Tailoring Electromagnetic Interference Shielding Properties in Sandwich Architectures made with Low-Concentration MWCNT-reinforced PDMS

Pavithra Ananthasubramanian^a, Pritom J. Bora^a, Chandana Gadadasu^{b,c}, Praveen C. Ramamurthy^{b,c}, Nagarajan Raghavan^{a†}

^anano-Macro Reliability Laboratory (nMRL), Engineering and Product Development Pillar (EPD), Singapore University of Technology and Design, 8, Somapah Road, 487372, Singapore.

^bOrganic Nano Electronics Laboratory (ONE Lab), Department of Materials Engineering, Indian Institute of Science, Bangalore, 560012, India.

^cInterdisciplinary Centre for Energy Research (ICER), Indian Institute of Science, Bangalore, 560012, India.

† Corresponding Author: <u>nagarajan@sutd.edu.sg</u>

Supplementary Information

S1. Comprehensive measurement of the thickness of the composite samples

A comprehensive physical thickness assessment using a screw gauge across more than 15 different points on the (1+1) LBL samples containing either 1 wt% or 3 wt% MWCNTs is conducted and compared with pristine PDMS samples. The pristine PDMS samples exhibited negligible thickness variation, consistently measuring 200 μ m in total (i.e., ~100 μ m per layer), as corroborated by the cross-sectional FESEM image in Figure 5(c) discussed in the manuscript. In contrast, the (1+1) LBL samples showed measurable variation in overall thickness due to the irregularity introduced by the MWCNT layer. These data points, summarized in the table below, reflect the non-uniform yet physically significant presence of the MWCNT network and its impact on the layered architecture.

Table S1.	Thickness	Measurement	of the	Composite	Samples
-----------	-----------	-------------	--------	-----------	---------

S.No.	Sample ID	Measurement No.	Total Thickness (µm)	PDMS Thickness (µm)	MWCNT layer thickness (μm)	Mean Total Thickness	Mean MWCNT layer
			Α	В	C= A-B	C = A - B (µm)	
1		1	394		194		
2		2	396		196		
3	1 wt% (i)	3	402	200	202	(400±10)	(200±10)
4		4	407	1	207		
5		5	410		210		
S.No.	Sample ID	Measurement	Total	PDMS	MWCNT layer	Mean Total	Mean

		No	Thickness (um)	Thickness (um)	thickness (μm)	Thickness	MWCNT layer
		190.	A	B	C= A-B	(µm)	thickness (µm)
6		1	390		190		
7		2	397		197		
8	1 wt% (ii)	3	405		205		
9		4	406		206		
10		5	398	200	198	(400±10)	(200±10)
11		1	399	200	199		
12		2	400		200		
13	1 wt% (iii)	3	398	-	198		
14		4	401		201		
15		5	397		197		
16		1	701		501		
17		2	693		493		
18	3 wt% (i)	3	709	-	509		
19		4	708		508		
20		5	684		484		
21		1	694		494		
22		2	685		485		
23	3 wt% (ii)	3	711	200	511	(700±10)	(500±10)
24		4	689		489		
25		5	700		500		
26		1	710		510		
27		2	699		499		
28	3 wt% (iii)	3	715		515		
29		4	697		497		
30]	5	705		505		

S2. Variability of total shielding effectiveness (SE_T) data collected from all the composite samples

The variability across samples shown in Figure 6 of the main manuscript is graphically represented in Figure S1 below.



Fig. S1. Range of EMI shielding effectiveness from the triplicate measurements collected from each sample in (a) X-band and (b) Ku-band. The group of data boxed inside blue colored borders is samples with 1 wt% CNT sandwiched between two PDMS layers, and the group of data boxed inside the red-colored border is samples with 3 wt% CNT sandwiched between two PDMS layers. The difference in the performance between the samples from the two different groups is larger than the variability in the performance of the samples within the same group.

As evident from Figure S1. (b), the inter-class variability significantly exceeds the intra-class variability, indicating that the performance variation within samples of the same composition is substantially lower than the variation observed between samples of different compositions. This establishes that the shielding effectiveness of the 3 wt% samples is markedly higher than that of the 1 wt% samples. Consequently, the minor fluctuations in performance within each group do not impact the robustness of the inference that sample composition, rather than sample-to-sample variability, governs the observed differences in shielding effectiveness. Although variability in CNT network density within and across sample groups can impact microstructural features and performance, the compositional parameter—specifically the CNT concentration—emerges as the primary determinant of the observed trends. This study focuses on elucidating the influence of CNT concentration in a layered architecture, while recognizing that network heterogeneity may warrant further dedicated investigation.

S3. How many MWCNTs are there per unit volume in composites with alternating layers of MWCNT and PMDS (LBL PNCs) and in bulk composites with MWCNT dispersed in the bulk of PDMS (Bulk PNCs)?

S3.1 Calculation of the weight of one MWCNT

The volume of a MWCNT can be calculated using its length (L) and diameter (d) by treating it as a hollow cylinder:

Volume = $\pi * (d/2)^2 * L$

For example, if we consider a multi-walled CNT (MWCNT) with a length of 1 μ m (1 μ m = 10⁻⁶ m) and a diameter of 1 nm (1 nm = 10⁻⁹ m), the volume would be:

Volume = $\pi * (1*(10^{-9})/2)^2 * 1 * 10^{-6}$

Volume = $7.85 * 10^{-25} \text{ m}^3$

Calculating the weight from volume and density,

The weight of the CNT can then be calculated by multiplying the volume by the density of the CNT material. The density of multi-walled CNTs (MWCNTs) with a diameter of 1 nm is approximately 1.3 g/cm³.

Converting the volume to cm³ and multiplying by the density:

Weight = Volume * Density

= $(7.85 * 10^{-25} \text{ m}^3) * (1.3 \text{ g/cm}^3) * (106 \text{ cm}^3/\text{m}^3)$

$$= 1.02 * 10^{-18} \text{ g}$$

Therefore, the weight of a single MWCNT with a length of 1 μ m and a diameter of 1 nm would be approximately 1.02 x 10⁻¹⁸ grams.

S3.2 Calculation of the numbers of MWCNT in one-layer LBL/ Bulk PNCs (0.5 wt%)

Let us consider two comparative composites from LBL and Bulk PNCs with 0.5 wt% MWCNT concentration.

Sample Name	Weight of MWCNT/ layer (mg)	Weight of MWCNT/ layer (g)	Weight of one MWCNT (g)	No. of MWCNT/ layer
LBL PNC (0.5 wt%)	1	0.001	1.02E10 ⁻¹⁸	9.8E+14
Bulk PNC (0.5 wt%)	1	0.001	1.02E10 ⁻¹⁸	9.8E+14

S3.3 Calculation of the numbers of MWCNT per unit volume in one layer of LBL vs Bulk PNCs

Sample Name	Surface area of sample (m ²)	Height of MWCNT dispersed region (m)	Volume of MWCNT dispersed region (m ³)	No. of MWCNT/ layer	No. MWCNT/ unit volume
LBL PNC (0.5 wt%)	0.002826	1E-6	2.83E-09	9.80392E+14	3.47E+23
Bulk PNC (0.5 wt%)	0.002826	25E-6	7.07E-08	9.80392E+14	1.39E+22

The number of MWCNT in the first 1 μ m depth in LBL PNC (0.5 wt%) is 25 times more than the same depth in Bulk PNC (0.5 wt%).