

Supplementary Information

Nanofibrous Aerogels with Continuous Conductivity Gradient for Ultrabroadband Absorption-Dominant Electromagnetic Interference Shielding

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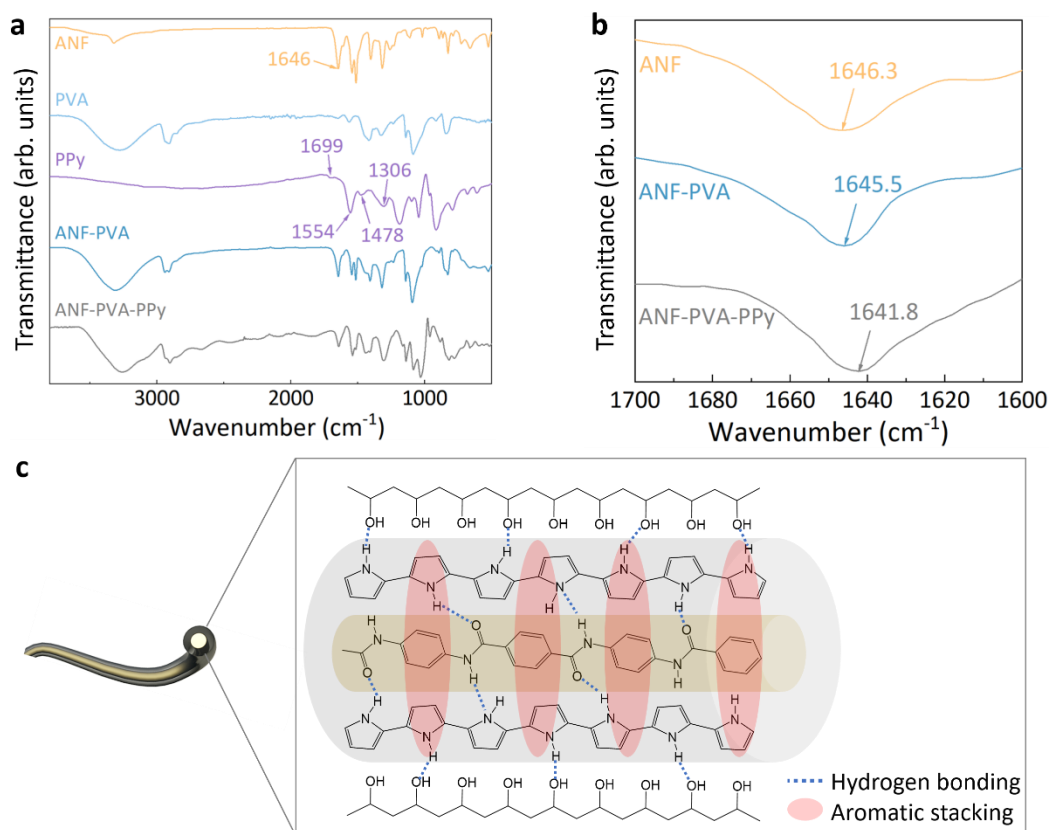
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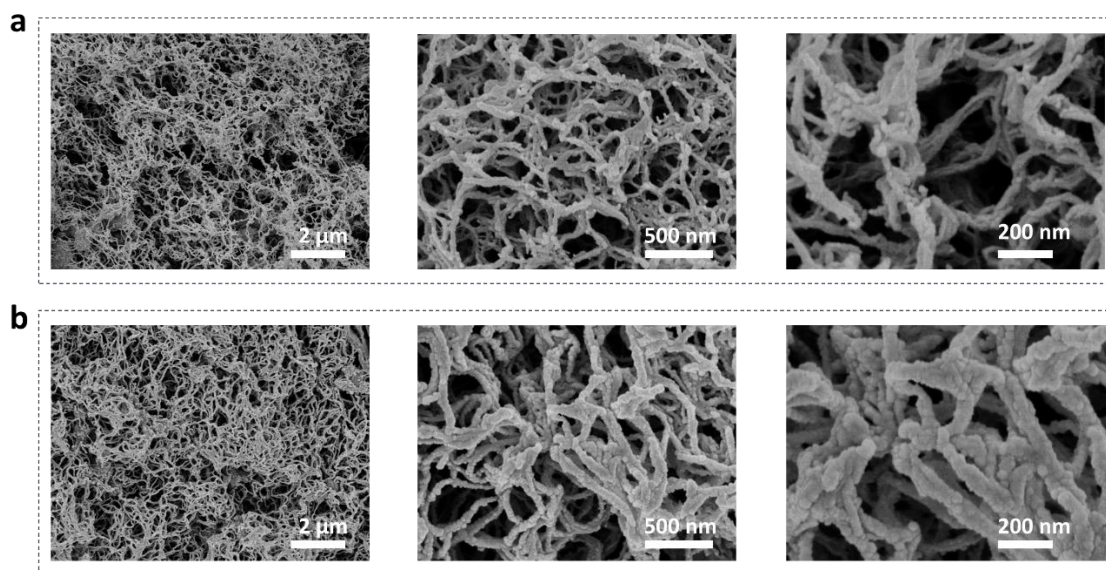
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Supplementary Figures 1-26

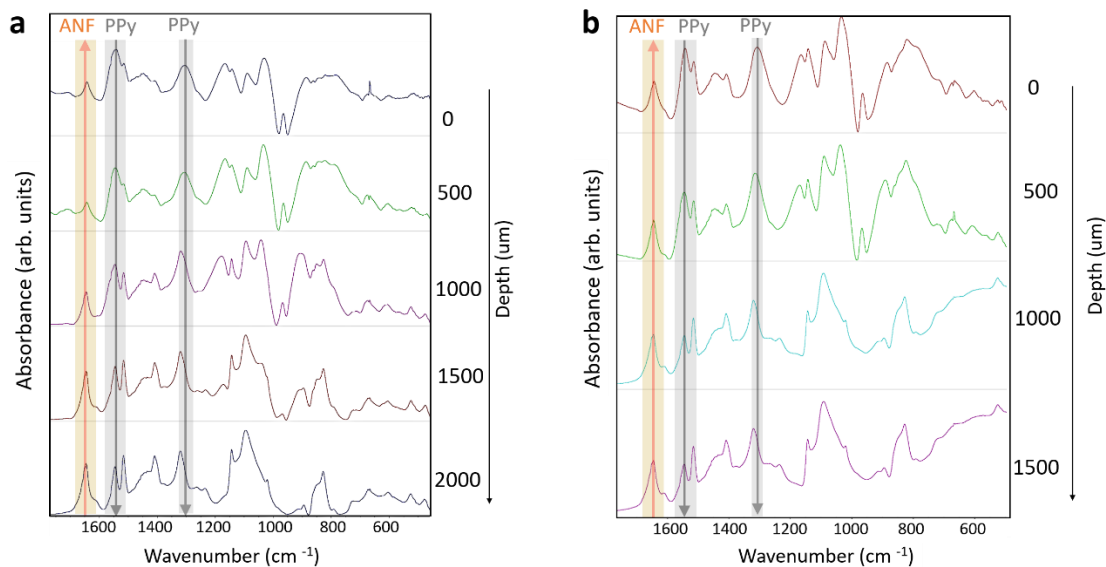
Supplementary Tables 1-3



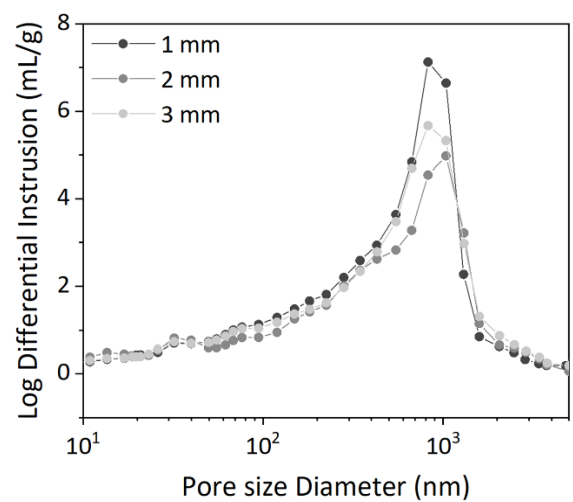
Supplementary Fig. 1 Chemical structures characterized by FTIR and intermolecular interactions of different polymers in MCAs (i.e., ANF-PVA-PPy). a. FTIR transmittance spectra of ANF, PVA, PPy, ANF-PVA, and ANF-PVA-PPy. For polypyrrole, the peaks at 1554 cm^{-1} and 1478 cm^{-1} correspond to the characteristic C=C stretching within the conjugated polymer backbone. Peaks at 1699 cm^{-1} and 1306 cm^{-1} are attributed to the stretching vibrations of C=N and C–N bonds, respectively.¹ For ANF, the peak at 1648 cm^{-1} is contributed by the amide C=O stretching. The spectral features observed in this study align well with previously reported data, validating the successful synthesis of polypyrrole. b. Enlarged view of the $1700\text{--}1600\text{ cm}^{-1}$ region, highlighting the evolution of the amide C=O stretching.² The characteristic peak at 1646.3 cm^{-1} for ANF red shifts slightly to 1645.5 cm^{-1} in ANF-PVA due to hydrogen bonding interactions between ANF and PVA. A more pronounced redshift to 1641.8 cm^{-1} in the ANF-PVA-PPy composite indicates enhanced hydrogen bonding between ANF and PPy. c. Schematic illustration of possible intermolecular interactions within the ANF-PVA-PPy network. π – π stacking and hydrogen bonding are proposed between the aromatic rings and amide groups of PPTA and the PPy chains. Concurrently, PVA forms hydrogen bonds with both PPTA and PPy, facilitating the formation of a physically interconnected hybrid network. FTIR spectra are vertically offset for clarity.



Supplementary Fig. 2 SEM images of ANF-PVA (a) and ANF-PVA-PPy (b) with 94% porosity.

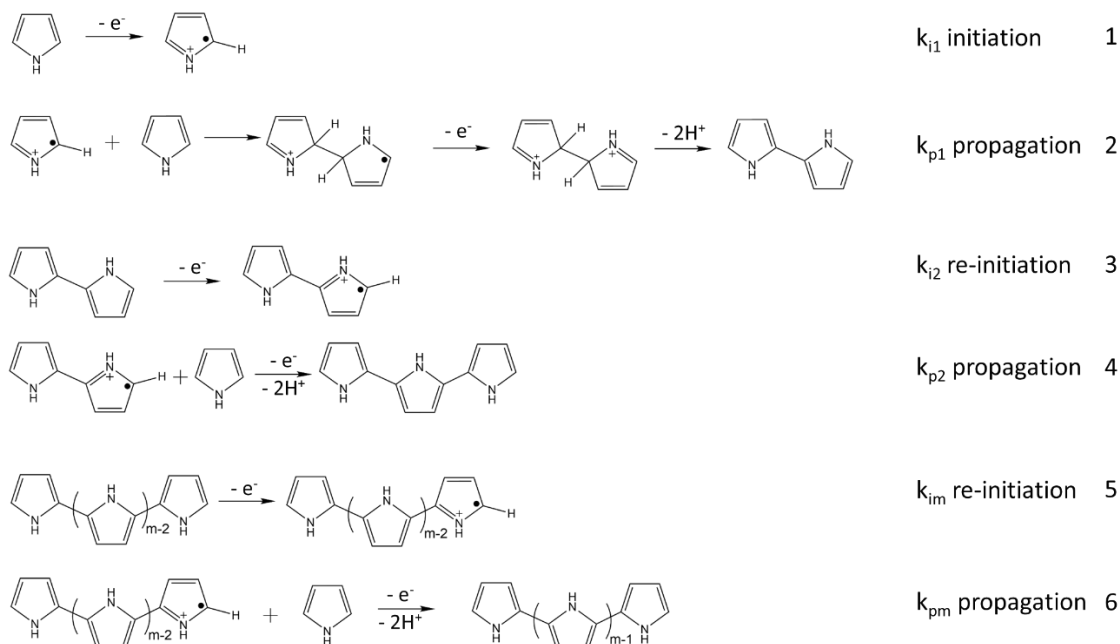


Supplementary Fig. 3 ATR-FTIR absorbance spectra of MCA-88 (a) and MCA-75 (b) samples with downward sliced layers along depth. Both figures exhibit decreasing characteristic peaks for PPy (1554 cm^{-1} for C=C stretching, 1306 cm^{-1} for C-N stretching) and increasing characteristic peaks for ANF (1646 cm^{-1} for C=O stretching). The variation in characteristic peaks is consistent across MCA-88 and MCA-75 samples, indicating a decreasing content of PPy and an increasing content of ANF in the depth direction for MCAs.



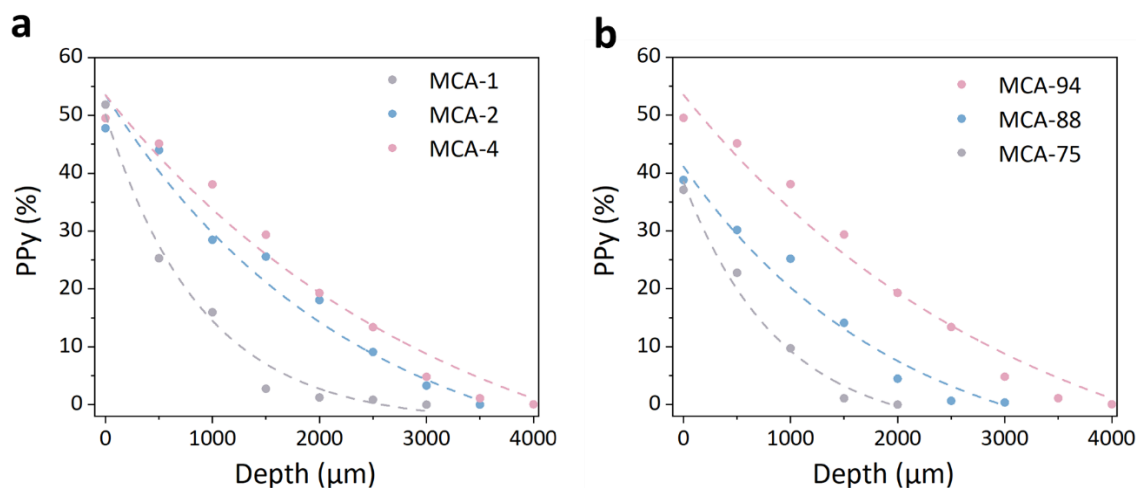
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62 **Supplementary Fig. 4** Pore size distribution of ANF-PVA with 94% porosity at various
 63 depths of the bulk sample.

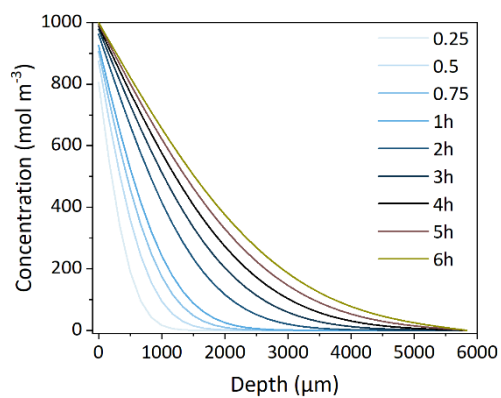


64

65 **Supplementary Fig. 5** The proposed mechanism of PPy polymerization. The radical cation
 66 reacts with a neutral monomer (reaction 1), followed by oxidation and deprotonation to
 67 yield a dimer (reaction 2). The dimer is then oxidized immediately, forming a dimeric
 68 radical cation (reaction 3), which attacks another monomer, leading to the formation of a
 69 trimer (reaction 4). This repeated process results in polymer chain growth and ultimately
 70 produces the polymer (reactions 5 and 6).³

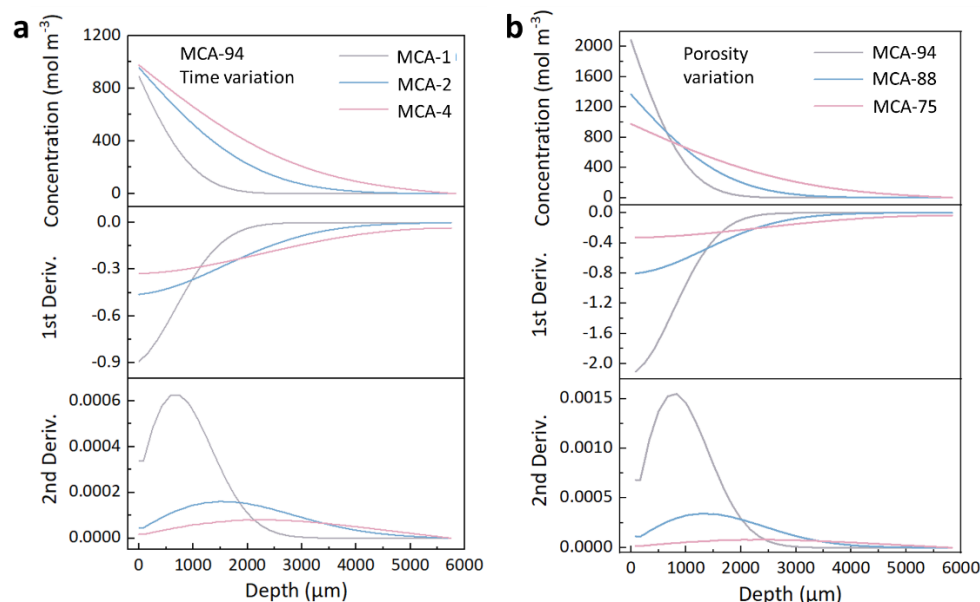


Supplementary Fig. 6 Experimental results of the variation of PPy content in weight percent units. a. The variation of PPy content (represented by PPy weight percent) at different depths of MCA-94 bulk samples with varying diffusion/reaction times. b. The variation of PPy content (represented by PPy weight percent) in MCAs with various porosities when the diffusion/reaction time is 4 h. (Dots represent experimental results; dashed lines indicate fitting curves.)

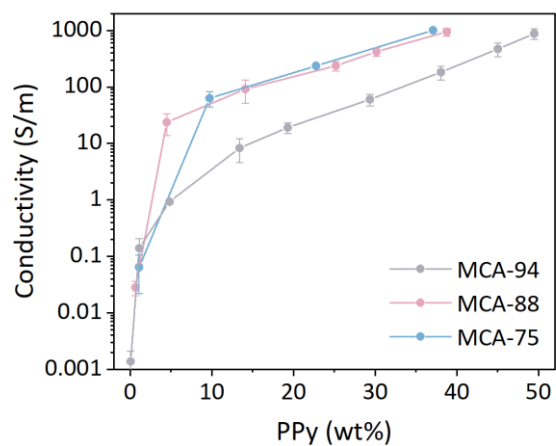


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79 **Supplementary Fig. 7** Simulation results of varied gradients of PPy content (represented
 80 by pyrrole molar concentration) at different depths of bulk aerogel with varying
 81 diffusion/reaction times.

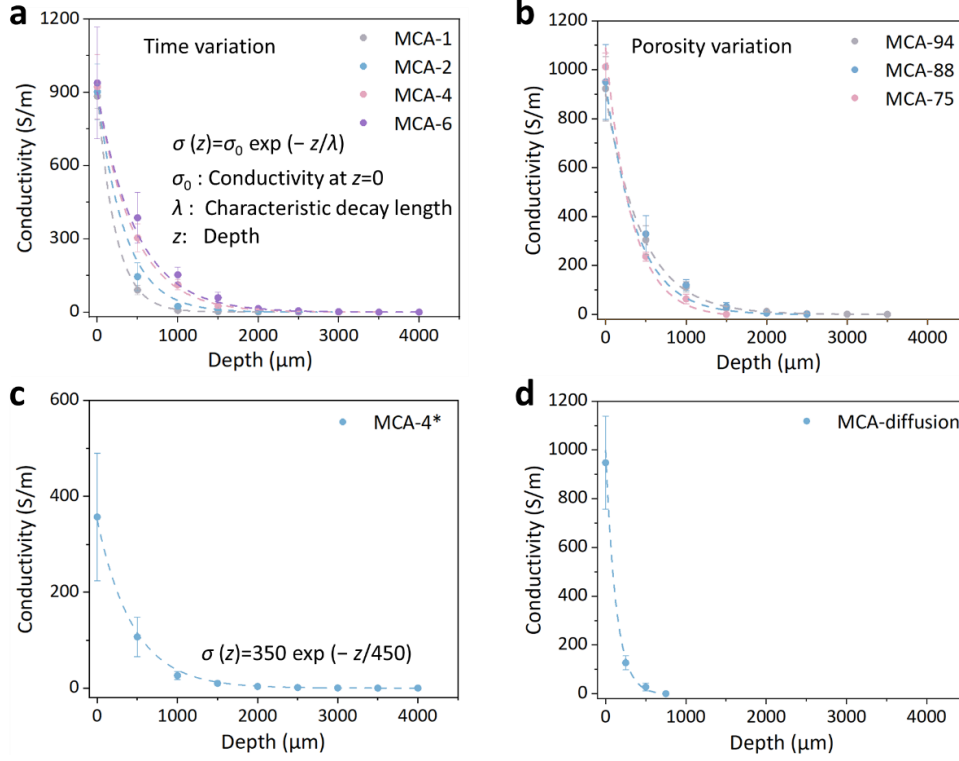


Supplementary Fig. 8 The characteristics of the simulated PPy content gradients (represented by pyrrole molar concentration). a. The concentration gradient, first and second derivatives of the PPy content gradient at downward depth with various diffusion/reaction times of the MCA-94 sample. In the first 1 mm below the top surface, a short reaction time (0.75 h) produces a sharper PPy-content drop, whereas a prolonged reaction time (4 h) yields a smoother concentration profile over the same region, corresponding to a smaller derivative magnitude and weaker curvature. b. The first and second derivatives of the PPy content gradient at a downward depth of MCAs with various porosities when the diffusion/reaction time is 4 h.



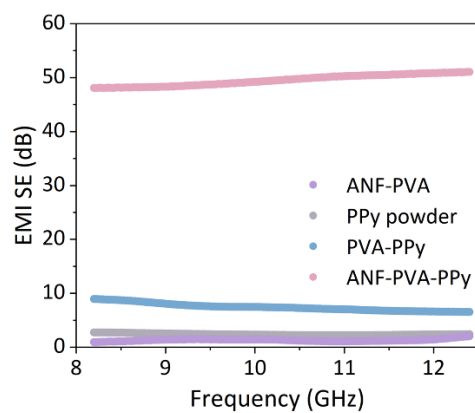
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94 **Supplementary Fig. 9** Conductivity variation as PPy wt% changes with different
 95 porosities. Data are presented as mean $\pm$ s.d. from three independent experiments (n=3);
 96 error bars represent s.d.



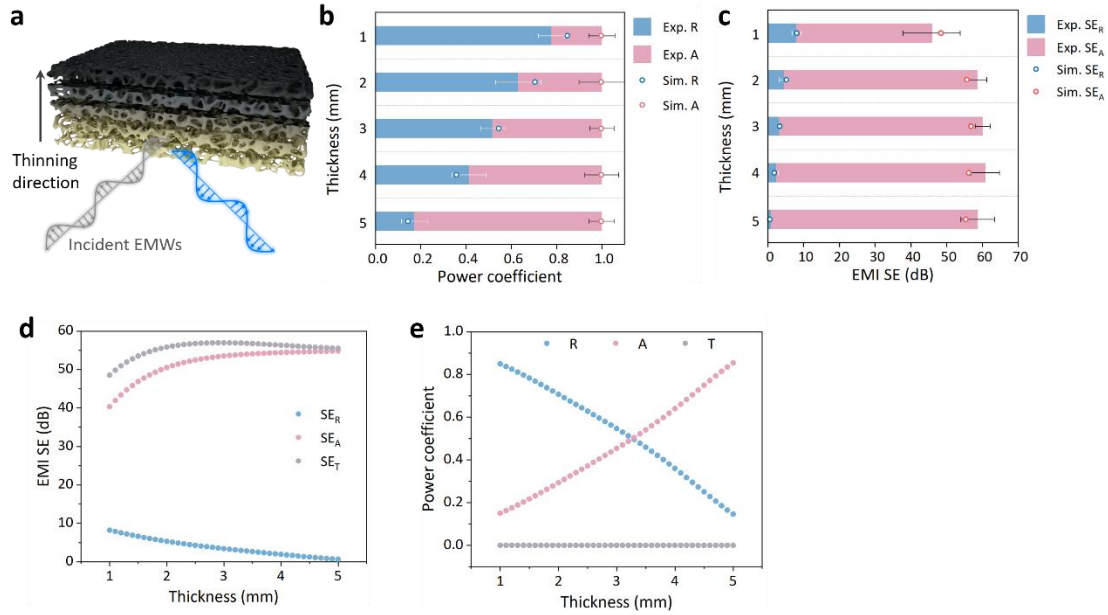
Supplementary Fig. 10 Through-thickness conductivity profiles of different MCAs plotted on a linear y-axis. a. Conductivity profiles versus depth for 94%-porosity of MCAs prepared with various diffusion/reaction times. b. Conductivity profiles versus depth for MCAs of different porosities prepared with a fixed diffusion/reaction time of 4 h. c. Conductivity profile of MCA-4* with reduced surface conductivity prepared in an oxygen-containing atmosphere. d. Conductivity profile of MCA-diffusion prepared by a non-gravity-driven reactant-permeation method and exhibiting a steeper gradient. Symbols represent experimental data and dashed lines are fits to $\sigma(z) = \sigma_0 \exp(-z/\lambda)$, where σ_0 is the surface conductivity at $z = 0$, λ is the characteristic decay length (μm), and z is measured from the high- σ surface toward the low- σ surface. Data points are presented as mean $\pm$ s.d. ($n = 3$ independent samples).

To assess thickness effects, we distinguish the sample thickness from the effective gradient length defined as the PPy-containing gradient region. Unless otherwise noted, the bulk MCAs compared in Fig. 3e–g and Supplementary Table 1 had the same thickness of ~ 5 mm. Shortening the diffusion/reaction time reduced the conductivity gradient length from ~ 4.0 to ~ 2.5 mm and decreased A from 0.84 to 0.76, while lowering the scaffold porosity further confined the gradient length to ~ 2.0 mm and reduced A to 0.67. In both cases, SE_T remained above 30 dB, indicating that millimeter-scale conductive thickness can provide strong attenuation but does not by itself ensure low reflection. In the present experimentally validated design, 5 mm is the minimum nominal thickness confirmed to satisfy both $R < 0.1$ and $SE_T > 30$ dB over 10.2–40 GHz.

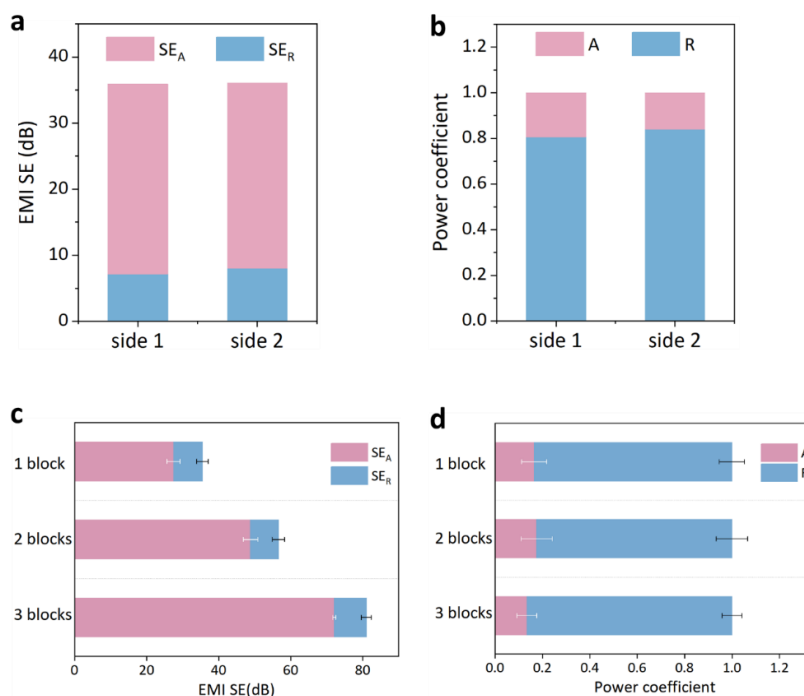


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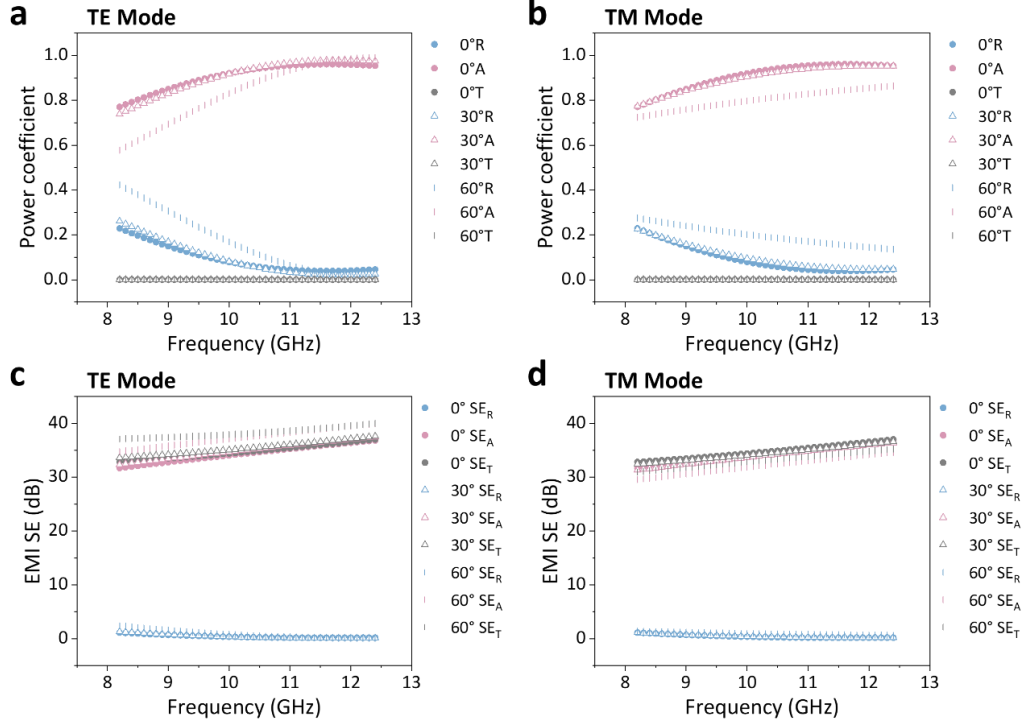
120 **Supplementary Fig. 11** EMI SE of ANF-PVA substrate, PPy powder, PVA-PPy by PPy in
 121 situ polymerization in PVA substrate, and ANF-PVA-PPy by PPy in situ polymerization in
 122 ANF-PVA substrate with porosity of 94%.



Supplementary Fig. 12 a. The thinning effect is conducted by sequentially cutting slices from the low- σ face. Power coefficients (b) and EMI SE (c) as a function of thickness when EMWs impinge from the low- σ face. The thickness is reduced from the low- σ face of sample MCA-94(4). Columns indicate experimental results; dots denote simulation results. For b and c, bars show the mean of three independently prepared specimens, each measured after sequential thinning, and error bars indicate s.d. ($n = 3$). The continuous thinning effect of EMI SE (d) and power coefficient (e) by simulation results.

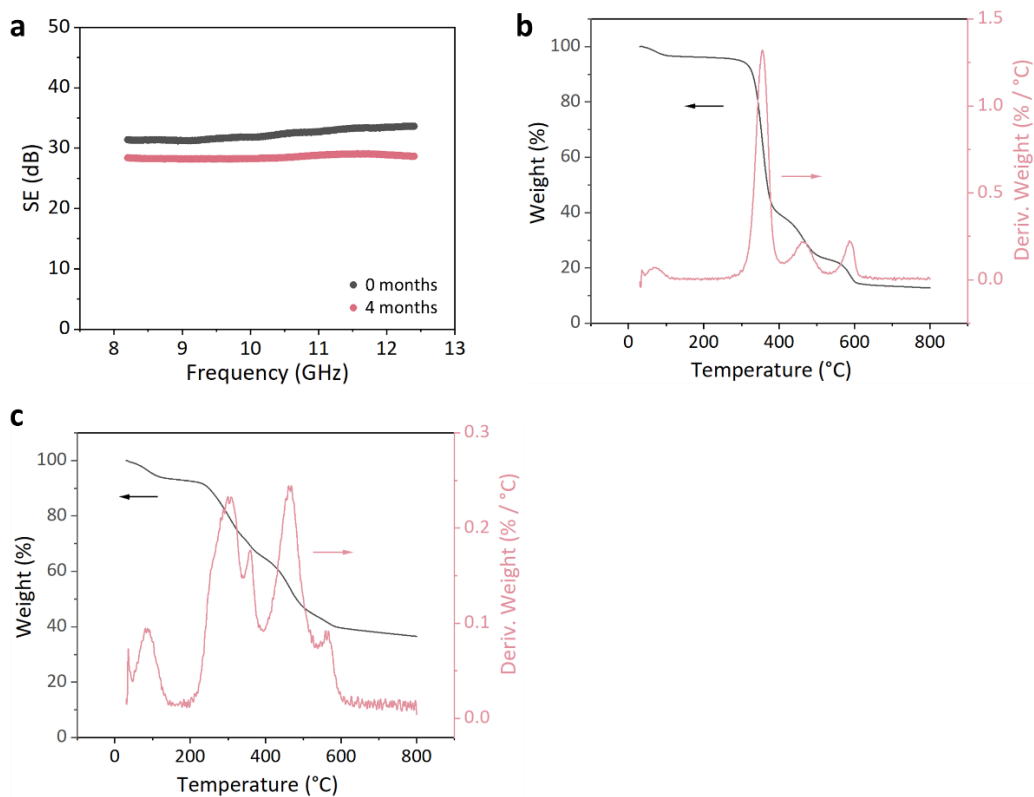


Supplementary Fig. 13 EMI shielding properties of ANF-PVA-PPy (94% porosity) aerogel blocks with uniformly distributed PPy throughout the depth. EMI SE_A , SE_R (a), and power coefficient (b) of a single block when incident from different faces. EMI SE_A , SE_R (c), and power coefficient (d) as a function of different numbers of stacking blocks. For c and d, stacked bars show the mean, and error bars indicate s.d.; $n=3$ independent specimens.

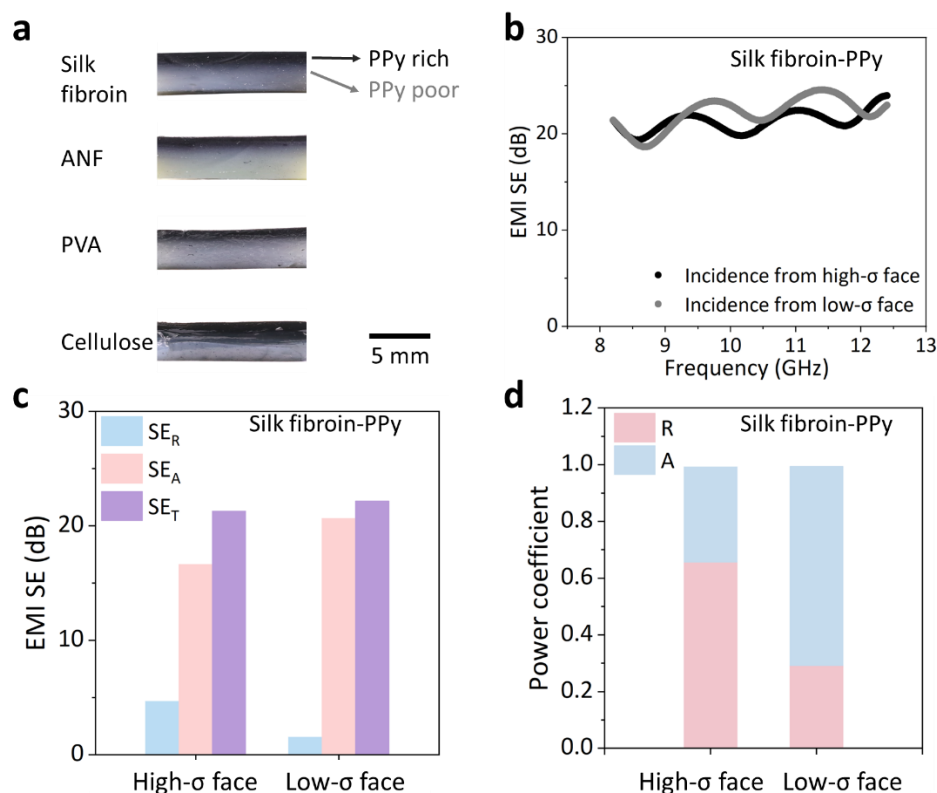


Supplementary Fig. 14 Simulated oblique-incidence EMI shielding response under TE and TM polarizations. Frequency-dependent absorption (A), reflection (R), and transmission (T) of the gradient shielding material under (a) TE and (b) TM polarizations, together with the corresponding shielding-effectiveness components, including reflection shielding effectiveness (SE_R), absorption shielding effectiveness (SE_A), and total shielding effectiveness (SE_T), under (c) TE and (d) TM polarizations. The data are shown at incident angles of 0° , 30° , and 60° over 8.2–12.4 GHz.

To evaluate the shielding robustness beyond the waveguide configuration, we performed full-wave simulations under oblique incidence at 0° , 30° , and 60° for both TE and TM polarizations. The frequency-dependent A , R , and T responses remain stable over 8.2–12.4 GHz. In all cases, T is strongly suppressed and remains negligible ($T < 0.08\%$), indicating that the incident electromagnetic waves are effectively attenuated within the gradient shielding material. Although the relative proportions of absorption and reflection vary with incident angle and polarization, absorption consistently dominates over reflection ($A > R$), confirming the absorption-dominated shielding mechanism. This trend is further supported by the corresponding shielding-effectiveness analysis, where the total shielding effectiveness (SE_T) is mainly contributed by absorption shielding effectiveness (SE_A), while reflection shielding effectiveness (SE_R) remains comparatively small. These results demonstrate that the gradient architecture maintains robust broadband EMI shielding under oblique incidence and different polarization states, highlighting its effectiveness in practical electromagnetic environments beyond the ideal waveguide configuration.



Supplementary Fig. 15 MCAs stability. a. EMI *SE* of MCAs tested in 0 month and in 4 months after formation. Thermogravimetric analysis (TGA) of the prepared samples of 94% porosity ANF-PVA aerogel (b) and the PPY integrated MCA-94 (c).

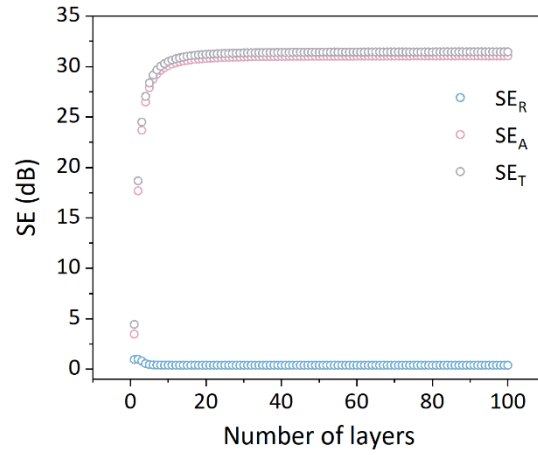


Supplementary Fig. 16 Scaffold-level compatibility of PPy gradient formation. a. In situ growth of PPy in different porous scaffolds, including silk fibroin, ANF, PVA, and bacterial-cellulose hydrogel, forming visible PPy gradients. b-d, EMI shielding properties of a representative silk fibroin–PPy gradient aerogel: (b) EMI SE across the X band; (c) SE_A, SE_R, and SE_T; and (d) power coefficients for incidence from the low-conductivity and high-conductivity faces.

To probe scaffold-level compatibility, we applied an analogous one-sided Py/FeCl₃ reaction–diffusion process to chemically distinct wettable porous scaffolds, including silk fibroin, ANF, PVA, and bacterial-cellulose hydrogel. All samples developed visible dark-to-light PPy distributions, consistent with gradient deposition. As a representative non-ANF system, the silk fibroin–PPy gradient aerogel was further evaluated for EMI shielding. The silk fibroin–PPy aerogel exhibited comparable X-band total SE_T under opposite incident directions, but showed direction-dependent shielding contributions: low- σ -face incidence resulted in lower reflection and higher absorption than high- σ -face incidence. These results support the scaffold-level compatibility of the PPy reaction–diffusion route and indicate that the asymmetric absorption-dominated shielding behavior can be retained in a non-ANF scaffold.

For the aqueous PPy system, successful gradient formation requires a wettable, structurally stable porous monolith that allows through-thickness monomer/oxidant transport and retains nascent PPy before complete precursor homogenization. The effective

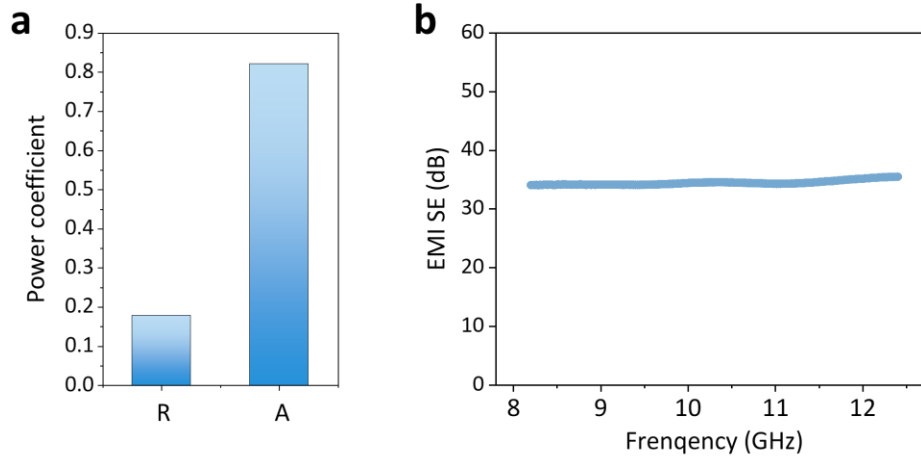
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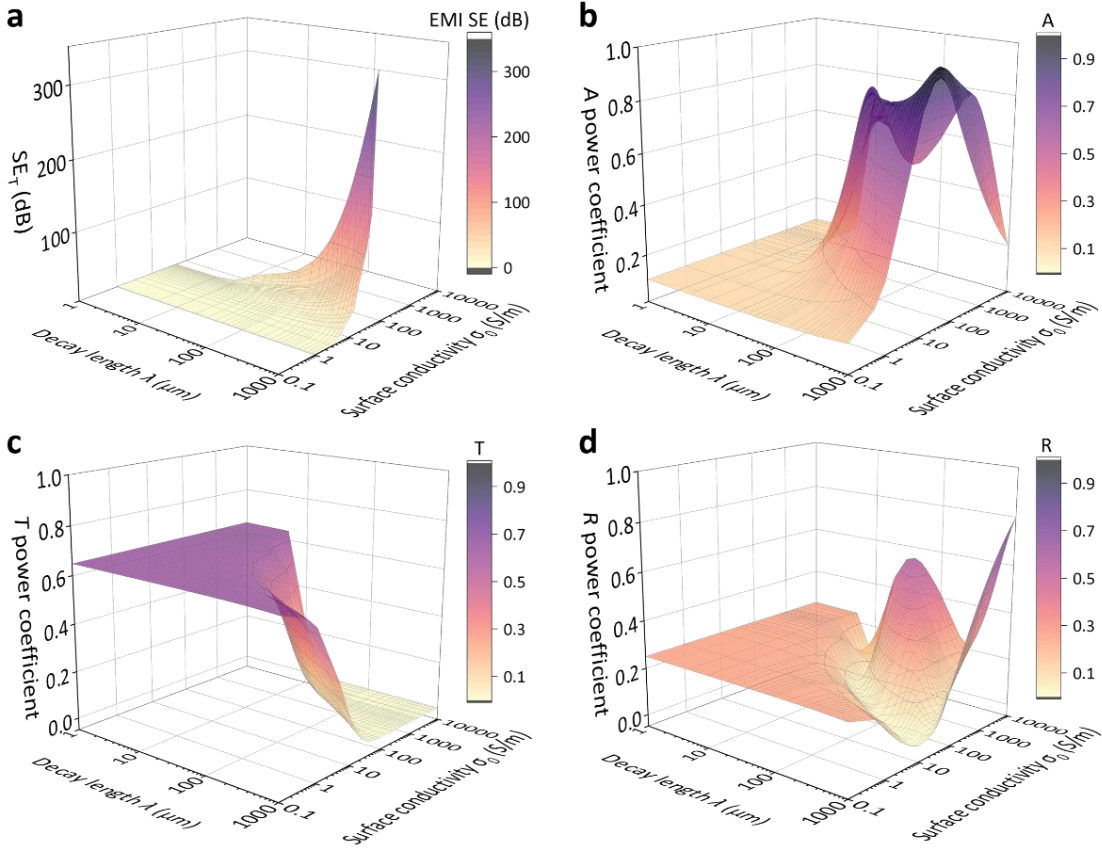
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213 **Supplementary Fig. 18** The continuous conductivity profile $\sigma=\sigma_0 \exp(-z/\lambda)$ was
 214 discretized into N uniform layers. EMI SE versus the number of gradient layers under
 215 incidence from the low- σ face, showing that increasing gradient continuity reduces
 216 reflection, increases absorption, and enhances total SE.

217



Supplementary Fig. 19 EMI shielding performance of a 3-layer stepwise conductivity-gradient control using the same ANF-PVA/PPy material system. a. Power coefficients R and A of the 3-layer stepwise conductivity-gradient control under incidence from the low- σ face (transmission is negligible). b. EMI shielding effectiveness (SE) of the same sample across the X band (8.2–12.4 GHz). The 3-layer stepwise control approximates the continuous conductivity profile with three discrete conductivity levels and exhibits effective shielding ($SE_T \sim 34$ dB) together with absorption-dominant behavior ($R \sim 0.18$, $A \sim 0.82$), while still showing higher reflection than the monolithic continuous-gradient MCA-4*.

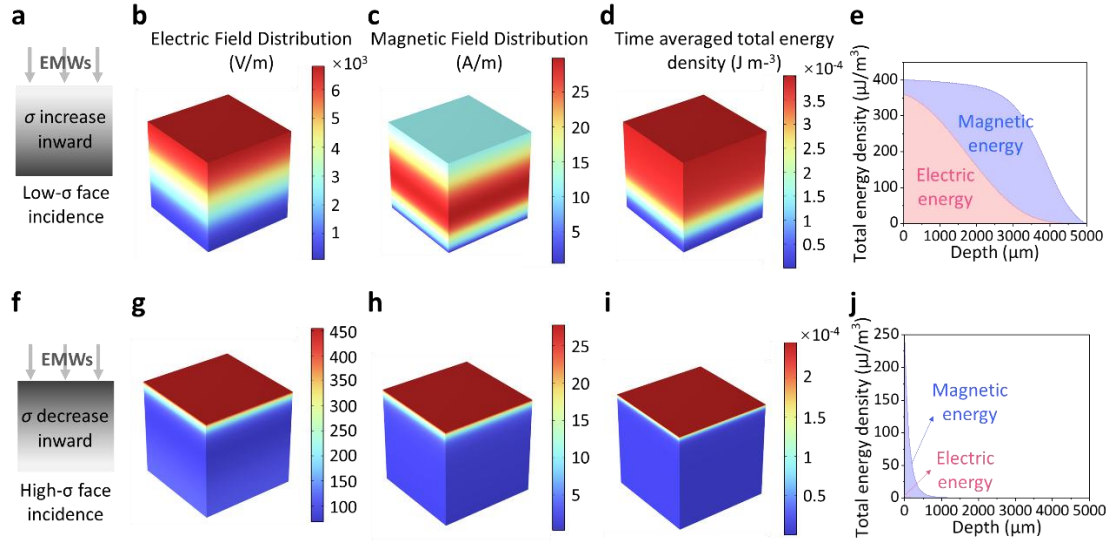


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Supplementary Fig. 20 Simulation-derived design maps for conductivity-gradient shields. 3D surface plots showing the dependence of (a) total shielding effectiveness SE_T , (b) absorption power coefficient A , (c) transmission power coefficient T , and (d) reflection power coefficient R on surface conductivity σ_0 and gradient decay length λ for an exponential conductivity profile $\sigma(z) = \sigma_0 \exp(-z/\lambda)$. Simulations were conducted at $d = 5$ mm and an incident EMW frequency of 10 GHz under low- σ -face incidence.

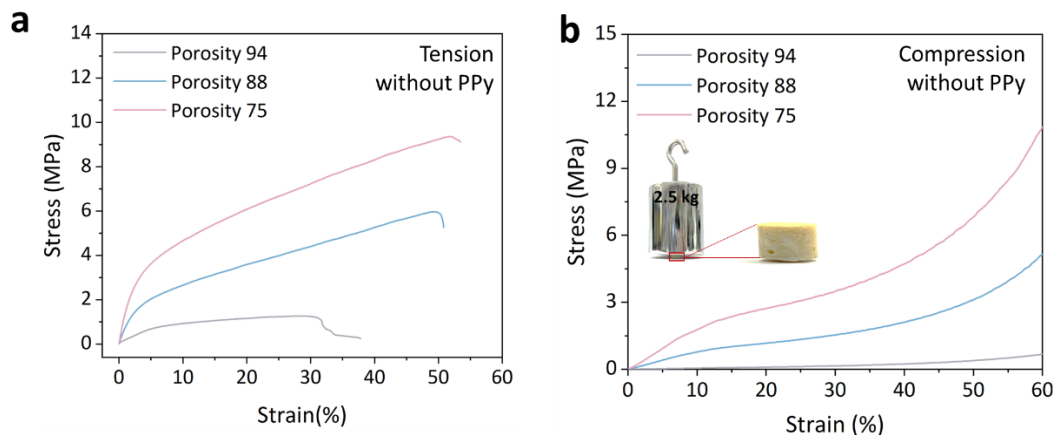
The maps reveal three design regimes. First, low σ_0 and/or overly small λ gives insufficient distributed loss, resulting in high T and inadequate SE_T . Second, excessive σ_0 or overly large λ suppresses T but increases incident-side σ , reintroducing entrance-side impedance mismatch thereby increasing R . Third, an intermediate σ_0 - λ window simultaneously suppresses T and R , producing high absorption. Because $A = 1 - R - T$, a SE_T target > 30 dB corresponds to $T \leq 10^{-3}$; therefore, within the low- T regime, the target $R \leq 0.1$ is essentially equivalent to $A \geq 0.899$, i.e., $A > 0.9$. In summary, the maps indicate that the optimal region of high SE_T (≥ 30 dB) with low- R (≤ 0.1)/high- A (> 0.9) occurs only at intermediate σ_0 and λ , where entrance-impedance matching and internal maximum dissipation are simultaneously satisfied.

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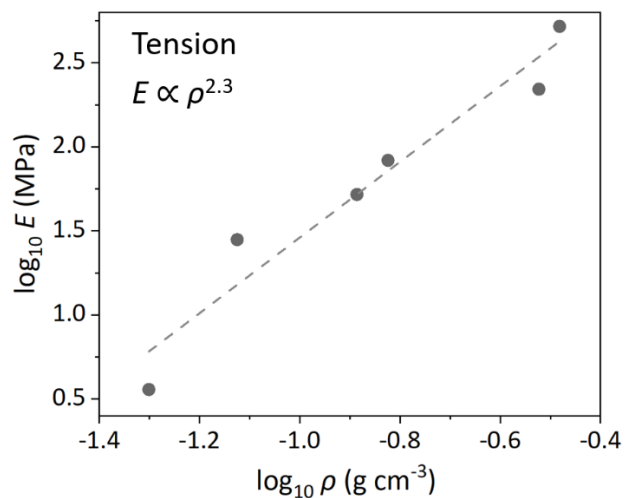
Supplementary Fig. 21 Simulated field, energy-density, and energy-partition distributions for opposite incidence directions in a representative conductivity-gradient MCA at 10 GHz. (a–e) Illustration and simulation results for low- σ face incidence. a, Schematic showing electromagnetic waves incident from the low- σ face, where conductivity increases toward the interior. b–d, Three-dimensional distributions of electric-field magnitude $|\mathbf{E}|$ (b), magnetic-field magnitude $|\mathbf{H}|$ (c), and time-averaged total electromagnetic energy density (d). e, Corresponding depth-dependent decomposition of total electromagnetic energy into electric and magnetic contributions. (f–j) Illustration and simulation results for high- σ face incidence. f, Schematic showing incidence from the high- σ surface, where conductivity decreases toward the interior. g–i, Three-dimensional distributions of $|\mathbf{E}|$ (g), $|\mathbf{H}|$ (h), and total electromagnetic energy density (i). j, Corresponding depth-dependent decomposition of total electromagnetic energy into electric and magnetic contributions.

For low- σ face incidence, $|\mathbf{E}|$ and the total electromagnetic energy density decay gradually through the aerogel, with the energy density decreasing from 4.01×10^{-4} to $1.29 \times 10^{-7} \text{ J m}^{-3}$ over 5 mm, corresponding to 34.9 dB attenuation and consistent with the experimentally measured SE_T . The attenuation is slow over the first $\sim 3000 \mu\text{m}$ and then accelerates as the wave reaches more conductive regions and accumulates dielectric and ohmic losses. By contrast, under high- σ face incidence, the field and energy density collapse near the entrance, with the energy density reaching its $1/e$ value within $\sim 150 \mu\text{m}$, consistent with strong entrance-side mismatch and near-surface field confinement. These results support a direction-dependent mechanism in which low- σ face incidence enables progressive impedance matching and absorption-dominated attenuation, whereas high- σ face incidence produces stronger front-surface reflection.



Supplementary Fig. 22 Effects of porosity on the mechanical properties of the pure ANF-PVA aerogel substrates. The tensile (a) and compression (b) responses of aerogels with varied porosities of ANF-PVA aerogel.

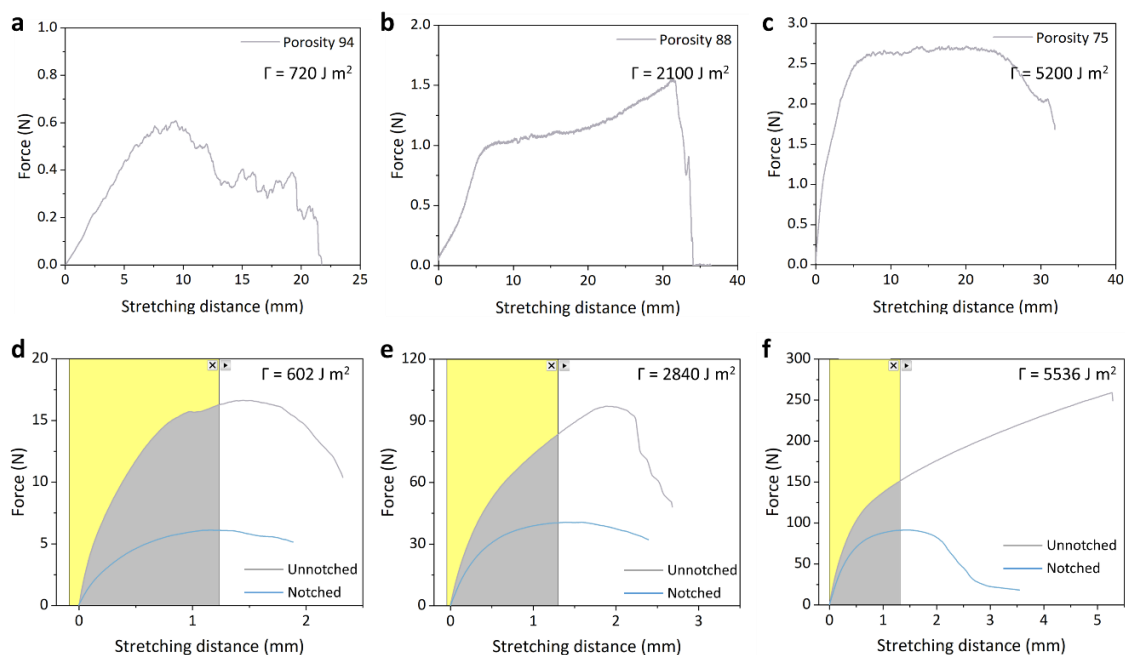
Porosity-mediated electromechanical trade-off in MCAs. In the 4 h, ~5 mm porosity-tuned MCA series, decreasing porosity increases diffusion tortuosity and restricts PPy penetration, thereby shortening the conductive-gradient transition and reducing λ . The 94% porosity sample prepared for 4 h, MCA-4/MCA-94, has σ_0 of 900 S m⁻¹ and λ of 450 μ m, giving SE_T of 49.5 dB, R of 0.16, and A of 0.84. By contrast, MCA-75 has σ_0 of 1100 S m⁻¹ and λ of 325 μ m, giving higher SE_T of 59.5 dB but larger R of 0.33 and lower A of 0.67. Thus, densification enhances transmission blocking but makes the conductivity transition steeper, weakens progressive impedance matching, and shifts the response from absorption toward reflection in the present processing window. Noted that it is not a universal consequence of densification itself, but a result of the σ_0 - λ pair moving away from the GSM-defined high-absorption/low-reflection window. Mechanically, however, the denser nanofibrillar network improves load bearing. The Young's modulus and tensile strength increase from 26 MPa and 1.27 MPa for MCA-94 to 205 MPa and 12.12 MPa for MCA-75. Therefore, MCA-4* represents the broadband low-reflection/high-absorption optimum, whereas MCA-75 prioritizes mechanical robustness and shielding effectiveness.



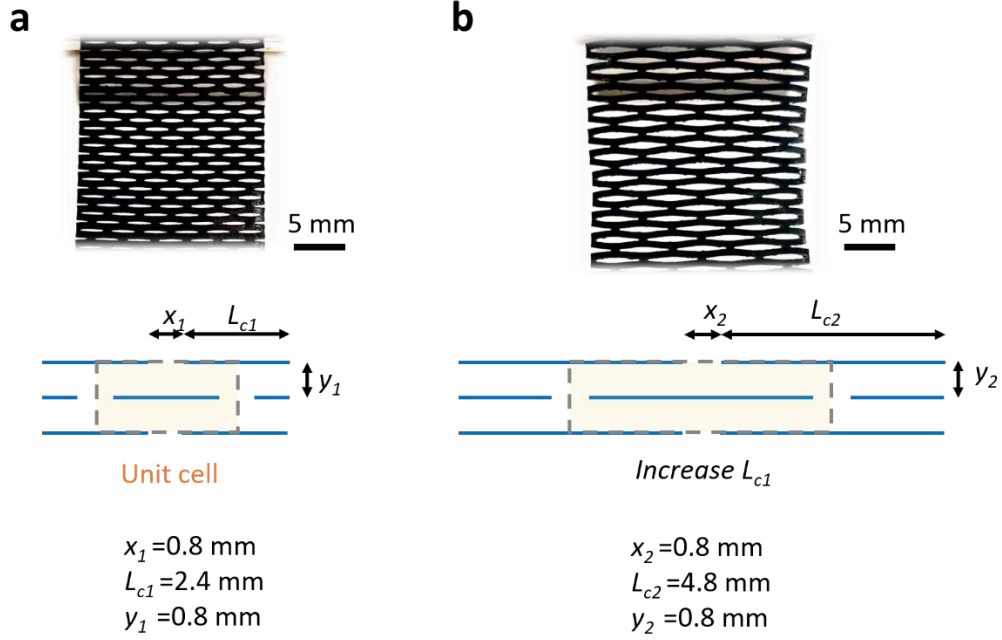
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293 **Supplementary Fig. 23** Tensile modulus of MCAs plotted against their densities, derived
 294 from experimental data (dots) and theoretical fitting (dashed lines). This function exhibits
 295 the typical power-law dependence observed in aerogel materials. This trend aligns with the
 296 behavior observed under tensile loading.

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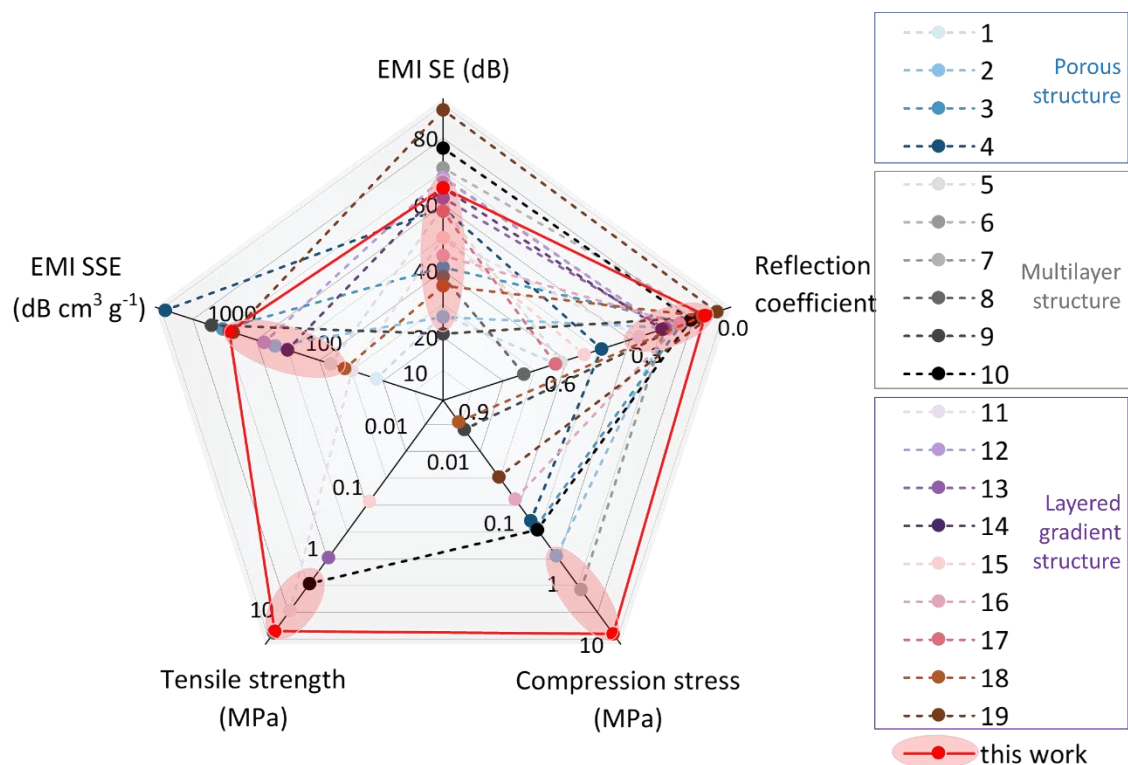
Supplementary Fig. 24 Fracture energies of MCAs characterized by tearing tests (a-c) and pure shear tests (d-f). Force-displacement curves of tearing test and the calculated tearing energies for MCA-94 (a), MCA-88 (b), and MCA-75 (c). Force-displacement curves of pure shear test for unnotched and notched samples, and the calculated fracture energies for MCA-94 (d), MCA-88 (e), and MCA-75 (f).



304

305 **Supplementary Fig. 25** MCA Kirigami with periodic laser-cut slits. a. Digital images of
 306 the kirigami pattern (upper panel) and cut-pattern unit cell (lower panel) marked in the
 307 yellow box. Cuts are indicated by the blue horizontal lines. b. Digital images (upper panel)
 308 and corresponding cut-pattern unit-cell schematic (lower panel) of the kirigami specimen
 309 with increased L_c . L_c indicates the length of the cut, x indicates the spacing in the transverse
 310 direction, and y indicates the spacing in the axial direction. Scale bars, 5 mm.

311 Periodic straight-line patterns created by laser cutting were designed with cut length L_c
 312 below one-quarter of the free-space wavelength at the upper band edge ($f = 12.4$ GHz,
 313 $\lambda_0/4 = 6.04$ mm) of the X-band measurement. This conservative rule keeps the slits well
 314 below the first slot/aperture resonance scale ($\sim \lambda_0/2$) and thereby limits aperture-induced
 315 scattering/transmission while preserving shielding performance.



Supplementary Fig. 26 Radar plot of MCAs compared with other porous EMI shielding materials with low reflectivity. The performance of the recently reported absorption-dominated EMI SE materials is summarized in Table 2.

Supplementary Table 1 Design table of conductivity gradient parameters and EMI shielding performance of key MCA specimens.

Specimen	Porosity (%)	Thickness (mm)	σ_0 (S/m)	λ (μm)	R^2	X band			Ka band		
						SE_T (dB)	R	A	SE_T (dB)	R	A
MCA-1	94	5	900	225	0.84	42.2	0.24	0.76	—	—	—
MCA-2	94	5	900	340	0.78	46.7	0.19	0.82	—	—	—
MCA-4	94	5	900	450	0.98	49.5	0.16	0.84	—	—	—
MCA-6	94	5	900	485	0.91	52.5	0.22	0.78	—	—	—
MCA-88	88	5	950	380	0.92	54.0	0.28	0.72	—	—	—
MCA-75	75	5	1100	325	0.99	59.5	0.33	0.67	—	—	—
MCA-4*	94	5	350	450	0.94	32.3	0.09	0.91	41.9	0.08	0.92

Note: Values are band-averaged under low- σ -face incidence. X-band: 8.2–12.4 GHz; Ka-band: 26.5–40.0 GHz. R^2 denotes the coefficient of determination for the exponential-profile fit. MCA-4/MCA-94 denotes the 94% porosity sample prepared with a 4 h diffusion/reaction time. MCA-4* denotes the intentionally reduced-surface-conductivity sample derived from MCA-4. “—”, not determined; Ka-band data were collected only for the optimized MCA-4* sample used for broadband validation; they were not measured for the X-band screening specimens.

331 **Supplementary Table 2** Mechanical performance of MCAs.

Sample	Young's modulus (MPa)	Tensile stress (MPa)	Elongation at fracture (%)	Toughness (J m^{-3})	Fracture energy (J m^{-2})	Compression stress @60% (MPa)
MCA-94	26	1.27	34.9	3.16×10^5	602	0.69
MCA-88	80	4.77	40.79	1.48×10^6	2840	5.51
MCA-75	205	12.12	50.55	4.29×10^6	5536	13.26

332

333 **Supplementary Table 3** Comprehensive performance of recent absorption-dominated
334 EMI shielding materials.

N o.	SE (dB)	R coef ficient	A coeff - icient	Band (GHz)	Band width (A> 0.9)	Compres s-ion stress (MPa) at specific strain	Tensi le streng th (MPa)	SSE (dB cm ³ g ⁻¹)	SSE/t (dB cm ² g ⁻¹)	SE/t (dB cm ⁻¹)	Thic- kness (mm)	Den sity (g/c m ³)	Stru- cture	Re - fer ence
1	26.8	0.61	0.39	8.2-12.4	0	—	—	26.8	127.62	134	2	1	porous	4
2	26.1	0.224	0.78	8.2-12.4	0	0.70 (70%)	—	342.7	1373-1518	104.4	2.5	0.076	porous	5
3	41.1	0.17	0.83	8.2-12.4	0	0.361 (80%)	—	1280	6400	205.5	2	0.032	porous	6
4	59	0.45	0.55	8.2-12.4	0	0.00187 (70%)	—	5420.5	10841	118	5	0.01	porous	7
5	21	0.11	0.89	18-26.5	0	0.006 (40%)	—	1690	3370	42	5	0.014	porous	8
6	59.4	0.59	0.41	8.2-12.4	0	—	—	84.5	422.5	297	2	0.723	multilayer	9
7	71	0.1	0.9	8.2-12.4	2.5	—	—	—	—	273.1	~2.6	—	multilayer	10
8	68	0.23	0.77	8.2-12.4	0	2.5 (60%)	—	—	—	123.6	5.5	—	multilayer	11
9	38.3	0.72	0.28	8.2-12.4	0	—	12.8	—	—	212.8	1.8	—	multilayer	12
10	77	0.14	0.86	8.2-18	0	0.264 (50%)	2	—	—	385	2	—	multilayer	13
11	50	0.32	0.68	8.2-12.4	0	—	5.5	49.6	12390	12500	0.04	1	gradient	14
12	68.2	0.23	0.77	8.2-12.4	0	—	—	448.7	897.4	136.4	5	0.152	gradient	15
13	62	0.23	0.77	8.2-12.4	0	—	0.75	—	—	310	2	—	gradient	16
14	65	0.24	0.76	8.2-12.4	0	—	—	250	500	130	5	0.26	gradient	17
15	50.0	0.51	0.49	8.2-12.4	0	—	0.07	—	—	2777.8	0.18	—	gradient	18
16	44.6	0.18	0.82	8.2-12.4	0	0.0833 (80%)	—	—	—	148.7	3	—	gradient	19
17	58	0.61	0.39	8.2-12.4	0	—	—	—	—	1705.9	0.34	—	gradient	20
18	35.5	0.085	0.915	8.2-12.4	0	4.5 (70%)	—	86.7	144.5	59.2	6	0.409	gradient	21

19	88.5	0.05	0.95	8.2-12.4	3.5	0.036 (50%)	—	983.3	2892.1	260.3	3.4	0.09	gradient	22
20	20-65	0.09-0.4	0.9	8.2-40	29.8	0.7-13.3 (60%)	1.3-12.1	67-1083	134-2166	60-600	~5	0.06-0.3	Continuous gradient	This work

335

336 Note: Bandwidth is defined as the continuous frequency window over which $A > 0.9$, unless
337 otherwise specified in the original reference. “—” denotes not determined.

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