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# Influence of the filler dimensionality on the electrical, mechanical and electromagnetic shielding properties of isoprene rubber-based flexible conductive composites

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# ABSTRACT

Rubber-based conductive polymer composites are deformable and flexible, which have been demonstrated tremendous applications in electromagnetic interference (EMI) shielding materials. Here, we filled different dimensional carbonaceous fillers, including zero-dimensional carbon black (0D CB), one-dimensional carbon nanotubes (1D CNTs), two-dimensional graphene (2D GP), and their combinations, into isoprene rubber (IR) to fabricate the flexible EMI shielding composites. Both the electrical, mechanical, and EMI shielding properties were investigated. It was found that the EMI shielding properties of IR-based composites mainly dominated by the microwave absorption consumption. Since 1D CNTs possess a highest EMI shielding effectiveness for absorption, IR/CNTs composites exhibit the best electromagnetic interference shielding properties compared to other carbonaceous nanoparticles filled composites. Moreover, partially replacing CNTs with CB particles to fabricate hybrid IR/CB/CNTs composite is an effective route to improve the electrical properties and lower the cost of the composites. However, it is found that hybrid IR/CB/CNTs composite has a lower EMI shielding effectiveness than IR/CNTs composites, indicating that EMI shielding properties depend on a lot more than just the electrical conductivity of the resulting composites. Nevertheless, based on comprehensive consideration of cost, electrical, mechanical and EMI shielding properties of the composites, it is still an effective to design the high-performance EMI shielding materials via the combination of different dimensional fillers.

**Keywords**: Electromagnetic interference shielding; Conductive polymer composites; Carbonaceous conductive particles; Conductive filler dimensionality

# 1. Introduction

In recent decades, the explosive growths of electronic devices in industrial, household, military and telecommunication bring us great convenience, while also bring the electromagnetic radiations. These electromagnetic radiations generated by various electronic devices not only interfere with the normal functioning of electronic devices but also have harmful effects on human beings, which are regarded as a new-type of pollution, i.e. electromagnetic pollution [1-6]. To solve this problem, electromagnetic interference (EMI) shielding materials were developed. Traditionally, metals are widely used as the EMI shielding materials due to their high electrical conductivity [7]. However, metals have many disadvantages, such as heavy weight, prone to corrosion and inconvenient for processing, which limit their practical applications as EMI shielding materials in many fields. On the other hand, conductive polymer composites (CPCs) are considered as the promising alternatives of metals in the electromagnetic shielding materials because of their light weight, high flexibility, resistance to corrosion, low cost and good processing performance [8-14]. In general, CPCs are fabricated by filling the conductive fillers, including metal nanoparticles and carbonaceous nanoparticles, into the polymer matrix.

Different dimensional carbonaceous nanoparticles, such as zero-dimensional (0D) carbon black (CB) [15, 16], one-dimensional (1D) carbon nanotubes (CNTs) [17, 18], carbon fiber [19, 20], and two-dimensional (2D) graphene (GP) [21-23] and MXene[24], have been widely used as the conductive fillers to fabricate the CPCs-based EMI shielding materials. For instance, Mondal et al. [25] filled 0D CB into chlorinated polyethylene to fabricate the flexible EMI shielding composites, which exhibited a highest EMI shielding effectiveness (EMI SE) value of 38.4 dB (at 30 wt% loading) in the X-band frequency range (8.2-12.4 GHz). Yang and coworkers [26] dispersed 1D CNTs at the interface of polyvinylidene difluoride and ethylene-a-octene block copolymer blend by melt compounding, which can significantly improve both the EMI SE and mechanical properties of the nanocomposites. Fan et al. [27] utilized the prefabricated ultrathin graphene papers to fabricate the EMI shielding film materials with EMI SE value of

46.3 dB at a thickness of 0.3 mm. In addition, a combination of different dimensional fillers together to construct the hybrid conductive network was also used to improve the EMI shielding properties of the CPCs-based composites [28-33]. Although it is obvious that EMI shielding properties of CPCs highly depend on the conductive filler dimensionalities, there is rare systematical investigation on this issue.

In this work, we blend different dimensional carbon materials, including 0D CB, 1D CNTs, 2D GP, and their combinations, into isoprene rubber (IR) by a simple and large-scale preparation way. The dependence of the microstructure, electrical conductivity, EMI shielding properties and mechanical properties of composites on the filler dimensionality are systematically studied. It is found that IR/CNTs composites exhibit the highest EMI shielding properties attributed to the large aspect ratio and good dispersion of CNTs in IR matrix. Although GP nanosheet also has large aspect ratio, its serious stacking in IR matrix results in the poor electrical conductivity and EMI shielding properties of the composite. Moreover, it is found that utilizing the cheap CB to replace part of expensive CNTs to construct a hybrid network in IR matrix can fabricate the high-performance EMI shielding materials, which are comparable to IR/CNTs composite.

## 2. Experimental

### *2.1 Materials*

Isoprene rubber (SKI-3, Russia) was chosen as the matrix of the rubber-based composite. The Mooney viscosity of raw rubber (ML1+4100 °C) is ca. 70. CB (Printex XE 2B) used here is a super conductive carbon black with a particle size of < 30 nm and surface area of 950 $m^2/g$. CNTs (TNM1, purity > 98%) with lengths of 10–30 μm, inside diameters of 2-5 nm, outside diameters < 8 nm, special surface area of 220-300 $m^2/g$, is provided by Chengdu Organic Chemistry Co. Ltd., Chinese Academy of Sciences. GP with a particle size about 5 μm, C/O ratio > 20 %, special surface area of 30 $m^2/g$, is supplied by Changchun Mote Technology Co., Ltd. The layer number in each single GP nanosheet is less than 10. Other compounding ingredients like stearic acid, sulfur, ZnO and accelerant CZ (N-Cyclohexyl-2-benzothiazolylsulfenamide) are of industrial grade.

### *2.2 Composites preparation*

The isoprene rubber-based composites filled with carbon materials were fabricated by a two-roll mill. For the detailed process, firstly, the IR was masticated for 1 min by a two-roll mill at the temperature of 70 °C to form a large IR thin sheet. Subsequently, the compounding ingredients of 2 wt% stearic acid, 5 wt% ZnO, 0.9 wt% accelerant CZ and 2.2 wt% sulfur were rapidly added into the IR. After mixing for 2 min, the carbon fillers of CB, CNTs, GP, CNTs/CB or CNTs/GP were added. The total blending time for all the isoprene rubber-based composites with carbon materials was kept at 10 min to get better dispersion of fillers. Then, the resulting composites were let stand 24 h for stress relaxation and diffusion of ingredients. After that, the composites were re-blended by a two-roll mill for 3 min. Finally, the isoprene rubber-based composites with carbon materials were vulcanized in a compression molding press at 150 °C and 10 MPa for 25 min to receive the sheets with a thickness of 1 mm. For simplification, individual samples are labeled as conductive filler type and content, for example, CB10 is expressed as the composite with 10 wt% CB nanoparticles.

### *2.3 Characterization*

Two different instruments were used to carry out the electrical conductivity measurements in this study. For the composites with conductivity lower than $10^{-4}$ S/m, the alternating-current (AC) conductivity measurements were performed at room temperature in the frequency range between $10^{0}$ and $10^{6}$ Hz by an Alpha high-performance frequency analyzer coupled to a Novocontrol Broadband Dielectric Spectrometer. For the composites with conductivity higher than $10^{-4}$ S/m, a digit precision multimeter (DMM7510, Keithley Instruments Inc., USA) was used to test the electrical resistance of the composites and the conductivity was calculated by the following equations: $\sigma=1/\rho$ and $\rho=RS/L$, where $\sigma$, R, S and L represented the electrical conductivity, electrical resistance, the cross-section area of the sample, and the gauge length between the two electrodes, respectively. Scanning electron microscopy (SEM, FEG, Micro FEI PHILIPS) was utilized to characterize the morphologies of the isoprene rubber-based composites with carbon materials. Before the SEM measurements, the samples were immersed in liquid nitrogen for 40 min and then fractured in liquid nitrogen. Subsequently, fracture surfaces of the samples were sputter-coated with a thin layer of gold for SEM measurements. Electromagnetic interference shielding performance and scattering parameters of the composites was measured with an Agilent N5230 vector network analyzer in accordance with ASTM ES7-83. All the samples with 10 mm diameter and 1.0 mm thickness were measured in the frequency range of 12-18 GHz. Mechanical testing was carried out on a universal test machine (Instron-5889, USA) at a rate of 500 mm/min. Specimens for the test were cut into a dumbbell-shape with a dimension of 50 mm (length overall) × 4 mm (width of the narrow section) × 1 mm (thickness).

## 3. Results and discussion

Electrical conductivity, one of the intrinsic requirements of electromagnetic interference shielding materials to absorb the electromagnetic energy, is crucial for electromagnetic interference shielding effectiveness. The dependence of the electrical conductivities of IR-based composites filled with different dimensional conductive fillers on the filler content is presented in Fig. 1. It is clear that the electrical properties of the IR-based composites highly depend on the filler dimensionality. It is observed that IR/CNTs composites exhibit superior electrical conductivity than those of the IR/CB and IR/GP composites at the same loading. This is because 1D CNTs have a large-aspect-ratio and outstanding electrical conductivity, which are easier to form the perfect conductive network inside polymer matrix. Yet surprisingly, GP/IR composites display very poor electrical properties even GP nanosheet also possesses excellent electrical conductivity and large-aspect-ratio. The electrical conductivity of GP/IR composites almost remains unchanged as the increasing of GP content. This implies that GP may have serious aggregation or stacking in IR matrix, which are not well dispersed during the blending. For CB/IR composites, it is observed that the electrical conductivity is increased slowly when CB content is lower than 20%. Thereafter, its electrical conductivity is rapidly increased with the further increase of CB loading. This indicates that it needs a relatively high loading of CB nanoparticles to construct a completely percolated network in IR matrix.

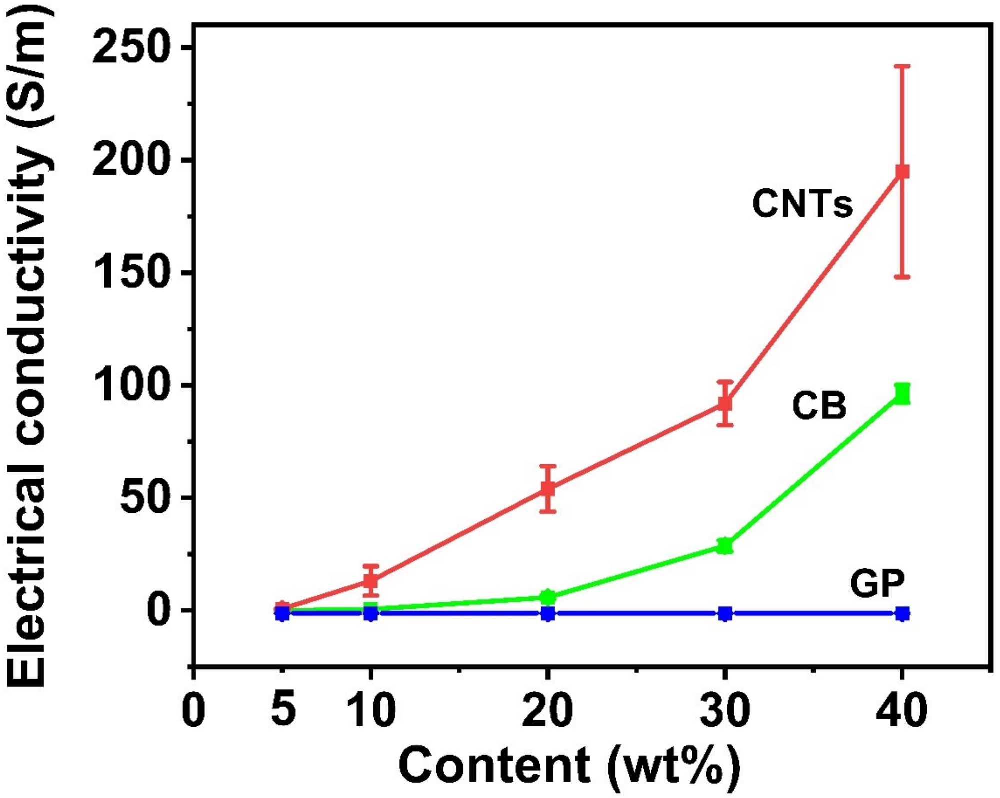

Fig. 1 Dependence of the electrical conductivity of IR-based composites filled with different carbon fillers on the filler content.

It is clear that the electrical properties of CPCs highly depend on the dispersion of filler inside polymer matrix. Generally, good dispersion of conductive fillers inside polymer matrix helps to the construction of percolated conductive network, which is critical to the electrical properties of the resulting composites. Figure 2 presents a series of SEM images to visualize the microstructures of IR-based composites filled with different conductive carbonaceous nanoparticles. For IR/CB composite, it is observed that 0D CB nanoparticles are well dispersed in IR matrix (Figure 2a1 and a2). Only few small spherical or cluster-like aggregations are observed. 1D CNTs exhibit an excellent dispersion in IR matrix, as shown in Figure 2b1 and b2. Even at a high loading of 20 wt% CNTs (Figure 2b2), no agglomeration of CNTs is observed. However, serious stacking of GP layers is observed from the SEM images of IR/GP composites (Figure 2c1 and c2). From the partial enlarged image (Insert in Figure 2c2), it can clearly observe the stacking of GP nanosheets. This maybe because that the original GP nanosheets possess the multi-layer structures, which cannot be exfoliated after by the blending in the two-roll mill. In addition, these stacked GP nanosheets also tend to aggregate into large-scale agglomerations instead of uniform dispersing in IR matrix. As a result, IR/GP composites show very poor electrical properties, as shown in Figure 1.

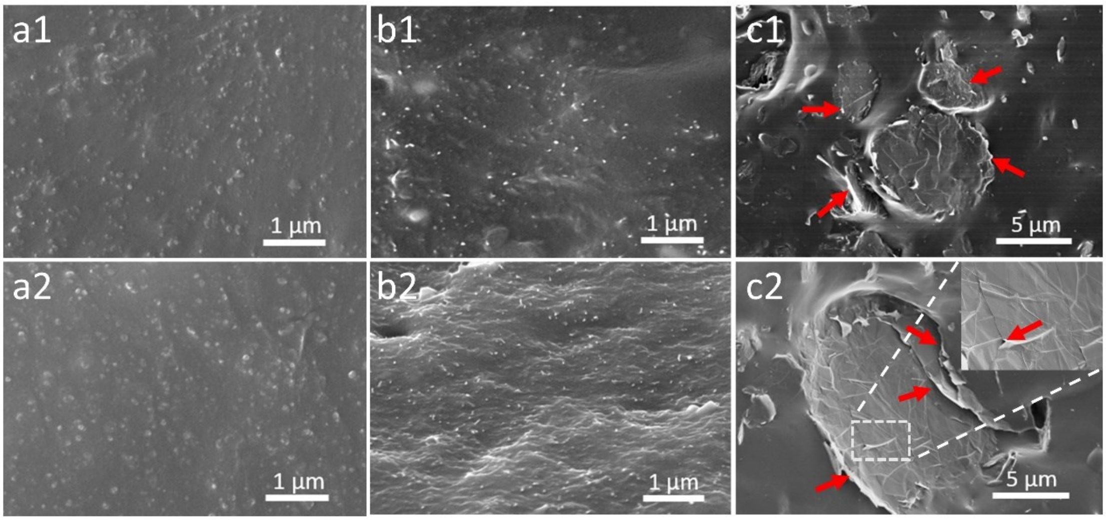

Fig. 2 SEM images of IR-based composites filled with different dimensional fillers. (a1) CB10 sample, 10 wt% CB; (b1) CNT10 sample, 10 wt% CNTs; (c1) GP10 sample, 10 wt% GP; (a2) CB20 sample, 20 wt% CB; (b2) CNT20 sample, 20 wt% CNTs; (c2) GP20 sample, 20 wt% GP.

EMI SE of the IR-based composites were measured in the frequency range of 12-18 GHz by an Agilent N5230 vector network analyzer at room temperature. The power coefficients of reflection (R), transmission (T), and absorption (A) can be determined by measuring the scattering parameters (S11 and S12), and their relationship can be described as R+A+T=1. Then, $SE_T$ (the total EMI SE), $SE_R$ (SE reflection), and $SE_A$ (SE absorption), can be calculated by the following equations [34]:

$$SE_T=-10\log T \qquad (1)$$

$$SE_R=-10\log(1-R) \qquad (2)$$

$$SE_A=SE_T-SE_R-SE_M \qquad (3)$$

where $SE_M$ is the microwave multiple internal reflections, which can be neglected when $SE_T \geqslant 15$dB [35]. $SE_T$ values of IR-based composites filled with different dimensional fillers in the range of 12-18 GHz are presented in Fig. 3. Basically, $SE_T$ is increased as the content of conductive filler is increased. However, there is a large difference between the $SE_T$ of the composites loaded with different dimensional fillers. At the same loading, 1D CNTs-filled composites exhibit a significantly higher $SE_T$ value than those of IR/CB and IR/GP composites. When the loading of CNTs is 30 wt% (CNT30), its $SE_T$ reaches up to 60 dB. At the same content, however, the $SE_T$ values of GP30 (30 wt% GP in IR) and CB30 (30 wt% CB in IR) are ca. 5 dB and 25 dB, respectively. It is worth to note that GP filled composites exhibit the worst EMI shielding properties because of their poor electrical properties. Compared Fig. 3 and 2, it is clear that EMI shielding properties of the composites correspond with their electrical properties, i.e., the higher the electrical conductivity, the higher the EMI SE.

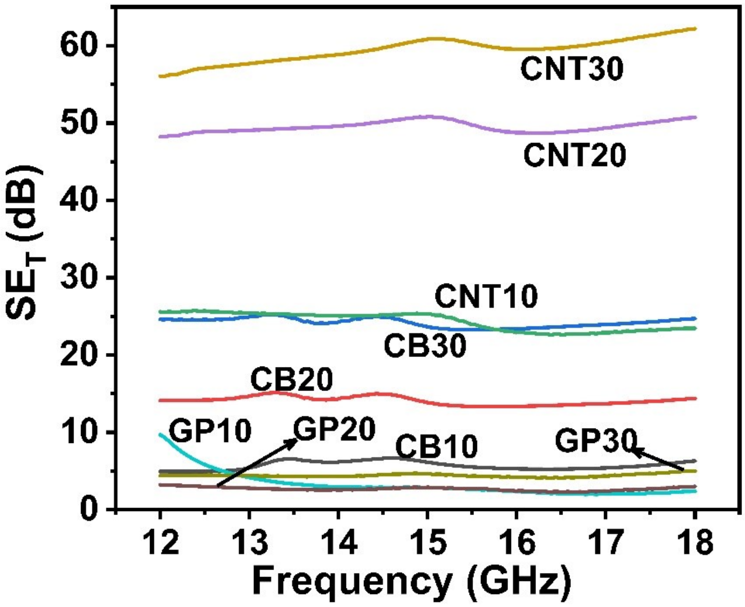

Fig. 3 $SE_T$ of IR-based composites filled with different dimensional conductive fillers.

In order to make clear the contributions of each shielding mechanism, i.e. absorption and reflection, the values of $SE_A$ and $SE_R$ of IR/CB and IR/CNTs composites are compared in Fig. 4a and 4b, respectively. It is clearly observed that $SE_A$ is much higher than $SE_R$, indicating that the electromagnetic interference shielding properties mainly dominated by the microwave absorption. This implies that absorption consumption is the main mechanism of electromagnetic shielding for carbon nanoparticles filled CPCs. For metal EMI shielding materials, their primary mechanism of electromagnetic shielding is the reflection mechanism. Moreover, the microwave absorption, i.e. $SE_A$, is rapidly increased as the increase of filler content. However, only a slight increase of

$SE_R$ with filler content is observed for both IR/CB and IR/CNTs composites. Take sample CNT30 as an example, the contribution of the absorption is ca. 9 times larger than that of the reflection at a frequency of 12 GHz.

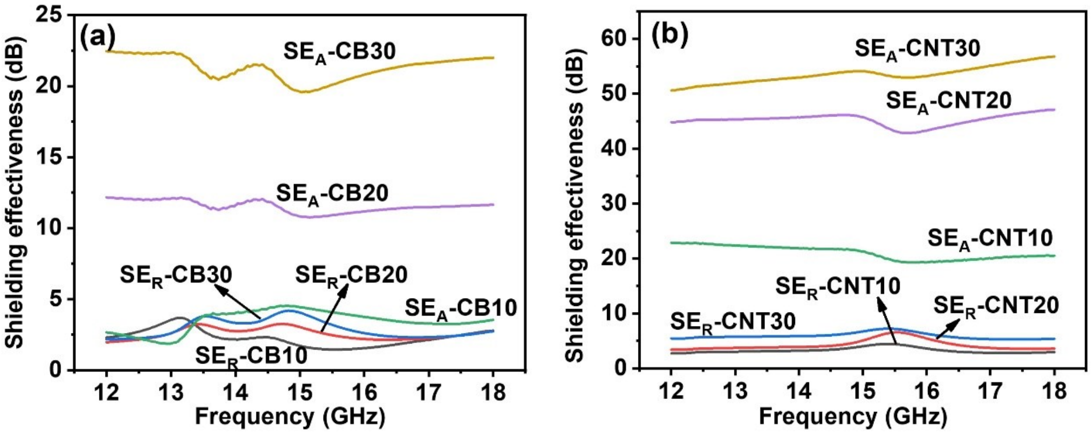

Fig. 4 $SE_A$ and $SE_R$ of IR/CB (a) and IR/CNTs (b) composites, respectively.

For the elastic materials, elongation at break is a very important parameter for their practical applications. In Fig. 5, we present the curves of young's modulus as a function of the strain for the composites containing with different conductive fillers, i.e. CB (Fig. 5a), CNTs (Fig. 5b), and GP (Fig. 5c), respectively. Clearly, the addition of conductive fillers into the IR matrix can largely enhance the young's modulus of materials. Moreover, it can be seen that 1D CNTs can bring a more significant enhancement in young's modulus compared to 0D CB and 2D GP. This may be attributed to the formation of perfect CNTs networks in IR matrix. However, there is an obvious decline of the elongation at break for IR/CB and IR/CNTs composites as the filler content is increased. Taking IR/CNTs composites as the example, the elongation at break of the material is declined from 1200% to 260% with the increasing of CNTs loading from 0 to 40 wt%. The reason why the elongation at break of the IR/CB and IR/CNTs composites decreases with the filler content may be attributed to the strong interaction between IR and filler particles, which limits the movement of IR chains. It is interesting to note that the elongation at break of IR/GP composites almost remain unchanged with the increasing of GP loading. This phenomenon is attributed to the serious stacking and aggregation of GP particles in IR matrix, which only has the negligible influence on the continuity of the rubber matrix and the movement of the IR chain.

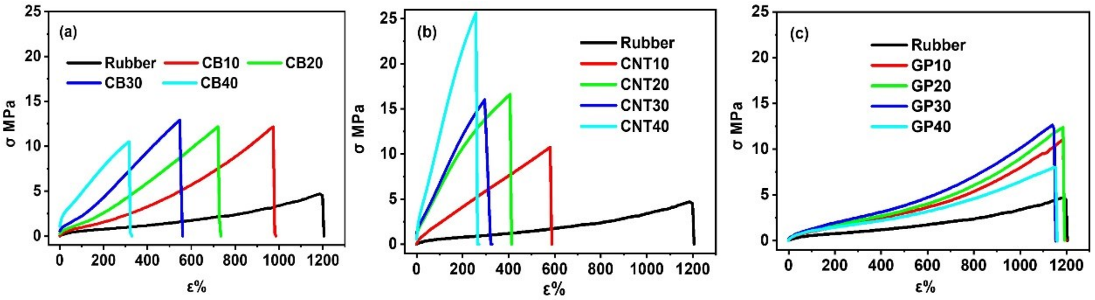

Fig. 5 The elongation at break of composites filled with different proportions of conductive fillers. (a) IR/CB composites with different CB loadings. (b) IR/CNTs composites with different CNTs loadings. (c) IR/GP composites with different GP loadings.

Combing different conductive fillers together to construct the hybrid conductive network is an effective strategy to improve the electrical properties of CPCs [36-39]. For instance, Zhao et al. observed that the percolation threshold of hybrid CB/MWCNTs fillers filled CPCs was much lower than MWCNTs or CB filled CPCs [40]. Moreover, it is also an effective strategy to reduce the cost of the CPCs by partially replacing expensive conductive fillers with the cheap conductive fillers. However, the influence of the hybrid conductive network on the EMI shielding properties is still not so clear. Herein, we combine 0D CB and 1D CNTs together to blend with IR to fabricate the hybrid IR/CB/CNTs composites. Their electrical properties, EMI shielding properties, and mechanical properties are compared with the corresponding CB or CNTs filled composites in Figure 6. Figure 6a shows the electrical conductivities of CB20, CNT20 and CB+CNT20 (CB/CNT=1:1) composites, respectively. It is worth to note that the hybrid CB+CNT20 composite even exhibits higher electrical conductivity than CNT20 composites. This may be attributed to the synergetic effect on the electrical properties when replacing part of CNTs by CB nanoparticles [32]. In Fig. 6b, we present the SEM image of the hybrid CB+CNT20 composite. It is observed that both CB and CNTs are homogeneously mixed together in IR matrix. Clearly, the combination of 0D CB and 1D CNTs can form a more perfect conductive network, as illustrated in Fig. 6c and 6d. First, the particulate CB can serve as “crosslinked points” to connect the neighboring CNTs (Fig. 6c). Second, CNT has large aspect ratio, which can act as long-distance charge ‘transporters’ to connect the isolated CB clusters (Fig. 6d). Attributed to the synergistic effect of hybrid CB-CNT nanofillers, CB+CNT20 (CB/CNT=1:1) composite possesses higher electrical conductivity than those of CB20 and CNT20 composites.

The similar synergetic effect of hybrid CB and CNTs on the electrical properties was reported in different polymer matrices [41-45]. However, it is still not clear whether hybrid CB-CNT network has the synergetic effect on the EMI shielding properties of the resulting CPCs. Therefore, we compare the electromagnetic interference shielding properties of IR/CB, IR/CNTs, and hybrid IR/CB/CNTs composites in Fig. 6e. It is found that the total EMI shielding effectiveness of hybrid CB+CNT20 composite is around 30 dB, which is between that of CB20 (ca.15 dB) and CNT20 (ca.50 dB). This implies that there is no synergetic effect of CNTs and CB on the EMI shielding properties of the CPCs and the electromagnetic interference shielding properties is not only determined by the electrical conductivity of the composites. To clearly understand the mechanisms, the microwave absorption ($SE_A$) and reflection ($SE_R$) of CB20, CNT20, and CB+CNT20 composites are compared in Figure 6f. For the microwave reflection, it is observed that the values of $SE_R$-CB20, $SE_R$-CNT20, and $SE_R$-CB+CNT20 are comparable. However, there are distinct gaps between the values of $SE_A$-CB20, $SE_A$-CNT20, and $SE_A$-CB+CNT20. At a frequency of 12 GHz, $SE_A$-CNT20 is almost 3.7 times of $SE_A$-CB20, while $SE_A$-CB+CNT20 is between $SE_A$-CB20 and $SE_A$-CNT20. This implies that the electromagnetic wave absorption loss highly depends on the conductive filler dimensionality. For example, Qin et al. [46] analyzed the microwave absorption in CPCs filled with different carbonaceous particles, which also found that CNTs were the better absorbing candidates than CB and graphite. In addition, the mechanical properties of the hybrid composite are also examined. Fig. 7 presents the elongation at break of CB20, CNT20, and CB+CNT20. It is observed that the elongation at break of CB+CNT20 is also between that of CB20 and CNT20. Based on a comprehensive consideration of cost, EMI and mechanical properties of the composites, it is an effective strategy to design the high-performance EMI shielding materials via the combination of different dimensional fillers.

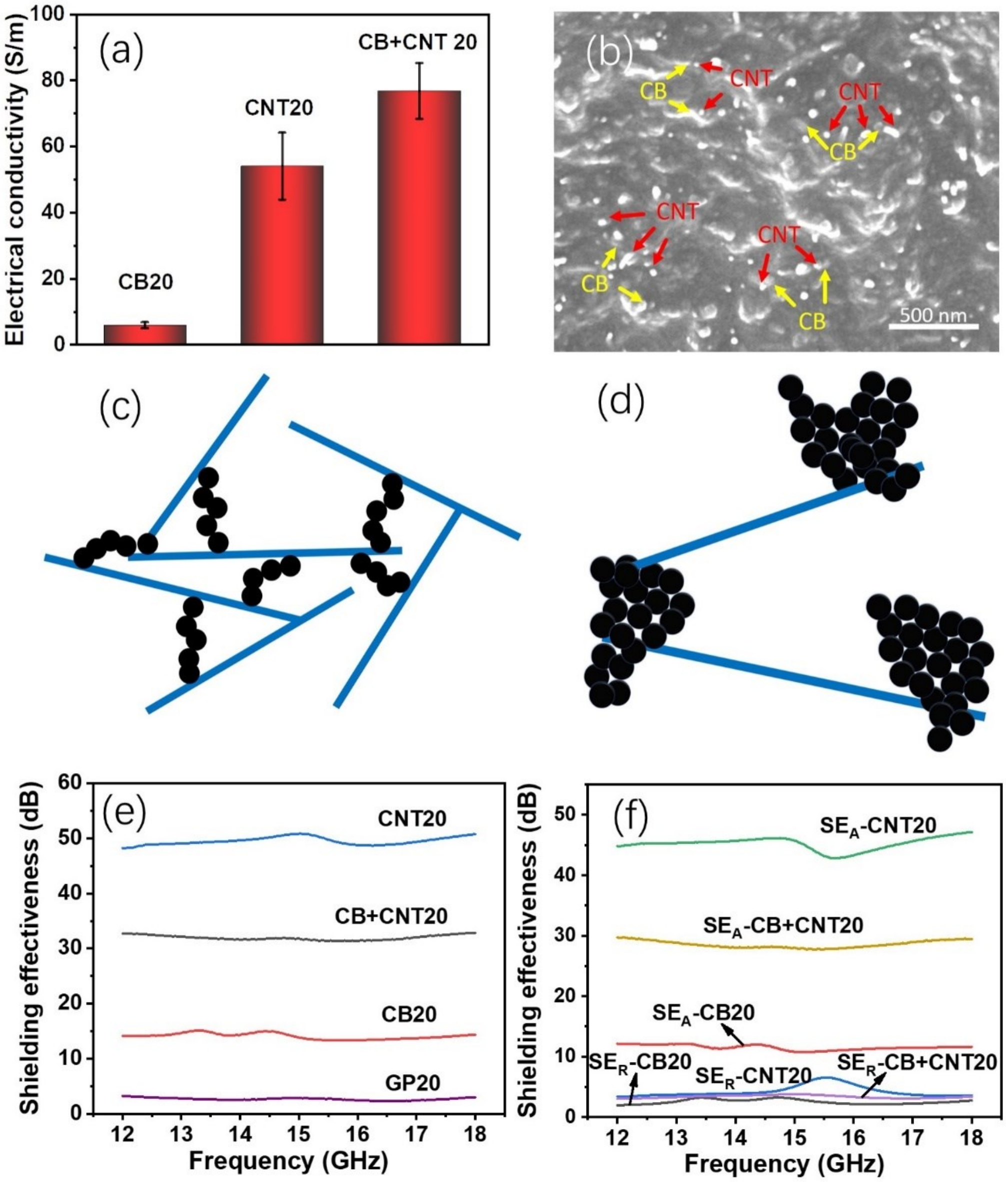

Fig. 6 Properties and morphologies of the composite of CB+CNT20: (a) Electrical conductivity; (b) SEM image showing the microstructures of hybrid CB+CNT20 composite; (c) Schematic diagram showing the CB particles act as the “cross-linked” points to connect the CNTs; (d) Schematic diagram showing large-aspect-ratio CNTs act as long-distance charge ‘transporters’ to connect the isolated CB clusters. (e) EMI shielding effectiveness of CB20, CNT20 and CB+CNT20; (f) The microwave absorption (SE$_A$) and reflection (SE$_R$) of CB20, CNT20 and CB+CNT20, respectively.

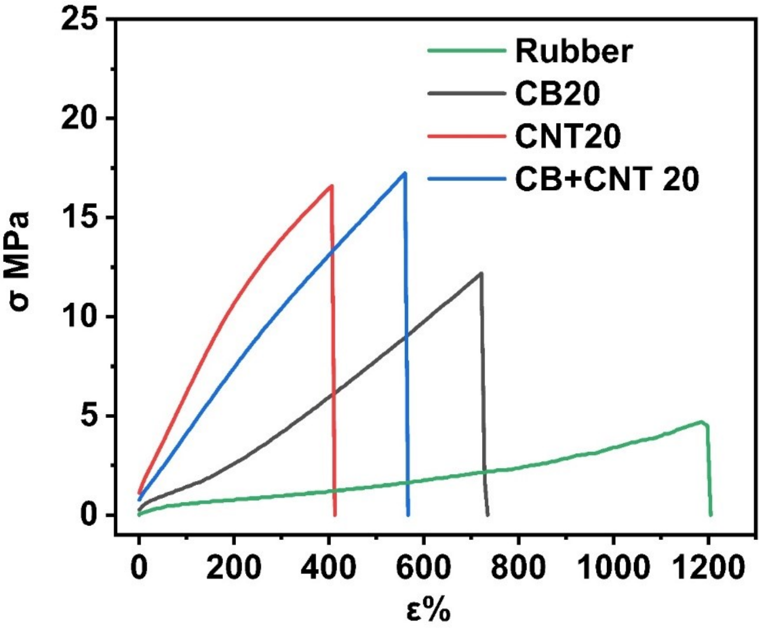

Fig. 7 The elongation at break of different conductive fillers with the same loading.

## 4. Conclusion

In the current study, we systematically investigate the mechanical, electrical, and EMI shielding properties of IR-based composites filled with different dimensional carbon nanoparticles, including 0D CB, 1D CNTs, and 2D GP. At the same loading, IR/CNTs composites always exhibit much higher EMI shielding effectiveness than IR/CB and IR/GP composites because of the outstanding electrical conductivity of CNTs and their good dispersion in IR matrix. In contrast, IR/GP composites possess the poor electrical conductivity and EMI shielding effectiveness because of the serious stacking and agglomeration of GP in IR. In addition, the EMI shielding mechanism of IR-based composites is investigated, which finds that the contribution of absorption consumption plays a decisive role on the EMI shielding properties of the composites. It is found that the electromagnetic wave absorption of CNTs is significantly higher than CB and GP, thus causing the higher EMI shielding effectiveness of IR/CNTs composites. Furthermore, it is interesting to note that hybrid IR/CB/CNTs (CB/CNTs: 1/1) composites have the comparable EMI shielding properties but higher mechanical properties and electrical conductivity compared to IR/CNTs composites. This proposes an effective route to fabricate the high-performance but low-cost EMI shielding materials by partially replacing expensive CNTs with CB.

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## References:

[1] F. Shahzad, M. Alhabeb, C.B. Hatter, B. Anasori, S.M. Hong, C.M. Koo, Y. Gogotsi, Electromagnetic interference shielding with 2D transition metal carbides (MXenes), Science 353 (2016) 1137-1140.

[2] W. Yang, Z. Zhao, K. Wu, R. Huang, T. Liu, H. Jiang, F. Chen, Q. Fu, Ultrathin flexible reduced graphene oxide/cellulose nanofiber composite films with strongly anisotropic thermal conductivity and efficient electromagnetic interference shielding, J. Mater. Chem. C 5 (2017) 3748-3756.

[3] M. Cao, C. Han, X. Wang, M. Zhang, Y. Zhang, J. Shu, H. Yang, X. Fang, J. Yuan, Graphene nanohybrids: excellent electromagnetic properties for the absorbing and shielding of electromagnetic waves, J. Mater. Chem. C 6 (2018) 4586-4602.

[4] H. G. Wei, H. Wang, A. Li, D. P. Cui, Z. N. Zhao, L. Q. Chu, X. Wei, Wei, L. Wei, D. Pan, J. C. Fan, Y. C. Li, J. X. Zhang, C. T. Liu, S. Y. Wei, Z. H. Guo, Multifunctions of Polymer Nanocomposites: Environmental Remediation, Electromagnetic Interference Shielding, And Sensing Applications. Chemnanomat 6 (2020) 1-12.

[5] B. Zhao, J. Deng, R. Zhang, L. Liang, B. Fan, Z. Bai, G. Shao, C. B. Park, Recent Advances on the Electromagnetic Wave Absorption Properties of Ni Based Materials. Eng. Sci. 3 (2018) 5-40.

[6] N. Wu, D. Xu, Z. Wang, F. Wang, J. Liu, W. Liu, Q. Shao, H. Liu, Q. Gao, Z. Guo, Achieving superior electromagnetic wave absorbers through the novel metal-organic frameworks derived magnetic porous carbon nanorods. Carbon 145 (2019) 433-444.

[7] Y. Wang, X. Jing, Intrinsically conducting polymers for electromagnetic interference shielding, Polym. Adv. Technol. 16 (2005) 344-351.

[8] X.H. Li, X. Li, K.N. Liao, P. Min, T. Liu, A. Dasari, Z.Z. Yu, Thermally annealed anisotropic graphene aerogels and their electrically conductive epoxy composites with excellent electromagnetic interference shielding efficiencies, ACS Appl. Mater. Interfaces 8 (2016) 33230-33239.

[9] Y. Chen, H.-B. Zhang, Y. Yang, M. Wang, A. Cao, Z.-Z. Yu, High-performance epoxy nanocomposites reinforced with three-dimensional carbon nanotube sponge for electromagnetic interference shielding, Adv. Funct. Mater. 26 (2016) 447-455.

[10] M. Tang, P. Zheng, Y. J. Wu, P. L. Zhu, Y. J. Qin, Y. Z. Jiang, R. Sun, C. P. Wong, Z. Li, Silver dendrites based electrically conductive composites, towards the application of stretchable conductors. Compos. Commun. 19 (2020) 121-126.

[11] Y.-J. Wan, G. Li, Y.-M. Yao, X.-L. Zeng, P.-L. Zhu, R. Sun, Recent advances in polymer-based electronic packaging materials. Compos. Commun. 19 (2020), 154-167.

[12] R. Sherman, V. Chalivendra, A. Hall, M. Haile, L. Nataraj, M. Coatney, Y. Kim, Electro-mechanical

characterization of three-dimensionally conductive graphite/epoxy composites under tensile and shear loading. Compos. Commun. 15 (2019), 30-33.

[13] C. Wang, V. Murugadoss, J. Kong, Z. He, X. Mai, Q. Shao, Y. Chen, L. Guo, C. Liu, S. Angaiah, Z. Guo, Overview of carbon nanostructures and nanocomposites for electromagnetic wave shielding. Carbon 140 (2018) 696-733.

[14] L. Lyu, J. Liu, H. Liu, C. Liu, Y. Lu, K. Sun, R. Fan, N. Wang, N. Lu, Z. Guo, E. K. Wujcik, An Overview of Electrically Conductive Polymer Nanocomposites toward Electromagnetic Interference Shielding. Eng. Sci. 2 (2018) 26-42.

[15] Q. Zhang, Y. Xu, Y. Yang, L. Li, C. Song, X. Su, Conductive mechanism of carbon black/polyimide composite films, J. Polym. Eng. 38 (2018) 147-156.

[16] S. Zheng, J. Deng, L. Yang, D. Ren, S. Huang, W. Yang, Z. Liu, M. Yang, Investigation on the piezoresistive behavior of high-density polyethylene/carbon black films in the elastic and plastic regimes, Compos. Sci. Technol. 97 (2014) 34-40.

[17] M.H. Aisaleh, U. Sundararaj, Electromagnetic interference shielding mechanisms of CNT/polymer composites, Carbon 47 (2009) 1738-1746.

[18] Y.A. Kim, T. Hayashi, M. Endo, Y. Gotoh, N. Wada, J. Seiyama, Fabrication of aligned carbon nanotube-filled rubber composite, Scr. Mater. 54 (2006) 31-35.

[19] A. Ameli, P.U. Jung, C.B. Park, Electrical properties and electromagnetic interference shielding effectiveness of polypropylene/carbon fiber composite foams, Carbon 60 (2013) 379-391.

[20] S.S. Tzeng, F.Y. Chang, EMI shielding effectiveness of metal-coated carbon fiber-reinforced ABS composites, Mater. Sci. Eng. A-Struct. Mater. Prop. Microstruct. Process. 302 (2001) 258-267.

[21] M.S. Cao, X.X. Wang, W.Q. Cao, J. Yuan, Ultrathin graphene: electrical properties and highly efficient electromagnetic interference shielding, J. Mater. Chem. C 3 (2015) 6589-6599.

[22] L. Jin, X. Zhao, J. Xu, Y. Luo, D. Chen, G. Chen, The synergistic effect of a graphene nanoplate/Fe3O4@BaTiO3 hybrid and MWCNTs on enhancing broadband electromagnetic interference shielding performance, RSC Adv. 8 (2018) 2065-2071.

[23] X. Z. Xu, G. M. Lin, D. Y. Liu, G. X. Sui, Y. Rui, Electrically conductive graphene-coated polyurethane foam and its epoxy composites. Compos. Commun. 7 (2018), 1-6.

[24] W. J. Yuan, J. Z. Yang, F. X. Yin, Y. B. Li, Y. Yuan, Flexible and stretchable MXene/Polyurethane fabrics with delicate wrinkle structure design for effective electromagnetic interference shielding at a dynamic stretching process. Compos. Commun. 19 (2020), 90-98.

[25] S. Mondal, S. Ganguly, M. Rahaman, A. Aldalbahi, T.K. Chaki, D. Khastgir, N. Das, A strategy to achieve enhanced electromagnetic interference shielding at low concentration with a new generation of conductive carbon black in a chlorinated polyethylene elastomeric matrix, Phys. Chem. Chem. Phys. 18 (2016) 24591-24599.

[26] X.J. Zha, J.H. Pu, L.F. Ma, T. Li, R.Y. Bao, L. Bai, Z.Y. Liu, M.B. Yang, W. Yang, A particular interfacial strategy in PVDF/OBC/MWCNT nanocomposites for high dielectric performance and electromagnetic interference shielding, Compos. Pt. A-Appl. Sci. Manuf. 105 (2018) 118-125.

[27] W.L. Song, L.Z. Fan, M.S. Cao, M.M. Lu, C.Y. Wang, J. Wang, T.T. Chen, Y. Li, Z.L. Hou, J. Liu, Y.P. Sun, Facile fabrication of ultrathin graphene papers for effective electromagnetic shielding, J. Mater. Chem. C 2 (2014) 5057-5064.

[28] M.H. Aisaleh, W.H. Saadeh, Hybrids of conductive polymer nanocomposites, Mater. Des. 52 (2013) 1071-1076.

[29] N. Bagotia, V. Choudhary, D.K. Sharma, Synergistic effect of graphene/multiwalled carbon nanotube

hybrid fillers on mechanical, electrical and EMI shielding properties of polycarbonate/ethylene methyl acrylate nanocomposites, Compos. Pt. B-Eng. 159 (2019) 378-388.

[30] E. Zhou, J. Xi, Y. Guo, Y. Liu, Z. Xu, L. Peng, W. Gao, J. Ying, Z. Chen, C. Gao, Synergistic effect of graphene and carbon nanotube for high-performance electromagnetic interference shielding films, Carbon 133 (2018) 316-322.

[31] Y. Yang, M.C. Gupta, K.L. Dudley, R.W. Lawrence, A comparative study of EMI shielding properties of carbon nanofiber and multi-walled carbon nanotube filled polymer composites, J. Nanosci. Nanotechnol. 5 (2005) 927-931.

[32] D.P. Schmitz, L.G. Ecco, S. Dul, E.C.L. Pereira, B.G. Soares, G.M.O. Barra, A. Pegoretti, Electromagnetic interference shielding effectiveness of ABS carbon-based composites manufactured via fused deposition modelling, Mater. Today Commun. 15 (2018) 70-80.

[33] W.L. Song, J. Wang, L.Z. Fan, Y. Li, C.Y. Wang, M.S. Cao, Interfacial engineering of carbon nanofiber-graphene-carbon nanofiber heterojunctions in flexible lightweight electromagnetic shielding networks, ACS Appl. Mater. Interfaces 6 (2014) 10516-10523.

[34] W. Yang, Y. Gong, X. Zhao, T. Liu, Y. Zhang, F. Chen, Q. Fu, Strong and highly conductive graphene composite film based on the nanocellulose-assisted dispersion of expanded graphite and incorporation of poly(ethylene oxide), ACS Sustain. Chem. Eng. 7 (2019) 5045-5056.

[35] N.C. Das, Y. Liu, K. Yang, W. Peng, S. Maiti, H. Wang, Single-walled carbon nanotube/poly(methyl methacrylate) composites for electromagnetic interference shielding, Polym. Eng. Sci. 49 (2009) 1627-1634.

[36] J. Chen, H. Li, Q. Yu, Y. Hu, X. Cui, Y. Zhu, W. Jiang, Strain sensing behaviors of stretchable conductive polymer composites loaded with different dimensional conductive fillers, Compos. Sci. Technol. 168 (2018) 388-396.

[37] Y. Nakaramontri, S. Pichaiyut, S. Wisunthorn, C. Nakason, Hybrid carbon nanotubes and conductive carbon black in natural rubber composites to enhance electrical conductivity by reducing gaps separating carbon nanotube encapsulates, Eur. Polym. J. 90 (2017) 467-484.

[38] V.H. Luan, H.N. Tien, T.V. Cuong, B.-S. Kong, J.S. Chung, E.J. Kim, S.H. Hur, Novel conductive epoxy composites composed of 2-D chemically reduced graphene and 1-D silver nanowire hybrid fillers, J. Mater. Chem. 22 (2012) 8649–8653.

[39] S. Zhu, C. Xing, F. Wu, X. Zuo, Y. Zhang, C. Yu, M. Chen, W. Li, Q. Li, L. Liu, Cake-like flexible carbon nanotubes/graphene composite prepared via a facile method for high-performance electromagnetic interference shielding, Carbon 145 (2019) 259-265.

[40] S. Zhang, L. Lin, H. Deng, X. Gao, E. Bilotti, T. Peijs, Q. Zhang, Q. Fu, Synergistic effect in conductive networks constructed with carbon nanofillers in different dimensions, Express Polym. Lett. 6 (2012) 159-168.

[41] Y.J. Zheng, Y.L. Li, K. Dai, Y. Wang, G.Q. Zheng, C.T. Liu, C.Y. Shen, A highly stretchable and stable strain sensor based on hybrid carbon nanofillers/polydimethylsiloxane conductive composites for large human motions monitoring, Compos. Sci. Technol. 156 (2018) 276-286.

[42] P. Coelho, V.A.D. Armellini, A.R. Morales, Assessment of percolation threshold simulation for individual and hybrid nanocomposites of carbon nanotubes and carbon Black, Mater. Res. 20 (2017) 1638-1649.

[43] M.H. Aisaleh, Synergistic effect of CNT/CB hybrid mixture on the electrical properties of conductive composites, Mater. Res. Express 6 (2019) 065011.

[44] G. Foteinidis, K. Tsirka, L. Tzounis, D. Baltzis, A.S. Paipetis, The role of synergies of MWCNTs and

carbon black in the enhancement of the electrical and mechanical response of modified epoxy resins, Applied Sciences-Basel 9 (2019) 3757.

[45] M.C. Qu, F. Nilsson, D.W. Schubert, Novel definition of the synergistic effect between carbon nanotubes and carbon black for electrical conductivity, Nanotechnology 30 (2019) 245703.

[46] F. Qin, C. Brosseau, A review and analysis of microwave absorption in polymer composites filled with carbonaceous particles, J. Appl. Phys. 111 (2012) 061301