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Supporting information

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³ Tunable electromagnetic interference shielding effectiveness
⁴ via multilayer assembly of regenerated cellulose as the
⁵ supporting substrate and carbon nanotube/polymer as the
⁶ functional layer

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2 Fig. S1 EMI shielding measurement setup and specimen.



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4 Fig. S2 SEM image of layer-structured CNTs/cellulose film.

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Fig. S2 presents the cross-section of layer-structured CNTs/cellulose film, the sample displays a multilayer structure composed of cellulose layers and PEO/CNTs layer as indicated. The sample for SEM observation was prepared by embedding a piece of layer-structured CNTs/cellulose composite film in epoxy and the cross section was obtained using a microtome equipped with a steel knife. It is worthy of mention that there is some delamination that may be due to the shear force during microtomy.



2 Fig. S3 SE_T, SE_A, SE_R of the layer and plain-structured CNT/cellulose composite films
3 at the frequency of 8.2 GHz.

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5 EMI shielding can be classified into three major mechanisms: reflection, absorption 6 and multiple reflections. Most of the multiply reflected power can be absorbed when 7 the shielding by absorption is higher than 10 dB, thus multiple reflections are often 8 ignored. In addition, total shielding (SE_T), absorption loss (SE_A), reflection loss (SE_R), 9 reflected power, transmitted power and absorbed power can be calculated based on the 10 S parameters obtained from the vector network analyzer.¹

From Fig. S3, we can see that SE_T , SE_A , and SE_R of the layer-structure film are both higher than those of the plain structure film. This result is consistent with that reported in other work.² SE_T , SE_A , and SE_R of the composites increase with increasing CNTs content. Although the total CNTs content in the layer-structure film is same as that in plain structure film, CNTs content in the conductive layer of the plain structure film is much more higher and the conductive network is perfect (as shown in Fig. S4B).

The number of conductive CNTs networks acting as dissipating mobile charge carriers increases and consequently leads to higher SE_A due to the increasing CNTs filler¹. SE_R often relies on the mobile charge carriers (electrons or holes) interacting with the incoming electromagnetic waves, which is related to the conductivity of conductive polymer composites^{3,4}; thus SE_R also increases with increasing CNTs mass ratios because of higher amount of mobile charge carriers at higher CNTs content.



2 Fig. S4 SEM images of freeze dried PEO/CNT composite aerogel at low (A) and high3 (B) magnifications.

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5 Fig. S4A presents the freeze dried PEO/CNT composite aerogel, the sample display a highly porous structure composed of interconnected micro-sheets. The three 6 dimensional micro-sheet scaffolds was formed during freeze-drying process. When the 7 PEO/CNTs/water solution was lyophilizated, the water is first frozen and then 8 9 sublimated from the frozen state, causing the formation of pores. At the same time, CNTs together with PEO connected and entangled with each other as a network (or 10 scaffold) to give a desirable strength, which effectively suppresses the shrinkage and 11 collapse of aerogel during drying. The solid CNTs tend to aggregate ascribed to the 12 interaction of PEO with CNTs. The micro-sheets at higher magnification is shown in 13 Fig.S4B. It is observed that CNTs coated with a PEO layer closely cohere with each 14 other to form dense and compact network structure which is ascribed to the strong 15 interaction of PEO with CNTs, as well as the PEO molecular chains, being long and 16 flexible, can act a binder role that attached to CNTs. It is worth noting that the PEO can 17 hardly be distinguished in the micro-sheets which demonstrates the homogenous 18 mixing of PEO with CNTs resulting from the solution mixing method. 19

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1 Table S1 The sizes of the samples in the mechanical measurement.

	Plain-structure					Layer-structure				
Sample	1	2	3	4	5	1	2	3	4	5
Width	9.85	9 90	9.85	9.92	9.95	9.95	9.95	9 98	9.84	9.97
(mm)	7.05).)0	7.05).)2).)5).)5).)5	9.90	7.04).)2
Thickness	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15
(mm)	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15

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3 References

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