

Nanofibrous aerogels with continuous conductivity gradient for ultrabroadband absorption-dominant electromagnetic interference shielding

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Electromagnetic interference (EMI) shields are limited by a severe impedance mismatch with free space, leading to strong reflections and secondary radiation. Here, we introduce ultralight all-polymer aerogels featuring a continuous through-thickness conductivity gradient (MCAs), fabricated in a single step via diffusion-driven oxidative polymerization of pyrrole within aramid nanofiber scaffolds. An exponentially decaying conductivity profile enables ultrabroadband, low-reflection performance in 5-mm-thick samples, achieving reflection power coefficient $R < 0.1$ across 10.2–40.0 GHz, while maintaining shielding effectiveness > 30 dB under low-conductivity face incidence. Waveguide measurements and simulations show that progressive impedance transition and internal field redistribution drive absorption-dominated ohmic loss rather than surface reflection. A gradient-stratified electromagnetic model delineates an optimal design window for key gradient parameter. Beyond EMI protection, this nanofibrous architecture provides mechanical robustness and facile processability, establishing a diffusion–reaction strategy for spatially programmed conductive networks relevant to electromagnetic, electronic, and energy systems.

The rapid proliferation of portable electronics and fifth-generation (5G) communications has intensified concerns about electromagnetic radiation and pollution, posing threats to human health and the reliability of electronic systems^{1,2}. Electromagnetic interference (EMI) can corrupt signals and trigger failures in devices ranging from biomedical pacemakers to aerospace platforms³. High-performance EMI shielding materials are therefore indispensable, yet remain challenging

to realize in complex electromagnetic environments. EMI shielding effectiveness (SE) is primarily governed by interactions between mobile charge carriers and incident electromagnetic waves (EMWs)⁴. Accordingly, conventional approaches employ highly conductive materials, e.g., metals^{5,6}, transition metal carbides/nitrides (MXenes)^{7–9}, carbon-based nanomaterials^{10,11}, liquid metals^{12,13}, to achieve high SE. However, their high conductivity inevitably produces

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severe impedance mismatch at air-shield interfaces, leading to a reflection coefficient (R) often exceeding 0.9, meaning that over 90% of incident electromagnetic waves (EMWs) are reflected. This not only generates secondary electromagnetic pollution but also raises the risk of information leakage¹⁴.

To mitigate impedance mismatch, multi-scale structural designs have been explored. Porous scaffolds alleviate impedance mismatch and introduce scattering centers and heterogeneous interfaces, thereby promoting absorption¹⁵. Laminated stacks or discrete conductivity gradients further suppress front-surface reflection^{16–18}. For instance, a three-dimensional (3D) printed MXene/carbon nanotube (CNT)/poly(amic acid) trilayer with CNT-graded, porous structure achieved SE of 68.2 dB with R of 0.23 at X band of 8.2–12.4 GHz¹⁸. An asymmetric laminate aerogel reached an average R of 0.1 at 8.2–12.4 GHz, which is achieved by an impedance-matching MXene@Ni/poly(p-phenylene-2,6-benzobisoxazole) nanofibers (PNF) absorbing layer and a MXene/PNF back-reflecting layer to guide incident EMW through an absorption-reflection-reabsorption pathway¹⁶. Despite these advances, the existing gradient shields are mainly constructed by stacking, asymmetric lamination, sequential deposition, or other discrete-gradient routes^{14,16,19}. As a result, their conductivity profiles are often stepwise rather than truly continuous, and the retained interlayer boundaries may introduce local impedance discontinuities, imperfect interfacial contact, and delamination risk. Moreover, their low-reflection response can be sensitive to layer thickness and stacking sequence, potentially limiting the effective matching bandwidth, while multistep assembly adds processing complexity. Furthermore, most shields rely on hybrid and heavy inorganic fillers^{1,20}, including MXenes^{21,22}, magnetic particles^{23,24}, carbon material^{25,26}, metal nanomaterials^{27–29}, which compromise structural and mechanical integrity due to poor interfacial adhesion and random dispersion, an issue that is especially critical in highly porous aerogels³⁰. Consequently, achieving ultrabroadband, absorption-dominated shielding with R below 0.1 while retaining low density and mechanical robustness remains a formidable challenge. Addressing this unmet need requires rethinking conductive architectures beyond discrete layers or random distribution of fillers, toward continuous, spatially programmable conductivity gradients.

Here, we develop all-polymer aerogels with monolithic, continuous conductivity gradients (MCAs), rather than stepwise architectures with discrete conductivity transitions and interlayer boundaries. MCAs feature a through-thickness exponential conductivity profile, synthesized via a one-step diffusion-controlled oxidative polymerization of polypyrrole (PPy) within aramid nanofiber scaffolds (Fig. 1a, b). By tuning scaffold porosity, diffusion time, and reaction conditions, we program PPy distribution and conductivity that exhibit exponential decay across thickness. The resulting MCAs deliver absorption-dominated EMI shielding with $R < 0.1$ across an ultrabroadband frequency range of 10.2–40 GHz, while maintaining SE >30 dB. Combined experimental-theoretical analysis shows that the continuous gradient minimizes interfacial impedance mismatch and redistributes internal electromagnetic fields, favoring conduction-driven dissipation over surface reflection. Based on Maxwell's electromagnetic theory, we further propose a gradient-stratified model that quantitatively describes the energy scattering behavior of MCAs. Our theoretical model further defines an optimal design window for conductivity gradient characteristics, providing general rules for low-reflection shielding. Beyond EMI protection, the all-polymer architecture provides elasticity, toughness, and facile processability (e.g., laser cutting), enabling integration onto complex geometries (Fig. 1c–e). These findings establish a lightweight, flexible, and robust paradigm for gradient-structured conductive networks, addressing the long-standing challenge of ultrabroadband low-reflection EMI shielding and providing guiding principles for the design of next-generation electromagnetic, electronic, and energy materials.

Results

Fabrication and structure of MCAs

The fabrication of MCAs begins with a highly porous scaffold, comprising a self-assembled hyperconnected aramid nanofiber (ANF) network³¹ cross-linked with poly(vinyl alcohol) (PVA). Briefly, Kevlar fibers (poly(p-phenylene terephthalamide), PPTA) are dispersed in DMSO with KOH to produce highly branched ANFs, which spontaneously form a hydrogel upon solvent exchange with water³². The integration of PVA during gelation welds together nanofiber junctions via hydrogen bonding, strengthening the network's nodal connectivity^{33,34}. This results in a self-assembled nanofibrous ANF-PVA hydrogel with extremely low solid content yet robust mechanical integrity, which serves as the template for subsequent in situ polymerization of PPy for creating the conductivity gradient structure (Fig. 1a)³⁵.

To introduce conductive polymer PPy, the ANF-PVA hydrogel is infused with an aqueous pyrrole (Py) solution from the top surface, and an FeCl₃ oxidant solution was subsequently applied. The oxidant slowly diffuses into the hydrogel, triggering oxidative polymerization of Py in a spatially confined manner. Polymerization commences immediately upon contact (the solution darkens without any induction period) and proceeds faster in regions with higher oxidant concentration (near the top). The open-pore architecture of the ANF-PVA network (surface area $\sim 111 \text{ m}^2 \text{ g}^{-1}$ at 94% porosity) ensures efficient mass transport and reactant diffusion³⁶. Consequently, PPy formation is concentration-diffusion-driven. The top layer, initially exposed to the most FeCl₃, forms the most PPy, while lower regions receive the oxidant later and form progressively less PPy. This one-step process yields a continuous gradient of PPy content throughout the monolith, with a highly conductive, PPy-rich top transitioning gradually to an insulating bottom (Figs. 1b and 2e). Hereafter, the PPy-rich and PPy-poor surfaces are denoted as the high- σ and low- σ faces, respectively, where σ is the electrical conductivity. After polymerization, the hydrogel is transformed into a lightweight aerogel (Fig. 1c) by critical point drying, preserving the monolithic porous structure.

Moreover, the strong affinity between PPy and the ANF framework enables the conformal, gradient distribution of PPy. Attenuated total reflectance Fourier-transform IR (ATR-FTIR) spectroscopy of the composite confirms successful in situ polymerization and reveals intermolecular interactions underlying the templating effect (Supplementary Fig. 1a). Specifically, C=O stretching of PPTA (ANFs) red shifts in the presence of PPy, indicating hydrogen bonding, and aromatic regions suggest π - π stacking between PPy chains and the benzene rings of PPTA (Supplementary Fig. 1)³⁷. Furthermore, pyrrole monomers adsorb strongly onto ANF surfaces, with a higher binding free energy than in water, which directs polymerization along the nanofibers as the oxidant permeates the network³⁸. Consistent with this, electron microscopy shows that PPy deposits as a thin coating on the ANF-PVA nanofibers, replicating the hyperbranched network topology without clogging the pores (Supplementary Fig. 2)³⁷. The interplay of diffusion kinetics and interfacial chemistry confines most PPy to the upper region of the monolith, leaving the deep interior as pure ANF-PVA, imprinting a continuous conductivity gradient into the dried aerogel.

The all-polymer fibrous network endows the MCAs with flexibility and processability. They can be bent, folded, or compressed without structural damage (Fig. 1d). Additionally, the aerogels can be patterned via laser cutting to create kirigami lattices or auxetic motifs, allowing for large tensile strains and conformal contact with curved or irregular surfaces (Fig. 1e). Leveraging a continuous through-thickness conductivity gradient that mitigates impedance mismatch at the air-shield interface, these MCAs deliver low-reflection, absorption-dominated EMI shielding (Fig. 1f), as shown in the following sections. This unique combination of low density, mechanical resilience, structural flexibility, straightforward processability, and tunable conductivity

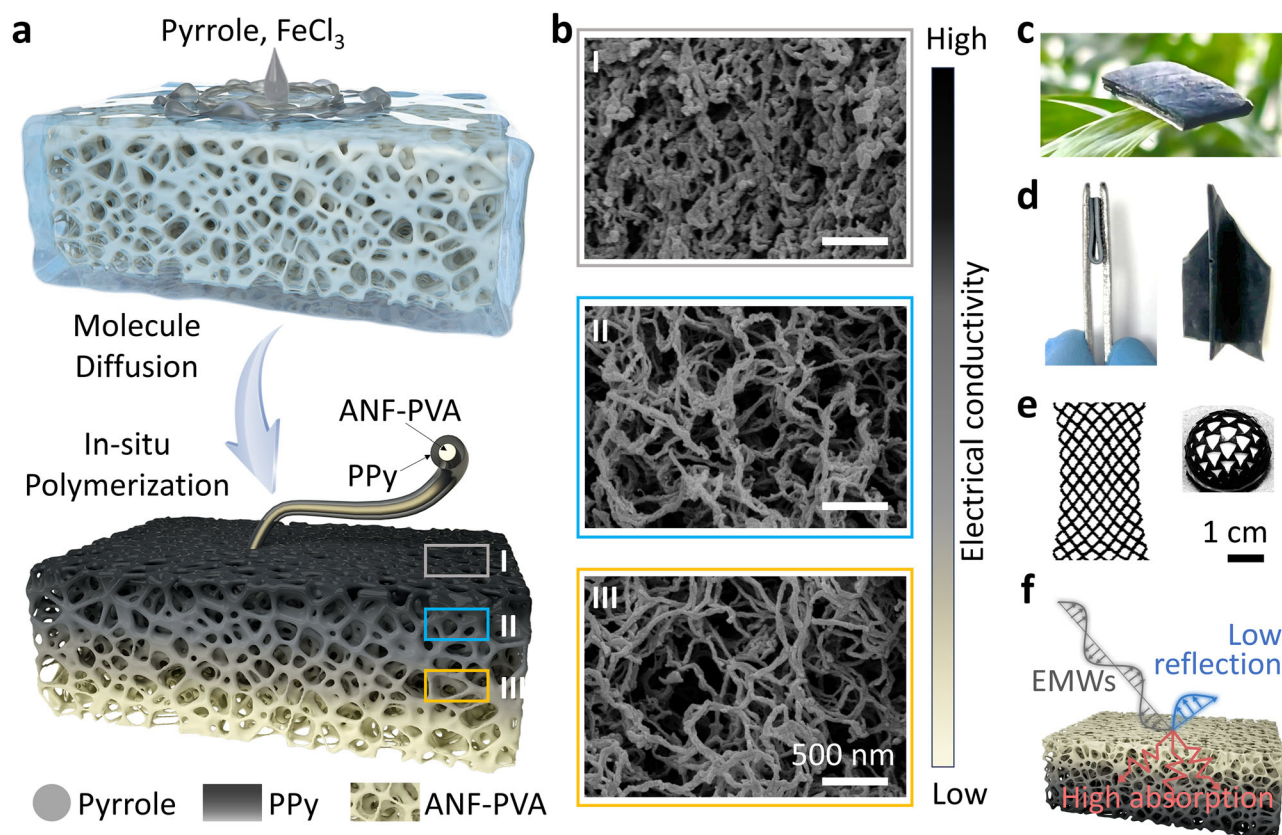


Fig. 1 | Fabrication and structure of monolithic conductivity-gradient aerogels. **a** Schematic illustrating the formation of monolithic continuous conductivity-gradient aerogels (MCAs) through diffusion-driven, in situ polymerization of polypyrrole within a porous ANF-PVA scaffold. **b** Cross-sectional SEM images of the top surface, mid-plane, and bottom, showing a decrease in PPY content and

conductivity across depth. **c–e** Digital images displaying macroscale features. MCAs exhibiting lightweight (**c**), foldable (**d**), stretchable, and readily processable properties (**e**). **f** MCAs serve as absorption-dominated, low-reflection EMI shields benefiting from the gradient conductivity.

profiles is unattainable in conventional EMI shields, positioning MCAs as a promising platform for next-generation flexible, low-reflectivity EMI protection.

Characterization of the gradient structure of MCAs

Depth-dependent measurements confirm the intended PPY gradient in the MCAs (Fig. 2). ATR-FTIR spectra collected at different depths show that the characteristic PPY peaks (e.g., C=C stretching at 1554 cm^{-1} and C-N stretching at 1306 cm^{-1}) progressively decrease in intensity with distance from the top surface, while the specific peak for ANF (C=O stretching at 1646 cm^{-1}) increases correspondingly (Fig. 2b and Supplementary Fig. 3). This indicates a gradual reduction of PPY content from the top to the bottom of the porous scaffold (Fig. 2a). Consistently, the pore structure varied with depth (Fig. 2c). The pristine ANF-PVA scaffold exhibits a relatively uniform pore size distribution (peak ~ $800\text{--}1100\text{ nm}$, Supplementary Fig. 4). After in situ PPY growth from the top down, the upper region shows significantly smaller effective pore sizes (peak ~ 400 nm), whereas the lower region retains much larger pores ($> 1000\text{ nm}$). This is attributed to PPY accumulating near the top, where PPY coats the nanofiber and partially fills the pore channels, thereby reducing the pore size in the upper region while leaving the deep interior largely unfilled.

The through-thickness concentration gradient is established by diffusion-reaction coupling. In aqueous solution, the pyrrole monomer is rapidly oxidized by FeCl_3 , following a well-established polymerization mechanism (see Methods and Supplementary Fig. 5)^{39–41}. When the monomer and oxidant are introduced sequentially from the top of the scaffold, their diffusion and reaction rates together govern the spatial PPY profile. This dynamic interplay between molecular diffusion

and reaction kinetics produces the smooth concentration gradient of PPY observed in the final aerogel. To quantitatively understand the gradient formation, we simulated the coupled diffusion-reaction process using COMSOL Multiphysics (see Methods). For direct comparison, the experimentally measured PPY wt% (Supplementary Fig. 6) was converted to molar concentrations of pyrrole. The simulation results (Fig. 2d) show a high PPY concentration near the top surface that gradually decays with depth, in good agreement with the experimental profile (Fig. 2f). Unlike conventional diffusion-induced concentration gradients, our gradient is encoded by diffusion-coupled in situ polymerization, which immobilizes insoluble PPY on the nanofibrous scaffold to form a continuous conductive network within a monolithic scaffold.

We systematically investigated how the diffusion time and porosity affect the gradient. MCAs with 94% porosity and different diffusion times are denoted MCA-1, MCA-2, and MCA-4 (0.75, 2, and 4 h, respectively), whereas MCAs with a fixed 4 h diffusion time and different porosities are labeled MCA-94, MCA-88, and MCA-75 (according to porosities of 94, 88, 75%, respectively). Experimentally, extending the polymerization time from 0.75 to 4 h increased the PPY penetration depth from ~2.5 to ~4.0 mm and made the gradient less steep (dots in Fig. 2f). The simulated PPY concentration profiles (solid lines in Fig. 2f) align with the measured data (Supplementary Fig. 7). Longer diffusion/reaction time increases the PPY penetration depth and broadens the gradient, whereas lower scaffold porosity confines PPY to a shallower depth and produces a steeper profile (Supplementary Fig. 8). The porosity of the nanofibrous template provides another lever to control the gradient. Lower matrix porosity, and thus higher tortuosity⁴², significantly limits the

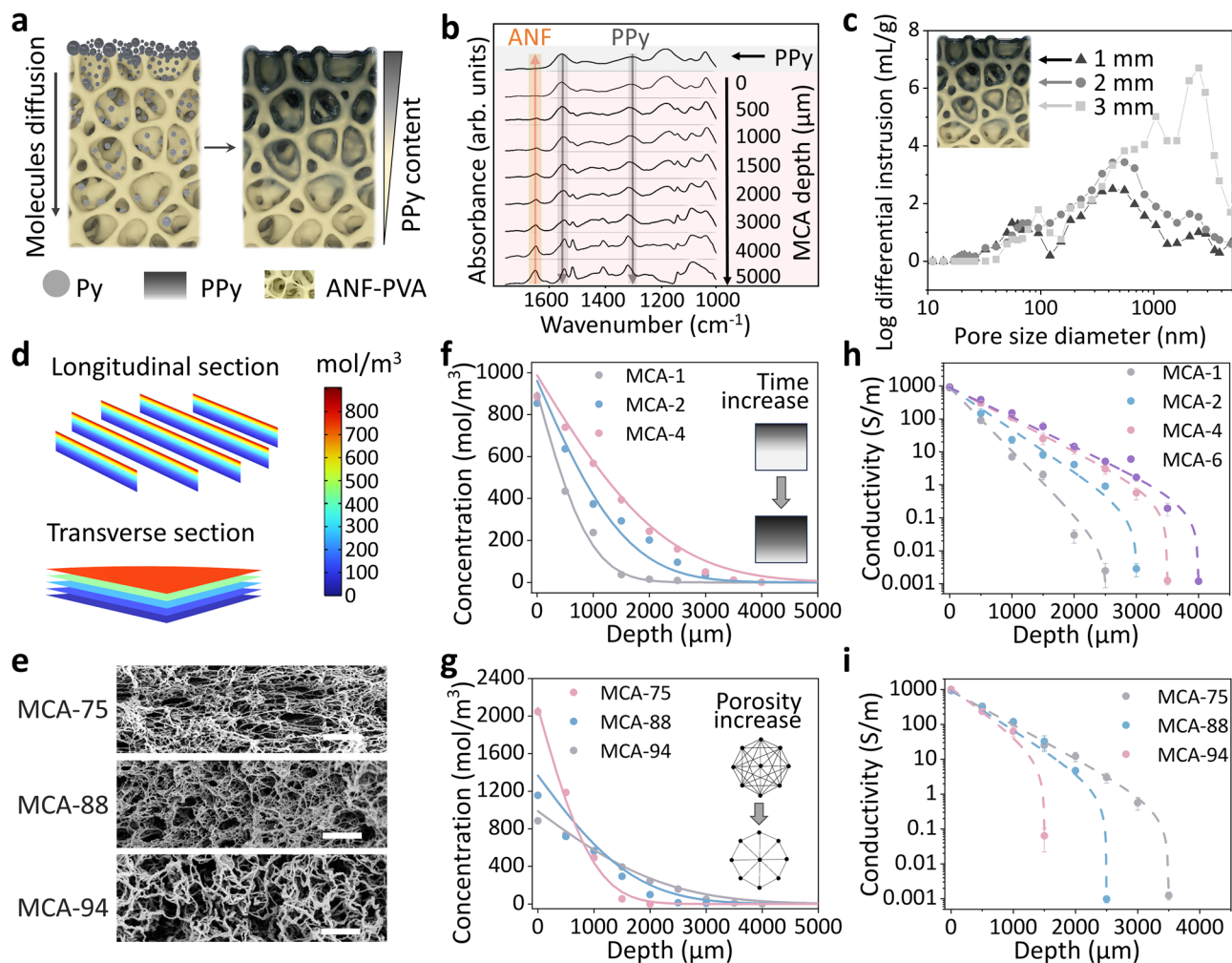


Fig. 2 | Characterization of the gradient structure. **a** Schematic of PPY gradient formation along the depth of ANF-PVA scaffold driven by the interplay between reactant diffusion and in situ polymerization. **b** FTIR spectra of MCA-94 serial sections showing progressive attenuation of PPY characteristic bands with depth, evidencing a downward PPY content gradient. **c** Pore size distribution for layers sampled at three depths. **d** COMSOL simulations of Py molar concentration maps in longitudinal (upper panel) and transverse sections (lower panel). **e** Cross-sectional SEM images of MCAs with varied porosities (75, 88, 94%). Scale bars: 1 μm . **f** Py molar concentration versus depth for 94% porosity MCAs with varied diffusion times (0.75, 2, 4 h) obtained from experimental characterization (in dots) and simulation

(in solid lines). **g** Py molar concentration versus depth for 4 h diffusion MCAs with varied porosities (75, 88, 94%) obtained from experimental characterization (in dots) and simulation (in solid lines). **h** Electrical conductivity gradient profiles versus depth for 94% porosity MCAs with varied diffusion times (0.75, 2, 4, 6 h) obtained from experimental characterization (in dots) and curve fitting (in dashed lines). **i** Electrical conductivity gradient profiles versus depth for 4 h diffusion MCAs with varied porosities (75, 88, 94%) obtained from experimental characterization (in dots) and curve fitting (in dashed lines). Data points in h and i are presented as mean $\pm$ s.d. ($n = 3$ independent samples).

depth of PPY infiltration. A dense template (75% porosity) showed PPY penetration confined to ~ 2.0 mm, with a sharp decline in PPY content (Fig. 2g and Supplementary Fig. 8b). The higher tortuosity and smaller pore size in the low-porosity matrix impede molecular transportation, causing most PPY to deposit near the top and leaving the deep interior nearly PPY-free. In contrast, a highly porous scaffold (94% porosity) enabled PPY to extend nearly 4 mm in thickness with a much smoother content gradient. The open, well-connected pores in the 94% porous matrix facilitate rapid mass transport, promoting deeper polymerization and a more gradual decrease in PPY concentration with depth. Notably, the experimentally measured PPY profiles under these different conditions (dots in Fig. 2f, g) align closely with simulation results (solid curves in Fig. 2f, g), further validating the diffusion-reaction model of gradient formation.

Since the electrical conductivity (σ) is directly associated with the PPY content, the through-depth conductivity profile likewise shows a continuous decrease from the top surface downward (Fig. 2h, i). Attributed to the hyperconnected ANF network, the aerogel shows a

low percolation threshold for electrical conductivity (< 3 wt% PPY, Supplementary Fig. 9). The through-thickness conductivity profile can be fitted by $\sigma(z) = \sigma_0 \exp(-z/\lambda)$, where σ_0 is the conductivity at the high- σ surface, z is the depth measured from that surface towards the low- σ face ($z = 0$ at the high- σ face), and λ (μm) is the characteristic decay length (Supplementary Fig. 10). The gradient arises from diffusion-reaction coupling. Rather than passive consequences of coupled diffusion and reaction, we intentionally engineer the conductivity gradient characteristics mainly by controlling (i) diffusion/reaction time, (ii) scaffold porosity/tortuosity, and (iii) reaction environment. Varying the diffusion time primarily alters the gradient's slope. Across the MCA-1/-2/-4/-6 series, increasing the diffusion/reaction time (from 0.75 to 6 h) broadens the conductivity gradient, with λ increasing from 225 to 340, 450, and 485 μm , while σ_0 remains nearly constant at ~ 900 S m^{-1} (Supplementary Fig. 10a). In contrast, decreasing the scaffold porosity from 94 to 88% and 75% slightly increases σ_0 but steepens the conductivity decay with depth. The σ_0/λ change from ~ 900 $\text{S m}^{-1}/450$ μm for MCA-4, also noted as MCA-94 in the porosity series, to

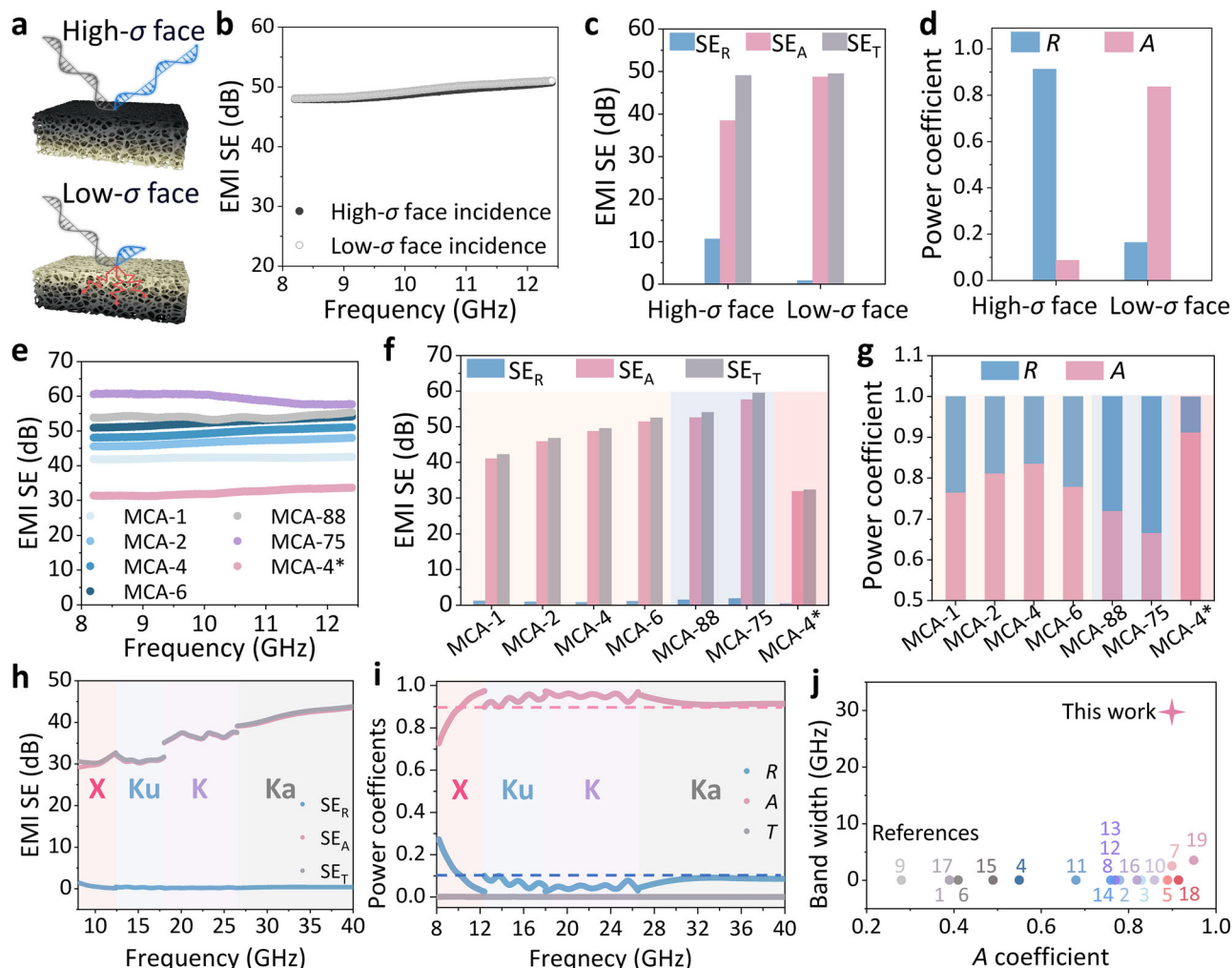


Fig. 3 | Modulation of EMI shielding performance of MCAs. **a** Schematic of EMW incidence from the high-conductivity (σ) face (upper panel) and the low- σ face (lower panel). **b–d** Direction-dependent EMI shielding performance of MCA-94. **b** EMI SE curves across the 8.2–12.4 GHz (X-band) frequency range for MCA-94 with EMWs incident from the high- σ and low- σ face, respectively. **c** SE_T , SE_R , and SE_A of MCA-94 with EMWs incident from the high- σ and low- σ face, respectively. **d** Power coefficients of reflectivity (R) and absorptivity (A) of MCA-94 as EMWs incident from the high- σ and low- σ face, respectively. **e–g** EMI shielding performance of MCAs modulated by varying the conductivity gradient profiles. EMWs incident from the low- σ face. **e** EMI SE curves over X-band for a series of MCAs with varied

gradients. SE_T , SE_R , and SE_A (**f**) and power coefficients of R , A (**g**) of a series of MCAs with varied gradients. **h, i** Ultrabroadband EMI shielding performance of MCA-4*. EMI SE (**h**) and power coefficients (**i**) of MCA-4* across 8.2–40 GHz of color-coded X, Ku, K, and Ka bands, with EMWs incident from the low- σ face. Dashed lines indicate $A = 0.9$ and $R = 0.1$. Here, MCA-4* denotes intentionally reduced surface-conductivity relative to MCA-4. All MCA samples in this series were ~5 mm thick. **j** Benchmarking the bandwidths with absorption $A > 0.9$ for previously reported EMI shielding materials. The numbers adjacent to the data points indicate the corresponding entry numbers in Supplementary Table 3.

~950 S m⁻¹/380 μ m for MCA-88 and ~1100 S m⁻¹/325 μ m for MCA-75, respectively (Fig. 2g, i and Supplementary Fig. 10b). MCA-4* denotes an intentionally surface-conductivity-reduced variant of MCA-4, prepared under an oxygen-containing polymerization atmosphere; σ_0 decreases to ~350 S m⁻¹ while λ remains ~450 μ m (Supplementary Fig. 10a, c and Supplementary Table 1)⁴³. Using a non-gravity-driven reactant infusion method yields a graded PPY profile with a reduced penetration depth (Supplementary Fig. 10d). In addition, varying the monomer and/or oxidant concentrations tunes the overall PPY loading and the gradient steepness.

Modulation of EMI shielding performance of MCAs

Incorporating continuous conductivity gradients into ultralight porous networks enables low-reflection, high-performance EMI shielding by improving impedance matching throughout the thickness. We first conducted comparative experiments without a conductivity gradient (Supplementary Fig. 11). The pristine ANF-PVA template, lacking conductive components, exhibits negligible shielding performance.

Densely packed PPY powders or PPY embedded in pure PVA matrices without a percolated network yield limited SE (<10 dB). Homogeneous integration of PPY without a gradient into ANF-PVA results in a substantial increase in SE to 50 dB but yields a symmetric, reflection-dominated shielding behavior.

In contrast, monolithic gradient MCAs maintain high SE (~50 dB) while exhibiting pronounced direction-dependent responses (Fig. 3a–d). When EMWs impinge on the high- σ face, the large impedance mismatch at the air-material interface results in strong reflection. Reversing the incidence to the low- σ face suppresses reflection and enhances absorption. This Janus shielding effect is evidenced by a decrease in R from 0.91 to 0.16 and a concomitant increase in the absorption coefficient A to 0.84 at X band (Fig. 3d), whereas the total SE remains nearly unchanged. To explore the effect of the conductivity-gradient region and thickness, we combined stepwise trimming experiments with simulations (Supplementary Fig. 12). In a representative ~5-mm-thick gradient MCA under low- σ -face incidence, R was initially ~0.15. Sequential removal from the low- σ face until only

the 1-mm-thick high- σ layer remained increased R from 0.15 to 0.8, while the SE changed only slightly. Thus, the gradient region governs the absorption-reflection partition, whereas the high- σ region largely preserves transmission blocking. By contrast, non-gradient controls of comparable thickness achieved $SE_T > 50$ dB but retained high reflectivity $R > 0.8$, showing little tunability in the absorption-reflection-transmission partitioning (Supplementary Fig. 13). These results indicate that the low-reflection response arises primarily from continuous-gradient enabled impedance matching rather than thickness alone.

We next characterized the X-band shielding response across a library of MCAs (thickness ~5 mm) with varied conductivity gradients (MCA-1, -2, -4, -6, -88, -75, and -4*; Fig. 3e–g). The depth-dependent conductivity $\sigma(z)$ is described by the exponential profile $\sigma(z) = \sigma_0 \exp(-z/\lambda)$ defined above. We observed two primary trends. First, increasing σ_0 at confined λ (~200–500 μm) raises the total SE (from ~30 to ~60 dB), consistent with a reduced skin depth that confines eddy-current dissipation to a $\sim 10^2 \mu\text{m}$ entrance layer. Second, progressively broadening the conductivity gradient (with larger λ) at near-constant σ_0 (MCA-1, 2, 4, 6) increases the absorption coefficient A from 0.76 to a maximum of 0.83 (MCA-4) before decreasing to 0.77 when λ becomes too large (MCA-6), while the SE_T increases near-monotonically at X band. This behavior suggests that an optimal λ exists to achieve low reflection. Second, holding λ near this optimum while lowering σ_0 from 900 to 350 S m^{-1} yields MCA-4*, which markedly increases A to 0.91, operates in an ultralow reflection (R of 0.09) with effective shielding (SE of 32 dB). Despite a decrease in SE, it still provides over 99.9% electromagnetic power attenuation by MCA-4*. Besides, porosity-tuned MCA-88 and MCA-75 deliver higher SE but slightly shift the balance toward reflection, lowering A (from 0.83 to 0.67). Hence, coordinated tuning of the conductivity gradient, i.e., σ_0 and λ , decouples high SE from high reflection and enables an absorption-dominated regime. The fitted σ_0 and λ values, together with porosity, thickness, and representative shielding metrics, are summarized in Supplementary Table 1.

Achieving $R < 0.1$ across a wide range, rather than at isolated frequencies, secures impedance-matched, absorption-dominated control in frequency-agile environments and mitigates backscatter-induced electromagnetic compatibility risks. Thus, broadband low- R should be regarded as a benchmark on par with SE for practical low-interference shields. However, sustaining low reflectivity across broadband is challenging because impedance matching to free space is highly frequency-dependent⁴⁴. Notably, the experimentally optimized MCA-4* operates in an ultrabroadband, absorption-dominated regime rather than exhibiting narrow resonance windows. In Fig. 3h, the shielding components are decomposed as SE_R , SE_A , and SE_T , over the color-coded X, Ku, K, and Ka bands. Across the entire 10.2–40 GHz window, the SE_T remains above 30 dB, while SE_A dominates and SE_R stays close to zero. This indicates that incident EMWs are primarily attenuated inside the gradient aerogel instead of being reflected to free space. The corresponding power coefficients further confirm this behavior (Fig. 3i). The reflectance R (solid blue dots) rapidly decreases below 0.1 (blue dashed line) at the low-frequency edge of the X band and then remains suppressed across Ku, K, and Ka, whereas the absorbance A (solid pink dots) exceeds 0.9 (pink dashed line) throughout the same range. The transmittance T is simultaneously minimized, consistent with the high SE_T . Therefore, the experimentally optimized gradient of MCA-4* achieves low-reflection shielding with $R < 0.1$ (or $A > 0.9$) while maintaining $SE > 30$ dB within an ultrabroadband range of 10.2–40 GHz (Fig. 3h, i). The bandwidth over which A exceeds 0.9 for our MCA-4* was benchmarked against reported EMI shielding materials (Fig. 3j and Supplementary Table 3)^{16–18,45–60}.

This Janus response enables orientation-specific shielding. Low- σ -face incidence is suited to suppressing secondary reflection, backscatter, and potential information leakage, whereas high- σ -face incidence can be used when front-surface rejection is desired

and reflected power is acceptable. The fixed gradient direction can be identified by intrinsic face-color contrast or rapid surface-conductivity checks. Angle- and polarization-resolved simulations confirmed absorption-dominated shielding ($SE_T > 30$ dB, $A > R$) across 8.2–12.4 GHz for both TE and TM incidence up to 60° (Supplementary Fig. 14). Preliminary stability tests showed that SE decreased from 32 to 28 dB after four months under ambient storage, with no major thermal decomposition below 200 °C (Supplementary Fig. 15). However, humidity-induced weakening of PPy-dopant interactions may cause conductivity drift⁶¹, so long-term humid-environment and cyclic hygrothermal stability remains to be established. Moisture-barrier encapsulation and dopant engineering may address this limitation^{62,63}. The PPy route also produced visible gradients in silk fibroin, ANF, PVA, and bacterial-cellulose scaffolds, indicating scaffold-level compatibility, although extension to other conductive polymers requires validation (Supplementary Fig. 16).

Theoretical modeling of shielding mechanisms governed by conductivity gradients

To move beyond discrete experiments and systematically investigate how conductivity gradients govern EMI shielding, quantified by absorption (A), reflection (R), and transmission (T), we developed a gradient-stratified model (GSM) for continuously graded conductors (see Methods). A target profile $\sigma(z)$ over thickness is represented by an N -layer, piecewise-constant approximation that preserves depth integral and boundary values (Methods, Supplementary Fig. 17). As the layer number (N) increases, the S-parameters of the staircase approximation systematically approach and converge to those of the continuous gradient (Fig. 4a). For sufficiently large N , the S-parameters predicted by our multilayer theory are numerically indistinguishable from the finite-element results for the continuous profile, thereby validating the GSM as an accurate and physically realizable surrogate of the ideal continuous gradient medium. As N increases (i.e., the discretization approaches a continuous gradient), the A coefficient rises from 0.44 at $N=1$ layer to 0.91 at $N=100$, and the total SE increases from 4.4 to 31.5 dB (Fig. 4a and Supplementary Fig. 18), confirming that gradient continuity (large N) suppresses internal impedance discontinuities and reflection. An experimentally implemented three-layer stepwise-gradient control shows intermediate performance ($R \sim 0.18$, $A \sim 0.82$, $SE_T \sim 34$ dB; Supplementary Fig. 19), confirming stepwise gradients only partially recover the impedance-matching benefit of a continuous gradient.

The model captures the pronounced directional dependence observed experimentally (Fig. 4b–d). For a representative exponential gradient, MCA-4* under low- σ face incidence shows $SE_R \sim 0.4$ dB and a $SE_A \sim 31.9$ dB, together with $R \sim 0.09$, $A \sim 0.91$, and negligible T . The high SE_A indicates strong internal attenuation of the coupled waves, mainly through PPy-network ohmic loss assisted by dielectric/interfacial polarization, whereas the low SE_R and R indicate suppressed front-surface reflection through impedance matching. Reversing the incidence to the high- σ face preserves transmission blocking ($SE_T \sim 32.4$ dB) but increases SE_R and R to ~ 9.5 dB and 0.89, respectively, while decreasing A to ~ 0.11 . Simulated and experimental trends agree in both orientation and magnitude, indicating that absorption-dominated shielding requires both strong internal attenuation and reflection suppression.

We model the through-thickness conductivity as $\sigma(z) = \sigma_0 \exp(-z/\lambda)$, with z measured from the high- σ surface (Fig. 4e). At fixed thickness ($d = 5$ mm), 10 GHz, under low- σ -face incidence, the GSM yields a design map in the explored σ_0 – λ space for SE_T , A , T , and R (Fig. 4f, g and Supplementary Fig. 20). Within this range, SE_T increases monotonically with σ_0 or λ (Fig. 4f and Supplementary Fig. 20a), as higher conductivity loading and longer conductive penetration suppress transmission⁶⁴. However, the A and R responses are non-monotonic. R and A maps show that high- A /low- R window ($A > 0.9$ or

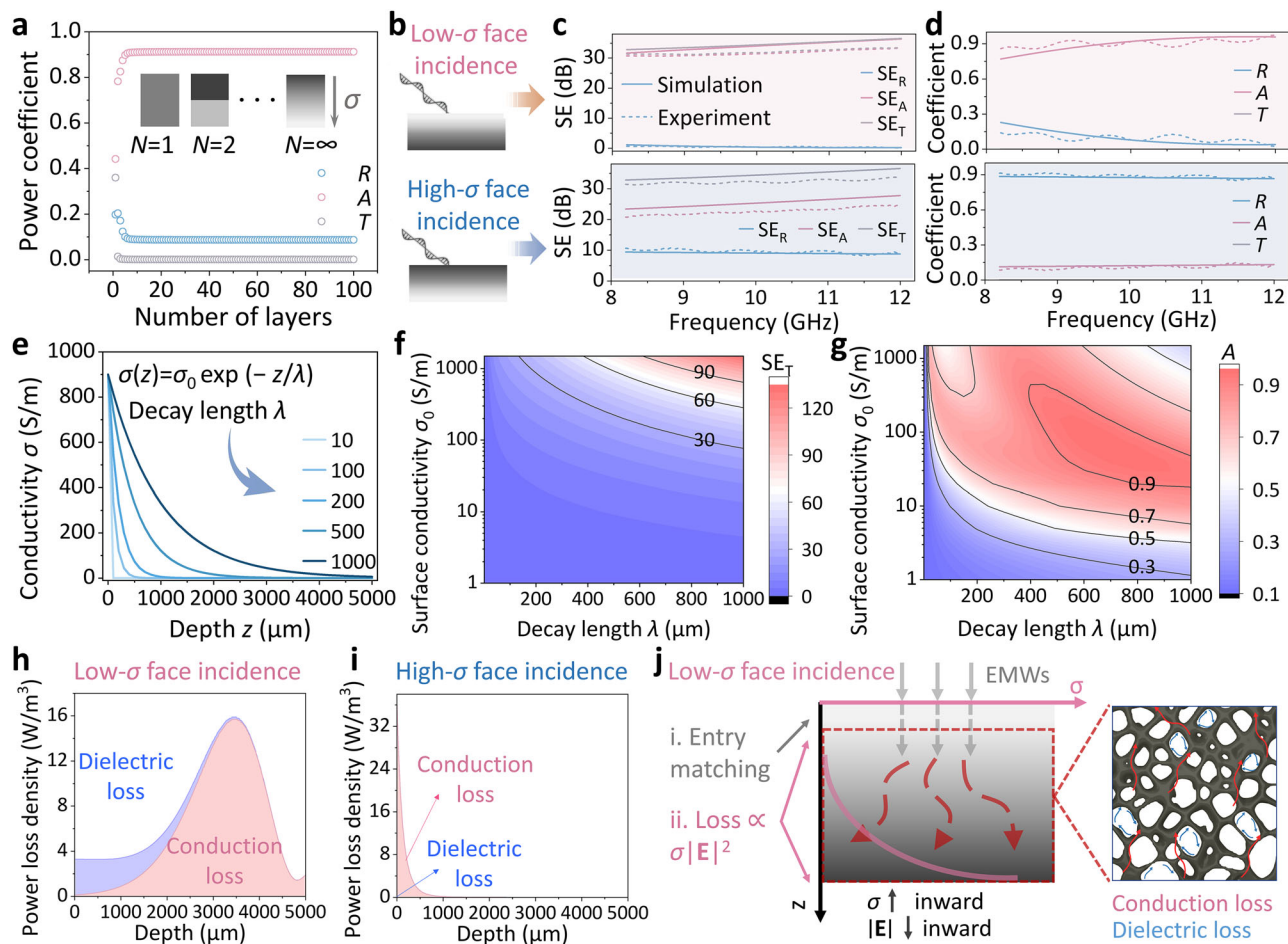


Fig. 4 | Theoretical modeling of shielding mechanisms governed by continuous conductivity gradients. **a** Power coefficients R , A , T of MCA-4* versus the number of gradient layers N under low- σ -face incidence, showing that increasing continuity (larger N) suppresses reflection and enhances absorption. Inset: discretizing the continuous conductivity $\sigma(z)$ into N equal-thickness layers, each assigned σ at the mid-depth of its interval. **b** Schematics of EM-wave incidence from the low- and high- σ faces. **c** Simulation (solid lines) and experimental (dashed lines) results of shielding effectiveness (SE) of MCA-4* when EMWs are incident from low- σ face (upper panel) and from high- σ face (lower panel) over X band. **d** Corresponding power coefficients (R , A , T) from simulation (solid lines) and experimental (dashed lines) results for MCA-4* when EMWs incident from low- σ face (upper panel) and

from high- σ face (lower panel) over X band. **e** Exponential conductivity profiles $\sigma(z) = \sigma_0 \exp(-z/\lambda)$, where z is measured from the high- σ surface. **f, g** Conditional design maps at fixed nominal thickness (~ 5 mm) and representative frequency (10 GHz), showing calculated SE_T (**f**) and absorption coefficient A (**g**) as functions of σ_0 and λ . **h, i** Depth-dependent power-loss density profiles, showing conduction loss and dielectric loss for low- σ -face incidence (**h**) and high- σ -face incidence (**i**). **j** Mechanistic schematic for low- σ -face incidence. Improved entrance matching enables deeper penetration, followed by inward shift of the $|\mathbf{E}| - \sigma$ overlap and the formation of an internal peak in $\sigma|\mathbf{E}|^2$, which promotes distributed loss and absorption-dominated shielding.

$R < 0.1$) emerges only at intermediate values of σ_0 and λ (Fig. 4g and Supplementary Fig. 20b, d). Too low σ_0 or too small λ weakens the distributed in-depth loss and leaves transmission insufficiently suppressed, whereas too high σ_0 or too large λ may reintroduce the entrance mismatch and reflection. Thus, the optimal design requires simultaneous entrance matching and internal dissipation. The experimentally optimized MCA-4* falls within this window, consistent with its low R and high SE_T . The GSM thus provides a quantitative basis for optimizing experimentally accessible σ_0 - λ combinations, in qualitative agreement with the experimental MCA library.

To reveal the mechanism of the conductivity-gradient-enabled electromagnetic behavior, we mapped the depth-dependent field, energy density, and loss profiles under opposite incidence directions (Fig. 4h, i and Supplementary Fig. 21). For incidence from the low- σ face, the power-loss density profiles show distinct regimes (Fig. 4h). Dielectric polarization dominates near the entrance with low- σ , where $|\mathbf{E}|$ is largest. Then, conduction loss prevails deeper inside, as the penetrating wave overlaps with increasing $\sigma(z)$. Notably, the local conductive loss density, $p_{\text{loss}}(z) \propto \sigma(z)|\mathbf{E}(z)|^2$, exhibits an internal

maximum, arising from the competition between the exponential growth of $\sigma(z)$ and the simultaneous decay of $|\mathbf{E}|$ (Supplementary Fig. 21b). This behavior indicates that the continuous gradient not only suppresses front-surface reflection, but also drives the field into the conductive interior, thereby enhancing internal attenuation. By contrast, for high- σ face incidence, the field energy is confined to a shallow entrance region and the loss density decays rapidly with depth (Fig. 4i and Supplementary Fig. 21f, g), consistent with strong interfacial mismatch and reflection-dominated shielding. Together, these results support a two-step mechanism for low- σ -face incidence (Fig. 4j). The low- σ entrance region first improves impedance matching and suppresses front-surface reflection, enabling deeper wave penetration. The penetrated field then overlaps with the inwardly increasing conductivity, causing the local power-loss density, $p_{\text{loss}}(z) \propto \sigma|\mathbf{E}|^2$, to reach an internal maximum and dissipate EM energy through enhanced ohmic loss. This process ultimately leads to absorption-dominated shielding.

We note that the present GSM is not intended as a full layer-resolved constitutive model. The constant ε^* , frequency-independent

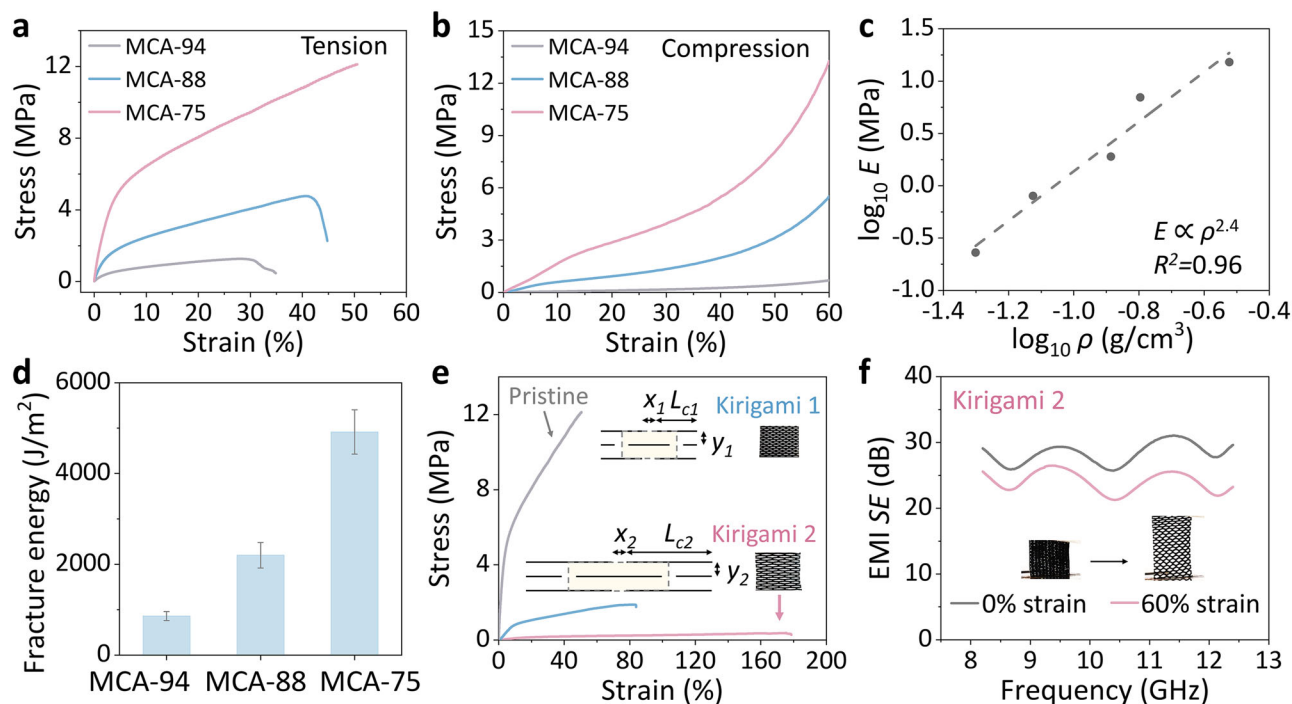


Fig. 5 | Mechanical tunability and structural adaptability of MCAs. **a** Tensile stress–strain curves of MCAs with varying porosities (94, 88, 75%). **b** Compressive stress–strain curves of MCAs with varying porosities. **c** Compressive modulus of MCAs as a function of their respective densities, obtained from experimental results (dots) and theoretical fitting (dashed lines). **d** Fracture energies of MCAs with different porosities determined by trouser-tear tests. Bars show the mean; error bars indicate s.d.; $n = 4$ independently prepared specimens per group.

e Tensile responses of kirigami-inspired MCA-75 films with different geometric dimensions. Insets are digital images of the kirigami patterns and the cut pattern unit cell in a yellow box. L_c , x , and y indicate the length of the cut, spacing in the transverse direction, and spacing in the axial direction, respectively. **f** EMI SE of MCA-75 kirigami 2 under 0 and 60% tensile strain.

σ approximation serves as a first-order conductivity-dominant description, which captures the main shielding physics in the X-band/10 GHz mechanism analysis but may become less accurate when stronger loss dispersion is involved, especially toward the Ka band and in the highly conductive deeper region, where interfacial polarization, hopping/tunneling conduction, and ϵ^* (zf) variation may become more important.

Mechanical tunability and structural adaptability of MCAs

While EMI performance is often emphasized, practical deployment also requires mechanical robustness, which remains challenging for porous shielding architectures. In the present MCA series, porosity serves as a common control parameter linking gradient formation, the reflection–absorption balance, and mechanical performance. Lower porosity increases tortuosity during in situ polymerization, shortens PPy penetration, and yields a steeper conductivity gradient with a smaller λ . In the current sample series, this shifts the σ_0 – λ pair of MCAs away from the high-absorption window identified by the GSM, whereas the denser nanofibrillar network simultaneously enhances load-bearing stiffness, strength, toughness, and film integrity.

The mechanical properties of MCAs can be systematically tuned via porosity (Fig. 5 and Supplementary Table 2). Incorporation of PPy into the ANF-PVA scaffold induces only modest stiffening (Fig. 5a, b and Supplementary Fig. 22), reflecting strong interfacial adhesion and effective stress transfer. Under uniaxial tension, decreasing porosity enhances modulus and strength (Fig. 5a). MCA-75 (75% porosity) exhibits a Young's modulus of 205 MPa and a tensile strength of 12.1 MPa, markedly higher than MCA-88 (80 and 4.77 MPa) and MCA-94 (26 and 1.3 MPa). The corresponding tensile toughness reaches $4.29 \times 10^6 \text{ J m}^{-3}$. Compression tests reveal analogous trends, with stress and modulus rising as porosity decreases (Fig. 5b). In particular, MCA-75 exhibits a typical three-stage deformation, with an initial elastic

regime ($< 10\%$ strain), a softening stage from fibrillar buckling, and final strain-hardening during network densification⁶⁵. Higher-porosity samples, with longer and less constrained fibrillar segments, undergo buckling at lower compressive stresses, leading to an earlier onset of softening. The compressive modulus (E) scales with density (ρ) as $E \propto \rho^{2.4}$ (Fig. 5c), closely matching the scaling observed in tensile tests, consistent with Gibson–Ashby models for open-cell cellular solids, validating the structural efficiency of the interconnected network (Supplementary Fig. 23)⁶⁶.

Fracture energy analysis (Fig. 5d and Supplementary Fig. 24) reveals that MCAs become markedly tougher as porosity decreases, reaching $\sim 5 \text{ kJ m}^{-2}$ for MCA-75, which is orders of magnitude higher than most reported EMI shielding aerogels. The denser network also restricts PPy penetration depth, yielding sub-millimeter MCA-75 films with preserved conductivity gradients for impedance matching and enhanced mechanical integrity. To further improve adaptability, kirigami structuring was incorporated into MCA-75. The slit geometry was chosen conservatively to minimize slit-induced transmission in the X band while permitting stretchability (Methods and Supplementary Fig. 25)^{67,68}. Increasing L_c while keeping x and y spacing constant reduced buckling stress and enhanced stretchability (Fig. 5e)⁶⁹. Although the patterned films exhibited a modest reduction in SE, the performance remained $> 26 \text{ dB}$ in the X-band and $> 20 \text{ dB}$ even under 60% strain (Fig. 5f). These results demonstrate that conductivity gradients embedded within flexible polymer networks sustain efficient electromagnetic shielding during deformation, and indicate suitability for both open (perforated/kirigami) and enclosed shielding architectures, advancing lightweight, conformal shields for dynamic, strain-varying operation.

A comparative benchmarking analysis (Supplementary Fig. 26 and Supplementary Table 3) underscores the multifunctionality of MCAs. Combining high total and specific SE (20–65 dB; 66 – $1083 \text{ dB cm}^3 \text{ g}^{-1}$),

low density ($0.06\text{--}0.3\text{ g cm}^{-3}$), strong mechanical integrity (tensile strength 12.1 MPa ; compressive stress 13.2 MPa at 60% strain), and low reflection (R of $0.09\text{--}0.4$), MCAs integrate high performance, scalability, and an all-organic composition, defining a promising platform for broadband, lightweight EMI protection.

Discussion

We have established a monolithic conductivity-gradient engineering strategy through diffusion-reaction coupled in situ polymerization to achieve ultrabroadband, absorption-dominated EMI shielding in lightweight all-polymer aerogels. Compared with stacked or stepwise-gradient architectures, the interface-free continuous gradient mitigates abrupt impedance discontinuities and eliminates interlayer delamination. In $\sim 5\text{-mm}$ -thick MCAs, the optimized gradient aerogel achieves ultralow reflection ($R < 0.1$) and high EMI shielding effectiveness ($SE > 30\text{ dB}$) across $10.2\text{--}40.0\text{ GHz}$ when irradiated from the low- σ face. Combined experimental and theoretical analyses reveal that progressive conductivity transitions and internal field redistribution govern wave attenuation, favoring conduction-dominated absorption over surface reflection. Our predictive gradient-stratified model delineates an optimal design window for key gradient parameters, providing quantitative guidance for broadband low-reflection architectures. The robust nanofibrous framework further imparts mechanical integrity, toughness, and adaptability under large deformations.

Methods

Preparation of monolithic gradient-continuous conductivity aerogels

Kevlar para-aramid pulp (Type 979, dry merge 1F538; DuPont; no numerical chemical-purity specification provided by the manufacturer) and poly(vinyl alcohol) (PVA; $M_w = 146,000\text{--}186,000$; $99\%+$ hydrolyzed; Sigma-Aldrich) were used as precursors to construct the porous framework. Kevlar pulp was dispersed in dimethyl sulfoxide (DMSO; Aladdin; AR grade, $\geq 99.7\%$) to obtain a $3\text{ wt}\%$ dispersion. Potassium hydroxide (KOH; mass equal to that of Kevlar pulp; Sigma-Aldrich, reagent grade, 90% purity) was added at a Kevlar:KOH mass ratio of $1:1$ for deprotonation. The mixture was magnetically stirred at 90°C for 7 days. PVA was dissolved in DMSO ($15\text{ wt}\%$) at 90°C under magnetic stirring for 7 days to ensure complete dissolution. The ANF dispersion and PVA solution were mixed at a $1:1$ mass ratio, and subsequent solvent exchange with water yielded an ANF-PVA hydrogel^{31,34}. The porosity of the hydrogel template was adjusted by diluting the ANF dispersion and PVA solution with additional DMSO while maintaining the final ANF:PVA mass ratio at $1:5$. The hydrogel served as the template for integrating the conductive polypyrrole polymer. In situ chemical oxidative polymerization of pyrrole within the ANF-PVA template yielded ANF-PVA-PPy composites. A certain volume of $7\text{ wt}\%$ pyrrole (Sigma-Aldrich, reagent grade, 98%) solution in degassed DI water was poured on top of the ANF-PVA hydrogel overnight in a sealed container. The same volume of iron(III) chloride (anhydrous FeCl_3 ; Aladdin, $>98\%$ purity) solution with a molar ratio of $1.2:1$ of FeCl_3 to Py was added on top of ANF-PVA. Polymerization was then conducted in an ice-water bath maintained at $0\text{--}5^\circ\text{C}$. The diffusion/reaction time was 0.75 , 2 , 4 , or 6 h , depending on the sample.

The resulting ANF-PVA-PPy hybrid hydrogel was then rinsed with water to remove residual species, followed by solvent exchange with ethanol three times to remove water, and dried by critical point drying (CPD, Tousimis Autosamdri 931) to produce monolithic gradient conductive aerogels (MCAs).

For the three-layer stepwise conductivity-gradient control, three ANF-PVA-PPy layers with different conductivities were prepared from the same material system and assembled in a low-to-high conductivity sequence through the thickness to approximate the continuous conductivity profile. The total thickness of the layered control was kept at

$\sim 5\text{ mm}$ for fair comparison in waveguide EMI measurements and simulation.

For kirigami processing, ANF-PVA-PPy hydrogel films (thickness $\sim 1\text{ mm}$; diffusion time 0.25 h) were patterned by laser cutting, solvent-exchanged with ethanol overnight, and dried by CPD to obtain kirigami MCAs.

Structural characterization of MCAs

Scanning electron microscopy (SEM) combined with an elemental analyzer was utilized to visualize the morphology and distribution of PPy across the depth of the composite. Fourier-transform infrared spectroscopy (FTIR) was performed to verify chemical interactions among ANF, PVA, and PPy. Thermal stability was characterized by thermogravimetric analysis (TGA). Pore size distribution was measured by mercury intrusion porosimeter (MIP, AutoPore V 9600) with pressure ranging from 0.1 to $6.1 \times 10^4\text{ psi}$ for samples from different depths and/or with different bulk densities. The specific surface area was determined by the Brunauer–Emmett–Teller (BET) method. The spatial distribution of PPy was determined by an elemental analyzer (EuroVector EA-Nu Perspective IRMS) after sectioning the bulk samples into serial slices along the thickness direction (PPy content was calculated from element content using a calibration and stoichiometric conversion). The depth-profiled PPy $\text{wt}\%$ was converted to pyrrole molar units for consistency and comparison with numerical simulation.

Porosity was calculated as

$$\text{Porosity} = \left(1 - \frac{\rho_{\text{bulk}}}{\rho_{\text{solid}}}\right) \times 100\% \quad (\text{S1})$$

where ρ_{bulk} is the measured bulk density of MCAs, ρ_{solid} is the density of the solid framework estimated from the mass fractions of ANF/PVA/PPy:

$$\rho_{\text{solid}} = \omega_{\text{ANF}}\rho_{\text{ANF}} + \omega_{\text{PVA}}\rho_{\text{PVA}} + \omega_{\text{PPy}}\rho_{\text{PPy}} \quad (\text{S2})$$

Here, ρ_{ANF} (or PPTA, 1.44 g cm^{-3}), ρ_{PVA} (1.19 g cm^{-3}) were taken from supplier datasheets, and $\omega_{\text{ANF}}:\omega_{\text{PVA}}:\omega_{\text{PPy}} = 13:67:20$ (by mass) for MCA-94.

Electrical conductivity measurement

Electrical conductivity was measured using a source meter (Keithley 2450) and a standard four-point probe system (HPS2661, Helpass electronic technologies). For sectioned strip specimens (length $l = 25\text{ mm}$, width $w = 4\text{ mm}$, thickness t), a two-probe end-to-end configuration was used by applying a bias voltage. The conductivity was calculated as

$$\sigma = \left(\frac{l}{V}\right) \left(\frac{l}{w \cdot t}\right) \quad (\text{S3})$$

where σ is the conductivity, V is the voltage, I is the current. Thickness t was measured for each specimen and used in the calculation. For square slices ($25\text{ mm} \times 25\text{ mm}$), the in-plane resistivity ρ was measured using the four-point probe, and the conductivity was calculated as $\sigma = 1/\rho$.³⁷

EMI shielding performance measurement

The electromagnetic interference (EMI) shielding performance of MCAs was quantitatively evaluated using a vector network analyzer (VNA, Agilent PNA N5244A) in conjunction with a waveguide measurement system. Before each measurement, a full two-port waveguide TRL calibration was performed for the corresponding frequency band. The calibration used standard Thru, Reflect, and Line waveguide standards: the Reflect standard was realized using short plates at the

two ports, the Thru standard was obtained by directly connecting the two waveguide ports, and the Line standard was an inserted waveguide section with a known electrical length. The calibration reference planes were set at the waveguide flanges. This procedure removed systematic errors from the VNA, waveguide adapters, and fixture, ensuring that the measured S-parameters were referenced to a well-defined baseline. MCA specimens were cut to match the internal cross-sectional dimensions of standard rectangular waveguides corresponding to the X (8.2–12.4 GHz), Ku (12.4–18.0 GHz), K (18.0–26.5 GHz), and Ka (26.5–40.0 GHz) bands. The in-plane dimensions of the samples were 22.9×10.2 mm, 15.8×7.9 mm, 10.7×4.3 mm, and 7.1×3.6 mm, respectively, to ensure precise fitting within the corresponding waveguide apertures and minimize possible edge leakage. To evaluate repeatability, 3–5 independent MCA specimens were measured under identical waveguide conditions after separate mounting. Only specimens that fully filled the waveguide cross-section without visible edge gaps were used. Unless otherwise noted, the bulk MCA specimens used for cross-sample comparison were prepared at the same thickness (~ 5 mm). Measurements were performed with incidence from the low- σ face unless otherwise noted. For the kirigami samples, the wavelength in the criterion $L_c < \lambda_0/4$ refers to the free-space wavelength λ_0 , not the guide wavelength λ_g . We used the upper X-band frequency (12.4 GHz) to define a conservative bound, giving $\lambda_0/4 = 6.04$ mm; thus, both kirigami patterns used here ($L_c = 2.4$ and 4.8 mm) satisfy this condition throughout 8.2–12.4 GHz. The scattering parameters (S-parameters) obtained from the VNA were used to calculate the total shielding effectiveness (SE_T), as well as its reflection and absorption components⁷⁰. The total shielding effectiveness (SE_T) is defined as:

$$SE_T = -10 \log(|S_{21}|^2) \quad (S4)$$

$$SE_R = -10 \log(1 - |S_{11}|^2) \quad (S5)$$

$$SE_A = -10 \log\left(\frac{|S_{21}|^2}{1 - |S_{11}|^2}\right) = SE_T - SE_R \quad (S6)$$

The reflection (R), absorption (A), and transmission (T) were calculated based on the measured S-parameters using the following equations:

$$R = |S_{11}|^2 \quad (S7)$$

$$T = |S_{21}|^2 \quad (S8)$$

$$A = 1 - R - T \quad (S9)$$

The skin depth δ , defined as the distance over which the field amplitude decays to $1/e$, was estimated for a good conductor as $\delta = \frac{1}{\sqrt{\pi\sigma\mu f}}$, where σ is the electrical conductivity, μ is the magnetic permeability, and f is the frequency.

Mechanical test

Uniaxial tensile properties of MCAs were evaluated using samples processed into a standard dumbbell shape (25 mm in length and 5 mm in width). Tests were performed on a Zwick Roell universal testing machine at a constant nominal strain rate of $100\% \text{ min}^{-1}$. Compressive behavior was characterized using cylindrical specimens (10 mm in diameter) subjected to a compressive deformation rate of $50\% \text{ min}^{-1}$. The mechanical toughness was estimated by calculating the area under the engineering stress–strain curve up to fracture, and the results were expressed in kJ m^{-3} .

To quantify the fracture energy (Γ), both trouser-tear and pure-shear tests were employed. For the tearing test, composite films (8 mm

in width and 50 mm in length) were fabricated and pre-notched (notch length: 20 mm) in a trouser-shaped configuration. The samples were loaded in tension at a constant crosshead speed of 1 mm s^{-1} , and the steady-state tearing force (F) was used to determine the fracture energy according to:

$$\Gamma = \frac{2F}{t} \quad (S10)$$

where t denotes the film thickness.

For the pure-shear test, rectangular samples (30 mm in width and 20 mm in length) were notched to a depth of 15 mm and mounted between grips with a fixed span of 15 mm. The samples were extended at a constant rate of 1 mm s^{-1} . The corresponding fracture energy was calculated from the mechanical work $U(L_c)$ done on the unnotched specimen up to the critical extension L_c , corresponding to the onset of crack propagation in the notched sample:

$$\Gamma = \frac{U(L_c)}{w \cdot t} \quad (S11)$$

where w and t represent the width and thickness of the sample, respectively. The two independent methods yielded consistent estimates of Γ , validating the reliability of the fracture energy measurements (Supplementary Fig. 24).

COMSOL multiphysics simulation of diffusion-reaction coupling

To model the depth-dependent PPy formation in porous ANF-PVA hydrogels, a time-dependent diffusion-reaction model was implemented in COMSOL Multiphysics (v6.1) using the transport of diluted species in porous media interface. A 3D cylindrical geometry was used to represent the hydrogel, with the thickness set to match the experimental sample. The transport of Py and Fe^{3+} was governed by Fick's second law:

$$\frac{\partial C_i}{\partial t} = \nabla \cdot (D_{\text{eff},i} \nabla C_i) - R_i \quad (S12)$$

The effective diffusion coefficient (D_{eff}) in the hydrogel matrix was computed using modified Einstein equation of Brownian motion and the equations for semi-empirical pore structure^{42,71}

$$D_{\text{eff}} = \frac{D_0}{1 + p(1 - \phi)} \quad (S13)$$

where D_0 is bulk solution diffusion coefficient, ϕ is porosity, and p is the structure factor calibrated to match the experimentally measured depth profiles, accounting for tortuosity-related transport hindrance.

Pyrrole polymerization follows a well-established sequence (Supplementary Fig. 5)^{39–41}. The reaction results in the rapid formation of high-molecular-weight, insoluble PPy. The high surface area of the ANF-PVA template provides abundant active reaction sites, which could accelerate polymerization⁷². The kinetics of this oxidative polymerization can be described by the following equation^{39,73}:

$$-\frac{d[\text{Py}]}{dt} = k_1 [\text{Fe}^{3+}] [\text{Py}] + k_2 [\text{Py}][\text{P}] \quad (S14)$$

with the assumption that the concentration of oxidized PPy oligomer be simplified as the same as the generated oligomer⁷³:

$$[\text{P}] = [\text{Py}]_0 - [\text{Py}] \quad (S15)$$

where $[\text{Fe}^{3+}]$, $[\text{Py}]$, and $[\text{P}]$, $[\text{Py}]_0$ are the concentrations of Fe^{3+} , Py monomer, polymerized oligomers, and the initial concentration of Py,

respectively. k_1 and k_2 represent the rate constants of the initiation (Supplementary Fig. 5, Eq. (1)) and chain propagation (Supplementary Fig. 5, Eq. (2), (4), and (6)), respectively. Thus, the rate equation can be simplified to:

$$-\frac{d[\text{Py}]}{dt} = k_1[\text{Fe}^{3+}][\text{Py}] + k_2[\text{Py}]_0[\text{Py}] - k_2[\text{Py}]^2 \quad (\text{S16})$$

Rate constants k_1 and k_2 were taken from the literature and further refined by fitting to the experimentally measured depth profiles.

Theoretical model of electromagnetic shielding effectiveness in near-continuous conductivity gradient structures

In the GSM, each stratum is treated as a homogeneous and isotropic effective medium. Each layer is $\mu_r \approx 1$, $\varepsilon_r' = 3.5$, and $\tan\delta = 0.07$ unless otherwise stated. The conductivity is introduced as the only depth-dependent parameter, following the experimentally fitted profile $\sigma(z) = \sigma_0 \exp(-z/\lambda)$; no explicit frequency dependence of σ is included. This reduced-order treatment absorbs the effects of porosity into the effective constitutive parameters, with the average contribution represented by background dielectric response, whereas sample-specific influence is captured by $\sigma(z)$, because porosity and connectivity regulate PPy penetration depth, conductive-network formation, and the resulting $\sigma_0 - \lambda$ gradient. This separation avoids double-counting the conductive contribution in both ε'' and $\sigma(z)$. The model is intended to isolate the first-order influence of the conductivity gradient on R , A , and T for mechanism analysis and parameter mapping, rather than to reconstruct the full local constitutive response of the graded porous composite.

For a passive, homogeneous medium characterized by permittivity ε , permeability μ , and electrical conductivity σ , the electromagnetic behavior is governed by Maxwell's equations:

$$\begin{aligned} \nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} \\ \nabla \times \mathbf{B} &= \mu\sigma\mathbf{E} + \mu\varepsilon\frac{\partial \mathbf{E}}{\partial t} \\ \nabla \cdot \mathbf{D} &= 0 \\ \nabla \cdot \mathbf{B} &= 0 \end{aligned} \quad (\text{S17})$$

By combining these equations, one obtains the general wave equation:

$$\begin{aligned} \nabla^2 \mathbf{E} &= \mu\sigma\frac{\partial \mathbf{E}}{\partial t} + \mu\varepsilon\frac{\partial^2 \mathbf{E}}{\partial t^2} \\ \nabla^2 \mathbf{H} &= \mu\sigma\frac{\partial \mathbf{H}}{\partial t} + \mu\varepsilon\frac{\partial^2 \mathbf{H}}{\partial t^2} \end{aligned} \quad (\text{S18})$$

This equation describes the behavior of electromagnetic fields in a passive, homogeneous, and conductive medium. For time-varying electromagnetic fields, it admits traveling wave solutions $Ae^{j\omega t}e^{\pm j\omega\sqrt{\mu(\varepsilon-j\frac{\sigma}{\omega})}z}$. The presence of a first-order derivative term in this second-order differential equation indicates energy loss during wave propagation through the medium. Considering a plane wave propagating in the z -direction, the electric and magnetic fields contain no longitudinal components. Such waves are referred to as transverse electromagnetic waves.

For the multilayer laminate structure illustrated in Supplementary Fig. 17, when a time-harmonic electromagnetic wave ($e^{j\omega t}$) is normally incident from air onto the stratified slab along the z -direction, multiple reflections and transmissions occur at each interface. In the m -th layer, these interactions result in waves with identical frequency but varying directions and phases. These waves can be equivalently expressed as a superposition of two waves traveling in the $+z$ and $-z$ directions. The electric field in the x -direction and the magnetic field in the y -direction

in the m -th layer can be written as:

$$E_m = A_m e^{-jk_m z} + B_m e^{jk_m z}, H_m = Y_m (A_m e^{-jk_m z} - B_m e^{jk_m z}) \quad (\text{S19})$$

where A_m and B_m denote the amplitudes of the forward and backward waves, respectively, k_m is the complex wave number, and Y_m is the intrinsic conductance of the m -th medium, given by:

$$Y_m = Y_0 \sqrt{\frac{(\varepsilon_m - j\frac{\sigma_m}{\omega\varepsilon_0})}{\mu_m}}, \quad Y_0 = \sqrt{\frac{\varepsilon_0}{\mu_0}} \quad (\text{S20})$$

Letting ε_0 , μ_0 , and Y_0 be the permittivity, permeability, and intrinsic conductance of free space, and ε_m , μ_m , and σ_m be the relative permittivity, permeability, and electrical conductivity of the m -th layer, we have:

$$\begin{aligned} \varepsilon_m &= \varepsilon_0(\varepsilon'_{m,r} - j\varepsilon''_{m,r}) \\ \mu_m &= \mu_0(\mu'_{m,r} - j\mu''_{m,r}) \end{aligned} \quad (\text{S21})$$

The wave number in air

$$k_m = k_0 \sqrt{(\varepsilon_m - j\frac{\sigma}{\omega\varepsilon_0})\mu_m}, \quad k_0 = \omega\sqrt{\varepsilon_0\mu_0} \quad (\text{S22})$$

To analyze reflection, transmission, and attenuation, we start from interface 0. At this interface, the continuity of the tangential components of $\mathbf{E}$ and $\mathbf{H}$ yields:

$$\begin{cases} A_1 e^{-jk_1 d_1} + B_1 e^{jk_1 d_1} = A_t \\ Y_1 A_1 e^{-jk_1 d_1} - Y_1 B_1 e^{jk_1 d_1} = Y_t A_t \end{cases} \quad (\text{S23})$$

At interface 1, a similar continuity condition gives:

$$\begin{cases} A_2 e^{-jk_2 d_2} + B_2 e^{jk_2 d_2} = A_1 + B_1 \\ Y_2 A_2 e^{-jk_2 d_2} - Y_2 B_2 e^{jk_2 d_2} = Y_1 (A_1 - B_1) \end{cases} \quad (\text{S24})$$

Let:

$$\begin{cases} a_2 = A_2 e^{-jk_2 d_2} \\ b_2 = B_2 e^{jk_2 d_2} \end{cases} \quad (\text{S25})$$

Then, the reflection and transmission coefficients are:

$$\begin{aligned} r_2 &= \frac{b_2}{a_2} = \frac{Z_{in,1} - Z_2}{Z_{in,1} + Z_2} \\ R_2 &= \frac{B_2}{A_2} = \frac{Z_{in,1} - Z_2}{Z_{in,1} + Z_2} e^{-2jk_2 d_2} \\ t_2 &= \frac{A_t}{a_2} = \frac{2Z_{tr,1}}{Z_{in,1} + Z_2} \\ T_2 &= \frac{A_t}{A_2} = \frac{2Z_{tr,1}}{Z_{in,1} + Z_2} e^{-jk_2 d_2} \end{aligned} \quad (\text{S26})$$

with:

$$\begin{aligned} Z_{in,1} &= Z_1 \frac{Z_1 + jZ_1 \tan(k_1 d_1)}{Z_1 + jZ_1 \tan(k_1 d_1)} \\ Z_{tr,1} &= \frac{Z_1 Z_t}{Z_1 \cos(k_1 t_1) + jZ_t \sin(k_1 t_1)} \end{aligned} \quad (\text{S27})$$

At interface $n-1$, we similarly define:

$$\begin{aligned} r_m &= \frac{b_m}{a_m} = \frac{Z_{in,m-1} - Z_m}{Z_{in,m-1} + Z_m} \\ R_m &= \frac{B_m}{A_m} = \frac{Z_{in,m-1} - Z_m}{Z_{in,m-1} + Z_m} e^{-2jk_m d_m} \\ t_m &= \frac{A_t}{a_m} = \frac{2Z_{tr,m-1}}{Z_{in,m-1} + Z_m} \\ T_m &= \frac{A_t}{A_m} = \frac{2Z_{tr,m-1}}{Z_{in,m-1} + Z_m} e^{-jk_m d_m} \end{aligned} \quad (\text{S28})$$

where:

$$\begin{aligned} Z_{in,n-1} &= Z_{n-1} \frac{Z_{in,n-2} + jZ_{n-1} \tan(k_{n-1}d_{n-1})}{Z_{n-1} + jZ_{in,n-2} \tan(k_{n-1}d_{n-1})} \\ Z_{tr,n-1} &= \frac{Z_{n-1} Z_{tr,n-2}}{Z_{n-1} \cos(k_{n-1}t_{n-1}) + jZ_{in,n-2} \sin(k_{n-1}t_{n-1})} \end{aligned} \quad (S29)$$

At interface m , the continuity conditions become:

$$\begin{cases} A_{m+1}e^{-jk_{m+1}d_{m+1}} + B_{m+1}e^{jk_{m+1}d_{m+1}} = A_m + B_m \\ Y_{m+1}A_{m+1}e^{-jk_{m+1}d_{m+1}} - Y_{m+1}B_{m+1}e^{jk_{m+1}d_{m+1}} = Y_m(A_m - B_m) \end{cases} \quad (S30)$$

Letting:

$$\begin{cases} a_{m+1} = A_{m+1}e^{-jk_{m+1}d_{m+1}} \\ b_{m+1} = B_{m+1}e^{jk_{m+1}d_{m+1}} \end{cases} \quad (S31)$$

Equation (S30) can be simplified and solved:

$$\begin{cases} a_{m+1} + b_{m+1} = A_m + B_m \\ Y_{m+1}a_{m+1} - Y_{m+1}b_{m+1} = Y_m(A_m - B_m) \end{cases} \quad (S32)$$

Solving (S32) yields:

$$r_{m+1} = \frac{b_{m+1}}{a_{m+1}} = \frac{Z_{in,m} - Z_{m+1}}{Z_{in,m} + Z_{m+1}} \quad (S33)$$

Further solving (S32) yields:

$$\frac{A_m}{a_{m+1}} = \frac{Z_{in,m} + Z_{m+1}}{Z_{in,m} + Z_{m+1}} \quad (S34)$$

Using Eq. (S28), we obtain:

$$t_{m+1} = \frac{A_t}{a_{m+1}} = \frac{A_m}{a_{m+1}} \frac{A_t}{A_m} = \frac{2Z_{tr,m}}{Z_{in,m} + Z_{m+1}} \quad (S35)$$

Thus, from Eq. (S28) at interface $m-1$, we derive the expressions at interface m :

$$\begin{aligned} r_{m+1} &= \frac{b_{m+1}}{a_{m+1}} = \frac{Z_{in,m} - Z_{m+1}}{Z_{in,m} + Z_{m+1}} \\ t_{m+1} &= \frac{A_t}{a_{m+1}} = \frac{2Z_{tr,m}}{Z_{in,m} + Z_{m+1}} \end{aligned} \quad (S36)$$

with:

$$\begin{aligned} Z_{in,m} &= \frac{Z_{in,m-1}Z_m + jZ_m^2 \tan(k_m d_m)}{jZ_{in,m-1} \tan(k_m d_m) + Z_m} \\ Z_{tr,m} &= \frac{Z_m Z_{tr,m-1}}{Z_m \cos(k_m t_m) + jZ_{in,m-1} \sin(k_m t_m)} \end{aligned} \quad (S37)$$

Equations (S36) and (S37) are structurally consistent with Eqs. (S28) and (S29), validating the recursive approach for multilayer systems.

Thus, a general method has been established to predict the shielding effectiveness of multilayer conductive media with reflection, attenuation, and transmission effects.

Numerical simulation of electromagnetic shielding effectiveness in layered conductive structures

Numerical simulations were carried out using the electromagnetic waves, frequency domain module of COMSOL Multiphysics, a finite-element analysis software, to solve the governing equation,

$$\nabla \times \frac{1}{\mu_r} (\nabla \times \mathbf{E}) - k_0^2 (\epsilon_r - \frac{j\sigma}{\omega\epsilon_0}) \mathbf{E} = 0 \quad (S38)$$

A two-dimensional homogenized geometric model was constructed, incorporating realistic physical parameters. Perfectly matched layers (PMLs) were applied at the boundaries of the air domain to absorb outgoing waves and suppress reflections. Floquet periodic boundary conditions were employed to reduce computational costs by leveraging the periodicity of the structure. To ensure numerical accuracy, the maximum mesh element size was set to be smaller than one-tenth of the minimum incident wavelength.

A waveguide port with a 1 W input power in the electric field mode was introduced in the air domain to excite the structure and calculate the scattering parameters (S-parameters) at 10 GHz. Spatial distributions of electric fields, magnetic fields, electromagnetic energy density, and electromagnetic loss were extracted to analyze and interpret the energy dissipation mechanisms. The simulated S-parameters were compared with theoretical predictions, demonstrating excellent agreement. This consistency validates the accuracy and robustness of the developed theoretical model.

Data availability

The datasets generated and/or analyzed during the current study have been deposited in the Figshare repository at <https://doi.org/10.6084/m9.figshare.32393211>. All data were available from the corresponding author upon request.

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Author contributions

L.X., Z.Z., X.L., and Y.C. designed the research. Y.C. carried out experimental investigations. Y.C. carried out a simulation on diffusion-reaction

coupling. C.W. and Y.C. designed and conducted theoretical modeling and simulation on continuously graded conductors. H.H., N.W., S.Z., J.Z., Y.L., and J.C. contributed to discussions, experimental support, and analysis. Y.C. drafted the manuscript, Z.Z., X.L., L.X., C.W., and H.H. reviewed and edited it. L.X. and Z.Z. supervised the whole project.

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Competing interests

The authors declare no competing interests.

Additional information

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