



# Liquid metal assisted dual hydroxide heterostructures of carbonyl iron powder for multifunctional electromagnetic wave absorption

Yunpeng Li<sup>1,2</sup>, Shenghao Yue<sup>3</sup>, Ting-An He<sup>1,2</sup>, Jiabao Zang<sup>1,2</sup>, Xuejun Bai<sup>1,2</sup>, Lihong Gao<sup>1,2,4</sup>, Zhuang Ma<sup>1,2,4</sup>, Qi Cao<sup>3</sup>, Miao Jiang<sup>1,2,4,5</sup>

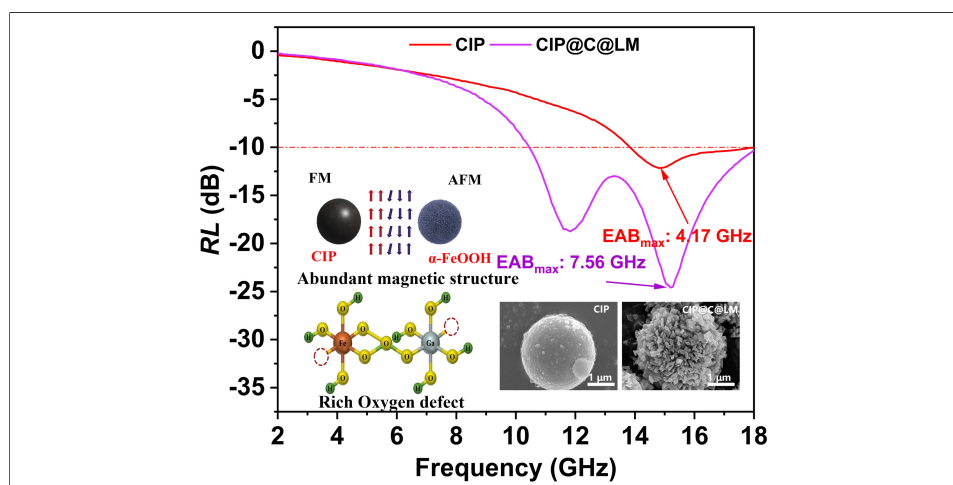
## Keywords:

Carbonyl iron powder, dual-hydroxide, oxygen defect, electromagnetic wave absorption, multifunctional

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## Abstract

Electromagnetic wave (EMW) absorbers for complex environments require appropriate impedance matching, strong absorption, and rapid heat dissipation. Although carbonyl iron powder (CIP) exhibits high magnetic loss, its practical application is hindered by intrinsic oxidation, as well as severe impedance mismatch and low thermal conductivity at low filler loadings. Herein, by applying an ultrasonic cavitation-driven Ga-based liquid metal (LM) encapsulation process, we constructed a core-shell heterostructure (CIP@C@LM), in which an intermediate C shell was introduced to protect the CIP core from severe oxidation and to improve impedance matching. During this process, an oxygen-vacancy rich  $\alpha$ -GaOOH outer shell and a small amount of interfacial  $\alpha$ -FeOOH form in situ, enabling dielectric-loss dominated attenuation through enhanced polarization relaxation and a secondary magnetic-loss contribution. Consequently, at a filler loading of 60 wt.%, the

<sup>1</sup>School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China.

<sup>2</sup>National Key Laboratory of Science and Technology on Materials under Shock and Impact, Beijing Institute of Technology, Beijing 100081, China.

<sup>3</sup>Key Laboratory of Energy Thermal Conversion and Control of Ministry of Education, Key Laboratory of Functional Polymers for Sustainability of Jiangsu Province, School of Energy and Environment, Southeast University, Nanjing 210096, Jiangsu, China.

<sup>4</sup>Materials Intelligent Innovation Laboratory (MIIL), Beijing Institute of Technology, Zhuhai 519088, Guangdong, China.

<sup>5</sup>Yangtze Delta Region Academy in Jiaxing, Beijing Institute of Technology, Jiaxing 314000, Zhejiang, China.

**Correspondence to:** Prof. Miao Jiang, School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China. E-mail: [jiangmiao@bit.edu.cn](mailto:jiangmiao@bit.edu.cn)

composite achieves a minimum reflection loss ( $RL_{\min}$ ) of -52.6 dB at a thickness of 2.31 mm and a maximum effective absorption bandwidth ( $EAB_{\max}$ ) of 7.56 GHz (10.44–18 GHz) at a thickness of 2.08 mm. The excellent performance significantly outperforms that of the pristine CIP ( $RL_{\min} = -16.29$  dB;  $EAB_{\max} = 4.17$  GHz). Furthermore, the C and dual-hydroxide shells act as a physical barrier and a heat-conducting network, providing strong corrosion resistance and rapid cooling ability (from 96.7 °C to 37.7 °C in 40 s). This work provides new insights into interfacial engineering, offering a viable route toward next-generation multifunctional EMW absorbers.

## INTRODUCTION

As electromagnetic interference increasingly compromises the reliability of wireless systems, the pursuit of efficient electromagnetic wave (EMW) absorbers has become a priority in materials science<sup>[1,2]</sup>. Among various candidates, magnetic metallic materials, particularly carbonyl iron powder (CIP), have attracted extensive attention for their high saturation magnetization ( $M_s$ ) and superior magnetic loss capabilities. However, the practical application of pristine CIP is severely hindered by its high density, susceptibility to oxidation, and intrinsic impedance mismatch at low filler loadings<sup>[3,4]</sup>. To overcome these limitations, reducing filler loading while simultaneously enriching the attenuation mechanisms via interfacial design has emerged as a critical route for advancing CIP-based absorbers<sup>[5]</sup>.

Core-shell heterostructure engineering provides a robust framework for addressing these limitations. By integrating a magnetic core with a dielectric or conductive shell, these architectures introduce abundant heterogeneous interfaces that strengthen interfacial polarization and relaxation losses. Specifically, at high filler loadings, a dielectric shell buffers excessive conductivity to optimize impedance matching; conversely, at low loadings, a conductive shell rebuilds the conductive network to maintain sufficient attenuation. In this context, significant efforts have been devoted to encapsulating CIP with diverse functional shells. For instance, inorganic oxides (e.g.,  $\text{SiO}_2$  and  $\text{TiO}_2$ ) have been widely employed to improve impedance matching and thermal stability<sup>[6–8]</sup>, while carbonaceous shells [e.g., carbon (C), graphene (GA), carbon nanotubes (CNTs)] are adopted to enhance dielectric loss and reduce the overall density<sup>[9–11]</sup>. Furthermore, conducting polymers such as polyaniline (PANI) and polypyrrole (PPy) have been utilized to construct conductive networks that boost conduction loss<sup>[12,13]</sup>. More recently, advanced yolk-shell structures featuring internal voids have been developed to induce multiple scattering and further lower the effective density<sup>[14,15]</sup>. Beyond electromagnetic performance, a rational shell can also protect oxidation-prone magnetic cores and enhance environmental tolerance, both of which are often necessary for realistic service conditions. However, conventional shell materials often rely on simple physical coatings, which struggle to construct strongly coupled heterointerfaces and lacks engineerable active defect sites. Consequently, there is a compelling need to design advanced multi-layer architectures - integrating magnetic cores, conductive buffer layers, and defect-rich dielectric shells - to strengthen electromagnetic attenuation and realize truly multifunctional absorbers.

Recently, Ga-based liquid metals (LM for short in this work) have emerged not only as conductive fluids, but also as highly active precursors for synthesizing defect-rich derivatives (e.g., metal oxides and hydroxides) to enhance the EMW absorption performance<sup>[16–21]</sup>. However, a critical bottleneck arises from the intrinsically high surface tension of LM ( $\sim 724 \text{ mN m}^{-1}$ )<sup>[22]</sup>, which drives the formation of macroscopic spherical droplets to minimize surface energy. Consequently, conventional mechanical mixing often fails to produce well-defined heterointerfaces, leading to unsatisfactory impedance matching and a limited bandwidth<sup>[23–25]</sup>. For instance, Zhao *et al.*<sup>[26]</sup> studied the construction of multi-layer heterogeneous interfaces in LM-GA mixed powder. The minimum reflection loss ( $RL_{\min}$ ) at a thickness of 3.3 mm was -42.68 dB, and the maximum effective absorption bandwidth ( $EAB_{\max}$ ) was 4.11 GHz. To overcome this limitation, ultrasonication offers a potent route by dispersing bulk LMs through high-intensity acoustic cavitation<sup>[26–28]</sup>. Cavitation fragments



bulk LMs via surface cavitation (shock-wave ejection upon near-surface bubble collapse) and internal cavitation (bubble nucleation within the LM followed by rupture and particle release)<sup>[29]</sup>. Crucially, the high-energy acoustic environment accelerates surface hydrolysis, driving the rapid transformation of metallic Ga into  $\text{Ga}_2\text{O}_3/\text{GaOOH}$ . This chemical conversion significantly lowers the interfacial energy<sup>[30]</sup>, thereby facilitating the formation of stable core-shell architectures while promoting the in-situ growth of anisotropic GaOOH phases enriched in oxygen vacancies ( $\text{O}_\text{v}$ )<sup>[31,32]</sup>. These vacancy-rich interfaces are pivotal for introducing defect states and enhancing dielectric loss via polarization relaxation<sup>[33–40]</sup>. Nevertheless, critical challenges persist, particularly the aggressive oxidative environment during sonication, which threatens the integrity of magnetic cores (e.g., CIP), as well as the need to further elucidate the mechanism underlying the relationship between intrinsic magnetic loss and vacancy-induced polarization.

Herein, we design a core-shell heterostructure (CIP@C@LM) via an ultrasonic cavitation-driven LM encapsulation process. Specifically, an intermediate C shell is introduced to protect the CIP core from severe oxidation during sonication and improve impedance matching. During this process, the outer LM shell undergoes in-situ oxidation to form an  $\alpha$ -GaOOH shell, which provides abundant  $\text{O}_\text{v}$  sites for defect-driven dielectric polarization. Concurrently, a small amount of interfacial antiferromagnetic (AFM)  $\alpha$ -FeOOH is formed, enriching the local magnetic structure and contributing to the effective magnetic response. Furthermore, the C and  $\alpha$ -GaOOH shells act as a physical barrier and heat-conducting network, equipping the absorbers with effective thermal management and corrosion resistance.

## MATERIALS AND METHODS

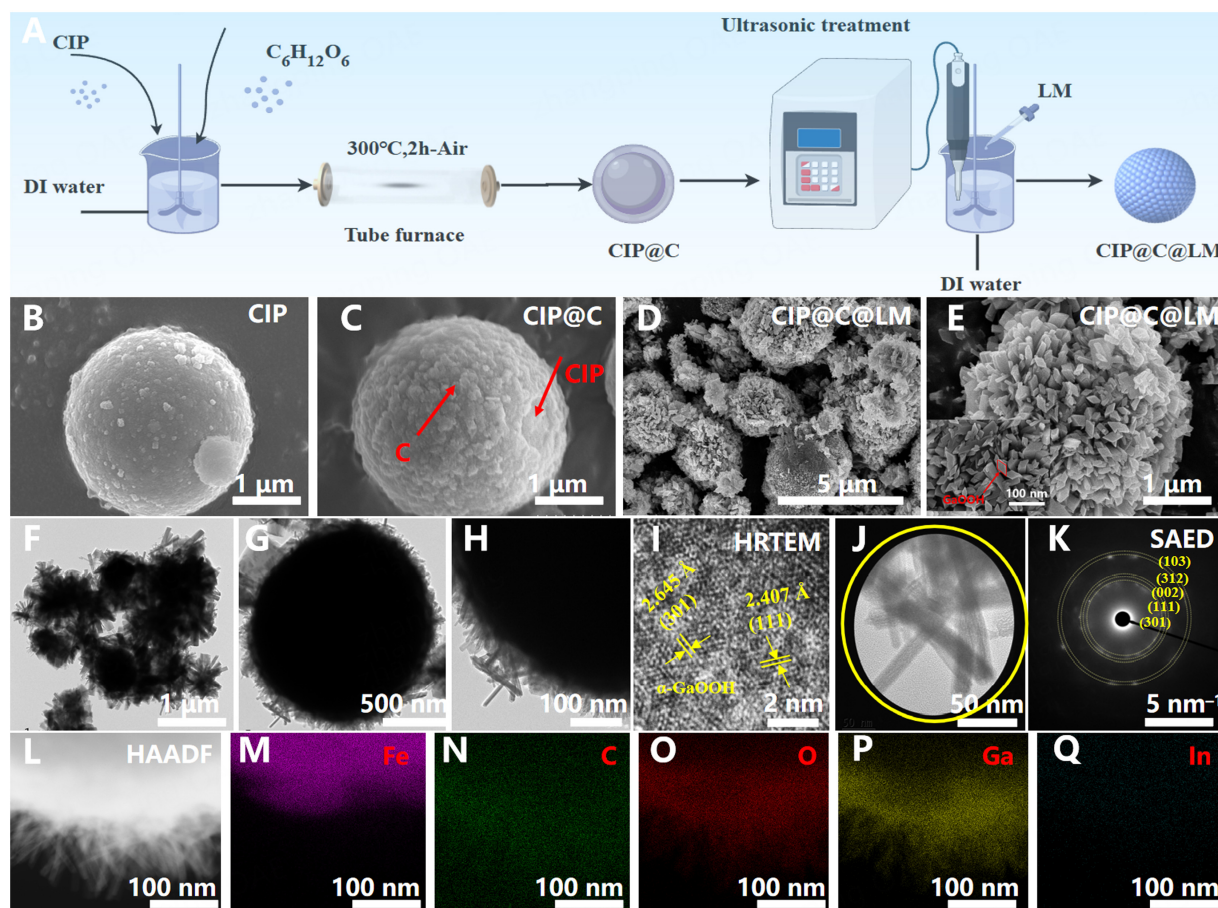
### Materials

Spherical CIP (purity > 99.9%) with an average particle size of 2–3  $\mu\text{m}$  was purchased from BASF SE (Ludwigshafen, Germany). LM (EGaIn, 78.5 wt.% Ga/21.5 wt.% In) was supplied by Beijing Zhongke Yannuo New Materials Technology Co., Ltd. Reagent-grade glucose (99.9%) and polyethylene glycol (PEG-400) were obtained from Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). Deionized (DI) water was provided by Beijing Jindong Tian Zheng Precision Chemical Co., Ltd. All chemicals were of analytical grade and used as received without further purification.

### Experiments

Figure 1A schematically illustrates the synthesis of CIP@C@LM core-shell particles. CIP (1.0 g) was dispersed in DI water (200 mL) containing glucose (9.0 g) under mechanical stirring (350 rpm) for 12 h. The precursor was magnetically collected, dried at 60 °C for 8 h, and subsequently calcined in air at 300 °C for 2 h (heating rate: 3 °C min<sup>−1</sup>, furnace cooling) to yield CIP@C particles. The obtained powder was resuspended in DI water (200 mL) containing PEG-400 (1 mL) and stirred for 2 h. Separately, LM (0.2 g) was sonicated in DI water (25 mL) at 600 W for 15 min to generate microdroplets. This LM suspension was added to the CIP@C dispersion and mechanically stirred for 8 h. During this process, intermittent ultrasonication (200 W, 30 °C) was applied. The CIP@C@LM sample was prepared using four ultrasonication cycles (15 min per cycle, with 30 min intervals), giving a total ultrasonication duration of 60 min. For comparison, CIP@C@LM1 and CIP@C@LM2 were prepared with total ultrasonication durations of 30 min and 120 min, respectively, under otherwise identical conditions. The final product was magnetically separated and dried at 60 °C for 8 h.

To prepare the coatings, the synthesized powders were blended with a silicate-based high-temperature adhesive (3:2 mass ratio) and homogenized. The slurry was applied via brush-coating, followed by conditioning in a humidity chamber [25 °C, 60% Relative Humidity (RH)] for 24 h. A stepwise thermal curing profile was implemented: 60 °C for 8 h, 80 °C for 6 h, and 100 °C for 2 h.



**Figure 1.** (A) Schematic illustration of the fabrication procedure for CIP@C@LM; (B–E) Representative SEM images tracking the morphological evolution: (B) pristine CIP, (C) CIP@C, and (D and E) the final CIP@C@LM product; (F–K) TEM analysis: (F) low-magnification overview, (G) image of a single particle, (H) magnified views of the shell edge structure, (I) HRTEM image showing lattice fringes (J) magnified view detailing the structural dimensions, and (K) the corresponding SAED pattern; (L–Q) HAADF-STEM image (L) and corresponding EDS elemental mappings of Fe (M), C (N), O (O), Ga (P), and In (Q) acquired from the shell region. CIP: Carbonyl iron powder; LM: liquid metal; SEM: scanning electron microscopy; TEM: transmission electron microscopy; DI: deionized; HRTEM: high-resolution transmission electron microscopy; SAED: selected area electron diffraction; HAADF-STEM: high-angle annular dark-field scanning transmission electron microscopy; EDS: energy-dispersive X-ray spectroscopy.

### Test methods

Morphological characteristics were analyzed using field-emission scanning electron microscopy (SEM, Hitachi S-4800) and transmission electron microscopy (TEM, JEOL JEM-F200). The phase composition was investigated by X-ray diffraction (XRD, Cu  $K\alpha_1$  radiation,  $\lambda = 1.54056 \text{ \AA}$ ) with a  $2\theta$  scanning range from  $5^\circ$  to  $90^\circ$  at a scan rate of  $10^\circ/\text{min}$ . Fourier transform infrared spectroscopy (FTIR,  $4,000\text{--}400 \text{ cm}^{-1}$ ) was conducted using a Bruker Vector33 spectrometer. Raman spectroscopy was performed on a Horiba LabRAM HR Evolution spectrometer with a 532 nm laser. The chemical states of the elements were analyzed by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, USA). Electron paramagnetic resonance (EPR) spectra were recorded using a Bruker EMX-10/12 EPR spectrometer. Magnetic properties were measured at room temperature with a vibrating sample magnetometer (VSM, Model 3105, East Changing Technologies, China). Electromagnetic properties in the frequency range of 2–18 GHz were characterized using a vector network analyzer (VNA, Agilent E5071C) via the coaxial line method. For this, CIP-based powders were mixed with paraffin wax (mass ratio of 3:2) and pressed into toroidal rings (inner diameter: 3.04 mm, outer diameter: 6.95 mm, thickness: 2.00 mm). Electrochemical corrosion and impedance measurements were carried out using an electrochemical workstation (CHI760E, Shanghai Chenhua, China), employing a three-electrode system in a 3.5 wt.% NaCl solution. The working electrode was a glassy carbon

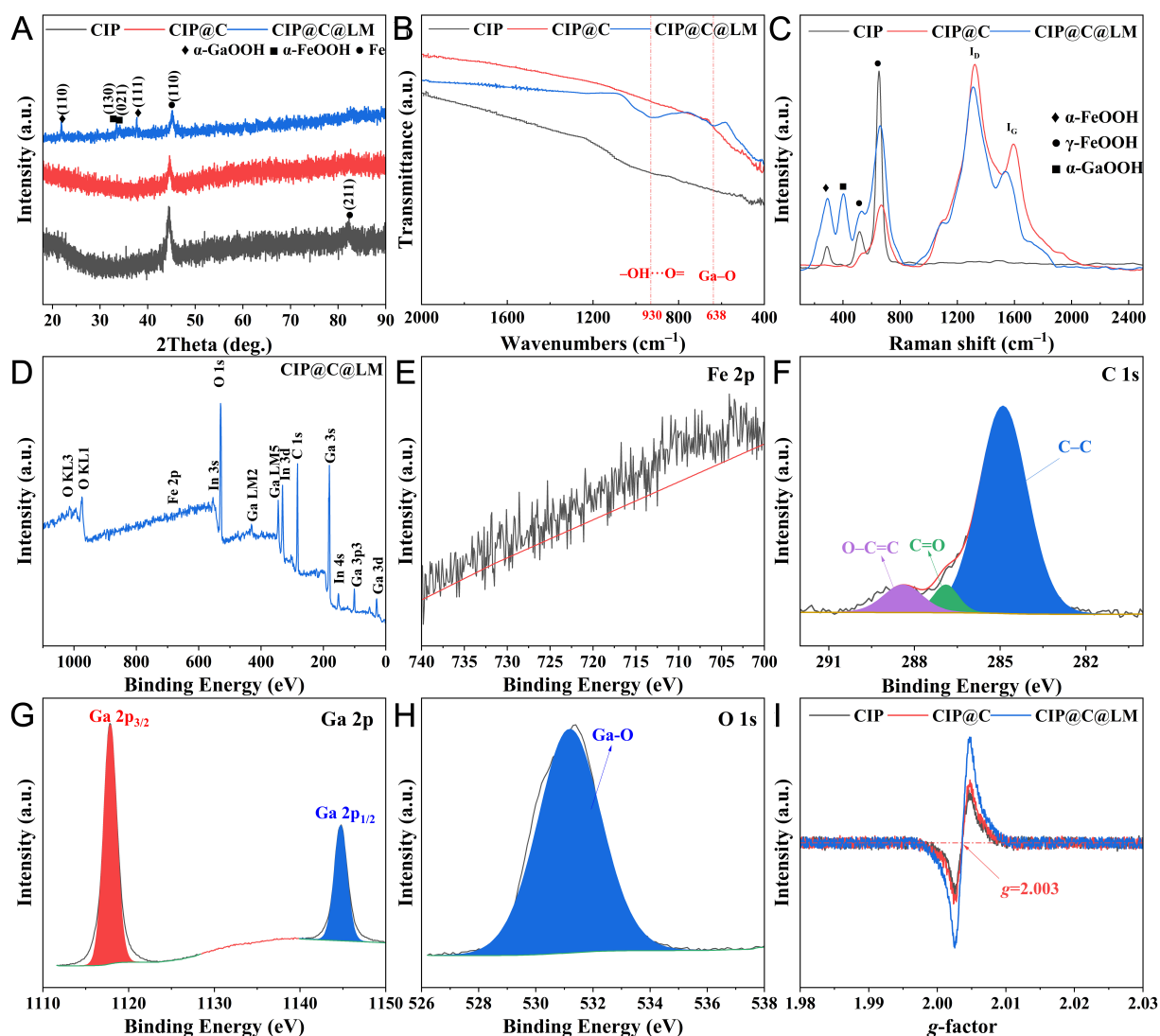
electrode, with an Ag/AgCl reference electrode and a platinum mesh counter electrode. Tafel polarization curves were recorded within a voltage range of -0.5 V to 0.5 V vs. Open circuit potential (OCP), at a scan rate of 5 mV/s. Thermal infrared radiation intensity of coatings was assessed using a thermal infrared imaging device (KREBOR FLIR ONE3, FLIR Systems, Inc., USA).

## RESULTS AND DISCUSSION

### Morphology and structural characterizations

Figure 1B–Q track the morphological evolution and interface assembly of the CIP@C@LM heterostructure. The pristine CIP [Figure 1B] exhibits a characteristic spherical geometry with relatively smooth surfaces. Upon C encapsulation, a textured, conformal coating emerges [Figure 1C], confirming the successful deposition of the C interlayer, which serves as a buffer against core oxidation. Subsequently, the introduction of LM via ultrasonication triggers a dramatic morphological transformation, yielding a well-defined core-shell architecture enveloped by a dense array of nanorod (needle-like) nanostructures [Figure 1D]. This hierarchical growth leads to a discernible increase in particle size, with the size distribution shifting notably toward the 2–3  $\mu\text{m}$  range [Supplementary Figure 1]. High-magnification SEM images [Figure 1E] reveal that the surface nanorods are self-assembled into rhomboidal patterns, uniformly distributed across the particle surface with discernible interparticle gaps. TEM analysis [Figure 1F–H] provides deeper insight into this microstructure, clearly resolving the radially oriented, rod-like crystallites that constitute the outer shell. Statistical analysis indicates that these crystallites possess an average length of  $\sim 62$  nm [Supplementary Figure 2]. The crystalline nature of the shell is corroborated by high-resolution transmission electron microscopy (HRTEM) [Figure 1I], where clear lattice fringes with  $d$ -spacings of 2.407 Å and 2.645 Å are observed. These correspond well to the (111) and (301) planes of orthorhombic  $\alpha$ -GaOOH<sup>[36]</sup>. Additionally, the precise dimensions of these nanostructures are detailed in the magnified view [Figure 1J]. Acquired from this specific region, the corresponding selected area electron diffraction (SAED) [Figure 1K] shows distinct polycrystalline diffraction rings. The calculated  $d$ -spacings from these concentric rings are 2.645, 2.407, 2.259, 1.575, and 1.489 Å, which are indexed to the (301), (111), (002), (312), and (103) planes of the standard  $\alpha$ -GaOOH phase (JCPDS PDF No. 54-0910)<sup>[37]</sup>, respectively. To elucidate the elemental distribution and interfacial chemistry, high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and energy-dispersive X-ray spectroscopy (EDS) mappings were performed [Figure 1L–Q]. The images distinctly delineate a sandwich-like structure: the Fe-rich core, the C interlayer, and the Ga/O-dominated outer shell. Notably, an O-enriched region is observed overlapping with the Fe/C interface, implying that partial surface oxidation of CIP occurred during the aqueous processing. Crucially, while Ga and O signals are strongly colocalized in the outer shell, the In signal is remarkably weak. This phenomenon can be attributed to the thermodynamically driven selective oxidation of the EGaIn alloy. Since Ga has a greater tendency to undergo oxidation than In<sup>[38]</sup>, it preferentially migrates to the surface to undergo hydrolysis and oxidation, forming the voluminous  $\alpha$ -GaOOH framework. The unoxidized In segregates into isolated metallic nanodroplets, which are subsequently removed by intense ultrasonic shear and washing processes, resulting in a diminished In signal in the final powder. Collectively, these microstructural and compositional analyses confirm the successful construction of a hierarchical magnetic-dielectric heterostructure, where the abundant heterogeneous interfaces and unique acicular shell are anticipated to play a critical role in optimizing electromagnetic response.

Figure 2 summarizes the phase composition and defect chemistry of CIP, CIP@C, and CIP@C@LM. The XRD patterns [Figure 2A] show that CIP exhibits the characteristic peaks of metallic Fe (body-centered cubic, bcc) at  $2\theta = 44.7^\circ$  and  $82.3^\circ$ , indexed to the (110) and (211) planes (JCPDS PDF No. 06-0696)<sup>[39]</sup>. After C coating, the diffraction features become noticeably weakened, and the Fe (211) reflection at  $82.33^\circ$  is strongly suppressed, indicating a reduced crystallinity. Upon subsequent LM-assisted treatment, distinct new peaks emerge at  $2\theta = 21.5^\circ$  and  $37.2^\circ$ , assignable to orthorhombic  $\alpha$ -GaOOH (110) and (111) (JCPDS



**Figure 2.** Characterization of phase composition and chemical states. (A) XRD patterns, (B) FTIR spectra, and (C) Raman spectra of CIP, CIP@C, and CIP@C@LM; (D-H) XPS analysis of the CIP@C@LM composite: (D) survey spectrum; (E-H) high-resolution spectra for Fe 2p, C 1s, Ga 2p, and O 1s, respectively; (I) Room-temperature EPR spectra of the samples showing the defect intensity variations. CIP: Carbonyl iron powder; XRD: X-ray diffraction; FTIR: fourier transform infrared spectroscopy; LM: liquid metal; EPR: electron paramagnetic resonance.

No.54-0910)<sup>[40]</sup>. In addition, minor peaks at  $2\theta = 33.5^\circ$  and  $34.1^\circ$  are detected, which match  $\alpha$ -FeOOH (130) and (021) (JCPDS PDF No. 29-0713)<sup>[41]</sup>. Combined with the nanorod shell morphology as shown in Figure 1M-P, these results indicate that aqueous ultrasonication drives  $\alpha$ -GaOOH growth while partially converting the Fe surface into  $\alpha$ -FeOOH. FTIR spectroscopy [Figure 2B] further supports the formation of the oxyhydroxides. The bands at  $930\text{ cm}^{-1}$  and  $638\text{ cm}^{-1}$  are attributed to the bending vibrations of  $-\text{OH}\cdots\text{O}=\text{}$ <sup>[42]</sup> and the stretching vibrations of  $\text{Ga}-\text{O}$ <sup>[40]</sup>, respectively, corroborating the presence of  $\alpha$ -GaOOH. Raman spectra [Figure 2C] further probe the surface oxyhydroxide species and the C shell. For pristine CIP, the distinct Raman scattering signals detected at  $\sim 286\text{ cm}^{-1}$  verify the existence of  $\alpha$ -FeOOH<sup>[43]</sup>, and the signals at  $\sim 528\text{ cm}^{-1}$  and  $\sim 650\text{ cm}^{-1}$  corresponded to  $\gamma$ -FeOOH phase<sup>[44]</sup>. The absence of  $\gamma$ -FeOOH peaks in XRD suggests a poorly crystalline and/or ultrathin surface layer. After C coating, the FeOOH-related peaks are markedly suppressed, while the carbon D and G bands emerge at  $\sim 1,330\text{ cm}^{-1}$  and  $\sim 1,590\text{ cm}^{-1}$ <sup>[45]</sup>, confirming C encapsulation. Following LM treatment, the  $\alpha$ -FeOOH band strengthens and slightly upshifts, whereas the  $\gamma$ -FeOOH features weaken, indicating  $\alpha$ -FeOOH enrichment near the interface. A new peak at  $\sim 394\text{ cm}^{-1}$  appears, consistent with the  $A_g$  mode of  $\alpha$ -GaOOH<sup>[41]</sup>. Furthermore, the redshifted D and G bands indicate increased defect density and interfacial charge transfer within the heterostructure.

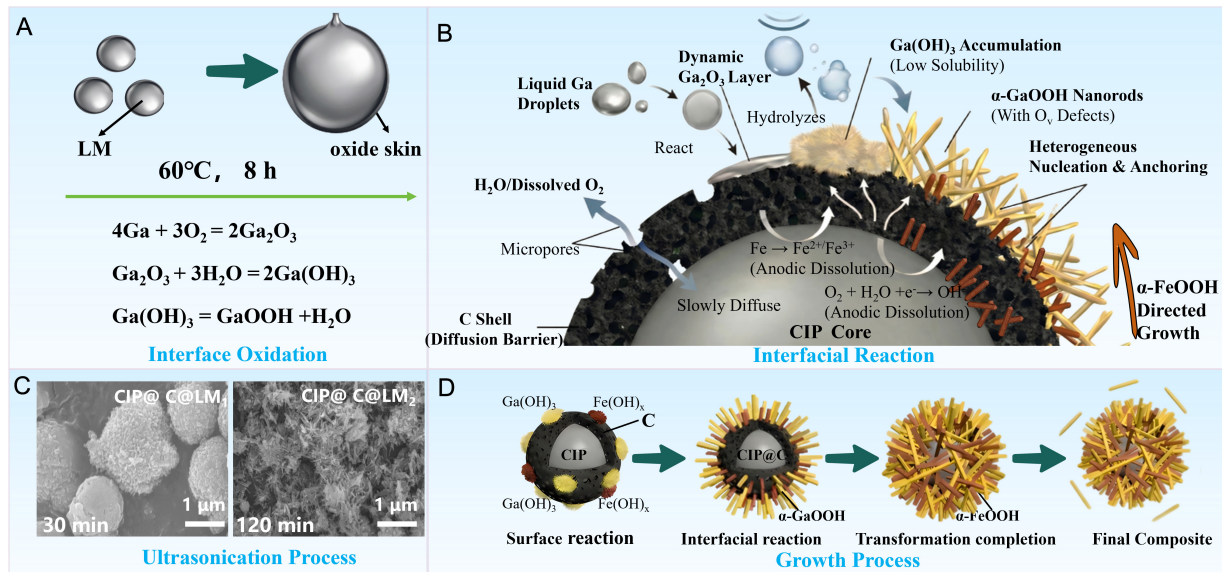


As shown in Figure 2D–H, the XPS results further confirm the surface composition and bonding environment of the synthesized composites. Figure 2D clearly identifies the presence of C, Ga, In, and O signals in CIP@C@LM. The Fe 2p region [Figure 2E] shows remarkably weakened and less-resolved features. This attenuation is anticipated due to the limited escape depth of photoelectrons. In the C 1s spectrum [Figure 2F], peaks deconvoluted at 284.9, 286.9, and 288.4 eV correspond to C–C, C=O, and O–C=O species<sup>[46]</sup>, respectively. The Ga 2p spectrum [Figure 2G] exhibits characteristic spin-orbit doublets components at 1,117.7 eV (Ga 2p<sub>3/2</sub>) and 1,144.6 eV (Ga 2p<sub>1/2</sub>)<sup>[47]</sup>, with a binding energy separation of ~26.9 eV, indicative of the Ga<sup>3+</sup> oxidation state in the  $\alpha$ -GaOOH lattice. In the O 1s spectrum [Figure 2H], the dominant peak at 531.2 eV is characteristic of Ga–O lattice oxygen<sup>[48]</sup>, indicating that the outer surface is dominated by Ga-based oxide/oxyhydroxide species formed during the sonochemical hydrolysis. To assess defect states and unpaired electrons, EPR measurements were performed [Figure 2I]. A distinct resonance signal at g-factor (g) = 2.003, characteristic of O<sub>v</sub> trapped with unpaired electrons<sup>[49]</sup>, is detected. Pristine CIP shows a weak O<sub>v</sub> response, consistent with the presence of a thin native oxidized surface layer (e.g., FeOOH). C coating produces negligible change. However, LM treatment leads to a pronounced increase in the O<sub>v</sub> signal intensity. This surge is consistent with the formation of oxyhydroxides ( $\alpha$ -FeOOH and  $\alpha$ -GaOOH) enriched with defect sites, likely originating from the rapid, non-equilibrium crystal growth driven by the high-energy sonochemical/aqueous conditions. Collectively, the XRD, FTIR, Raman, XPS, and EPR results confirm the coexistence of  $\alpha$ -GaOOH and interfacial  $\alpha$ -FeOOH. Furthermore, they demonstrate the defect-rich nature of the shell/interface<sup>[50]</sup>.

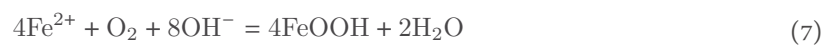
### Dual-hydroxide growth mechanism

As shown in Figure 3, the concurrent growth of nanorods  $\alpha$ -GaOOH/ $\alpha$ -FeOOH shells can be attributed to interfacial oxidation, hydrolysis, and heterogeneous nucleation. First, regarding the Ga evolution [Figure 3A]: Upon ultrasonic dispersion, the liquid Ga droplets undergo interfacial oxidation to form a dynamic Ga<sub>2</sub>O<sub>3</sub> skin (Equation 1). The high-intensity acoustic cavitation continuously renews the gas-liquid and liquid-solid interfaces, accelerating mass transport and disrupting the oxide skin. This promotes the hydration of Ga<sub>2</sub>O<sub>3</sub> into low-solubility Ga(OH)<sub>3</sub> (Equation 2)<sup>[51]</sup>, which subsequently converts to  $\alpha$ -GaOOH via a dissolution-recrystallization and dehydration process<sup>[52]</sup> (Equation 3). The cavitation environment also introduces abundant defect sites (e.g., O<sub>v</sub>) and active hydroxyl groups on the Ga-based surface. Simultaneously, the Fe pathway is modulated by the C shell. The micropores and defects in the C coating act as diffusion valves, allowing limited access of H<sub>2</sub>O and dissolved O<sub>2</sub> to the CIP surface [Figure 3B]. This restricts the anodic dissolution of Fe<sup>0</sup> (Fe<sup>0</sup> → Fe<sup>2+</sup>/Fe<sup>3+</sup>) (Equation 4) to a rate-controlled regime, preventing the structural collapse of the magnetic core [Supplementary Figure 3]. Coupled with the cathodic reduction of dissolved oxygen, hydroxide ions (OH<sup>-</sup>) are generated (Equation 5), establishing a localized alkaline microenvironment that facilitates the hydrolysis of Fe ions into FeOOH intermediates (Equations 6–7). Crucially, the synergistic growth is driven by structural and energetic compatibility. Since both  $\alpha$ -GaOOH and  $\alpha$ -FeOOH crystallize in the isostructural diasporite/goethite framework<sup>[53]</sup> (built from edge-sharing octahedral chains), the in-situ formed Ga-based species serve as ideal templates. The Ga<sub>2</sub>O<sub>3</sub>/Ga(OH)<sub>3</sub> interphase not only lowers the interfacial energy but also provides a hydrophilic, hydroxylated surface that facilitates the heterogeneous nucleation of Fe(OH)<sub>x</sub>. Furthermore, the defect sites (O<sub>v</sub>) serve as active anchoring centers, lowering the nucleation barrier and promoting the epitaxial-like assembly of  $\alpha$ -FeOOH along the  $\alpha$ -GaOOH lattice. However, this self-assembly process is strictly governed by reaction kinetics. As schematically illustrated in Figure 3C, the morphological evolution is highly dependent on the ultrasonication duration: sparse initial growth is observed at 30 min, which develops into dense, ordered nanorod arrays at 60 min [Figure 1D and E], while prolonged sonication to 120 min leads to disordered and detached nanostructures. Correspondingly, increasing the ultrasonication duration induces a morphological transition from ordered nanorod arrays to disordered textures, eventually leading to discrete, detached nanostructures [Figure 3D].





**Figure 3.** Proposed growth mechanism of the CIP@C@LM heterostructure. (A) The Ga evolution pathway via ultrasonic cavitation-driven interfacial oxidation and hydrolysis; (B) The Fe evolution pathway modulated by the intermediate C shell and the subsequent synergistic heterogeneous nucleation; (C) SEM images displaying the time-dependent microscopic morphological evolution of the composite at ultrasonication durations of 30 and 120 min; (D) Schematic diagram illustrating the four-stage morphological evolution of the composite shell. CIP: Carbonyl iron powder; LM: liquid metal; SEM: scanning electron microscopy.



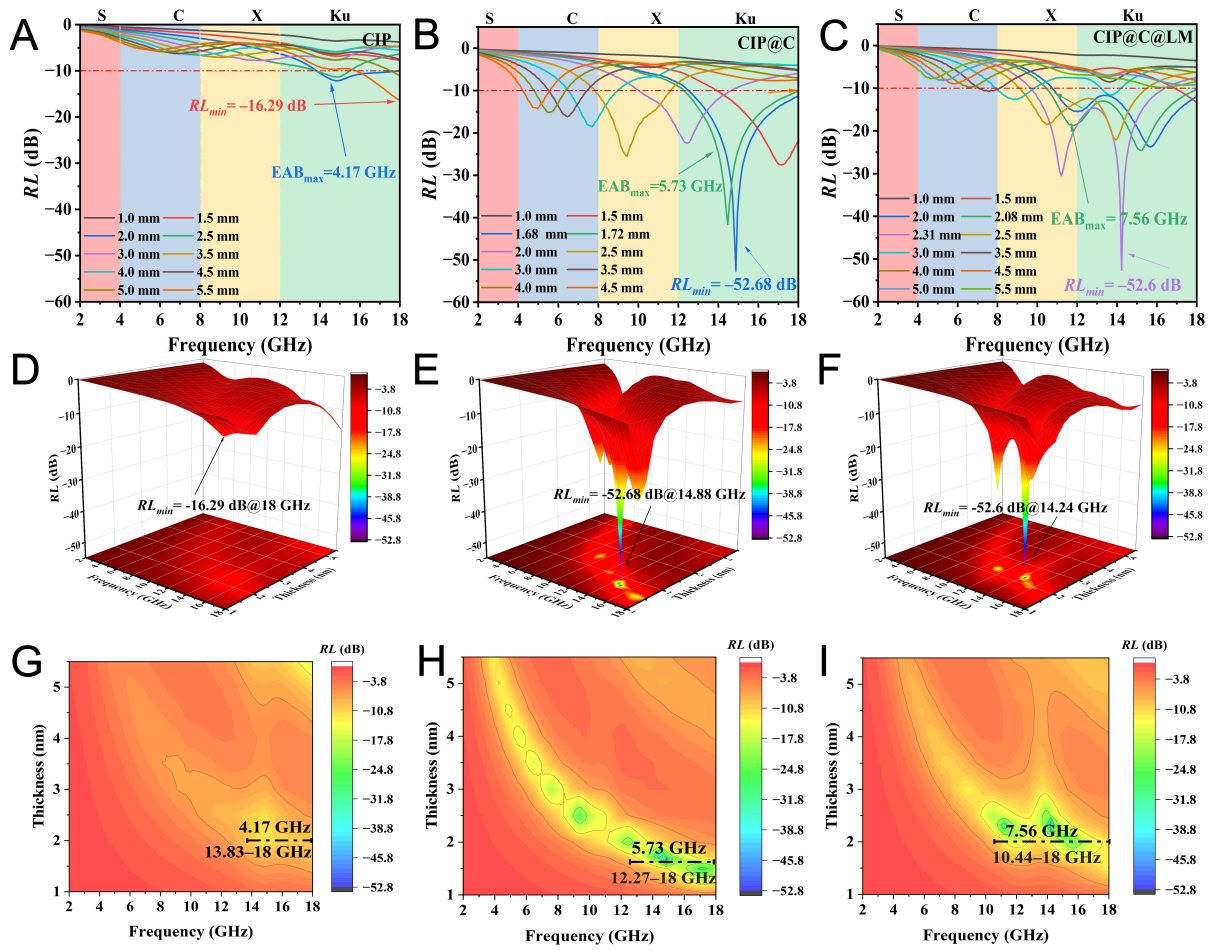
### EMW absorption performance

To characterize the EMW absorption performance,  $RL$  and impedance matching are generally calculated based on experimentally measured  $f$  dependent electromagnetic parameters. According to the transmission line theory, the relevant calculation formulas are expressed as follows<sup>[54]</sup>:

$$RL = 20 \lg \left| \frac{Z - 1}{Z + 1} \right| \quad (8)$$

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

Here,  $Z$  represents the normalized input impedance relative to free space,  $Z_{in}$  is the input impedance of the material,  $Z_0$  is the characteristic impedance of free space,  $d$  is the absorber thickness, and  $\epsilon_r$  ( $\epsilon_r = \epsilon' - j\epsilon''$ ) and  $\mu_r$  ( $\mu_r = \mu' - j\mu''$ ) are the complex permittivity and permeability. Figure 4 compares the EMW absorption performance of CIP, CIP@C and CIP@C@LM. For pristine CIP, the EMW absorption remains modest, with an  $RL_{\min}$  of -16.29 dB at 18 GHz ( $d = 5.5$  mm) and an  $EAB_{\max}$  of 4.17 GHz (13.83–18 GHz) at a  $d$  of 2.0 mm.

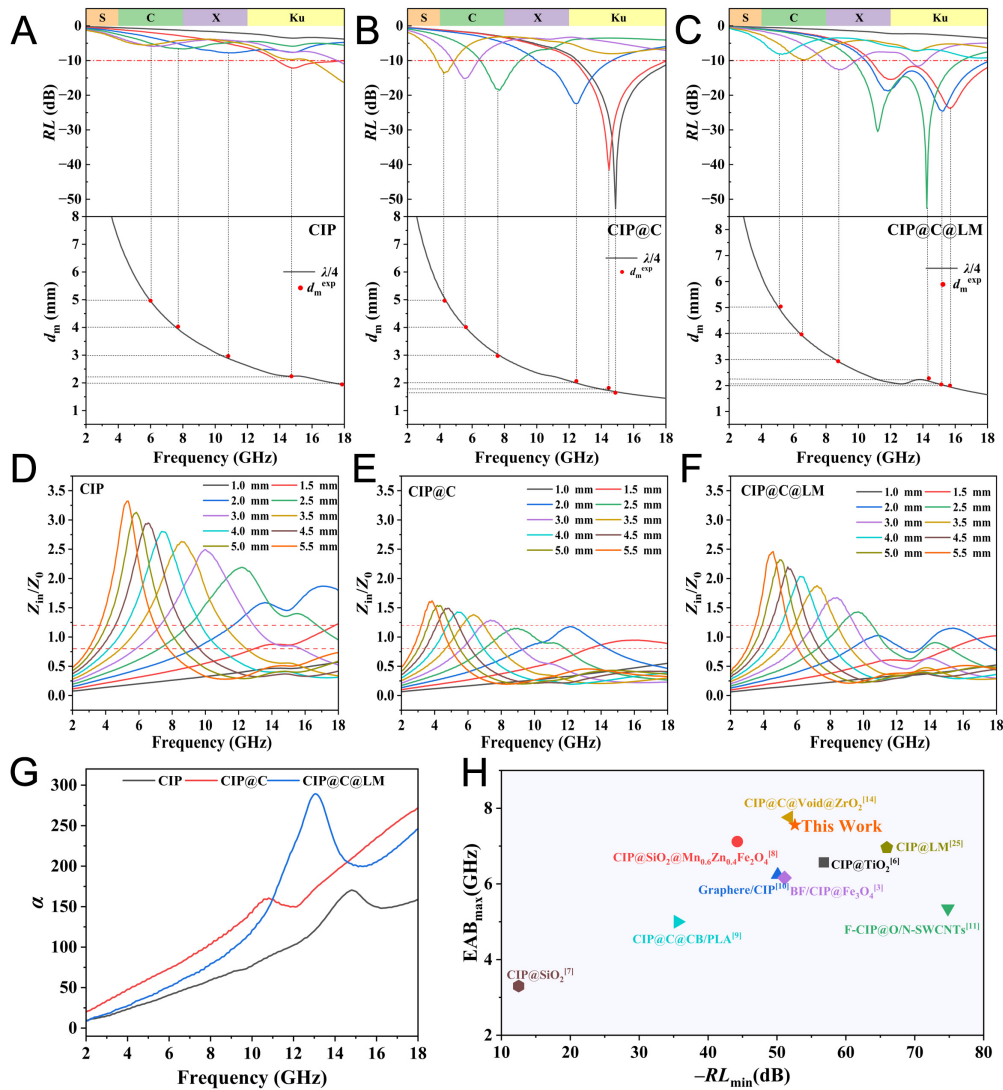


**Figure 4.** (A–C) Frequency ( $f$ ) dependent reflection loss (RL) and effective absorption bandwidth (EAB) at the different thickness of CIP, CIP@C, and CIP@C@LM; (D–F) Three-dimensional (3D) RL plots; (G–I) Two-dimensional (2D) projection plots of RL and  $EAB_{max}$ . CIP: Carbonyl iron powder; LM: liquid metal;  $EAB_{max}$ : maximum effective absorption bandwidth.

This limited performance is attributable to the low dielectric response of the CIP/paraffin composites at a filler ratio of 60 wt.%. The C coating significantly improves the attenuation capability, with CIP@C delivering an  $RL_{min}$  of -52.68 dB at 14.88 GHz ( $d = 1.68$  mm). More importantly, after the LM-assisted conversion, the final CIP@C@LM heterostructure achieves an  $RL_{min}$  of -52.60 dB at 14.24 GHz ( $d = 2.31$  mm), together with an  $EAB_{max}$  of 7.56 GHz (10.44–18 GHz) at a  $d$  of 2.08 mm. The EMW absorption maps of CIP@C@LM show a double-peak feature, indicating the concurrent contributions of multiple loss processes. Furthermore, EMW absorption performance deteriorates when the ultrasonication time is either insufficient or excessive, which can be attributed to inappropriate microstructures as shown in Figure 3C. When the treatment time is too short, it is difficult to form effective heterointerfaces for interfacial polarization. Meanwhile, too long treatment generates redundant non-conductive phases and lowers electrical conductivity [Supplementary Figure 4]. The corresponding electromagnetic-parameter comparison further confirms that the optimized oxidation state at 60 min results in dielectric-loss-dominated attenuation [Supplementary Figure 5].

Overall, as shown in Figure 5A–C, the EMW absorption maxima of all samples follow the quarter-wavelength ( $l/4$ ) cancellation principle. When the absorber thickness satisfies<sup>[55]</sup>:

$$d_m = \frac{n\lambda}{4} = \frac{nc}{4f_m\sqrt{|\epsilon_r\mu_r|}} \quad (n = 1, 3, 5 \dots) \quad (10)$$



**Figure 5.** Correlation between reflection loss (RL) and the quarter-wavelength ( $\lambda/4$ ) matching condition for (A) CIP, (B) CIP@C, and (C) CIP@C@LM. Normalized input impedance (Z) maps of (D) CIP, (E) CIP@C, and (F) CIP@C@LM; (G) Attenuation constant ( $\alpha$ ) of the three composites; (H) Performance benchmarking of representative CIP-based absorbers reported recently in terms of  $RL_{\min}$  and  $EAB_{\max}$ . CIP: Carbonyl iron powder; LM: liquid metal;  $EAB_{\max}$ : maximum effective absorption bandwidth;  $RL_{\min}$ : minimum reflection loss.

where  $d_m$  is the matching thickness of  $RL_{\min}$ ,  $c$  is the velocity of light in a vacuum,  $n$  is the odd-order matching number and  $f_m$  is the matching frequency corresponding to the absorption peak. The reflected wave at the absorber/air interface is approximately out of phase with the incident wave, leading to destructive interference and a pronounced absorption peak. Accordingly, to achieve effective EMW absorption at lower  $f_m$ , a larger  $d_m$  is generally required to satisfy the phase cancellation condition. This thickness- $f$  correlation also indicates that the absorption behavior of the system is governed not only by intrinsic dielectric/magnetic losses, but also by the combined effects of phase delay during wave propagation and impedance matching. As the  $d_m$  increases, the effective propagation path lengthens and multiple reflections/interference become more prominent, driving the peak to lower  $f$  and, in some cases, giving rise to secondary peaks. Notably, CIP@C@LM exhibits dual absorption peaks at a fixed  $d_m$ , suggesting that the material simultaneously satisfies the phase cancellation condition at different orders (e.g.,  $n = 1$  and  $n = 3$ ), resulting in multi-order matching. In addition, concurrent intrinsic loss processes, such as polarization relaxation and magnetic resonance, further enable different  $f$  windows to reach more favorable impedance matching and energy dissipation<sup>[56]</sup>. Meanwhile, impedance matching is a prerequisite for efficient EMW absorption because it determines the

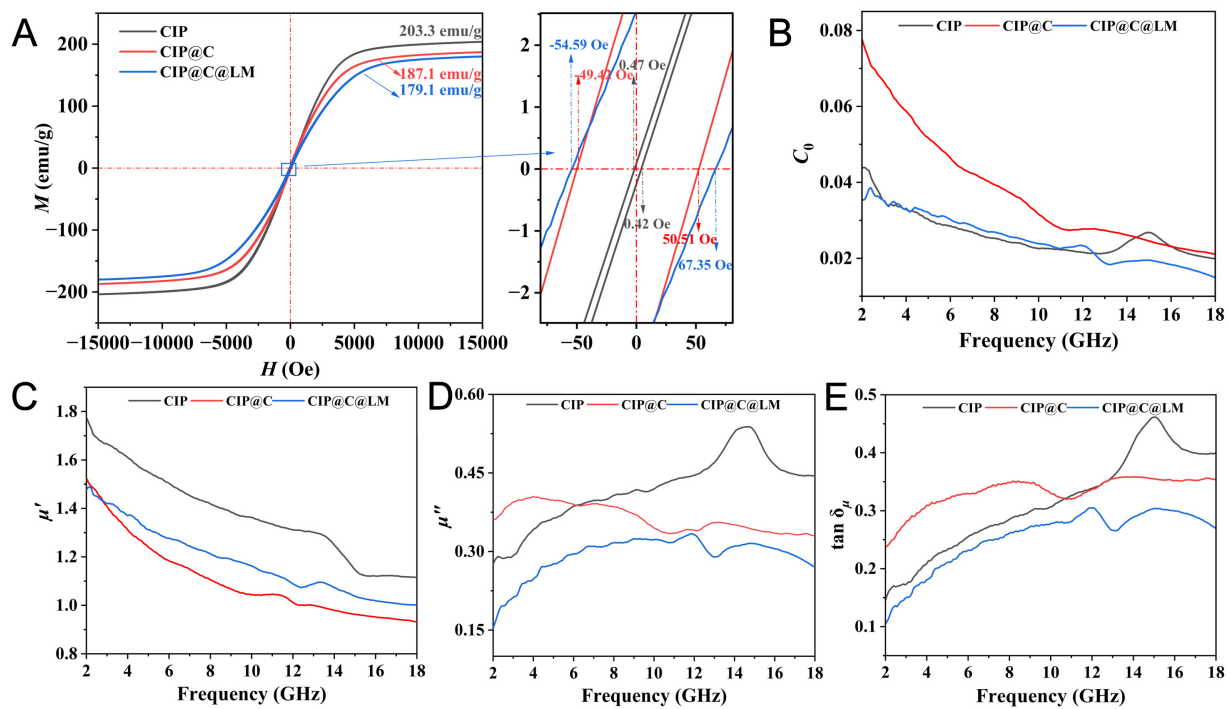
fraction of incident electromagnetic energy that enters the absorber rather than being reflected at the surface. When the normalized input impedance  $Z$  approaches 1, more EMW can penetrate the material; in practice, the region  $0.8 < Z < 1.2$  is commonly regarded as indicative of good matching<sup>[57]</sup>. As shown in Figure 5D–F, the normalized input impedance curves reveal a frequency-dependent matching behavior after heterostructure construction. Compared with CIP@C, CIP@C@LM exhibits somewhat weakened impedance matching in the low-frequency region, whereas more favorable matching behavior is observed in the middle and high frequency regions, particularly within its main EAB of 10.44–18 GHz [Supplementary Table 1]. This improvement is mainly attributed to the  $\alpha$ -GaOOH-rich shell, which reduces excessive dielectric storage and alleviates the impedance mismatch caused by overly high  $\epsilon'$  in the middle and high frequency regions. Meanwhile, the introduced heterogeneous interfaces, hydroxyl-related dipoles, and defect-associated polarization centers maintain sufficient dielectric loss, thereby enabling a better balance between electromagnetic-wave entry and internal dissipation in the relevant absorption region. Beyond wave entry, EMW absorption fundamentally relies on the dissipation of the transmitted energy through dielectric and magnetic losses, ultimately converting EMW energy into heat. A key descriptor of dissipation strength is the attenuation constant  $\alpha$ , which quantifies the exponential decay rate of the wave amplitude during propagation in the medium. Physically,  $\alpha$  is closely associated with intrinsic loss terms and is further amplified by multiple reflections and an extended effective propagation path within the absorber, as expressed by<sup>[58]</sup>:

$$\alpha = \frac{\sqrt{2}\pi f}{c} \times \sqrt{(\mu''\epsilon'' - \mu'\epsilon') + \sqrt{(\mu''\epsilon'' - \mu'\epsilon')^2 + (\mu'\epsilon'' - \mu''\epsilon')^2}} \quad (11)$$

As shown in Figure 5G,  $\alpha$  differs substantially among the three materials. CIP exhibits the lowest  $\alpha$  across the measured range, indicating the weakest overall dissipation capability. In the 11–15 GHz region,  $\alpha$  of CIP@C@LM exceeds that of CIP@C, demonstrating a stronger attenuation ability in this band, consistent with its improved absorption response. To benchmark the performance of CIP@C@LM, we further compare it with representative CIP-based composites reported recently<sup>[3,6,7,8,9,11,14,25]</sup> [Figure 5H]. In comparison, despite the simpler structural design and the relatively low filler loading (60 wt.%), our CIP@C@LM still delivers a competitive EAB<sub>max</sub> of 7.56 GHz, underscoring the effectiveness of the interfacial architecture in balancing impedance matching and attenuation. For a normalized evaluation of the bandwidth performance, the fractional bandwidth is calculated as  $\text{FBW} = (f_H - f_L) / [(f_H + f_L)/2] \times 100\%$ , where  $f_H$  and  $f_L$  represent the upper and lower frequencies of the effective absorption bandwidth, respectively. Based on the EABmax ranges shown in Figure 4G–I, the FBW values of CIP, CIP@C, and CIP@C@LM are 26.2%, 37.9%, and 53.2%, respectively. Notably, CIP@C@LM achieves the highest FBW of 53.2% over 10.44–18.00 GHz, confirming its substantially broadened effective absorption coverage after heterostructure construction. Loading-dependent measurements confirm that, in the present system, strong and broadband absorption is achieved at 60 wt.% [Supplementary Figure 6].

The EMW absorption behavior is intrinsically governed by the dielectric and magnetic loss capabilities, as detailed in Figures 6 and 7. As shown in Figure 6A, VSM measurements reveal high  $M_s$  for all samples. As expected, introducing non-magnetic shells progressively dilutes the magnetic phase, leading to a stepwise decrease in  $M_s$  from 203.3 emu/g (CIP) to 187.1 emu/g (CIP@C) and 179.1 emu/g (CIP@C@LM). The corresponding coercivities ( $H_c$ ) are 0.29, 49.96, and 60.97 Oe, respectively. The increase in  $H_c$  indicates enhanced resistance to magnetization reversal after interfacial reconstruction. Although  $\alpha$ -FeOOH is present only as a minor interfacial phase, its formation at the CIP surface introduces an additional magnetic component into the heterogeneous interface, thereby enriching the local magnetic structure of CIP@C@LM<sup>[59–61]</sup>. To assess the magnetic loss contribution, the eddy-current loss coefficient ( $C_o$ ) is

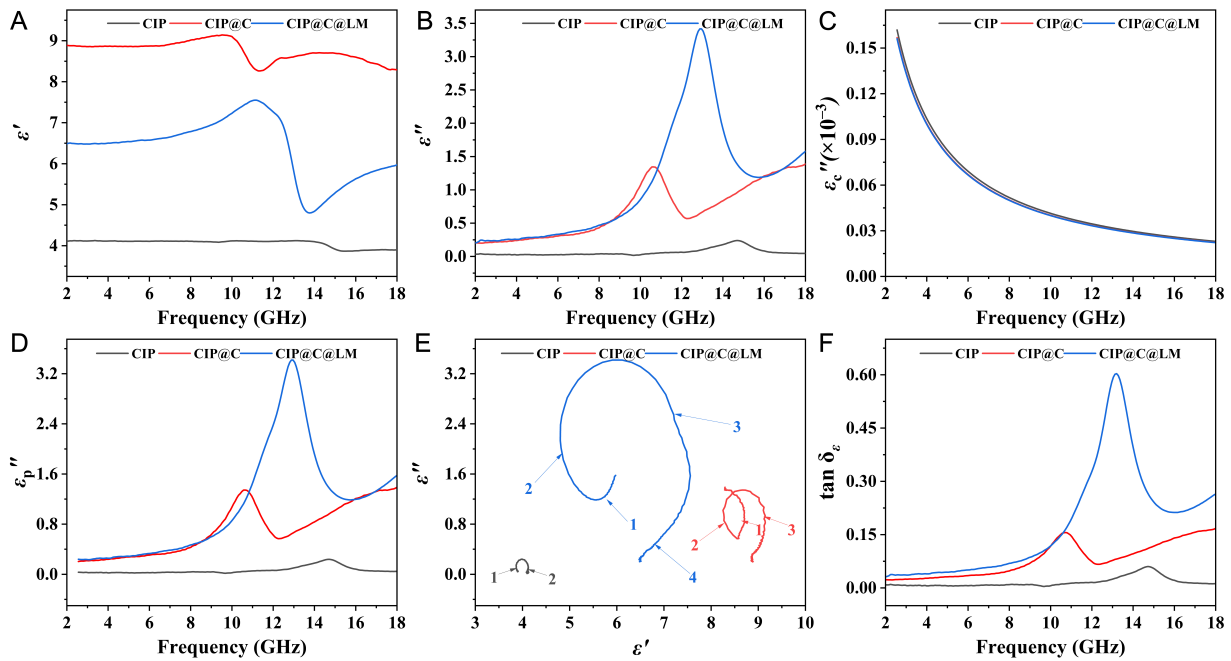




**Figure 6.** (A) Magnetic hysteresis loops; (B)  $C_0$  value (C) Real part of permeability ( $\mu'$ ); (D) Imaginary part of permeability ( $\mu''$ ); (E) Magnetic loss tangent ( $\tan \delta_\mu$ ). CIP: Carbonyl iron powder; LM: liquid metal.

commonly used and defined as  $C_0 = \mu''(\mu')^{-2}f^{-1}$  [62], where  $\mu'$  and  $\mu''$  are the real and imaginary parts of the relative complex permeability, respectively. When  $C_0$  remains nearly constant with  $f$ ,  $C_0$  is considered dominant. As shown in Figure 6B, magnetic loss in the 2–18 GHz range is mainly associated with eddy-current loss and natural resonance in pristine CIP. For CIP@C@LM,  $C_0$  shows only minor variation as  $f$  increases, suggesting that eddy-current loss governs the magnetic dissipation, while a weak resonance feature appears in the 10–13 GHz range, indicative of a relatively modest natural resonance contribution. The  $f$ -dependent  $\mu_r$  further supports these interpretations. In Figure 6C,  $\mu'$  follows the order CIP > CIP@C@LM > CIP@C. The decrease in  $\mu'$  after C encapsulation originates from the dilution of the magnetic CIP phase, whereas the higher  $\mu'$  value of CIP@C@LM relative to CIP@C indicates that the  $\alpha$ -FeOOH-containing heterogeneous interface contributes to the enhanced effective permeability response after LM-assisted interfacial reconstruction. The  $\mu''$  spectra [Figure 6D] show that CIP@C exhibits the highest  $\mu''$  in the 2–6 GHz region, whereas CIP dominates in the 6–18 GHz range, confirming that pristine CIP retains the strongest intrinsic magnetic loss at higher  $f$ . However, despite its favorable magnetic loss, CIP alone still delivers limited EMW absorption because the composite suffers from insufficient dielectric loss and poor impedance matching under the given loading, highlighting the need to enhance the dielectric contribution. Consistently, the magnetic loss tangent ( $\tan \delta_\mu = 0.24 \sim 0.35$ , Figure 6E) of CIP@C remains relatively high below  $\sim 13$  GHz, while CIP shows the largest  $\tan \delta_\mu$  at 13–18 GHz, reflecting the high- $f$  nature of its dominant resonance-related losses. For CIP@C@LM, the frequency-dependent  $\mu''$  and  $\tan \delta_\mu$  responses reflect the modified permeability behavior and retained magnetic-loss contribution after interfacial reconstruction. Together with its higher  $\mu'$  than CIP@C, these results indicate that the minor interfacial  $\alpha$ -FeOOH enriches the local magnetic structure and contributes to the magnetic response of CIP@C@LM through the heterogeneous interface.

As for the dielectric loss contribution, the pristine CIP exhibits the lowest  $\epsilon'$  at a filler loading of 60 wt.%, reflecting its weak dielectric contribution and the difficulty in establishing an effective polarization/conduction response in the paraffin matrix as shown in Figure 7A. After introducing the C



**Figure 7.** (A) Real part of complex permittivity ( $\epsilon'$ ); (B) Imaginary part of complex permittivity ( $\epsilon''$ ); (C) Conduction-loss component ( $\epsilon_c''$ ); (D) Polarization-loss component ( $\epsilon_p''$ ); (E) Cole-Cole plots; (F) Dielectric loss tangent ( $\tan \delta_\epsilon$ ). CIP: Carbonyl iron powder; LM: liquid metal.

interlayer, the real part of complex permittivity ( $\epsilon'$ ) increases markedly, indicating enhanced charge-storage capability and stronger interfacial/electronic polarization. Upon further formation of  $\alpha$ -GaOOH,  $\epsilon'$  decreases to some extent, suggesting that the outer oxidized/hydroxylated shell partially suppresses the overall dielectric response by reducing the effective conductivity. The evolution of imaginary part of complex permittivity ( $\epsilon''$ ) [Figure 7B] more directly reflects dielectric dissipation. CIP shows the smallest  $\epsilon''$ , indicating negligible dielectric loss. In contrast, CIP@C exhibits a pronounced enhancement in  $\epsilon''$  and presents a clear relaxation feature in the 9–13 GHz range, implying activation of polarization relaxation processes after C encapsulation. Notably, CIP@C@LM displays a further, substantial increase in  $\epsilon''$ , with the most prominent relaxation peak located at 11–15 GHz and reaching a maximum value of 3.5. This enhanced dielectric dissipation is mainly associated with the  $\alpha$ -GaOOH-rich nanorod shell, which increases the heterogeneous interfacial area and introduces polar Ga–O/OH environments, thereby promoting Maxwell–Wagner interfacial polarization<sup>[63]</sup>. Under an alternating electromagnetic field, charge carriers accumulate at heterogeneous boundaries due to contrasts in conductivity and permittivity, leading to dynamic charge redistribution and polarization relaxation; the associated lag of polarization behind the field converts electromagnetic energy into Joule heating, thereby enhancing attenuation and suppressing reflection and secondary interference. To further deconvolute the dielectric-loss origins,  $\epsilon''$  was separated into the conduction-loss term ( $\epsilon_c''$ ) and polarization-loss term ( $\epsilon_p''$ ) based on the Debye relaxation framework (method described in [Supplementary Section 8](#)). As shown in Figure 7C, all three samples exhibit relatively weak  $\epsilon_c''$ , implying that long-range charge transport is not the dominant dissipation pathway. This observation aligns with the largely amorphous and discontinuous nature of the C layer, as well as the insulating effect of the  $\alpha$ -GaOOH-rich shell, which further suppresses electrical conductivity. In contrast, the trend of  $\epsilon_p''$  [Figure 7D] closely follows that of  $\epsilon''$  for CIP@C and CIP@C@LM, indicating that polarization-dominated loss is the primary contributor. Here, interfacial polarization mainly arises from the effective dipole layers formed at boundaries with large contrasts in permittivity/conductivity. Meanwhile, oxygen-containing functional groups [Figure 2B] and  $O_v$ -related defect dipoles [Figure 2I] within the  $\alpha$ -GaOOH/ $\alpha$ -FeOOH-containing shell/interface introduce localized dipole moments and provide additional pathways for

defect-induced polarization relaxation. Although the reduced  $\varepsilon'$  leads to slightly weakened matching at low frequencies, it improves the impedance balance in the middle and high frequency absorption region. Together with the enhanced  $\varepsilon''$  and  $\varepsilon_p''$ , this result indicates that the  $\alpha$ -GaOOH-rich shell enables favorable wave entry while retaining strong polarization-dominated dissipation.

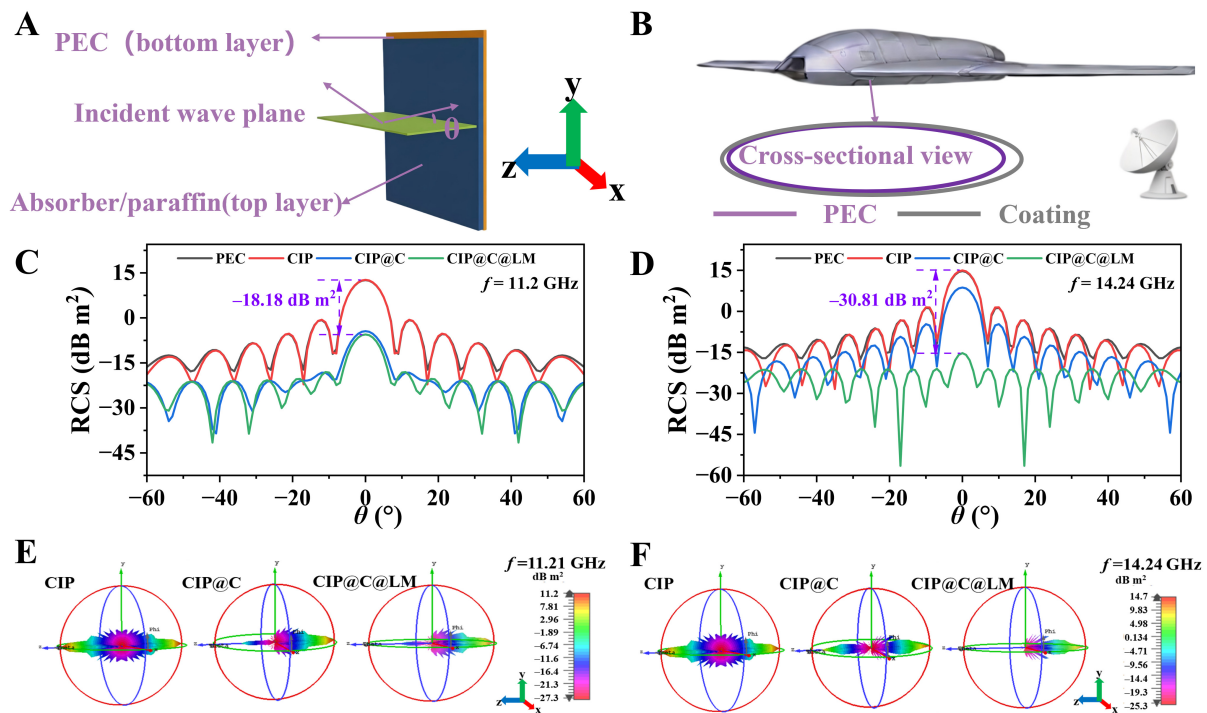
To visualize relaxation behavior, Cole-Cole plots ( $\varepsilon''$  vs.  $\varepsilon'$ ) were analyzed according to Debye theory, using the fitting relation<sup>[64]</sup>:

$$\left(\varepsilon' - \frac{\varepsilon_s + \varepsilon_\infty}{2}\right)^2 + (\varepsilon'')^2 = \left(\frac{\varepsilon_s - \varepsilon_\infty}{2}\right)^2 \quad (12)$$

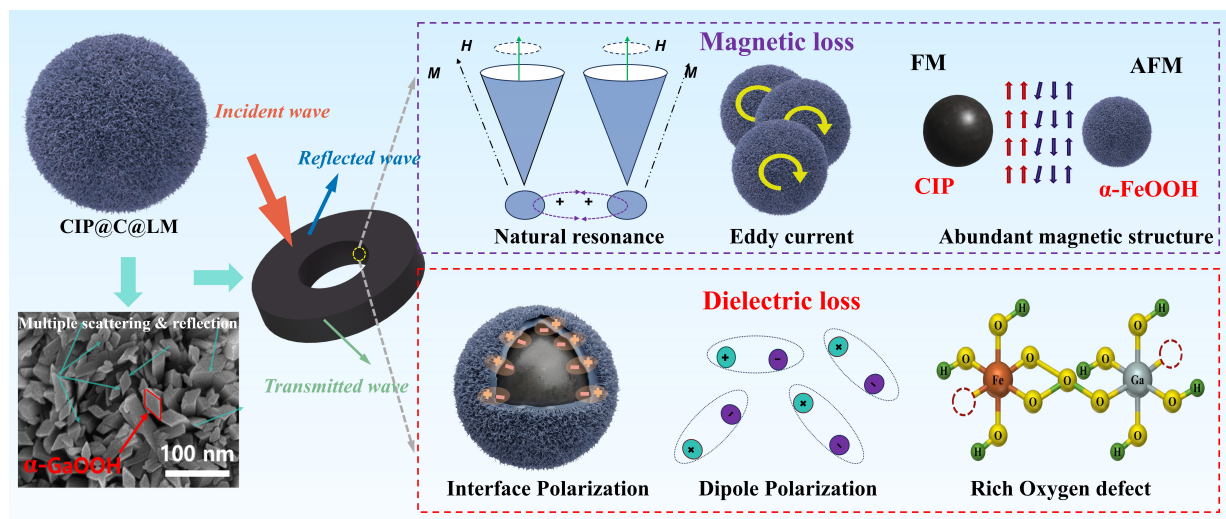
where  $\varepsilon_s$  and  $\varepsilon_\infty$  are the static and high- $f$  limit permittivity, respectively. An ideal single Debye relaxation corresponds to one semicircle, deviations from a single semicircle generally indicate multiple relaxation processes. As shown in **Figure 7E**, CIP, CIP@C, and CIP@C@LM display approximately 2, 3, and 4 semicircles, respectively, suggesting progressively more complex relaxation behavior with increasing interfacial/defect complexity. The larger number and broader distribution of semicircles for CIP@C@LM indicate coexisting polarization processes and more developed interfacial polarization network enabled by the enriched surface dipoles and abundant heterogeneous interfaces. Finally, the dielectric loss tangent ( $\tan \delta_\varepsilon$ , see **Figure 7F**) provides a direct comparison with  $\tan \delta_\mu$  [**Figure 6E**]. In the principal absorption frequency range of 10–16 GHz,  $\tan \delta_\varepsilon$  is substantially higher than  $\tan \delta_\mu$ , indicating that dielectric loss dominates the electromagnetic attenuation of CIP@C@LM. This result is consistent with the enhanced polarization-loss component  $\varepsilon_p''$  and the increased number of Cole-Cole semicircles, confirming that abundant polarization-relaxation processes are primarily responsible for its improved EMW absorption performance. Magnetic loss, as reflected by the  $\mu''$  and  $\tan \delta_\mu$  responses, remains a secondary contribution, while optimized impedance matching further facilitates the broadband absorption of CIP@C@LM.

To substantiate the broadband and wide-angle EMW attenuation capability of CIP@C@LM composite, we further evaluated its stealth-related performance through radar cross-section (RCS) simulations<sup>[65]</sup> [**Figure 8**]. **Figure 8A** presents the reference model constructed with a perfect electric conductor (PEC) plate, while **Figure 8B** shows the unmanned aerial vehicle (UAV) model coated with the as-prepared absorber, highlighting its potential for practical platform-level EMW suppression. In general, the RCS quantitatively represents the intensity of reflected EMW detected from a target under coating conditions; a smaller RCS corresponds to weaker backscattering and thus improved stealth performance. **Figure 8C** and **D** present the RCS simulations at a specific resonance  $f$ . The coated model achieves minimum RCS values (with respect to the PEC reference) of -18.18 dB m<sup>2</sup> at 11.21 GHz and -30.81 dB m<sup>2</sup> at 14.24 GHz, representing a significant reduction in radar backscattering. The 3D far-field scattering patterns at 11.21 GHz and 14.24 GHz [**Figure 8E** and **F**] further visualize this effect, where the scattering “lobe” area is markedly reduced for the coated model, confirming effective suppression of reflected waves at both  $f$ . Such a combined attenuation strategy suggests that this material can effectively regulate radio-wave absorption under complex operating conditions, supporting its promise for practical stealth-oriented EMW mitigation applications<sup>[66]</sup>.

Based on the above analysis, the EMW absorption mechanism in CIP@C@LM can be summarized as shown in **Figure 9**. First, the hierarchical nanorod/porous architecture extends the transmission path of incident waves via multiple scattering and reflection, thereby increasing the dissipation probability. Second, the intrinsic magnetic performance of the CIP core contributes to the magnetic loss by natural resonance. The reconstructed heterostructure helps induce the eddy-current dissipation by regulating the local electromagnetic environment. Furthermore, the minor interfacial AFM  $\alpha$ -FeOOH introduces an additional magnetic component, thereby enriching the local magnetic structure and contributing to the effective



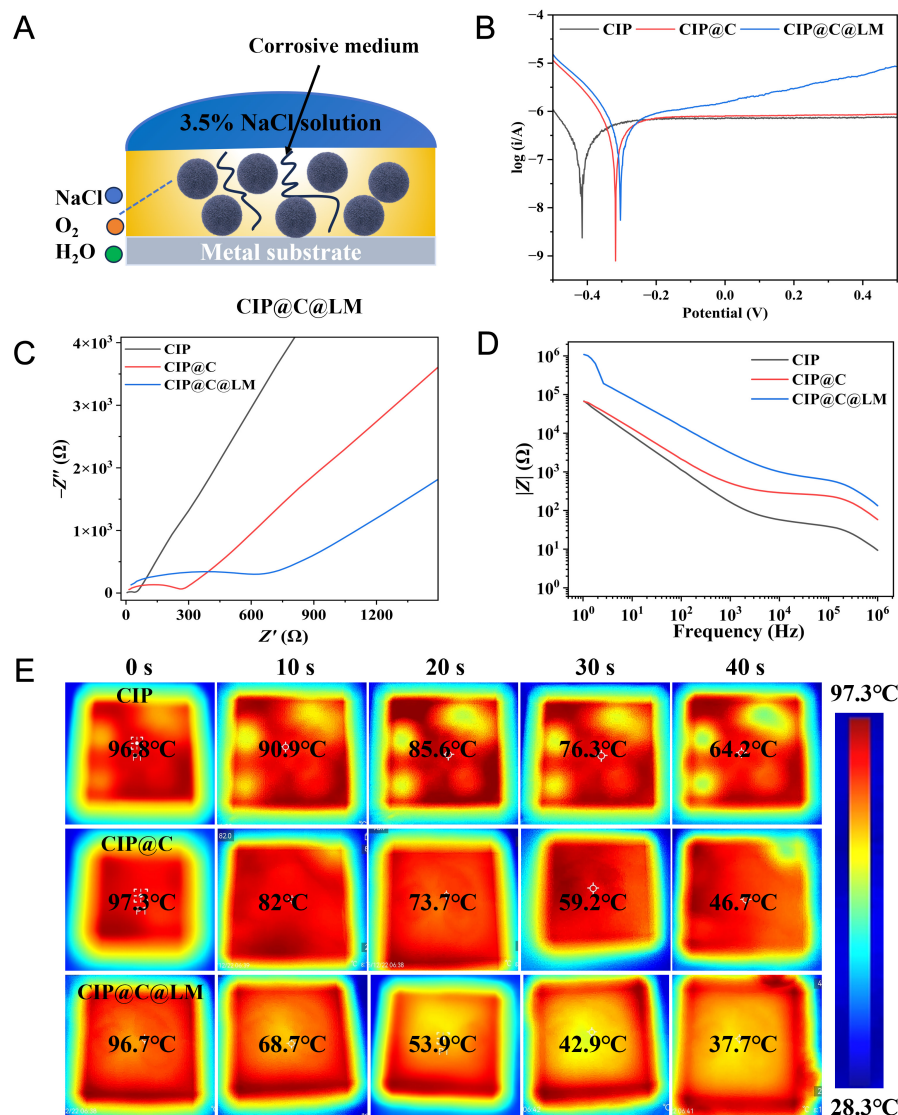
**Figure 8.** Radar cross-section (RCS) simulation of the composite absorbers. (A) Theta polarization model, (B) Drone model with ultra-wideband EMW absorption performance; (C and D) RCS simulation curves of PEC in the test range of  $-60.0^\circ < \theta \leq 60.0^\circ$  at 11.21 GHz and 14.24 GHz, (E and F) CST far-field simulation results of CIP, CIP@C and CIP@C@LM. CIP: Carbonyl iron powder; LM: liquid metal; EMW: electromagnetic wave; PEC: perfect electric conductor; RCS: radar cross-section; CST: computer simulation technology.



**Figure 9.** Proposed EMW absorption mechanism of CIP@C@LM composite. CIP: Carbonyl iron powder; LM: liquid metal; EMW: electromagnetic wave; AFM: antiferromagnetic; FM: ferromagnetism.

magnetic response of CIP@C@LM. Third, dielectric loss is significantly enhanced by the synergistic effect of interfacial and dipolar polarizations. The multiple heterogeneous boundaries (CIP/C, C/ $\alpha$ -GaOOH,  $\alpha$ -FeOOH/ $\alpha$ -GaOOH) induce strong Maxwell-Wagner relaxation, while the abundant defects ( $O_v$ ) and functional groups within the oxyhydroxide shell serve as active dipolar centers, converting electromagnetic energy into heat. The integration of these loss channels enables the CIP@C@LM composite to achieve superior broadband absorption.





**Figure 10.** Corrosion resistance and heat-dissipation performance of CIP, CIP@C, and CIP@C@LM coatings. (A) Schematic illustration of the electrochemical corrosion test setup; (B) Tafel polarization curves obtained in 3.5 wt.% NaCl solution; (C) Nyquist plots in the low-impedance region obtained from electrochemical impedance spectroscopy measurements; (D) Bode impedance modulus plots; (E) Infrared thermal images and corresponding surface-temperature evolution showing the heat dissipation behavior of the three coatings during cooling from approximately 97 °C. CIP: Carbonyl iron powder; LM: liquid metal.

### Corrosion resistance and heat dissipation capacity

With the expanding deployment of EMW absorbing materials, their corrosion resistance and heat-dissipation capability under harsh environments have become increasingly critical. The formation of C and  $\alpha$ -GaOOH on the CIP surface can act as an effective protective barrier, thereby improving the oxidation resistance and corrosion tolerance of CIP. Figure 10A and B schematically illustrate the electrochemical measurement configuration and the corresponding Tafel polarization curves obtained in 3.5 wt.% NaCl solution using glassy C working electrodes coated with CIP, CIP@C, and CIP@C@LM, respectively. Compared with pristine CIP, the polarization curves of CIP@C and CIP@C@LM shift toward more positive corrosion potentials ( $E_{\text{corr}}$ :  $-0.415 \text{ V} \rightarrow -0.319 \text{ V} \rightarrow -0.301 \text{ V}$ ), accompanied by a pronounced decrease in corrosion current density ( $I_{\text{corr}}$ :  $3.16 \times 10^{-6} \text{ A} \rightarrow 1.99 \times 10^{-7} \text{ A} \rightarrow 1.28 \times 10^{-7} \text{ A}$ ). This trend indicates that both the C shell and the  $\alpha$ -GaOOH shell promote the formation of a compact protective film, which suppresses charge transfer at the metal/electrolyte interface and thus retards the corrosion process. The fitted

parameters derived from Tafel<sup>[67]</sup> analyses are summarized in [Supplementary Table 2](#). To complement the Tafel polarization analysis, electrochemical impedance spectroscopy (EIS) measurements were further performed. As shown in the Nyquist plot in [Figure 10C](#), compared with CIP and CIP@C, CIP@C@LM exhibits a more pronounced impedance response. More importantly, the Bode impedance modulus plots in [Figure 10D](#) show that CIP@C@LM possesses a markedly higher  $|Z|$  value (impedance modulus) in the low-frequency region, indicating enhanced resistance to electrolyte penetration and interfacial charge transfer. Together with the positively shifted  $E_{\text{corr}}$  and reduced  $I_{\text{corr}}$  values obtained from the Tafel analysis, these EIS results further support the improved corrosion resistance of CIP@C@LM under the present testing conditions. The heat-dissipation capability of the coatings is compared in [Figure 10E](#). All coatings were heated to an initial temperature of approximately 97 °C and the surface temperature evolution was tracked within 40 s. The CIP coating develops visible bulging during cooling, indicative of thermal-stress accumulation caused by interfacial mismatch and a relatively high effective thermal expansion. In contrast, CIP@C and CIP@C@LM coatings remain intact during the cooling process, indicating better coating integrity under the present test condition and more efficient heat dissipation. Quantitatively, after 10 s, the surface temperatures of CIP, CIP@C, and CIP@C@LM decrease to 90.9, 82, and 68.7 °C, respectively. After 40 s, the temperatures further drop to 64.2, 46.7, and 37.7 °C, demonstrating a substantially accelerated cooling rate for the coated architectures. These results suggest that the introduction of the C interlayer and the  $\alpha$ -GaOOH shell reduces the interfacial thermal resistance and facilitates heat transport across the coating, thereby enhancing the heat-dissipation capability of the composite coating.

## CONCLUSIONS

In summary, we propose a heterostructure engineering strategy to construct CIP@C@LM featuring abundant magnetic structure and  $O_v$  defects, enabling integrated regulation of interfacial architecture and electromagnetic response, and addressing key limitations of CIP-based absorbers in EMW attenuation, heat dissipation, and corrosion resistance. The introduction of a C shell not only protects the magnetic core but also enables controlled interfacial oxidation of CIP to generate an AFM  $\alpha$ -FeOOH, while simultaneously promoting the growth of a rhombus-like  $\alpha$ -GaOOH shell enriched with  $O_v$ . In parallel, the multi-shell interfaces together with defect-related dipoles markedly intensify polarization loss and optimize impedance matching, thereby boosting EMW absorption. As a result, the composite delivers an  $RL_{\text{min}}$  of -52.6 dB at a thickness of 2.31 mm and achieves an  $EAB_{\text{max}}$  of 7.56 GHz (10.44–18 GHz) at 2.08 mm. In addition, the heterostructure exhibits improved environmental adaptability, including enhanced corrosion resistance and rapid heat dissipation, cooling from 96.7 °C to 37.7 °C in 40 s. Collectively, these findings provide new insights into interface engineering of CIP-based EMW absorbers and offer a viable design route toward next-generation multifunctional EMW attenuation materials.

## DECLARATIONS

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### Authors' contributions

Design: Li, Y.; Zang, J.; Bai, X.;

Experiments: Li, Y.; Yue, S.; He, T. A.

Manuscript writing and revision: Li, Y.; Gao, L.; Ma, Z.; Cao, Q.; Jiang, M.;

Supervision: Jiang, M.

### Availability of data and materials

The raw data supporting the conclusions of this article are available from the corresponding author upon reasonable request.

### AI and AI-assisted tools statement

During the preparation of this manuscript, the AI tool ChatGPT (version GPT-5.4 Thinking, released 2026-03-05) was used solely for language editing, and Gemini (version Gemini 3.1 Pro, released 2026-02-19) was used to assist in the preparation of certain non-data illustrative elements in the schematic representation of [Figure 3](#). These tools did not influence the study design, data collection, analysis, interpretation, or the scientific content of the work. All authors take full responsibility for the accuracy, integrity, and final content of the manuscript.

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### Conflicts of interest

All authors declared that there are no conflicts of interest.

### Ethical approval and consent to participate

Not applicable.

### Consent for publication

Not applicable.

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### Supplementary Materials

[Supplementary Materials](#)

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