

## Supporting Information

### Symmetric gradient structure enables robust CNF/FeCo/LM composite film with excellent electromagnetic interference shielding and electrical insulation

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**Table S1** The formula of the C/F<sub>7.3</sub>/L<sub>m:n</sub>-20 composite films with different LM gradient ratio.

Sample	Absorption layer	Transition layer	Reflection layer	Transition layer	Absorption layer
C/F <sub>7.3</sub> /L <sub>1:1</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.0126	0.0054	0.0054	0.0126
	LM (g)		0.0126	0.0126	0.0126
C/F <sub>7.3</sub> /L <sub>1:2</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.0126	0.0054	0.0054	0.0126
	LM (g)		0.009	0.018	0.009
C/F <sub>7.3</sub> /L <sub>1:3</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.0126	0.0054	0.0054	0.0126
	LM (g)		0.0072	0.0216	0.0072
C/F <sub>7.3</sub> /L <sub>1:4</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.0126	0.0054	0.0054	0.0126
	LM (g)		0.006	0.024	0.006
C/F <sub>7.3</sub> /L <sub>1:5</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.0126	0.0054	0.0054	0.0126
	LM (g)		0.0051	0.0255	0.0051

**Table S2** The formula of the C/F<sub>x:y</sub>/L<sub>1:3</sub>-20 composite films with different mass ratio of FeCo

Sample	Absorption layer	Transition layer	Reflection layer	Transition layer	Absorption layer
C/F <sub>5.5</sub> /L <sub>1:3</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.009	0.009	0.009	0.009
	LM (g)		0.0072	0.0216	0.0072
C/F <sub>6.4</sub> /L <sub>1:3</sub> -20	CNF (g)	3.75	3.75	3.75	3.75
	FeCo (g)	0.0108	0.0072	0.0072	0.0108

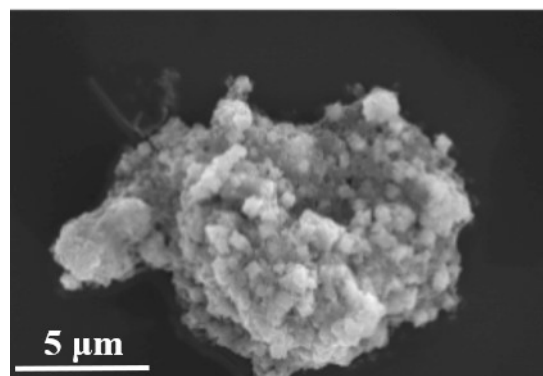
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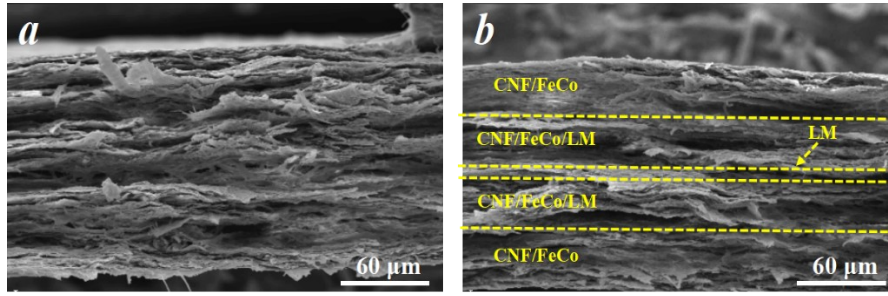
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>7.3</sub> /L <sub>1.3</sub> -20	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0126	0.0054		0.0054	0.0126
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>8.2</sub> /L <sub>1.3</sub> -20	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0144	0.0036		0.0036	0.0144
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>9.1</sub> /L <sub>1.3</sub> -20	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0162	0.0018		0.0018	0.0162
	LM (g)		0.0072	0.0216	0.0072	

**Table S3** The formula of the C/F<sub>8.2</sub>/L<sub>1.3</sub>-z composite films with different content of LM

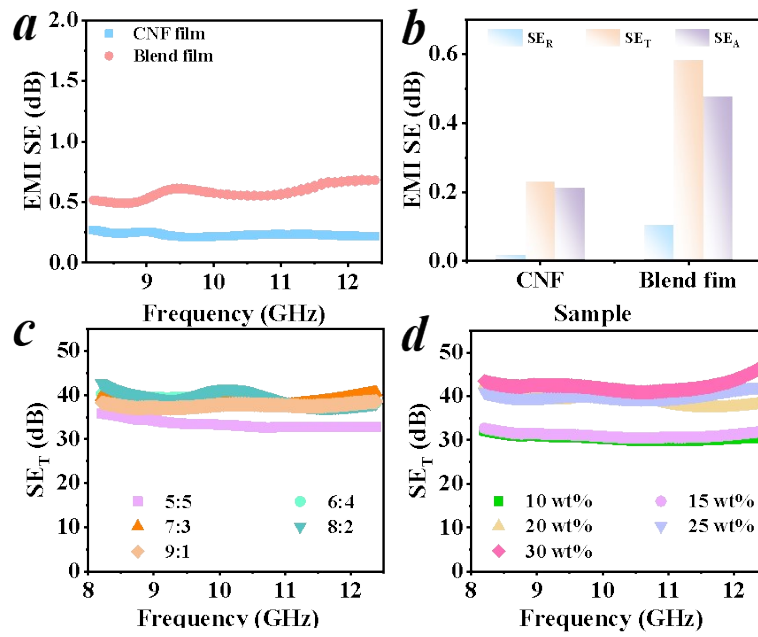
Sample		Absorption layer	Transition layer	Reflection layer	Transition layer	Absorption layer
C/F <sub>8.2</sub> /L <sub>1.3</sub> -10	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.009	0.009		0.009	0.009
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>8.2</sub> /L <sub>1.3</sub> -15	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0108	0.0072		0.0072	0.0108
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>8.2</sub> /L <sub>1.3</sub> -20	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0126	0.0054		0.0054	0.0126
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>8.2</sub> /L <sub>1.3</sub> -25	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0144	0.0036		0.0036	0.0144
	LM (g)		0.0072	0.0216	0.0072	
C/F <sub>8.2</sub> /L <sub>1.3</sub> -30	CNF (g)	3.75	3.75		3.75	3.75
	FeCo (g)	0.0162	0.0018		0.0018	0.0162
	LM (g)		0.0072	0.0216	0.0072	



**Fig. S1** EDS mapping images of the scanning of FeCo nanoparticles.



**Fig. S2** EDS mapping images of the scanning of blend C/F/L-20 film and C/F<sub>8:2</sub>/L<sub>1:3</sub>-20 composite films

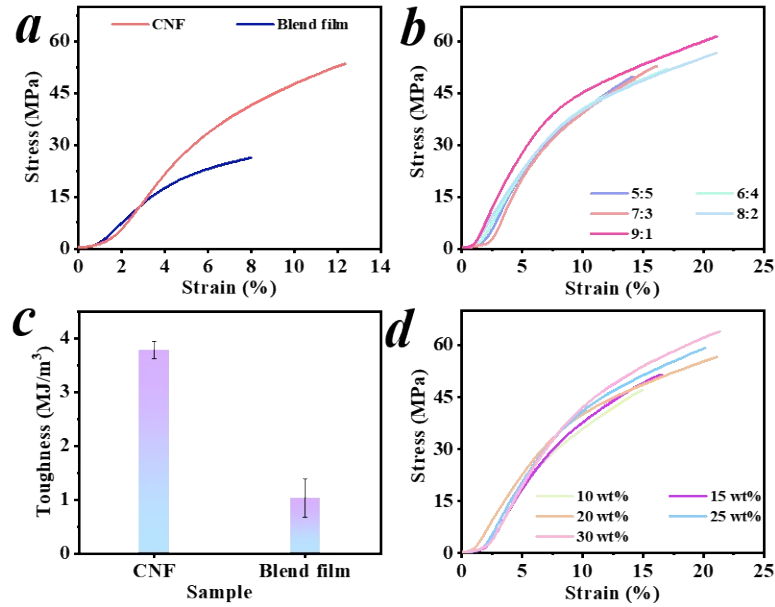


**Fig. S3** (a) EMI SE, (b)  $SE_T$ ,  $SE_R$ , and  $SE_A$  of pure CNF and blend C/F/L-20 film. (c) EMI SE of C/F<sub>x:y</sub>/L<sub>1:3</sub>-20 composite films. (d) EMI SE of C/F<sub>8:2</sub>/L<sub>1:3</sub>-z composite films with different content of LM.

**Table S4** The detailed information is listed of the filler content, thickness, and normalized SE of composite film.

Sample	Conductive Filler	Filler Content (wt%)	Thickness (mm)	EMI SE (dB)	Normalized SE (dB mm <sup>-1</sup> )	Ref.
CP/PGEF/Fe <sub>3</sub> O <sub>4</sub>	CP	30	1	48.9	48.9	[32]
PLLA/graphene	Graphene	5.61	0.3	41.7	139	[33]
PVDF/graphene/Ni	Graphene	10	0.7	51.4	73.43	[34]
Cu-Ni/Wood	Cu	23	0.4	57.4	143.5	[35]
CI/Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /PVDF	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	10	0.4	42.8	107	[36]
CNF/Fe <sub>3</sub> O <sub>4</sub> /LM/GNPs	LM/GNPs	40	0.233	46.6	202.6	[37]
SR/graphene	Graphene	3	0.35	30.42	86.9	[38]

CNF/rGO	rGO	50	3.023	32	10.59	[39]
PI/BNNS/Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	6	0.3	49.2	164	[40]
CNF/FeCo/LM	LM	20	0.170	39.32	231.29	This work



**Fig. S4** (a) Stress-strain curves of pure CNF and blend films. (b) Stress-strain curves of the  $C/F_{x,y}/L_{1:3}-20$  composite films. (c) Toughness of pure CNF and blend film. (d) Stress-strain curves of the  $C/F_{8:2}/L_{1:3}-z$  composite films with different LM content.

**Table S5** The detailed information is listed of the stress, strain, toughness, filler content, and EMI SE/filler content

Sample	Stress (MPa)	Strain (%)	Toughness (M.J. m <sup>-3</sup> )	Conductivity filler content (wt%)	EMI/conductive filler content (dB/%)	Ref.
CNF/Fe <sub>3</sub> O <sub>4</sub> /LM/GNPs	55.27	12.47	2.72	40	1.17	[33]
MXene-xanthan	116.48	1.22	0.84	67	0.46	[42]
CNF@MXene	112.5	4.3	2.7	50	0.79	[43]
NFC/Fe <sub>3</sub> O <sub>4</sub> &CNT/PEO	36.03	19.1	2.98	38.7	0.88	[44]
MXene/ANF	124.1	8.1	6.3	40	1.02	[45]
MXene/UANF	156.9	3.34	2.9	60	0.53	[46]
MXene@HCNT	126.2	3.2	5.5	50	1.44	[47]
PEDOT: PSS/MXene	38.6	0.28	~0.05	30	1.35	[48]
MXene/ANF	158.3	5.03	5.77	80	0.67	[49]
$C/F_{x,y}/L_{m:n}-z$	56.28	21.47	7.2	20	1.97	This work