Supporting Information for

Large-Area Potassium-Doped Highly Conductive Graphene Films for Electromagnetic Interference Shielding

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Characterizations. Renishaw in *Via*-Rlflex Raman Microscropy recorded Raman spectra at an excitation wavelength of 532 nm. To avoid laser heating effect, the laser power at sample was kept below 0.5 mW. X-ray diffraction (XRD) measurement were carried out on an X'Pert Pro diffractometer (PANalytical) using Cu Kαl radiation (λ=1.5406 Å) with scanning speed of 6 degree min^{-1} at room temperature. To avoid the sample being oxidized, the test samples of Raman and XRD were sealed within the commercial scotch tapes. The scanning electron microscopy (SEM) images and energy dispersive spectroscopy (EDS) elemental mapping were taken on a Hitachi S4800 field-emission SEM system. The SEM samples were characterized at once after they were taken out of the glovebox.

Electrical Performance Testing. A four-probe method on an electrical transport properties measurement system comprising a Keithley 2400 multiple-function source-meter was used to measure the electrical transport properties. For the air and water sensitive samples, the GF-K film was tested in the glove box.

EMI SE Testing. SE in 2-18 GHz was tested with a modified version of the flanged coaxial sample holder. The modified sample holder is smaller in dimensions (inner diameter=3 mm, outer diameter=7 mm, flange edge diameter= 80 mm). The two sample holders are connected with a ZNB 40 vector network analyzer (Rohde & Schwarz, Germany) through a pair of 50 Ω double-shielded coaxial cables. The samples were tested immediately after taking out of the glovebox.

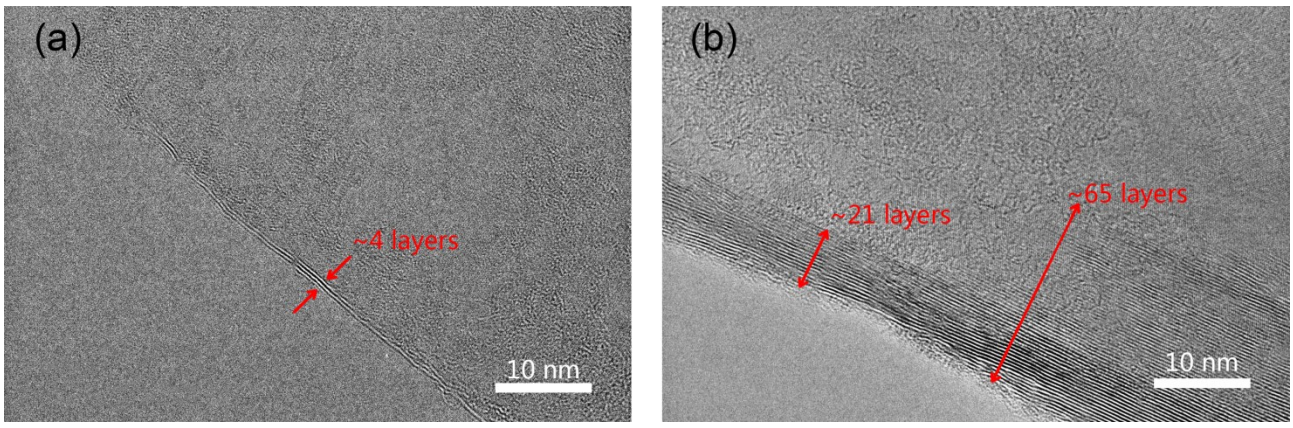

Figure S1. TEM image of GF.

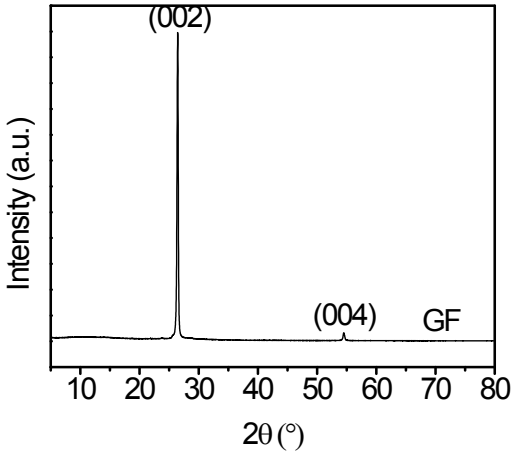

Figure S2. XRD pattern of GF.

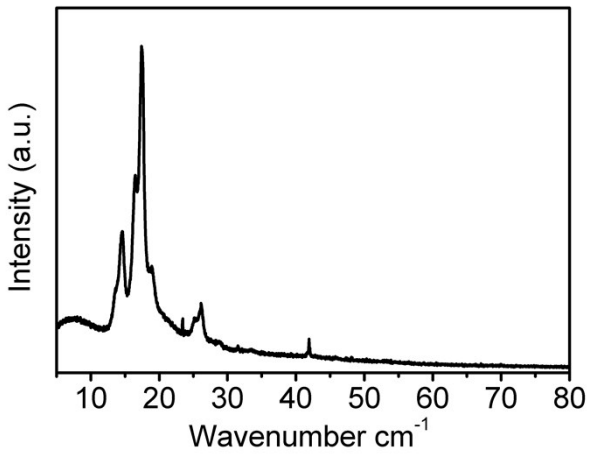

Figure S3. XRD pattern of potassium

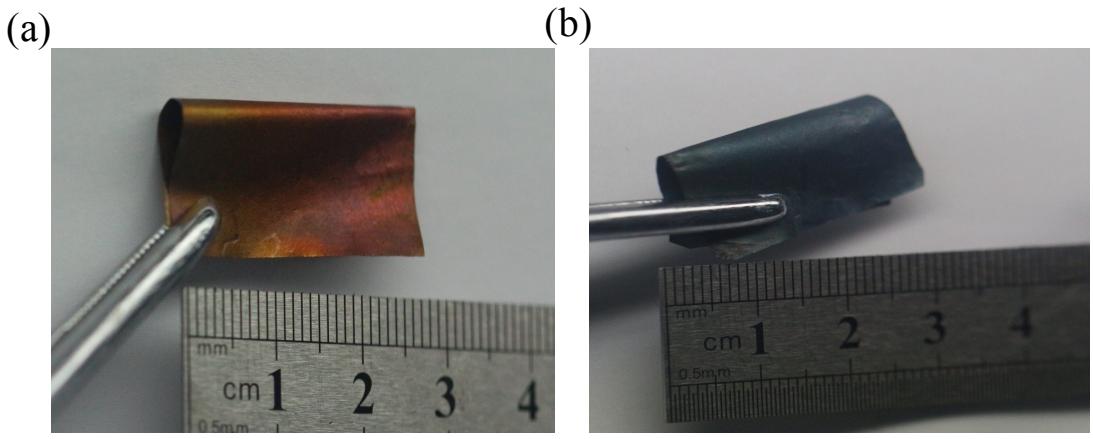

Figure S4. a) Photograph of the bent GF-K1 film, b) photograph of the bent GF-K2 film.

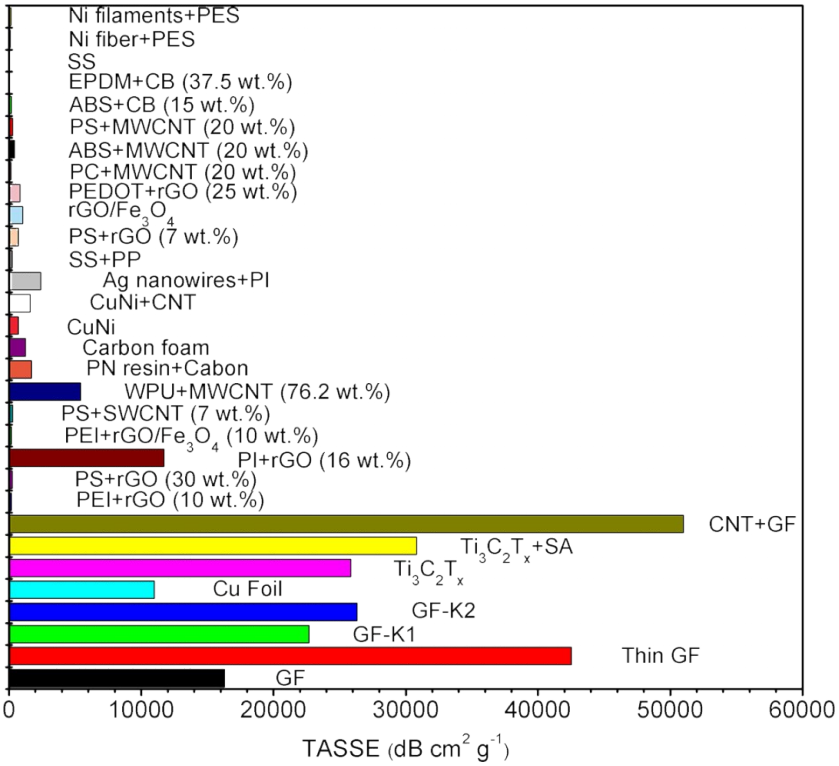

Figure S5. Thickness-averaged specific shielding effectiveness (TASSE) of various shielding materials.

Table S1. TASSE of various shielding materials.

Type		Filler	Filler [wt.%]	Matrix	t [cm]	SE [dB]	SSE/t [dB cm^2g^{-1}]	Ref
Foam Structure	Carbon Based	rGO	10	PEI	0.23	12.8	191.3	1
		rGO	30	PS	0.20	29	257.6	2
		rGO	16	PI	0.08	21	11712	3
		rGO/Fe_3O_4	10	PEI	0.25	18	176	4
		SWCNT	7	PS	0.12	18.5	275	5
		MWCNT	76.2	WPU	0.1	21.1	5410	6
		Carbon	/	PN resin	0.2	51.2	1705	7
		Carbon foam	Bulk	/	0.2	40	1250	8
		GF		CNT	0.08	23.7	510000	9
	Metal	CuNi	Bulk	/	0.15	25	690	10
		CuNi-CNT	Bulk	/	0.15	54.6	1580	10
		Ag nanowires	4.5	PI	0.5	35	2416	11
		SS	1.1#	PP	0.31	48	241.9	12
Solid Structures	Carbon Based	rGO	7	PS	0.25	45.1	692	13
		rGO/Fe_3O_4	Bulk	/	0.03	24	1033	14
		rGO	25	PEDOT	0.08	70	841	15
		MWCNT	20	PC	0.21	39	164	16
		MWCNT	15	ABS	0.11	50	432.7	17
		MWCNT	20	PS	0.2	30	285	6
		CB	15	ABS	0.11	20	190	17
		CB	37.5	EPDM	0.2	18	15.1	18
	Metal	SS	Bulk	/	0.4	89	27.5	19
		Ni fiber	7#	PES	0.285	58	108.7	19
		Ni filaments	7#	PES	0.285	87	164.9	19
		Cu Foil	Bulk	/	0.0013	127	10976.7	This work
	MXene	$Ti_3C_2T_x$	Bulk	/	0.0011	68	25863	20
		$Ti_3C_2T_x$	90	SA	0.0008	57	30830	
	GF-K	GF-K1	Bulk	/	0.0038	>125	>22686	This work
		GF-K2	Bulk	/	0.0031	>130	>26321	
	GF	GF	Bulk	/	0.0027	74	15572.4	
		Thin GF	Bulk	/	0.0012	50	42500	

Notes: / indicates that the values were either not available or impossible to calculate; # -Vol. %; SS – Stainless steel; Bulk - 100% pure material with no polymeric binder; Thin GF shows a better TASSE.

The choice of the standard frequency, thickness and areal density in VASE and WASE

When evaluating VASE and WASE with Simon formula, the error is inevitable since the structural information is not included in this equation. To limit the error in calculation, the frequency and thickness should be closed to the test values. In the existing EMI shielding researches, most tested the shielding performance in 2-18 GHz by coaxial line system or 8-12 GHz by waveguide system, so 10 GHz is always the median in these two frequency bands. That's why 10 GHz is used as a standard frequency.

For the thickness selection, we classified the former reports into two categories: conductive films and polymer-based composites. In the former case, the tested thickness is several micrometers while the tested thickness of composites always approaches 1 millimeter. However, when we select a thickness more than 10 μm, the VASE and WASE will be so high that far beyond the test limits of commercial test machines (130 dB for our case). Therefore, we chose 10 μm as the standard thickness for VASE, and 8.9×10^{-2} g cm^{-2} (10 μm for copper) as the standard areal density for WASE.

The calculation of VASE and WASE

VASE is defined as EMI SE at 10 μm thickness, and WASE is defined as EMI SE at the areal density of 8.9×10^{-3} g cm^{-2}, that is the SE at the thickness of (89/ρ) μm, where ρ(g cm^{-3}) is the density of the shielding material.

The deduction of VASE and WASE was based on simon formula in the text:

$$SE = 90 + 0.017 \times d\sqrt{f\sigma} + 10\log_{10}\left(\frac{\sigma}{f}\right) = f(d,f,\sigma) \quad \text{(S1)}$$

When a serie of data including tested frequency (f), shielding effectiveness (SE), thickness (d) and density (ρ) in known, the value of σ could be obtained by calculating the inverse function of Simon equation:

$$\sigma_s = f^{-1}(d,f,SE) \quad \text{(S2)}$$

Typically, the obtained conductivity σ_s is not accurately the conductivity of the material. For the materials with specific structure, σ_s is the conductivity containing the structure information, and it directly correlates with EMI shielding performance, so we name it as "specific conductivity".

After that, VASE and WASE can be obtained:

$$VASE = f(10\ \mu m, 10\ GHz, \sigma_s) \quad \text{(S3)}$$

$$WASE = f(^{89}/_{\rho}\ \mu m, 10\ GHz, \sigma_s) \quad \text{(S4)}$$

Since Simon formula itself is not accurate when evaluating lowly conductive materials, the VASE and WASE values we give in the tables and figures are not calculated by equation (S1). The actual formula we use is equation (S5), the pre-simplified form of Simon formula:

$$SE = 20log_{10}\frac{2\sqrt{\mu_0/\varepsilon_0}\cdot\sqrt{i\cdot 2\pi\mu_0 f/\sigma}}{\left(\frac{\mu_0}{\varepsilon_0}+i\cdot\frac{2\pi\mu_0 f}{\sigma}\right)\cdot\sinh\left(\sqrt{i\cdot 2\pi\mu_0 f}\cdot d\right)+\left(2\sqrt{\frac{\mu_0}{\varepsilon_0}}\cdot\sqrt{i\cdot\frac{2\pi\mu_0 f}{\sigma}}\right)\cdot cosh\left(\sqrt{i\cdot 2\pi\mu_0 f}\cdot d\right)} = f(d,f,\sigma) \quad \text{(S5)}$$

Where ε_0 is vacuum permittivity, μ_0 is vacuum permeability, i is imaginary units. The rest of the steps are the same as described above.

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