Responses to Reviewers' Comments and Revisions on the Manuscript

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We sincerely appreciate the Reviewers for these constructive remarks and useful suggestions. We addressed each comment below and highlighted where the changes were made in manuscript with blue color. We wish the revised manuscript could meet the requirements of Nanomaterials. The point-by-point responses and revisions are as follows.

Response to Reviewer 1 Comments

Point 1: Title: The title "Desirable Mechanical Properties…by Incorporating Nanosized Carbon Black" should be reconsidered. From Fig. 6, the tensile strength and elongation at break of UHMWPE/CCB composites have not improved much, and the mechanical properties of most composites have even deteriorated. And from SEM, the Carbon Black did not achieve nanosized dispersion in composites, so the title is inappropriate.

Response 1: We thank the Reviewer for his/her professional advice. In our work, the tensile strength and elongation at break of UHMWPE/h-CCB composites have not improved much as compared with pure UHMWPE. However, their tensile strength show a relatively high retention rate relative to those of UHMWPE/l-CCB composites with high CCB loading content. As displayed in Figure S1, the tensile strength of UHMWPE/CCB composites with low loading content possesses a distinctly increased trend, which is closely related to the similar SEM morphology of UHMWPE/CCB composites. We further analyzed the improved effectiveness and retention rate of the tensile strength of UHMWPE/h-CCB and UHMWPE/l-CCB composites in detail. It is clear that the retention rate of the tensile strength UHMWPE/h-CCB_{10} composites is 96.4 %, which is higher than of UHMWPE/1-CCB_{10} composites (50.8%). Moreover, we also found high loading l-CCB particles are more liable to deteriorate the mechanical properties of the segregated conducting UHMWPE-based composites compared with the high loading h-CCB particles at the same filler content. These results show the segregated conducting UHMWPE based composites with high loading content beyond 10 wt% have the acceptable mechanical properties as well, which can meet the requirement of commercial electromagnetic interference (EMI) shielding material applications *(High Perform. Polym. 2020, 32,1140-1149; Polym. Eng. Sci. 2020, 60, 2414-2427; Composites, Part B, 2017, 119, 41-56).* As suggested by the Reviewer, we have changed our title to "Endowing Acceptable Mechanical Properties of Segregated Conductive Polymer Composites with Enhanced Filler-Matrix Interfacial Interactions by Incorporating High Specific Surface Area Nanosized Carbon Black".

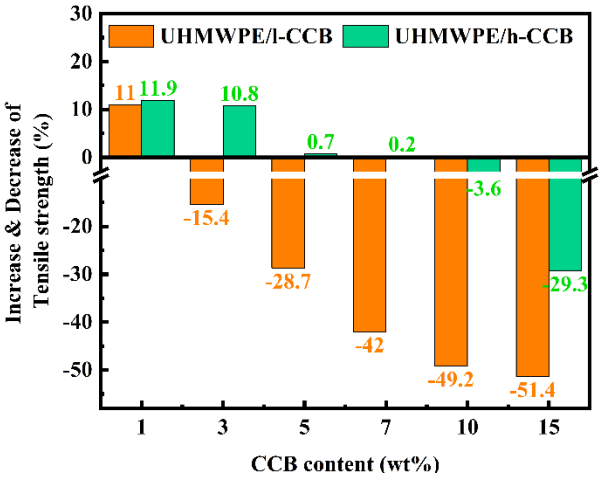


Figure S1 Comparison of increase and decrease trend of tensile strength for UHMWPE/l-CCB15 and UHMWPE/h-CCB composites

Point 2: Introduction: The authors mentioned that "The segregated conductive pathway has proven to outperform in enlarging conductive loss mechanism, particularly for polymer-based EMI shielding materials". The topic background of the manuscript needs to be lighted further to broaden the impact, related literature about constructing segregated conductive pathway like: Composites Part A: Applied Science and Manufacturing 2021): 106376 is recommended to cite

Response 2: We thank the Reviewer for our work's comment. To address the concern of the Reviewer, we have summarized the advantages of the segregated conductive pathway and cited the related literature about constructing segregated conductive pathways in the introduction section to broaden the impact, and forecasts the future challenges and opportunities of polymer-based EMI shielding materials *(Composites, Part A Appl. Sci. Manuf. 2021: 106376； Prog. Polym. Sci. 2014, 39, 627-655； Prog. Polym. Sci. 2014, 39, 1908-1933)* in Line 65, Page 2.

Point 3: Results: "In Figure 4(d-f), as the amount of h-CCB increases from 0.5 to 3.0 wt%, h-CCB particles remain orderly dispersed at the interfaces of UHMWPE granules instead of penetrating their interior", "And the h-CCB lamella structures appear a compact segregated structure with different thicknesses, Figure 4(f)". These conclusions cannot be obtained from Fig. 4, the SEM images of UHMWPE/l-CCB and UHMWPE/h-CCB with different filler content are very similar, which cannot prove the conclusion obtained by the authors. The authors are suggested to focus more on analyzing the Raman mapping images at different magnification to get more reliable conclusions.

Response 3: We thank the Reviewer for our work's comment. As suggested by the Reviewer, we have tried our best to modify these conclusions in the revised manuscript. The modified conclusions are as follow: In Figure 4(d-f), as the amount of h-CCB increases from 0.5 to 3.0 wt%, we observed that both h-CCB and l-CCB particles were orderly dispersed at the interfaces of UHMWPE granules, which is attributed to the strong volume exclusion effect of the UHMWPE matrix with high viscosity. Moreover, the SEM morphology images of UHMWPE/l-CCB and UHMWPE/h-CCB with different filler content are very similar, and no voids and interfacial defects are found. These results suggested that UHMWPE/l-CCB and UHMWPE/h-CCB composites have excellent mechanical properties at low loading content. (Line 260-267, Page 7). Moreover, we further focus more on analyzing the Raman mapping

images of UHMWPE/h-CCB0.5 composite at a different magnification of 10x (a) and 50x(a) objectives to get more reliable conclusions, as presented in Figure S2. It is found that the segregated h-CCB lamella structures appear a compact segregated structure and interfacial bonding layer with different thicknesses, its h-CCB lamella thickness mainly depends on the content of h-CCB as observed on the cross-sectional surface of UHMWPE/h-CCB composites as shown in Figure 5(c-d) (Line 324-326, Page 9). In Figure 5(c-d), as the h-CCB content increases from 1 wt% to 3 wt%, the thickness of the compact segregated h-CCB lamella increase from 4.44 μm to 22.65 μm, while the interfacial bonding layer is slightly enhanced, but the improved trend is no obvious (Line 296-303, Page 8). And such plentiful h-CCB nanoparticles as enhanced transition layer is positively contributed to the retention of the mechanical properties of high-loading polymeric-based composites. Hence high-loading polymeric-based composites with acceptable mechanical performance are mainly dependent on filler-matrix interface structure and filler distribution.

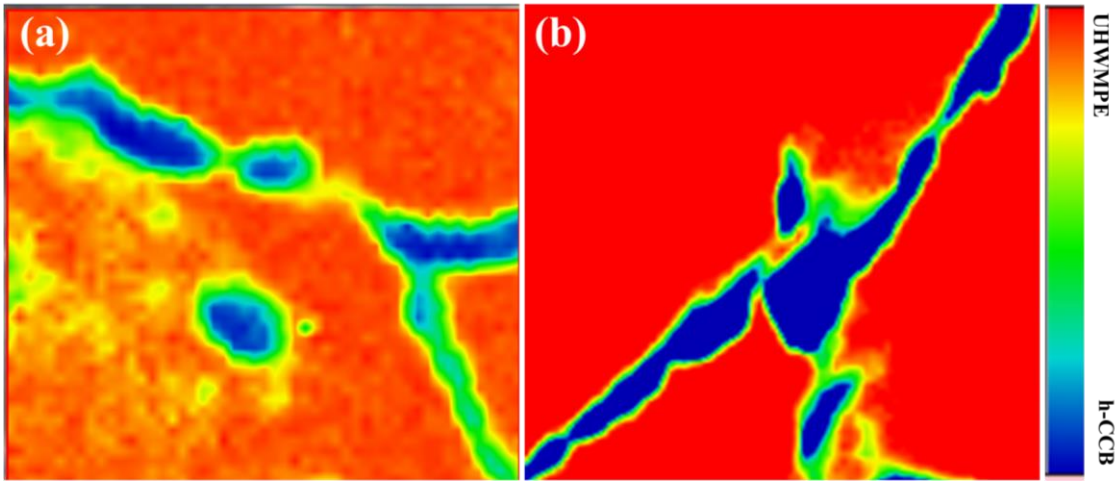


Figure S2 Raman mapping images of UHMWPE/h-CCB0.5 composite at a different magnification of 10x (a) and 50x(a) objectives, the scan area of Raman mapping images is 50×50 μm.

Point 4: The unit of electrical conductivity is S/m in the context in section 3.4, but the unit of electrical conductivity is S/cm in Fig. 8. The unit should be consistent in the paper.

Response 4: We thank the Reviewer for our work's comment. We have revised the unit of electrical conductivity (S/cm) in the context of section 3.4 (Line 449-452, Page 12).

Point 5: There are some relevant literature to be cited, such as Composites Part A: 2021, 145, 106376; Polymer, 2011, 52 (18), 4001-4010; Nanotechnology, 2013, 24 (12), 125704.

Response 5: We thank the Reviewer for his/her comment. Some relevant works have been cited in the introduction section, such as Composites Part A: Applied Science and Manufacturing, 2021, 145, 106376; Polymer, 2011, 52 (18), 4001-4010; Nanotechnology, 2013, 24 (12), 125704. (Line 62-65, Page 2).