Microwave-Assisted Sintering to Rapidly Construct a Segregated Structure in Low-Melt-Viscosity Poly(Lactic Acid) for Electromagnetic Interference Shielding

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ABSTRACT: Formation of a segregated structure in conductive polymer composites is one of the most effective strategies for achieving good electrical conductivity and electromagnetic interference (EMI) shielding performance. Nevertheless, for low-melt-viscosity poly(lactic acid) (PLA), intense molecular motion occurs at the molding temperature, which is detrimental to the fixation of the conductive networks. In this study, a novel molding technique assisted by microwave heating was proposed to construct a segregated structure in a PLA/carbon nanotube (CNT) composite. The coating layer of CNTs acted as the microwave absorber and caused intense localized heating of PLA surfaces upon microwave irradiation. The surface temperature of the PLA granule was precisely regulated by adjusting the coated CNT content, microwave power, and irradiation time. Thus, the coated granules were softened and fused at an optimal sintering zone, which effectively hindered the excessive migration of CNT strips into the interior of PLA phases, and a majority retained the original CNT network in the molded composite. Meanwhile, benefiting from microwave sintering, sufficient chain diffusion and entanglement occurred in the interfacial regions, enhancing the adhesion strength among the neighboring PLA phases. The prepared PLA/CNT composite with only 5.0 wt % CNTs exhibited a high electrical conductivity of 16.3 S/m and an excellent EMI shielding effectiveness (EMI SE) of 36.7 dB at a frequency of 10.0 GHz. The results indicate that microwave-assisted sintering might be a promising alternative for constructing a segregated structure in low-melt-viscosity polymers.

1. INTRODUCTION

Nowadays, the global popularization of portable electronic devices brings about serious electromagnetic radiation, which not only profoundly endangers human health but also threatens national security.1−3 In view of this, it is urgently needed to develop novel and highly efficient electromagnetic interference (EMI) shielding materials. Among various EMI shielding materials, conductive polymer composites (CPCs) possess unique advantages (e.g., low density, tunable performance, and good processability), and they have been recognized as promising candidates for EMI shielding applications.4−6 Recently, constructing the segregated conductive networks in polymer composites has been proven to be an effective strategy to improve filler utilization efficiency and provide CPCs with simultaneously enhanced electrical conductivity and EMI shielding performance.7−9 Yu et al.10 innovatively created the oriented segregated networks in ultrahigh molecular weight polyethylene (UHMWPE)/carbon nanotube (CNT) composites via solid-phase stretching. Only 2.0 wt % CNTs endowed the composite with an EMI shielding effectiveness (EMI SE) of 35.5 dB at a draw ratio of 6. In Jia’s work,11 waste tire rubber (WTR) was converted into a low-cost, efficient, and flexible EMI shielding material by designing a typical segregated structure in WTR/CNT composites. The segregated WTR composite containing 5.0 wt % CNTs showed an excellent electrical conductivity of 109.3 S/m and an EMI SE as high as 66.9 dB, superior to most of CNT-based CPCs. In short, the cell-like structure with conductive layers as cell walls provides abundant interfaces that can effectively reflect, scatter, and absorb the microwave (MW), thus endowing the segregated CPCs with excellent EMI shielding performance.4,10 Besides, incorporating nanopores,12 microsphere,13 and solid phase14,15 in CPCs to create multiple interfaces enhances the EMI shielding performance caused by the multiple reflection and/or scattering. Some novel materials, such as MXene,16,17 conductive ink,18 silver nanowire (AgNW),19,20 and carbonyl
iron–nickel alloy powder\textsuperscript{21} were also used to induce excellent EMI shielding effectiveness for CPCs.

However, it should be noted that the segregated networks always form in polymers with high melt viscosity, including UHMWPE\textsuperscript{22–24} and cross-linked polymers (e.g., cross-linked POE\textsuperscript{25}, natural rubber\textsuperscript{26}, etc.). When it comes to low-melt-viscosity and semi-crystalline polymers, the construction of a segregated structure is still a challenge because of the fact that conductive fillers tend to intermix into the polymer phases rapidly once the molding temperature exceeds the melting point of the polymer matrix.\textsuperscript{27} Furthermore, limited by the traditional heating method, heat transfers from the surface to the center, showing low heating efficiency and uneven temperature distribution. To address these issues, an advanced molding method (e.g., solid phase molding\textsuperscript{28}), structural design (e.g., double percolation conducting network\textsuperscript{25,30}), and cross-linking technique (e.g., chemically cross-linked elastomers\textsuperscript{31}) were proposed to form a segregated structure in CPCs. Nevertheless, these strategies always involved high-pressure operation and a complex and multistep fabrication process. It is necessary to develop novel, facile, and energy-saving methods for constructing the perfect segregated structure in CPCs, particularly for these low-melt-viscosity polymers.

MW with a frequency ranging from 300 MHz to 300 GHz is one of the most commonly used electromagnetic waves.\textsuperscript{32} When the MW absorber is exposed to MW irradiation, it will absorb MW and convert the energy into heat.\textsuperscript{33–35} Because of its unique energy-transforming characteristic, MW heating shows enormous potential in polymer materials processing. Up to now, MW heating technology has been widely applied to prepare some functional composites.\textsuperscript{36–42} For instance, Sweeney et al.\textsuperscript{43} utilized intense localized heating of CNTs to further weld the interfaces of 3D-printed thermoplastic parts upon exposure to MW irradiation, and the results indicated that the weld fracture strength was enhanced by 275% after MW irradiation. MW heating could be used to effectively overcome the nonuniformity and thickness restriction of conventional heating methods. In Rezvanpanah’s work,\textsuperscript{44} CNTs acted as the MW absorber and caused a rise in the local temperature of the polystyrene/carbon dioxide (PS/CO\textsubscript{2}) system, allowing local PS softening and CO\textsubscript{2} foaming in a one-step process. Compared with traditional thermal curing, MW curing provided uniform temperature distribution and reduced energy consumption and cost of preparing high-performance carbon fiber-reinforced polymer composites.\textsuperscript{45} More importantly, MW possesses selective heating behavior for materials depending on different values of dielectric loss. Generally, most thermoplastic polymers show an extremely weak interaction with MW, while the carbon materials, especially CNTs and graphene, are the excellent absorbents of MW and can be intensely heated upon exposure to MW radiation.\textsuperscript{33,37} By utilizing a different response behavior of polymers and carbon materials, selective heating occurs at the interfacial regions in their binary system via converting the MW energy into heat by MW absorbers. Similar to the traditional metal soldering process, CNTs play the role of a solder to provide vigorous, targeted heating and melt the polymer surface under the MW field. Thus, the composite parts with strong interfacial bonding can be manufactured by MW-assisted molding.

Poly(lactic acid) (PLA), a typical low-melt-viscosity semi-crystalline polymer,\textsuperscript{46} cannot meet the requirements as the suitable matrix in segregated CPCs prepared by the conventional hot compression molding. In this study, we proposed a new strategy assisted by MW heating to avoid the conductive fillers migrating into the interior of PLA during the molding process. A CNT, an excellent MW absorbent, was selectively distributed on the surface of PLA granules. The coating layer of CNTs induced intense heating under MW irradiation, and heat transfer from CNTs to the surfaces of PLA granules occurred in the interfacial regions. When the surface temperature of polymer particles increased to around 120 °C, a slight thermal deformation occurred and the gaps between particles gradually diminished when external compaction was applied, thus forming a dense segregated conductive network structure. Nevertheless, the weak interface bonding between adjacent polymer granules will always appear, and it is virtually needed to conduct heat treatment above the melting point to further enhance the mechanical properties. Herein, the fabrication process was divided into two steps. Initially, the processing condition of high pressure as well as temperature between $T_g$ and $T_m$ was chosen to densify the polymer. In this step, migration of CNTs can be suppressed because of the high melt strength. Second, the processing condition of low pressure as well as temperature above $T_m$ was chosen to improve interface bonding. Finally, the PLA/CNT composites featured enhanced mechanical properties, good electrical conductivity, and EMI SE. This facile technology showed great potential in constructing segregated conductive networks in low-melt-viscosity polymers.

2. RESULTS AND DISCUSSION

2.1. MW Sintering for Segregated PLA/CNT Composites. PLA, an important biodegradable polymer, possesses a glass transition temperature ($T_g$) of 65 °C and a melting point ($T_m$) of 155.3 °C. For the preparation of the segregated CPCs, polymers with high melt strength are required to prohibit the intense intermixing between the conductive fillers and polymer domains. Moreover, it is desirable to minimize energy

![Figure 1. Heating behavior of PLA/CNT (5 wt %) complex powder as a function of MW power and irradiation time (a) and infrared thermal image of samples during a compaction procedure (b) and sintering procedure (c).](https://dx.doi.org/10.1021/acsomega.0c03704)
consumption for sintering the samples from an economic point of view. Therefore, it is necessary to exploit the optimal MW power and irradiation time for MW-assisted molding. The temperature rise of the complex granule is also related to the content of coated CNTs. According to the results obtained in our previous work, the higher mass fraction of CNTs caused a faster heating rate and more generated heat upon exposure to MW irradiation. The surface temperature of coated granules with 5% CNTs was detected by an infrared thermal imager at different MW powers and irradiation times, and the related results are illustrated in Figure 1. The surface temperature of the complex particle increased significantly with the increase of MW power or irradiation time. The MW power and radiation time are the critical parameters for sintering quality of the complex powders. Considering the softening and melting behavior of PLA, the sintering zone of complex granules could be interpreted from the temperature diagram (Figure 1a). For 5 wt % CNT-coated granules, they should be compacted at 400 W for 10 s and then sintered at 500 W for 12 s. In this way, to obtain high-quality welding among granules, the optimal sintering condition for different samples, including compaction and sintering process, is given in Table 1. In contrast with other conventional strategies, the MW-assisted sintering proposed in this study involves a much shorter period (shown in Table 2), which might be a facile and energy-saving method for constructing the segregated structure in CPCs.

Table 1. Optimal MW Power and Irradiation Time for Different Complex Granules with Various CNT Contents

| CNT content (wt %) | compaction procedure | sintering procedure |
|--------------------|----------------------|---------------------|
| 1                  | 800 W, 15 s          | 1000 W, 20 s        |
| 3                  | 600 W, 10 s          | 750 W, 15 s         |
| 5                  | 400 W, 10 s          | 500 W, 12 s         |

Table 2. Some Strategies for Constructing the Segregated Structure in CPCs

| segregated CPCs       | method                  | process period | refs  |
|-----------------------|-------------------------|----------------|-------|
| CNTs/PLA              | MW-assisted sintering    | tens of seconds| our work|
| G-CNT/UHMWPE          | compression molding      | 10 min         | 1     |
| rGO/PS                | high-pressure solid-phase compression molding | 10 min | 7 |
| CNT/UHMWPE            | compression molding      | 5 min          | 10    |
| CNT/GTR               | compression molding      | 10 min         | 11    |
| Fe₃O₄-rGO/NR          | compression molding      | 8 min          | 26    |
| CNT/PLA               | compression molding      | 15 min         | 27    |

“Note: G-CNT, graphite-CNT; rGO, reduced graphene oxide; ground tire rubber; natural rubber.”

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2.2. Morphology and Microstructure. Figure 2a shows the scanning electron microscopy (SEM) morphology of 5.0 wt % CNT-coated PLA complex granules. The as-prepared PLA particles present irregular geometry with a mean size of 240 μm. The irregular geometry of PLA granules might induce obvious gaps among neighboring granules and reduce bulk density of composites during the sintering process. As shown in Figure 2b, numerous CNTs uniformly adhere to the surfaces of PLA granules, which favors the fabrication of segregated CNT/PLA composites in the following process.

Morphological comparison of samples prepared by two different methods was performed via optical microscope (OM) observation, providing the intuitive insights into the conductive networks of composites. The OM images shown in Figure 3 show the morphology of the 5.0 wt % segregated PLA/CNT composite slice prepared by the one-step method and two-step method. The black domains represent the CNT phases, while the white ones indicate the PLA phases because light cannot be transmitted through the CNT domains. As shown in Figure 3a, the CNT-conductive pathways became indistinct, indicating the destruction of the original CNT network. This could be due to the migration of CNT fillers from the surface into the interior of the PLA phases under intense MW sintering conditions. However, as shown in Figure 3b, the 2-SM samples displayed a perfect network configuration. The intermixing between CNT layers and PLA matrix was effectively hindered during the compaction process, and the segregated conductive pathways were retained in the composites.

To further reveal the superiority of the compaction process, the morphology and interfacial structure of samples prepared by different procedures were comparatively investigated via SEM observation. Because of the high molding temperature above the melting point of PLA, the obvious migration of CNTs into the low-viscosity matrix occurred and the predesigned CNT conductive pathways were deteriorated dramatically in one-step sintered samples. As shown in Figure 4, the thickness of the interfacial layer was up to 20 μm and CNTs were uniformly dispersed in the interfacial regions, which further demonstrated the intense migration of CNTs. In contrast, during the compaction stage, the surface temperature of polymer granules was controlled at around 120 °C. Thus, PLA particles were in a rubber state throughout the molding process.
When the particles were compressed by the external force, they experienced plastic deformation to form polyhedral PLA domains. From Figure 4b, the samples broke from the interface between polymer particles, indicating weak interface adhesion and CNTs enriched on the particle surface without penetrating into the interior of PLA granules. After the sintering procedure, the compaction sample underwent high-quality welding and acquired a typical segregated structure as shown in Figure 4c. A closer inspection revealed that the cryofractured surface became smooth, and the CNT fillers distributed at the interfacial regions among the adjacent PLA granules. The above results indicated that this two-step method is a promising alternative for the construction of the intact and compact conductive networks in the CPCs, particularly for these low-melt-viscosity polymers.

In terms of the traditional melt blending system, when CNTs are added into matrix resin, the majority of polymer chains adhere to the fillers, which will produce a significant barrier to the motion and lead to an obvious increase in the relaxation time and thus a shift of a terminal relaxation to low frequencies. Therefore, for more detailed information about the continuous segregated conductive network structure of fillers in composites, the frequency (Hz) dependence of the storage modulus ($G'$) and complex viscosity ($\eta^*$) for composites is tested and shown in Figure 5. Compared with that for neat PLA, a higher $G'$ was observed for s-CNT/PLA because of the presence of CNTs in the matrix. With the increasing CNT content, a more prominent increase of $G'$ for s-CNT/PLA composites at the low frequency region occurred, indicating the largely enhanced network density. Expectedly, the incorporation of CNTs induces a pronounced enhancement in $\eta^*$ (Figure 5b), confirming the formation of an unbroken network structure once again.

2.3. EMI Shielding Performance. Figure 6a shows the electrical conductivity of 1-MS and 2-MS composites as a function of CNT volume fraction. The electrical conductivity showed a dramatic increase with the increase in CNT content from 1 to 5 wt %. Furthermore, at a given CNT content, the electrical conductivity of the 2-MS sample was much higher than that of the 1-MS sample, for example, the electrical conductivity values of 1-MS$_5$ and 2-MS$_5$ are 2.2 and 16.3 S/m, respectively. Normally, the EMI shielding performance of the
CPCs is tightly related to the electrical conductivity. Figure 6b shows the EMI SE of the 1-MS and 2-MS composites as a function of CNT content. The composites exhibited gradually increasing EMI SE with CNT content. The EMI SE of the 2-MS composite with 5.0 wt % CNTs achieved 36.5 dB at a frequency of 10 GHz, much higher than the required value (20 dB) for commercial EMI shielding devices. In sharp contrast, the 1-MS5 composite showed an EMI SE value of 25.2 dB, lower than that of the 2-MS sample with the equivalent CNT content. The segregated samples exhibited higher EMI SE than the randomly distributed PLA/CNT composite with same CNT loading (5 wt %). We all know that EMI shielding mechanisms mainly include absorption (A), reflection (R), and transmission (T). Thus, it is of great significance to probe into the contributions of A, R, and T. Figure 6c shows the T, R, and A of the 2-MS composites as a function of CNT content. As CNT content increases, T always keeps at quite a low level. R exhibits a considerable increase with CNT content, which is attributed to the impedance mismatch at interfaces caused by the increasing conductivity. A gradually decreasing trend is observed for A values caused by the huge increase of R. Besides, the 2-MS composites have a much larger SEA than SER, showing an absorption-dominated mechanism. The skin depth is another important parameter to evaluate the EMI shielding properties. Figure 6e shows the skin depth of one-step and two-step composites as a function of CNT volume content at a frequency of 10 GHz.

Figure 7. Stress-strain curves of pure PLA and PLA/CNT composites with different CNT contents (a) and the corresponding tensile strength, modulus, and elongation at break of pure PLA and segregated PLA/CNTs composites (b).
shielding performance. As is known to all, high EMI shielding efficiency occurs at the thickness of the samples beyond the skin depth. That is to say, a higher skin depth value indicates worse EMI shielding performance of the materials. From Figure 6, the skin depth showed a decrease with the increase of CNT loading. In addition, the skin depth of the 2-MS composite was lower than that of the 1-MS composite at the same CNT content, which indicated that 2-MS composites possessed better EMI shielding performance. For example, the skin depth values of 1-MS and 2-MS composites with 5.0 wt % CNTs were 0.78 and 0.43 mm, respectively, which were much smaller than the thickness of the tested sample (2.0 mm).

2.4. Mechanical Properties. Apart from the high electrical conductivity and EMI SE, segregated PLA/CNT composites also showed excellent mechanical properties, which was important for the materials in practical shielding applications. Typical stress–strain curves of 2-SM composites and extruded 5% PLA/CNT composite are plotted in Figure 7a. As shown in Figure 7a, the tensile strength of extruded 5% PLA/CNT composite is higher than that of pure PLA, showing an obvious enhancement effect caused by the randomly distributed CNTs. For segregated CPCs prepared by conventional methods, the poor interfacial adhesion always deteriorates their mechanical properties. From the results shown in Figure 7a, 3 wt % can be considered to be the optimal CNT content for enhancement in mechanical properties. With CNT content increasing to 5 wt %, the tensile strength of 2-SM composites decreased from 60.3 to 55.8 MPa (Figure 7b). The incorporation of CNTs significantly enhanced the stiffness of the composites, for example, when the content of CNTs was 3%, tensile modulus achieved a high level of 2693 MPa, 28.5% higher than that of pure samples. In addition, the ductility characterized by the elongation at break declined from 4.8 to 3.2% (Figure 7b), which could be mainly attributed to the inherent brittleness of PLA.

3. CONCLUSIONS

In summary, we reported a facile, eco-friendly, and energy-saving strategy based on MW heating for the scalable fabrication of low-melt-viscosity polymer composites with perfect segregated conductive networks. Benefiting from this novel molding method, the integration of excellent EMI shielding performance and superior mechanical property were achieved in PLA/CNT composites. For example, with the CNT content of 5 wt %, the 2-MS sample was endowed with the maximum EMI SE of 36.7 dB, far superior to that of 1-MS. The tensile strength and tensile modulus of 2-SM, composites are 61.5 and 2693 MPa, respectively, higher than those of neat PLA, revealing an enhancing effect of CNT networks. In addition, PLA is derived from renewable sources and can be completely biodegraded. The proposed design principle is not only of profound significance to environmental protection, but also a great promotion to the fabrication of segregated CPCs based on low-melt-viscosity polymers.

4. EXPERIMENTAL SECTION

4.1. Materials. PLA (4032D) with a weight-average molecular weight ($M_w$) of 2.23 × 10^5 g/mol, a melting temperature ($T_m$) of 155.3 °C, and a density of 1.24 g/cm3 was purchased from Nature Works. CNTs (NC7000) were obtained from Nanocyl S.A., Belgium, and their average diameter and length were 9.5 nm and 1.5 μm, respectively.

4.2. Fabrication of the Segregated PLA/CNT Composites. The fabrication of the segregated PLA/CNT composite is schematically depicted in Figure 8. Initially, PLA granules were mixed with CNTs in a ball-milling machine (QM-3SP4, Nanjing NanDa Instrument Plant, Nanjing, China) at 500 rpm for 0.5 h to obtain CNT-coated PLA granules with CNT contents of 1.0, 3.0, and 5.0 wt % by weight ratio (0.60, 1.8, and 3 vol % by volume ratio). Then, the complex granules were filled into Teflon mold which can be found in our previous work, followed by MW sintering in an MW oven. The sintering process was divided into two steps. First step: the CNT-coated PLA complex granules were irradiated at a low MW power for a short period of time at the temperature of the surface of the complex granules between $T_g$ and $T_m$ of PLA (around 120 °C), and then the Teflon mold was quickly taken out and immediately exposed to an external force to compact the granules. Second step: the pre-impacted sample was further irradiated at a higher MW power for a longer time, and then the mold was taken out and pressed gently. Finally, the dense segregated PLA/CNT composite was obtained after ejection of the molded parts. For convenience, the obtained composites with different CNT loadings are denoted as 2-MS, 2-MS, and
As a control, the segregated PLA/CNT composites prepared by one-step MW sintering were also prepared (samples denoted as 1-MS and 1-MS). As a contrast, the pure PLA and randomly distributed PLA/CNT composite with 5 wt% CNT's were prepared through melting extrusion, followed by hot compression molding.

4.3. Temperature Distribution Test. The infrared thermal imaging of samples was measured using a Testo infrared thermal imager (Testo 870-2). In order to ensure the accuracy of the experiment, the images were collected immediately after MW was turned off.

4.4. SEM Observation. The micro-morphologies of the CNT-coated PLA complex granules and the cryo-fractured surface of PLA/CNT composites were observed under a SEM instrument (FEI Co., Ltd., USA) with an accelerating voltage of 4.7 kV. The fractured surface was prepared by immersing PLA/CNT composites in liquid nitrogen for 20 min and then broken. Before observation, PLA granules and the fractured surfaces were sputtered with a very thin golden layer.

4.5. Rheological Measurement. Rheological properties were analyzed using a stress-controlled dynamic rheometer AR2000ex (TA Instruments) with a parallel plate geometry (diameter: 25 mm) under the protection of a nitrogen atmosphere. The samples were dried in a vacuum at 60 °C for 6 h before measurement. Oscillatory frequency sweeps were performed from 0.01 to 100 Hz at 180 °C with a strain of 1%. All the specimens were disk-shaped with a diameter of 25 mm and a thickness of 2 mm.

4.6. Optical Microscopy Observation. For OM measurements, the PLA/CNT composites were cut into a film with a thickness of 20 μm; the film was observed under a Leica DM2500P OM, and then the OM images were taken by a pixel camera (Leica, USA). The thin PLA/CNT film tends to curl in order for us to observe a clear segregated structure; the film must be sandwiched between two glass slides to remain flat.

4.7. Electrical Property Test. The electrical conductivities were measured using a high-performance four-point probe instrument (RK-FA, Ningbo Ruikeweiye instrument Co., Ltd., China). In all cases, at least five sample replications were performed for each case, and the average values were reported.

4.8. EMI Shielding Test. The EMI shielding performance in the X-band (8.2–12.4 GHz) was characterized using an Agilent N5230 vector network analyzer (USA), with the APC-7 connector as a coaxial test cell, and the scattering parameters performed for each case, and the average values were reported.

4.9. Mechanical Property Test. Tensile strength testing of pure PLA and PLA/CNT composites (5 samples for each) were carried out by an Instron 5567 tensile instrument (Instron Co., Ltd, USA) with the sample size of 40 × 10 × 2 mm³ in dimension and the cross-head speed of 50 mm/min. The pure PLA samples were prepared by hot compression molding.

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REFERENCES

(1) Jia, L.-C.; Yan, D.-X.; Jiang, X.; Pang, H.; Gao, J.-F.; Ren, P.-G.; Li, Z.-M. Synergistic Effect of Graphite and Carbon Nanotubes on Improved Electromagnetic Interference Shielding Performance in Segregated Composites. Ind. Eng. Chem. Res. 2018, 57, 11929–11938.
(2) Li, J.; Peng, W.-J.; Fu, Z.-J.; Tang, X.-H.; Wu, H.; Guo, S.; Wang, M. Achieving high electrical conductivity and excellent electromagnetic interference shielding in poly(lactic acid)/silver nanocomposites by constructing large-area silver nanoplates in polymer matrix. Composites, Part B 2019, 171, 204–213.
(3) Liu, J.; Zhang, H.-B.; Sun, R.; Liu, Y.; Liu, Z.; Zhou, A.; Yu, Z.-Z. Hydrophobic, Flexible, and Lightweight MXene Foams for High-Performance Electromagnetic-Interference Shielding. Adv. Mater. 2017, 29, 1703267.
(4) Wu, H.-Y.; Zhang, Y.-P.; Jia, L.-C.; Yan, D.-X.; Gao, J.-F.; Li, Z.-M. Injection Molded Segregated Carbon Nanotube/Polypyrrole Composite for Efficient Electromagnetic Interference Shielding. Ind. Eng. Chem. Res. 2018, 57, 12378–12385.
(5) Yu, W.-C.; Xu, J.-Z.; Wang, Z.-G.; Huang, Y.-F.; Yin, H.-M.; Xu, L.; Chen, Y.-W.; Yan, D.-X.; Li, Z.-M. Constructing highly oriented segregated structure towards high-strength carbon nanotube/ultra-high-molecular-weight polyethylene composites for electromagnetic interference shielding. Composites, Part A 2018, 110, 237–245.
(6) Zeng, Z.; Jin, H.; Chen, M.; Li, W.; Zhou, L.; Zhang, Z. Lightweight and Anisotropic Porous MWCNT/PU Composites for...
Ultrahigh Performance Electromagnetic Interference Shielding. Adv. Funct. Mater. 2016, 26, 303–310.

(7) Yan, D.-X.; Pang, H.; Li, B.; Vajtai, R.; Xu, L.; Ren, P.-G.; Wang, J.-H.; Li, Z.-M. Structured Reduced Graphene Oxide/Polymer Composites for Ultra-Efficient Electromagnetic Interference Shielding. Adv. Funct. Mater. 2015, 25, 559–566.

(8) Pang, H.; Xu, L.; Yan, D.-X.; Li, Z.-M. Conductive polymer composites with segregated structures. Prog. Polym. Sci. 2014, 39, 1908–1933.

(9) Sun, R.; Zhang, H.-B.; Liu, J.; Xie, X.; Yang, R.; Li, Y.; Hong, S.; Yu, Z.-Z. Highly Conductive Transition Metal Carbide/Carbonitride (MXene)@poly styrene Nanocomposites Fabricated by Electrostatic Assembly for Highly Efficient Electromagnetic Interference Shielding. Adv. Funct. Mater. 2017, 27, 1702807.

(10) Yu, W.-C.; Zhang, G.-Q.; Liu, Y.-H.; Xu, L.; Yan, D.-X.; Huang, H.-D.; Tang, J.-H.; Xu, J.-Z.; Li, Z.-M. Selective electromagnetic interference shielding performance and superior mechanical strength of conductive polymer composites with oriented segregated conductive networks. Chem. Eng. J. 2019, 373, 556–564.

(11) Jia, L.-C.; Li, Y.-K.; Yan, D.-X. Flexible and efficient electromagnetic interference shielding materials from ground tire rubber. Carbon 2017, 121, 267–273.

(12) Li, J.; Chen, J.-L.; Tang, X.-H.; Cai, J.-H.; Liu, J.-H.; Wang, M. Constructing nanopoies in poly(oxy m ethylene)/multi-wall carbon nanotube nanocomposites via poly(l-lactide) assisting for improving electromagnetic interference shielding and mechanical enhancement. J. Coll. Interf. Sci. 2020, 565, 536–545.

(13) Cai, J.-H.; Li, J.; Chen, X.-D.; Wang, M. Multifunctional polydimethylsiloxane foam with multi-walled carbon nanotube and thermo-expandable microsphere for temperature sensing, microwave shielding and piezoresistive sensor. Chem. Eng. J. 2020, 393, 124805.

(14) Li, J.; Tan, Y.-J.; Chen, Y.-F.; Wu, H.; Guo, S.; Wang, M. Constructing multiple interfaces in polydimethylsiloxane/multi-walled carbon nanotube composites by the incorporation of cotton fibers for high-performance electromagnetic interference shielding and mechanical enhancement. Appl. Surf. Sci. 2019, 466, 657–665.

(15) Tan, Y. J.; Li, J.; Cai, J. H.; Tang, X. H.; Liu, J. H.; Hu, Z. Q.; Wang, M. Comparative study on solid and hollow glass microspheres for enhanced electromagnetic interference shielding in polydimethylsiloxane/multi-walled carbon nanotube composites. Composites, Part B 2019, 177, 107378.

(16) Li, L.; Cao, Y.; Liu, X.; Wang, J.; Yang, Y.; Wang, W. Multifunctional MXene-Based Fireproof Electro magnetic Shielding Films with Exceptional Anisotropic Heat Dissipation Capability and Joule Heating Performance. ACS Appl. Mater. Interfaces 2020, 12, 27350–27360.

(17) Jin, X.; Wang, J.; Dai, L.; Liu, X.; Li, L.; Yang, Y.; Cao, Y.; Wang, W.; Wu, H.; Guo, S. Flame-retardant poly(vinyl alcohol)/MXene multilayered films with outstanding electromagnetic interference shielding and thermal conductive performances. Chem. Eng. J. 2020, 380, 124275.

(18) Jia, L.-C.; Zhou, C. G.; Sun, W. J.; Xu, L.; Yan, D. X.; Li, Z. M.; Water-based conductive ink for highly efficient electromagnetic interference shielding coating. Chem. Eng. J. 2020, 384, No. 123368. DOI: 10.1016/j.cej.2019.123368.

(19) Jia, L.-C.; Zhang, G.; Xu, L.; Sun, W.-J.; Zhong, G.-J.; Lei, J.; Yan, D.-X.; Li, Z.-M. Robustly Superhydrophobic Conductive Cloth for Efficient Electromagnetic Interference Shielding. ACS Appl. Mater. Interfaces 2019, 11, 1680–1688.

(20) Jia, L.-C.; Xu, L.; Ren, F.; Ren, P.-G.; Yan, D.-X.; Li, Z.-M. Stretchable and durable conductive fabric for ultrahigh performance electromagnetic interference shielding. Carbon 2019, 144, 101–108.

(21) Ren, F.; Song, D.; Li, Z.; Jia, L.; Zhao, Y.; Yan, D.; Ren, P. Synergistic effect of graphene nanosheets and carbonyl iron–nickel alloy hybrid filler on electromagnetic interference shielding and thermal conductivity of cyanate ester composites. J. Mater. Chem. C 2018, 6, 1476–1486.

(22) Cui, C.-H.; Pang, H.; Yan, D.-X.; Jia, L.-C.; Jiang, X.; Lei, J.; Li, Z.-M. Percolation and resistivity-temperature behaviours of carbon nanotube–carbon black hybrid loaded ultrahigh molecular weight polyethylene composites with segregated structures. RSC Adv. 2015, 5, 61318–61323.

(23) Duan, H.; Xu, Y.; Yan, D.-X.; Yang, Y.; Zhao, G.; Liu, Y. Ultrahigh molecular weight polyethylene composites with segregated nickel conductive network for highly efficient electromagnetic interference shielding. Mater. Lett. 2017, 209, 353–356.

(24) Wang, Z.-G.; Gong, F.; Yu, W.-C.; Huang, Y.-F.; Zha, L.; Lei, J.; Xu, J.-Z.; Li, Z.-M. Synergetic enhancement of thermal conductivity by constructing hybrid conductive network in the segregated polymer composites. Compos. Sci. Technol. 2018, 162, 7–13.

(25) Liu, Y.-F.; Feng, L.-M.; Chen, Y.-F.; Shi, Y.-D.; Chen, X.-D.; Wang, M. Segregated polypolypropylene/cross-linked poly(ethylene-co-1-octene)/multi-walled carbon nanotube nanocomposites with low percolation threshold and dominated negative temperature coefficient effect: Towards electromagnetic interference shielding and thermists. Compos. Sci. Technol. 2018, 159, 152–161.

(26) Zhan, Y.; Wang, J.; Zhang, K.; Li, Y.; Meng, Y.; Yan, N.; Wei, W.; Peng, F.; Xia, H. Fabrication of a flexible electromagnetic interference shielding Fe3O4@reduced graphene oxide/natural rubber composite with segregated network. Chem. Eng. J. 2018, 344, 184–193.

(27) Cui, C.-H.; Yan, D.-X.; Pang, H.; Xu, X.; Jia, L.-C.; Li, Z.-M. Formation of a Segregated Electrically Conductive Network Structure in a Low-Melt-Viscosity Polymer for Highly Efficient Electromagnetic Interference Shielding. ACS Sustain. Chem. Eng. 2016, 4, 4137–4145.

(28) Wu, H.-Y.; Jia, L.-C.; Yan, D.-X.; Gao, J.-L.; Zhang, X.-P.; Ren, P.-G.; Li, Z.-M. Simultaneously improved electromagnetic interference shielding and mechanical performance of segregated carbon nanotube/polypolypropylene composite via solid phase molding. Compos. Sci. Technol. 2018, 156, 87–94.

(29) Zhang, K.; Li, G.-H.; Feng, L.-M.; Wang, N.; Guo, J.; Sun, K.; Yu, K.-X.; Zeng, J.-B.; Li, T.; Guo, Z.; Wang, M. Ultralow percolation threshold and enhanced electromagnetic interference shielding in poly(l-lactide)/multi-walled carbon nanotube nanocomposites with electrically conductive segregated networks. J. Mater. Chem. C 2017, 5, 9359–9369.

(30) Ren, F.; Li, Z.; Xu, L.; Sun, Z.; Ren, P.; Yan, D.; Li, Z. Large-scale preparation of segregated PLA/carbon nanotube composite with high efficient electromagnetic interference shielding and favourable mechanical properties. Composites, Part B 2018, 155, 405–413.

(31) Li, T.; Ma, L.-F.; Bao, R.-Y.; Qi, G.-Q.; Yang, W.; Xie, B.-H.; Yang, M.-B. A new approach to construct segregated structures in thermoplastic polyolefin elastomers towards improved conductive and mechanical properties. J. Mater. Chem. A 2015, 3, 5482–5490.

(32) Wang, C. Y.; Chen, T. H.; Chang, S. C.; Cheng, S. Y.; Chin, T. S. Strong Carbon-Nanotube-Polymer Bonding by Microwave Irradiation. Adv. Funct. Mater. 2007, 17, 1979–1983.

(33) Zhang, M.; Fang, S. I.; Zakhidov, A. A.; Lee, S. B.; Alley, A. B.; Williams, C. D.; Atkinson, K. R.; Baughman, R. H. Strong, transparent, multifunctional, carbon nanotube sheets. Science 2005, 309, 1215–1219.

(34) Vázquez, E.; Prato, M. Carbon Nanotubes and Microwaves: Interactions, Responses, and Applications. ACS Nano 2009, 3, 3819–3824.

(35) Qin, F.; Brousseau, C. A review and analysis of microwave absorption in polymer composites filled with carbonaceous particles. J. Appl. Phys. 2012, 111, 061301.

(36) Xu, X.; Wang, X.; Liu, W.; Zhang, X.; Li, Z.; Du, S. Microwave curing of carbon fiber/bismaleimide composite laminates: Material characterization and hot pressing pretreatment. Mater. Des. 2016, 97, 316–323.

(37) Xie, R.; Wang, J.; Yang, Y.; Jiang, K.; Li, Q.; Fan, S. Aligned carbon nanotube coating on polyethylene surface formed by microwave radiation. Compos. Sci. Technol. 2011, 72, 85–90.

(38) Wang, C. Y.; Chen, T. H.; Chang, S. C.; Chin, T. S.; Cheng, S. Y. Flexible field emitter made of carbon nanotubes microwave welded onto polymer substrates. Appl. Phys. Lett. 2007, 90, 103111.
Sun, X.; Yu, J.; Wu, G. Study on microwave welding of polypropylene by carbon nanotube. *Integr. Ferroelectr.* 2019, 197, 16–22.

Sun, X.; Wu, G.; Yu, J.; Du, C. Efficient microwave welding of polypropylene using graphite coating as primers. *Mater. Lett.* 2018, 220, 245–248.

Song, S. A.; Oh, H. J.; Kim, B. G.; Kim, S. S. Novel foaming methods to fabricate activated carbon reinforced microcellular phenolic foams. *Compos. Sci. Technol.* 2013, 76, 45–51.

Poyraz, S.; Zhang, L.; Schroder, A.; Zhang, X. Ultrafast Microwave Welding/Reinforcing Approach at the Interface of Thermoplastic Materials. *ACS Appl. Mater. Interfaces* 2015, 7, 22469–22477.

Sweeney, C. B.; Lackey, B. A.; Pospisil, M. J.; Achee, T. C.; Hicks, V. K.; Moran, A. G.; Teipel, B. R.; Saed, M. A.; Green, M. J. Welding of 3D-printed carbon nanotube-polymer composites by locally induced microwave heating. *Sci. Adv.* 2017, 3, No. e1700262.

Rezvanpanah, E.; Ghaffarian Anbaran, S. R.; Di Maio, E. Carbon nanotubes in microwave foaming of thermoplastics. *Carbon* 2017, 125, 32–38.

Li, N.; Li, Y.; Jelonnek, J.; Link, G.; Gao, J. A new process control method for microwave curing of carbon fibre reinforced composites in aerospace applications. *Composites, Part B* 2017, 122, 61–70.

Liu, Z.; Luo, Y.; Bai, H.; Zhang, Q.; Fu, Q. Remarkably Enhanced Impact Toughness and Heat Resistance of poly(L-Lactide)/Thermoplastic Polyurethane Blends by Constructing Stereocomplex Crystallites in the Matrix. *ACS Sustain. Chem. Eng.* 2016, 4, 111–120.

Feng, D.; Wang, Q.; Xu, D.; Liu, P. Microwave assisted sinter molding of polyetherimide/carbon nanotubes composites with segregated structure for high-performance EMI shielding applications. *Compos. Sci. Technol.* 2019, 182, 107753.

Wu, K.; Lei, C.; Huang, R.; Yang, W.; Chai, S.; Geng, C.; Chen, F.; Fu, Q. Design and Preparation of a Unique Segregated Double Network with Excellent Thermal Conductive Property. *ACS Appl. Mater. Interfaces* 2017, 9, 7637–7647.

Gupta, T. K.; Singh, B. P.; Dhakate, S. R.; Singh, V. N.; Mathur, R. B. Improved nanoindentation and microwave shielding properties of modified MWCNT reinforced polyurethane composites. *J. Mater. Chem. A* 2013, 1, 9138–9149.