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

A Strong and Tough Polymer-carbon Nanotube Film for Flexible and Efficient Electromagnetic Interference Shielding

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1. The modification of CNT

The original CNT (1 g) was first chemically modified with 60 ml of H_2SO_4/HNO_3 mixture (3:1) under reflux at 60 ^oC for 4 h. Then the mixture was diluted with distilled water and filtered repeatedly until the pH of the filtrate was neutral. The sample was then dried under vacuum at 60 oC. Fig. S1a shows the dispersion of original CNT and modified CNT in distilled water. The original CNT aggregates and precipitates completely within only 30 min after the sonication process. In comparison, the modified CNT dispersion is homogeneous even standing for 48 h. This implies that the chemical modification effectively improves the hydrophilicity of CNT and thus ensures the stable CNT dispersion. FTIR spectroscopy was carried out to determine the surface structure of CNT before and after modification, as shown in Fig. S1b. In the case of modified CNT, the emerging peak at 1731 cm^{-1} is assigned to the characteristic stretching vibration of carbonyl and the peak at 3449 cm^{-1} assigned to hydroxyl is strengthened compared to original CNT. Both affirm the introduction of carboxyl groups on the surface of modified CNT after the acid treatment. Structural changes are further confirmed by Raman spectra, as shown in Fig. S1c. Two characteristic bands are observed: one related to the graphitic structure (G-band) at ~1568 cm^{-1} and the other corresponded to defect (D-band) at ~1334 cm^{-1}. The intensity ratio of D-band to G-band (i.e. I_D/I_G) is used to evaluate the relative extent of structural defects on the surfaces the original and modified CNT. The I_D/I_G ratio increases to 1.27 for modified CNT from 0.98 for original CNT, indicating the intensified surface functionalization and increased structural defects caused by the acid treatment. The degree of functionality was characterized using TGA analysis. As presented in Fig. S1d, the degree of modification is calculated to about 11.1 wt% according to the yields of residual carbon at 600 ^{o}C.

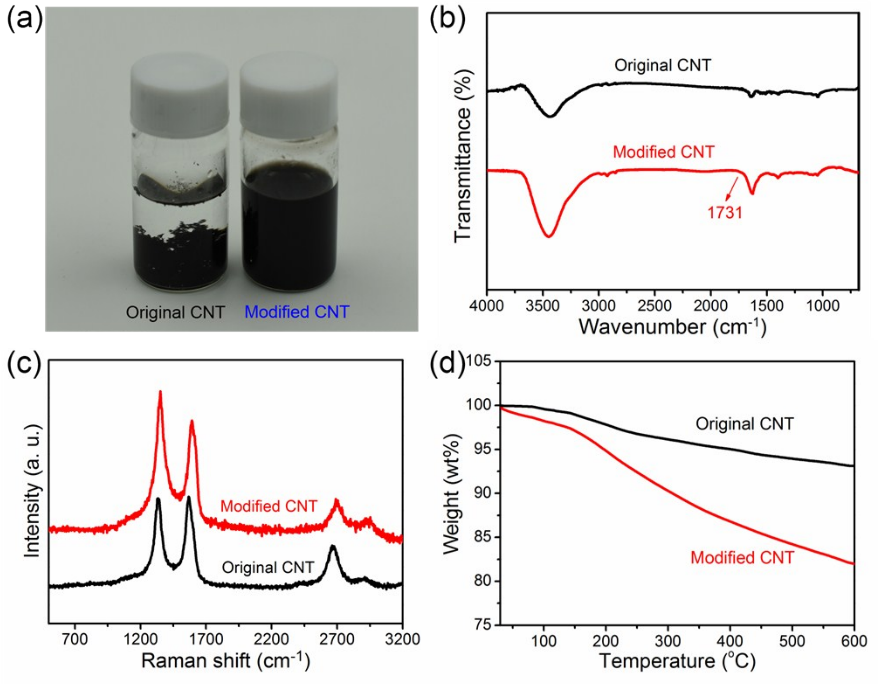

Fig. S1 (a) Digital photographs of original CNT and modified CNT in distilled water taken after 48 hours. FTIR spectra (b), Raman spectra (c) and TGA curves (d) of original CNT and modified CNT.

2. The bending property of modified pure CNT film

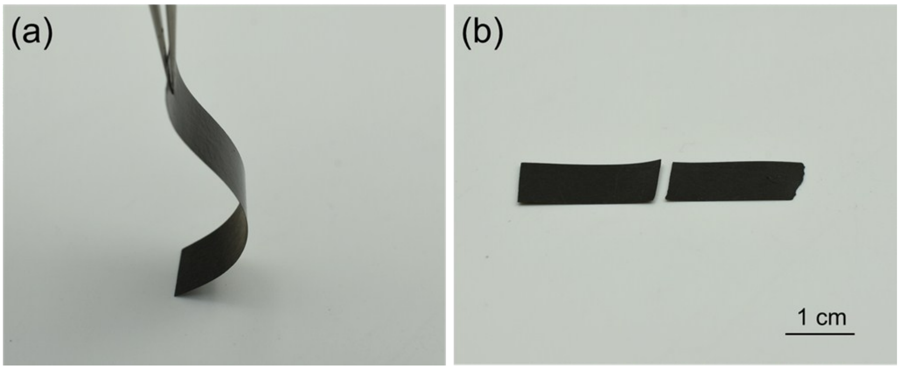

Fig. S2 The bending state of modified pure CNT film (a), and it collapses after bending to a radius of ~13 mm (b).

3. The size distribution of NR latex particles

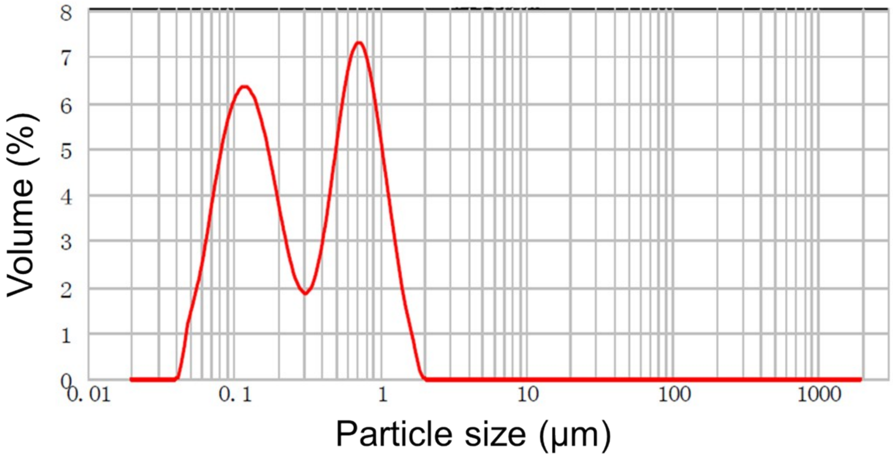

Fig. S3 The size distribution of NR latex particles.

4. The power coefficients of the CNT and CNR films

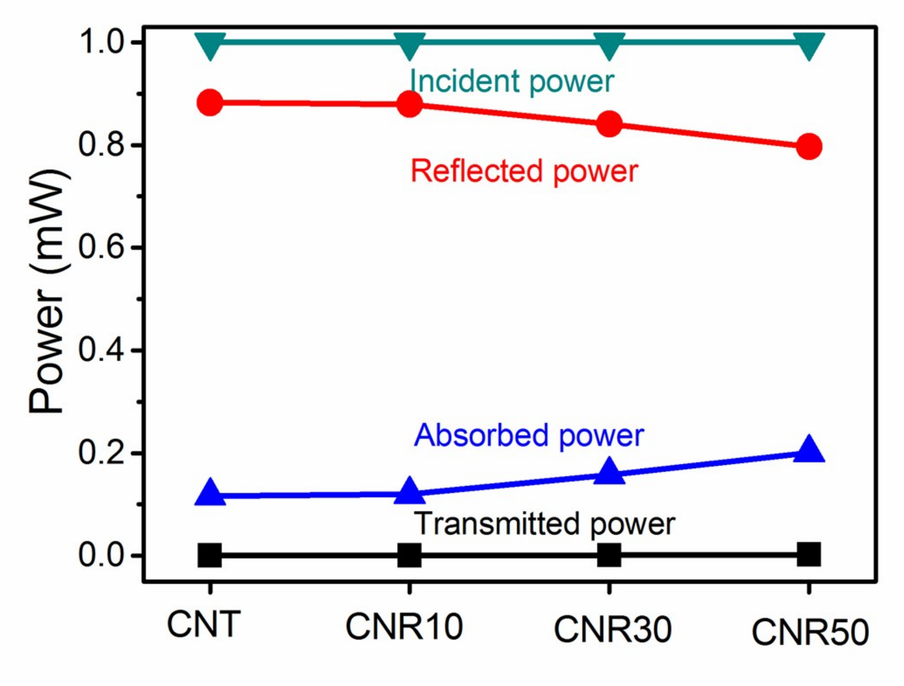

Fig. S4 The power balance at the frequency of 10.3 GHz for the CNT and CNR films.

5. The comparison of SE_{total}, SE_A, and SE_R of the CNR films

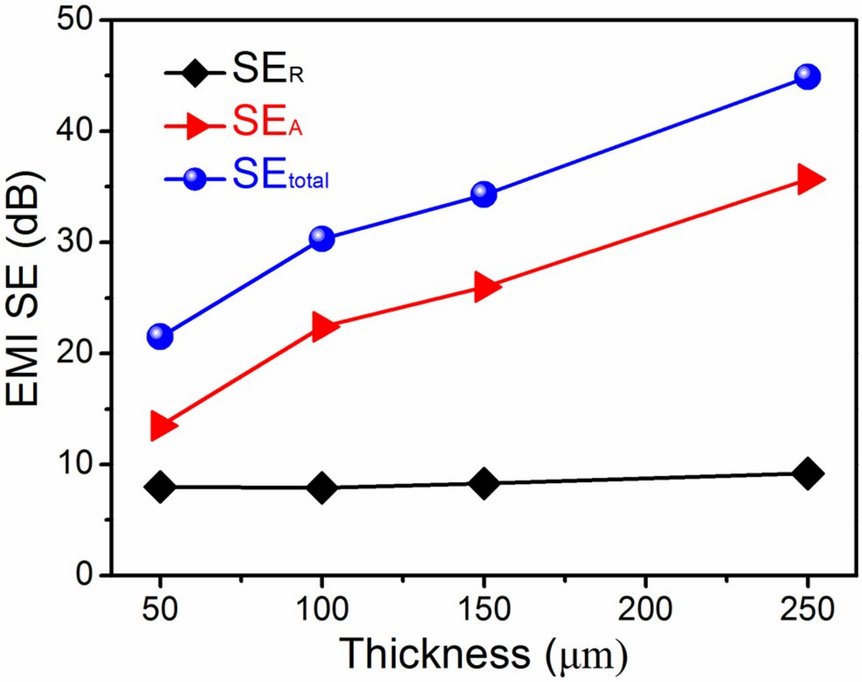

Fig. S5 Comparison of SE_{total}, SE_A, and SE_R of the CNR30 films with various sample thicknesses at a frequency of 10.3 GHz.

6. Comparison of EMI SE of CNR films in this work with the literature.

Table S1 EMI shielding performance for CNR films compared with the CNT and graphene films reported in the literature.

Materials	Thickness (μm)	EMI SE (dB)	Specific EMI SE (dB mm^{-1})	Reference
CNT	**100**	**35.6**	**356**	**This work**
CNR10	**100**	**34.3**	**343**	**This work**
CNR30	**100**	**30.1**	**301**	**This work**
CNR30	**50**	**21.4**	**428**	**This work**
Pd-CNT-Cu	200	35	175	1
Co/Ni-CNT	1000	12	12	2
CNT/PU[a)]	320	49	153	3

CNT/cellulose	170	20.3	119	4
GNS[a]	100	19	190	5
GNS/Fe_3O_4[a]	200-250	22	88-110	6
GNS/EVA[a]	350	25	71	7
GNS/PVA[a]	220	10	45	8
CNF−GNS−CNF[a]	260	27	104	9

[a] PU, GNS, EVA, PVA, GNS, and CNF indicate polyurethane, graphene, ethylene vinyl acetate, polyvinyl alcohol, graphene, and carbon nanofiber, respectively.

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