Supporting Information for

Multi-interfaced Graphene Aerogel/Polydimethylsiloxane Metacomposites with Tunable Electrical Conductivity for Enhanced Electromagnetic Interference Shielding

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The fabrication process of GA/PPyNRs/PDMS composites is stated as follows. In a typical synthesis, 0.06 M CTAB was dispersed in HCl solution (1 M, 120 mL) at 40 °C, and 0.03 M ammonium persulfate (APS) was added and stirred for 15 minutes until white precipitates were formed. Then, 0.24 M pyrrole monomer was added, when the mixture was continuously stirred at about 0 °C for 2 h until the black precipitates were formed, which were washed with HCl solution (0.5 M) and acetone. The obtained resultant polypyrrole nano rings (PPyNRs) were freeze-dried for 12 hours and ground into powders. In the GO solution (7.5 mg/mL), PPyNRs were added in different mass ratios of 6:1, 4:1, 3:1 and 2:1, simultaneously. Finally, the obtained hybrid graphene aerogels were filled by PDMS through the process of section **2.3** as detailed in the main text, and were named as GA/PPyNRs(6:1)/PDMS-7.5, GA/PPyNRs(4:1)/PDMS-7.5, GA/PPyNRs(3:1)/PDMS-7.5 and GA/PPyNRs(2:1)/PDMS-7.5.

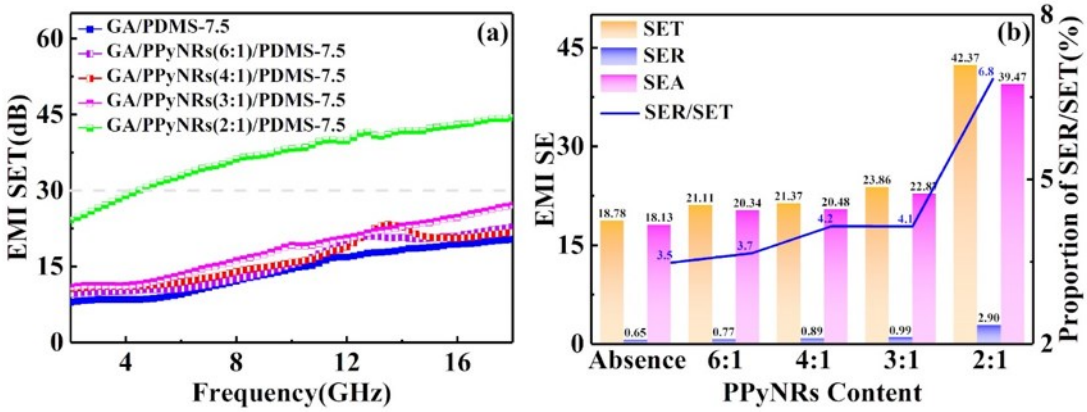

Fig. S1 (a) Frequency dependences of EMI shielding for GA/PDMS-7.5 composites with different PPyNRs contents in 2-18 GHz, (b) the average value of SET, SEA and SER for the GA/PPyNRs/PDMS-7.5 samples with different PPyNRs contents in 12-18 GHz. The blue line represents the proportion of SER/SEA for the GA/PPyNRs/PDMS-7.5 composites with different PPyNRs contents.

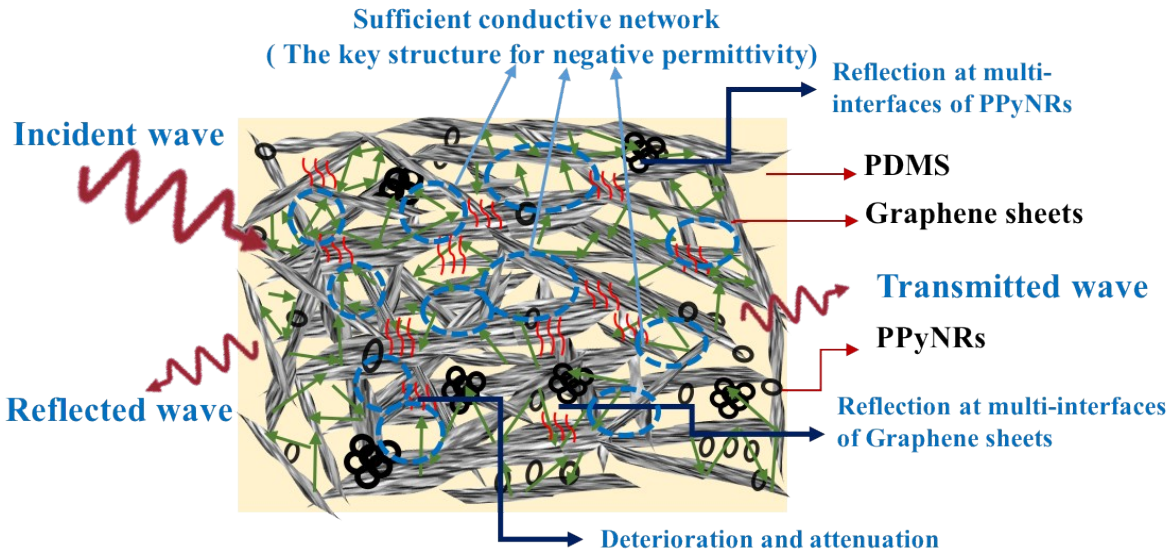

Fig. S2 Scheme of EMI shielding for the GA/PDMS and GA/PPyNRs/PDMS composites.

The electromagnetic interference (EMI) of the GA/PDMS samples with varied PPyNRs contents is shown in **Fig. S2(a)**. The EMI total shielding effectiveness (SET) of GA/PPyNRs/PDMS-7.5 increases continuously as the PPyNRs content increases. The increment of EMI SET is less marked as the mass ratios of graphene to PPyNRs are 6:1, 4:1 and 3:1, Yet, it changes obviously as the mass ratio reaches 2:1. The probable reason is that with the assistance of PPyNRs' bridge, the conductive network becomes more complete at the mass ratio of 2:1 and the ring structure of PPyNRs also provides more interfaces shown in **Fig. S2**, thereby enhancing the microwave deterioration and attenuation. Hence, in **Fig. S1(b)**, the SER/SET ratio of the GA/PPyNRs (2:1)/PDMS-7.5 composite is improved from 3.5% to 6.8% compared to the GA/PDMS-7.5 composites. This improvement may be assigned to the more contact surfaces provided by PPyNRs. However, the EM waves absorption is still very high (the SEA/SET ratios are all over 93.2%) in both kinds of composites.