheres through a process of seed-induced deposition. As

hollow spherical structure support and substrates for ferromagnetic nickel assembly, the HGMs were surface-activated before plating according to protocol[13]. With this activation, the immobilized nano catalysts catalyzed the reduction of nickel ions to metallic species at the interface, which initiates the directed assembly and triggers subsequent autocatalytic nickel deposition to form well defined nickel-glass dual-shell hollow microspheres (DSHM).

The plating solution contained: $\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$ (40 g/L), $\text{NaKC}_4\text{H}_4\text{O}_6 \cdot 4\text{H}_2\text{O}$, 90 g/L, $(\text{NH}_4)_2\text{SO}_4$ (60 g/L), $\text{NaH}_2\text{PO}_2 \cdot \text{H}_2\text{O}$ (50 g/L), and $\text{NH}_3 \cdot \text{H}_2\text{O}$ (50 mL/L). Typically, 4 g activated HGMs were added to 200 mL solution (60 °C, pre-stirred 1 min). After 10 min reaction, powders were filtered, washed 3× with deionized water, and immediately transferred to fresh plating solution for secondary assembly under identical conditions. Final products were separated by filtration, ethanol-washed and air-dried.

2.3 Fabrication of CNGAT-X and chitosan-derived carbon aerogels

The DSHM was uniformly mixed with chitosan acetate solution at different volume ratios (1:49, 1:19, 2:23, and 1:9), then poured into a custom axial freeze-casting mold (PTFE inner wall, copper base, $4 \times 4 \times 1 \text{ cm}^3$). The mold was immersed in liquid nitrogen for 10 minutes to induce oriented ice crystal growth. Subsequently, the mixture was freeze-dried at -50°C for 48 hours to remove ice crystals. The resulting CNGA intermediate aerogel samples with increasing DSHM volume ratios were designated CNGA-1 to CNGA-4[30]. The pure chitosan aerogel without the addition of DSHM was designated CA.

Finally, the obtained CNGA intermediate and CA were calcinated in a high-purity nitrogen atmosphere for two hours at 600°C , 650°C , 675°C , or 700°C , respectively with a constant heating rate of $10^\circ\text{C}/\text{min}$. Samples are denoted as CNGAT-X where T and X stand for the calcination temperature and the above mentioned CNGA intermediate number, respectively (e.g., CNGA-1 treated at 600°C : CNGA600-1). Chitosan-derived carbon aerogels are denoted as CAT where T stand for the calcination temperature.

2.4 Fabrication of R/C-X

The CNGA675-1, CNGA675-2, CNGA675-3, CNGA675-4 were selected as a model

to fabricate resin matrix composite (the corresponding samples were named R/C-1, 2, 3 and 4). A hybrid resin was employed. Specifically, Liquid A is prepared by thoroughly mixing methylstyrene and benzoyl peroxide in a mass ratio of 100:1.5. Liquid B is obtained by thoroughly mixing TDE85 and MHHPA in a mass ratio of 1:1. These two liquids are then blended in a 1:1 mass ratio to form a low-viscosity resin. Pour enough of the resin prepared in the previous step into the aerogels which were suspended at this stage. Subsequently, place the mixture in a vacuum oven to replace the air in the open cells by the resin until the aerogels were completely submerged. Repeat three times to ensure thorough resin penetration.

Finally, the following curing procedure was applied: 50°C (2 h) - 80°C (2 h) - 120°C (2 h) - 160°C (3 h) - natural cooling. A heating rate of 10 °C/min was maintained throughout the entire process.

2.5 Characterizations

The morphology of all samples was investigated using field-emission scanning electron microscopy (Hitachi-S4800, Japan). The EDS test results were obtained using an Oxford X-Max N 80 T energy dispersive spectrometer. TEM and (HRTEM) characterizations were carried out with an FEI Tecnai G2 F20 S-Twin (E.A. Fischione Instruments, Inc., USA) transmission electron microscope at 200 kV. The crystal structure of the as-obtained materials was examined via X-ray powder diffraction (XRD, D8Advance, Bruker, Germany). The magnetic hysteresis was measured on a vibrating sample magnetometer (Lake Shore 7404, USA) at room temperature. The XPS characterization was achieved on an X-ray photoelectron spectrometer (K-Alpha, ThermoFisher Scientific). Raman spectra were performed on a confocal Raman spectroscopic system (inVia-Qontor, Renishaw) using a 532 nm laser. The fourier transform infrared spectroscopy (FTIR) of aerogels were measured by attenuated total reflection (ATR) mode on Excalibur 3100 (Varian, USA), and the wave number range was 4000 cm^{-1} -500 cm^{-1} . The contact angle was measured by a contact angle measuring device (OCA20, German). The axial thermal conductivity of the aerogels was measured by Hot Disk (TPS 2500 s, USA). The surface temperature of CNGA675-1 was

measured using a handheld infrared thermal imager (DS2TPH10-3AUF, Hikvision). True density of DSHM was measured using an automatic density analyzer (Quantachrome Instruments, PentaPyc 5200e, USA). Electrical conductivities of aerogels were measured with a Dual-Type Four-Probe conductivity tester (Four Probes Tech, RTS-9, China). According to the GBT1041 standard, the compression tests of the resin/CNGA675-1 were carried out by a universal testing machine (HUALONG WDW-50C) and the compression rate was set to 1 mm/min. The dimensions of compressive specimens were $7 \times 7 \times 7 \text{ mm}^3$.

2.6 Electromagnetic performance measurement

The microwave absorbing performance of the aerogels was evaluated based on their electromagnetic parameters, which were measured using a vector network analyzer (VNA, Anritsu MS4642A) over the frequency range of 2–18 GHz via the coaxial method. To maintain structural integrity during testing, the aerogel was first infiltrated with paraffin wax. The wax was melted in an oven at 70°C, and the aerogel was immersed in the molten wax before being transferred to a vacuum oven to ensure complete pore infiltration. The sample was then cut into a concentric ring geometry with an outer diameter of 7 mm and an inner diameter of 3 mm suitable for coaxial testing. The filling ratio (FL) of the aerogel was defined as:

$$FL (\%) = (\rho_{aerogel} \times V_{ring}) / m_{ring} \times 100\%$$

where $\rho_{aerogel}$ denotes the bulk density of the original aerogel, V_{ring} , and m_{ring} represent volume and mass of the wax-infiltrated ring sample, respectively.

In addition, the electromagnetic properties of the resin/CNGA675-1 composite were measured by the waveguide method in the Ku-band (12.4–18.0 GHz). The sample was prepared with dimensions of 15.24 mm $\times$ 7.62 mm $\times$ 2.00 mm to fit the waveguide fixture.

2.7 Numerical simulation

The infinite two-dimensional array of honeycomb structures embedded with microspheres was modeled using a basic unit cell with Floquet periodic boundary conditions applied on all four sides. All models employed identical mesh and sphere

materials (complex permittivity and complex permeability values derived from transmission line theory at corresponding test frequencies), with air-filled gaps and perfect matching layers (PMLs) above and below the models. The absorber's base was modelled using a perfect electronic conductor boundary (PEC) condition to simulate the conductive coating used in practical applications. Port boundary conditions were applied at the inner boundaries of the PML and air domains. An orthogonal electromagnetic wave (1 W power, 12.0 GHz frequency) propagated along the Z-axis from the source port (with electric field polarization along the X-axis and magnetic field polarization along the Y-axis), then passed through the electromagnetic wave absorption material before exiting via the monitor port[27, 31, 32].

Results and discussion

3.1. Fabrication process and formation mechanisms

The synthesis of the hierarchical composite aerogel is fundamentally governed by an ice-templated heterogeneous nucleation strategy, as schematically illustrated in Figure 1a. The process commences with the self-assembly of metallic nickel onto dielectric glass cores to fabricate the dual-shell hollow microspheres (DSHMs)[33], which are subsequently dispersed homogeneously within a chitosan-acetic acid precursor. Following directional freezing and subsequent calcination-induced carbonization, the tailored composite aerogel exhibits ultra-low density (enabling it to rest seamlessly on delicate petals, Figure S1a), magnetic responsiveness (Figure S1c), and inherent self-cleaning attributes derived from its highly hydrophobic surface (Figure S1c-d).

Morphological evaluations of the pristine DSHMs (Figure S2a) confirm their well-defined spherical geometry and uniform exterior. Cross-sectional analysis of a fractured microsphere (Figure S2b) explicitly reveals the rationally designed core-shell architecture, comprising a 375 nm-thick inner silicate supporting shell encapsulated by a 210 nm-thick functional nickel layer, which is further corroborated by element mapping (Figure S2c). Dynamic light scattering (Figure S2d) confirms a normal size distribution with D10, D50, and D90 values of 35.88, 64.38, and 110.2 μm , respectively.

Crystallographically, XRD analysis of the DSHMs (Figure S3a) presents three sharp diffraction peaks at $2\theta = 44.65^\circ$, 51.37° , and 76.48° , corresponding to the (111), (200), and (222) planes of face-centered cubic nickel (PDF#70-1849), alongside a broad halo around 21.85° ascribed to the inner amorphous SiO_2^{30} . High-resolution TEM (HRTEM) further visualizes distinct lattice fringes with d-spacings of 2.07 Å and 1.79 Å (Figure S3b), substantiating the highly crystalline nature of the assembled nickel shell.

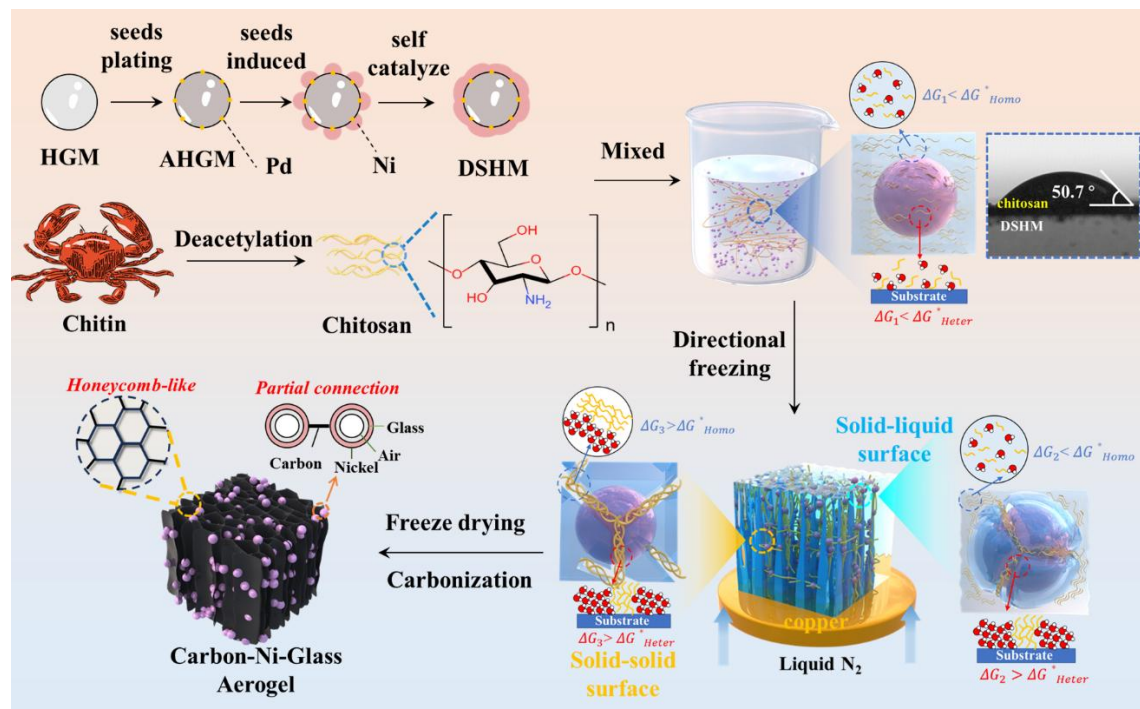


Figure.1 Schematic illustration of the fabricating process of CNGA.

The contact angle between DSHM and the chitosan–acetic acid solution (Fig. S4(a)) varied from 87.44° at 0 seconds to 50.70° after 90 seconds, indicating favorable wetting and ensuring uniform dispersion of DSHM within the solution. The mixture of chitosan solution and DSHM particles remained stable without stratification for over one hour (Fig. S4(b))[34-36]. Thermodynamically, this favorable liquid-solid interface significantly diminishes the Gibbs free energy barrier required for heterogeneous nucleation compared to homogenous nucleation within the bulk liquid[25]. Consequently, upon supercooling, ice crystals preferentially nucleate directly on and propagate radially outward from the DSHM surfaces (Figure S5). This spatially confined nucleation forcefully completely phase-separates the advancing ice front and the chitosan chains. Rather than forming continuous coats, the biomolecules are

templated to solely bridge the adjacent microspheres. Following calcination, this programmed connectivity precisely realizes a geometry where conductive nitrogen-doped carbon nanosheets (N-CNSs) bridge isolated micro-units, establishing the topological prerequisite for a distributed RLC electromagnetic network.

3.2 Morphology and composition

To structurally decode this distributed RLC network, we evaluated how DSHM volume fraction programs the topological connectivity of the composite. Radial and axial cross-sections (Figure S6 and Figure 2) unveil that the DSHM concentration fundamentally alters the ice crystal propagation kinetics. At an optimal low filling fraction (CNGA675-1, Figure 2a₁-a₂), the isolated DSHMs act as discrete nodes beautifully bridged by intact bi-dimensional N-CNS matrices, maintaining the original cellular architecture. As the DSHM loading increases (CNGA675-2, Figure 2b₁-b₂), adjacent microspheres initiate localized contacts without catastrophic network collapse. However, excessive loading (CNGA675-4, Figure 2c₁-c₂) overwhelmingly forces inter-microsphere aggregation, disrupting the heterogeneous nucleation equilibrium and merging the scaffold into continuous conductive lamellar blocks—a state bound to severely mismatch free-space impedance. Unobstructed by continuous aggregation, micro-CT reconstruction perfectly visualizes the optimized state in CNGA675-1 (Figure S7): the high-density DSHMs (bright regions) are homogeneously anchored across the highly porous, longitudinally oriented carbon framework (dark regions) in a mesoscopically decoupled yet electrically bridged state. Detailed compositional analyses untangled the localized physics across the heterogeneous interfaces. EDS mapping (Figure 2d) validates that the un-aggregated nickel microspheres are tightly embraced by the N-CNS bridges. More fundamentally, HRTEM examination of this metal/carbon boundary (Figure 2e–g) exposes an oriented graphitic lattice fringe (0.345 nm) immediately adjacent to the well-defined Ni (111) lattice (0.2085 nm) within the predominantly amorphous framework. This confirms that metallic nickel catalytically induces localized graphitization of the amorphous carbon, essentially providing swift electron tunneling channels that elevate intrinsic conduction loss.

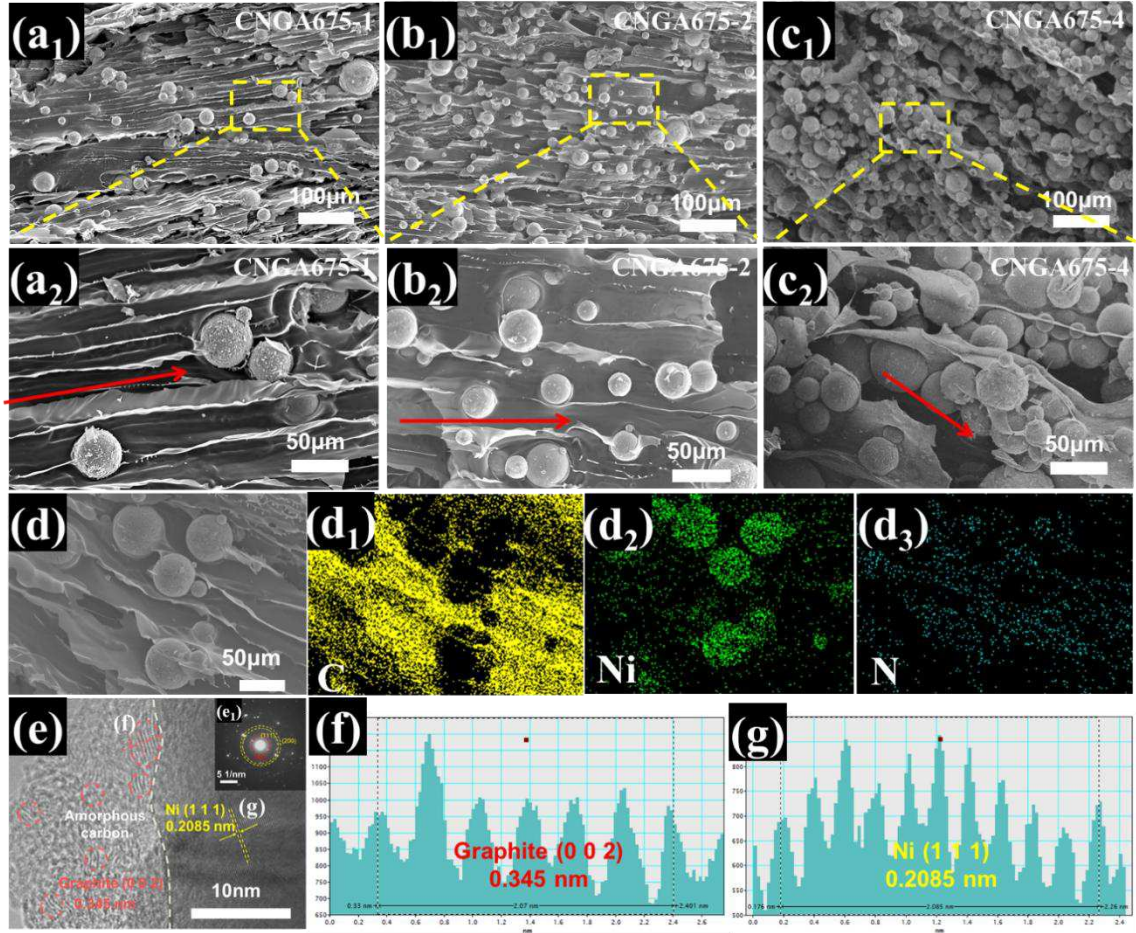


Figure.2 Comprehensive characterization of CNGA: SEM images of (a₁-a₂) CNGA675-1, (b₁-b₂) CNGA675-2, (c₁-c₂) CNGA675-4; (d-d₃) EDS mapping of CNGA675-1; (e) HRTEM images; (f-g) lattice spacing measurements of graphite (0 0 2) and nickel (1 1 1).

Simultaneously, the chemical bonding evolution was mapped via XRD, FTIR, and XPS. As shown in Fig.3a, the XRD spectra of CA/CA675/CNGA-1/CNGA675-1 were presented to illustrate the crystal structure changes of chitosan aerogel (CA) with the addition of DSHM and calcination. The wide diffraction peaks at around 21° correspond to amorphous SiO₂ peaks, which needs to be distinguished from CA characteristic peak. And a prominent broad diffraction peak at 25° can be attributed to the (0 0 2) graphite carbon structure[21]. Notably, three intense peaks are observed at 44.8°, 51.8° and 76.3°, which can be indexed to the (1 1 1), (2 0 0), (2 2 0) crystal planes of metallic Ni (JCPDS No.70-1849), respectively[34]. All distinctive FTIR macromolecular vibrations of chitosan vanish. Importantly, a preceding C–N redshift

in the precursor (Figure 3b) indicates strong $\text{N} \rightarrow \text{Ni}^{2+}$ coordination bonds[35], which subsequently evolve into robust interfacial C–O–Ni linkages (669 cm^{-1} and 1069 cm^{-1}) in CNGA675-1. High-resolution C1s XPS spectra strictly track extensive structural deoxygenation (Figure 3d) [36]. More critically, within the carbonized lattice (Figure 3e), the initial amine and glycosidic nitrogen species are successfully transformed into pyridinic N (398.4 eV), pyrrolic N (400.7 eV), and graphitic N (402.6 eV)[22]. These precise nitrogen dopants profoundly skew the regional charge symmetry, acting as prolific stable dipole centers that are heavily polarized under alternating electromagnetic fields, thus fundamentally bolstering the dielectric polarization loss capability of the continuous bridges. Ni 2p high-resolution spectra (Figure S8) confirm a dynamic coexistence of catalytic metallic Ni^0 states ($853.1/870.6\text{ eV}$) and surface-passivated NiO layers ($856.7/874.5\text{ eV}$).

Furthermore, Raman spectroscopy tracks the defect dynamics (Figure 3h). With escalating DSHM content, the D-band (1340 cm^{-1} , structural defects) to G-band (1580 cm^{-1} , sp^2 graphitic domains) intensity ratio (I_D/I_G) slightly intensifies from 0.96 to 1.04, reflecting amplified hetero-interfacial lattice distortions that further contribute to defect-induced polarization⁴². The hierarchical microporosity accommodating this composite was accurately parameterized via mercury intrusion porosimetry (Figure 3f, g). The saturated intrusion volume drops successively from 9.28 mL/g down to 4.71 mL/g as the microsphere density increases, coinciding with a macroscopic pore size reduction from $10.8\text{ }\mu\text{m}$ to $6.83\text{ }\mu\text{m}$. Interestingly, nanoscale void imperfections ($10\text{--}100\text{ nm}$) densely accumulate at higher DSHM loadings, primarily introduced by water infiltrating the rough nanoscopic crevices of aggregating microspheres prior to sublimation.

Ultimately, this bottom-up programmed connectivity rigorously tailors macroscopic electromagnetic parameters. As captured in Figure 3i, climbing DSHM fractions gradually improve overarching conductivity up to a critical percolation transition between CNGA675-2 and CNGA675-3. Beyond this threshold, the bridging isolation is forfeited to continuous conductive routes, fatally shifting the composite from a

distributed attenuation network to an EM reflector[37]. Concurrently, static magnetic properties (Figure 3j) document a steady ascent in saturation magnetization (9.8 to 14.4 emu/g) and robust coercivity (111.2 to 141.2 Oe). The amplified coercivity conceptually roots in the multidomain geometry and heterogeneous pinning centers physically restricting domain wall propagation, compelling magnetization reversal to proceed via arduous coherent moment rotations[38]. Crucially, this forcibly elevated magnetic anisotropy shifts natural resonances up to the targeted GHz microwave region, securing dynamic magnetic dissipation cooperativity within the integrated RLC network[39].

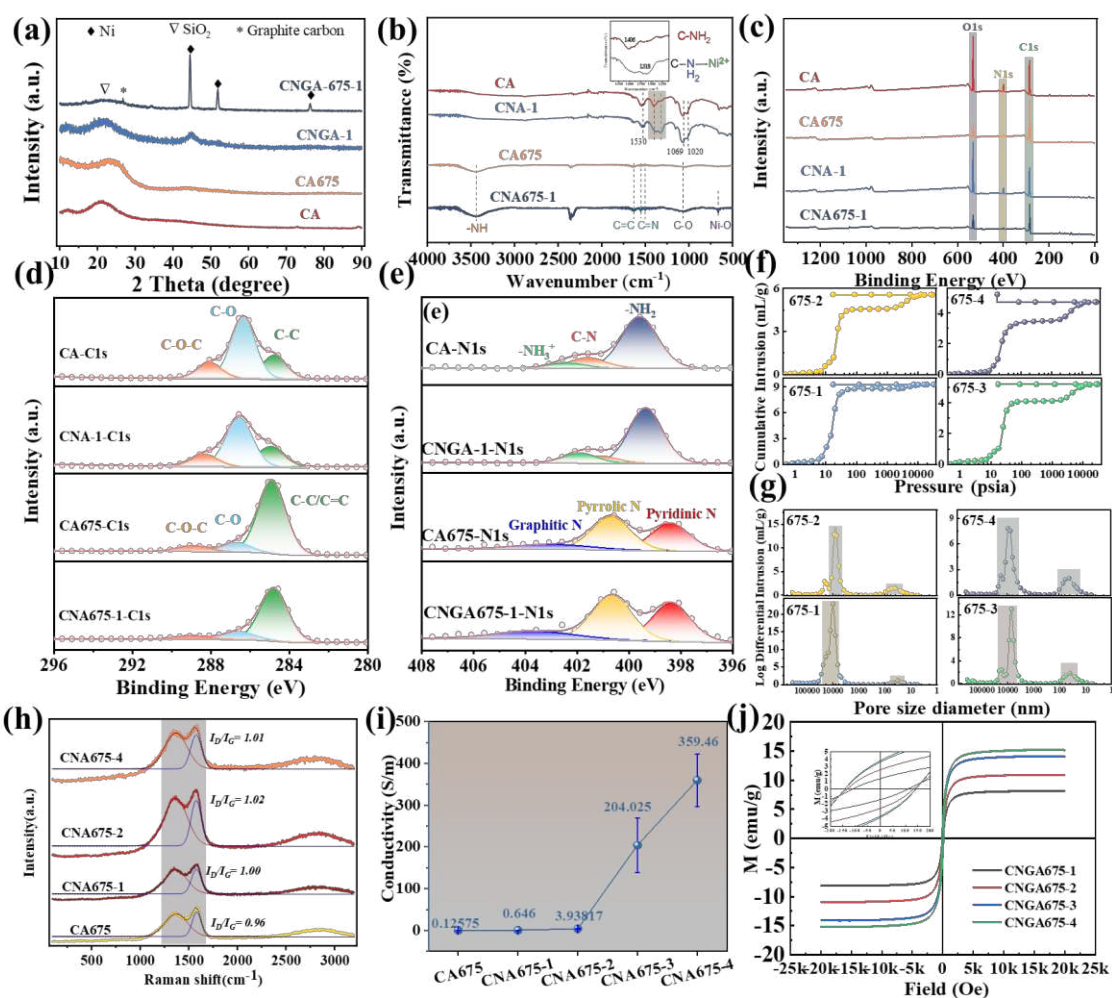


Figure.3 (a) XRD curves, (b) FT-IR, (c) XPS full spectra, (d) C1s region, (e) N1s region of CA, CA675, CNGA-1 and CNGA675-1; (f) Mercury intrusion-extrusion isotherms, (g) Mercury intrusion-extrusion isotherms, (h) Raman spectra, (i) electrical conductivity and (j) Magnetic hysteresis loops of CNGA675-1 to CNGA675-4

3.3. Electromagnetic wave absorption performance

To elucidate the intrinsic electromagnetic trade-off comprehensively, we first investigated the microwave absorption of paraffin-based mixtures containing either pure DSHMs or pure carbon aerogels (CAs). For conductive fillers, the spatial volume fraction dictates interparticle contact, heavily influencing the effective permittivity. According to effective medium theory, adding more conductive particles boosts the system's ability to store and dissipate electromagnetic energy. However, as the fraction approaches the percolation threshold, the emerging interconnected conductive network inevitably triggers severe impedance mismatch. To quantitatively evaluate this electromagnetic interaction, the reflection (R), absorption (A), and transmission (T) coefficients were derived from the scattering parameters (S_{11} , S_{22} , S_{12} , S_{21}) via vector network analyzer (VNA) measurements:[14]

$$T = S_{12}^2 = S_{21}^2 \quad (1)$$

$$R = S_{11}^2 = S_{22}^2 \quad (2)$$

$$A = 1 - T - R \quad (3)$$

As mapped in Figure S9a–c, a low DSHM fraction (15.4%) results in dominant transmission because the particles are too scattered to build a valid scattering network. When the loading hits 26.4%, it nears the theoretical infinite percolation threshold for ideal spheres (~ 0.29)[37, 40]. At this point, the DSHMs begin to make localized microscopic contact. Consequently, transmittance drops steeply, and reflectance starts to govern the high-frequency range. Pushing the loading further to 41.2% completely blocks wave penetration due to overwhelming interfacial reflection. To dissipate waves efficiently, the impedance matching ratio ($Z = Z_{in}/Z_0$) must stay close to 1.0 (Figure S9d–f). A highly continuous network makes the material too conductive, driving Z toward zero and bouncing the incoming waves back. The core material challenge was finding a way to exploit the strong attenuation of DSHMs without suffering from aggregation-induced mismatch.

Concurrently, the microstructural evolution of the continuous chitosan-derived carbon bridging networks essentially relies on the carbonization temperature[41]. As

investigated (Figure S10), although the CA matrix carbonized at 675 °C (CA675) presents visually superior matching over CA650 and CA700, its relatively weak intrinsic polarization loss restrains its practical absorption magnitude, manifesting in a prominent transmission tail (Figure S10d–f). This confirms that pristine CAs natively possess favorable characteristic impedance but critically lack dense attenuation motifs, thereby verifying the necessity of topologically weaving the highly lossy DSHM micro-units into this 3D carbon scaffold via our ice-templated heterogeneous nucleation strategy.

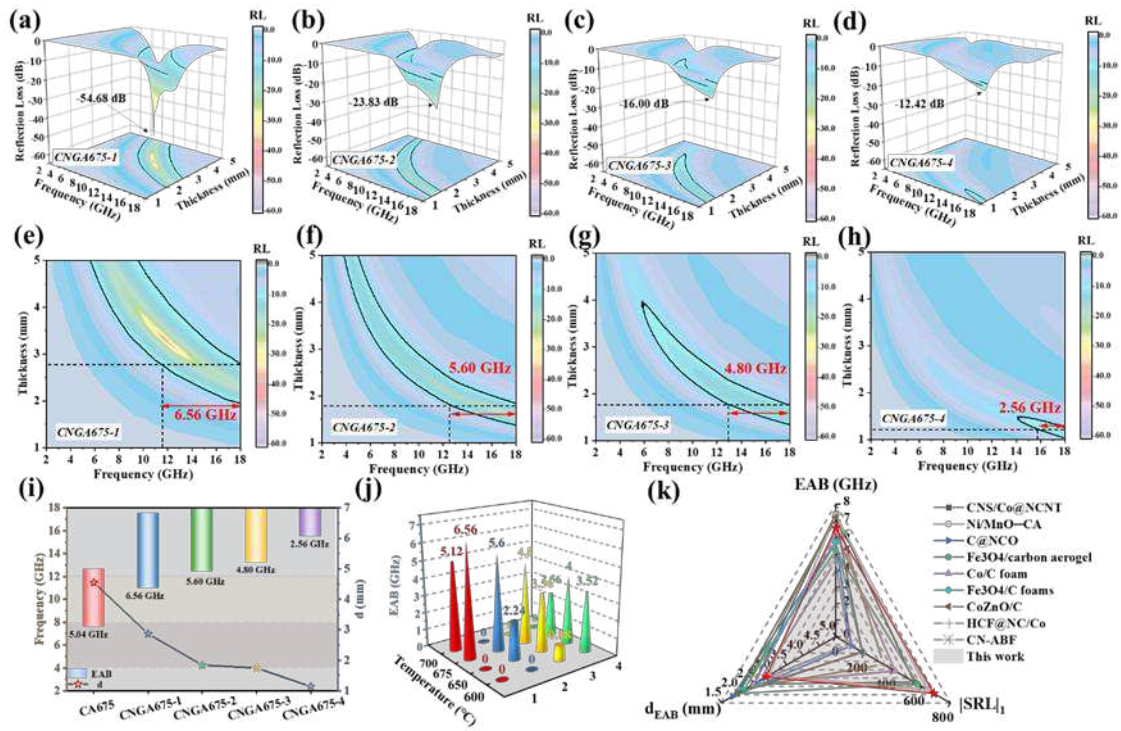


Figure.4 (a-d) 3D RL contour maps of the aerogels; (e)-(h) 2D RL contour maps of the aerogels; (i) Summary of EABs and corresponding thicknesses for aerogels; (j) Comparison of EABs at different carbonization temperatures and DSHM volume fractions; (k) Radar chart of the properties for magnetic metal-carbon aerogels in recent years[42-45, 11, 46-50].

By elegantly averting the formation of continuous conductive macroscopic shells, this trade-off is exquisitely resolved in the integrated architecture. Figure 4a–d maps the 3D RL contours for the hierarchical CNGA architectures, alongside their 2D projections (Figure 4e–h). CNGA675-1 exhibits state-of-the-art microwave absorption

capabilities, delivering an optimal $RL_{\min}$ of -54.86 dB at a thickness of merely 3.32 mm, and an ultra-broad effective absorption bandwidth (EAB) of 6.56 GHz (10.96 – 17.52 GHz)—covering over 91% of the Ku-band—at a thin matching thickness of 2.87 mm. As the DSHM loading further increases (CNGA675-3 and CNGA675-4), the EAB shrinks dramatically to 4.8 GHz and 2.56 GHz, respectively, consistent with the premature impedance mismatch discussed earlier. As shown in Figure S11a-d, the R-A-T coefficients curves exhibit a clear pattern of variation: the rise in the R curve confirms the impedance mismatch caused by excessive contact between the microspheres. Crucially, thermogravimetric analysis (TGA, Figure S12) and theoretical tracking (Equations S1–10, Figure S13) mathematically confirm identical phase volume fractions across variable synthesis temperatures. These systematic evaluations substantiate that precisely programming the geometric isolation of thin-shell DSHMs via the ultra-lightweight N-CNS bridging architecture effectively constructs a macroscopic spatially distributed lossy network. It is noteworthy that CNGA675-1 features a thin-shell structure composed of magnetic metal, accounting for only 0.26% of the total volume, and hollow microspheres, accounting for 7.32% of the total volume. This design has been shown to enhance the spatial utilization efficiency of the metal, thereby facilitating its controlled aggregation. Compared with recently reported 3D porous absorbing materials (Figure 4o), CNGA675-1 yields an overwhelming specific reflection loss ($SRL[27] = RL/\text{filler loading}$) of -685.75 dB/g, powerfully confirming its superiority as an ultra-lightweight, high-efficiency electromagnetic absorber.

3.4. Electromagnetic Wave Absorption Mechanism

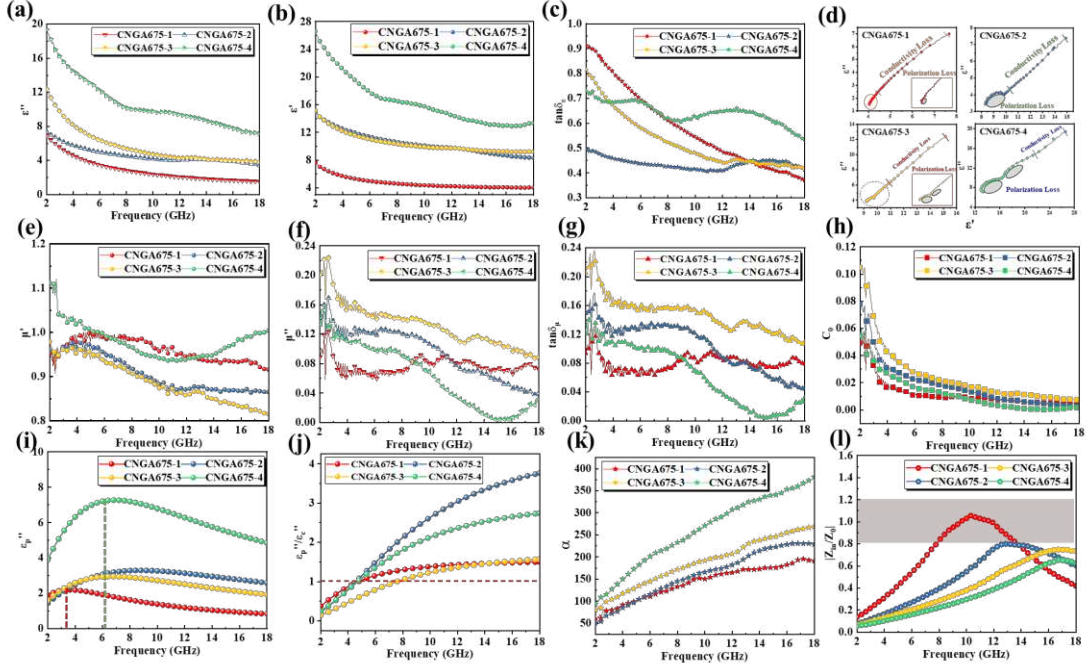


Figure. 5 (a-b) Real and imaginary parts of permittivity; (c) Dielectric loss tangent. (d) Cole-cole curves; (e-f) Real and imaginary parts of magnetic permeability. (g) magnetic loss tangent; (h) Eddy current loss; (i) Polarization loss; (j) The ratio of polarization loss to conduction loss; (k) The attenuation constant; (l) The impedance matching performance at the thickness achieving RL_{min} of CNGA675-1 to CNGA675-4

According to the free electron theory ($\epsilon'' = \sigma/2\pi\epsilon_0 f$), the prominent elevation in both the real (ϵ') and imaginary (ϵ'') parts of complex permittivity with increasing DSHM content (Figure 5a–b) underscores an enhanced capacity for electric energy storage and conduction loss. In similar, dielectric loss tangent ($\tan \delta\epsilon$) for CNGA675-1 to CNGA675-4 exhibit pronounced frequency dispersion (Figure 5c). The dense proliferation of multiphase heterointerfaces specifically intensifies spatial charge accumulation. Using the Cole–Cole relation derived from Debye relaxation (Equation 4) the $\epsilon' - \epsilon''$ plots (Figure 5d) present explicit semicircles correlating to distinctive relaxation behaviors[51]:

$$\left(\epsilon' - \frac{\epsilon_s + \epsilon_\infty}{2}\right)^2 + (\epsilon'')^2 = \left(\frac{\epsilon_s - \epsilon_\infty}{2}\right)^2 \quad (4)$$

For CNGA675-1 and CNGA675-2, the single semicircle implies a unified interfacial polarization mainly localized across the spatially isolated DSHM hetero-shells. However, overloaded samples (CNGA675-3 and CNGA675-4) display dual semicircles accompanied by a steep linear tail, indicating aggressive inter-microsphere aggregation that triggers secondary structural relaxations and massive continuum leakage currents.

For magnetic attributes, the real (μ') and imaginary (μ'') permeability parts confirm typical high-frequency dispersion (Figure 5e–f). Initially, the incorporation of magnetic DSHMs elevates μ'' ; yet, exceeding the geometric percolation threshold severely degrades magnetic attenuation, as excessively strong induced conduction currents generate counteracting demagnetization fields. Examining the eddy current loss evaluation ($C_0 = \mu''(\mu')^{-2}f^{-1}$, Figure 5g–h) clarifies that magnetic dissipation in the 2–6 GHz range primarily originates from geometric natural and exchange resonances. Across 6–18 GHz, C_0 stabilizes for CNGA675-1 (~ 0.08), validating the dominance of eddy current loss. Conversely, heavily loaded samples rapidly lose magnetic performance due to their excessive intrinsic conductivity triggering adverse skin effects.

By mathematically decoupling the experimental dielectric polarization (ε_p'') and conduction (ε_c'') losses (Equation 5)[52], we clarify that at high-frequency ranges ($f > f_0$), extensive conduction loss dominates the macro-attenuation, which is forcefully magnified by the localized Ni-induced graphitization tunneling channels.

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

Meanwhile, interfacial polarization governs the low-frequency dissipation (Figure 5i–j). Notably, the stable electric dipoles introduced by localized pyridinic and pyrrolic N-dopants fundamentally bolster these polarization magnitudes by aggressively skewing the molecular charge symmetry. The attenuation constant (α , Figure 5k) escalates with frequency across all samples, but impedance matching (Z , Figure 5l) reveals that only CNGA675-1 safely resides within the ideal optimal gap ($Z = 0.8\text{--}1.2$). Overloaded networks endure severe impedance mismatch, virtually shielding out incoming waves via surface reflection.

Crucially, the underlying physical interplay in CNGA675-1 is exquisitely

abstracted as a distributed RLC micro-circuit network, intimately mirroring our ice-templated architecture. Total effective impedance is formulated as[53]:

$$Z_{in} = R + j\omega L + \frac{1}{j\omega C} \quad (4)$$

To explore the complex parameter state space of the distributed RLC network, we conducted a systematic study on how synthesis thermodynamics (specifically carbonization temperature) and spatial filler loading (DSHM volume fraction) influence the macroscopic electromagnetic properties. In this framework, the carbonization temperature serves as a critical control for the graphitization degree, which in turn regulates the intrinsic ohmic resistance (R) of the N-CNS bridges. Meanwhile, the DSHM fraction is used to fine-tune the localized capacitance (C) and inductance (L)[54]. At lower temperatures (600 °C and 650 °C), the N-CNS bridging skeleton does not reach a high level of graphitization, leading to high intrinsic macroscopic resistivity. As a result, for samples with low DSHM content like CNGA600-1 and CNGA600-2, the complex permittivity is notably suppressed (Figure S14a–b), which keeps energy dissipation low and limits overall attenuation. Following the RLC resonance principle ($\omega_0 = 1/\sqrt{LC}$) [55], we observed that increasing the DSHM concentration forces the resonance frequency to drop from 15.92 GHz down to 10.4 GHz (Figure S14c). In such a high-resistance matrix, strong microwave absorption only appears when DSHM loading reaches extreme levels. At these limits, the microspheres are forced to touch locally, compensating for the poorly conductive carbon bridges. A clear example is at 650 °C, where high DSHM volume fractions trigger a unique inverse dispersion (Figure S15a–c). This behavior activates stronger resonance above 18 GHz, allowing CNGA650-4 to show a reflection loss of –54.86 dB at 12.16 GHz with a very thin matching thickness of 1.47 mm. In this state, because the N-CNS bridges are under-active, the overloaded DSHMs must carry the burden for both conduction and polarization. On the other hand, aggressive heating at 700 °C leads to an opposite problem. Excessive graphitization and significant volume shrinkage cause the intrinsic resistivity of the carbon networks to plummet. This thermal over-activation turns the interconnected skeleton into a continuous, macroscopic conductive reflector,

regardless of the DSHM loading. Such a change creates a severe impedance mismatch originating from the matrix itself. Adding DSHMs to this 700 °C environment only worsens the "skin effect," which blocks microwave penetration and ruins the absorption performance (Figure S16). Our tests on pure carbon aerogels (CAs) confirm this limitation: CA650 lacks enough dielectric loss, while CA700 fails because its unmatched impedance leads to absolute reflection (Figure S10f). From a thermodynamic perspective, setting the synthesis at 675 °C and using a decoupled DSHM geometry (CNGA675-1) allows us to hit the ideal phase "sweet spot." Here, the N-CNS bridges offer just enough localized conduction (R) to turn microwave energy into micro-currents without causing heavy surface reflection. At the same time, the well-isolated microspheres provide the best localized interfacial polarization (C) without forming short-circuit paths. Ultimately, this detailed temperature-concentration study proves that the superior performance of CNGA675-1 is not a random result. Instead, it is the direct outcome of reaching a carefully programmed electromagnetic RLC equilibrium.

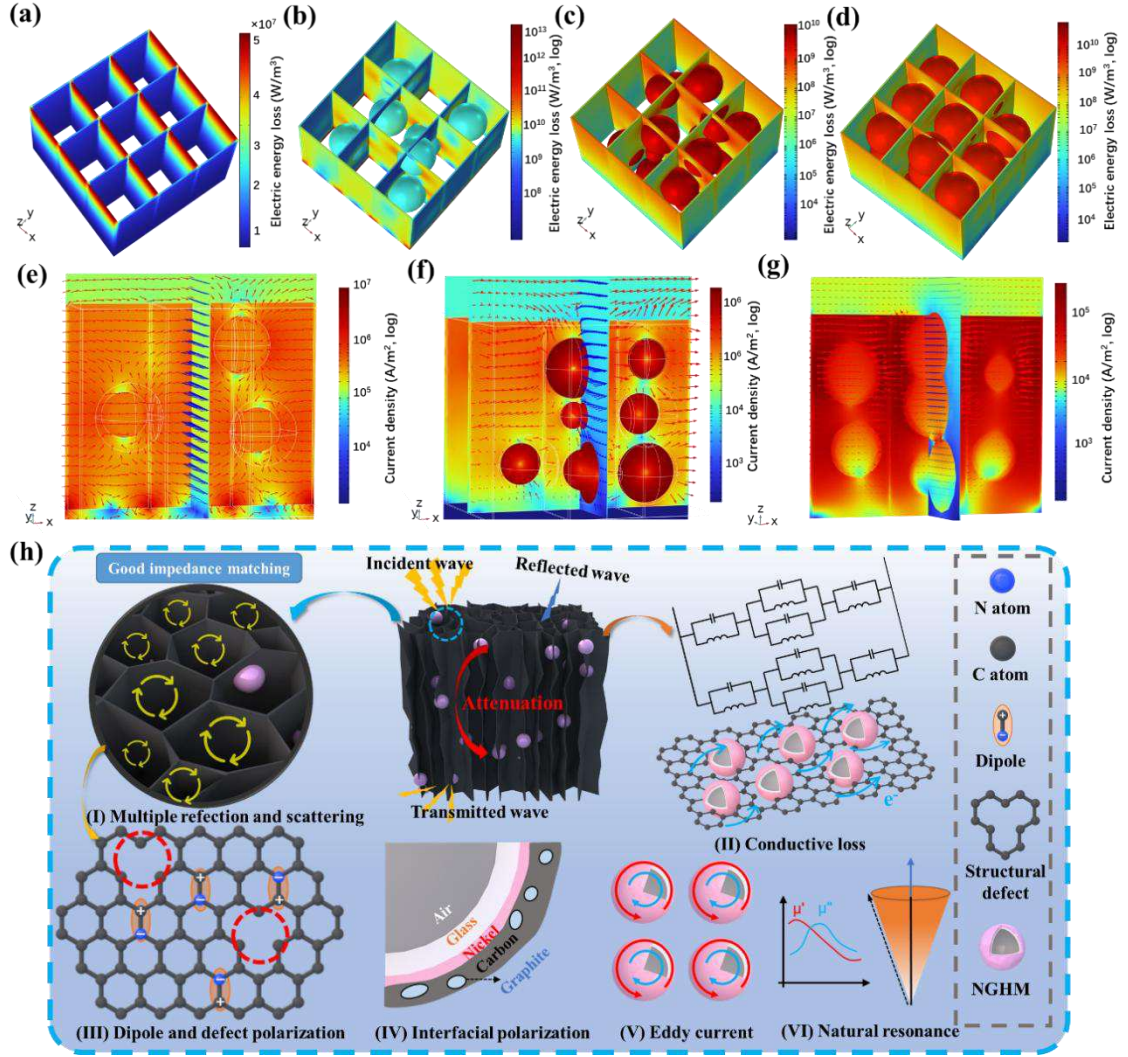


Figure. 6 Electromagnetic loss power density distribution of (a) CA675, (b) CNGA675-1, (c) CNGA675-2, (d) CNGA675-4. Cross-sectional diagram of current density and direction for (e) CNGA675-1, (f) CNGA675-2, and (g) CNGA675-4. (h) Schematic diagram of the mechanism for absorbing electromagnetic waves in CNGA675-1.

To visually corroborate the topological evolution of this distributed RLC mechanism, finite element method (FEM) simulations mapped both the overarching spatial power dissipation and localized induced current densities[56]. For the pristine matrix (CA675, Figure 6a), electromagnetic energy dissipation is strictly ohmic and structurally planar. Power loss concentrates predominantly along the electric field polarization axis (x-axis) on the macro-surface, lacking deep-tissue penetration, which fundamentally restricts its attenuation magnitude. Upon configuring the optimally isolated RLC network

(CNGA675-1, Figure 6b), the spatial dissipation geometry broadens into a volumetric distribution. Intense dissipation zones emerge not only around the distributed heterointerfaces but also firmly at the bottom boundary against the perfectly electrically conducting (PEC) substrate. This deep-tissue bottom loss physically verifies an excellent impedance matching state: incident waves smoothly traverse the bulk aerogel, allowing robust secondary reflections from the metal backing to be trapped and consumed within the 3D depth of the absorber. However, as the DSHM loading escalates to enforce localized geometric contacts (CNGA675-4, Figure 6c–d), the macroscopic attenuation paradigm shifts. The profound absence of bottom-region dissipation explicitly indicates that incident waves are blocked from entering the matrix, regressing the loss strictly back to the incident surface due to macroscopic impedance mismatch. The exact dynamic behaviors of the RLC equivalent circuit are profoundly elucidated by the induced current density vectors (Figure 6e–g). In the perfectly matched CNGA675-1 architecture, induced current densities peak specifically at the equatorial regions of the uniformly dispersed DSHMs while diminishing at the polar tips. This localized charge accumulation directly illustrates the dynamic alternating charging/discharging cycles of the discrete micro-capacitors (C). Simultaneously, these localized displacement currents are safely guided through the structurally tortuous N-CNS bridges (L and R), verifying a synergistic capacitive-inductive resonant network. In contrast, when filler overloading obliterates the physical isolation boundaries (CNGA675-4, Figure 6g), the mesoscopic electron transport pathways are rewired. Given the inherently higher conductivity of metallic nickel shells, induced currents tend to flow directly between the physically touching microspheres. This trans-microsphere conduction structurally short-circuits the discrete micro-capacitive loops, amplifying continuous surface currents. Consequently, the 3D resonance network degrades into a conductive surface boundary, physically barring internal EM energy entry.

As schematically synthesized in Figure 6h, the mesoscopic synergy achieved via heterogeneous nucleation rigorously avoids adverse continuous conduction. The vertically aligned N-CNS scaffolds offer robust macroscopic impedance matching

pathways, while the discrete DSHMs behave as micro-capacitors to intensify localized polarization. Together, this structurally decoupled topology orchestrates unparalleled broad-band dielectric-magnetic attenuation through tightly confined eddy-current dissipation, natural resonance, and supreme interfacial polarization within an optimum RLC loop.

3.5 Heat transfer and infrared radiation shielding performance

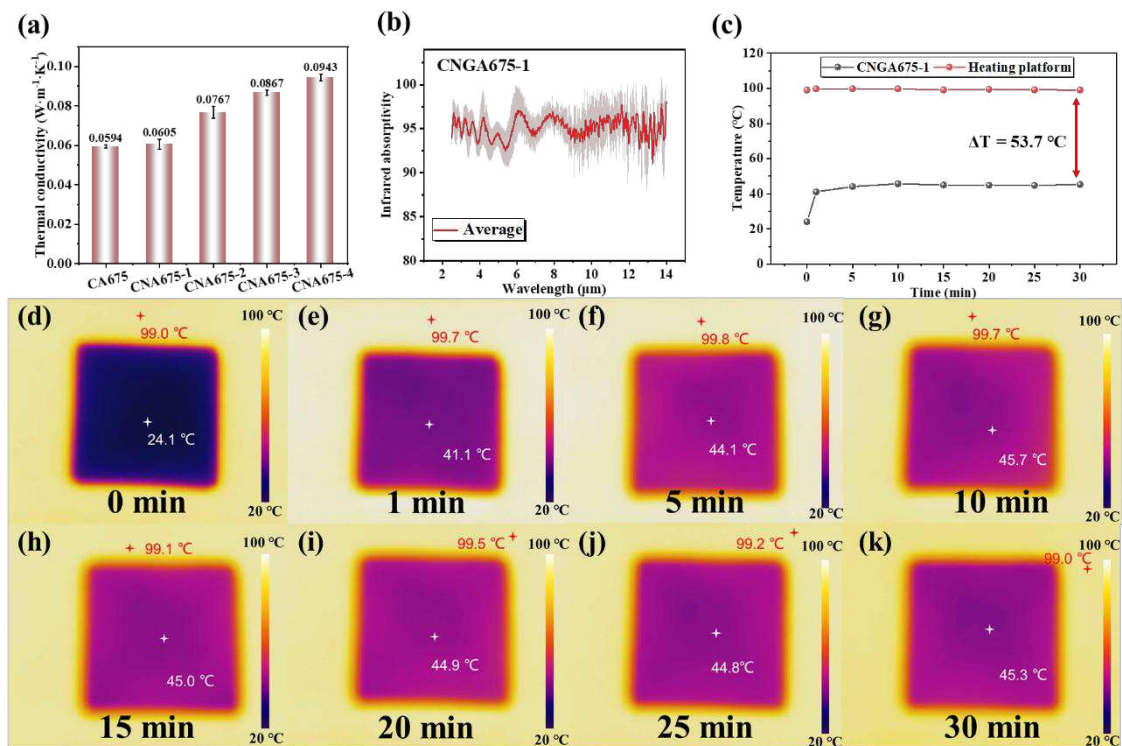


Figure.7 (a) Axial thermal conductivity. (b) Infrared emissivity at the bottom and top of CNGA675-1. (c) Temperature variation at different times. (d-k) Thermal insulation performance of CNGA675-1

To comprehensively evaluate the multi-environment adaptability of the composite, its steady-state thermal transport and dynamic infrared radiation shielding behaviors were systematically investigated. As depicted in Figure 7a, the axial thermal conductivity was mapped across varying DSHM spatial loadings. Notably, CNGA675-1, which operates at the optimal electromagnetic phase equilibrium, concurrently delivers an ultra-low thermal conductivity of merely $0.0605 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$. Furthermore, the structurally optimized composite yields a high macroscopic infrared spectral absorptivity of 95% (Figure 7b), effectively restricting the continuous transmission of

incident thermal radiation. During a dynamic thermal exposure mapping (Figure 7c), after maintaining the localized background temperature at approximately 100 °C for 30 minutes, the topmost surface of CNGA675-1 stably equilibrated near 45 °C, establishing a robust steady-state temperature gradient of 53.7 °C. It must be clarified that the inherent 95% high infrared absorption (corresponding to a high emissivity, $\epsilon \approx 0.95$) typically renders carbon-based materials potent thermal radiators. However, in our CNGA architecture, effective infrared stealth is achieved uniquely via a temperature-suppression paradigm. According to the Stefan–Boltzmann law ($E = \epsilon\sigma T^4$)[57], the macroscopic thermal radiation outflux is exponentially governed by the surface temperature. The ultra-low thermal conductivity of our continuous pore structure rigorously blocks bottom-up heat tunneling, drastically depressing the operating surface temperature (T) and thereby fundamentally overriding the high native emissivity. This thermal-insulation-driven radiation blockade provides a highly reliable stealth strategy in severe externally heated environments.

3.6 Air replacement with resin for mechanical reinforcement

While the macroscopic aerogel configuration delivers ultra-lightweight functionality, effectively translating these highly interconnected continuous skeletons into load-bearing structural components requires resolving their intrinsic mechanical fragility[58, 28]. Herein, replacing the internal air with an EM-transparent resin medium via vacuum-assisted impregnation safely reconfigures the skeletal phase into a robust 3D matrix. Standard static compression testing (Figure 8a) reveals that the highly integrated composite (hereafter denoted as R/C-1) achieves a resilient compressive strength of 45.70 ± 1.86 MPa. By computing the theoretical and experimental densities (Figure 8b), the highly consistent correlation confirms that the low-viscosity resin accurately infiltrates the open-cell mesoporous regions while perfectly preserving the structurally enclosed structural-vacuum interiors of the pristine DSHMs, forming a highly synergistic triple-phase heterogeneous bulk. However, as the localized DSHM volume fraction excessively increases (Figure 8c), the structural compressive strength progressively declines. This mechanical regression primarily corresponds to the

proliferation of rigid, defect-abundant discrete heterojunctions, shifting the dominant mechanical failure paradigm from a ductile, matrix-yielding mode directly toward a brittle, interface-fracturing mode.

Crucially, the structurally encapsulated RLC topological network securely maintains its superior electromagnetic dissipating capacity even after transferring into the solid bulk matrix. Notably, replacing pervasive air ($\epsilon_r \approx 1$) with the solid resin matrix ($\epsilon_r \approx 2.5\text{--}3.0$) deliberately shifts the background dielectric environment. From the perspective of our developed RLC distributed network, this matrix transition essentially intensifies the localized capacitance (C) of the inter-microsphere heterojunctions (as $C = \epsilon S/d$) while maintaining the topological continuity of the resistive-inductive ($R - L$) carbon framework. The sustained superior microwave attenuation in R/C-1 profoundly verifies that our spatially decoupled geometrical design possesses broad matrix-independent tunability, ensuring deterministic multi-band stealth functionality across varying dielectric service media.

Evaluating the Ku-band EM parameters of R/C-1 (Figure 8d₁–d₂), the real permittivity stably anchors near 4, while the imaginary part exhibits notable frequency dispersion. The combined Cole-Cole analytical profiles (Figure 8e) delineate robust multi-stage Debye semicircles, confirming that intense dipole relaxations localized across the structurally preserved hierarchical heterointerfaces remain fully active. Consequently, the modeled macroscopic impedance matching (Figure 8g) reaches an optimal operational state near 2.5 mm, ultimately orchestrating exceptional broadband attenuation (Figure 8h–i). The reflection loss minimum (RL_{min}) reaches -74.68 dB, and the effective absorption comprehensively spans the entire Ku band.

To explicitly validate that the exceptional macroscopic EM capacity genuinely originates from the tightly programmed 3D topological bridging rather than merely the physical presence of heterogeneous fillers, a deliberately destructive protocol was implemented. Specifically, the intact CNGA porous scaffold was physically crushed to disrupt the spatial networks and subsequently re-dispersed into a matrix under an identical mass fractional layout. Following fundamental topological devastation, both

the macroscopic dielectric storage and dissipation capabilities of the dispersed counterparts uniformly collapsed (Figure S17a₁–a₂). The corresponding loss tangents severely plunged (Figure S17b₁–b₂), inducing a critical degradation of the overarching wave-absorbing profile (Figure S17c). This fundamental verification precisely proves that preserving the geometrically decoupled, continuous carbon-bridging framework is the absolute prerequisite for successfully activating and stabilizing the distributed macroscopic RLC equivalent circuit. While mechanical crushing inevitably induces secondary damage to localized micro-interfaces, this catastrophic reduction in attenuation predominantly verifies that isolating fillers without a coherently programmed conducting topological network renders the individual loss capacities functionally isolated and impotent on a macroscopic scale.

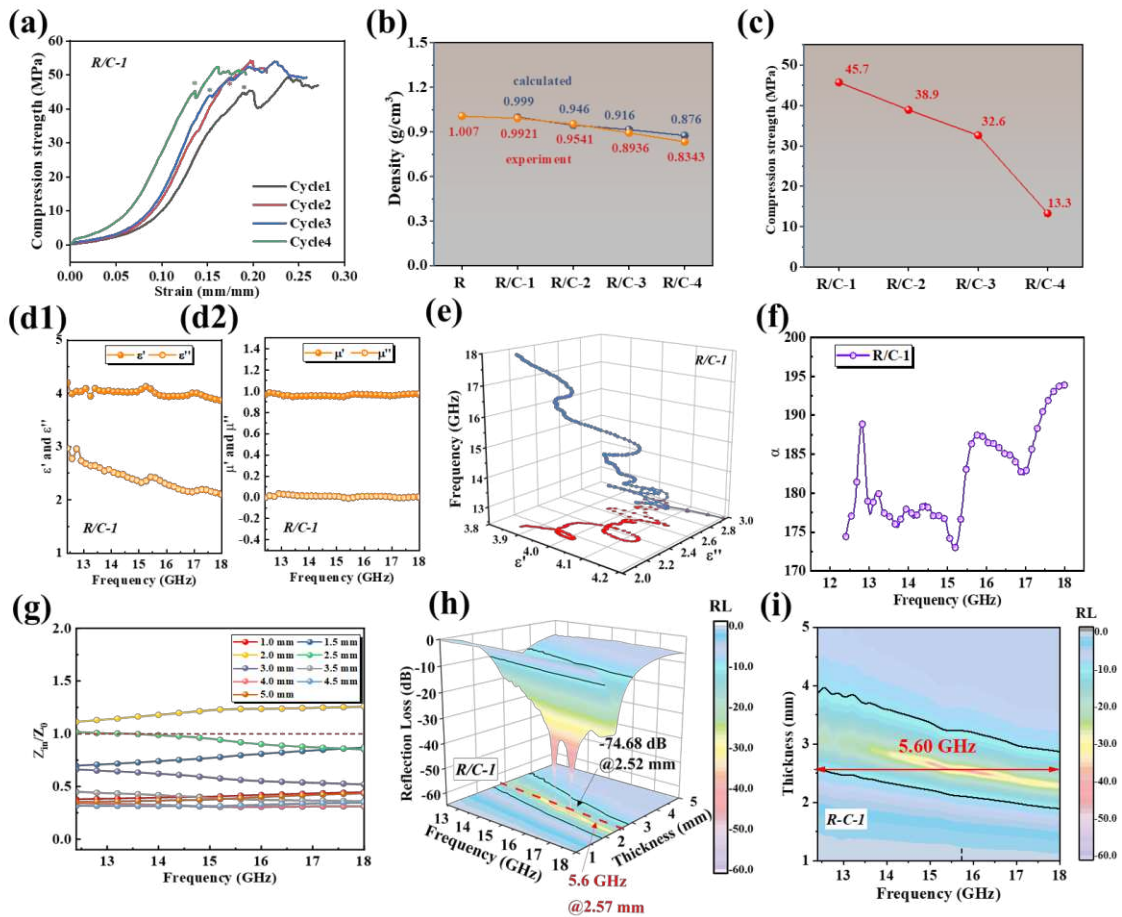


Figure.8 (a) Compression performance testing curves of R/C-1. (b) Calculated and experimental R/C-x density variation curves. (c) R/C-x compressive strength variation curve. (d₁) and (d₂) Permittivity and magnetic permeability of R/C-1. (e) Cole-cole

curve. (f) The 3D RL diagrams of R/C-1. (g) The impedance matching. (h) The attenuation constant. (i) The 2D RL map of R/C-1.

4. Conclusion

In summary, we realized a structurally programmed composite aerogel (CNGA) that resolves the intrinsic physical contradiction between macro-impedance mismatch and optimal internal energy dissipation. Utilizing an ice-templated freeze-induced phase separation paradigm, we physically governed the spatial isolation of core-shell DSHMs—preventing contiguous conductive aggregation—while systematically controlling the interrelated connective strength of the N-CNS bridging networks via carbonization thermodynamics. This precise morphological decoupling mechanism enabled a high proportion of metallic spherical attenuation units to operate optimally at an ultra-low physical volume fraction (0.26 vol%), thereby constructing a geometrically balanced equivalent RLC network. Consequently, the optimal architectural composite (CNGA675-1) effectively leverages synergistic dielectric and magnetic loss mechanisms, delivering an exceptional effective absorption bandwidth of 6.56 GHz, a robust minimum reflection loss of -54.86 dB at an ultralow 8 wt% filler loading, alongside simultaneously achieving deep thermal insulation characterized by an ultra-low thermal conductivity of $0.0605 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ and excellent high-temperature infrared shielding. Finite element modeling visibly substantiates that spatially isolating the distribution of specialized loss units fundamentally dictates the trajectory of macroscopic energy dissipation, visually validating the internal operational dynamics of the macroscopic RLC loop model. Additionally, coupling the functional networks via vacuum impregnation enables robust state transfer; the resin-integrated triple-phase composite yields remarkable load-bearing robustness (45.70 MPa) without compromising density limits ($0.999 \text{ g} \cdot \text{cm}^{-3}$) or sacrificing EM broadband properties. This physically decoupled phase-topology architecture delivers highly generalizable guidelines for programming extreme-environment multi-functional stealth composites.

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