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Supporting Information

Microstructure controllable polyimide/MXene composite aerogels for high-temperature thermal insulation and microwave absorption

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Fig S1. Photos of the setup of the ultrasonic-assisted liquid nitrogen freezing process.

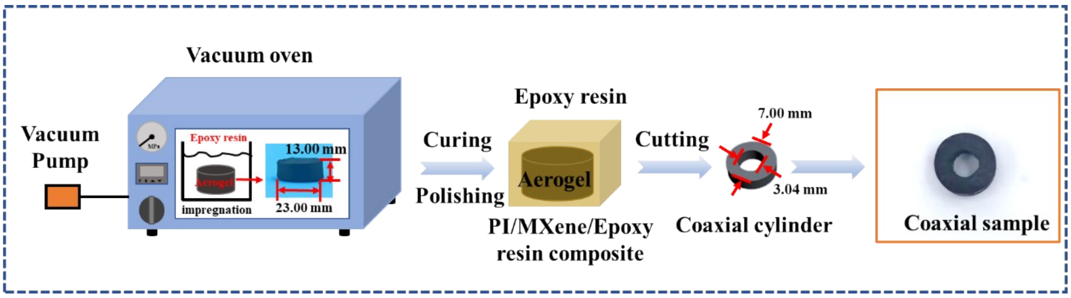

Fig. S2. Schematic illustration of the fabrication of a coaxial cylinder for microwave absorption measurement.

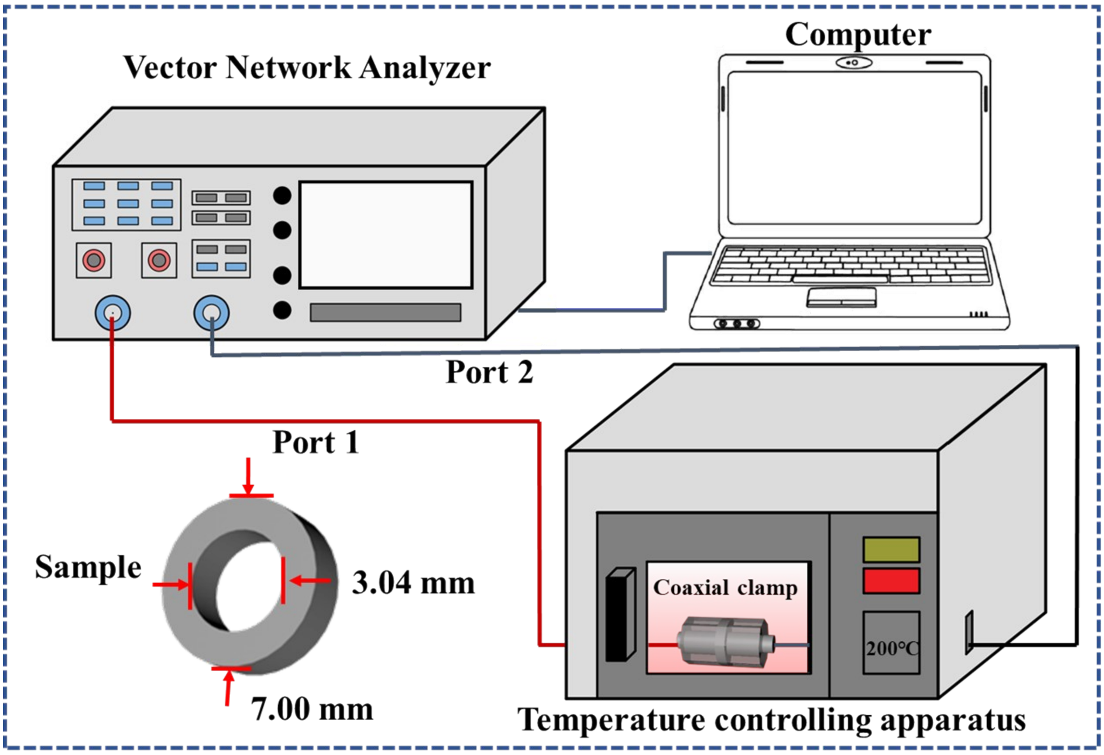

Fig S3. Schematic illustration of the high-temperature coaxial line setup.

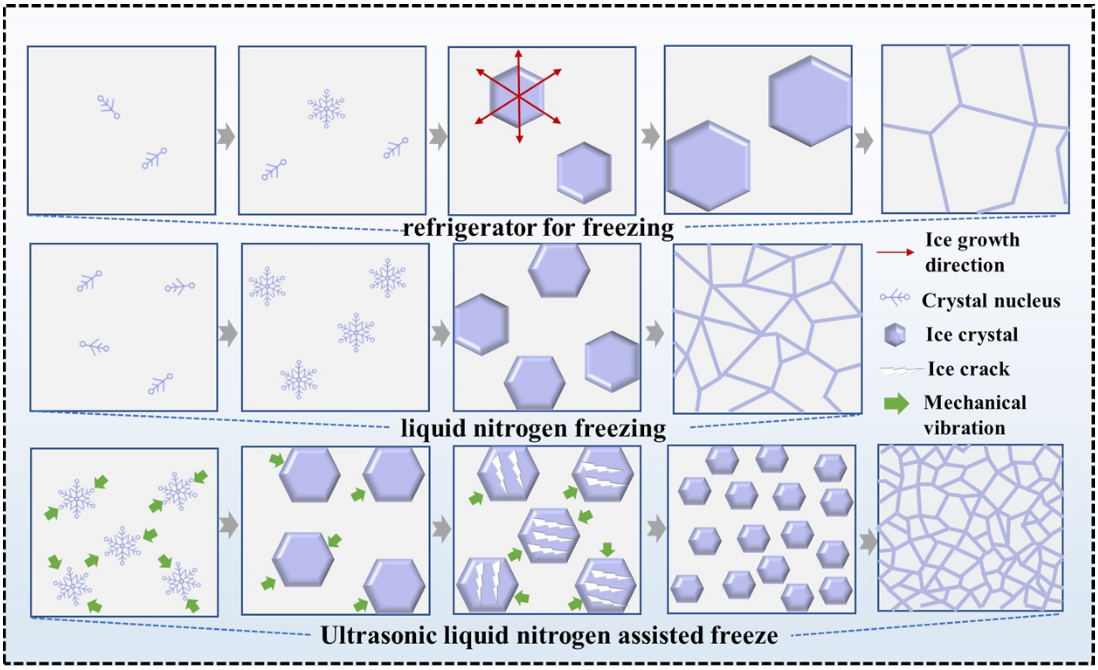

Fig S4. Mechanism diagrams of different freezing processes.

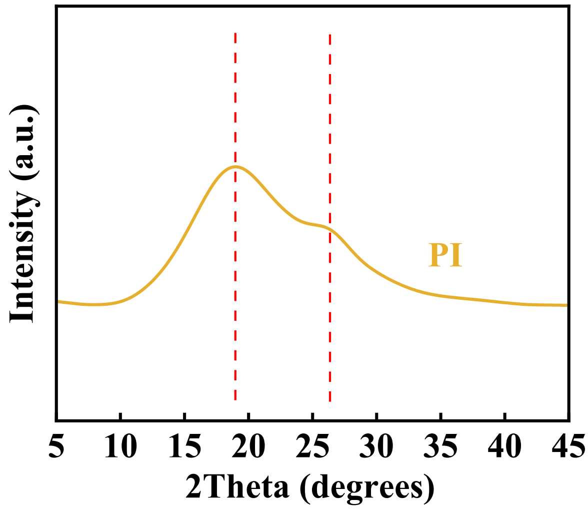

Fig. S5. XRD pattern of neat PI.

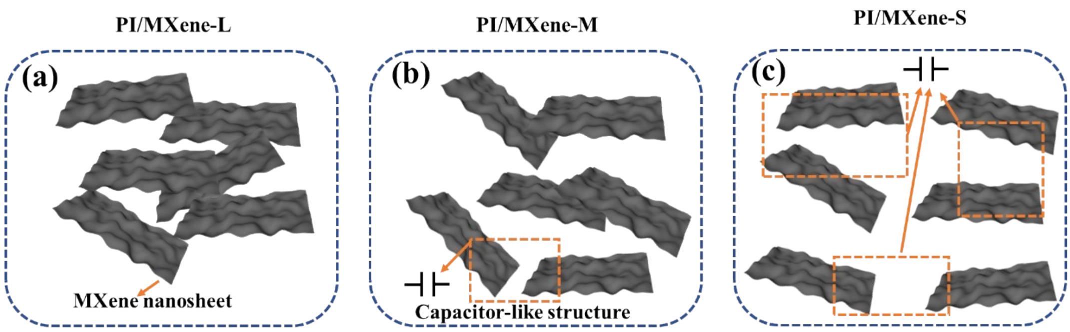

Fig. S6. Formation of capacitor-like structures of (a) PI/MXene-L, (b) PI/MXene-M and (c) PI/MXene-S.

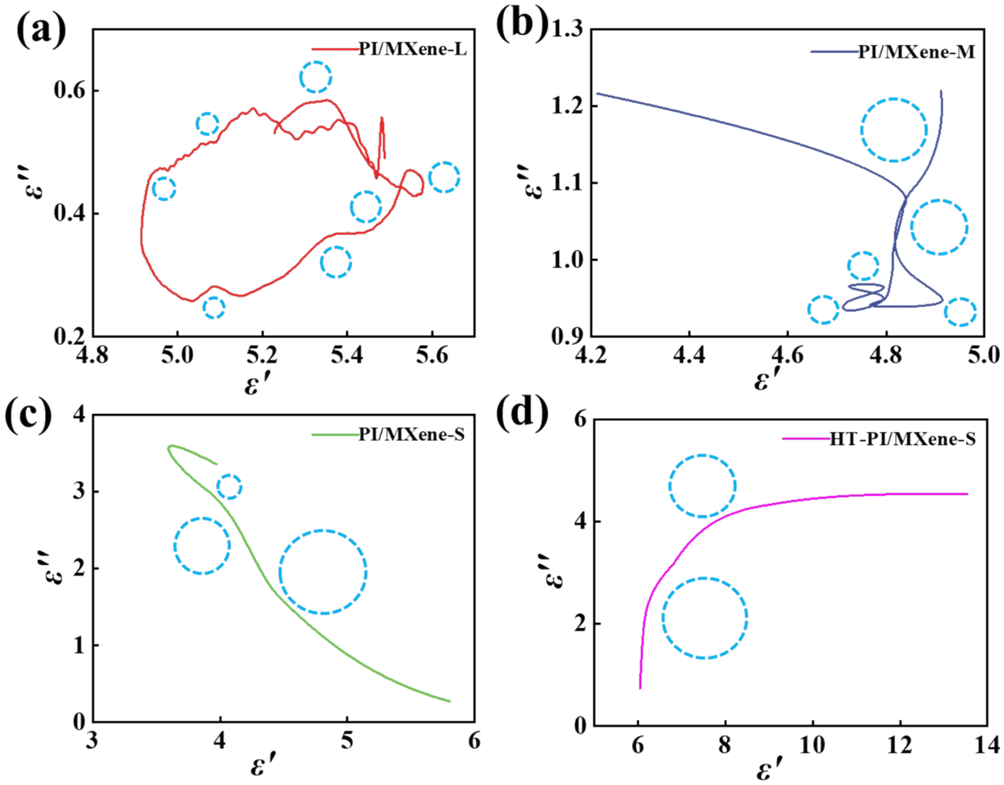

Fig. S7. Cole-Cole curves of (a) PI/MXene-L, (b) PI/MXene-M, (c) PI/MXene-S and (d) HT-PI/MXene-S.

Table S1. The values of $\lambda_0(T)$, $\lambda_{air}(T)$, and $\Lambda_g(T)$

Temperature	$\lambda_0(T)$ **(W/(m·K))**	$\lambda_{air}(T)$ **(W/(m·K))**	$\Lambda_{air}(T)$ **(nm)**
25 °C	0.7	0.026	70
200 °C	0.9	0.038	121.3

Table S2. Apparent densities, shrinkage and porosities of composite aerogels

	PI/MXene-L	**PI/MXene-M**	**PI/MXene-S**
Density (ρ)	0.083 g/cm^3	0.087 g/cm^3	0.092 g/cm^3
Shrinkage	30.00%	33.36%	36.92%
Porosity (Φ)	95.59%	95.37%	95.12%

Table S3. The values of λ_g, λ_s, and λ_r under ambient temperature

Sample	λ_g **(25 °C) (mW/(m·K))**	λ_s **(25 °C) (mW/(m·K))**	λ_r **(25 °C) (mW/(m·K))**
PI/MXene-L	24.8	14.7	8.3
PI/MXene-M	24.7	9.7	16.2
PI/MXene-S	24.5	3.4	23.7

Table S4. The values of λ_g, λ_s, and λ_r at 200 °C

Sample	λ_g **(200 °C) (mW/(m·K))**	λ_s **(200 °C) (mW/(m·K))**	λ_r **(200 °C) (mW/(m·K))**
PI/MXene-L	36.5	19.1	8.2
PI/MXene-M	36.4	12.5	16.4
PI/MXene-S	35.7	4.4	25.7

Table S5. Comparison of the microwave absorption performance of different samples at room temperature.

Samples	**Thickness (mm)**	***EAB* (GHz)**	**RL_{min} (dB)**	**Ref**
rGO/MXene/FeS foam	4.1	11.2	-50.7	[43]
Porous carbon foam	1.5	6.2	-46.9	[44]
GN/Fe_2O_3 foam	4.8	6.2	-60.1	[45]
N-CQDs/rGO aerogel	4.1	7.4	-69.4	[46]
Carbon foam/CNT@Si_3N_4 hybrid	3.2	8.6	-56.3	[47]
Carbon fiber/PMI foam	1.0	7.5	-32.0	[48]
C-wood/CI foam	1.5	5.2	-15.9	[49]
PI/ANF/MXene aerogel	2.6	2.8	-28.0	[23]
PI/MXene-S aerogel	4.0	4.1	-30.3	This work

(rGO: reduced graphene oxide; CF: Carbon foam; GN: graphene; N-CQDs: nitrogen doped carbon quantum dots; CNT: carbon nanotube; PMI: polymethacrylimide; C-wood: carbonized wood; CI: carbonyl iron; ANF: aramid nanofiber)

Appendix:

According to Debye theory, the dielectric loss can be divided into polarization loss (ε_p'') and conductive loss (ε_c''), which can be expressed by the following equations:

$$\varepsilon_p'' = (\varepsilon_s - \varepsilon_\infty)\frac{\omega\tau_{(T)}}{1 + \omega^2{\tau_{(T)}}^2} \qquad (1)$$

$$\varepsilon_c'' = \frac{\sigma_{(T)}}{\varepsilon_0\omega} \qquad (2)$$

where ε_s is the static dielectric constant, ε_∞ is the optical dielectric constant, $\omega = 2\pi f$ is the angular frequency, ε_0 is the vacuum permittivity, $\tau_{(T)}$ is the temperature-dependent relaxation time and $\sigma_{(T)}$ represents the temperature-dependent conductivity.

The model function in this work is $\varepsilon = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + i\omega\tau} - \frac{i\sigma}{\omega\varepsilon_0}$. The nonlinear squares fitting method was used to fit parameters τ, σ, ε_s, and ε_∞, which are signed as a group γ. The sum of squares is $S = \sum_{i=1}^{m} r_i^2$, where r_i is $\varepsilon_{fit} - \varepsilon$. The goal is to minimize S by adjusting γ. The code, which was run by using MATLAB, is shown below.

```
clc;clear;
format long

% Set initial parameter values and read in data
p0 = [50,1,1,1e-12];
data = readmatrix('data.xlsx');
fcost = 0;          % cost of fitting
fsig = 0;           % fitting conductivity
ft = 0;             % fitting relaxation time
fplsq = [];          % fitting results of data
fs = 0;              % static dielectric constant
fu = 0;              % optical dielectric constant
fconduct_loss = 0;    % fitting conducting loss
frelaxation_loss = 0;  % fitting polarization loss

% Read data
start_index = 1;
end_index = size(data);
```

```
xdata = data(start_index:end_index,1) * 1e9;
ydata_1 = data(start_index:end_index,2);  % real permitivity
ydata_2 = data(start_index:end_index,3);  % imaginary permitivity
ydata = complex(ydata_1, -ydata_2);

% Perform the fitting
options = optimoptions('lsqnonlin', 'MaxFunctionEvaluations', 100000000);
plsq = lsqnonlin(@(p) residuals(p,ydata,xdata), p0, [0,0,0,0], [200,200,200,1e-5], options);
fplsq = [fplsq; plsq];

% Compute various fitting statistics
conduct_loss = mean(condut_loss_result(xdata, plsq));
fconduct_loss = fconduct_loss + conduct_loss;
relaxation_loss = mean(ydata_2) - conduct_loss;
frelaxation_loss = frelaxation_loss + relaxation_loss;

fs = fs + plsq(1);
fu = fu + plsq(2);
fsig = fsig + plsq(3);
ft = ft + plsq(4);

% Define the realimag function
function result = realimag(array)
result = zeros(length(array), 2);
for i = 1:length(array)
    result(i,:) = [real(array(i)),-imag(array(i))];
end
end

% Define the condut_loss_result function
function result = condut_loss_result(x,p)
s = p(1);  % static dielectric constant
u = p(2);  % optical dielectric constant
sigma = p(3);  % conductivity
t = p(4);  % relaxation time
o = 8.854187817e-12;
result = sigma./(x.*o);
end

% Define the residuals function
function result = residuals(p,y,x)
s = p(1);  % static dielectric constant
```

```
u = p(2);  % optical dielectric constant
sigma = p(3);  % conductivity
t = p(4);  % relaxation time
d = 1i;
o = 8.854187817e-12;
result = (realimag(y) - realimag(u + (s-u)./(1+d*(x.*t)) -
    d.*(sigma./(x*o))));
result = result(:);
end
```

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