Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting information

Lightweight flexible carbon nanotube/polyaniline films with outstanding EMI shielding property

Hui Li, Xuehong Lu, Du Yuan, Jiaotong Sun, Fuat Erden, Fuke Wang, Chaobin He*

*Correspondence to: msehc@nus.edu.sg

1. Supplementary Text

The electromagnetic interference shielding effectiveness (EMI SE), is the ability to attenuate EM radiation. It is measured in decibels (dB) and defined as the logarithmic ratio of incoming power (P_I) to transmitted power (P_T) as [1]

$$SE(dB) = 10\log\left(P_I/P_T\right)$$

Total EMI SE of a material consists of three mechanisms: reflection (SE(R)), absorption (SE(A)), and multiple reflections (SE(M)) of electromagnetic radiation.[2-4] SE(R) is mainly attributed to the interactions between the EM waves and free charges on the surface of the materials.[5] Thus conductive materials with high amount of mobile charge carriers are beneficial for EMI shielding by reflection. For absorption, the shield should have electric and/or magnetic dipoles which could interact with EM waves to attenuate EM energies into thermal and/or internal energies.[6] The SE(M) represents the internal reflections within the shielding materials, typically for the materials with large surface area and interface area.[5] When SE(Total) is greater than 15 dB, SE(M) can be neglected.[7] Thus SE(R) and SE(A) are the dominate mechanisms for EMI shielding. For conductive monolithic materials, EMI SE can be expressed using the following equations,[8-9]

$$SE(R) = 39.5 + 10log(\sigma/(2\pi f\mu))$$

$$SE(A) = 8.7t\sqrt{\pi f\mu\sigma}$$

$$SE = 39.5 + 10log(\sigma/(2\pi f\mu)) + 8.7t\sqrt{\pi f\mu\sigma}$$

while f is the frequency of the electromagnetic wave, μ the magnetic permeability, σ the conductivity, and t the sample thickness. Obviously, both SE(R) and SE(A) increase with conductivity, while absorption is also proportional to sample thickness.

In a vector network analyzer, four scattering parameters (S_{11}, S_{12}, S_{21}, S_{22}) were obtained. And SE(Total), SE(R), and SE(A) could be calculated from these parameters using the following equations[1, 10]

$$SE(Total) = 10log\frac{1}{|S_{12}|^2} = 10log\frac{1}{|S_{21}|^2}$$

$$SE(R) = 10 \times log(1/(1 - {S_{11}}^2))$$

$$SE(A) = 10 \times log((1 - {S_{11}}^2)/{S_{12}}^2)$$

where S_{11}, S_{12} and S_{21} are the forward reflection coefficient, reverse transmission coefficient and forward transmission coefficient, respectively.

Specific shielding effectiveness (SSE) is derived to compare the effectiveness of shielding materials taking into account of the density and thickness. It can be calculated as follows[9]:

$$SSE(dB\ cm^2 g^{-1}) = \frac{SE}{\rho t}$$

ρ (g cm^{-3}) is the mass density of the materials and *t* (cm) is the thickness.

It is more reality to evaluate the shielding efficiency of materials for the practical applications by taking into account of density and thickness.

EMI shielding efficiency presents the material ability to block waves in terms of percentage. For commercial application, EMI SE of 20 dB is required, which corresponds to 99% blockage of incident radiation. EMI shielding effectiveness [dB] is converted into EMI shielding efficiency [%] by the equation as follow:

$$Shielding\ efficiency\ (\%) = 100 - \left(\frac{1}{10^{\frac{SE}{10}}}\right) \times 100$$

The relationship between shielding efficiency (dB) and shielding efficiency (%) is displayed as below:

Shielding Effectiveness (dB)	Shielding Efficiency (%)
10	90
20	99
30	99.9
40	99.99
50	99.999
60	99.9999
70	99.99999
74.9	99.999997

2. Experimental

2.1 Materials

Aniline, ammonium peroxidisulfate (APS), m-cresol, and camphor sulfonic acid (CSA) were all purchased from Sigma-Aldrich. Aniline was purified by vacuum distillation before use. Amine functionalized SWCNT/DWCNT (A-CNT) and unmodified SWCNT/DWCNT (U-CNT) were purchased from Cheaptubes with purity > 99%, out diameter:1-4nm, length:3-30 μm.

2.2 Synthesis of CNT/PANI composite powders

The CNT/PANI composite was synthesized by in situ polymerization of aniline with ammonium peroxidisulfate (APS) as the oxidant in the presence of CNT. First, 50 mg CNT was ultrasonic for 15min and then disperse in 1M HCl solution at 0 ^{o}C in an ice bath.

Second, a different amount of aniline was added by stirring for 0.5 h. Then the precooled APS in 1 M HCl solution was slowly added dropwise to the reaction mixture. The polymerization reaction was performed for 24 h by stirring in an ice bath. After that, the products were filtrated and washed with deionized water and ethanol. Next, the as-prepared products were dedoped in 0.1 M ammonia solution for 24h. Finally, the products were filtrated, washed with deionized water and ethanol, and then drying at 60 ^{o}C in the vacuum. The CNT contents were determined on the basis of the initial weight of the CNT and the final dry weight of the composites.[11-13] The PANI powders were synthesized by the same process.

The CNT/PANI composite was doped with camphor sulfonic acid (CSA) according to a PANI : CSA mole ratio of 2 : 1 in an appropriate amount of m-cresol. The mixture was stirred at room temperature for overnight. CNT/PANI composite films were prepared by casting the mixed solution on glass substrates and drying at 40 ^{o}C in air. The film thickness in the range of 5-30 μm was obtained via the amount of the solution cast.

2.3 Characterization

Attenuated total reflection Fourier transform infrared spectra (ATR-FT-IR) were obtained using a Shimadzu IR Tracer-100 spectrometer with the wavenumber range of 600-4000 cm^{-1}. The morphology of the synthesized CNT/PANI composite was investigated under Zeiss Supra 40 field-emission scanning electron microscope (SEM) at an accelerating voltage of 5 kV and JEOL2010F transmission electron microscopy (JEM-2010F). Temperature dependences of the resistivity of the films were measured by Janis Research VPF-475 dewar with liquid nitrogen as the coolant. The thicknesses of the films were confirmed by a surface profiler. The conductivities of the polymer films were measured by

the van der Pauw four-point probe technique with Keithley 2400 source/meter.[14-15] The CNT/PANI composite films were prepared on square glass substrates of 1.3 × 1.3 cm^2 by drop coating CNT/PANI composite solution. The electrical contacts were made by coating silver paint on the four corners of each film on glass substrate. The probes were contacted on the four corners of the films to obtain the resistance of the films and the distance between adjacent probes was 1.3 cm. The conductivity (σ) was calculated from $\sigma = 1/(R_S d)$, where R_S is sheet resistance and d is film thickness. At least 5 samples for each condition were measured for the test. EMI shielding measurements were conducted using Agilent/HP 8510C Vector Network Analyzer (VNA) in the frequency range of 8.2–12.4 GHz (X-band). The film was prepared with size of 2.5 cm × 2.0 cm which is larger than the tunnel size of the sample holder of 2.28 cm × 1.01 cm. And then the sample was attached on the sample holder with scotch tape. Finally, the sample holder was tightly fixed to the waveguide adapters with screws for the measurement. The photograph of the measurement for EMI shielding effectiveness was showed as Figure S1.

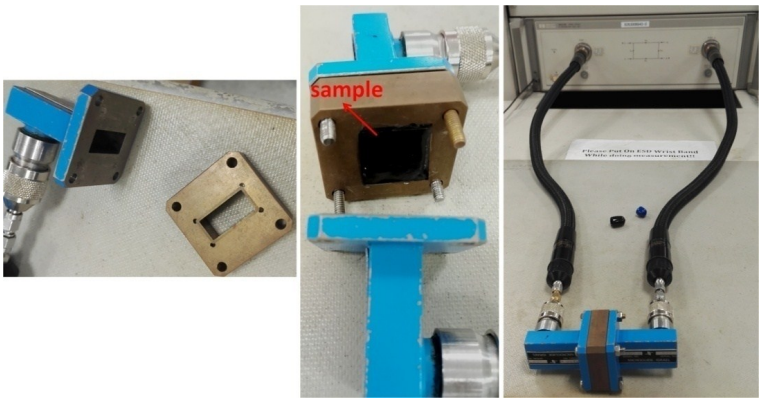

Fig. S1. The photograph configuration of the measurement for EMI shielding effectiveness.

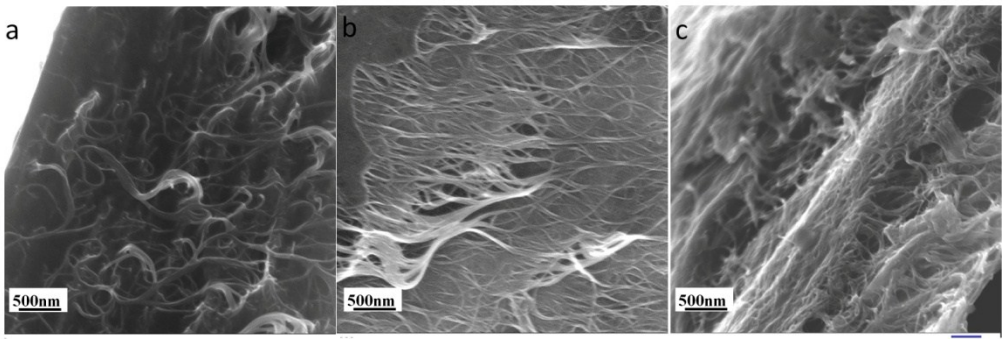

Fig.S2. Cross-sectional SEM images of A-CNT/PANI films with A-CNT content of 13 wt% (a), 53 wt% (b), and 70 wt% (c).

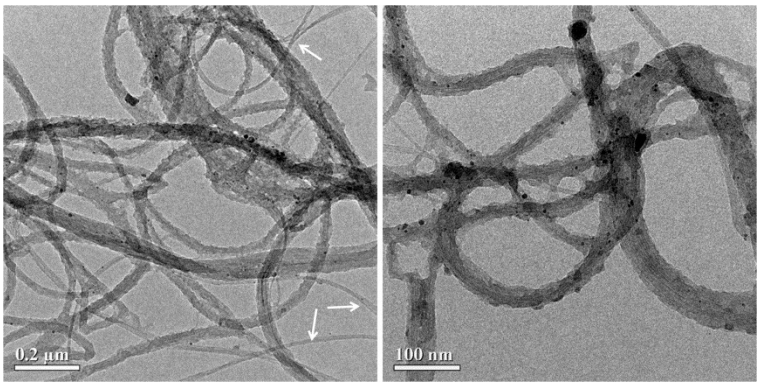

Fig. S3 TEM morphology of 53 wt% U-CNT/PANI composites. As shown in the image, some naked CNT is observed in the composites (indicated by the white arrows). Besides, the PANI is not homogeneous coated on the surface of U-CNT. Some nodules of PANI were observed on U-CNT.

Table S1. The electrical conductivity of CNT/PANI and graphene/PANI composites.

Composites	**Filler content(wt%)**	**σ (S cm^{-1})**	**References**
SWCNT/PANI	65	1440	[11]
SWCNT/PANI	-	1138	[16]
SWCNT/PANI	-	61.5	[13]
MWCNT/PANI	10	152.8	[17]
CNT/PANI	40	17.1	[18]
SMCNT/PANI	41.4	125	[12]
MWCNT/Porous PANI	1	14.1	[19]
DWCNT/PANI	30	610	[20]
PANI-CNT / PANI	30	28	[21]
SWCNT/PANI	64	769	[22]
rGO /PANI	50	18	[23]
Graphene/PANI	48	814 856	[24-25]
Graphene/PANI	16.7	116	[26]
GNP/PANI	43	59 174	[27]
A-CNT/PANI	**70**	**3009**	**Our work**

GNP: graphene nanoplatelets

Table S2. The sheet resistance and conductivity of 70 wt% A-CNT/PANI films with different film thickness.

	5 μm	10 μm	20 μm	29 μm
Sheet resistance (Ω sq^{-1})	0.66 ± 0.040	0.34 ± 0.027	0.17 ± 0.011	0.11±0.0064
Conductivity (S cm^{-1})	3009±252	2960±235	2968±190	3067±198

Table S3 EMI SE of various shielding materials.

	Materials	Filler content/ wt%	Conductivity/ S cm^{-1}	Thickness/ mm	SE/dB	Band range/GHz	References
CNT	MWCNT/PU	22	0.1	0.1	20	8-12	[28]
	SWCNT/PU	20	2.2 x 10^{-4}	2	17	8-12	[29]
	CNT/cellulose paper	8.32	2	0.45	20	15-40	[30]
	CNT-PEO/cellulose	40	20.0	0.15	35.0	8.2–12.4	[31]
	SWCNT/epoxy	15	0.2	2	25	8-12	[32]
	MWCNT/WPU	76.2	21	0.8	80	8-12	[33]
	MWCNT/PS-PMMA	1	1x10(-4)	5	24.1	8-18	[34]
	COOH-MWCNT/PU	10	0.124	1.5	29	8-12	[35]
	MWCNT/ABS	15	1.2	1.1	50	8-12	[36]
	SWCNT/PU	5	1x10-7	2	21.9	8-12	[37]
	MWCNT/LDPE	10	0.63	1.65	22.4	12.4-18	[38]
	MWCNT/UHMWPE	10	~2	1.0	50	8-12.4	[39]
	MWCNT/TPU	10	0.456	2	21.8	8-12	[40]
	MWCNT/WPU	76.2	0.446	4.5	52	8-12	[41]
	MWCNT/PU	10	7.9	2.5	41.6	8-12	[42]
	MWCNT/PS	7	0.01	-	18.6	8-12	[43]
	MWCNT/PC	20	~3	2.15	43	8-12	[44]
	MWCNT/PS	20	1	2	60	8-12	[45]
Graphene	r GO/PS	7	0.435	2.5	45.1	8-12	[7]
	Graphene	100	1000	0.0084	20	8-12	[46]
	Graphene/PDMS	0.8	2	1	20	8-12	[4]
	rGO/SiO2	20	-	1.5	38	8-12	[47]
	Graphene/WPU	7.7	0.051	2	32	8-12	[48]
	r GO/PE	0.66vol%	0.034	2.5	32.4	8-12	[49]
	Graphene/PS	30	0.0125	2.5	29	8-12	[50]

Metal	Al foil	100	$2.8x10^5$	0.008	66	8-12	[9]
	Cu Foil	100	$8.0x10^5$	0.01	70	8-12	[9]
	Copper	100	$6.0x10^5$	3.1	90.2	8-12	[51]
	Nickel	100	$1.2x10^5$	2.85	82.1	8-12	[51]
	Stainless steel	100	$1.4x10^4$	4	88.9	8-12	[51]
	Ni filaments/PES	19	-	2.85	91.7	8-12	[51]
	Ni fibers/PES	43	-	2.85	73.7	8-12	[51]
	Ni-CB/Resin	50	0.32	1	85	8-12	[52]
	Ag Nanowires/PANI	14	5300	0.013	50	8-12	[53]
	Cu Nanowires/PS	2.1	-	0.2	35	8-12	[54]
	Ag Nanowires/PS	2.5	19	0.8	33	8-12	[55]
Others	PEDOT:PSS/WPU	20	77	0.15	62	8-12	[8]
	PANI/PU	50		1.9	26.7	8-12	[56]
	Ag@graphene/PANI	5	20.32	1	29.33	8-12	[57]
	MWCNT/PANI	25	19.7	2	39.2	12.4–18.0	[58]
	MWCNT/PANI		14.6	0.5	47	0.8-3	[59]
	Graphite/PANI	15.6	12.5	2.02.7	33.6	8.2–12.4	[60]
	Graphite/PANI	17.4	67.4	2.0	39.7	8.2–12.4	[61]
	Graphene/PANI	33	19	2.4	34.2	2.0-18	[62]
	SWCNT/PANI	25	10	2.4	31.5	2.0-18	[62]
	Graphite-CNT/PANI	15	29.5	2.50	98	12.4-18	[63]
	Graphene/PANI	16.7	116	0.005	~42 ~32	4-8 8-12	[26]
	PANI nanofibers	100	18.5	1	74	8.2-18	[64]
	Graphite-PANI nanofiber	2.3	24.0	1	87	8.2-18	[65]
	DBSA doped PANI	100	2.09	2.7	55.04	12.4-18.0	[66]
	DBSA doped PANI	100		0.017	21.6	30M-	[67]

						1.5G	
	CSA doped PANI	100	10-100	0.001-0.030	~35	0.1M-1000M	[68]
	Graphene foam/PEDOT:PSS	82	43.2	1.5	91.9	8-12	[5]
	r GO/PEDOT	23		2	35.5	2-18	[69]
	rGO/PEDOT:PSS	25	6.84	0.8	70	8-12	[70]
	Ti3C2Tx	100 90	4800 2900	0.045 0.008	92 57	8-12	[9]
Our work	A-CNT/PANI	70	3009	0.005	50.2	8.2-12.4	
		70	3009	0.029	74.9	8.2-12.4	

Table S4. EMI SSE of various shielding materials.

Materials	Filler content/ wt%	Conductivity/S cm^{-1}	Thickness/ mm	Density(g cm^{-3})	SE/ dB	SSE [dB cm^2g^{-1}]	References
SWCNT/epoxy	15	0.2	2	1.3	25	96.2	[32]
MWCNT/WPU	76.2	21	0.05	1.2	24	4000	[33]
MWCNT/ABS	15	1.2	1.1	1.05	50	432.7	[36]
MWCNT/WPU	76.2	0.446	1	0.039	21.1	5410	[41]
MWCNT/PC	20	~3	0.43	~1.1	19	401.7	[44]
CNT Spong	100	2.78	2.38	0.02	22	4622	[71]
CNT-PEO/Cellulose	40	20.0	0.15	~1.7	35	1372.4	[31]
MWCNT/PS	20	1	2	~1.05	60	285.7	[45]
r GO/PS	7	0.435	2.5	0.26	45.1	692	[7]
Graphene/PEI	10	$2.2x10^{-5}$	2.3	0.29	12.8	191.7	[72]
Graphene/PS	30	0.0125	2.5	0.45	29	257.8	[50]
rGO/PI	16	0.008	0.8	0.28	21	937.5	[73]
rGO/PEDOT	25	6.84	0.8	1.04	70	841.3	[70]
Graphene/PDMS	0.8	2	1	0.06	20	3333	[4]
Graphene/WPU	7.7	0.051	2	~1.05	32	152.5	[48]
Al foil	100	$2.8x10^5$	0.008	2.7	66	30556	[9]
Cu Foil	100	$8.0x10^5$	0.01	8.92	70	7848	[9]
Copper	100	$6.0x10^5$	3.1	8.92	90.2	32.6	[51]
Nickel	100	$1.2x10^5$	2.85	8.9	82.1	32.4	[51]
Stainless	100	$1.4x10^4$	4	7.9	88.9	28.1	[51]

steel							
Ni filaments/PES	7	-	2.85	1.87	87	163.2	[51]
Ni fibers/PES	7	-	2.85	1.87	58.1	109.0	[51]
Ti3C2Tx	90 100	2900 4800	0.008 0.011	2.31 2.39	57 68	30830 25863	[9]
PEDOT:PSS/ WPU	20	77	0.15	~1.0	62	4133	[8]
Graphene foam/PEDOT :PSS	82	43.2	1.5	0.022	69.1	20827	[5]
A-CNT/PANI	70	3009	0.005 0.010 0.020 0.029	1.34	50.2 57.5 67.4 74.9	74900 42900 25100 19300	**Our work**

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