

Towards Efficient Electromagnetic Interference Shielding Performance for Polyethylene Composites by Structuring Segregated Carbon Black/Graphite Networks*

Cheng-hua Cui, Ding-xiang Yan**, Huan Pang, Li-chuan Jia, Yu Bao
Xin Jiang and Zhong-ming Li

*College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials Engineering,
Sichuan University, Chengdu 610065, China*

Abstract An electromagnetic interference (EMI) shielding composite based on ultrahigh molecular weight polyethylene (UHMWPE) loaded with economical graphite-carbon black (CB) hybrid fillers was prepared *via* a green and facile methodology, *i.e.*, high-speed mechanical mixing combined with hot compression thus avoiding the assistance of the intensive ultrasound dispersion in volatile organic solvents. In this composite, the graphite-CB hybrid fillers were selectively distributed in the interfacial regions of UHMWPE domains resulting a typical segregated structure. Thanks to the specific morphology of segregated conductive networks along with the synergetic effect of large-sized graphite flakes and small-sized CB nanoparticles, a low filler loading of 7.7 vol% (15 wt%) yielded the graphite-CB/UHMWPE composites with a satisfactory electrical conductivity of 33.9 S/m and a superior shielding effectiveness of 40.2 dB, manifesting the comparable value of the pricey large-aspect-ratio carbon nanofillers (*e.g.*, carbon nanotubes and graphene nanosheets) based polymer composites. More interestingly, with the addition of 15 wt% graphite-CB (1/3, *W/W*) hybrid fillers, the tensile strength and elongation at break of the composite reached 25.3 MPa and 126%, respectively; with a remarkable increase of 58.1% and 2420% over the conventional segregated graphite/UHMWPE composites. The mechanical reinforcement could be attributed to the favor of the small-sized CB particles in the polymer molecular diffusion between UHMWPE domains which in turn provided a stronger interfacial adhesion. This work provides a facile, green and affordable strategy to obtain the polymer composites with high electrical conductivity, efficient EMI shielding, and balanced mechanical performance.

Keywords: Graphite; Carbon black; Segregated structure; Electromagnetic interference; Synergistic effect.

INTRODUCTION

Due to the widespread use of transient power sources, the intensive electromagnetic interference (EMI) has been recognized as the hazardous threat that has significant effects on the interference or malfunction of highly sensitive precision electronic instruments (*e.g.*, cell phones, computers, laptops, *etc.*)^[1–3]. Over the past decades, conductive polymer composites (CPCs) comprising of the insulating polymer matrices and conducting fillers have attracted enormous research interests as promising advanced materials for the EMI protection, due to their light weight, resistance to corrosion, versatility, and processability^[3–7]. To satisfy the essential EMI shielding effectiveness (SE) requirement in the commercial applications (20 dB), an electrical conductivity of at least

* This work was financially supported by the National Natural Science Foundation of China (Nos. 51421061, 51120135002 and 51473102), the Innovation Team Program of Science and Technology Department of Sichuan Province (No. 2014TD0002), and the China Postdoctoral Science Foundation (Nos. 2015M572474 and 2016T90848).

** Corresponding author: Ding-xiang Yan (鄢定祥), E-mail: yandingxiang@scu.edu.cn

Received June 16, 2016; Revised July 21, 2016; Accepted July 25, 2016

doi: 10.1007/s10118-016-1849-6

1 S/m is typically required, which can only be achieved in CPCs with well-developed conductive interconnected networks at relatively high filler loadings^[8]. Nevertheless, high concentration of conductive fillers usually gives rise to the inferior mechanical performance, complex processing, and high cost. Efforts have been made to achieve the practical application requirements of CPC based EMI shielding materials (*i.e.*, high performance balanced with low product cost). Three frequently used methods are introduced: (i) the improvement of the filler dispersion in polymer matrices by ultrasound-assistant solution dispersion or surface modification techniques^[9, 10]; (ii) structuring perfect conductive networks to improve the efficiency of conductive fillers in constructing conductive pathways^[11–14]; (iii) the cooperative effect of hybrid fillers with different size and geometric dimensionality on facilitating the formation of conductive networks^[15–17].

Achieving a highly efficient shielding performance by means of improving filler dispersion in the entire polymer matrix at relatively low filler loading is extremely difficult, even for large-aspect-ratio conductive fillers (*e.g.*, carbon nanotubes (CNTs) and graphene), that could be attributed to the high degree of agglomeration. For example, Al-Saleh and Sundararaj reported an EMI SE of only 24 dB for 5 vol% CNT/polypropylene (PP) composite^[5]. Liang *et al.* fabricated an EMI shielding graphene/epoxy composite with a relatively low EMI SE of 21 dB even at 8.8 vol% graphene loading^[18]. Alternatively, graphite flakes, as naturally abundant and economical natural materials which own the multilayered structure of carbon atoms and also possess high electrical conductivity of 10^3 S/cm^[19]. Owing to the acceptable comprehensive performance accompanied by the economical affordability, graphite flakes have attracted enormous research interests as reinforcing fillers for EMI shielding materials^[20–26]. For example, Sawai and Banerjee fabricated the graphite/PP and graphite/poly(ether imide) composites with the EMI SE of ~19 and 40 dB at 60 wt% graphite loading^[26]. The addition of such high graphite loadings usually sacrifices the process ability and mechanical properties (*e.g.*, toughness and ductility) of graphite based CPCs. For instance, Zhao and Ye found that the ultimate strain of 10 wt% graphite/POM CPCs was only 3.4%^[27]. Krupa and Chodak loaded 30% graphite into high density polyethylene, causing three orders of magnitude drop in ultimate strain compared with neat HDPE^[28].

Structuring segregated networks in CPCs is regarded as one of the most effective approaches for constructing well-developed conductive networks at ultralow filler loadings^[8, 9, 29, 30]. In this specific structure, conductive fillers are predominately located in the interfacial regions of polymer domains, that largely enhances the efficiency of conductive fillers on formalizing continuous conductive channels throughout CPCs. Sachdev *et al.* formalized a segregated graphite conductive network in poly(vinyl chloride) (PVC) matrix to achieve an EMI SE value of ~25 dB at 15 wt% graphite content at the frequency of 8.54 GHz^[31]. In our previous work, segregated graphite/ultrahigh molecular weight polyethylene (UHMWPE) composites have been fabricated, revealing a superior EMI SE^[32]. However, constrained by the maximum filler loading (usually less than 10 vol%) of composites with segregated structure and low-aspect-ratio of graphite flakes^[30], the EMI SE of graphite loaded segregated CPCs (sCPCs) is difficult to realize a further enhancement.

The synergistic effect of two dimensional (2D) graphite flakes/zero dimensional (0D) carbon black (CB) is another successful methodology to construct efficient conductive networks at relatively low filler loadings^[16, 33, 34]. The small-sized CB nanoparticles occupy the interspaces between the adjacent large-sized graphite flakes, enhancing the transport behaviors of conductive channels. After the incorporation of 25 wt% CB and 55 wt% graphite into the PP, the electrical conductivity (3.5×10^3 S/m) of CB/graphite/PP composite exhibited more than five times larger than that of graphite/PP one^[31]. Furthermore, compared to the large-sized 2D graphite flakes, the small-sized CB nanoparticles based conductive networks are beneficial to the mechanical reinforcement of polymer composites. Chen *et al.* incorporated 15 phr CB into PVC matrix by melt blending and as a result, the tensile strength and modulus of the composite increased from 40 and 950 MPa to 46 and 1180 MPa, respectively^[35]. Likewise, the tensile strength and modulus of high density polyethylene were also increased by about 24% and 65% with the incorporation of 28 wt% CB^[36].

Therefore, in order to further improve the EMI shielding performance of CPCs without sacrificing the mechanical properties, the combination of the specific segregated structure and the synergistic effect of hybrid conductive fillers were utilized to fabricate the highly efficient EMI shielding materials in this work. We chose

the ultrahigh melt-viscosity UHMWPE as the polymer matrix scaffold whereas large-sized graphite flakes and small-sized CB were selected as the hybrid conductive fillers to formalize the segregated conductive networks. The 15 wt% graphite-CB/UHMWPE sCPCs exhibited the desirable electrical conductivity of 33.9 S/m and EMI SE of 40.2 dB, superior to that of graphite/UHMWPE (10.2 S/m and 29.0 dB) or CB/UHMWPE sCPCs (14.1 S/m and 33.3 dB) for the same filler loading. Additionally, it is noteworthy that the mechanical properties of the graphite-CB/UHMWPE sCPCs were also dramatically improved compared with graphite/UHMWPE sCPCs. By the combination of segregated structures and synergistic effect of hybrid fillers, this work provided a green and facile method to fabricate the low-cost graphite-CB/UHMWPE sCPCs with the satisfactory electrical properties, EMI shielding performance, and balanced mechanical properties.

EXPERIMENTAL

Materials

Graphite platelets with a density of 2.2 g/cm³ and lateral dimension of 20 μm, were supplied by Beishu Graphite Co. Shangdong, China. CB (VXC-605) was purchased from Cabot Co. Ltd, Shanghai, China, with the average particle size of 25 nm. UHMWPE powders, with the density of 0.94 g/cm³ and resistivity of 10¹⁷ Ω·cm, were provided by Beijing No.2 Auxiliary Agent Factory, Beijing, China.

Fabrication of sCPCs

The fabrication of graphite-CB/UHMWPE sCPCs includes two steps, as shown schematically in Fig. 1. Firstly, the quantified graphite, CB and UHMWPE were mechanically mixed for 4 min at an ultrahigh speed of 24000 r/min to realize the decoration of hybrid fillers on the surfaces of UHMWPE particles. Then the obtained graphite-CB/UHMWPE particles were compressed at 200 °C at a pressure of 10 MPa for 5 min after preheating for 5 min. The graphite₃-CB₁/UHMWPE, graphite₁-CB₁/UHMWPE and graphite₁-CB₃/UHMWPE sCPCs (the weight ratios of graphite and CB were 3/1, 1/1 and 1/3, respectively) with varying filler loadings from 0.1 wt% to 15 wt% were fabricated. As the control sample, the single graphite or CB loaded sCPCs were also prepared under the same processing conditions.

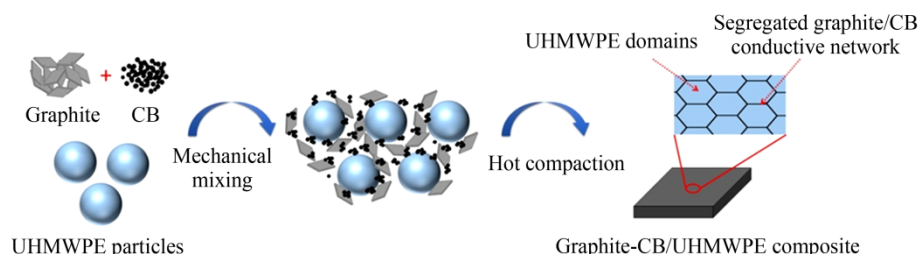


Fig. 1 Schematic representation for the fabrication of segregated graphite-CB/UHMWPE composites

Characterizations

To clarify the morphology and the segregated conductive networks of sCPCs, the films with the thickness of 20 μm cut at room temperature with an ultramicrotome (Leica EM UC6, Germany) were observed using the optical microscope (OM). Scanning electron microscope (SEM, Inspect F, FEI, Finland) was employed to observe the distribution of conductive fillers in the composites with an accelerated voltage of 20 kV. The samples were rapidly fractured after immersing in liquid nitrogen for 30 min and the fractured surfaces were sputter-coated with gold before the SEM observation. When the hybrid filler content was below 4 wt%, the electrical conductivity was measured adopting a two-point method using a Keithley electrometer Model 4200-SCS. The samples were cut into rectangular sheets with silver paste coated on the sides of the sheets to eliminate the contact resistance. The electrical conductivity of CPCs (2.1 mm sample thickness) with 8 wt% and 15 wt% filler loadings were conducted using a four-probe method by a RST-9 resistivity measurement system. The EMI SE of sCPCs was measured in the X-band (8.2–12.4 GHz) using an Agilent N5230 vector network analyser.

connected with a coaxial test cell (APC-7 connector). The thickness of specimens for EMI shielding measurement was 2.1 mm. The tensile testing was carried out at room temperature using Model 5576, Instron Instruments, America, with a drawing rate of 50 mm/min and a gauge length of 20 mm.

RESULTS AND DISCUSSION

The OM images exhibited the morphological evolution of the conductive networks along with increasing hybrid filler loadings (see Fig. 2). It can be clearly seen that hybrid fillers were selectively distributed at the interfacial regions of UHMWPE domains, forming a typical segregated structure. When the filler loading was just 0.1 wt%, the conductive fillers could not afford the continuous conducting pathways (Figs. 2a, 2d and 2g). For 0.5 wt% hybrids loaded ones (Figs. 2b, 2e and 2h), the thickness of conductive pathways increased, resulting in the formation of the well-developed conductive networks. Increasing filler loading up to 2 wt%, the well-defined conductive pathways could be formalized. In addition, the graphite₃-CB₁ sCPCs exhibited much looser conductive pathways than the graphite₁-CB₃ ones (Figs. 2c and 2i), because of the weak interfacial interaction between UHMWPE domains.

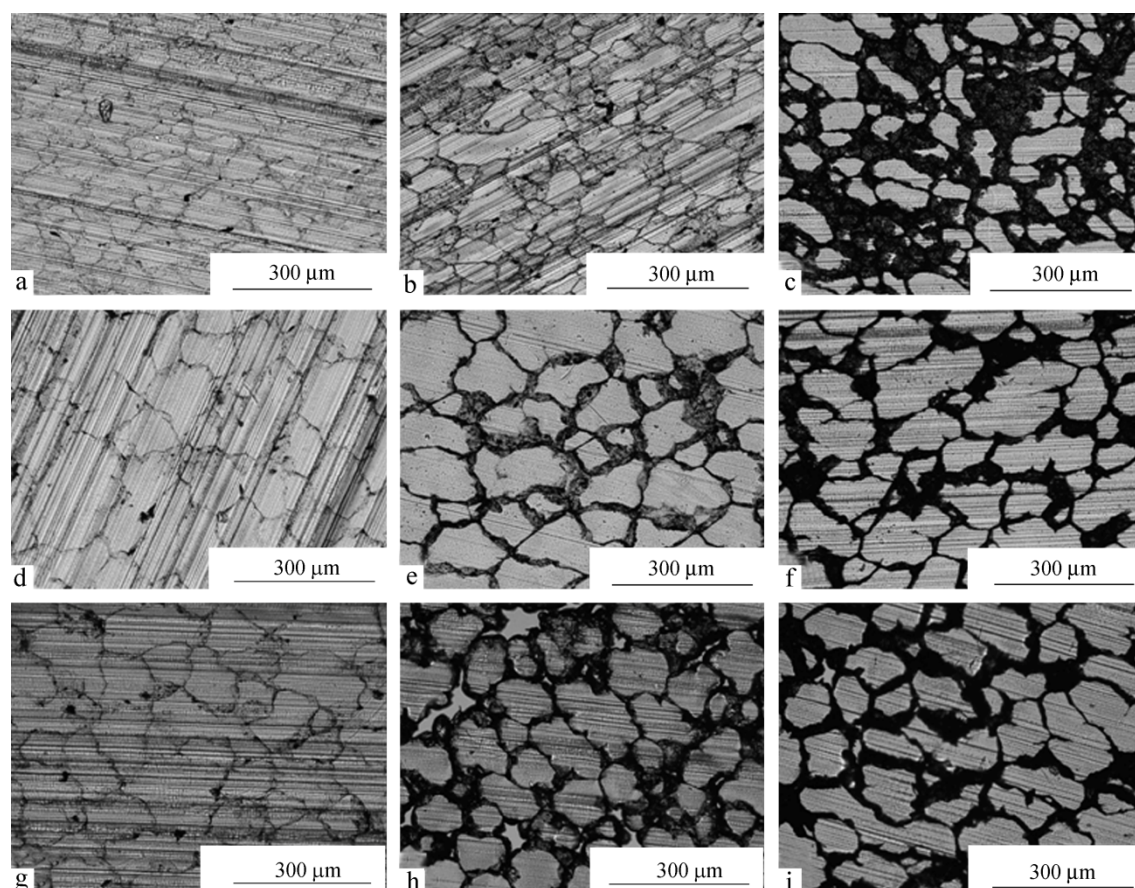


Fig. 2 OM images of graphite₃-CB₁/UHMWPE (a) (b) (c), graphite₁-CB₁/UHMWPE (d) (e) (f), and graphite₁-CB₃/UHMWPE sCPCs (g) (h) (i), with different filler content of 0.1 wt% (a) (d) (g), 0.5 wt% (b) (e) (h), and 2 wt% (c) (f) (i)

OM images provided a powerful and direct proof of the segregated conductive network formation throughout the entire UHMWPE matrix, but the dispersion state of graphite-CB hybrid fillers still remained inexplicit. Then SEM characterization was employed to observe the detailed morphology of the graphite-CB

based segregated structure. Figure 3 displays the surface of the graphite-CB hybrid fillers decorated UHMWPE particles, in which numerous graphite and CB particles overlapped each other to construct the continuous conductive channels. The graphite-CB conducting pathways were well preserved in the subsequent compression moulding, because of the weak shear during the melt process, the ultrahigh melt viscosity and melt strength of UHMWPE. This specific morphology provided a well prerequisite for the construction of a segregated graphite-CB conductive network. As shown in Fig. 4, the UHMWPE domains merely occurred slight plastic deformation transforming into irregular polyhedrons during the hot compression to effectively prevent hybrid fillers from penetrating into the interior of polymer domains^[17]. With the UHMWPE crystallization during the cooling process, graphite-CB decorating UHMWPE particles were preserved. Finally, segregated conductive networks of graphite-CB hybrid fillers were formalized in the interfacial regions of UHMWPE domains. Additionally, in comparison with the morphology of graphite₃-CB₁/UHMWPE, the graphite₁-CB₃/UHMWPE sCPCs experienced much larger deformation during the hot compaction process, resulting in the stretched UHMWPE polyhedrons (see Figs. 4a and 4g). This phenomenon could be explained by the barrier effect of the large-sized graphite flakes, which dramatically limited the intensity of shear flow and the deformation of the molten UHMWPE domains. Owing to the small-sized 0D CB agglomerates, UHMWPE chains more easily diffused into the CB dominated conductive pathways, as a result of the relatively intensive heat-sealing between UHMWPE granules. Observed from the magnified SEM images (Figs. 4c, 4f and 4i), the small-sized CB particles bridged the large-sized adjacent graphite flakes, indicating the synergistic effect of 0D CB agglomerates and 2D graphite flakes on the formation of continuous conducting channels.

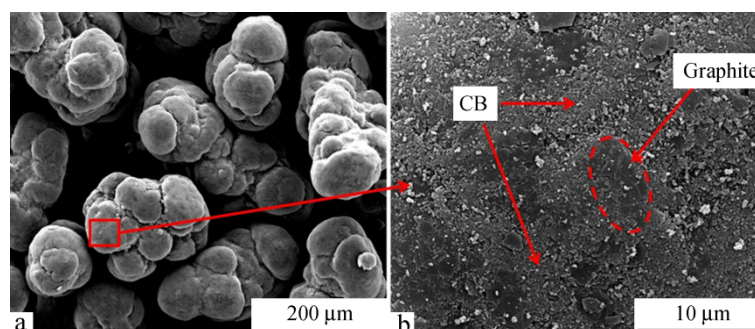


Fig. 3 SEM images of segregated graphite₁-CB₁/UHMWPE complex particles with 2 wt% graphite-CB loading (a); the graphite-CB fillers decorated on the UHMWPE granule surface (b)

Figure 5 depicts the electrical conductivity of sCPCs with varying graphite-CB hybrid filler content. The sCPCs exhibited quite low electrical conductivity below 10^{-9} S/m when the loadings of hybrid fillers were below 0.3 wt%, illustrating the incomplete formation of conductive networks throughout the insulating UHMWPE matrix. With further increase in the hybrid filler concentration to 0.5 wt%, the sCPC electrical conductivity dramatically raised, indicative of the appearance of percolation behaviors, *i.e.*, transformation from semiconductors to conductors. The percolation threshold can be obtained according to the classical percolation theory: $\sigma = \sigma_0 (\varphi - \varphi_c)^t$, where σ represents the sCPC electrical conductivity, σ_0 is a constant concerned with the intrinsic conductivity of conductive fillers, φ indicates the volume fraction of conducting fillers, φ_c is the volume percolation threshold and t suggests the critical exponent related to the conductive network dimensionality. Table 1 provides the percolation thresholds of sCPCs with the different rate of graphite and CB hybrid fillers. The graphite/UHMWPE sCPCs achieved the lowest φ_c (about 0.12 vol%); while the CB/UHMWPE ones exhibited a little higher value φ_c of about 0.16 vol%. These results indicate that the plane-to-plane contact in 2D graphite flakes facilitated the formation of the segregated percolating pathways compared to the point-to-point contact mechanism of 0D CB agglomerates^[37]. As for the hybrid filler systems, they seemed to be difficult to form the initial conducting pathway at relatively low filler loadings, showing higher percolation thresholds. Referring to the electrical conductivity of sCPCs at 15 wt% filler loading, evident synergistic effect was found:

graphite₁-CB₁/UHMWPE sCPC showed the maximum electrical conductivity of 33.9 S/m, corresponding to nearly 232% and 140% increase over that of graphite/UHMWPE (10.2 S/m) and CB/UHMWPE (14.1 S/m), respectively. The t values of all systems were in the range of 2.08–2.78, revealing the formation of 3D conductive networks. This attractive phenomenon could be ascribed to the cooperative effect of the large-sized 2D graphite/small-sized 0D CB hybrid fillers on the formation of segregated conductive networks. As shown in the schematic illustrating the graphite-CB hybrid conductive networks (Fig. 6), the polymer molecules hardly permeated into the interspace between the adjacent graphite flakes, forming the micro-voids among the

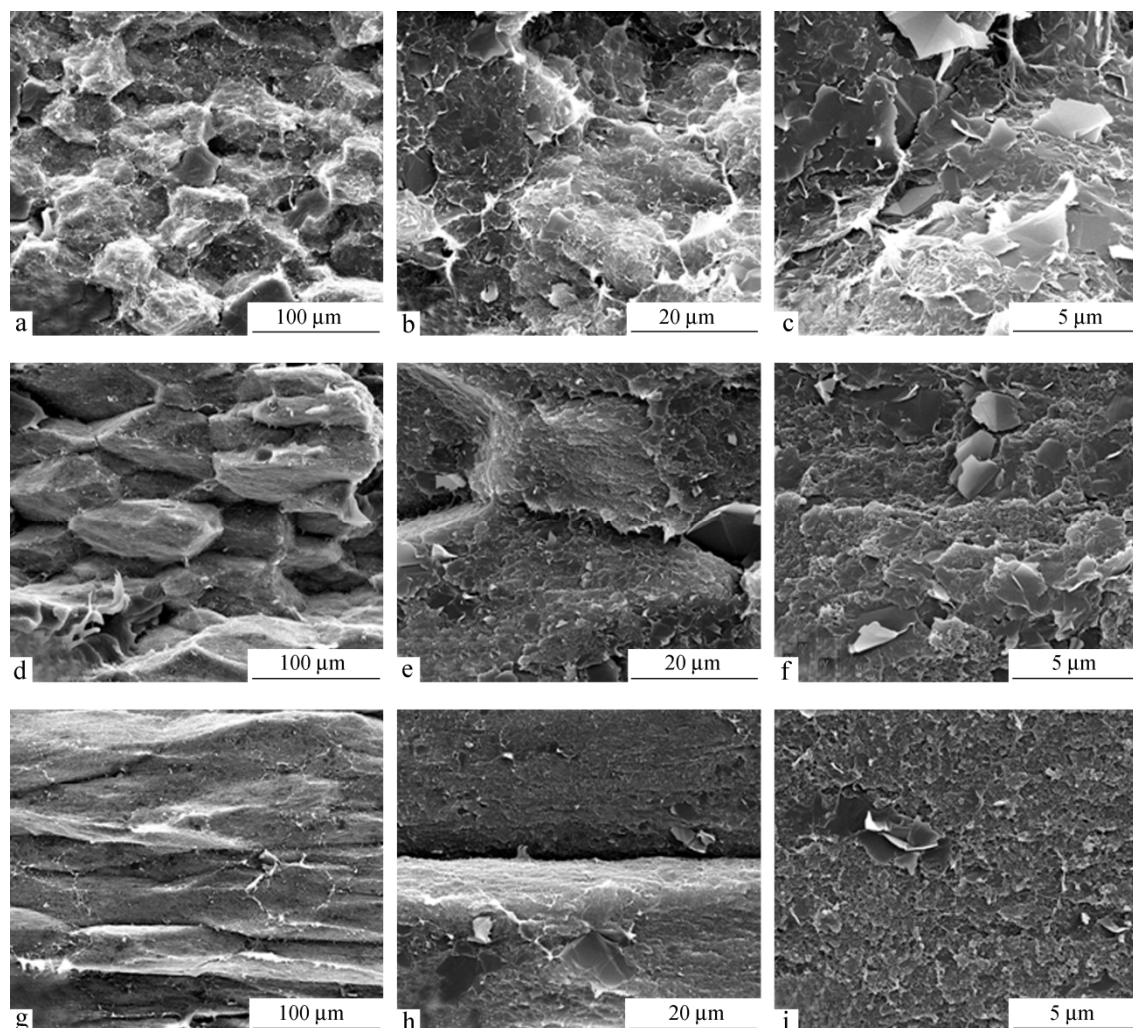


Fig. 4 SEM images of segregated graphite₃-CB₁/UHMWPE (a) (b) (c), graphite₁-CB₁/UHMWPE (d) (e) (f), and graphite₁-CB₃/UHMWPE CPCs (g) (h) (i), with same filler content of 2 wt%

Table 1. Percolation thresholds and electrical conductivities of segregated composites at 15 wt% filler content

Conductive filler	Percolation threshold (vol%)	Electrical conductivity (S/m)	t
Graphite	0.12	10.2	2.58
Graphite ₃ -CB ₁	0.63	20.8	2.55
Graphite ₁ -CB ₁	0.24	33.9	2.78
Graphite ₁ -CB ₃	0.23	18.9	2.08
CB	0.16	14.1	2.30

segregated conducting pathways. With the addition of small-sized CB nanoparticles, they occupied the microvoids between the large-sized graphite flakes, increasing the density of conductive pathways and decreasing the contact resistance among graphite flakes.

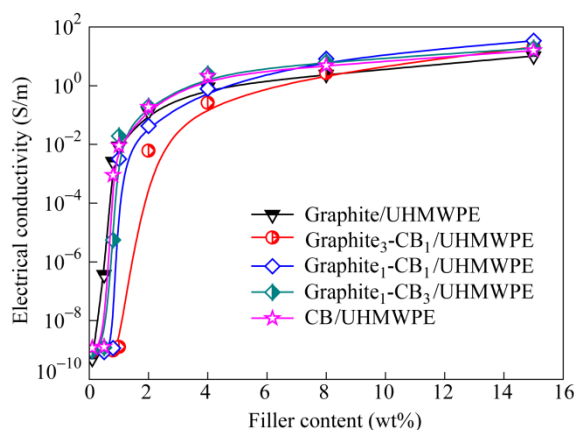


Fig. 5 Electrical conductivities of segregated graphite-CB/UHMWPE, graphite/UHMWPE and CB/UHMWPE composites as a function of filler loadings

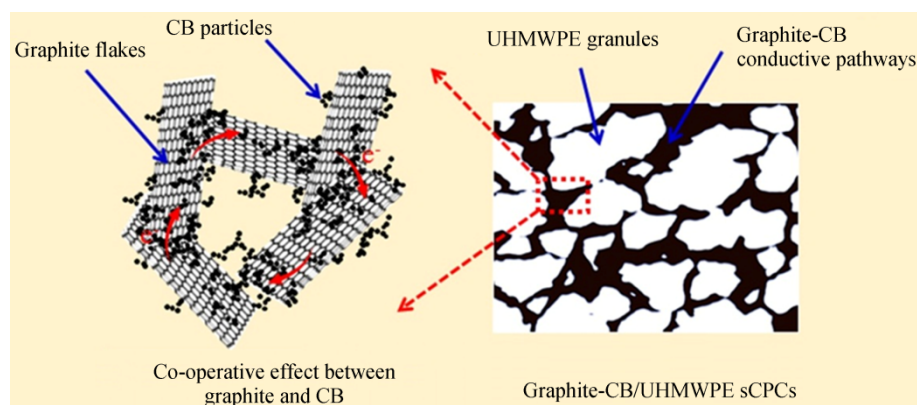


Fig. 6 Schematic representation of graphite-CB hybrid conductive networks

In this section, the EMI shielding performance of graphite/UHMWPE, CB/UHMWPE and graphite-CB/UHMWPE sCPCs in X-band at 15 wt% filler loading was investigated, as shown in Fig. 7. Thanks to the synergistic effect of graphite and CB on the formation of conductive networks, the graphite-CB/UHMWPE sCPCs manifested the higher EMI SE value (36.5–40.2 dB) than that (29.0–33.3 dB) of the single filler ones. Further, the graphite₁-CB₁/UHMWPE sCPC exhibited the optimal shielding performance (40.2 dB), comparable to or even better than that of large-aspect-ratio CNT or graphene loaded composites. For example, Al-Saleh and Sundararaj reported an EMI SE of 34.8 dB for 7.5 vol% CNT/PP composites^[5]. Arjmand *et al.* prepared the CNT/polystyrene composites through injection molding, illustrating an EMI SE of about 45 dB at 20 wt% CNT loading^[38]. Liang *et al.* incorporated the graphene into epoxy resin which resulted in an EMI SE of only 21 dB at 8.8 vol% filler loading^[18]. The emerged synergistic effect on EMI shielding performance originated from two features of graphite-CB hybrid based segregated conductive networks: i) the highly conductive graphite-CB hybrid conductive networks displayed the superior charge storage capacity to the single graphite or CB networks, in favour of absorbing more incident electromagnetic microwave energy by the polarization of the electric field^[39]; ii) the specific morphology-the small-sized CB particles bridged the large-sized graphite flakes, resulting in the larger polarized interfacial interaction area^[40].

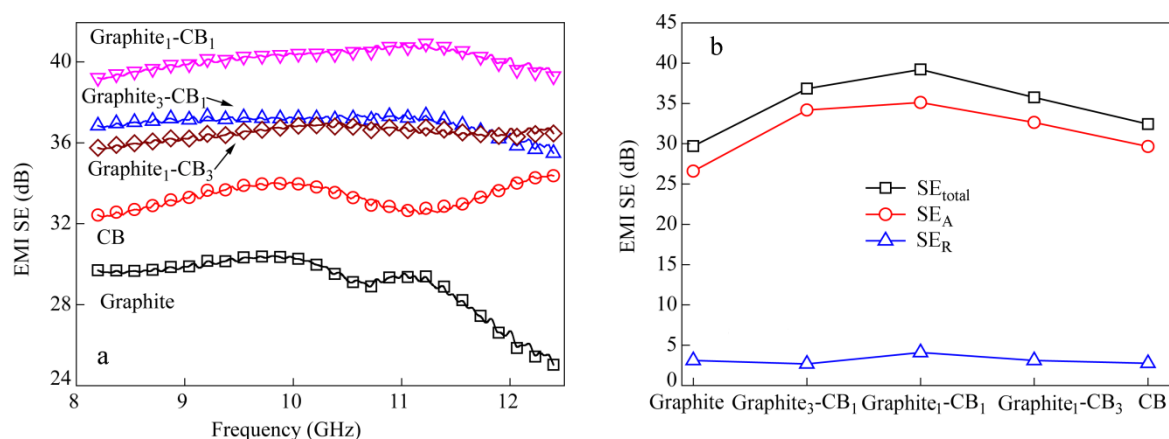


Fig. 7 EMI SE in the X band for composites at 15 wt% filler loadings (a); The total EMI SE (SE_{total}), microwave absorption (SE_A) and microwave reflection (SE_R) at the frequency of 8.2 GHz for segregated CPCs with various rates of graphite and CB (b)

To clarify the EMI shielding mechanism in graphite-CB/UHMWPE sCPCs, the total EMI SE (SE_{total}), the microwave absorption (SE_A) and microwave reflection (SE_R) of sCPCs as a function of graphite-CB hybrid fillers at the frequency of 8.2 GHz were plotted in Fig. 7(b). It is apparent that SE_{total} and SE_A presented a similar variation tendency with the varying graphite-CB ratio, while the SE_R of sCPCs maintained a relatively low level (less than 4.1 dB), which could be negligible on the basis of its contribution to SE_{total} . For example, the SE_{total} , SE_A and SE_R of graphite₁-CB₁ loaded sCPCs were 39.2, 35.1 and 4.1 dB, respectively, which suggested that the contribution of absorption to SE_{total} was 89.5%, indicative of the absorption dominant shielding mechanism. To better understand the absorption dominant shielding mechanism, the conductive layers comprised of the closely packed hybrid conductive fillers could be considered as the highly conducting “hole-walls”, while the UHMWPE domains were regarded as the insulating “holes”, which were almost transparent to microwaves. Hence, the conductive network morphology of sCPCs could be equivalent to the specific “foam” structure. The incident electromagnetic microwaves were gradually dissipated and attenuated in the form of heat by interaction (*i.e.*, reflection, scattering, and absorption) with the highly conductive and interconnected “hole-walls” in the unique “foam” structure.

Apart from the excellent EMI SE, mechanical properties played an important role in the practical applications of shielding materials as well. It is known that the mechanical performance is predominately influenced by the interfacial adhesion of the adjacent polymer domains in sCPC materials. Figure 8 shows the representative engineering strain-stress curves for graphite/UHMWPE, graphite-CB/UHMWPE and CB/UHMWPE samples at 15 wt% filler loading. The graphite/UHMWPE sCPC presented a brittle failure in virtue of the very low elongation at break of only 5.0%, and the tensile strength was as low as 16.0 MPa. Such poor tensile properties of graphite/UHMWPE sCPCs were originated from the weak inter-diffusion and heat-sealing between the adjacent UHMWPE granules caused by the blocking effect of 2D large-sized graphite flakes. With gradually replacing graphite with small-sized CB nanoparticles, molten polymer chains gradually diffused into the hybrid conductive layers giving rise to a tremendous increase in the elongation at break, indicating the transformation from brittle to ductile failure in sCPCs. For instance, the ultimate strain of graphite₁-CB₃/UHMWPE sCPCs achieved 126%, almost 25 times higher than that of graphite/UHMWPE ones (Although this value was lower than that of neat UHMWPE sample, it is sufficient for practical applications). Likewise, the tensile strength of graphite-CB/UHMWPE sCPCs also showed the evident enhancement in comparison with graphite/UHMWPE composite, especially for graphite₁-CB₃/UHMWPE system, the value of tensile strength increased up to 25.3 MPa, corresponding to 58.1% increase. Therefore, the incorporation of small-sized CB particles effectively enhanced the mechanical properties of graphite/UHMWPE sCPCs. The

mechanical reinforcement can be further understood by the stronger interfacial adhesion of UHMWPE domains after the partial replacement of 2D large-sized graphite flakes by small-sized CB nanoparticles. Moreover, SEM images in Fig. 4 also testified that the CB dominated segregated conductive networks experienced the obvious ductile deformation, confirming the intensive interfacial adhesion between the adjacent UHMWPE domains.

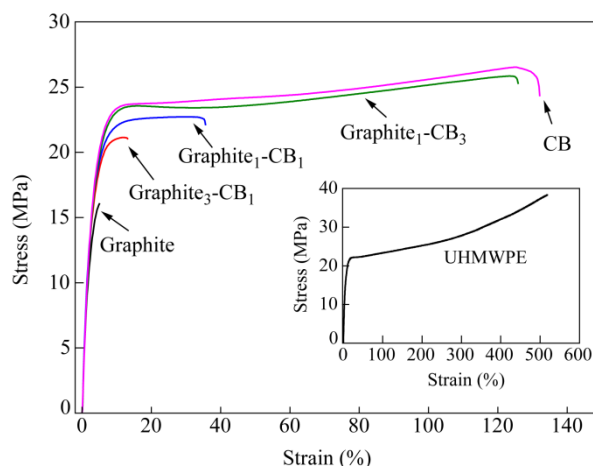


Fig. 8 Comparison of strain-stress curves for pure UHMWPE, segregated graphite/UHMWPE, graphite-CB/UHMWPE and CB/UHMWPE composites at 15 wt% filler loading

CONCLUSIONS

By employing the small-sized CB particles to replace the large-sized graphite flakes, we utilized a green and facile methodology to fabricate graphite-CB/UHMWPE sCPCs with the desirable electrical conductivity and EMI SE of 33.9 S/m and 40.2 dB, respectively, at a fairly low filler concentration of 7.7 vol% (15 wt%). This impressive performance primarily appraised from the synergistic effect of 2D graphite flakes and 0D CB agglomerates on the formation of conductive pathways and the selective distribution of hybrid fillers at the interfaces among UHMWPE domains. In addition, the tensile strength and elongation at break of graphite₁-CB₃/UHMWPE systems were remarkably improved compared with those of graphite/UHMWPE system, increased by 58.1% and 2420%, respectively. Our technology can pave the way to the preparation of environmentally friendly, low-cost, and high performance (*i.e.*, high electrical conductivity, EMI SE, and balanced mechanical performance, *etc.*) polymer composite materials.

REFERENCES

- 1 Chen, Z.P., Xu, C., Ma, C.Q., Ren, W.C. and Cheng, H.M., *Adv. Mater.*, 2013, 25: 1296
- 2 Shen, B., Zhai, W.T. and Zheng, W.G., *Adv. Funct. Mater.*, 2014, 24: 4542
- 3 Al-Saleh, M.H., Saadeh, W.H. and Sundararaj, U., *Carbon*, 2013, 60: 146
- 4 Yan, D.X., Ren, P.G., Pang, H., Fu, Q., Yang, M.B. and Li, Z.M., *J. Mater. Chem.*, 2012, 22: 18772
- 5 Al-Saleh, M.H. and Sundararaj, U., *Carbon*, 2009, 47: 1738
- 6 Zeng, Z.H., Jin, H., Chen, M.G., Li, W.W., Zhou, L.C. and Zhang, Z., *Adv. Funct. Mater.*, 2016, 26: 303
- 7 Zhang, Y., Fang, X.X. and Wen, B.Y., *Chinese J. Polym. Sci.*, 2015, 33: 899
- 8 Yan, D.X., Pang, H., Li, B., Vajtai, R., Xu, L., Ren, P.G., Wang, J.H. and Li, Z.M., *Adv. Funct. Mater.*, 2015, 25: 559
- 9 Pang, H., Piao, Y.Y., Cui, C.H., Bao, Y., Lei, J., Yuan, G.P. and Zhang, C.L., *J. Polym. Res.*, 2013, 20: 304
- 10 Hsiao, S.T., Ma, C.C., Liao, W.H., Wang, Y.S., Li, S.M., Huang, Y.C., Yang, R.B. and Liang, W.F., *ACS Appl. Mater. Interfaces*, 2014, 6: 10667

- 11 Rohini, R. and Bose, S., *ACS Appl. Mater. Interfaces*, 2014, 6: 11302
- 12 Zhang, S.M., Deng, H., Zhang, Q. and Fu, Q., *ACS Appl. Mater. Interfaces*, 2014, 6: 6835
- 13 Lin, C.Y., Zuo, M., Li, H.H., Liu, T. and Zheng, Q., *Chinese J. Polym. Sci.*, 2015, 33: 1162
- 14 Wang, H.S., Zhang, X.H., Zhu, Y.L., Song, Z.H. and Qiao, J.L., *Chinese J. Polym. Sci.*, 2012, 30: 138
- 15 Zhang, S.M., Lin, L., Deng, H., Gao, X., Bilotti, E., Peijs, T., Zhang, Q. and Fu, Q., *Express Polym. Lett.*, 2011, 6:159
- 16 Wei, T., Song, L.P., Zheng, C., Wang, K., Yan, J., Shao, B. and Fan, Z.J., *Mater. Lett.*, 2010, 64: 2376
- 17 Cui, C.H., Pang, H., Yan, D.X., Jia, L.C., Jiang, X., Lei, J. and Li, Z.M., *RSC Adv.*, 2015, 5: 61318
- 18 Liang, J.J., Wang, Y., Huang, Y., Ma, Y.F., Liu, Z.F., Cai, J.M., Zhang, C.D., Gao, H.J. and Chen, Y.S., *Carbon*, 2009, 47: 922
- 19 Li, B. and Zhong, W.H., *J. Mater. Sci.*, 2011, 46: 5595
- 20 Heo, S.I., Yun, J.C., Oh, K.S. and Han, K.S., *Adv. Compos. Mater.*, 2006, 15: 115
- 21 Cho, E.A., Jeon, U.S., Ha, H.Y., Hong, S.A. and Oh, I.H., *J. Power Sources*, 2004, 125: 178
- 22 Kuan, H.C., Ma, C.C.M., Chen, K.H. and Chen, S.M., *J. Power Sources*, 2004, 134: 7
- 23 Sachdev, V.K., Patel, K., Bhattacharya, S. and Tandon, R.P., *J. Appl. Polym. Sci.*, 2011, 120: 1100
- 24 Panwar, V., Mehra, R.M., Park, J.O. and Park, S.h., *J. Appl. Polym. Sci.*, 2012, 125: E610
- 25 Dhakate, S.R., Sharma, S., Chauhan, N., Seth, R.K. and Mathur, R.B., *Int. J. Hydrogen Energ.*, 2010, 35: 4195
- 26 Sawai, P. and Banerjee, S., *J. Appl. Polym. Sci.*, 2008, 109: 2054
- 27 Zhao, X.W. and Ye, L., *J. Appl. Polym. Sci.*, 2009, 111: 759
- 28 Krupa, I. and Chodák, I., *Eur. Polym. J.*, 2001, 37: 2159
- 29 Yan, D.X., Pang, H., Xu, L., Bao, Y., Ren, P.G., Lei, J. and Li, Z.M., *Nanotechnology*, 2014, 25: 145705
- 30 Pang, H., Xu, L., Yan, D.X. and Li, Z.M., *Prog. Polym. Sci.*, 2014, 39: 1908
- 31 Sachdev, V., Srivastava, N., Kumar, K. and Mehra, R., *Mater. Sci-Poland*, 2005, 23: 269
- 32 Jiang, X., Yan, D.X., Bao, Y., Pang, H., Ji, X. and Li, Z.M., *RSC Adv.*, 2015, 5: 22587
- 33 Song, J.B., Zhang, W.B., Yang, W.B., Xu, J.F. and Lai, J.J., *J. Polym. Res.*, 2014, 21: 556
- 34 Dweiri, R. and Sahari, J., *J. Power Sources*, 2007, 171: 424
- 35 Chen, C.H., Li, H.C., Teng, C.C. and Yang, C.H., *J. Appl. Polym. Sci.*, 2006, 99: 2167
- 36 Wang, L.W., Hong, J.B. and Chen, G.H., *Polym. Eng. Sci.*, 2010, 50: 2176
- 37 Song, W.L., Wang, J., Fan, L.Z., Li, Y., Wang, C.Y. and Cao, M.S., *ACS Appl. Mater. Interfaces*, 2014, 6: 10516
- 38 Arjmand, M., Apperley, T., Okoniewski, M. and Sundararaj, U., *Carbon*, 2012, 50: 5126
- 39 Yousefi, N., Sun, X.Y., Lin, X.Y., Shen, X., Jia, J.J., Zhang, B., Tang, B.Z., Chan, M.S. and Kim, J.K., *Adv. Mater.*, 2014, 26: 5480
- 40 Shen, B., Zhai, W.T., Tao, M.M., Ling, J.Q. and Zheng, W.G., *ACS Appl. Mater. Interfaces*, 2013, 5: 11383