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## **Electronic supplementary information**

Single-layer Copper Particles Integrated with Carbon Nanotube Film for Flexible

Electromagnetic Interference Shielding

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Fig. S1. The schematic diagram of the measurement of electromagnetic interference shielding setup

A rectangular waveguide via a two-port network analyzer system was used for measurement the EMI SE of the different samples. After the sample was cut into a rectangular sheet with an area slightly larger than the corresponding holder, it was held on the holder for S-parameter measurements ( $S_{11}$  and  $S_{21}$ ).



Fig. S2. The photograph of the continuous CNT macroscopic tube



Fig. S3. The surface of the copper foil

Lot of microbumps with height of about  $\sim 1 \ \mu m$  on the surface of the copper foil that used as the substrate can be found obviously. These microbumps were embedded in the surface of the assembled macroscopic tube when it was been squeezed, thereby obtained CCTFs replicate the surface topography of the substrate.



Fig. S4. The schematic diagram of electro-deposition setup



**Fig. S5.** The photographs of the CCTF-Cu and their flexibility, (a) the morphologies of Cu foil, CCTF-Cu and CCTF, qualitative demonstration of insensitivity of the composite towards (b) folding and (c) curling



Fig. S6. The XRD patterns of CCTF-Cu composite film at different side

The different sides of the composite were checked by XRD. The diffraction characteristic peak for CCTF side located at 26.2° is corresponding to the (002) for graphite. In addition, the diffraction characteristic peaks 43.3°, 50.5° and 74.4° correspond to (111), (200), (101) and (220) plane for Cu, respectively. And copper also has been demonstrated by XRD pattern of the other side of the composite. The results indicated the copper particles have been successfully deposited onto the CCTF.



**Fig. S7.** The typical SEM images of the copper deposited on the CCTF's surface, (a) the low and (b) magnified images of copper particles inside the porous surface of the CCTF after deposition 10 seconds, the SEM image of the cross section of the copper deposited on the CCTF after (c) 60 and (d) 120 seconds

After electro-deposition 10 s, the particle on the surface of CCTF can be observed. And the magnified SEM images indicated the particles have infiltrated into the CCTF's surface. The porous surface provides enough positions for the sub-granular area of the obtained particles with diameter of ~ 100 nm, and thereby enhanced the contact area between the particles and CCTF. Moreover, the single-layer of particles with ~ 400 nm could be completely covered on the surface after deposition 60 s. It is interesting that the single-layer of particles on the surface also can be obtained even 120 s later. The difference is that the particles with long time deposition have integrated into bigger and flatter particles. The thickness of the composite film is ~ 2  $\mu$ m can be seen clearly on the image.



**Fig. S8.** The surface morphologies of the CNF assembled with (a) ethanol and (b) CCTF from compress macroscopic tube that deposited copper particles with 60 s

The morphologies of the samples that deposited copper with 60 s on CNFs and CCTFs show different colors. The CNF-Cu shows black on most of the areas, and small areas shows pale purple. The results demonstrate little copper particles have deposited. However, the CCTF-Cu shows uniform purple on the surface, which indicate copper particle deposited homogenous on the surface. In addition, the surface of the CCTF-Cu shows similar morphology to that of the CCTF.



Fig. S9. The thermogravimetric curves of CCTF and CCTF-Cu (electro-deposition 60 s)

The content of Fe and Cu elements in the CCTF-Cu was tested by thermogravimetric analysis (Diamond TG/DTA6300) with the heating rate of 10 °C min<sup>-1</sup> from room temperature to 1000 °C at an air flow rate of 100 ml min<sup>-1</sup>. The weight loss below 400 °C could be attributed to the evaporation of H<sub>2</sub>O and the oxidation of amorphous carbon. From 400 to 600 °C, there is a sharp weight loss in the samples, which is related to the consumption of graphitic walls of CNTs. And the oxidized process indicates the weight increase in the curve. After burning at high temperature, the weight of the residual material, which is presumably Fe<sub>2</sub>O<sub>3</sub> and CuO. Therefore, the Cu content in the composite of ~ 80 wt% can be estimated.



Fig. 10. The EMI SE performance of CCTF from electro-deposition copper particles within 60 s

The EMI SE increases obviously with the prolongation of the deposition until the time reaching 60 s. The SE is ~40 dB after the sample deposition Cu particles with 10 s, and the value suddenly increase to ~50 dB after the time prolong to 20 s. Moreover, the SE will increase to ~60 dB as the electro-deposition time extends to 60 s.



**Fig. S11.** Comparison of EMI SE performance of CCTF-Cu (a) under different electro-deposition time exceeds 60 s under a relative long electro-deposition time

After the electro-deposition time exceeds 60 s, the improvement of EMI SE performance of the CCTF-Cu composite increases slowly. The improved shielding performances are less than 10 dB as the electro-deposition time extended from 60 s to 240 s. The results indicate that it is difficult to achieving continuous SE improvement by prolonging the electro-deposition time and the thickness of the film.



Fig. S12. Comparison of MSE (a) and VSE (b) for various flexible materials that have been reported in recent years



**Fig. S13.** The electromagnetic shielding performance (a), MSE (b) and VSE (c) of CFC and CFC-Cu, the insert images are the photographs of CFC and the obtained CFC-Cu with different deposited time. The area of the CFC as substrate is  $5 \text{ cm} \times 8 \text{ cm}$ .

Resistance	R1 $\Omega$ sq <sup>-1</sup>	R2 $\Omega$ sq <sup>-1</sup>	R3 $\Omega$ sq <sup>-1</sup>	Raverage
0 s	4.27	4.36	4.32	4.32
10 s	1.82	1.79	1.80	1.80
20 s	0.48	0.45	0.47	0.47
30 s	0.31	0.33	0.32	0.32
40 s	0.08	0.07	0.08	0.08
50 s	0.06	0.05	0.04	0.05
60 s	0.02	0.02	0.02	0.02

Table S1. The sheet electrical resistance of the CCTF-Cu composite after different electro-deposition time

The sheet resistance the CCTF-Cu on Cu side was measured by a four-probe system, and the last column represents the average values of sheet electrical resistance of the different electro-deposition times. The results indicated CCTF-Cu has a significant improvement (from  $4.32 \Omega \text{ sq}^{-1}$  increased to  $0.02 \Omega \text{ sq}^{-1}$ ) in electrical conductivity during the electro-deposition time.

pe	Materials	Frequency	Thickness	Density	EMI SE	MSE	VSE	Ref.
Tyl		range (GHz)	mm	g cm <sup>-3</sup>	dB	dB g <sup>-1</sup>	dB cm <sup>-3</sup>	
	Cu foil	8.2-12.4	0.01	8.97	70	318.2	2800	[1]
	Al foil	8.2-12.4	0.008	2.7	66	1245.2	3370.1	[1]
	Ni	8.2-12.4	1.95	N/A	23	N/A	N/A	[23]
	Ni-Co Fiber	8.2-12.4	2.5	N/A	41.2	N/A	N/A	[24]
sed	Ag/CF	8.2-12.4	2.5	N/A	38	N/A	N/A	[25]
l Bas	Ag Nanowires	8.2-12.4	0.8	N/A	33	N/A	N/A	[26]
Meta	PP-SSF	8.2-12.4	3.1	0.64	48	9.8	6.3	[2]
	CuNi	8.2-12.4	1.5	0.24	25	28.4	6.8	[3]
	CuNi-CNT	8.2-12.4	1.5	0.23	54.6	65.0	14.9	[3]
	CuS/PANI	0.5-3	3	N/A	45.2	N/A	N/A	[37]
	CoFe <sub>2</sub> O <sub>4</sub> /PANI-PTSA	8.2-12.4	4	N/A	22.5	N/A	N/A	[34]
	NiFe <sub>2</sub> O <sub>4</sub> -BaTiO <sub>3</sub> ceramics	8.2-12.4	1.2	N/A	34	N/A	N/A	[36]
	Ti <sub>3</sub> C <sub>2</sub> Tx-SA	8.2-12.4	0.045	N/A	90	N/A	N/A	[38]
ed	MXene foam	8.2-12.4	0.006	0.39	32	5614.0	2178.6	[4]
MXene Base	Ti <sub>3</sub> C <sub>2</sub> Tx	8.2-12.4	0.045	N/A	92	N/A	N/A	[1]
	MXene/PS	8.2-12.4	2	N/A	62	N/A	N/A	[30]
	MXene/CNF	8.2-12.4	0.047	0.19	24	1097.9	208.6	[31]
	CF/PC/Ni film	0.03-1.5	0.31	1.7	72.7	56.4	95.8	[5]

 Table S2. Comparison of EMI shielding performance for various flexible materials

MWCNT/WPU	8-12	2.3	0.032	46.7	259.4	8.3	[6]
GF-K2	2-18	0.031	1.63	130	1048.4	1713.1	[7]
MCMBs/MWCNTs/Fe <sub>3</sub> O <sub>4</sub>	8.2-12.4	0.5	0.5	80	130.7	65.4	[8]
Flexible graphite	0.03-1.5	0.2	1.1	110	203.7	224.7	[9]
MWCNT/PLA foam	8.2-12.4	2.5	0.3	23	12.8	3.8	[11]
PVDF/CNT	18-26.5	0.1	N/A	25	N/A	N/A	[27]
PVDF/CNT/graphene	18-26.5	0.1	N/A	27.6	N/A	N/A	[27]
CNT/CS foam	8.2-12.4	2.5	0.0176	37.6	349.1	8.47	[28]
CNT/cellulose	8.2-12.4	20	0.095	20.8	4.5	0.42	[29]
MWCNTs/Mn(nps)	8.2-12.4	0.13	0.711	42	185.6	131.9	[32]
CF/Fe <sub>3</sub> O <sub>4</sub>	1-18	13	N/A	20	N/A	N/A	[39]
CF/CNT	12.4-18	1	N/A	21.9	N/A	N/A	[40]
Graphene aerogel films	2-8	0.12	0.02	105	16422	328.5	[41]
CNT sponge	8.2-12.4	1	0.032	30	382.9	12.3	[42]
CNT/PDMS	8.2-12.4	2	N/A	46.3	N/A	N/A	[42]
CNT film	8.2-12.4	0.13	0.011	65	18568	204.2	[43]
CNT/PP	8.2-12.4	2	N/A	43.1	N/A	N/A	[44]
GF	2-18	0.013	0.91	48.9	1688.5	1536.5	[45]
GCF	2-18	0.015	1.45	57.6	1248.2	1809.9	[45]
C/SiO <sub>2</sub> /SiC aerogels	12.4-18	10	0.223	24	4.4	0.98	[33]
rGO-PDMS	8-12	1	0.06	20	133.3	8.2	[10]
rGO-PS	8.2-12.4	2.5	0.26	45.1	28.4	7.4	[14]

	rGO-PEI	8.2-12.4	2.3	0.92	22	4.25	3.9	[15]
	rGO/Fe <sub>3</sub> O <sub>4</sub> -PEI	8-12	2.5	0.4	18	7.35	2.9	[16]
	Graphene foam	8.2-12.4	0.3	0.06	25	568.2	34	[17]
	Graphene film	8-12	0.05	0.15	60	3260.8	490.2	[18]
	rGO/γ-Fe <sub>2</sub> O <sub>3</sub> -PANI	8-12.4	2.5	N/A	51	N/A	N/A	[19]
	rGO/ MnO <sub>2</sub>	12.4-18	3	N/A	57	N/A	N/A	[20]
	rGO-BaTiO <sub>3</sub>	8.2-12.4	1.5	N/A	41.7	N/A	N/A	[21]
	rGO/CF/γ-Fe <sub>2</sub> O <sub>3</sub>	8.2-12.4	0.4	N/A	42.8	N/A	N/A	[22]
	rGO/CA film	8.2-12.4	0.012	N/A	25.7	N/A	N/A	[35]
	GNRs/MnO <sub>2</sub>	12.4-18	3	N/A	57	N/A	N/A	[46]
	GNPs/rGO/EP	8.2-12.4	3	N/A	51	N/A	N/A	[47]
	PEI-rGO	8.2-12.4	4.5	N/A	26	N/A	N/A	[48]
	PI-rGO	8.2-12.4	0.8	0.28	21	38.3	10.7	[49]
sed	CNF	8.0-12.6	~0.004	0.1	67	68367.4	6842.3	[12]
film Bat	CNF	40-60	~0.004	0.1	57	58163.3	5821.1	[13]
CNT	CCTF-Cu	2.6-26.5	~0.002	0.12	84	151724.1	17973.9	This work



Movie S1. The light and flexible CCTF-Cu composite film.



**Movie S2.** The demonstration of CCTF-Cu composite and commercial Cu foil wound on cables (CNT face (a, b) or Cu-face (c, d) toward outer side.



**Movie S3.** The demonstration of mechanical flexibility of CCTF-Cu composite (CNT face (a-c) and Cu-face (d-f) toward outer side)



**Movie S4.** The demonstration of the morphologies of CCTF-Cu composite (CNT face (a-c) and Cu-face (d-f)) and commercial Cu foil after deformation



Movie S5. The demonstration of electromagnetic wave shielding with CCTF-Cu composite.

When the mobile phone was not been covered by the CCTF-Cu, the electromagnetic wave detector detected that the highest electromagnetic wave intensity is 17.80  $\mu$ T. The screen of the mobile phone displays the calling successes, which indicates that the electromagnetic wave for communication has not been shielded. While the mobile phone was covered with the CCTF-Cu composite, the electromagnetic wave detector indicates that the electromagnetic wave intensity is 0  $\mu$ T. This phenomenon suggests that the electromagnetic wave for communication was shielded successfully by the CCTF-Cu composite.

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