

Strain-induced defects suppress conductivity to enhance microwave absorption in high-entropy hollow microspheres

Lili Li^{1,2}, Pingan Chen^{1,2}(✉), Yingli Zhu^{1,2}, Fu Chen^{1,2}, Mengke Qiao^{1,2}, Jiang Wu^{1,2}, Gangtao Luo^{1,2}, Xiangcheng Li^{1,2}(✉)

¹ State Key Laboratory of Advanced Refractories, Wuhan University of Science and Technology, Wuhan 430081, China

² Key Laboratory of High Temperature Electromagnetic Materials and Structure of MOE, Wuhan University of Science and Technology, Wuhan 430081, China

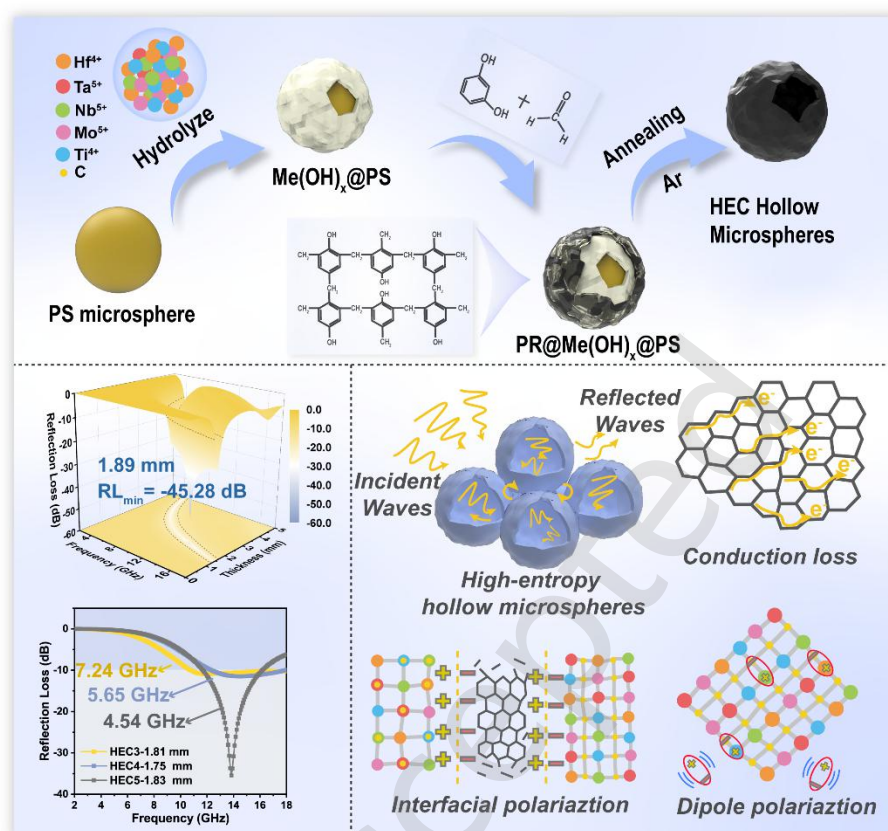
Nano Res., **Just Accepted Manuscript** • <https://doi.org/10.26599/NR.2026.94909033>

<https://www.sciopen.com/journal/1998-0124> on Jul. 16, 2026

© The Authors(s)

Just Accepted

This is a “Just Accepted” manuscript, which has been examined by the peer-review process and has been accepted for publication. A “Just Accepted” manuscript is published online shortly after its acceptance, which is prior to technical editing and formatting and author proofing. Tsinghua University Press (TUP) provides “Just Accepted” as an optional and free service which allows authors to make their results available to the research community as soon as possible after acceptance. After a manuscript has been technically edited and formatted, and the page proofs have been corrected, it will be removed from the “Just Accepted” web site and published officially with volume and article number (e.g., *Nano Research*, **2025**, *18*, 94906990). Please note that technical editing may introduce minor changes to the manuscript text and/or graphics which may affect the content, and all legal disclaimers that apply to the journal pertain. In no event shall TUP be held responsible for errors or consequences arising from the use of any information contained in these “Just Accepted” manuscripts. To cite this manuscript please use its Digital Object Identifier (DOI®), which is identical for all formats of publication.



Strain-induced defects in high-entropy carbide/carbon hollow spheres effectively suppress excessive conductivity, triggering a transition from conduction loss to polarization relaxation. This entropy-driven modulation enables an outstanding microwave absorption bandwidth of 7.24 GHz at a thin thickness of 1.81 mm.

Strain-induced defects suppress conductivity to enhance microwave absorption in high-entropy hollow microspheres

Lili Li ^{1,2}, Pingan Chen ^{1,2} , Yingli Zhu ^{1,2}, Fu Chen ^{1,2}, Mengke Qiao ^{1,2}, Jiang Wu ^{1,2}, Gangtao Luo ^{1,2}, Xiangcheng Li ^{1,2} 

¹ State Key Laboratory of Advanced Refractories, Wuhan University of Science and Technology, Wuhan 430081, China

² Key Laboratory of High Temperature Electromagnetic Materials and Structure of MOE, Wuhan University of Science and Technology, Wuhan 430081, China

Received: 15 May 2026; **Revised:** 13 July 2026; **Accepted:** 16 July 2026

 Address correspondence to Pingan Chen, pinganchen@wust.edu.cn; Xiangcheng Li, lixiangcheng@wust.edu.cn

 **Cite this article:** *Nano Research*, 2026, 19, 94909033 <https://doi.org/10.26599/NR.2026.94909033>

ABSTRACT: Carbon-based materials are extensively utilized for electromagnetic wave (EMW) absorption due to their low density and high stability. However, their absorption capacity is often hindered by impedance mismatch originating from high intrinsic conductivity. Here, we synthesize high-entropy carbide nanoparticles with in-situ growth on hollow carbon spheres (HECs@HCSs) to form core-shell composites via entropy engineering. The results show that the high configurational entropy in the carbides leads to an expansion of the HEC lattice constant from 4.309 Å to 4.401 Å, yielding a great lattice strain from 3.55% to 13.45%. The atomic-scale strain field promotes defect formation and suppresses the long-range ordering of the carbon matrix, leading to the disruption of the conductive carbon network. Together with enhanced carrier scattering and interfacial transport barriers, the electrical conductivity decreases from 20.46 S/m to 0.27 S/m. As a result, the HECs@HCSs composites exhibit outstanding microwave absorption with an effective absorption bandwidth of 7.24 GHz at 1.81 mm and a minimum reflection loss of -45.28 dB at 1.89 mm. Furthermore, radar cross-section simulations demonstrate that the HEC5 coating reduces the monostatic RCS by 18.85 dB·m² relative to the bare PEC plate at normal incidence, confirming its excellent radar stealth capability. The superior EMW absorption properties are attributed to the enhanced impedance matching and the mechanistic transition from conduction loss to polarization relaxation. This work provides novel design insights and theoretical criteria for developing lightweight, wideband, and high-efficiency EMW absorbing protection materials.

KEYWORDS: high-entropy carbides; hollow spheres; lattice distortion; electromagnetic wave absorption; impedance matching

1 Introduction

Electromagnetic wave (EMW) absorbing materials are essential for mitigating electromagnetic pollution to protect human health and equipment operation [1-3]. Among various candidates, carbon-based absorbers have been extensively utilized due to their low density and structural diversity. However, the high conductivity of traditional carbon materials induces severe impedance mismatch, which aggravates surface reflection rather than internal dissipation of the incident waves [4-7]. To circumvent this limitation, researchers have employed methods to reduce the real part of the dielectric constant for carbon materials, such as heterostructure construction, defect engineering, and heteroatom doping [8-10]. In particular, the suppression of long-range ordering in the carbon matrix via amorphous domains or structural defects has emerged as a critical approach [11-14]. This method leverages interfacial and dipole polarizations to increase energy loss while simultaneously optimizing impedance matching, which is key to achieving high-performance EMW absorption.

The geometric curvature of hollow carbon spheres (HCSs) could effectively inhibit graphitization by inducing local strain, leading to a defect-rich amorphous structure. These microscopic defects, accompanied by the heterogeneous interfaces, serve as active polarization sites to dissipate energy through polarization relaxation under an alternating electromagnetic field [15-18].

For example, Wang et al. [19] found that the curvature-induced lattice mismatch and interfacial stress could generate high-density defects while suppressing graphitization in hollow FeCo@C heterostructures, which yields a minimum reflection loss (RL_{min}) of -50.57 dB and a 5.35 GHz effective absorption bandwidth (EAB) at 1.8 mm. Similarly, Sun et al. [20] fabricated Pb/N-doped graphene hollow spheres, where curvature-induced lattice distortions and carbon vacancies were used to enhance dielectric and conduction losses. At 10 wt% loading, this defect-driven polarization enabled a -47.2 dB RL_{min} and a 4.64 GHz EAB at 1.53 mm. Zhang et al. [21] revealed that the synergy between curvature and P-doping promoted sp³ hybridization and structural defects in P-doped hollow porous carbon spheres, resulting in an RL_{min} of -34.05 dB at 1.8 mm. These studies demonstrate that hollow curved designs could regulate the defect states of the carbon matrix to optimize dielectric polarization and impedance matching. Furthermore, the incorporation of magnetic, dielectric, or atomic-scale components has also proven essential to enhance interfacial polarization and broaden the absorption bandwidth [22,23]. However, conventional doping methods often fail to induce substantial lattice strain across a large scale, limiting their ability to control the crystallinity and conductivity of the carbon matrix [24-26]. Therefore, it is imperative to explore a novel way to generate a synergistic strain effect with the hollow architecture, achieving precise structural modulation and superior microwave

absorption.

High-entropy materials (HEMs) could produce severe lattice distortions to drive the structural evolution and dielectric modulation of the carbon matrix [27,28]. For instance, Gang et al. [29] revealed that lattice distortions and nanotwins induced by HE Ni-Mn-Co-Cu-Al alloys drive abundant vacancies and defects within the carbon shell. This multiscale defect boosts polarization loss, enabling a strong reflection loss of -52.36 dB and a broad bandwidth of 5.01 GHz at a thickness of 1.9 mm. Similarly, Hassan et al. [30] illustrated that lattice distortion suppressed the long-range ordering of the carbon shell and promoted sp^3 defect formation in carbon-coated FeCoNiCuMn_x particles, which enhanced dipole polarization while reducing conductivity, thereby optimizing impedance matching and yielding an EAB of 5.52 GHz at 2.0 mm. Liu et al. [31] reported that the lattice distortion and chemical disorder in FeCoNi/carbon heterostructures could generate high-density hetero-interfaces and defects, leading to an RL_{min} of -57.5 dB and an EAB of 5.67 GHz at 2.2 mm. These studies reveal that the lattice distortion and chemical disorder in HEMs disrupt the periodic arrangement of carbon atoms. Thus, it induces multiple polarization relaxations and enables macroscopic control over complex permittivity and impedance matching. Furthermore, the high-entropy structural disorder intensifies carrier scattering to reduce the intrinsic conductivity [32,33]. As a result, the synergy effect between high-entropy features and hollow architectures could yield outstanding EMW absorption performance in the composites.

Thus, the regulation of carbon matrix defects via hollow core-shell structures and HE-induced lattice distortion provides an effective strategy for tailoring the microwave absorption performance of composites. This study synthesizes a series of composition- tunable high- entropy carbide nanoparticles (HECs), which grow in situ on hollow carbon spheres (HCSs), forming composite powders with a core- shell heterostructure (HECs@HCSs). The results show that a high configurational entropy in the carbides leads to an expansion of the HEC lattice constant and great lattice strain. This atomic-scale strain field disturbs the symmetry of atomic arrangements in the hollow carbon shell, introducing a high density of vacancy defects and restraining the development of graphitized ordered structures. Consequently, the electrical conductivity drastically drops from 20.46 S/m to 0.27 S/m, creating a transition of the dissipation mechanism from conduction loss to polarization relaxation. The synergy between entropy-induced polarization enhancement and impedance optimization of the hollow architecture enables HECs@HCSs composite to achieve an EAB of 7.24 GHz at 1.81 mm and an RL_{min} of -45.28 dB at 1.89 mm. Radar cross-section (RCS) simulation results further confirm the superior stealth potential of HECs@HCSs composites across the Ku-band. This work provides a valuable reference for the design and fabrication of lightweight, wideband, and high-efficiency high-entropy EMW absorbing materials.

2 Experimental

2.1 Cell fabrication

Fabrication of Polystyrene (PS) microspheres: styrene (St, inhibitor-removed) and polyvinylpyrrolidone (PVP) (5:1 w/w) were dissolved in 300 mL of anhydrous ethanol within a three-neck round-bottom flask under mechanical stirring (500 rpm).

The solution was deoxygenated by purging with Ar for 30 min and subsequently maintained under an N₂ atmosphere. The mixture was then heated to 70°C in an oil bath, followed by the addition of azobisisobutyronitrile (AIBN, 0.8 g) to initiate the polymerization. After reacting at 70°C for 24 h, monodisperse polystyrene (PS) microspheres were successfully obtained.

Fabrication of Me(OH)_x@PS microsphere: Polystyrene (PS) microspheres (0.4 g) were dispersed in 150 mL of anhydrous ethanol within a three-neck flask via 10 min of ultrasonication, followed by mechanical stirring at 500 rpm. Subsequently, 1.5 mL of NH₄OH (28%) was injected into the suspension. Separately, stoichiometric amounts of TBOT, MoCl₅, NbCl₅, TaCl₅, and HfCl₅ (molar ratios detailed in Table S1) were dissolved in 6 mL of anhydrous acetonitrile to form a precursor solution. This solution was then introduced dropwise into the PS suspension at a controlled rate of 0.5 mL/min under continuous stirring. After reacting at room temperature for 4 h, the resulting product was collected by centrifugation (2000 rpm, 3 min), washed three times with ethanol, and vacuum-dried at 60°C for 12 h to obtain a free-flowing powder.

Fabrication of PR@Me(OH)_x@PS microsphere: 1 g of dried Me(OH)_x@PS powder was dispersed in a mixture of anhydrous ethanol and deionized water (300 mL) via ultrasonication. Under continuous mechanical stirring, 0.6 mL of formaldehyde solution was added and stirred for 10 min, followed by the introduction of 0.35 g of resorcinol. After another 20 min of stirring, 0.85 mL of ammonium hydroxide was added to catalyze the polymerization. The reaction lasted for 24 h to form the PR@Me(OH)_x@PS precursor. The product was collected via centrifugation, washed alternately with deionized water and ethanol for four cycles, and vacuum-dried. Finally, the obtained powders were carbonized at 1500°C for 2 h under an Ar atmosphere, yielding the HECs@HCSs composites.

2.2 Materials characterizations

The phase constitutions of the HECs@HCSs composites were identified by X-ray diffraction (XRD, XPert PRO MPD, Netherlands) and further analyzed via Rietveld refinement using GSAS II software. The microstructure was characterized using field emission scanning electron microscopy (FESEM, Nano SEM 400, America) and transmission electron microscopy (TEM, JROL JEM-F200, Japan). X-ray photoelectron spectroscopy (XPS, AXIS SUPRA+, Japan) was employed to investigate the chemical states of the constituent elements, while the vibrational modes of chemical bonds were captured by Raman spectroscopy (Horiba LabRAM HR Evolution, Japan). For electromagnetic characterization, the composites were uniformly mixed with paraffin wax in a mass ratio of 15:85. The mixture was then pressed into a toroidal ring (inner diameter: 3 mm, outer diameter: 7 mm, thickness: 3 mm) for testing with a vector network analyzer (VNA, Keysight E5071c).

The lattice strain was evaluated using the Williamson–Hall (W–H) method based on the broadening of X-ray diffraction peaks. The full width at half maximum (FWHM) of the diffraction peaks was fitted according to the Williamson–Hall equation [34,35]:

$$\beta \cos \theta = \left(\frac{k\lambda}{D} \right) + (4\varepsilon \sin \theta) \quad (1)$$

Where β is the full width at half maximum (FWHM), θ is the Bragg diffraction angle, k is the Scherrer constant, λ is the X-ray wavelength, D is the crystallite size, and ε is the lattice strain. The lattice strain was determined from the slope of the linear fitting of $\beta \cos \theta$ versus $4 \sin \theta$.

The far-field simulations were performed using the CST Studio Suite. The simulation model consisted of a square Perfect Electric Conductor (PEC) plate (100×100×1 mm³) serving as the

reflective substrate. A coating layer with a thickness of 1.81 mm was modeled on the PEC surface, where the material properties were defined by the experimentally measured complex permittivity and permeability of the HECs@HCSs composites.

3 Results and discussion

The schematic fabrication process of the HECs@HCSs composites is illustrated in Figure 1a. Initially, the polystyrene (PS) microspheres are coated with a layer of metal hydroxides ($\text{Me}(\text{OH})_x$) derived from the hydrolysis of metal ions (Me^{x+} : Ti^{4+} , Mo^{5+} , Nb^{5+} , Ta^{5+} , Hf^{4+}) (Figures 1b, 1c, and S1). Subsequently, the $\text{Me}(\text{OH})_x$ @PS microspheres are encapsulated by a layer of phenolic resin (PR) through the condensation reaction of formaldehyde and resorcinol (Figure 1d), which serves as the carbon precursor. Finally, the $\text{PR@Me}(\text{OH})_x$ @PS microspheres are ultimately converted into hollow carbide structures via

thermal treatment, during which the PS templates are eliminated by pyrolysis as the PR and $\text{Me}(\text{OH})_x$ undergo a carbothermal reduction (Figure 1e). Furthermore, an entropy-increasing method is applied to the carbides by modulating the variety of metal ions. Based on the number of principal metallic elements, the samples are designated as HEC2, HEC3, HEC4, and HEC5, corresponding to $(\text{Ti}_{0.5}\text{Mo}_{0.5})\text{C}$, $(\text{Ti}_{1/3}\text{Mo}_{1/3}\text{Nb}_{1/3})\text{C}$, $(\text{Ti}_{0.25}\text{Mo}_{0.25}\text{Nb}_{0.25}\text{Ta}_{0.25})\text{C}$, and $(\text{Ti}_{0.2}\text{Mo}_{0.2}\text{Nb}_{0.2}\text{Ta}_{0.2}\text{Hf}_{0.2})\text{C}$, respectively. Comparative experiments (Figure S2) show that the $\text{Me}(\text{OH})_x$ @PS microspheres without the PR coating collapse into irregular micrometer-sized particles under identical thermal conditions, failing to retain a spherical morphology. This observation indicates that the PR plays a dual role by functioning as both the carbon source for the carbothermal reduction reaction and a structural scaffold that maintains the hollow architecture of the microspheres

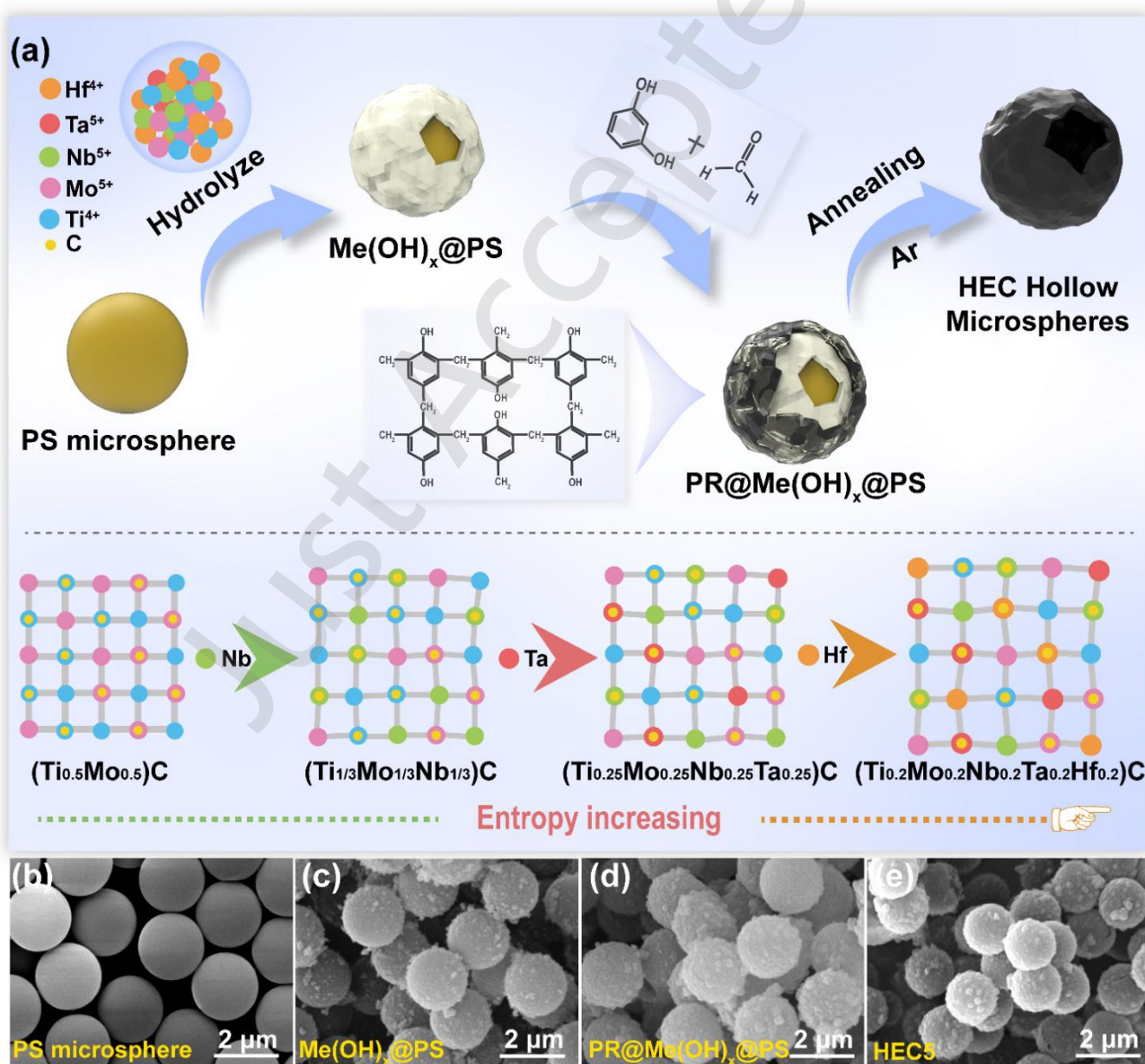


Figure 1 Design and preparation of HECs@HCSs composites. (a) Process flowchart and entropy increase design diagram; (b) SEM image of PS microspheres; (c) SEM image of $\text{Me}(\text{OH})_x$ @PS microspheres; (d) SEM image of $\text{PR@Me}(\text{OH})_x$ @PS microspheres; (e) SEM image of HECs@HCSs composites

X-ray diffraction (XRD) is employed to characterize the phase and crystalline structure of HECs@HCSs composites in Figure 2a. All Bragg diffraction peaks align with the standard pattern of face-centered cubic (FCC) TiC (ICDD No. 96-153-9506), indicating that the metallic atoms are solubilized into the TiC lattice, forming a stable, single-phase solid solution. From HEC2 to HEC5, all diffraction peaks shift toward low angles, which signifies an expansion of the lattice constant with increasing entropy. The diffraction peaks of the (111) lattice plane reveal that the full width at half maximum (FWHM) also increases alongside the peak shift, suggesting obvious lattice distortion in the crystal. Rietveld refinement results (Figure 2b, 2c, and S3) further confirm that the lattice constant increases sequentially with entropy. In detail, the values of the lattice constant rise from 4.309 ± 0.00012 Å (HEC2), 4.356 ± 0.00015 Å

(HEC3), 4.401 ± 0.00011 Å (HEC4) to 4.461 ± 0.00016 Å for HEC5. The low reliability factors compiled in Table S2 demonstrate the high quality and reliability of the refinement fitting. Driven by this lattice dilation, the derived lattice strain (ϵ) exhibits a synchronized upward trend from $3.55\% \pm 0.021\%$ in HEC2 to $13.45\% \pm 0.016\%$ in HEC5 (Figure 2d). The increment of lattice constants and lattice strain (Figure 2e) demonstrates that the carbide structure undergoes pronounced lattice distortion as metallic atoms incorporate into the solid solution [36,37]. This result arises from the lattice stress induced by disparities in atomic radii among Ti, Mo, Nb, Ta, and Hf. The occupation of FCC lattice sites by these diverse metallic atoms produces fluctuations in the crystal planes and results in the shift and broadening of the diffraction peaks.

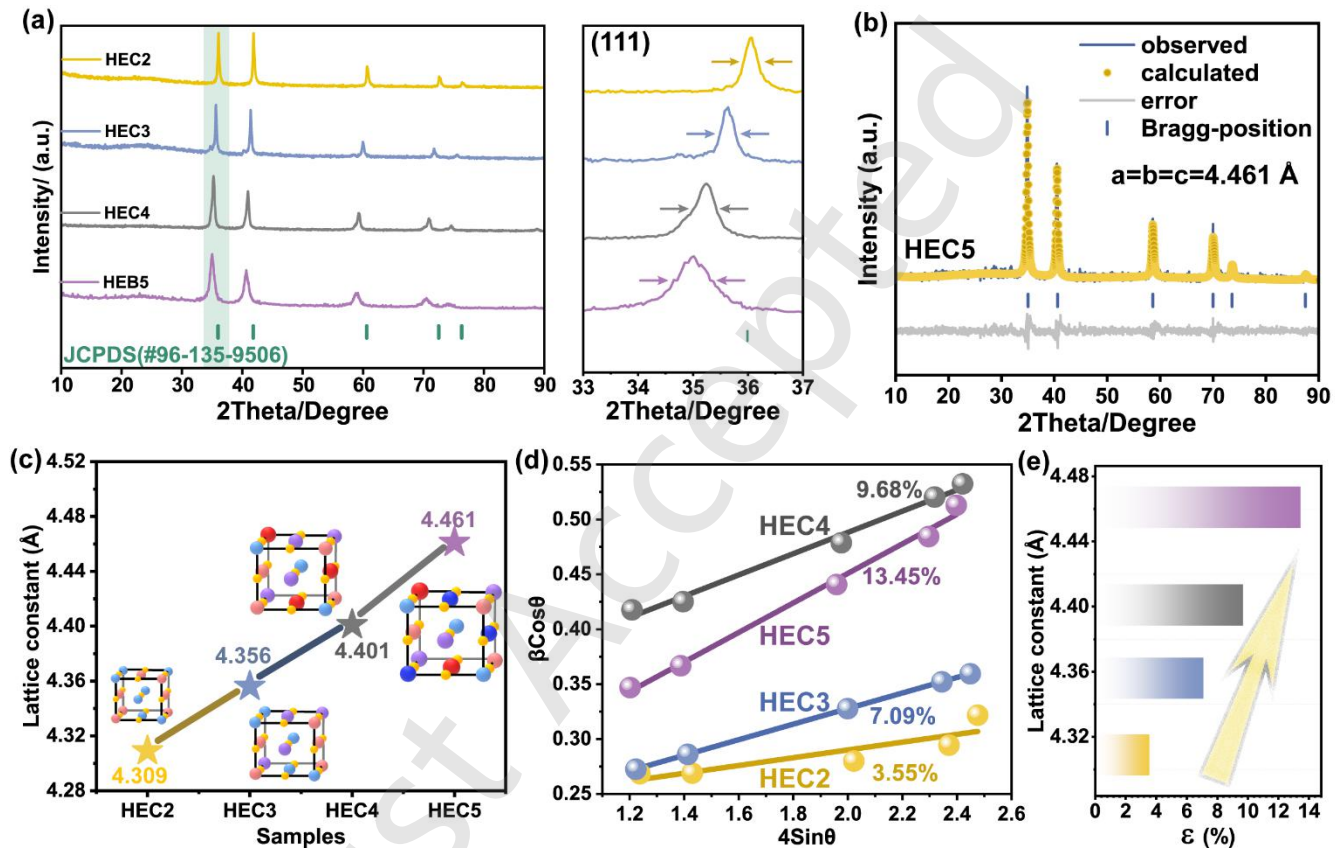


Figure 2 Physical phases and crystal structure of HECs@HCSs composites. (a) XRD; (b) XRD refined pattern of HEC5; (c) Lattice constants obtained from XRD refinement; (d) (e) The variation of lattice constant with ϵ

The X-ray photoelectron spectroscopy (XPS) survey spectra (Figures 3a-d and S4) reveal characteristic signals for all constituent metallic elements (Ti, Mo, Nb, Ta, and Hf) consistent with their compositions in HEC samples. These findings verify the formation of a homogeneous solid solution at the atomic scale in conjunction with the XRD results. In the Ti 2p, Ta 4f, and Hf 4f spectra, the peaks correspond to metal- carbon (Me-C) bonds, along with a minor contribution of metal-oxygen (Me-O) bonds. The Mo 3d spectra not only display the characteristic Mo-C peaks at 232.48 eV and 229.28 eV, but also reveal additional peaks attributed to Mo ions in valence states, located at 226.88/224.58 eV, 235.98/233.08 eV, and 229.98/227.18 eV. Meanwhile, as the entropy increases, the Nb 3d spectra evolve from a single characteristic peak to multiple peaks, and three groups of characteristic peaks appear in the HEC5 sample. The XPS spectra evolution of HEC can be attributed to the severe atomic size mismatch and inhomogeneous charge distribution within the high-entropy lattice [38,39]. Specifically, electronegativity disparities among

the diverse metallic atoms, coupled with lattice distortion, result in a rearrangement of the electron cloud, which leads to the coexistence of multiple valence states for Mo and Nb.

Figures 3e-h display the high-resolution C 1s XPS spectra of HECs@HCSs composites. The C 1s spectra are resolved into C=O, C-O, sp^3 , sp^2 -hybridized carbon, and the Me-C bond located between 283.68 and 283.98 eV. Simultaneously, Raman spectra (Figures 3i-l) are fitted into I, D, A, and G peaks located at approximately 1230, 1350, 1500, and 1590 cm^{-1} , respectively. Specifically, the G peaks arise from the in-plane stretching vibration of the E_{2g} in graphite, indicating long-range order within the carbon lattice. The D peaks result from phonon scattering at Brillouin zone boundaries due to structural defects or disorder. Furthermore, the I peaks are attributed to the stretching vibrations of C-C and C=C in sp^2 - sp^3 mixed bonds, while the A peaks stem from amorphous carbon, graphene fragments, and residual functional groups [40,41]. Quantitative results in Figure 3m and S5 indicate that the sp^3/sp^2 integrated area ratio rises significantly from 0.593 ± 0.021 for HEC2 to

1.597 ± 0.021 for HEC5, as systematically summarized in Table S3.. The ratios of I/D, I/G, and I/A in the Raman spectra also exhibit a monotonic upward trend with increasing entropy. These variations indicate that the entropy increment enhances the degree of disorder and the proportion of sp^3 bonding within the carbon matrix, which provides a high density of defects as

polarization centers to attenuate the EMW [42]. Furthermore, the interfaces between the disordered carbon matrix and the high-entropy ceramic phase are expected to broaden the effective absorption bandwidth through the enhancement of both interfacial and dipole polarization.

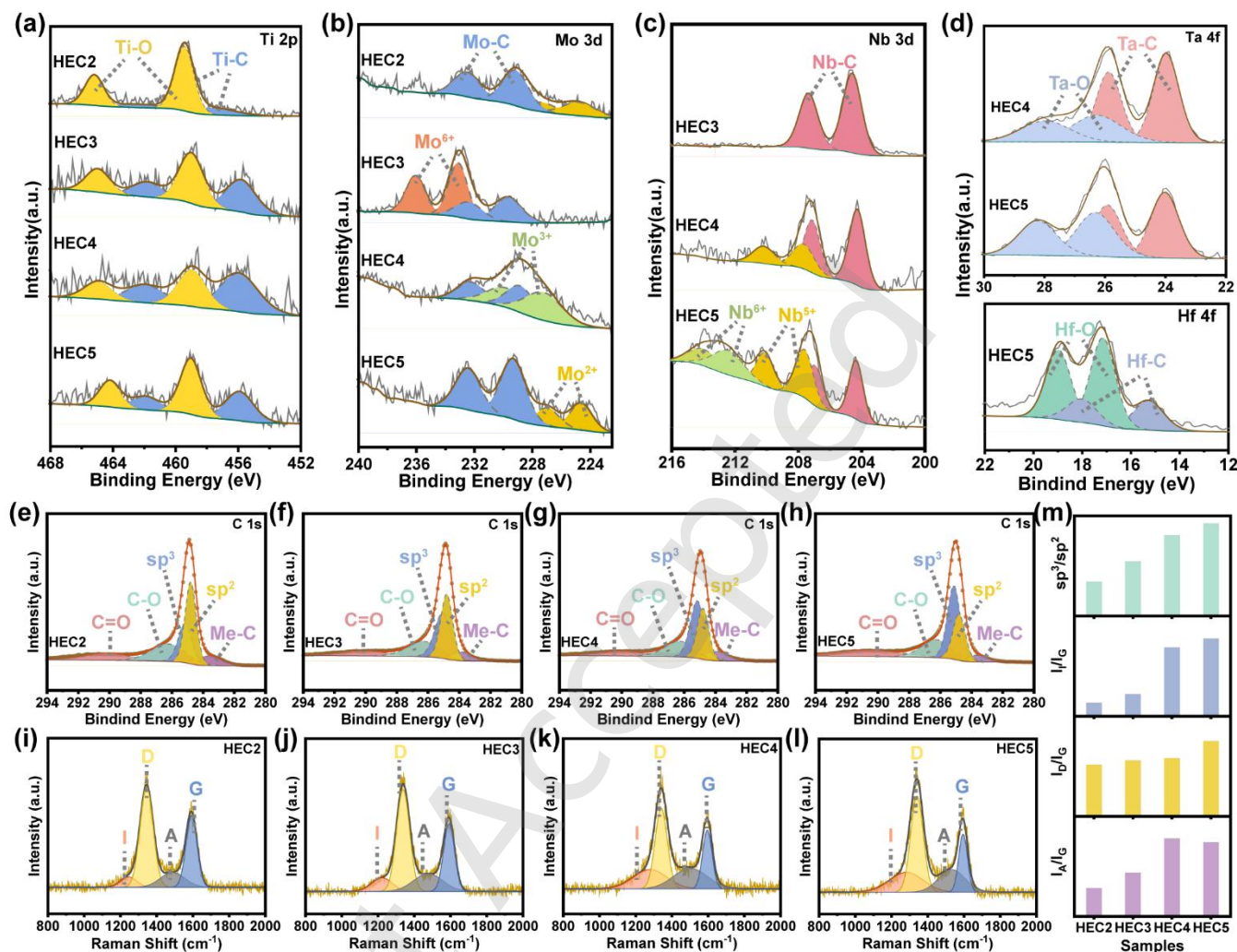


Figure 3 Chemical bond characterization of HECs@HCSs composites. (a) XPS fine spectra of Ti 2p; (b) XPS fine spectra of Mo 3d; (c) XPS fine spectra of Nb 3d; (d) XPS fine spectra of Ta 4f and Hf 4f; (e-h) XPS fine spectra of C 1s; (i-l) Raman spectra; (m) sp^3/sp^2 , I_A/I_G , I_D/I_G , and I_D/I_A

The high-resolution transmission electron microscopy (HRTEM) exhibits a monodispersed hollow spherical structure with an average diameter of 1.8 μm , as illustrated in Figure S6 and Figure 4a. Energy-dispersive X-ray spectroscopy (EDS) elemental mapping of a single sphere (Figures 4b and S7) demonstrates that all constituent elements are uniformly distributed throughout the hollow shell. HRTEM images (Figures 4c-f) reveal that the hollow shell consists of an amorphous carbon matrix embedded with carbide nanoparticles (10–100 nm), and the particle size decreases gradually as the entropy rises. The corresponding selected area electron diffraction (SAED) patterns display clearly polycrystalline diffraction rings, and the lattice space values are consistent with the XRD results. Interestingly, the diffraction ring (002) of graphitic carbon is visible in the SAED patterns of HEC2 and HEC3, suggesting a high crystalline degree of the carbon matrix in the low-entropy components. The spacing of the (111) planes widens from 0.254 nm in HEC2 to 0.261 nm in HEC5 as metallic atoms are introduced (Figures 4g-i), confirming the lattice distortion during the formation of the high-entropy solid solution, which is consistent with the Rietveld refinement results.

Figures 4k-n illustrate atomic-scale structures of the HECs@HCSs composites. In HEC2, the carbide particles are surrounded by highly ordered, ribbon-like graphite layers, and HEC3 maintains a partially graphitized structure. In contrast, the carbon matrix in HEC4 and HEC5 transforms into a turbostratic graphite with a highly distorted, vortex-like distribution. This structural evolution can be understood from the perspective of interfacial misfit strain relaxation. During high-temperature carbonization and subsequent cooling, the thermal expansion mismatch between carbide nanoparticles and the carbon matrix, together with the intrinsic lattice strain induced by the high-entropy effect, generates significant residual interfacial stresses at the carbide/carbon boundaries [43,44]. The accumulated interfacial stress can be transferred into the adjacent carbon layers, inducing distortion, bending, and partial fragmentation of the graphitic frameworks [45,46]. To partially release the localized strain energy, the distorted carbon networks may undergo local bonding reconstruction, resulting in an increased fraction of tetrahedral sp^3 configurations. This strain-induced carbon structural evolution is consistent with the XPS results, where the sp^3/sp^2 ratio increases from 0.593 ± 0.021 to $1.597 \pm$

0.021 with increasing configurational entropy. [47]. The hierarchical heterostructure, comprising high-entropy nanocrystals, turbostratic graphite, and amorphous carbon matrix, introduces a wealth of phase interfaces and structural

defects. Such a configuration not only induces strong interfacial polarization via the Maxwell-Wagner effect but also extends the propagation path of EMWs within the hollow shells through scattering and multiple reflections.

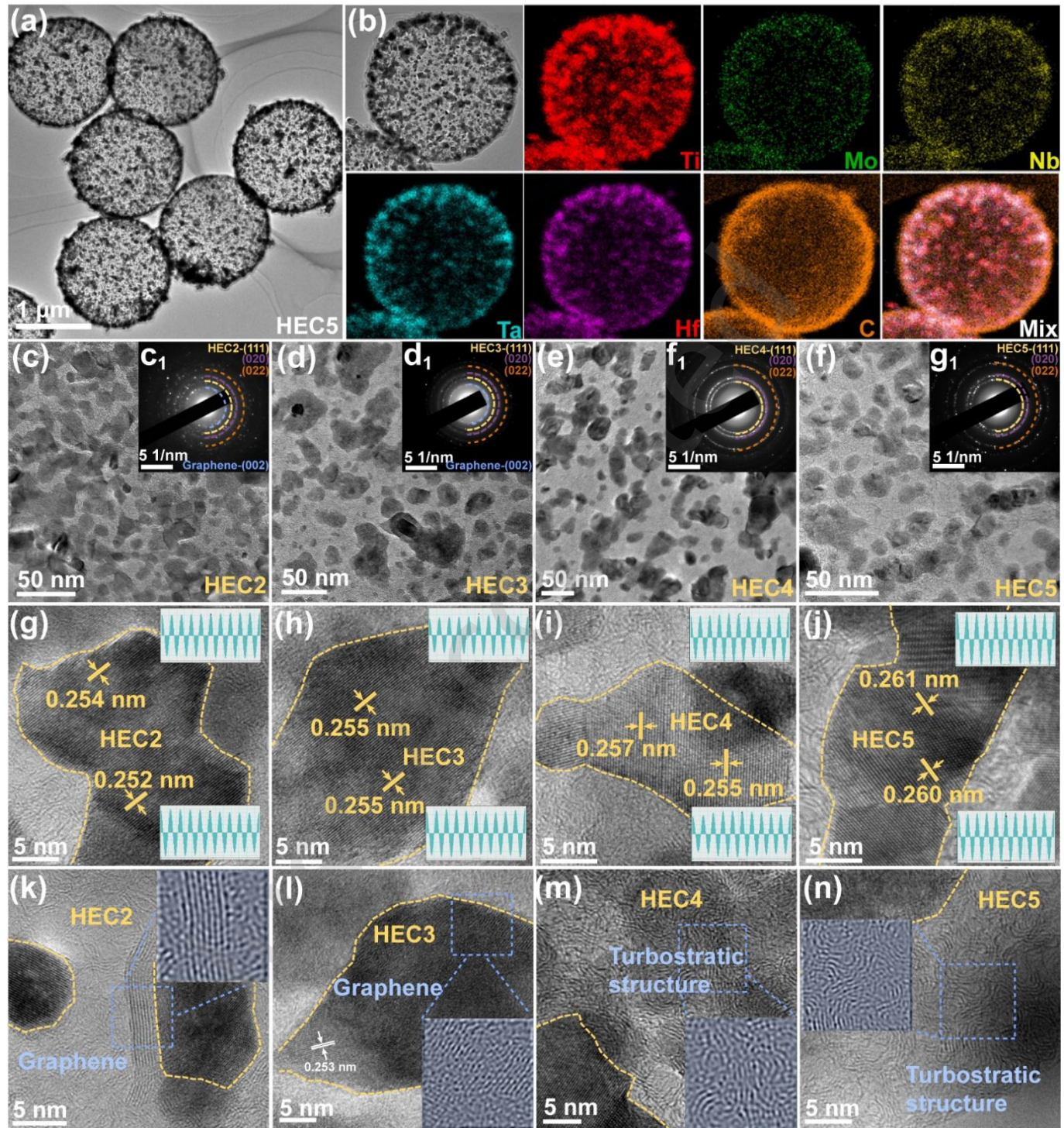


Figure 4 Microstructure of HECs@HCSs composites. (a) TEM image of HEC5; (b) TEM image and EDS area scan of a single HEC5 hollow sphere; (c-f) TEM images of carbide particle distribution; (g-j) High-resolution TEM images of carbide particles; (k-n) High-resolution TEM images of the carbon matrix

The real (ϵ') and imaginary (ϵ'') parts of the complex permittivity for HECs@HCSs composites are measured and illustrated in Figures 5a and 5b. The real permittivity (ϵ') of each sample declines as frequency rises, showing characteristic frequency dispersion. Meanwhile, the ϵ'' curves display fluctuating characteristics, which signify the presence of multiple polarization relaxation processes in the composites. Specifically, HEC2 exhibits high ϵ' and ϵ'' values of approximately 46.1 and 186.7 in the low-frequency region,

respectively. With increasing entropy, the permittivity of the composites decreases significantly, with the ϵ' and ϵ'' of HEC5 dropping to approximately 14.1 and 4.7, respectively. Figure 5c illustrates the frequency dependence of the dielectric loss tangent ($\tan\delta$) for the HEC2 to HEC5 within the 2-18 GHz range. HEC2 exhibits the most robust dielectric dissipation, maintaining an average $\tan\delta$ value of approximately 2.31. As the entropy increases, the average $\tan\delta$ values for HEC3 and HEC4 decrease to 0.61 and 0.58, respectively, while HEC5 further

declines to 0.39.

The attenuation constant (α) is calculated to evaluate the absorbing capacity of the HECs@HCSs composites. As shown in Figure 5d, the α values increase with frequency across the 2–18 GHz. Specifically, HEC2 exhibits the highest attenuation constant, rising from 358.18 to 1017.05, which demonstrates a powerful EMW loss capability. For HEC3, the α stays between 250 and 305 across the Ku-band. HEC4 reaches 382.43 at 18 GHz. In contrast, HEC5 exhibits the lowest α values in the composites.

To investigate the dielectric loss mechanisms of the HECs@HCSs composites, the Cole-Cole plots are analyzed in Figures 4e–h. The plot for HEC2 exhibits a nearly linear upward trajectory within an exceptionally high dielectric range, with ϵ' values significantly surpassing those of ϵ'' , indicating that conduction loss dominates the HEC2; the long-range migration of charge carriers effectively masks localized polarization relaxation [48,49]. With increasing entropy, the plots shift toward low dielectric values. Specifically, HEC3 displays multiple overlapping and distorted semicircles alongside a residual tail of conduction loss. In contrast, the curves of HEC4 and HEC5 maintain arc-like features without a linear rise at the higher frequency end. The Cole-Cole plots confirm that the relative contribution of polarization loss to the total dissipation increases with the entropy rising.

The conductivity of HECs@HCSs composites is measured to reveal the regulatory mechanisms of the microstructure on charge transport, as shown in Figure 5i. The conductivity decreases continuously by nearly two orders of magnitude with increasing configurational entropy, dropping from 20.46 S/m for HEC2 to only 0.27 S/m for HEC5. The relatively high conductivity of HEC2 originates from its well-developed graphitic carbon framework, which provides continuous sp^2 -conjugated pathways for electron transport. As the configurational entropy increases, however, HRTEM observations reveal that the graphitic layers gradually evolve into fragmented turbostratic carbon, while Raman and XPS analyses indicate a continuous increase in the sp^3/sp^2 ratio, indicating suppressed graphitization and the progressive disruption of the conductive carbon network [50,51]. Thus, electron transport through the conductive carbon framework becomes increasingly hindered, resulting in higher contact resistance and deteriorated long-range charge transport [52,53]. Importantly, the conductivity suppression cannot be solely attributed to the disordering of the carbon matrix. The severe lattice distortion associated with the high-entropy carbide nanoparticles generates local strain fields and chemical disorder, which enhance carrier scattering and reduce carrier mobility. At

the same time, the increasing number of carbide and carbon heterointerfaces introduces additional potential barriers for interfacial charge transfer, further impeding electron migration throughout the conductive network. Therefore, the significant reduction in electrical conductivity results from the synergistic effects of conductive-network disruption, lattice-distortion-induced carrier scattering, and interfacial transport barriers.

To further explore the electromagnetic energy dissipation mechanisms of the HECs@HCSs composites, the ϵ'' is decomposed into conduction loss (ϵ_c'') and polarization relaxation loss (ϵ_p''). Figure 5j illustrates that the ϵ_c'' values decrease as frequency increases, which is consistent with the physical law derived from Maxwell equations, and the conduction loss is inversely proportional to frequency [54]. HEC2 exhibits the most significant conduction loss. In accordance with the entropy increase, ϵ_c'' gradually declines, and the HEC5 reaches a minimum of approximately 0.27 in the high-frequency band, confirming that the continuous conductive network in HEC2 facilitates efficient energy dissipation via free charge migration. In contrast, the disruption of conductive pathways in the high-entropy components leads to a substantial attenuation of conduction loss.

The contribution of polarization relaxation to the overall energy dissipation is reflected in the ϵ_p'' (Figure 5k). HEC2 exhibits a prominent relaxation peak at approximately 3.28 GHz, indicating a robust dipole or interfacial polarization response in the low-frequency region. As the entropy increases, the relaxation loss for HEC3, HEC4, and HEC5 remains weak at low frequencies, then gradually rises and stabilizes, with peaks identified near 6.76, 5.68, and 4.27 GHz, respectively. A comparison of the two loss ways shows that conduction loss contributes less to overall attenuation as frequency rises [55]. In particular, for HEC3 to HEC5, energy dissipation above 8 GHz shifts toward a regime governed mainly by relaxation loss, confirming that the polarization relaxation serves as the dominant process for EMW attenuation in these high-entropy materials.

The relaxation time (τ) is calculated to further investigate the dipole orientation and interfacial polarization kinetics of the HECs@HCSs composites. As shown in Figure 5l, HEC2 shows the longest relaxation τ value of 4.53×10^{-10} s, while HEC3 shortens to 1.33×10^{-10} s. As the entropy continually increases, the τ values for HEC4 and HEC5 rise again to 1.4×10^{-10} s and 1.89×10^{-10} s. This originates from the reduction in charge transport resistance due to carbon matrix fragmentation and the increased rotational barriers for dipoles induced by severe lattice distortion.

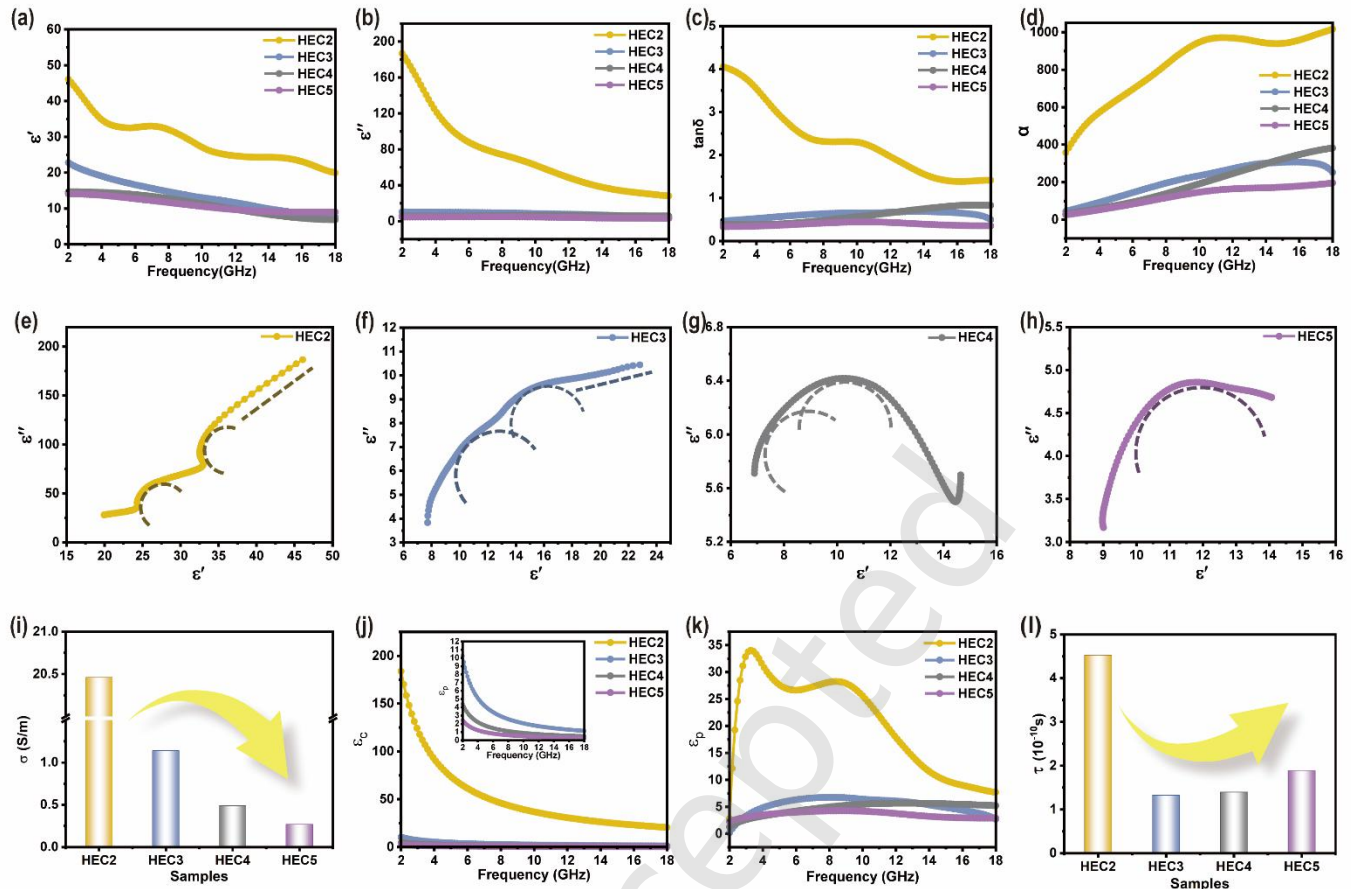


Figure 5 EM parameters of HECs@HCSs composites. (a) ϵ' ; (b) ϵ'' ; (c) $\tan\delta$; (d) α ; (e-h) Cole-Cole; (i) Electrical conductivity; (j) ϵ'' ; (k) ϵ'' ; (l) τ

The reflection loss (RL) of the HECs@HCSs composites is calculated within the 2-18 GHz range based on transmission line theory. As illustrated in Figures 6a-d, the HEC2 exhibits an $RL_{\min}$ of -3.43 dB. The excessive conductivity leads to a severe impedance mismatch, preventing EMWs from entering the interior for dissipation. In contrast, HEC5 exhibits the most intense absorption peak, reaching as low as -45.28 dB at a thickness of 1.89 mm. The low conduction loss and moderate relaxation time of HEC5 create optimal impedance matching, allowing the maximum EMW into the composites and subsequent conversion into thermal energy via polarization relaxation mechanisms [56]. Meanwhile, HEC3 exhibits the most superior EAB in the composites. As shown in Figures 6e-f, the EAB of HEC3 reaches 7.24 GHz at a thickness of 1.81 mm. This disparity is attributed to their different relaxation kinetics, as HEC3 exhibits the shortest relaxation time to absorb EMWs over a broader frequency spectrum.

Figures 6i and 6j compare the RL and the EAB for HEC3, HEC4, and HEC5 across a thickness range of 0-5 mm. HEC5 exhibits super RL intensity at various thicknesses, highlighting its outstanding performance in electromagnetic energy dissipation. Simultaneously, the EAB coverage for three samples is concentrated in the Ku-band. These results indicate that through compositional regulation, the HECs@HCSs composites achieve a significant transition in electromagnetic characteristics, moving from strong reflection to high absorption and broadband coverage [57]. Specifically, HEC5 prioritizes breakthroughs in absorption intensity, while HEC3 achieves effective absorption over a broader frequency range, owing to its fast relaxation response and moderate conduction loss. Compared with the relevant reports on high-entropy materials, hollow structures, and carbon-based composites (Figures 6k, 6l, and Table S4), the HECs@HCSs composites achieve a markedly

low filler loading without compromising the wide effective absorption bandwidth. This advantage is attributed to the effective density control provided by the hollow structure, as well as the fine-tuning of the permittivity via the high-entropy effect and atomic size mismatch.

Figures 6m-p present 2D maps of the normalized impedance ($|Z_{in}/Z_0|$) as a function of thickness for the HECs@HCSs composites, revealing the capacity for EMW absorption. The $|Z_{in}/Z_0|$ values for HEC2 are below 0.5, signifying intense surface reflection that impedes the transmission of EMWs into the interior. While the $|Z_{in}/Z_0|$ values for HEC3 are generally below 0.8, the impedance matching characteristics improve significantly for the HEC4, which exhibits regions approaching the ideal $|Z_{in}/Z_0|=1$ within the 2-8 GHz range. The HEC5 manifests the most superior impedance matching, characterized by a markedly expanded area within the optimal $|Z_{in}/Z_0|$ range of 0.8-1.2, which is a prerequisite for high-performance microwave absorption [58]. By precisely tailoring the high-entropy phase, HEC5 achieves a significantly optimized conduction loss of 0.27 at 18 GHz. This dielectric loss ensures that a maximum volume of energy can penetrate the material rather than being reflected at the surface [59]. Consequently, HEC5 leverages the quarter-wavelength interference effect to the fullest extent, yielding an intensive reflection loss of -45.28 dB at 1.89 mm. This underscores the efficacy of high-entropy integration in achieving near-perfect impedance matching and high-intensity absorption in thin-layered absorbers.

It should be emphasized that the microwave absorption performance is not determined by impedance matching or attenuation capability individually, but by their synergistic balance. Although HEC2 exhibits relatively strong attenuation capability due to its larger dielectric loss contribution, the severe

impedance mismatch restricts the effective penetration of incident electromagnetic waves into the absorber. In contrast, HEC5 maintains sufficient attenuation capability while exhibiting the impedance matching coefficient closest to unity over the broadest frequency range. Consequently, a larger proportion of the incident electromagnetic waves can enter the absorber and be effectively dissipated through conduction and

polarization losses, resulting in the broadest effective absorption bandwidth and the strongest reflection loss. Therefore, the superior microwave absorption performance of HEC5 arises from the optimized balance between attenuation capability and impedance matching rather than the maximization of either electromagnetic parameter individually.

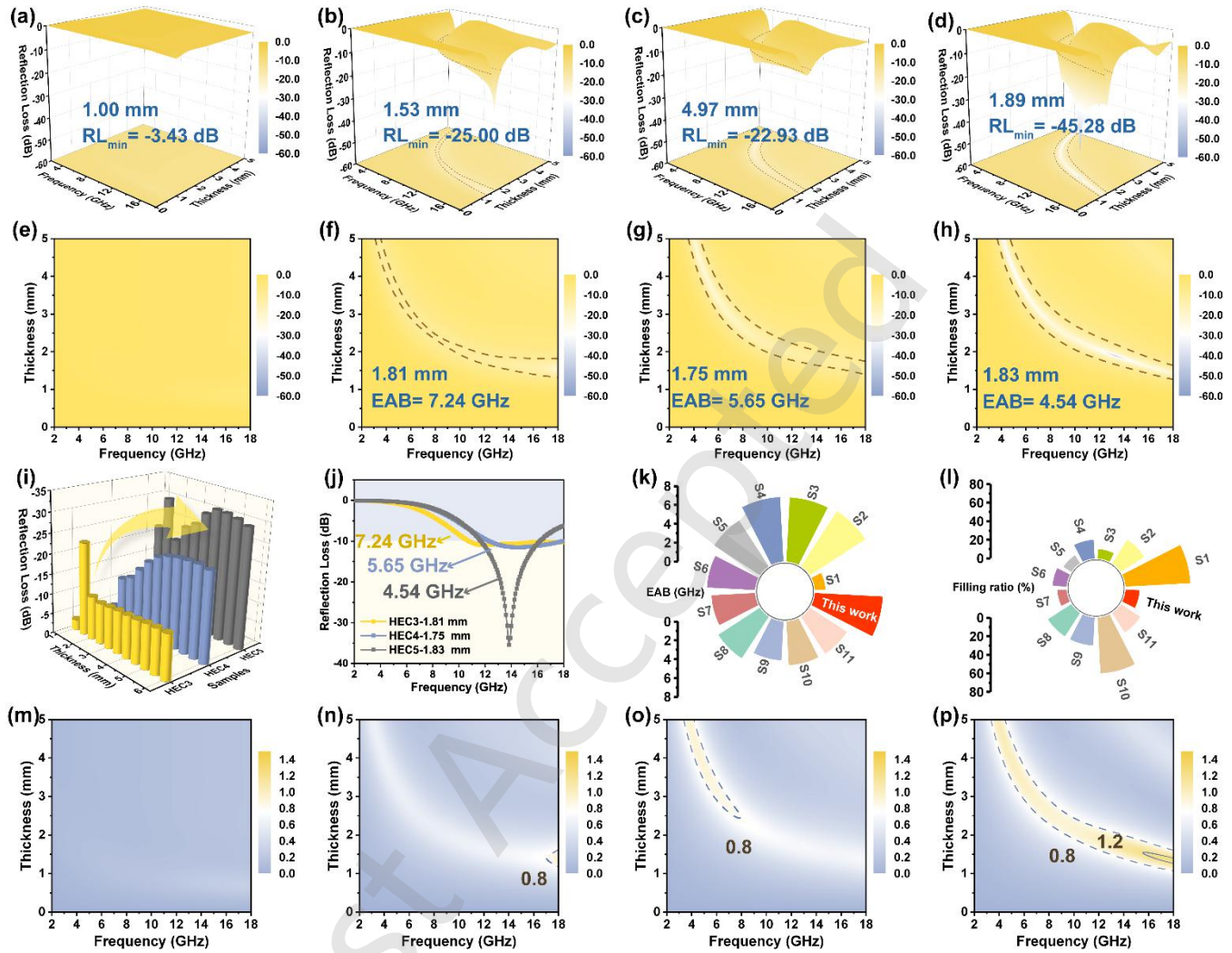


Figure 6 EMW absorption performance of HECs@HCSs composites. (a-d) 3D RL diagram; (e-h) 2D RL diagram; (i) RL comparison chart for HEC3-HEC5 at different thicknesses; (j) EAB comparison chart; (k, l) Comparison of EAB and filler ratios with previously reported materials of the same type; (m-p) 2D $|Z_{in}/Z_0|$ diagram

The practical application of the HECs@HCSs composites is performed with CST Studio Suite to evaluate the radar scattering signals in a far-field environment. A pure Perfect Electric Conductor (PEC) plate exhibits intense scattering signals at a specific frequency of 13.36 GHz in Figures 7a-f. This frequency is selected because it corresponds to the minimum reflection loss frequency of HEC5, where the strongest electromagnetic wave absorption is achieved. Therefore, the RCS simulation at this representative frequency provides a direct evaluation of the radar scattering suppression capability under the optimal absorption condition. The radar cross-section (RCS) signals are attenuated to varying degrees upon coating the PEC surface with the composite materials [60,61]. Notably, HEC5 demonstrates the most pronounced reduction in the RCS signal. Figure 7g-h presents the simulated monostatic RCS distributions of the bare PEC plate and the corresponding HEC2-HEC5 coated structures. Compared with the bare PEC plate, all HECs@HCSs coatings effectively

suppress radar backscattering. At normal incidence (0°), the RCS decreases from $13.98 \text{ dB}\cdot\text{m}^2$ for the bare PEC plate to 11.82 , 3.61 , 5.22 , and $-4.87 \text{ dB}\cdot\text{m}^2$ for HEC2, HEC3, HEC4, and HEC5, respectively, corresponding to RCS reductions of 2.16 , 10.37 , 8.76 , and $18.85 \text{ dB}\cdot\text{m}^2$. Although slight fluctuations are observed between the intermediate compositions, the HEC5 coating exhibits the most pronounced reduction in radar scattering. This trend agrees well with the microwave absorption results discussed above, indicating that entropy engineering effectively optimizes the balance between impedance matching and electromagnetic attenuation, thereby enabling more incident electromagnetic waves to enter the absorber and be dissipated before being reflected.

The exceptional EMW absorption performance of the HECs@HCSs composites originates from the synergy between the microscopic electromagnetic evolution driven by high-entropy components and the macroscopic wave-dynamic response of the hollow structure. The underlying physical

mechanisms are illustrated in Figure 7i. Initially, the hollow core-shell architecture facilitates multiple reflections and scattering of EMWs in the internal cavities, effectively extending the propagation path and optimizing macroscopic impedance matching [62]. At the atomic scale, the lattice distortion of HECs establishes energy barriers for electron hopping, which regulates long-range conduction while preserving the intrinsic dissipative capability of crystalline carbon domains, thereby addressing the impedance mismatch challenge [63]. Crucially, the distorted HECs lattice acts as a RCS reduction and wide-angle electromagnetic protection

prolific source of dipole polarization, creating a high density of dipoles that undergo ultra-fast relaxation under alternating electric fields. Simultaneously, the significant disparities in atomic radii and the presence of complex hetero-interfaces create a high density of dipole polarization centers, leading to an intense Maxwell-Wagner effect under alternating electric fields [64]. As a result, HECs not only ensure high-performance absorption under near-field conditions but also provide a robust foundation for far-field

applications.

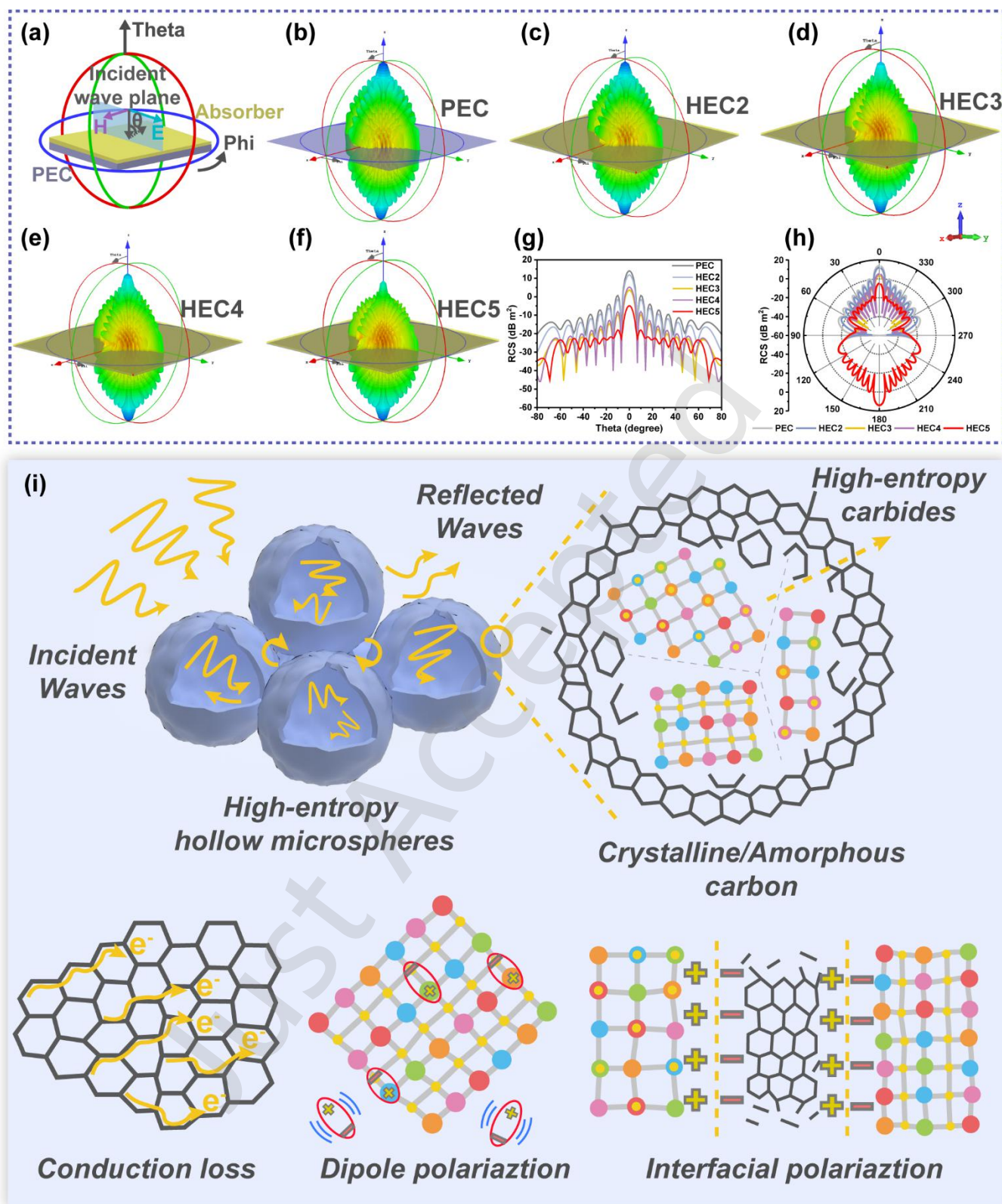


Figure 7 RCS simulation and mechanism diagram. (a) RCS calculation schematic diagram; (b-f) RCS of PEC plates and RCS after coating PEC surfaces with HEC2-HEC5; (g) RCS between $\pm 80^\circ$; (h) Compared to a 2D RCS between $\pm 90^\circ$; (i) Schematic diagram of the physical mechanism for EMW absorption in HECs@HCSs composite

4 Conclusions

In summary, this study synthesizes HECs@HCMs core-shell composite powders via the in-situ construction of composition-tunable high-entropy carbide (HEC) nanoparticles on hollow carbon spheres (HCSs). The results reveal that the high configurational entropy of the carbides induces significant lattice distortion, suppresses the graphitization of the carbon matrix, and promotes defect formation. The resulting disruption of the conductive carbon network, together with enhanced carrier scattering and interfacial transport resistance, reduces the intrinsic electrical conductivity of the nanocomposites from 20.46 S/m to 0.27 S/m. This decline promotes a shift in the dominant loss mechanism from conduction loss to polarization relaxation. The evolution of high-entropy components, the structural remodeling of the carbon matrix, and the cavity effect of the hollow architecture lower the complex permittivity and construct abundant hetero-interfaces, thereby significantly improving impedance matching. Benefiting from this synergy, at a low filler loading of 15 wt%, the HECs@HCSs composites achieve an effective absorption bandwidth of 7.24 GHz at 1.81 mm and a strong reflection loss of -45.28 dB. RCS simulations further demonstrate that the optimized HEC5 coating effectively suppresses radar scattering, achieving an RCS reduction of 18.85 dB·m² relative to the bare PEC plate at normal incidence. This research employs a dual-modulation strategy that combines compositional high-entropy and hollow-structure optimization, providing a novel paradigm for designing lightweight, wideband EMW-absorbing materials.

Electronic Supplementary Material: Supplementary material is available in the online version of this article.

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.

Acknowledgements

This work is supported by the National Key Research and Development Program of China (2024YFB3714603), National Natural Science Foundation of China (U2541259, 52304410), Major Project of Hubei Province (Functional Coating and Materials, 2023BAA003-1), and Hubei Province Innovation Research Group Project (JCZRQT202600033).

Declaration of competing interest

All the contributing author(s) report(s) no conflict of interests in this work.

Author contribution statement

Lili Li: Investigation, methodology, writing - original draft, validation, formal analysis, writing - review & editing. Pingan Chen: Conceptualization, writing - original draft, data curation, formal analysis, funding acquisition. Yingli Zhu: Software, funding acquisition. Fu Chen: Validation. Mengke Qiao: Formal analysis. Jiang Wu: Data Curation. Gangtao Luo: Visualization. Xiangcheng Li: Conceptualization, resources, data curation, project administration, funding acquisition, writing - review & editing. All the authors have approved the final manuscript.

Informed consent

Not applicable.

Ethics statement

Not applicable.

Use of AI statement

None.

References

- [1] Kang, G.; Kwon, G.; Jeon, J.; Kwon, J.; Kim, M. K.; Hong, J.; Lee, A. S.; Lee, S.; Lee, B.; Kim, Y.; Lee, M.; Choi, S.; Jeong, I.; Kang, C.; Kim, D.-A.; Park, H.; Joo, Y. C.; Yeon, H. Electromagnetic interference shielding using metal and MXene thin films. *Nature* **2025**, 647, 356-363.
- [2] Zhou, X.; Min, P.; Liu, Y.; Jin, M.; Yu, Z. Z.; Zhang, H.-B. Insulating electromagnetic-shielding silicone compound enables direct potting electronics. *Science* **2024**, 385, 1205-1210.
- [3] Zhao, B.; Yan, Z.; Du, Y.; Rao, L.; Chen, G.; Wu, Y.; Yang, L.; Zhang, J.; Wu, L.; Zhang, D. W.; Che, R. High-entropy enhanced microwave attenuation in titanate perovskites. *Adv. Mater.* **2023**, 35, 2210243.
- [4] Wang, S.; Li, Y.; Lei, D.; Ma, M.; He, X. Butterfly wing-inspired carbon fiber with confined growth of wave-trapping architecture for broadband electromagnetic wave absorption. *Adv. Funct. Mater.* **2025**, n/a, e26212.
- [5] Cao, X.; Wu, X.; Wang, X.; Luo, J.; Zhang, Z.; Xue, Y.; Zhang, G.; Zhang, L.; Zhang, H.; Yu, J. Mechanistic insights into the structural evolution of ZIF-67 via electrospinning strategy toward high electromagnetic wave absorption performance of ZIF-67-derived carbon nanofibers. *Adv. Sci.* **2025**, 12, 2502560.
- [6] Zhao, L.; Qian, Y.; Xu, X.; Wan, G.; Yu, L.; Yan, Y.; Zhao, J.; Wu, L.; Dou, Z.; Wang, G. Thermal-insulating carbon foam for efficient electromagnetic energy absorption and conversion. *Nano Energy* **2025**, 143, 111313.
- [7] Wang, J.; Liu, M.; Liu, X.; Zhang, R.; Zhong, S.; Wang, X.; Yang, Y.; Zhang, P.; Lv, D.; Xia, L. Dual-Control strategy of structure and polarization for tunable electromagnetic wave absorption in the S-Ku band. *Chem. Eng. J.* **2025**, 509, 161193.
- [8] Cui, A.; Miao, Y.; Wang, C.; Tian, Z.; Jia, Q.; Liu, J.; Song, G.; Wang, T.; Li, Z.; Zhang, M. NiFe₂O₄/Ni₃Fe nanoparticles Decorate wood carbon strategies for efficient electromagnetic absorption. *Chem. Eng. J.* **2025**, 507, 160354.
- [9] Liu, X.; Tian, F.; Sheng, J.; Yu, Y.; Yang, W. High-entropy porous spinel ferrite @ amorphous carbon nanocomposites with abundant structural defects for wide-band electromagnetic wave absorption. *Chem. Eng. J.* **2024**, 490, 151848.
- [10] Liu, J.; Zhang, L.; Wu, H. Anion-doping-induced vacancy engineering of cobalt sulfoselenide for boosting electromagnetic wave absorption. *Adv. Funct. Mater.* **2022**, 32, 2200544.
- [11] Huang, B.; Ye, F.; Cao, Y.; Liang, J.; Li, C.; Zhang, W.; Fan, X.; Xiang, L. Hierarchical CoNi@C microspheres assembled by high-content CoNi nanoparticles interpenetrated into amorphous carbon for high-performance electromagnetic wave absorption. *J. Mater. Sci. Technol.* **2026**, 253, 1-11.
- [12] Tang, Z.; Xu, L.; Xie, C.; Guo, L.; Zhang, L.; Guo, S.; Peng, J. Synthesis of CuCo₂S₄@expanded graphite with crystal/amorphous heterointerface and defects for electromagnetic wave absorption. *Nat. Commun.* **2023**, 14, 5951.
- [13] Abdalla, I.; Elhassan, A.; Ali, S.; Saty, M. Y. H.; Adam, E.; Zou, L.; Ni, Q.; Xu, Z. Impact of defect-rich carbon nanofibers combined with magnetic materials on broadband electromagnetic wave absorption

- and radar cross-section reduction. *Small Struct.* **2025**, 6, 2400624.
- [14] Jiang, T.; Zhao, T.; Jalil, A.; Li, X.; Ma, M. Pillared amorphous carbon nanotube/MXene nanohybrid for ultrathin broadband electromagnetic wave absorption. *Chem. Eng. J.* **2026**, 528, 172520.
- [15] Ge, J.; Liu, S.; Liu, L.; Cui, Y.; Meng, F.; Li, Y.; Zhang, X.; Wang, F. Optimizing the electromagnetic wave absorption performance of designed hollow $\text{CoFe}_2\text{O}_4/\text{CoFe@C}$ microspheres. *J. Mater. Sci. Technol.* **2021**, 81, 190-202.
- [16] Ding, H.; Song, W.; Chen, Y.; Wang, Y.; Wang, Y.; Tian, C.; Du, Y. Hollow engineering of carbon-based microspheres: Microstructural modulation for advanced electromagnetic wave absorption. *Carbon* **2025**, 244, 120621.
- [17] Zhang, Y.; Liu, X.; Guo, Z.; Jia, C.; Lu, F.; Jia, Z.; Wu, G. MXene@Co hollow spheres structure boosts interfacial polarization for broadband electromagnetic wave absorption. *J. Mater. Sci. Technol.* **2024**, 176, 167-175.
- [18] Ban, Q.; Li, L.; Liu, H.; Zhou, D.; Zheng, Y.; Qin, Y.; Xing, R.; Kong, J. Polymerization-induced assembly-etching engineering to hollow Co@N -doped carbon microcages for superior electromagnetic wave absorption. *Carbon* **2023**, 215, 118506.
- [19] Wang, R.; He, Y.; Tang, C.; Wu, X.; Zhuang, Q.; Liu, X.; Liu, X.; Zuo, P. Constructing crystalline/amorphous heterojunction in FeCo@C nanoboxes for enhanced electromagnetic wave absorption. *Carbon* **2024**, 229, 119494.
- [20] Sun, Y.; Shi, Y.; Zhang, X.; Cao, F.; Huang, L.; Chen, Y. Pb single-atoms on nitrogen-doped graphene hollow spheres for electromagnetic wave absorption. *Carbon* **2025**, 240, 120352.
- [21] Zhang, S.; Zheng, J.; Liang, X.; Lan, D.; Niu, L.; Zhao, X.; Zhao, Z.; Zhang, S.; Wu, G.; Li, X. Phosphorus-driven heteroatom doping in mesoporous carbon hollow platelets enables efficient electromagnetic wave absorption. *Small* **2025**, 21, e09237.
- [22] Chen, J.; Zheng, J.; Huang, Q.; Wang, G.; Ji, G. Carbon fibers@Co-ZIFs derivations composites as highly efficient electromagnetic wave absorbers. *J. Mater. Sci. Technol.* **2021**, 94, 239-246.
- [23] Luo, J.; Yan, W.; Li, X.; Shu, P.; Mei, J.; Shi, Y. Carbon nanotubes decorated FeNi /nitrogen-doped carbon composites for lightweight and broadband electromagnetic wave absorption. *J. Mater. Sci. Technol.* **2023**, 158, 207-217.
- [24] Yang, Y.; Qiao, J.; Zhang, X.; Zhang, B.; Liu, J. Electrospun magnetic carbon nanofibers for electromagnetic wave absorption: A review. *J. Mater. Sci. Technol.* **2026**, 265, 1-22.
- [25] Dong, Y.; Lan, D.; Xu, S.; Gu, J.; Jia, Z.; Wu, G. Controllable fiberization engineering of cobalt anchored mesoporous hollow carbon spheres for positive feedback to electromagnetic wave absorption. *Carbon* **2024**, 228, 119339.
- [26] Dou, Y.; Liu, N.; Zhang, X.; Jiang, W.; Jiang, X.; Yu, L. Synthesis of polymer-derived N,O-doped bowl-like hollow carbon microspheres for improved electromagnetic wave absorption using controlled template pyrolysis. *Chem. Eng. J.* **2023**, 463, 142398.
- [27] Wang, S.; Liu, Q.; Li, S.; Huang, F.; Zhang, H. Joule-Heating-driven synthesis of a honeycomb-like porous carbon nanofiber/high entropy alloy composite as an ultralightweight electromagnetic wave absorber. *ACS Nano* **2024**, 18, 5040-5050.
- [28] Zhang, X.; Hu, J.; Li, W.; Zhang, J.; Xu, X.; Ying, Y.; Yu, J.; Zheng, J.; Qiao, L.; Li, J.; Inoue, A.; Che, S. Synergistic dynamic phase transformation and multistage structure enabling ultra-broadband electromagnetic wave attenuation in high entropy alloy. *Adv. Funct. Mater.* **2026**, 36, e16735.
- [29] Gang, S.; Qian, Y.; Xiong, T.; Li, Y.; Li, W.; Ma, Z.; Zhang, W.; Long, H.; Wei, Y.; Li, X.; Jiang, Q.; Luo, Y.; Yang, J. High electromagnetic wave absorption performance from all-scale hierarchically defective Ni-Mn-Co-Cu-Al high entropy alloys@carbon. *Chem. Eng. J.* **2024**, 499, 156254.
- [30] Hassan, S. u.; Hou, L.; Yang, Y.; Qamar, T. H.; Wang, S. Optimizing the electromagnetic wave attenuation properties of carbon encapsulated FeCoNiCuMn_x high entropy alloy nanoparticles. *Carbon* **2024**, 229, 119502.
- [31] Liu, Y.; Yao, J.; Shen, Z.; Zhang, M.; Xu, D.; Chen, P. Magnetic quantum dots modified hollow carbon spheres and controllable electromagnetic wave absorption performance. *Composites, Part A* **2024**, 179, 108056.
- [32] Jiang, B.; Wang, W.; Liu, S.; Wang, Y.; Wang, C.; Chen, Y.; Xie, L.; Huang, M.; He, J. High figure-of-merit and power generation in high-entropy GeTe-based thermoelectrics. *Science* **2022**, 377, 208-213.
- [33] Yang, B.; Zhang, Q.; Huang, H.; Pan, H.; Zhu, W.; Meng, F.; Lan, S.; Liu, Y.; Wei, B.; Liu, Y.; Yang, L.; Gu, L.; Chen, L.-Q.; Nan, C.-W.; Lin, Y.-H. Engineering relaxors by entropy for high energy storage performance. *Nature Energy* **2023**, 8, 956-964.
- [34] Williamson, G. K.; Hall, W. H. X-ray line broadening from filed aluminium and wolfram. *Acta Metall.* **1953**, 1, 22-31.
- [35] Langford, J. I.; Wilson, A. J. C. Scherrer after sixty years: A survey and some new results in the determination of crystallite size. *J. Appl. Crystallogr.* **1978**, 11, 102-113.
- [36] Philo, D.; Luo, S.; He, C.; Wang, Q.; Ichihara, F.; Jia, L.; Oshikiri, M.; Pang, H.; Wang, Y.; Li, S.; Yang, G.; Ren, X.; Lin, H.; Ye, J. Lattice Distortion Engineering over Ultrathin Monoclinic BiVO_4 Nanoflakes Triggering AQE up to 69.4% in Visible-Light-Driven Water Oxidation. *Adv. Funct. Mater.* **2022**, 32, 2206811.
- [37] Wan, Q.; Jia, B. Y.; Liu, P.; Luo, Y.; Chen, J.; Zhang, X. Y.; Xiao, Y. Y.; Abdelkader, T. K.; Refai, M.; Zhang, J.; Yang, B. Microstructure and mechanical properties of $\text{FeCoCrNiAl}_{0.1}\text{N}$ high entropy alloy nitride coatings synthesized by cathodic arc ion plating using alloy target. *Surf. Coat. Technol.* **2023**, 457, 129305.
- [38] Xiao, X.; Zhang, H.; Xiong, Y.; Liang, F.; Yang, Y.-W. Iridium-doped N-rich mesoporous carbon electrocatalyst with synthetic macrocycles as carbon source for hydrogen evolution reaction. *Adv. Funct. Mater.* **2021**, 31, 2105562.
- [39] Gao, Y.; Huang, L.; Tong, Z.; Liu, J.; Zhang, Q.; Zhang, H.; Jia, Q.; Zhang, S. Low-temperature synthesis of high-entropy $(\text{Hf}_{0.2}\text{Ti}_{0.2}\text{Mo}_{0.2}\text{Ta}_{0.2}\text{Nb}_{0.2})\text{B}_2$ powders combined with theoretical forecast of its elastic and thermal properties. *J. Am. Ceram. Soc.* **2022**, 105, 6370-6383.
- [40] Chen, J.; Chen, P.; Zhu, Y.; Chen, F.; Qiao, M.; Wu, J.; Li, X. Modulation of carbon atom hybridization structure and electromagnetic absorption properties in carbon fiber composites. *Carbon* **2025**, 245, 120823.
- [41] Chen, F.; Zhang, S.; Guo, R.; Ma, B.; Xiong, Y.; Luo, H.; Cheng, Y.; Wang, X.; Gong, R. 1D magnetic nitrogen doped carbon-based fibers derived from NiFe Prussian blue analogues embedded polyacrylonitrile via electrospinning with tunable microwave absorption. *Composites, Part B* **2021**, 224, 109161.
- [42] Yang, Q.; Chen, P.; Li, X.; Zhu, Y. Rational design of SiBCN ceramics with excellent attenuation to strong electromagnetic-wave-absorbing properties at low frequency. *Composites, Part B* **2024**, 280, 111486.
- [43] Agrawal, P.; Conlon, K.; Bowman, K. J.; Sun, C. T.; Cichocki, F. R.; Trumble, K. P. Thermal residual stresses in co-continuous composites. *Acta Mater.* **2003**, 51, 1143-1156.
- [44] Bao Y.; Su S.; Huang J. An uneven strain model for analysis of residual stress and interface stress in laminate composites. *J. Compos. Mater.* **2002**, 36, 1689-1706.
- [45] Wang, H.; Yang, P. Y.; Zhao, W. J.; Ma, S. H.; Hou, J. H.; He, Q. F.; Wu, C. L.; Chen, H. A.; Wang, Q.; Cheng, Q.; Guo, B. S.; Qiao, J. C.; Lu, W. J.; Zhao, S. J.; Xu, X. D.; Liu, C. T.; Liu, Y.; Pao, C. W.;

- Yang, Y. Lattice distortion enabling enhanced strength and plasticity in high entropy intermetallic alloy. *Nat. Commun.* **2024**, 15, 6782.
- [46] Hofmann, T.; Ren, X.; Weymouth, A.; Meuer, D.; Liebig, A.; Donarini, A.; Giessibl, F. Evidence for temporary and local transition of sp² graphite-type to sp³ diamond-type bonding induced by the tip of an atomic force microscope. *New J. Phys.* **2022**, 24.
- [47] Sun, X.; Li, X.; Chen, P.; Zhu, Y.; Qiao, M. Progressive hollow engineering induced raspberry-like magnetoelectric microspheres via synergistic etching-assembly strategy toward boosted microwave attenuation. *Chem. Eng. J.* **2024**, 499, 155857.
- [48] Zhang, S.; Liu, G.; Lv, S.; Zhu, Y.; Chen, P.; Luo, H.; Li, X.; Chen, F. Ti-MOF@Metal-polyphenol network derived TiN_{0.9}@NC/magnetic MWCNTs composites for microwave absorption. *Chem. Eng. J.* **2023**, 468, 143763.
- [49] Li, Y.; Zhu, Y.; Luo, G.; Chen, P.; Qiao, M.; Chen, F.; Li, X. Fe-induced stacking faults engineering for breakthrough broadband absorption in SiBCN ceramics. *J. Mater. Sci. Technol.* **2026**, 258, 1-11.
- [50] Ferrari, A. C.; Robertson, J. Interpretation of Raman spectra of disordered and amorphous carbon. *Phys. Rev. B* **2000**, 61, 14095-14107.
- [51] Robertson, J. Diamond-like amorphous carbon. *Mater. Sci. Eng. R Rep.* **2002**, 37, 129-281.
- [52] Wang, C.; Chen, P.; Li, X.; Zhu, Y.; Zhu, B. Enhanced electromagnetic wave absorption for Y₂O₃-doped SiBCN ceramics. *ACS Appl. Mater. Interfaces* **2021**, 13, 55440-55453.
- [53] Li, T.; Qiao, M.; Chen, F.; Li, X. Electrostatic self-assembled CS/Ti₃C₂T_x/Co@CNT composites with gradient carbon structure and wideband microwave absorption. *Nano Research* **2025**, 18, 94907696.
- [54] Hu, S.; Chen, P.; Li, X.; Zhu, Y.; Zhu, B. A competitive reaction strategy toward dielectric phases for enhancing electromagnetic wave absorption of polymer-derived ceramics. *J. Mater. Chem. A* **2024**, 12, 19298-19309.
- [55] Chang, L.; Zhang, X.; Liu, T.; Li, B.; Ji, Y.; Sun, G.; Wang, Z.; Zhang, X.; Cao, M.; Li, L. Multifunctional three-dimensional porous MXene-based film with superior electromagnetic wave absorption and flexible electronics performance. *Nano-Micro Lett.* **2026**, 18, 184.
- [56] Luo, G.; Zhu, Y.; Chen, P.; Chen, F.; Li, X.; Che, R. Electromagnetic attenuation of Sm-doped alloys modified by interface exchange coupling and magnetocrystalline stress anisotropy. *Adv. Funct. Mater.* **2025**, 35, 2425298.
- [57] Chen, J.; Chen, P.; Li, X.; Zhu, Y.; Zhu, B. Enhancement of electromagnetic attenuation of network CNT-Carbon microsphere nanocomposites derived from phenolic resin. *Carbon* **2024**, 219, 118789.
- [58] Luo, H.; Chen, F.; Wang, X.; Dai, W.; Xiong, Y.; Yang, J.; Gong, R. A novel two-layer honeycomb sandwich structure absorber with high-performance microwave absorption. *Composites, Part A* **2019**, 119, 1-7.
- [59] Xu, L.; Liu, X.; Si, Z.; Liao, H.; Li, D.; Zhan, G.; Wan, G.; Wu, L.; Wang, G. Dual-interface charge migration polarization boosting broadband electromagnetic wave absorption. *Journal of Energy Chemistry* **2026**, 116, 144-154.
- [60] Ren, B.; Jia, Y.; Qi, L.; Fu, Q.; Lin, Y.; Yu, H.; Cheng, J.; Dai, M.; Song, Q.; Zhang, Y.; Hou, X.; Li, H. Broadband electromagnetic absorption up to 1473 K enabled by dielectric frequency-dispersion engineering in ceramic composites. *Nat. Commun.* **2025**, 17, 318.
- [61] Liu, P.; He, Z.; Li, X.; Ding, L.; Liu, S.; Kong, J. Multifunctional hollow carbon microspheres enable superior electromagnetic wave response and corrosion barrier. *Adv. Mater.* **2025**, 37, 2500646.
- [62] Guan, X.; Jiang, M.; Shen, Z.; Tan, S.; Ji, G.; Xu, Z. J. High-entropy interface engineering in multifunctional green fiber aerogels for coupled electromagnetic and waste-heat management. *Adv. Funct. Mater.* **2025**, n/a, e27596.
- [63] Zhang, S.; Wang, C.; Yu, M. Design and synthesis of core-shell Fe₄N@C nanowires for efficient electromagnetic wave absorption and corrosion resistance. *Carbon* **2026**, 250, 121306.
- [64] Li, T.; Qiao, M.; Chen, F.; Chen, P.; Li, X. Efficient electromagnetic wave absorption of Ti₃C₂T_x composite hollow spheres via multilayer heterostructure impedance matching. *Carbon* **2026**, 250, 121286.

© The Author(s) 2026. *Nano Research* published by Tsinghua University Press. The articles published in this open access journal are distributed under the terms of the Creative Commons Attribution 4.0 International License (<http://creativecommons.org/licenses/by/4.0/>), which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Strain-induced defects suppress conductivity to enhance microwave absorption in high-entropy hollow microspheres

Lili Li^{1,2}, Pingan Chen^{1,2}  , Yingli Zhu^{1,2}, Fu Chen^{1,2}, Mengke Qiao^{1,2}, Jiang Wu^{1,2}, Gangtao Luo^{1,2}, Xiangcheng Li^{1,2}  

¹ State Key Laboratory of Advanced Refractories, Wuhan University of Science and Technology, Wuhan 430081, China

² Key Laboratory of High Temperature Electromagnetic Materials and Structure of MOE, Wuhan University of Science and Technology, Wuhan 430081, China

Supporting information to <https://doi.org/10.26599/NR.2026.94909033>

Equations

All calculation formulas used in this article are as follows:

$$\epsilon' = \epsilon_{\infty} + \frac{\epsilon_s - \epsilon_{\infty}}{1 + (2\pi f)^2 \tau^2} \quad (1)$$

$$\epsilon'' = \frac{2\pi f \tau (\epsilon_s - \epsilon_{\infty})}{1 + (2\pi f)^2 \tau^2} \quad (2)$$

$$\left(\epsilon' - \frac{(\epsilon_s - \epsilon_{\infty})}{2} \right) + (\epsilon'')^2 = \frac{(\epsilon_s - \epsilon_{\infty})^2}{2} \quad (3)$$

$$\epsilon'' = \epsilon_c'' + \epsilon_p'' \quad (4)$$

$$\epsilon_c'' = \frac{\sigma}{\omega \epsilon_0} \quad (5)$$

$$\epsilon_p'' = \frac{(\epsilon_s - \epsilon_{\infty}) \omega \tau}{(1 + \omega^2 \tau^2)} \quad (6)$$

Where f is frequency, ϵ_{∞} is the optical dielectric constant, ϵ_s is the static dielectric constant, ϵ_c'' is the contribution of conductive loss, ϵ_p'' is the contribution of dielectric loss, σ is the leakage conductivity and τ is the relaxation time.

$$R_L = 20 \ln \left| \frac{Z_{in} + 1}{Z_{in} - 1} \right| \quad (7)$$

$$Z_{in} = \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh h \left[j \frac{2\pi f d}{c} \sqrt{\epsilon_r \mu_r} \right] \quad (8)$$

Where Z_{in} is the normalized input impedance of the material, ϵ_r is complex permittivity, μ_r is magnetic permeability, and c is the propagation velocity of the EMW in vacuum.

This file includes:

Figures. S1 to S7

Tables. S1 to S4

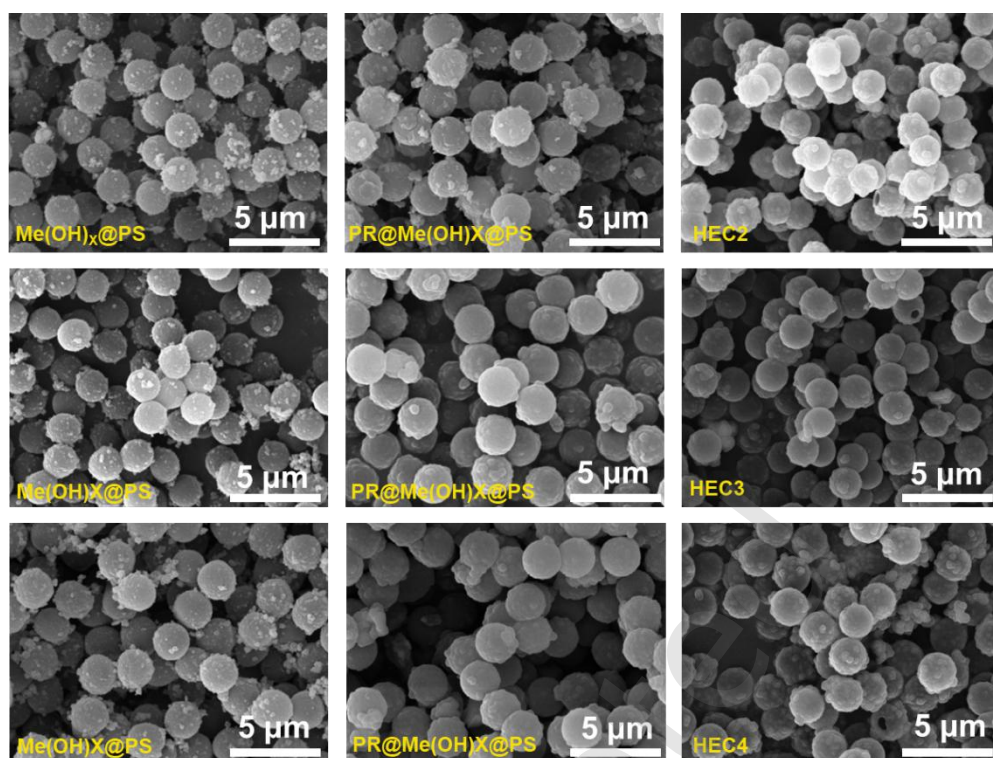


Figure S1 SEM image of $\text{Me(OH)}_x\text{@PS}$ microspheres, $\text{PR@Me(OH)}_x\text{@PS}$ microspheres, HEC2, HEC3 and HEC4

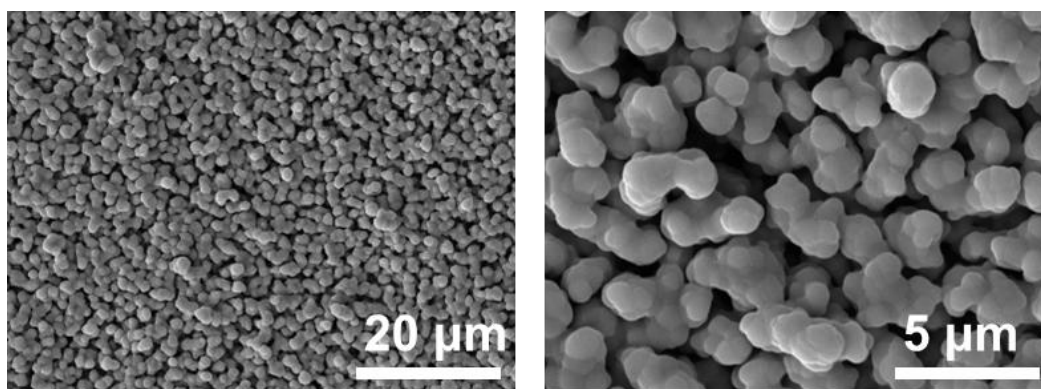


Figure S2 SEM image of Me(OH)_x@PS microspheres after heat treatment

Just Accepted

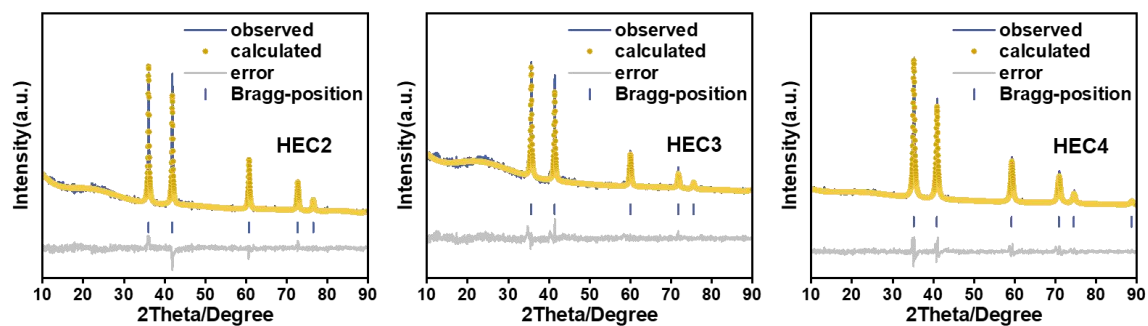


Figure S3 XRD refinement of HEC2, HEC3 and HEC4

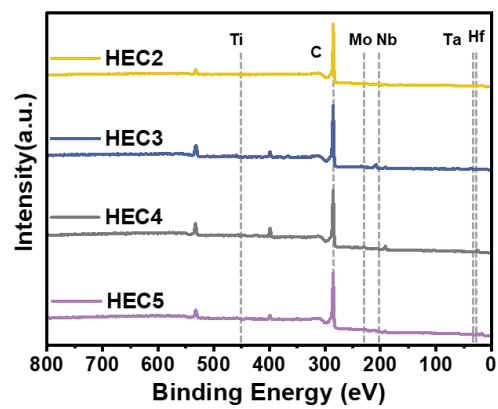


Figure S4 XPS spectra diagram

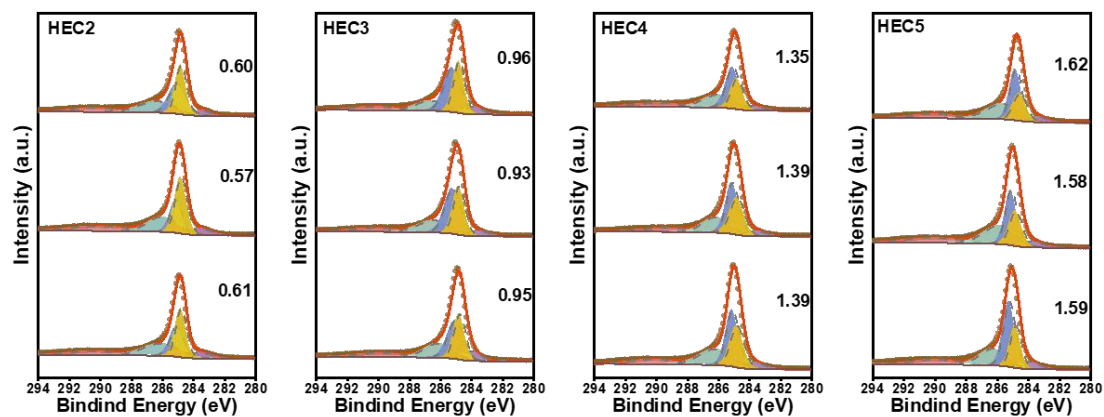


Figure S5 High-resolution C 1s XPS spectra of HECs@HCSs composites

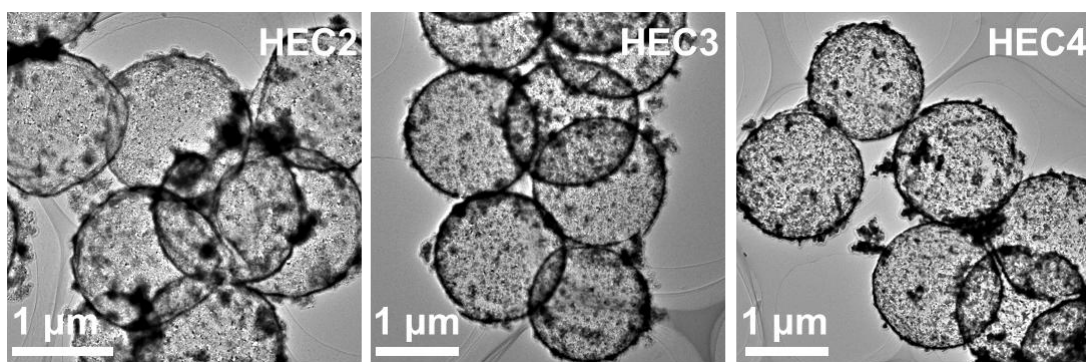


Figure S6 TEM of HEC2, HEC3 and HEC4

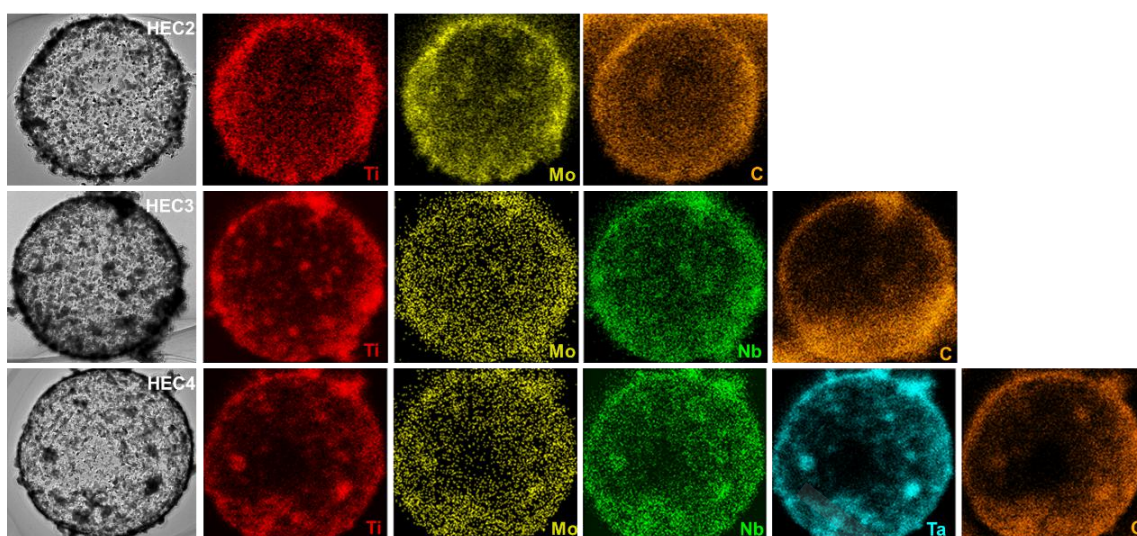


Figure S7 EDS of HEC2, HEC3 and HEC4

Table S1. Synthesis conditions for carbides	
carbides	Molar ratio
HEC2	Ti: Mo=1:1
HEC3	Ti: Mo: Nb=1:1:1
HEC4	Ti: Mo: Nb: Ta=1:1:1:1
HEC5	Ti: Mo: Nb: Ta: Hf=1:1:1:1:1

Just Accepted

Table S2 Crystallographic parameters, derived intrinsic lattice strains, and Rietveld refinement reliability factors for HECs@HCSs composites

Samples	Rwp (%)	χ^2	ε (%)
HEC2	8.87	1.22	3.551±0.000515
HEC3	8.16	1.13	7.090±0.000141
HEC4	7.68	1.44	9.684±0.000401
HEC5	9.02	1.36	13.451±0.000165

Just Accepted

Table S3 Deconvoluted peak positions, areas, and relative atomic percentages of carbon species in the high-resolution C 1s XPS spectra for HECs@HCSs composites

Sample	Trial 1 (sp ³ /sp ²)	Trial 2 (sp ³ /sp ²)	Trial 3 (sp ³ /sp ²)	Statistical Value	Original Value
HEC2	0.60	0.57	0.61	0.593±0.021	0.59
HEC3	0.96	0.93	0.95	0.947±0.015	0.94
HEC4	1.36	1.39	1.39	1.380±0.017	1.39
HEC5	1.62	1.58	1.59	1.597±0.021	1.59

Table S4. Comparison with similar jobs

Samples	EAB (GHz)	Filling ratio (%)	Reference
S1	1.12	70	[1]
S2	7	30	[2]
S3	6.81	10	[3]
S4	6.85	20	[4]
S5	5.8	10	[5]
S6	5.12	15	[6]
S7	4.64	10	[7]
S8	5.26	30	[8]
S9	4.13	30	[9]
S10	4.64	60	[10]
S11	4.68	25	[11]
This work	7.24	15	/

Reference

- [1] Yang, H.; Zou, J.; Zhang, N.; Li, L.; Liu, J.; Ma, H.; Bao, W.; Ji, W.; Wang, W.; Fu, Z. Effects of temperature-induced lattice distortion and phase evolution on electromagnetic wave absorption in high-entropy powders. *J. Mater. Chem. C* **2026**, 14, 196-204.
- [2] Chai, W.; Yang, H.; Ma, Y.; Cai, Z.; Zhou, H.; Liu, Y.; Lin, Y. Hierarchical structure SiC@CoNi-layered double hydroxide fibers for high performance electromagnetic wave absorption. *J. Mater. Chem. A* **2025**, 13, 17689-17698.
- [3] Liu, Z.; Zhao, X.; Jiang, X.; Yan, X.; Yu, L. Carbon microspheres/carbon sheets as efficient electromagnetic wave absorbers. *Microporous and Mesoporous Materials* **2024**, 372, 113090.
- [4] Mo, P.; Shui, A.; Yu, H.; Qian, J. Facile synthesis of MoO₂/S-doped carbon nanofibers for ultra-high electromagnetic wave absorption. *Carbon* **2025**, 237, 120121.
- [5] Han, X.; Feng, Y.; Zhang, N.; Du, W.; Zhang, W.; Yu, Q.; Liu, Y.; Wang, B.; Jiang, F.; Liu, L.; Xu, L.; Zhang, X. Bio-based multifunctional carbon aerogels for ultrawide electromagnetic wave absorption and thermal insulation. *Carbon* **2025**, 238, 120231.
- [6] Wang, C.; Hua, L.; Li, J.; Bai, H.; Li, J.; Ibragimov, A. B.; Khalilov, U.; Yusupboyevich, Y. Y.; Lu, H. Electromagnetic wave absorption and Joule heating of carbon fiber/carbon nanotube composites via solid carbon source-chemical vapor deposition method. *Carbon* **2026**, 247, 121049.
- [7] Sun, Y.; Shi, Y.; Zhang, X.; Cao, F.; Huang, L.; Chen, Y. Pb single-atoms on nitrogen-doped graphene hollow spheres for electromagnetic wave absorption. *Carbon* **2025**, 240, 120352.
- [8] Wei, P.; Liu, Y.; Bai, H.; Zhuang, L.; Yu, H.; Chen, Z.; Chu, Y. A universal laser-driven controllable synthesis methodology enabling electromagnetic absorption in high-entropy rare-earth oxides. *Adv. Mater.* **2026**, 38, e20122.
- [9] Lei, B.; Zhou, C.; Zhang, K.; Li, J.; Miao, H.; Wang, H.; Fan, Q.; Yang, Z.; Wang, G.; Li, Y.; Kong, C. Plasma-induced geometry engineering in high-entropy oxide composites for superior electromagnetic absorption. *Adv. Funct. Mater.* **2025**, 35, 2425262.
- [10] Liu, Y.; Liu, J.; Zhou, E.; Liu, D.; Cui, J.; Li, B.; Du, Y.; Yang, L.; Liang, G.; Lv, H.; Che, R. Entropy-driven multi-ion coexistence at heterogeneous nanoscale interfaces of transition metal sulfides with anomalous electronic transport-enhancement. *Adv. Funct. Mater.* **2026**, 36, e14873.
- [11] Zhu, Y.; Guan, L.; Duan, C.; Zhang, J.; Yan, Z.; Wen, L.; Wang, Z.; Sun, X.; Yao, Y.; Guo, X.; Zhang, R.; Zhao, B. High-entropy transition metal carbide nanowires with enhanced microwave absorption properties. *J. Mater. Sci. Technol.* **2025**, 224, 302-311.