Supporting Information

Laminated Structural Engineering Strategy toward Carbon Nanotube-Based Aerogel Films

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1. Electromagnetic Interference (EMI) Shielding Measurements

Electromagnetic interference shielding measurements of CNT-based aerogel films were carried out in a WR-90 rectangular waveguide using a 2-port network analyzer (N5227A) in the X-band frequency range (8.2-12.4 GHz). The samples were cut into a rectangular shape in a dimension of 24 × 12 mm\textsuperscript{2}, which is slightly larger than the sample holder dimension (22.84 × 10.14 mm\textsuperscript{2}). Electromagnetic interference shielding efficiency can be calculated according to the following formula:\textsuperscript{1}

\begin{align*}
R &= |s_{11}|^2 = |s_{22}|^2 \quad (1) \\
T &= |s_{12}|^2 = |s_{21}|^2 \quad (2) \\
SE_R &= 10\log\left(\frac{1}{1-R}\right) = 10\log\left(\frac{1}{1-|s_{11}|^2}\right) \quad (3)
\end{align*}
\[
SE_A = 10 \log \left( \frac{1-R}{T} \right) = 10 \log \left( \frac{1-|s_{11}|^2}{|s_{21}|^2} \right)
\]

(4)

\[
SE_T = SE_R + SE_A + SE_{MR}
\]

(5)

where \( R, A, T \) are the reflection coefficient, absorption coefficient, and transmission coefficient, respectively; \( S_{ij} \) represents the s-parameter obtained by the network analyzer; \( SE_R, SE_A, SE_T, \) and \( SE_{MR} \) are the reflection effectiveness, absorption effectiveness, total shielding effectiveness, and multiple internal reflection effectiveness, respectively.

Specific shielding effectiveness (SSE) is derived to compare the effectiveness of shielding materials by taking into account the density, which can be obtained by dividing the EMI SE by the density of materials as follows:

\[
SSE = \frac{\text{EMI SE}}{\text{density}} = \text{dB cm}^3 \text{ g}^{-1}.
\]

(6)

However, SSE has a basic limitation, that is, it does not account for the thickness information. So, the absolute effectiveness (SSE/\(t\)) was obtained, which is take into account the thickness:

\[
SSE_t = \frac{\text{SSE}}{t} = \text{dB cm}^2 \text{ g}^{-1}.
\]

(7)

2. Orientation measurements for aerogel film

Herman's orientation factor (\( f \)) was calculated to describe the degree of orientation of the nanofibers as well as CNTs relative to the film plane using Equation (3):

\[
f = \frac{3(\cos^2 \theta) - 1}{2}
\]

(8)

where the mean-square cosine is calculated from the scattered intensity \( I(\theta) \) by integrating over the azimuthal angle \( \theta \) according to Equation (4):

\[
\langle \cos^2 \theta \rangle = \frac{\int_0^\pi I(\theta) \sin \theta \cos^2 \theta d\theta}{\int_0^\pi I(\theta) \sin \theta d\theta}
\]

(9)

where \( \theta \) is the angle between the film plane and the nanofibers as well as CNTs. \( f = 1 \) when all nanofibers and CNTs are parallel to the film plane and \( f = 0 \) for random orientation of
nanofibers and CNTs.

3. Input power density for electrothermal conversion

The voltage was input by Precision Measurement DC Supply and the corresponding current could be obtained simultaneously. The CNT-based aerogel film was cut into a certain size (1×2 cm²). Assuming that electrical energy is completely converted into heat, the input power density ($P_A$) can be calculated by Joule’s law with the following formula:

$$P_A = \frac{UI}{A}$$  \hspace{1cm} (10)

Where $U$ is the input voltage, $I$ is the current through the aerogel film and $A$ is the area of the aerogel film.

![Figure S1. Typical WAXS patterns (a) and its relative radial profiles (b) of D-ANF/CNT film.](image)

Figure S2. SEM images of ANF/CNT aerogel films with different compressive treatments and their corresponding fast Fourier transform (FFT) frequency domain images: 0 MPa (a and d), 10 MPa (b and e), and 20 MPa (c and f).

The fast Fourier transform (FFT) method was used to analyze the structural evolution of ANF/CNT aerogel films with different compressive strain treatments. Orientation Factor was calculated from the standard formulation as defined by $F = \langle \cos^2 \phi \rangle = \frac{\int_0^\pi I(\phi) \cos^2 \phi d\phi}{\int_0^\pi I(\phi) d\phi}$, where $I$ is the intensity profile as a function of the angle between the structural unit vector and the reference direction. In this case, the intensity profile was derived from the FFT of side-view SEM images. Without compression, the nanofibers are randomly oriented and there is a circular spot on the FFT frequency domain image (Figure S2d). After compressive treatment under 10 MPa and 20 MPa, the randomly aligned nanofibers deformed into the orientated structure (Figure S2b and S2c). From the FFT frequency domain images, the plot also has a prominent orientation (Figure S2e and S2f).
Figure S3. Tensile stress-strain curves of ANF/CNT aerogel film, D-ANF/CNT film and CANF/CNT aerogel films at different temperatures.

Figure S4. The surface morphology of ANF/CNT (a) and CANF/CNT with different carbonization temperatures: 550 °C (b), 650 °C (c), 750 °C (d), 850 °C (e), and 950 °C (f), (scale bar: 1 μm).

Without carbonization, the ANF/CNT aerogel film exhibit a smooth surface structure with
some small pores (Figure S4a). Following the carbonized temperature increase, the pores on the CANF/CNT films got bigger and bigger, due to the pyrolysis of ANF and the generated gases. For CANF/CNT-550 (Figure S4b) and CANF/CNT-650 (Figure S4c), a continuous smooth surface can be observed. When the carbonization temperature increases to 750 °C, a more porous structure can be observed on the surface (Figure S4d). For CANF/CNT-850 (Figure S4e) and CANF/CNT-950 (Figure S4f), the smooth surface structures were destroyed and obvious nanofibers network structure can be observed.

**Figure S5.** The SEM images of CANF/CNT-550 (a), CANF/CNT-650 (b), CANF/CNT-750 (c), CANF/CNT-850 (d), and CANF/CNT-950 (e), and their Orientation factors (f, insets: 2D FFT of the SEM images).
**Figure S6.** The SEM images of pure ANF aerogel film before (a) and after carbonization at 950 °C (b).

In order to investigate the morphology of ANF after carbonization, we prepared a pure ANF aerogel film without CNTs and the ANF aerogel film was carbonized under 950 °C by the protection of Ar. From the SEM image, the pure ANF aerogel film shows a three-dimensional network structure of nanofibers (Figure S6a), and the nanofiber morphology of ANF was maintained after carbonization treatment (Figure S6b).
Figure S7. The FTIR spectrum of decomposed gases at 638 °C.

Figure S8. The elemental content of C, N, and O for ANF/CNT, CANF/CNT-550, CANF/CNT-650, CANF/CNT-750, CANF/CNT-850, and CANF/CNT-950, respectively.
Figure S9. The FTIR spectrum of CH₄ and CO₂ is generated during the pyrolysis of ANF/CNT.

Figure S10. The Raman spectrum of CANF/CNT with different carbonization temperatures.
Figure S11. The XRD spectra of CANF/CNT with different carbonization temperatures.

Figure S12. The specific surface area of ANF/CNT, D-ANF/CNT, and CANF/CNT with different carbonization temperatures.
S13. The pore size distribution curves (a), micropore distribution (b), and mesoporous (c) of CNT-based aerogel films carbonized at different temperatures.

Figure S14. The contact angle of CANF/CNT aerogel films with different carbonization temperatures.
Figure S15. The comparison of experimentally and theoretically results for CANF/CNT-750.

Figure S16. the cross-section SEM image of CANF/CNT-d.
**Figure S17.** The density (a) and electric conductivity (b) of the carbon film without aerogel progress.

**Figure S18.** The EMI SE of the CNT-based film without aerogel progress before (RT) and after carbonization progress (750°C).
Figure S19. The EMI $S_E$ (a) and electrical conductivity (b) of CANF/CNT aerogel films with different ANF:CNT mass ratios in the processing recipe.

Figure S20. The I-V curves for CANF/CNT-750 aerogel film.
Figure S21. The IR camera images of CANF/CNT-750 aerogel film under different input voltages and its photo.

Table S1. EMI SE of various aerogel shielding materials.

| Sample                        | Density (g·cm⁻³) | Thickness (mm) | Electrical conductivity (S·m⁻¹) | EMI SE (dB) | SSE/t (dB·cm²·g⁻¹) | Ref. |
|-------------------------------|------------------|----------------|-------------------------------|-------------|-------------------|------|
| Graphene/polyimide            | 0.076            | 2.5            | 2.58                          | 28.8        | 1515.8            | 4    |
| Graphene/carbon               | 0.002            | 2              | 1000                          | 21.3        | 53250.0           | 5    |
| Graphene/texture              | 0.07             | 2              | 8000                          | 27          | 1928.6            | 6    |
| Graphene/cellulose            | 0.00283          | 5              | 2.57                          | 47.8        | 33780.9           | 7    |
| Graphene                      | 0.41             | 0.12           | 8000                          | 105         | 21341.5           | 8    |
| Graphene                      | 0.006            | 5              | 78.3                          | 40.5        | 13500.0           | 9    |
| Cu Nanowire@Graphene          | 0.0165           | 9.46           | 2083                          | 52.5        | 3363.4            | 10   |
| Cellulose nanofibril/RGO      | 0.0058           | 3              | 64.49                         | 33          | 18965.5           | 11   |
| Graphene                      | 0.122            | 1.547          |                               | 53          | 2808.2            | 12   |
| Graphene/silver nanowires     | 0.019            | 5              |                               | 45.2        | 4757.9            | 13   |
| Carbon                        | 0.079            | 2              | 44.3                          | 93.1        | 5892.4            | 14   |
| CNF@Co/C                      | 0.023            | 4              | 0.47                          | 56.07       | 6094.6            | 15   |
| Carbon                        | 0.0544           | 0.162          | 1029.5                        | 41.4        | 46977.1           | 16   |
| Carbon                        | 0.00125          | 4              |                               | 36.75       | 73500.0           | 17   |
| Carbon                        | 0.057            | 4              | 1.35                          | 40          | 1754.4            | 18   |
| MXene@wood                    | 0.108            | 10             | 37.04                         | 71.8        | 664.8             | 19   |
| PVA/MXene                     | 0.034            | 3.49           | 314                           | 53.3        | 4491.8            | 20   |
| MXene/CNT                     | 0.0091           | 2              | 447.2                         | 51          | 28022.0           | 21   |
|                | Cellulose nanofibril/MXene | MXene/rGO | MXene/CNT | Nanocellulose-MXene | MXene/Carbon | MXene | MXene/PEDOT:PSS | CNT Nanocellulose/CNT | Aramid Nanofiber/CNT | MWCNT/Cellulose | Others Nanocellulose/Silver Nanowire | CNT CANF/CNT-550 | CANF/CNT-650 | CANF/CNT-750 | CANF/CNT-850 | CANF/CNT-950 |
|----------------|---------------------------|-----------|-----------|---------------------|--------------|-------|----------------|----------------------|-----------------------|-----------------|--------------------------------------|----------------|-------------|-------------|-------------|-------------|
|                | 0.0073                    | 0.032     | 0.042     | 0.0015              | 0.2483       | 0.0055 | 0.012          | 0.072                | 0.043                  | 0.095          | 0.02                   | 0.017           | 0.15         | 0.12         | 0.087        | 0.058        |
|                | 2                         | 2         | 3         | 1                   | 3.23         | 1     | 4.5            | 2                    | 0.568                  | 2.5             | 6                      | 0.024           | 0.024        | 0.024        | 0.024        | 0.024        |
|                | 226.4                     | 1085      | 943       | 27.6                | 28.34        | 48.5  | 3.097          | 26.2                 | 230                    | 1.8             | 416                    | 1408.5          | 3449.4       | 8540.5       | 6561.6       | 2490.9       |
|                | 22.8                      | 56.4      | 103.9     | 28.41               | 70           | 59    | 59             | 39.8                 | 54.4                   | 20.8            | 30.3                    | 25.97           | 31.33        | 35.95        | 34.72        | 27.87        |
|                | 15620.0                   | 8812.5    | 8246.0    | 189400.0            | 872.8        | 88181.8| 10841.0        | 2763.9               | 22273.2                | 875.8           | 28323.3                  | 63652.0         | 87027.8     | 124826.4     | 166283.5     | 200215.5     |
|                |                           |           |           |                     |              |       |                |                      |                        |                 |                         | This work       |             |             |             |             |

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