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Phase Structure Formed by Rubber and Rigid Particles in a Polymer and Its Effect on the Composite Rigidity and Toughness

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Abstract Various phase structures can be formed by rubber and rigid particles in a polymer. Which is more beneficial to the impact toughness and rigidity, as well as their balance. This has been unclear until now. In this study, polymer composites with typical three-phase structures, that is, hard core-soft shell particles, soft core-hard shell particles, and two types of separated particles, were studied theoretically. The calculated results indicate that the soft core-hard shell structure is the best for enhancing the modulus in these three-phase structures, whereas the hard core-soft shell structure is the worst. On the other hand, both core-shell structures are beneficial for enhancing toughness, whereas the two types of particle-separated structures are not. These theoretical results are supported by the experimental results for polypropylene (PP), ethylene-propylene-diene monomer (EPDM) rubber, and SiO_2 composites.

Keywords Polymer composites; Polymer processing; Phase structure; Rigidity; Toughness

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INTRODUCTION

Rigidity and toughness are the two key parameters that determine whether a polymer can be used as an engineering material. Rubber and rigid particles are generally used to toughen and stiffen polymers, respectively.[1–4] However, improving the rigidity and toughness simultaneously using rubber or rigid particles alone is difficult.[5–9] It has been reported that the impact strength increases from 5 kJ/m^2 to 37 kJ/m^2 when 15 wt% ethylene-octene copolymer (POE) is added to polypropylene (PP), whereas the Young's modulus decreases from 1060 MPa to 670 MPa, which is 36.8% less than that of pure PP.[10] Introduction of rigid particles alone can lead to an increase in rigidity; however, it generally results in a decrease in toughness. Notably, in some special cases, polymers can be toughened by adding rigid particles.[2,11] However, ductile fracture can be obtained only at room temperature or above. Therefore, rubber and rigid particles have been used together to overcome the trade-off between rigidity and toughness, which has become a favorable topic in both polymer science and engineering.[12–17]

It is well known that the mechanical properties of a polymer composite strongly depend on its phase structure formed by rubber and rigid particles in the polymer matrix. Dubnikova *et al*. prepared PP/glass bead/ethylene-propylene rubber composites with various phase structures and reported that an optimal stiffness–toughness balance can be obtained by coating rigid particles with an elastomer shell.[18] Similarly, Wang *et al*. reported that both the impact toughness and rigidity of PP/POE/SiO_2 composites were synchronously enhanced when SiO_2 particles were surrounded by an ethylene-octene copolymer (POE) elastomer in the PP matrix. The notch impact strength and flexural modulus of the toughened PP can reach 44.6 kJ/m^2, which is 5.4 times greater than that of PP, and 1.6 GPa, which is 1.4 times greater than that of PP.[19] Moreover, Yang and Fu *et al*. reported a unique structure in which the majority of EPDM particles were surrounded by SiO_2 particles in the PP matrix, resulting in a dramatic increase in the Izod impact strength as the rubber content increased in the range of brittle–ductile transition (15 wt%–20 wt%).[20] Furthermore, Hajibabazadeh *et al*. reported that a phase structure in which rubber domains and nanoparticles were separately distributed in the PP matrix resulted in the lowest fracture parameters in the PP/EPDM/SiO_2 composite.[21] These experimental results clearly show that both the toughness and rigidity of polymer/rubber/rigid particle composites depend on their phase structures. However, the structure is more rigid. Which structure is more beneficial for the toughness. Which structure is more beneficial for balancing the toughness and rigidity. Such questions have not yet been answered. The purpose of this study is to answer these questions through theoretical and experimental investigations.

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EXPERIMENTAL

Raw Materials

Commercial-grade isotactic polypropylene (iPP) (marked as T30; Fujian Zhongjing Petrochemical. Co., Ltd.) with a melt flow index of 3.0 g/10min (230 °C, 2.16 kg) was used as the primary component. An ethylene-propylene-diene monomer (EPDM) (EP35, Japan Synthetic Rubber Co., Ltd.) was used. The Mooney viscosity was 83 (100 °C), the PP content was 43%, and the third monomer was ethylidene norbornene (ENB), hydrophilic silica (SiO_2, AEROSIL200) was purchased from Evonik Degussa, Germany), and the particle size ranged of 10–30 nm.

Preparation of PP/SiO_2/EPDM Composites

First, PP, SiO_2, and EPDM were dried in a vacuum oven at 80 °C for 4 h to remove the adsorbed moisture. To obtain different phase structures, one- and two-step processing methods were employed. In the one-step method, PP, EPDM, and SiO_2 were mixed by melting for 10 min. In the two-step method, PP and SiO_2 were first blended by melting for 6 min, and EPDM rubber was subsequently added and blended for 10 min. They were melt-blended in a HAAKE XSS-300 internal mixer (KCCK, Shanghai, China) at different weight ratios to prepare the PP/SiO_2/EPDM composites. Blending was performed at 60 r/min and 190 °C.

Mechanical Properties

The prepared samples were compression molded into 3.2 mm and 1 mm sheets using a plate vulcanizing machine (XLB, Qingdao, China) with a holding pressure of 10 MPa at 190 °C for approximately 5 min. The standard samples for the notched Izod impact test were rectangular bars with dimensions of 63.5 mm × 12.7 mm × 3.2 mm (length × width × thickness). The samples for the tensile test were cut into dumbbell shapes with dimensions of 50 mm × 4 mm × 1 mm (length × width × thickness). The notched Izod impact strengths of the composites were measured according to the ASTM D256-10 test method using an Izod impact test machine (XJUD-5.5; Jinjian Testing Instrument Co., Ltd., China). A 45° V-shaped notch (depth of 2.5 mm) was cut into samples using an autocycle test sample notching cutter. Tensile tests were performed on a universal tensile machine (Instron-1121, UK) at a cross-head speed of 50 mm/min following ASTM D638-14 at room temperature (23±2 °C), and the dimensions of the samples were 50 mm × 4 mm × 1 mm (length × width × thickness). The mechanical properties of each sample were determined using five replicates. All samples were maintained at 23 °C for 24 h before the test.

Microstructure of PP/SiO_2/EPDM Composites

Scanning electron microscopy (SEM, Thermo Scientific Apreo 2S HiVac) was used to observe the morphology of the prepared composites. The samples were immersed in liquid nitrogen and cryogenically fractured. The fracture surface was then etched with *n*-heptane at 60 °C for 12 h to clearly observe the EPDM phase dispersed in the composite. Finally, the etched fracture surface was dried and sputter-coated with gold powder before SEM measurement. The SEM measurements were performed at an acceleration voltage of 2.0 kV.

MODELS AND THEORIES

The composites we studied contained three components: a matrix polymer, rubber, and rigid particles. Their moduli were obtained based on the three-phase model developed by Takayanagi's two-phase model.[22,23] The schematic diagram of three typical phase structures formed by rubber or elastomer and rigid particles and the equivalent model for their moduli and the contribution of each phase to the modulus under uniaxial tension T is shown in Fig. 1.

First, we consider the case in which rubber or elastomer and rigid particles are separated in a polymer matrix, as shown in Fig. 1(a). The calculation of the Young's modulus of the composite can be simplified to that shown in Fig. 1(b) and further divided into different elements, as shown in Fig. 1(c), based on the phase relation in the tensile test. The two dispersed phases are assumed to be anisotropic, that is, the height and width are equal for each phase region, as indicated in Fig.1(b), and the area of each phase corresponds to its volume fraction, that is,

$$\varphi_\alpha = \alpha^2 \text{ and } \varphi_\beta = \beta^2 \tag{1}$$

$$\varphi = \varphi_\alpha + \varphi_\beta \tag{2}$$

where φ refers to the total volume fraction of α and β phases.

The relative modulus of the composite can be obtained from Fig. 1(c):

$$\frac{E_c}{E_m} = \left(1 - \alpha + \frac{\alpha - \beta}{(1-\alpha) + \alpha\frac{E_\alpha}{E_m}} + \frac{\beta}{(1-\alpha-\beta) + \alpha\frac{E_\alpha}{E_m} + \beta\frac{E_\beta}{E_m}}\right)^{-1} \tag{3}$$

where E_c, E_m, E_α and E_β are the moduli of the composite, matrix polymer, α phase, and β phase, respectively.

Second, we studied the phase structure in which rubber or elastomer particles were surrounded by rigid particles in a polymer matrix. This structure was obtained in a PP/EPDM rubber/SiO_2 rigid-particle system by Fu *et al*.[20] To perform quantitative calculations, we consider an ideal case, that is, rigid particles located near rubber particles, as shown in Fig. 1(d). Notably, encapsulated rubber particles can deform under stress. Similarly, the equivalent model of the modulus and contribution of each phase to the modulus under uniaxial tension T are shown in Figs. 1(e) and 1(f), respectively. The modulus of a composite with this phase structure can be obtained as follows:

$$\frac{E_c}{E_m} = \left(1 - \alpha + \frac{\alpha - \beta}{(1-\alpha) + \alpha\frac{E_\alpha}{E_m}} + \frac{\beta}{(1-\alpha) + (\alpha-\beta)\frac{E_\alpha}{E_m} + \beta\frac{E_\beta}{E_m}}\right)^{-1} \tag{4}$$

Third, we studied the phase structure in which rigid particles were covered by rubber or elastomer, forming core-shell particles, as shown in Fig. 1(g). This structure was obtained and studied in a PP/POE elastomer/SiO_2 rigid-particle system by Dizaji *et al*. and Wang *et al*.[19,24] For comparison with the above two-phase structures, the equivalent model for the modulus and contribution of each phase to the modulus under uniaxial tension T are given in Figs. 1(h) and 1(i), respectively. The modulus of a composite with this phase structure can be obtained as follows:

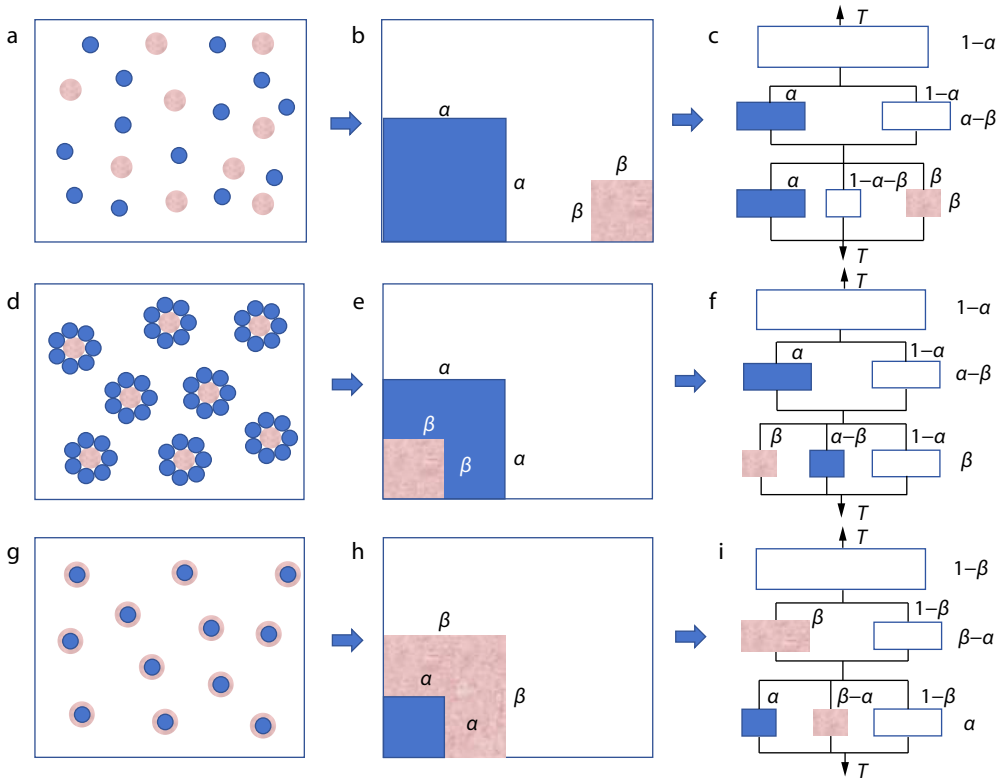

Fig. 1 Schematic diagram showing three typical phase structures formed by the rubber (pink circle) or elastomer and rigid particles (bule circle) and the equivalent model for their moduli and the contribution of each phase to the modulus under uniaxial tension T (connected in series). (a) Rubber or elastomer and rigid particles dispersed separately in a polymer matrix and its equivalent model for the modulus (b) and the contribution of each phase to the modulus (c); (d) Rigid particles near the rubber or elastomer particles in a polymer matrix and its equivalent model for the modulus (e) and the contribution of each phase to the modulus (f); (g) Rigid particles encapsulated by rubber or elastomer forming core-shell particles and its equivalent model for the modulus (h) and the contribution of each phase to the modulus (i).

$$\frac{E_c}{E_m} = \left(1 - \beta + \frac{\beta - \alpha}{(1-\beta) + \beta\frac{E_\beta}{E_m}} + \frac{\alpha}{(1-\beta) + (\beta-\alpha)\frac{E_\beta}{E_m} + \alpha\frac{E_\alpha}{E_m}}\right)^{-1} \quad (5)$$

THEORETICAL RESULTS AND DISCUSSION

In this study, we used PP/rubber/SiO_2 rigid particle composites as examples and set E_α/E_m = 90/1 (α refers to the SiO_2 phase) and E_β/E_m =1/300 (β refers to the rubber phase). From Eqs. (1)–(5), we can calculate the variation in the relative modulus (E_c/E_m) with respect to the total volume fraction of the two dispersed phases (φ) for the three-phase structures.

Rubber or Elastomer and Rigid Particles Dispersed Separately in a Polymer Matrix

The results calculated by Eqs. (1)–(3) are shown in Fig. 2. The modulus increased monotonically with φ if $\varphi_\alpha/\varphi_\beta$>1. However, the modulus increased to a peak value and then decreased with increasing φ if $\varphi_\alpha/\varphi_\beta$<1. Moreover, the peak position shifted in the higher-φ direction with increasing $\varphi_\alpha/\varphi_\beta$.

Let us now consider the toughness of the composites. In the case in which rubber or elastomer and rigid particles are dispersed separately in a polymer matrix, the increase in toughness is attributed mainly to rubber particles. Fu *et al*. reported that adding SiO_2 particles to pure PP leads to a reduction in toughness. On the other hand, the toughness increased with the addition of pure EPDM rubber, and a sharp brittle ductile transition occurred. The fracture is ductile if the EPDM content is greater than 15 wt%; otherwise, it is brittle. With respect to ductile fracture, that is, when the EPDM content was 30 wt%, the addition of SiO_2 had less effect on the toughness. Notably, the highest SiO_2 content added was only 5 wt%, that is, the results for higher SiO_2 contents have not been reported.[20]

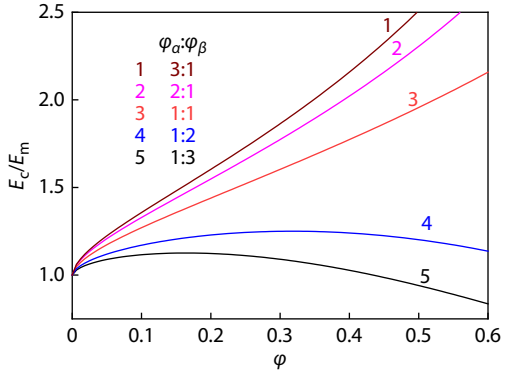

Fig. 2 Variation in the relative modulus (E_c/E_m) with respect to the total volume fraction of the two dispersed phases (φ) for various $\varphi_\alpha/\varphi_\beta$ ratios, in which E_α/E_m = 90 and E_β/E_m =1/300.

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Rigid Particles Near the Rubber or Elastomer Particles in a Polymer Matrix

The results calculated using Eqs. (1), (2) and (4) are shown in Fig. 3. The modulus increased monotonically with increasing φ for all $\varphi_\alpha/\varphi_\beta$ ratios, even when the ratio was as low as 1/3. Moreover, the modulus in Fig. 3 is higher than that in Fig. 2 for the same values of φ and $\varphi_\alpha/\varphi_\beta$. In Fig. 3, E_c/E_m can reach 2.52, whereas it is 1.77 in Fig. 2, for φ=0.4 and $\varphi_\alpha/\varphi_\beta$=1. The results indicate that the phase structure in which rigid particles are near the rubber particles is more effective in enhancing the composite rigidity than the structure in which the rubber and rigid particles are dispersed.

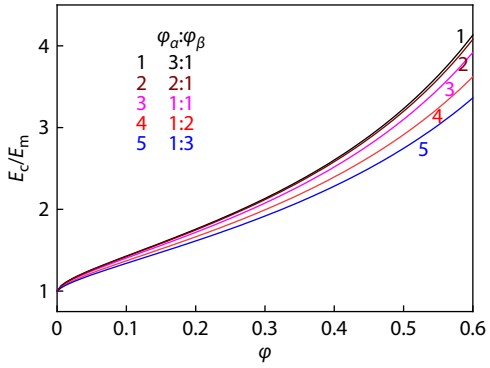

Fig. 3 Variation in the relative modulus (E_c/E_m) with respect to the total volume fraction of the two dispersed phases (φ) for various $\varphi_\alpha/\varphi_\beta$ ratios, in which E_α/E_m = 90 and E_β/E_m =1/300.

To determine the composite toughness, we considered rubber particles surrounded by rigid particles as spherical toughness modifiers. Based on previous theoretical and experimental studies, the moduli of the modifiers ($E_{modifier}$) should be less than 1/10 that of the matrix, that is,[25–27]

$$E_{modifier}/E_m \le 1/10 \quad (6)$$

Considering the modifier as a sphere composed of two phases (Fig. 1d), the modulus can be obtained as follows:

$$E_{modifier} = \left(\frac{V_\alpha}{E_\alpha} + \frac{(1-V_\alpha)}{E_\beta}\right)^{-1} \quad (7)$$

where V_α is the volume fraction of α phase in the modifier. Taking E_α/E_m = 90 and E_β/E_m =1/300, $V_\alpha \le$ 96.7% according to Eqs. (6) and (7). This means that the volume fraction of SiO_2 in the modifier should be less than 96.7% to effectively toughen PP. In contrast, as shown in Fig. 1(d), the diameter of the modifier is $2d_\alpha + d_\beta$, where d_α and d_β are the diameters of the SiO_2 and rubber particles, respectively. Therefore, the diameter of the modifier for such a structure should be larger than the diameter (d_α + rubber shell thickness) of the rubber-covered rigid particles, leading to a decrease in the toughening efficiency. Therefore, nanoscale SiO_2 particles should be used to increase the toughness. Fu *et al.* reported that EPDM and SiO_2 particles (10–30 nm in diameter) could form a phase structure in which the EPDM particles were surrounded by SiO_2 particles. The addition of SiO_2 particles can increase the toughness of the composites. However, the highest SiO_2 content was 5 wt%, and the SiO_2 particles tended to aggregate together. This indicates that obtaining such a phase structure is difficult for higher SiO_2 contents.

From the above results and discussion, the phase structure in which rigid nanoscale particles are near the rubber particles in a polymer matrix, as shown in Fig. 1(d), is more beneficial for enhancing rigidity and toughness.

Rigid Particles Encapsulated by Rubber or Elastomer, Forming Core-Shell Particles

The results calculated using Eqs. (1), (2) and (5) are shown in Fig. 4. As shown in Fig. 4(a), the modulus increased to a peak value and then decreased continuously with increasing φ for all $\varphi_\alpha/\varphi_\beta$ ratios. Moreover, both the peak position (φ_{peak}) and value increased with increasing $\varphi_\alpha/\varphi_\beta$, as shown in Figs. 4(b) and 4(c). Notably, it is quite difficult for E_c/E_m to reach 2 even for greater contents of core-shell particles if $\varphi_\alpha/\varphi_\beta$ is smaller than 5.

For comparison, the variation in the relative modulus (E_c/E_m) with respect to the total volume fraction of the two dispersed phases (φ) for the three-phase structures with $\varphi_\alpha/\varphi_\beta$=1 is shown in Fig. 5. A phase structure in which rubber particles are closely surrounded by rigid particles is the most effective for enhancing rigidity, whereas a phase structure with rigid particle-covered rubber is the least effective. Moreover, the difference in the rigidity of the three-phase structures increases with an increase in the total volume fraction of the two dispersed phases (φ).

The studied phase structures (Fig. 1) are ideal models. For example, a phase structure in which rigid particles are covered by rubber means that all rigid particles are covered by rubber. However, it was difficult to ensure that 100% of the rigid particles were covered. With an increase in the rigid particle content, some rigid particles are separated in the matrix, forming coexisting phase structures of covered rubber and two separated phases. As a result, the modules at higher SiO_2 contents should be higher than those predicted from the

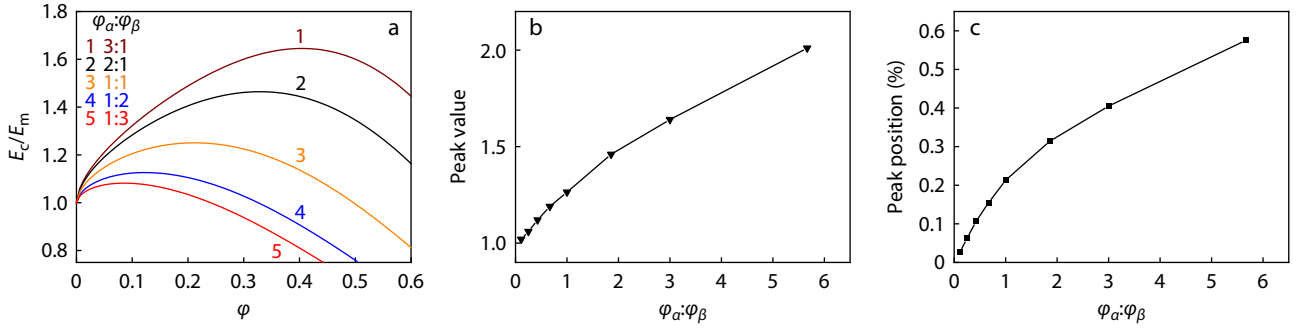

Fig. 4 Variation in the relative modulus (E_c/E_m) with respect to the total volume fraction of the two dispersed phases (φ) for various $\varphi_\alpha/\varphi_\beta$ ratios (a); variations in the peak position (b) and peak value (c) with $\varphi_\alpha/\varphi_\beta$.

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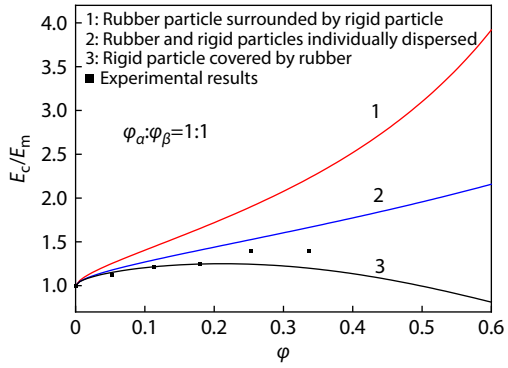

Fig. 5 Variation in the relative modulus (E_c/E_m) with respect to the total volume fraction of the two dispersed phases (φ) for the three phase structures in which $\varphi_\alpha/\varphi_\beta$=1. For the purpose of comparison, the experimental results of Fig. 6(a) in Ref. [19] are plotted.

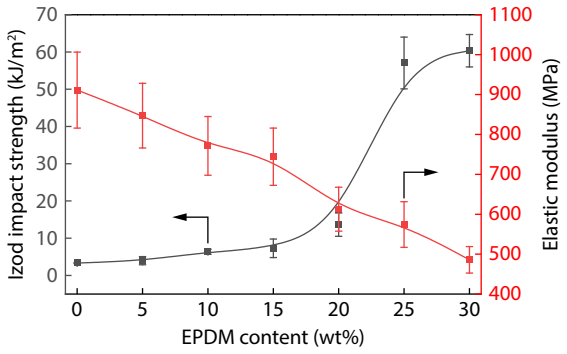

Fig. 6 Variation in the Young's modulus and notch Izod impact strength of the PP/EPDM blend with EPDM content.

model in which 100% of the rigid particles are covered by rubber. However, it should be lower than that predicted by the model in which the rubber and rigid particles are completely separated. For comparison with the experimental results, the experimental results in Fig. 6(a) in Ref. [19] are plotted in Fig. 5. The volume fraction or ratio was used in this theoretical study, whereas the weight fraction or ratio was used in the experiment. [19] For example, if the weight ratio of POE to SiO_2 is 1:3, the volume ratio of POE to SiO_2 is approximately 1:1, that is, φ_α:φ_β=1 in this study. From Fig. 5, it can be observed that the experimental results are in good agreement with the theoretical prediction (Line 3) at lower contents, indicating that most SiO_2 particles are covered by POE. However, the experimental results are clearly higher than the theoretical prediction (Line 3) at higher content, however, they are lower than the theoretical prediction (Line 2), indicating that more SiO_2 particles are individually separated in the PP matrix at higher SiO_2 contents.

For composite toughness, the modulus of the modifier, as shown in Fig. 1(g), should be less than 1/10 that of the matrix. Similarly, according to Eqs. (6) and (7), the volume fraction of SiO_2 in the modifier should be less than 96.7% to effectively toughen the PP. Moreover, both experimental and theoretical studies indicate that a soft shell and hard core are more beneficial for balancing the impact toughness and rigidity.[28,29] The notch impact strength and flexural modulus are 584.9 J/m and 953 MPa, respectively, for the PE core-EPR shell rubber particle-toughened PP and 609.6 J/m and 1100 MPa, respectively, for the PP core-EPR shell rubber particle-toughened PP.[28] However, the impact toughness and rigidity of the PP/POE/SiO_2 composite can reach 44.6 kJ/m^2 (approximately 446 J/m) and 1600 MPa, respectively, for the SiO_2 core-POE shell particle-toughened PP, indicating an excellent balance between the impact toughness and rigidity.[19]

EXPERIMENTAL RESULTS AND DISCUSSION

First, we experimentally studied the binary PP/EPDM system. The variation in the Young's modulus and notch Izod impact strength of the PP/EPDM blend with respect to the EPDM content is shown in Fig. 6. Young's modulus decreased from 911 MPa to 612 MPa as the EPDM content increased from 0 wt% to 20 wt%. However, the notch Izod impact strength tends to increase with increasing EPDM content, and a sharp brittle ductile transition occurs. The fracture is ductile if the EPDM content is greater than 20 wt%; otherwise, it is brittle, indicating that 20 wt% is the critical rubber content for brittle duction transition. In subsequent studies, the EPDM content in the PP matrix was fixed at 20 wt%, and the SiO_2 content was variable.

For the PP/SiO_2/EPDM composites, compared with those in the one-step processing, the SiO_2 particles in the two-step processing have a greater possibility of achieving separate distributions in the PP matrix. This affects the modulus and toughness of the composites, as described below. The notch Izod impact strength of the PP(80)/EPDM(20)/SiO_2 composite with respect to SiO_2 content is shown in Fig. 7(a). The intro-

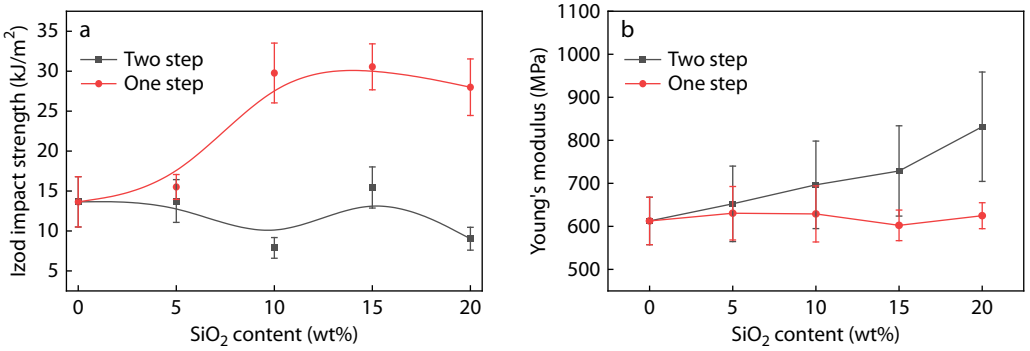

Fig. 7 Variations in notch Izod impact strength (a) and Young's modulus (b) of the PP(80)/EPDM(20)/SiO_2 composite with respect to the SiO_2 content.

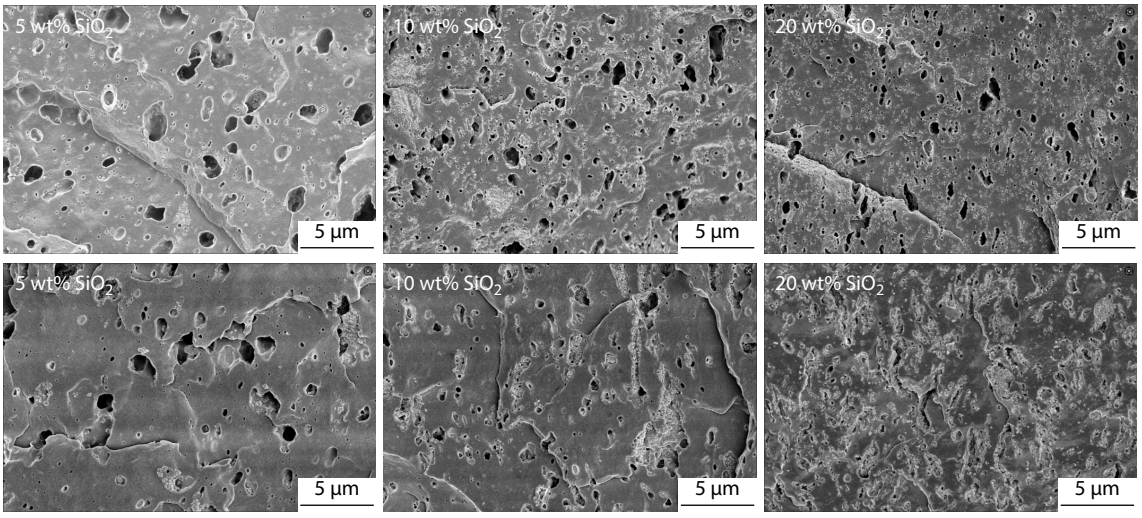

Fig. 8 SEM images of the cryofractured surfaces of the PP(80)/EPDM(20)/SiO_2 composites etched by n-heptane for different SiO_2 contents and melting methods (top row: two steps; bottom row: one step).

duction of SiO_2 can lead to an increase in the notch Izod impact strength in one step; however, this cannot be achieved in the two-step process. The variation in Young's modulus of the PP(80)/EPDM(20)/SiO_2 composite with respect to the SiO_2 content is shown in Fig. 7(b). The modulus of the composite prepared in two steps was obviously greater than that of the composite prepared in one step. As the materials and their compositions remained unchanged, this difference in moduli likely resulted mainly from the phase structure formed by EPDM and SiO_2 in the PP matrix.

To reveal the effect of the phase structure formed by the EPDM and SiO_2 on the mechanical properties of the PP matrix, SEM images of the cryofractured surfaces of the PP(80)/EPDM(20)/SiO_2 composites etched with *n*-heptane are shown in Fig. 8. More SiO_2 particles were separately distributed in the PP matrix for the composite prepared by the two steps, that is, blending PP and SiO_2 first. Therefore, the theoretical and experimental results (Figs. 2 and 7b) show that the modulus increases with increasing SiO_2 content for such phase structures. Moreover, such a phase structure is not beneficial for improving toughness, as indicated by the experimental results (Fig. 7a). However, the SiO_2 particles in the composite produced in one step were more likely to be encapsulated by rubber. Such a phase structure is beneficial for toughness, as shown in Fig. 7(a), but does not increase rigidity, as shown in Fig. 7(b). Moreover, as shown in Fig. 7(b), the modulus of the composite obtained in one step was less than that of the composite obtained in two steps, particularly at high SiO_2 content. This finding is in agreement with the theoretical results (Fig. 5). Finally, as shown in Fig. 8, an increasing number of SiO_2 particles tend to aggregate with increasing SiO_2 particle content. Therefore, the impact strength of the composite prepared in one step is clearly lower than that of the composite toughened by SiO_2 core-POE shell rubber particles but higher than that of the composite prepared in two steps.[19]

Finally, it is noted that the interface between the phases is not considered in this study based on the following considerations: Previous studies have shown that the brittle ductile transition can be determined by modifier size and content, modifier and matrix polymer properties, rather than the interfacial bonding between the phases;[6,30,31] comparing with the modifier size, the interface thickness is so small that its effect can be approximatively ignored.

CONCLUSIONS

The effects of the phase structures formed by rubber and rigid particles in a polymer matrix on the rigidity and toughness of polymer/rubber/rigid particle composites were studied. The results show that the polymer rigidity and toughness can be simultaneously enhanced for a phase structure in which rubber particles are closely surrounded by rigid particles or a phase structure in which rigid particles are covered by rubber; however, it is difficult to simultaneously enhance the rigidity and toughness of a phase structure in which rubber and rigid particles are separately dispersed. Moreover, the phase structure in which rubber particles are closely surrounded by smaller rigid particles is more effective in simultaneously increasing the rigidity and toughness of the polymer. However, it is difficult to obtain such a phase structure experimentally. Compared with one phase modifier (only rubber or rigid particles), the impact toughness and modulus can be increased synchronously by the two phase modifiers with core shell structures, whereas they cannot generally be increased by a single-phase modifier. This study provides new insights into the relationship between the mechanical properties and phase structure of polymer composites and is helpful for designing and preparing polymer materials with high rigidity and toughness.

Conflict of Interests

The authors declare no interest conflict.

Data Availability Statement

The data in this study can be available from the corresponding

author upon reasonable reasons.

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REFERENCES

1 Sharma, A.; Joshi, S. C. Effect of core-shell micro-fillers on impact performance and damage progression in bi-directional carbon thermoplastic composites. *Compos. Part B Eng.* **2025**, *293*, 112127.
2 Zuiderduin, W. C. J.; Huétink, J.; Gaymans, R. J. Rigid particle toughening of aliphatic polyketone. *Polymer* **2006**, *47*, 5880–5887.
3 Lule, Z.; Kim, J. Thermally conductive and highly rigid polylactic acid (PLA) hybrid composite filled with surface treated alumina/nano-sized aluminum nitride. *Compos. Part A Appl. Sci. Manuf.* **2019**, *124*, 105506.
4 Wee, J.-W.; Chudnovsky, A.; Choi, B.-H. Brittle–ductile transitions of rubber toughened polypropylene blends: a review. *Int. J. Precis. Eng. Manuf. Green Technol.* **2024**, *11*, 1361–1402.
5 Wang, J.; Zhang, X.; Jiang, L.; Qiao, J. Advances in toughened polymer materials by structured rubber particles. *Prog. Polym. Sci.* **2019**, *98*, 101160.
6 Gao, Y.; Jiang, Z.; Zhang, J.; Tan, H.; Jin, J.; Jiang, W. Models and theories regarding the brittle-ductile transition of rubber-toughened thermoplastics and their application in the design of high-impact polymer composites with balanced toughness and rigidity. *Polym. Sci. Technol.* **2025**, *1*, 314–329.
7 Shangguan, Y.; Chen, F.; Yang, J.; Jia, E.; Zheng, Q. A new approach to fabricate polypropylene alloy with excellent low-temperature toughness and balanced toughness-rigidity through unmatched thermal expansion coefficients between components. *Polymer* **2017**, *112*, 318–324.
8 Corté, L.; Rebizant, V.; Hochstetter, G.; Tournilhac, F.; Leibler, L. Toughening with little stiffness loss: polyamide filled with ABC triblock copolymers. *Macromolecules* **2006**, *39*, 9365–9374.
9 Shirvanimoghaddam, K.; Balaji, K. V.; Yadav, R.; Zabihi, O.; Ahmadi, M.; Adetunji, P.; Naebe, M. Balancing the toughness and strength in polypropylene composites. *Compos. Part B Eng.* **2021**, *223*, 109121.
10 Fasihi, M.; Mansouri, H. Effect of rubber interparticle distance distribution on toughening behavior of thermoplastic polyolefin elastomer toughened polypropylene. *J. Appl. Polym. Sci.* **2016**, *133*, app.44068.
11 Zuiderduin, W. C. J.; Westzaan, C.; Huétink, J.; Gaymans, R. J. Toughening of polypropylene with calcium carbonate particles. *Polymer* **2003**, *44*, 261–275.
12 Li, Z.; Shen, J.; Ma, H.; Liu, F.; Yu, W.; Shangguan, Y.; Zheng, Q. Effect of partially crosslinked nanoparticle cluster networks in dispersed rubber phase on the toughness and stiffness of polypropylene composites. *Compos. Part B Eng.* **2025**, *304*, 112669.
13 Rouhi, E.; Bahrami, K.; Poorabdollah, M. Remarkable enhancement of physical-mechanical properties in unsaturated polyester resin composites using SiO_2@ CTBN hybrid particles. *Polym. Compos.* **2025**, *46*, 10908–10922.
14 Zou, M.; Chen, J.; Wei, Z.; Lei, W.; You, J.; Liu, J.; Zhang, Q.; Shi, D. Preparation of SiO_2@EVA core–shell particles *via* post-emulsification method with polydisperse SiO_2 particle sizes and the critical thickness-to-diameter ratio in PA6/SiO_2@EVA system. *Appl. Surf. Sci.* **2023**, *614*, 156051.
15 Hajibabazadeh, S.; Razavi Aghjeh, M. K.; Palahang, M. Study on the fracture toughness and deformation micro-mechanisms of PP/EPDM/SiO_2 ternary blend-nanocomposites. *J. Compos. Mater.* **2020**, *54*, 591–605.
16 Jancar, J.; Zidek, J. Effect of *in-situ* formed core–shell inclusions on the mechanical properties and impact fracture of polypropylene. *Eur. Polym. J.* **2015**, *71*, 221–230.
17 Yang, L.; Hu, Y.; Guo, H.; Song, L.; Chen, Z.; Fan, W. Toughening and reinforcement of rigid PVC with silicone rubber/nano-$CACO_3$ shell-core structured fillers. *J. Appl. Polym. Sci.* **2006**, *102*,

Graphical Abstract

Phase Structure Formed by Rubber and Rigid Particles in a Polymer and Its Effect on the Composite Rigidity and Toughness

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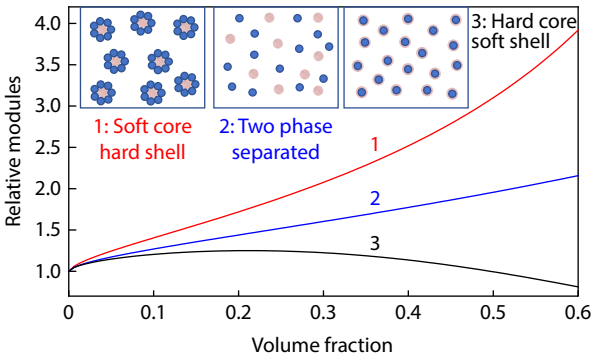
The soft core-hard shell structure is the best, whereas the hard core-soft shell structure is the worst for enhancing the modulus in three-phase structures.



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2560–2567.
18 Dubnikova, I. L.; Berezina, S. M.; Antonov, A. V. The effect of morphology of ternary-phase polypropylene/glass bead/ethylene-propylene rubber composites on the toughness and brittle-ductile transition. *J. Appl. Polym. Sci.* **2002**, *85*, 1911–1928.
19 Wang, X.; Gao, Y.; Jin, J.; Jiang, W. A strategy to develop homo-polypropylene composites with high impact and high rigidity. *Macromolecules* **2024**, *57*, 4576–4583.
20 Yang, H.; Zhang, Q.; Guo, M.; Wang, C.; Du, R.; Fu, Q. Study on the phase structures and toughening mechanism in PP/EPDM/SiO_2 ternary composites. *Polymer* **2006**, *47*, 2106–2115.
21 Hajibabazadeh, S.; Razavi Aghjeh, M. K.; Mazidi, M. M. Processing–microstructure–fracture toughness relationships in PP/EPDM/SiO_2 blend-nanocomposites: effect of mixing sequence. *Polym. Eng. Sci.* **2024**, *64*, 2513–2529.
22 Ji, X.-L.; Jing, J.-K.; Jiang, W.; Jiang, B.-Z. Tensile modulus of polymer nanocomposites. *Polym. Eng. Sci.* **2002**, *42*, 983–993.
23 Takayanagi, M.; Uemura, S.; Minami, S. Application of equivalent model method to dynamic rheo-optical properties of crystalline polymer. *J. Polym. Sci., C Polym. Symp.* **1964**, *5*, 113–122.
24 Mokhtari Dizaji, S.; Katbab, A. A.; Hajibabazadeh, S. Role of interfacial compatibilizer and functionality of rubber phase on micromorphology development and mechanical properties of PP/EPDM/nanosilica ternary nanocomposite. *Polym. Bull.* **2023**, *80*, 1353–1368.
25 Yang, J.; Li, F.; Guan, C.; Xu, X.; Zhong, L.; Gao, Y.; Han, Y.; Yan, N.; Zhao, G.; Jiang, W. Brittle-ductile transition of impact PP blends: effect of modulus ratio of PP matrix to impact modifier. *Polym. Bull.* **2023**, *80*, 4459–4471.
26 Borggreve, R. J. M.; Gaymans, R. J.; Schuijer, J. Impact behaviour of nylon-rubber blends: 5. Influence of the mechanical properties of the elastomer. *Polymer* **1989**, *30*, 71–77.
27 Jiang, W.; An, L.; Jiang, B. Brittle–tough transition in elastomer toughening thermoplastics: effects of the elastomer stiffness. *Polymer* **2001**, *42*, 4777–4780.
28 Li, C.; Wang, Z.; Liu, W.; Ji, X.; Su, Z. Copolymer distribution in core–shell rubber particles in high-impact polypropylene investigated by atomic force microscopy–infrared. *Macromolecules* **2020**, *53*, 2686–2693.
29 Li, F.; Gao, Y.; Zhang, C.; Jin, J.; Ji, X.; Zhang, Y.; Zhang, X.; Jiang, W. Design of high impact thermal plastic polymer composites with balanced toughness and rigidity: effect of matrix polymer molecular weight. *Polymer* **2020**, *208*, 122957.
30 Wu, S. Phase structure and adhesion in polymer blends: a criterion for rubber toughening. *Polymer* **1985**, *26*, 1855–1863.
31 Borggreve, R. J. M.; Gaymans, R. J. Impact behaviour of nylon-rubber blends: 4. Effect of the coupling agent, maleic anhydride. *Polymer* **1989**, *30*, 63–70.

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