ORIGINAL PAPER

Enhanced EMI shielding and conductive network formation in segregated polyethylene/carbon black composites governed by polymer particle size distribution

Fatemeh Najarnia, Jalil Vahdati Khaki, Samaneh Sahebian*, Ali Shajari

Department of Materials Science and Engineering, Ferdowsi University of Mashhad, Mashhad, Iran

*Corresponding Email: Email: s.sahebian@um.ac.ir

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ABSTRACT

As a novel class of lightweight conductive material, segregated polymer composites are widely regarded as their unique architecture enables the formation of electrical networks at exceptionally low filler loadings. In such configuration, the conductive phase is preferentially localized at the polymer microgranule boundaries, forming continuous channels for electron transport. This promotes superior performance, particularly in electromagnetic interference shielding. The influence of initial polyethylene particle size on electrical conductivity and electromagnetic interference shielding performance of polyethylene/carbon black segregated structured composites was studied in the present research. Composite samples were fabricated using polymer particles with varying size distributions (105–150, 150–250, 250–500, and mixed <500 μm). Characterization Results demonstrated that increase in initial polymer particle size enhances electrical conductivity; as an illustration, conductivity improved from 94.74 S/m at 105-150 μm to 163.31 S/m at 250-500 μm. In terms of shielding behavior, absorption ratio of 69-76%, confirms a dominant absorptive contribution in segregated structure regardless of initial particle size. The mixed-particle sample (<500 μm) exhibited the best overall performance, combining a high conductivity of 158.40 S/m with the highest total and absorption shielding effectiveness of 11.07 and 7.70 dB, respectively. The incorporation of finer particles not only improved interfacial adhesion among larger polymer particles, but also increased the number of "cages" that trap incident waves and prolong multiple scattering pathways.

Keywords: Segregated structure composites; polyethylene; carbon black; shielding effectiveness; electrical conductivity.

INTRODUCTION

As a new generation of multifunctional materials, conductive polymer composites have attracted significant attention, primarily due to their low density, excellent processability and property tunability via additive incorporation [1]. Recently, a special class of materials known as segregated polymer composites has been developed, distinguished by their ability to form efficient conductive networks at significantly reduced filler loadings relative to conventional composites with randomly dispersed fillers [2].

This effect attributed to the selective localization of the conductive phase at the interfaces between polymer particles in segregated structures. Such configuration facilitates the formation of continuous conductive pathways at very low filler loadings, providing desirable electrical and electromagnetic performance through a simple and cost-efficient processing approach [3, 4].

Beyond the benefits of reduced filler content and improved electrical performance [5], segregated structures demonstrate broad multifunction including their application in strain and stress sensors [6], self-regulating thermal components [7] and energy storage systems [8]. Furthermore, the rapid expansion of modern communication technologies has intensified exposure to electromagnetic waves, with unavoidable adverse consequences such as interference with electronic equipment [9, 10], tissue heating, and other biological effects in living organisms [11]. In this regard segregated polymer composites represent a promising candidate for efficient electromagnetic wave absorption [12, 13].

In the field of electromagnetic shielding applications, segregated polymer composites exhibit superior performance owing to their efficient conductive networks together with the relative transparency of the polymer matrix to incident waves. This dual characteristic enables the activation of both absorption and reflection mechanisms. The formation of conductive layers along particle boundaries facilitates energy dissipation of electromagnetic fields through multiple scattering and the subsequent transformation of electromagnetic energy into heat inside the material [14, 15]. As a result, these materials exhibit superior shielding effectiveness (SE) compared to conventional composites with randomly distributed fillers [13, 16]. Figure1 depicts a schematic illustration of electromagnetic attenuation inside the "cages" of a segregated polystyrene/graphene composite [17].

Among multiple factors influencing the properties of segregated-structure polymer composites, three principal categories of parameters play the most critical roles in defining the segregated architecture: (i) characteristics of the conductive filler, including shape, aspect ratio, and intrinsic conductivity; (ii) the physical and chemical features of the polymer matrix, such as particle size and molecular weight; and (iii) the processing conditions, such as method of mixing, temperature, pressure, and duration [17].

Yu et al. investigated the mechanical and electrical properties of segregated ultrahigh molecular weight polyethylene (UHMWPE)/ carbon nanotube (CNT) composites, highlighting their improved electromagnetic interference (EMI) shielding effectiveness [18]. Cheng et al. fabricated UHMWPE/ground tire rubber (GTR) segregated composites of hybrid carbon black fillers. They showed that tuning filler type and content improved conductivity and achieved effective EMI shielding at low filler loadings while maintaining desirable mechanical properties [19]. While polymer particle size is one of the very important parameters governing the development of conductive networks in segregated polymer composites, its direct impact on the overall performance of the structure has received comparatively limited attention.

Zhang et al. investigated the effect of polypropylene particle size in the millimeter range (≈0.9–3.6 mm), revealing that larger particle sizes in segregated polypropylene (PP)/CNT composites markedly reduced the electrical percolation threshold from 0.33 to 0.08 wt%. This reduction facilitated the formation of more concentrated conductive pathways, leading to development of electrical conductivity and electromagnetic wave absorption capability [20]. Similarly, Li et al. investigated PP/CNT segregated composites with polymer particle sizes ranging from 20 to 1200 μm and reported a decrease in percolation threshold from 1.32 to 0.44 vol% with increasing particle size, confirming the inverse correlation between particle size and electrical percolation threshold [21]. Although numerous research has investigated various influencing parameters on segregated structures, the specific effect of sub-millimeter polymer particle sizes on the formation and performance of segregated architecture remains largely unexplored.

Considering the functional advantages of segregated structures and the critical role of microstructural parameters in determining their performance, the present study focuses on the effect of polymer particle size in the sub-millimeter range on the resulting microstructure, electrical conductivity, and EMI shielding effectiveness. The objective is to establish a clearer understanding of the relationships between polymer particle size, conductive network formation, and the mechanisms of absorption and reflection, thereby providing insights for optimizing the design and fabrication of this new class of advanced composites.

EXPERIMENTAL

Materials

High-density polyethylene (HDPE) powder as the polymer matrix, and carbon black (CB) N220 as the conductive filler were purchased from Baspar Tejarat Asia Co. and Carbon Simorgh Co., respectively. The as received HDPE powder was sieved using laboratory mesh screens numbered 35, 60, 100, and 140, which were sequentially mounted on a mechanical shaker to achieve precise particle size separation.

Consequently, four particle size ranges were selected: 105–150, 150–250, 250–500 μm, and a mixed sample (<500 μm). Particles exceeding 500 μm were excluded due to the 1 mm thickness limitation of the molded samples and potential issues related to incomplete mold filling. The mesh numbers and their corresponding

particle size ranges used for HDPE powder classification are summarized in Table 1.

Composite Preparation

Initially, the composite powder was prepared by combining the HDPE powder with 5 wt% carbon black nanoparticles [22], a loading that supports the formation of a continuous coating of the reinforcing phase on the polymer particles. The mixture was processed in a Retsch mixer mill at 20 Hz for 10 minutes, a duration sufficient for carbon black to distribute uniformly and completely surround the polymer granules. The resulting composite powder was hot-pressed in a steel mold (1 × 40 × 10 mm^3) at 124 °C for 180 s. The sample codes related to the prepared composites are listed in Table2. An overview of the experimental process followed in this study is illustrated in Figure2.

Characterization

The distribution of the conductive phase and the surface coverage of polymer particles was studied using a field emission scanning electron microscopy (FESEM) (model: LEO VP1450 microscope, Germany) operating at an accelerating voltage of 20 kV. Prior to imaging, all samples were sputter-coated with a thin layer of gold to enhance surface conductivity. The performance of the composite samples was evaluated by measuring their electrical conductivity and electromagnetic shielding effectiveness. Electrical resistance measurements were conducted at room temperature using a Keithley four-point probe system.

To evaluate the electrical conductivity of the composite samples, data was collected from 40 randomly selected points on each specimen. The measured resistance values were converted to electrical conductivity and averaged to yield a representative value that served as the basis for the analysis. The electromagnetic shielding effectiveness was analyzed in the 8–12 GHz frequency range using an Agilent Vector Network Analyzer (VNA, model E8363B).

RESULTS AND DISCUSSION

Figure 3 demonstrates FESEM micrograph of the HDPE/CB composite powder after the mixing process. The images reveal that the surface of the polyethylene particles is completely coated with carbon black nanoparticles. The absence of uncovered regions or excessive agglomeration indicates that the conductive phase has formed an interfacial conductivity, which can further make a continuous network in contact with other coated polymer particles [23]. This homogeneous coverage confirms the effective dispersion of the conductive filler during the mixing process, forming the selective localization of carbon black on the outer layer of polymer particles, prior to hot pressing. Such a microstructural arrangement is characteristic of an ideal segregated structure, facilitating effective electron transport pathways and contributing to the high electrical conductivity observed in the composites.

The average electrical conductivity of the composites was calculated from measurements taken at forty random points on each sample and is summarized in Table 2. The results indicate that the particle size of the polymer plays an important role in determining the electrical conductivity of HDPE/CB segregated composites. For the PE105/150 sample, an electrical conductivity of 94.74 S/m was obtained, confirming the formation of a conductive network. However, the smaller particle size and the resulting increase in interparticle boundaries, higher contact resistance occurs along the electron transport paths. As the particle size increases to the 250–500 μm range, the conductivity significantly rises to 163.31 S/m.

Regarding the conduction mechanism of segregated structures, the conductive phase selectively accumulates at the interfaces between polyethylene particles [24, 25]. As the polymer particle size increases, the specific surface area per unit volume decreases, leading to the formation of a thicker carbon black coating layer at the interfacial regions [8]. This thicker conductive layer improves network continuity the development of more continuous conductive networks, stabilizing electron transport and reducing the overall resistance of the composite.

For the mixed-size sample (PE5C-<500), the electrical conductivity inconsiderably decreased to 158.40 S/m. Although this value is marginally lower than that of the PE5C-250/500, it remains relatively high. This small reduction is attributed to the presence of smaller particles, which introduce additional interfacial boundaries and contact resistances. However, the relatively large average particle size helps maintain a strong and continuous conductive network.

On the other hand, the presence of smaller particles within the polymer powder distribution can act as a connecting agent, filling the gaps between larger particles and enhancing interparticle contact. Consequently, in addition to maintaining a relatively high level of conductivity, the structural integrity and interfacial adhesion are also improved. Therefore, while uniform and larger particle sizes provide the highest electrical conductivity, a mixed particle-size distribution offers a desirable combination of desirable electrical performance and improved macroscopic structural integrity, making it a more practical choice for composite fabrication.

The variation of electromagnetic shielding effectiveness with frequency for the PE5C-105/150, PE5C-150/250, PE5C-250/500, and PE5C-<500 samples are depicted in Figure 4. The results obtained from the vector network analyzer (VNA) measurements reveal that the mixed-size sample (PE5C-<500) exhibits the highest overall shielding effectiveness, with an average total SE of 11.07 dB. This sample not only demonstrates the maximum total shielding performance, but also achieves the highest absorption shielding effectiveness (SEA ≈ 7.70 dB), indicating a significant enhancement in EMI shielding efficiency through the tailored polymer particle size distribution.

Figure 5 represents the average electromagnetic shielding effectiveness of the PE5C-105/150, PE5C-150/250, PE5C-250/500, and PE5C-<500 samples. Mainly, absorption was recognized as the predominant mechanism, while reflection made a smaller contribution.

This behavior is consistent with the expected characteristics of segregated structures, where the formation of conductive networks along polymer particle boundaries provides multiple pathways for electromagnetic energy absorption and dissipation. In such architecture, each polymer microgranule surrounded by conductive carbon black acts as a "cage," where incident electromagnetic waves undergo multiple reflections and progressive energy loss [17]. Therefore, the segregated structure provides an effective framework for absorption-dominated electromagnetic shielding [26].

While only a few studies have addressed this specific topic, the available evidence suggests that although smaller polymer particles promote a more uniform filler distribution, they require a higher filler content to establish an effective conductive network [20]. In the mixed-size sample (PE5C-<500), both structural robustness and electromagnetic shielding performance were found to be optimized. The slight decrease in electrical conductivity observed for this sample can be attributed to the increased specific surface area of smaller particles, which require a higher amount of conductive filler content to maintain efficient interfacial contact.

As shown in Figure 6, the absorption fraction (SEA/SET) decreases slightly as the initial polymer particle size increases. Although the numerical differences vary within a narrow range, the trend remains meaningful: larger polymer granules create fewer segregated microdomains per unit volume, resulting in slightly lower absorption contributions. The slight reduction in absorption fraction does not undermine the overall enhancement in shielding performance, since both SEA and SET increase with particle size as a consequence of forming thicker and more continuous conductive pathways around the polymer granules.

Another size-dependent trend is demonstrated in Figure 7. In this figure, the electrical conductivity and the corresponding shielding effectiveness are presented as functions of the initial average polymer particle size. The results reveal an overall positive correlation, indicating that improved electrical pathways enhance the electromagnetic shielding performance of the segregated HDPE/CB composite. As the polymer particle size increases, the reduced specific surface area leads to fewer interfacial boundaries and lower contact resistance. This improves more continuous conductive networks, which facilitate the dissipation of electromagnetic energy across the composite. Consequently, a steady increase in total shielding effectiveness with electrical conductivity is observed for samples containing larger polymer particles, consistent with the conductive network formation mechanism inherent to segregated structures.

Interestingly, the mixed-particle sample (PE5C-<500) slightly deviates from this general trend. Despite exhibiting a marginally lower conductivity than the largest particle-size sample (PE5C-250/500), it demonstrates the highest overall shielding effectiveness. This enhanced performance can be attributed to the synergistic microstructure formed by the coexistence of fine and coarse polymer particles. The smaller particles fill the space between larger ones, increasing the number of reflective interfaces and multiple scattering sites, while the larger particles maintain a relatively thick and continuous conductive layer along intergranular boundaries. These properties enable effective electromagnetic energy absorption, yielding an optimal balance between electrical and shielding performance, particularly desirable for advanced lightweight EMI shielding materials.

Overall, the results confirmed that absorption loss is the dominant mechanism in all segregated structures. The fine balance between electrical conduction and electromagnetic absorption achieved in the mixed-particle sample highlights the importance of tuning particle size distribution as a design parameter. By optimizing this parameter, it is possible to maximize shielding performance without degrading mechanical integrity or requiring excess conductive filler.

CONCLUSION

In this study, the effect of polymer particle size distribution on the electrical conductivity and EMI shielding effectiveness of segregated high-density HDPE/CB composites was investigated. The results prove that polymer particle size plays a crucial role in determining the functional performance of segregated composites. As the particle size increased from 105–150 μm to 250–500 μm, the electrical conductivity notably enhanced from 94.7 S/m to 163.3 S/m, accompanied by an increase in total shielding effectiveness (SET) from 6.6 dB to 8.8 dB.

The shielding performance of the samples followed a generally positive correlation with electrical conductivity, where higher conductivity corresponded to improved electromagnetic shielding effectiveness. However, the mixed-size sample (PE5C-<500) exhibited a positive deviation from this trend. Despite its slightly lower conductivity (158.4 S/m) compared to the large-particle sample, it demonstrated the highest total shielding effectiveness (SET = 11.07 dB) and maximum absorption component (SEA ≈ 7.7 dB).

In summary, the present study proved that controlling the polymer particle size ranges represents an effective strategy to tailor both electrical and EMI shielding properties of lightweight segregated polymer composites. Especially The mixed-size configuration offers a synergistic improvement in conductivity and absorption-dominant shielding, providing a potential approach for further development of lightweight conductive materials.

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Figures

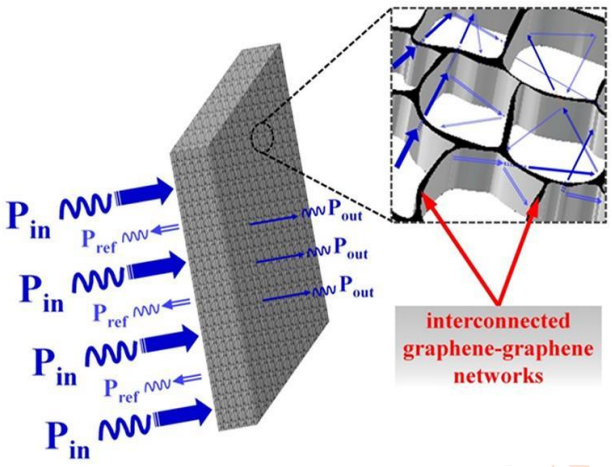


Figure1. Schematic illustration of electromagnetic wave energy attenuation in segregated structure cages [17].

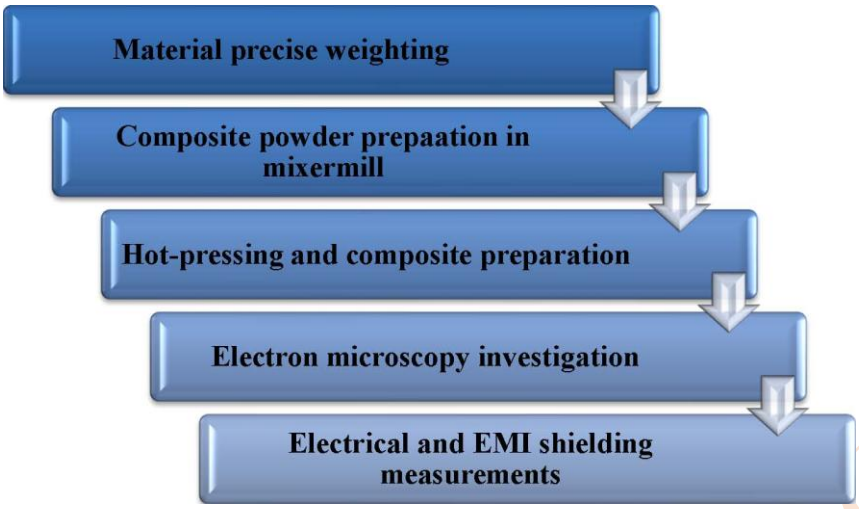


Figure 2. Schematic representation of the experimental workflow.

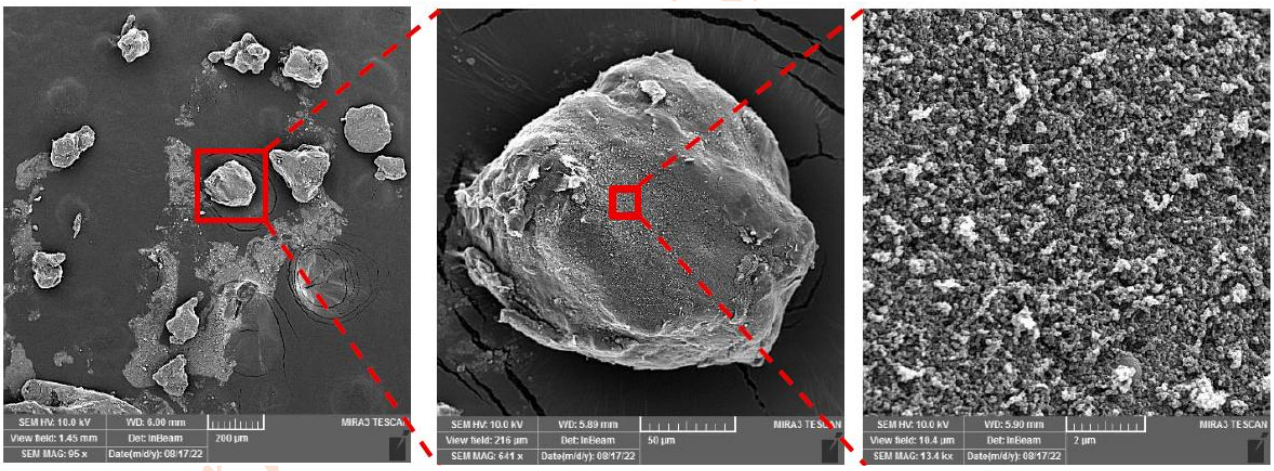


Figure 3. FESEM images of HDPE/5wt%CB composite powder after mixing process, illustrating uniform coating of polyethylene particles with carbon black prior to hot pressing.

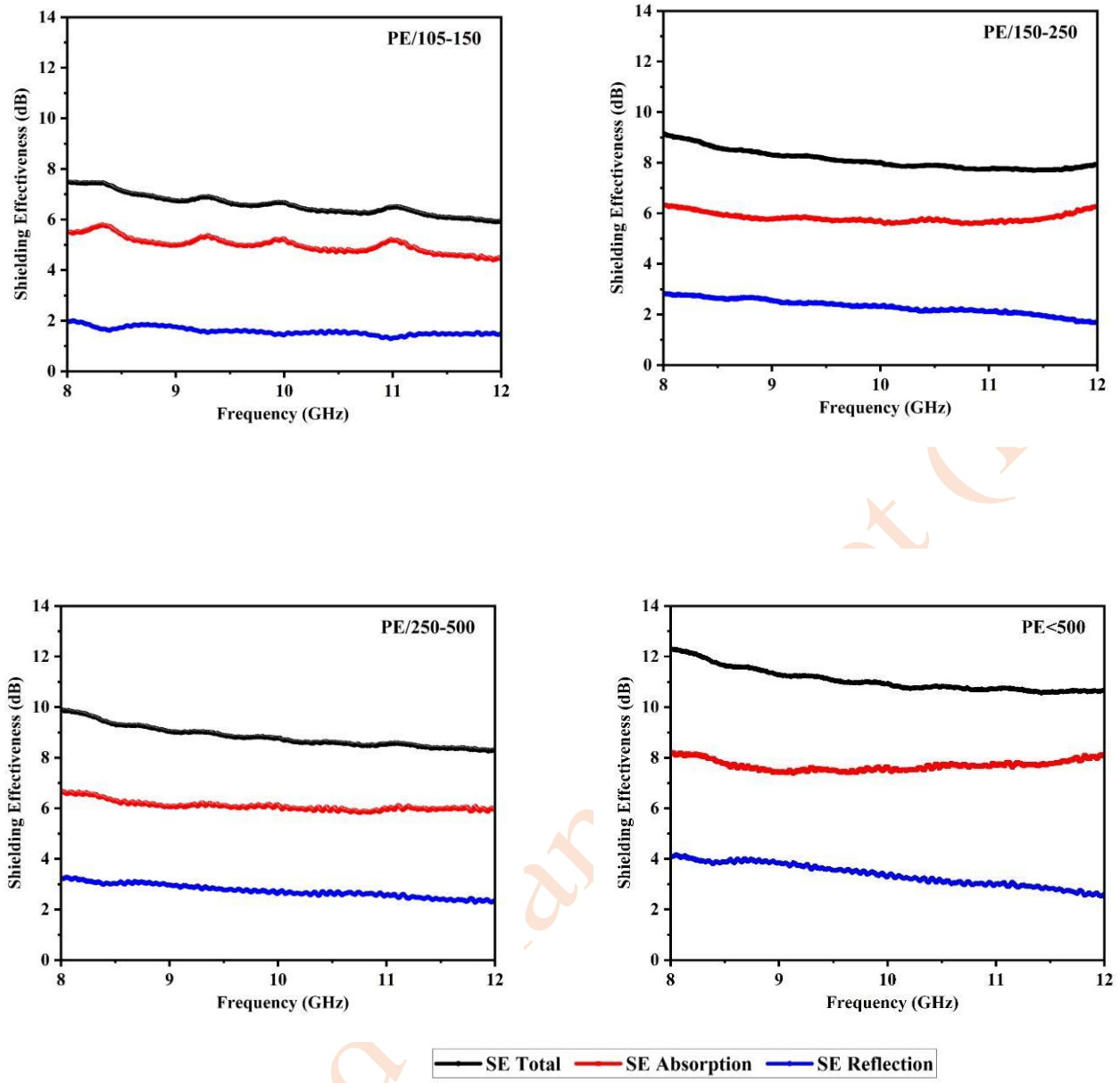
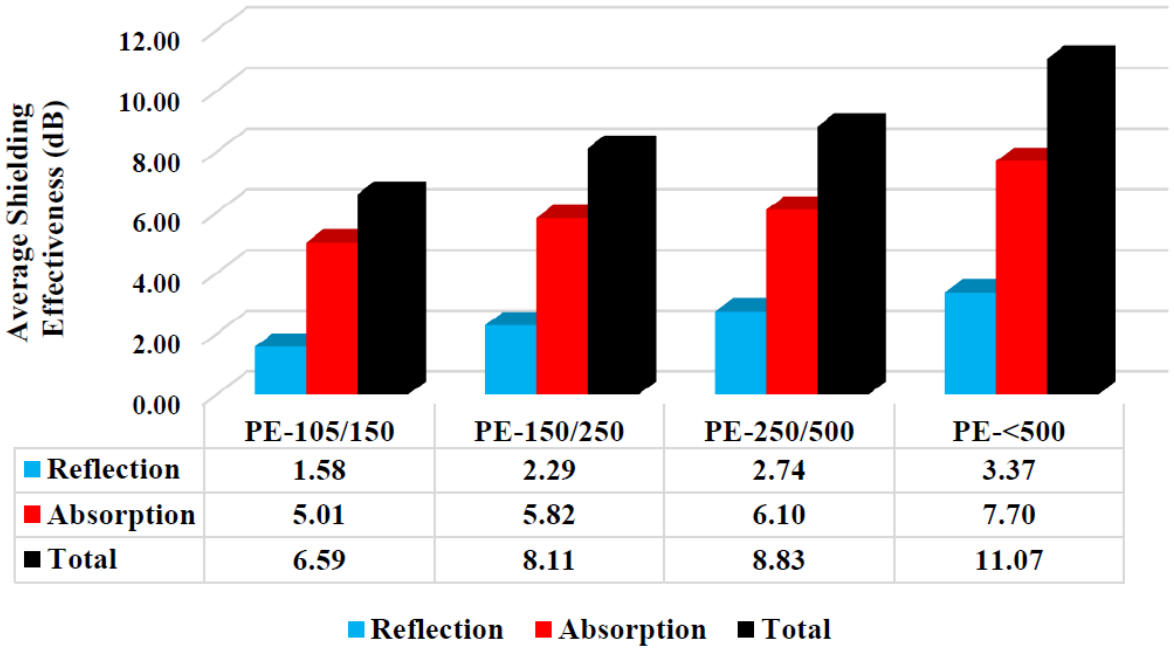


Figure 4. Absorption, reflection and total shielding effectiveness of the segregated HDPE/CB composites in the X-band frequency.



	PE-105/150	PE-150/250	PE-250/500	PE-<500
Reflection	1.58	2.29	2.74	3.37
Absorption	5.01	5.82	6.10	7.70
Total	6.59	8.11	8.83	11.07

Figure 5. Average total, absorption, and reflection EMI shielding effectiveness of PE/CB segregated composites in the X-band region versus polymer particle size.

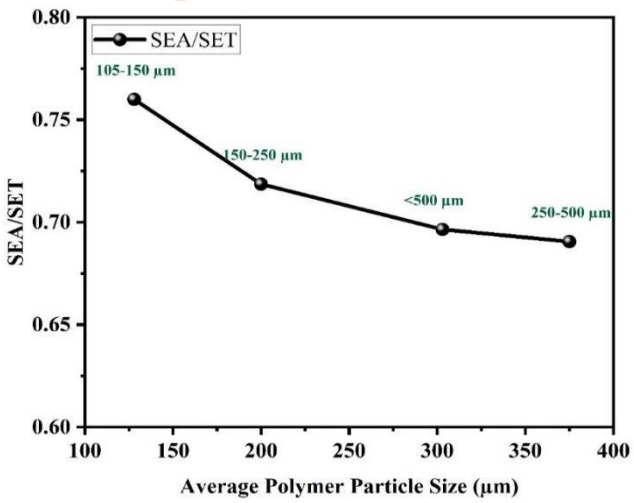


Figure 6. Variation of absorption fraction as a function of initial average polymer particle size in segregated HDPE/CB composites.

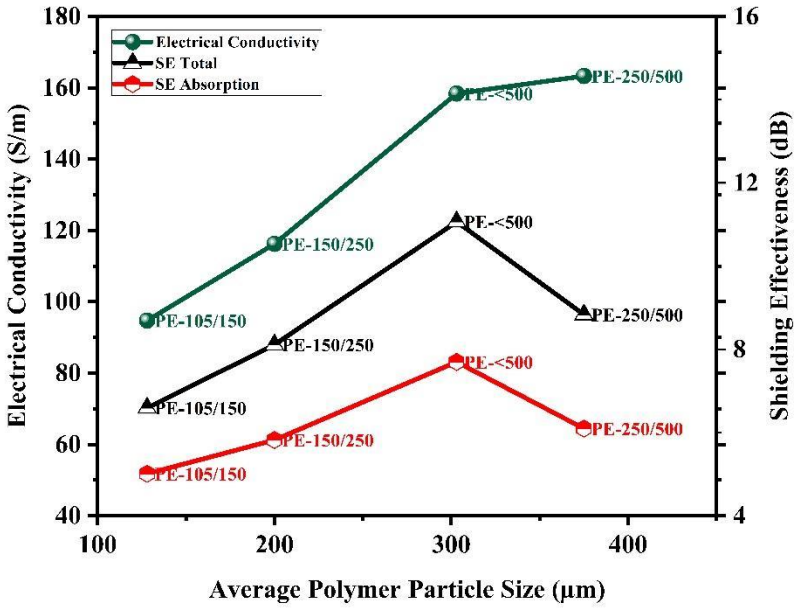


Figure 7. Variation of electrical conductivity and shielding effectiveness with the initial polymer particle size, averaged assuming a linear particle size distribution within the specified range.

Tables

Table1. Mesh specifications used for HDPE powder particle size separation.

Mesh number	140	100	60	35
Polymer Powder Size (micron)	105	150	250	500

Table2. Sample coding and electrical conductivity of the HDPE/CB composites

Sample code	Polymer paticle (µm)	Electrical conductivity (S/m)
94.7	105-150	PE5C-105/150
116.2	150-250	PE5C-150/250
163.3	250-500	PE5C-250/500
158.4	<500	PE5C-<500