

Structural Evolution in Flowing Immiscible Blends in the Presence of Rough Particles: Dependence of Shear Rate and Blend Ratio

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Abstract The influence of polystyrene particles with different nanoscale roughnesses on the morphology of polyisobutylene/polydimethylsiloxane blends was studied under shear flow by using confocal laser scanning microscopy. It was found that the surface roughness of particles strongly affected their diffusion and distribution behaviors, thereby determining the size and spatial arrangement of droplets in the blends. The roughness effect of particles was found to possess a strong dependence on both the blend ratio and the shear rate. The result suggested that the particle roughness can serve as a new parameter to control the structure-property correlation in particle-filled polymer blends, especially under slow flow.

Keywords Rough particles; Immiscible polymer blends; Shear flow

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INTRODUCTION

The presence of micro- or nanoscale solid particles can usually alter the structural evolution dynamics of immiscible polymer blends under flow.^[1–3] Especially, the selective distribution of solid particles in the immiscible blends, which is closely related to their affinity for each component and could be described by the Young's equation,^[4] is reported to play a vital role. When solid particles are located at the fluid/fluid interface, they usually have higher morphological refinement and stabilization efficiencies than when they are distributed in the droplet or matrix phase.^[5] The underlying mechanism can be attributed to the reduction in the apparent interfacial tension^[6] or the formation of a dense rigid particle layer at the interface which can act as a space barrier to effectively prevent flow-induced droplet coalescence.^[7] Some studies have found that interfacial particles can also promote the coalescence of droplets.^[8] When the interfacial particles show better affinity towards the droplet phase, they promote the coalescence of the droplets, which can be explained by a "bridging-dewetting" mechanism;^[9] when they show better affinity towards the matrix phase, the coalescence of droplets can also be promoted by bringing the droplets close enough to collide under flowing.^[10] Therefore, the asymmetric affinity of the interfacial particles toward blend components will lead to quite different morphological evolutions in the blend when the composition

ratio changes.^[11]

During the synthesis process, the surface of particles will inevitably have a certain degree of nanoscale roughness. As a result, the diffusion and aggregation behavior of particles is expected to show a dependence on the microscopic particle-particle and particle-fluid interactions mediated by the surface roughness. Some studies in water/oil mixtures have shown that the surface roughness of particles has a significant impact on the diffusion behavior,^[12,13] interaction force^[14] and wettability^[15] of particles. For interfacial particles, their adsorption,^[16,17] movement^[18] and interaction^[19] are also affected by the surface roughness of particles due to the contact-line pinning effect. Therefore, the particle roughness is expected to be a crucial factor in the structural evolution of polymer blends besides the surface wettability, size and concentration of particles.

In previous work, we studied the effect of roughness on the morphology of a fixed-ratio blend under a low-speed flow field, which is the first study discussing the particle roughness-induced microstructure in flowing polymer blends. In order to broaden the application of rough particles during polymer processing, in this contribution, the effect of particle roughness on the morphology of immiscible polymer blends was explored at different shear rates and blend ratios. Fluorescently labeled smooth and rough polystyrene (PS) particles were prepared and added into immiscible blends of polyisobutylene (PIB) and polydimethylsiloxane (PDMS). The morphology evolution of blends in the presence of smooth and rough particles was studied by using confocal laser scanning microscopy and discussed in terms of particle diffusion and distribution behavior.

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EXPERIMENTAL

Preparation of Fluorescent Microsphere

Monodisperse and spherical PS particles (diameter=3.1±0.1 μm) with different roughnesses but similar surface chemistry were purchased from Knowledge & Benefit Sphere Tech. Co., Ltd., China (please refer to Ref. [20] for detailed SEM pictures of the particles). The “smooth” ones and rough ones have a root mean square (RMS) roughness of 2.56±0.65 and 12.65±2.1 nm, respectively, which are measured by atomic force microscopy (AFM). Through a swelling method,^[21] fluorescent particles labeled with Rhodamine B with an excitation wavelength of 552 nm were prepared.

Preparation of Blends

Particle-free blends consisted of polyisobutylene (PIB, Daelim Industrial Co., Ltd., Korea, number-average molecular weight M_n =~1300, ρ_{PIB} =0.901 g/cm³) and immiscible polydimethylsiloxane (PDMS, Aldrich, America, M_n =4×10⁴~5×10⁴, ρ_{PDMS} =0.971 g/cm³) in a weight ratio of 90:10 or 10:90. The zero shear viscosity of PIB and PDMS is 50.3 and 93.1 Pa·s at 30 °C, respectively. Particle-filled blends consisted of PS particles, PIB and PDMS at a certain ratio (up to 2 wt%). Blend components were mixed simultaneously at room temperature using a scraper for 15 min. Then, all the samples were degassed in a vacuum oven at 40 °C for 12 h before use.

Shearing Experiments

The morphology of blends under shearing was observed by a confocal laser scanning microscopy (Leica, TCS SP8) equipped with a shearing stage (Linkam, CSS450). All the samples were pre-sheared at 20 s⁻¹ and 30 °C for 300 s to eliminate the sample loading history and then coalesced at 0.05 or 0.5 s⁻¹ for 1.8×10⁴ s. Images were taken every 60 s under a 20× objective lens by a CCD camera. The droplet radii were measured and counted by the free ImageJ software. The volume average radius R_v of 200 droplets per sample was calculated using the following equation:

$$R_v = \frac{\sum_{i=1}^{\infty} n_i R_i^4}{\sum_{i=1}^{\infty} n_i R_i^3} \quad (1)$$

where R_i is the radius of each droplet and n_i is the number of droplets having radius R_i . To avoid the limited number of droplets in the late stage of coalescence, we performed repetitive experiments or counted the droplets in several pictures around a specific time point.

2D Particle Tracking Experiments

The movement of individual particles under shearing was tracked by using a Tracking Particle module equipped on the confocal laser scanning microscope (CLSM). Firstly, 0.01 wt% rough or smooth particles were separately added into the pure PIB or PDMS phase and stirred for 10 min to form a very dilute suspension. Then, the suspension was degassed in a vacuum oven at 40 °C for 12 h. The shear condition was the same as that used in shearing experiments. A careful control of the stage position guaranteed the same confocal slice and the accuracy of fluid rate for different samples. By analyzing the trajectory and the mean-square displacement of individual particles, the particle velocities under different conditions were obtained and compared.

RESULTS AND DISCUSSION

PIB/PDMS (90/10) Blends

This study used large PS particles (~3 μm) to facilitate the observation at the single-particle level. This will to a certain extent affect the compatibilization efficiency of the particles toward the blends although our focus will be on the effect of particle roughness. At slow shear rates, the effect of structural properties of particles (such as surface roughness) rather than the deformation rate of blends should be prominent on the dynamics and distribution of particles in the blends. Figs. 1(a)–1(d) and 1(a')–1(d') show that after a long period of low-speed shearing (0.05 s⁻¹, t =300 min), the PIB/PDMS (90/10) blends containing rough or smooth PS particles differ largely in the size and spatial arrangement of droplets. In general, the addition of smooth particles promotes the flow-induced droplet coalescence in the PIB/PDMS (90/10) blends (Figs. 1e and 1f). The droplet size grows gradually from 20.1 μm for the blend with 0.1 wt% particles to 29.8 μm for that with 0.5 wt% particles. After that, the coalescence-promoting effect of smooth particles decreases as more particles are incorporated. When the particle concentration increases to 2.0 wt%, the droplet radius decreases to 22.9 μm. In contrast, the addition of rough particles only refines the morphology slightly and the droplet radius is about 18 μm which changes little with the particle concentration.

Besides the droplet size, the addition of particles also causes a change in the spatial arrangement of droplets in the PIB/PDMS (90/10) blends. Figs. 1(A) and 1(A') show that upon the addition of 0.1 wt% particles, the droplets are mainly dispersed randomly in an individual form in the matrix regardless of the surface roughness of particles. As the particle content increases to 1.0 wt%–2.0 wt% (Figs. 1c and 1d), the droplets mainly appear in the form of clusters of 3–6 droplets in blends containing smooth particles. For blends containing rough particles, however, similar droplet bridging behavior does not appear, and only randomly dispersed droplets are found within the particle concentration range investigated.

The difference in the influence of smooth or rough particles on the morphology of the blends is closely related to their selective distribution and aggregation behavior in the blends. Confocal micrographs with a higher magnification (Fig. 2a) show that at a low particle content (e.g. 0.1 wt%), smooth particles tend to reside at the PIB/PDMS interface and show a higher affinity towards the PIB matrix phase. As the particle content increases (e.g. 2 wt%), smooth particles form multilayer aggregations at the PIB/PDMS interface and no particles can be found in droplets (Fig. 2b). For rough particles, however, most of them are distributed in the matrix phase at low particle contents; only at higher particle contents, some rough particles can be found at the interface in the form of a monolayer (Fig. 2b') and no obvious multilayer aggregation is formed at the interface. This formation of particle monolayer rather than multilayer may be related to the strong contact line-pinning effect of rough particles,^[17] which severely traps the motion and rearrangement of particles at the PIB/PDMS interface.

For smooth and rough particles, the large difference in their distribution behavior in blends can be understood by their diffusion behavior under the flow. According to our previous work,^[20] smooth particles are found to possess higher

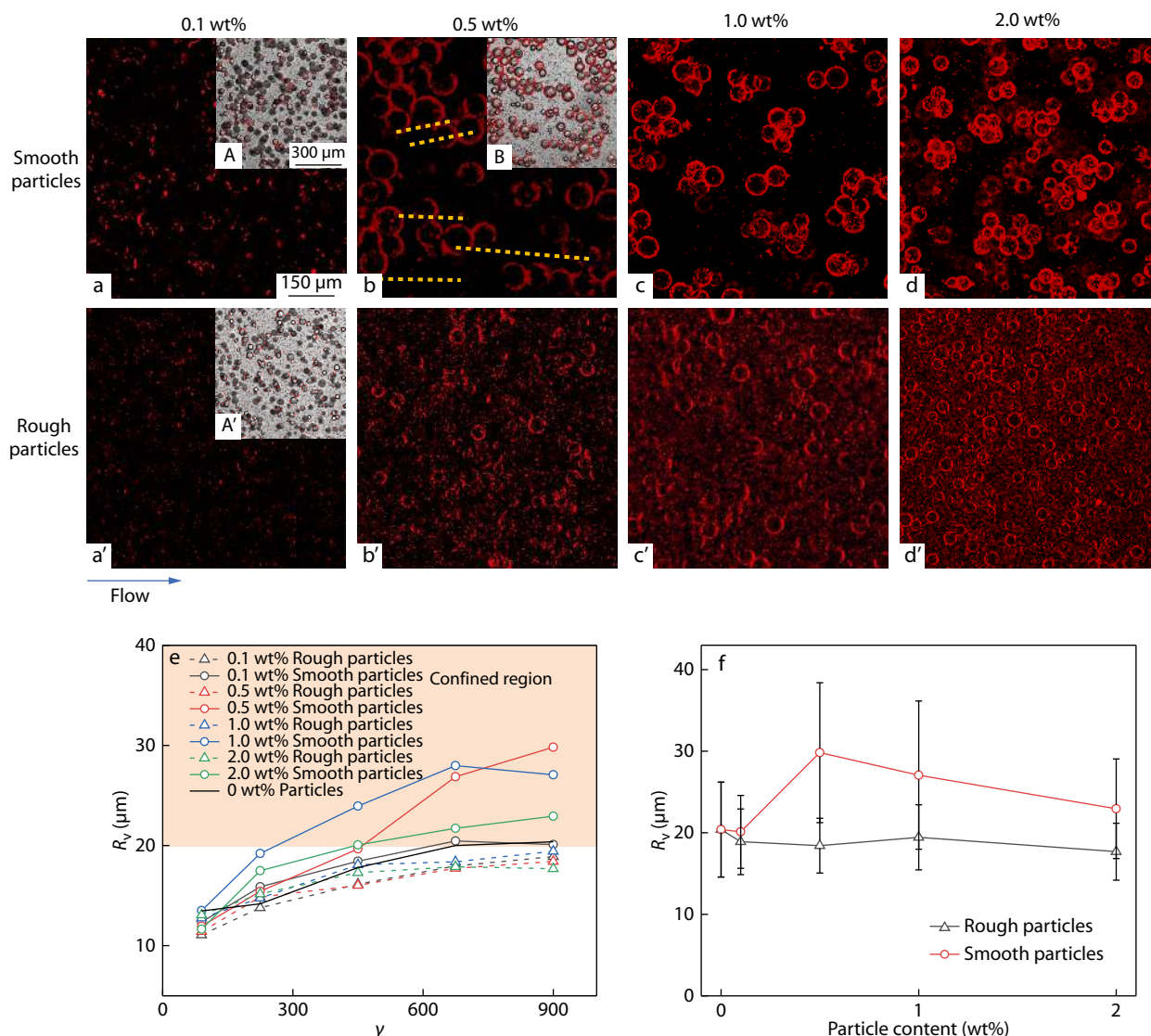


Fig. 1 Morphology of PIB/PDMS (90/10) blends with (a–d) smooth particles and (a'–d') rough particles after shearing at 0.05 s^{-1} for $1.8 \times 10^4 \text{ s}$ ($\gamma=900$): (a,a') 0.1 wt%; (b,b') 0.5 wt%; (c,c') 1.0 wt%; (d,d') 2.0 wt%. Insets A, A' and B are corresponding overlay images of (a,a') and (b). The yellow dash lines denote the droplet structures and the blue arrow denotes the flow direction. The gap space of the shear cell is $200 \mu\text{m}$. (e) Growth of droplet radius (R_v) in PIB/PDMS (90/10) blends with different rough or smooth PS particle concentrations as a function of shear strain at 0.05 s^{-1} . (f) Dependence of R_v of PDMS droplets in PIB/PDMS (90/10) blends on particle content after shearing at 0.05 s^{-1} for $1.8 \times 10^4 \text{ s}$.

mobility in the slowly flowing fluids than those rough ones. Consequently, they can easily migrate to the PIB/PDMS interface under slow shear flow. At low loadings (0.1 wt%–0.5 wt%), these particles could bridge adjacent droplets into clusters.^[22] The particle-free area near the bridging point has a higher probability to coalesce during shearing,^[9] thereby increasing the droplet size. When the particle concentration increases to 1.0 wt%–2.0 wt%, more particles cover the droplet surfaces and gradually hinder droplet aggregation during shearing,^[23] thereby leading to reduced droplet sizes. The higher the particle content is, the earlier this hindrance appears (Fig. 1e). Unlike those smooth particles, rough particles exhibit lower mobility under slow flow,^[13,20] and most of them distribute within the continuous PIB phase rather than migrating to the fluid/fluid interface. Even if a few of them migrate to the interface, they just stagnate there in the form of a

monolayer (Fig. 2b') due to the high energy required to break through the fluid/fluid interface.^[24] These large rough particles appear a marginal compatibilization effect on the 90/10 blends (Fig. 1f) related to their weak viscosity enhancement and interface hindrance effect toward droplet coalescence.^[25] Therefore, the droplet size of blends with rough particles is smaller than that of blends with smooth particles.

The spatial arrangement of droplets in the blends is closely related to the droplet size and geometrical confinement effect exerted by shear walls. The confinement degree of droplets between the walls can be given by R_v/h ,^[26] where h is half the distance between the upper and lower plates of the shear cell. The droplets will enter a confined state when $R_v/h > 0.2$. Here, h is $100 \mu\text{m}$, and the corresponding droplet radius will be $20 \mu\text{m}$ when the geometrical confinement occurs. As shown in the orange area in Fig. 1(e), the droplets and their

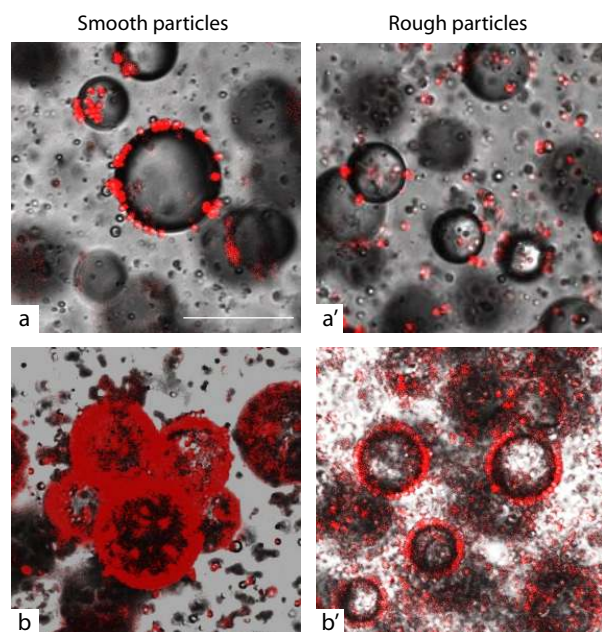


Fig. 2 Overlay fluorescent images showing the distribution of (a, b) smooth and (a', b') rough PS particles in PIB/PDMS (90/10) blends after shearing at 0.05 s^{-1} for $1.8 \times 10^4 \text{ s}$: (a, a') 0.1 wt%; (b, b') 2.0 wt%. The scale bar in (a) is $50 \mu\text{m}$.

clusters in blends containing 0.5 wt%–2.0 wt% smooth particles should be in a confined state due to the bridging effect of smooth particles.^[11] When a blend flows in a confined geometry, the distortion of velocity fields around droplets will

produce attractive interactions between droplets,^[27] thereby leading to the formation of droplet chains in the flow direction. With the increasing particle content, the cluster-like structure replaces the droplet chains. This is because the higher particle content increases the particle coverage of the droplets and provides more bridging sites between droplets.^[11] After a long period of shear flow, the droplets finally form stable droplet chains (Fig. 1b) or cluster-like structure (Figs. 1c–1d). In blends filled with rough particles, the droplets are always small in size and therefore are not in a confined state. The schematic diagram in Fig. 3 details the difference in the morphological evolution of different blends under slow shear flow.

Interestingly, the discrepancy in the morphological evolution of blends induced by smooth and rough particles is found to diminish as the shear rate increases. As shown in Fig. 4, the shear-induced morphology of particle-filled PIB/PDMS (90/10) blends at 0.5 s^{-1} significantly differs from that observed at 0.05 s^{-1} . Firstly, the increase in the shear rate leads to a decline in the droplet size. Secondly, the addition of any PS particles further inhibits the coalescence of droplets. Adding 0.1 wt%–2.0 wt% of smooth or rough particles, the droplets are always dispersed independently in the matrix phase and the droplet radius after shearing at 0.5 s^{-1} maintains at $\sim 9 \mu\text{m}$, which is slightly smaller than that of particle-free blends. No droplet bridging can be found even in the blends filled with smooth particles. Confocal micrographs reveal that most of the smooth or rough particles are distributed in the matrix phase at high-rate shearing. Only at high particle contents (2 wt%), a small number of smooth particles can be found at the thermodynamically favorable PIB/PDMS

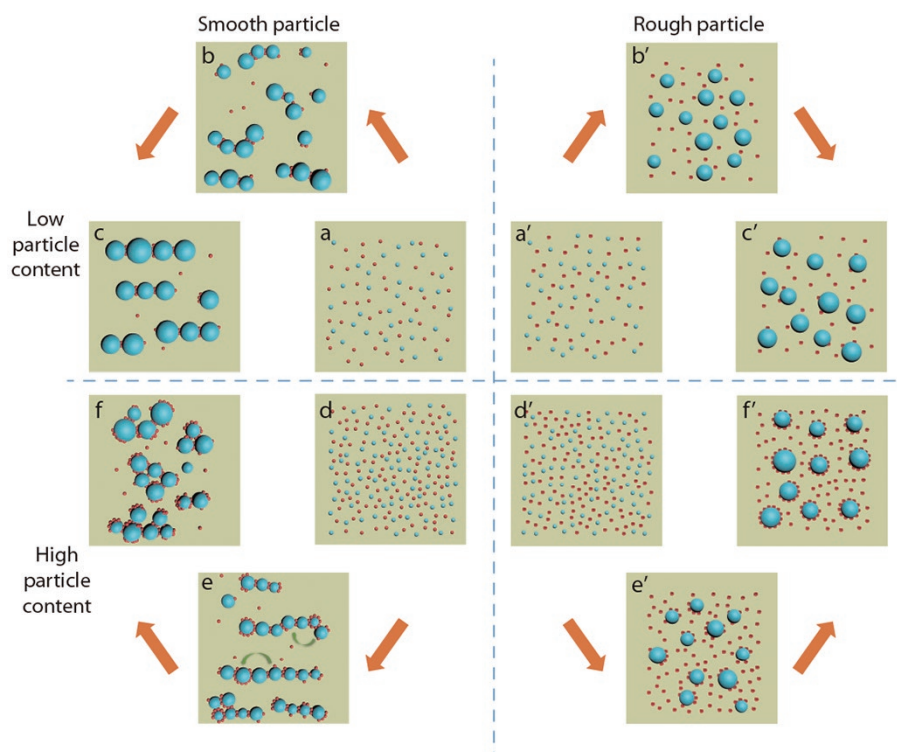


Fig. 3 Schematic depiction of the formation of different droplet structures in PIB/PDMS (90/10) blends with different particles at 0.05 s^{-1} , where particles are in red and PIB is in blue.

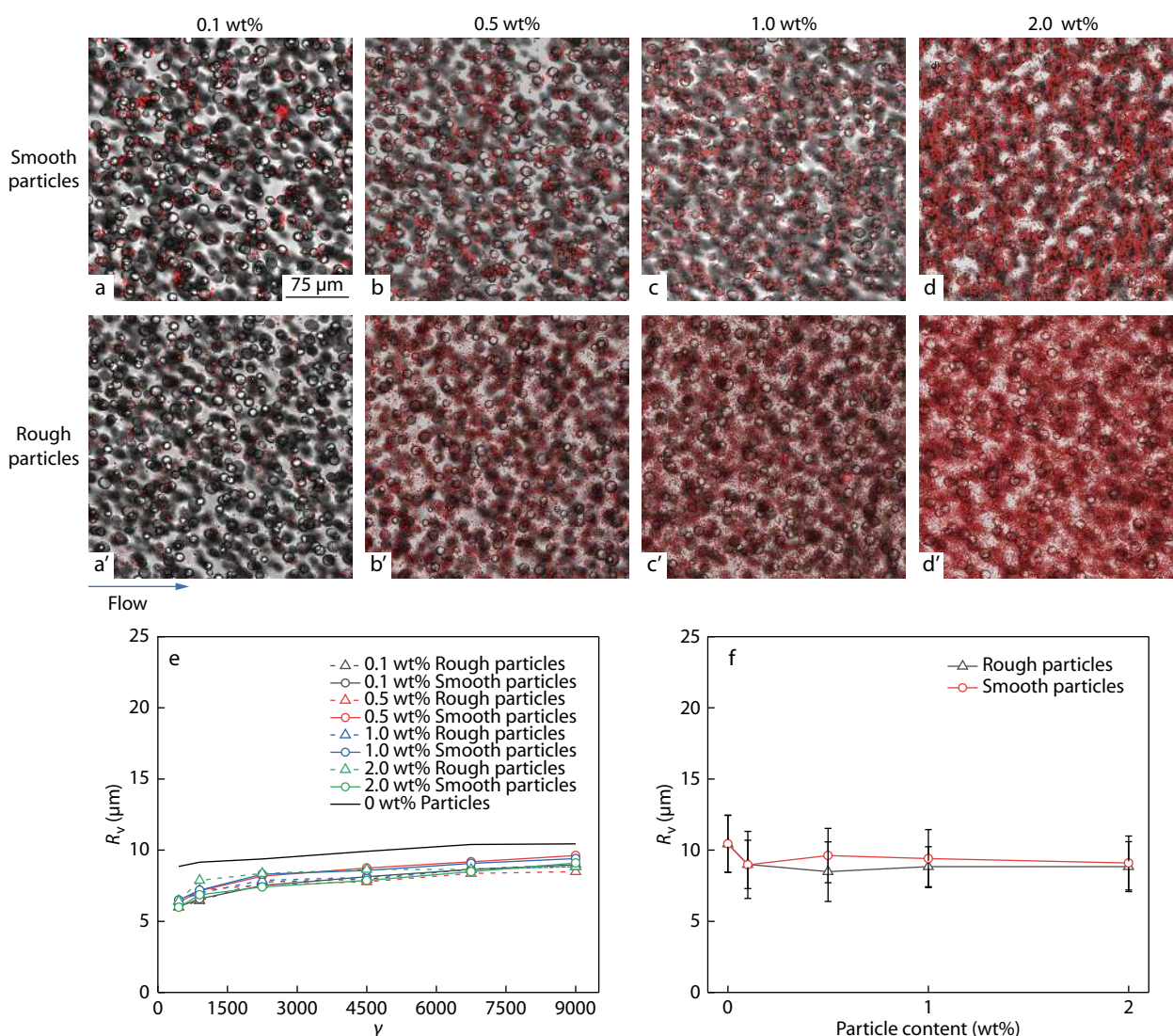


Fig. 4 Morphology of PIB/PDMS (90/10) blends with (a–d) smooth particles and (a'–d') rough particles after shearing at 0.5 s⁻¹ for 1.8×10⁴ s (γ =9000): (a, a') 0.1 wt%; (b, b') 0.5 wt%; (c, c') 1.0 wt%; (d, d') 2.0 wt%. The scale bar in (a) is 75 μm and the blue arrow denotes the flow direction. The gap space of shear cell is 200 μm. (e) Growth of droplet radius (R_v) in PIB/PDMS (90/10) blends with different particle concentrations at 0.5 s⁻¹ as a function of shear strain. (f) Dependence of R_v of PDMS droplets in PIB/PDMS (90/10) blends on particle content after shearing at 0.5 s⁻¹ for 1.8×10⁴ s.

interface (Fig. 4d). Besides, no droplet cluster structure is observed possibly due to the stronger shear force at 0.5 s⁻¹ which may exceed the weak bridging effect of particles.^[10]

The difference in the morphology caused by the difference in the particle distribution can be understood from thermodynamics and dynamics perspectives. From the perspective of thermodynamics, the different wettabilities of particles caused by roughness may affect their distribution. In our study, as the wetting parameter of smooth PS particles in the PIB/PDMS blends is -0.48,^[11] PS particles tend to be distributed at the PIB/PDMS interface. Theoretically, Nonomura *et al.*^[16] introduced the roughness into the Wenzel model for a solid particle at the liquid/liquid interface system based on an area amplification factor (γ). They found that the microstructure on the surface changed the wettability and the equilibrium position of the adsorbed particle drastically,

which prevents the particle from adsorption at the interface. According to the modified Wenzel model, as the γ (namely roughness) increases, particles with a negative wetting parameter (here -0.48) will change from tending to be distributed at the interface to tending to be distributed in one of the liquids. In this sense, rough PS particles should show a better apparent affinity with the PIB phase than smooth particles ones do, which is consistent with Fig. 2.

To reveal the underlying reason for the diminishing difference in the morphology tailoring effect of the two types of particles, the diffusion dynamics of these particles was investigated at 0.05 and 0.5 s⁻¹, respectively, by single-particle tracking experiments. To exclude the possibility of wall slip during the single-particle tracking experiments, the particle velocity at 0.5 s⁻¹ in the flow cell was measured at different heights. As shown in Fig. 5 below, the particle velocity dis-

plays a linear relationship with the height value and the experimental curve overlaps satisfactorily with the theoretical curve within experimental error. Moreover, the extrapolated displacement $\langle r \rangle$ approaches 0 when the height becomes 0, implying that no wall slip occurs near the two plates of the shear cell. There should be no wall slip at an even lower shear rate of 0.05 s^{-1} . According to the Péclet (Pe) number^[28] of particles, the Brownian motion of particles can be ignored under such experimental conditions, and the convect of particles is dominated ($Pe \gg 1$). Fig. 6(a) shows that the velocity of rough particles is slower than that of fluid and smooth particles at a low shear rate (fluid rate = $5.74 \mu\text{m/s}$). The difference may be explained by the larger effective hydrodynamic radius of rough particles due to the presence of a bound layer on particles.^[20] On the other hand, Hsiao *et al.*^[12] found that the presence of roughness can slow down the rotation of particles in Brownian motion. Here, similar particle rotation should also exist under shear flow, and therefore rough particles may move slower than smooth ones under flow. However, at a higher fluid rate of $52.0 \mu\text{m/s}$, the two types of particles show a similar motion rate of $50 \pm 0.5 \mu\text{m/s}$ in the PIB or PDMS fluid close to that of fluids (Fig. 6b). This suggests that in terms of diffusion behavior, the contribution of the larger shear force generated by a faster fluid flow rate should

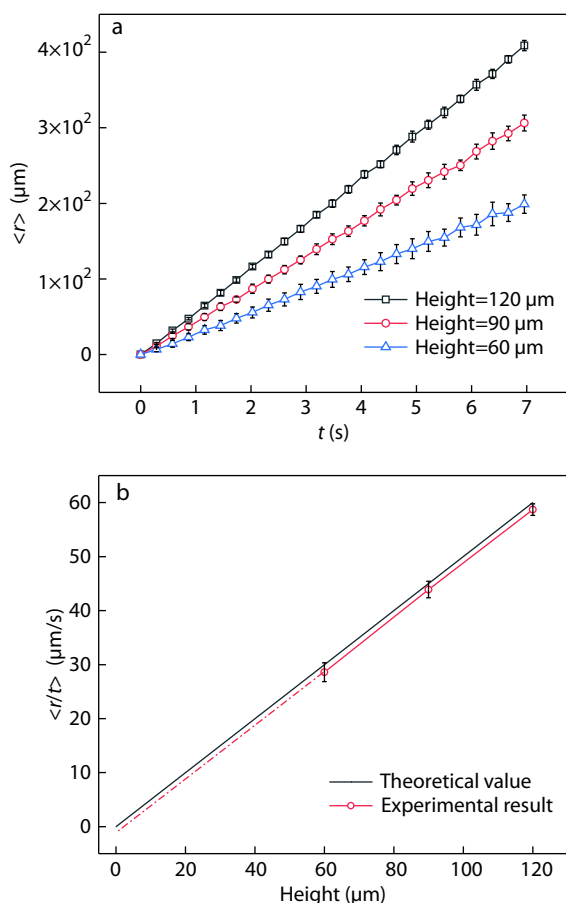


Fig. 5 (a) Displacement $\langle r \rangle$ of individual “smooth” PS particles in the PDMS fluid at different heights; (b) The velocity of particles at different heights compared with the fluid rates calculated from the uniform velocity profile without wall slip.

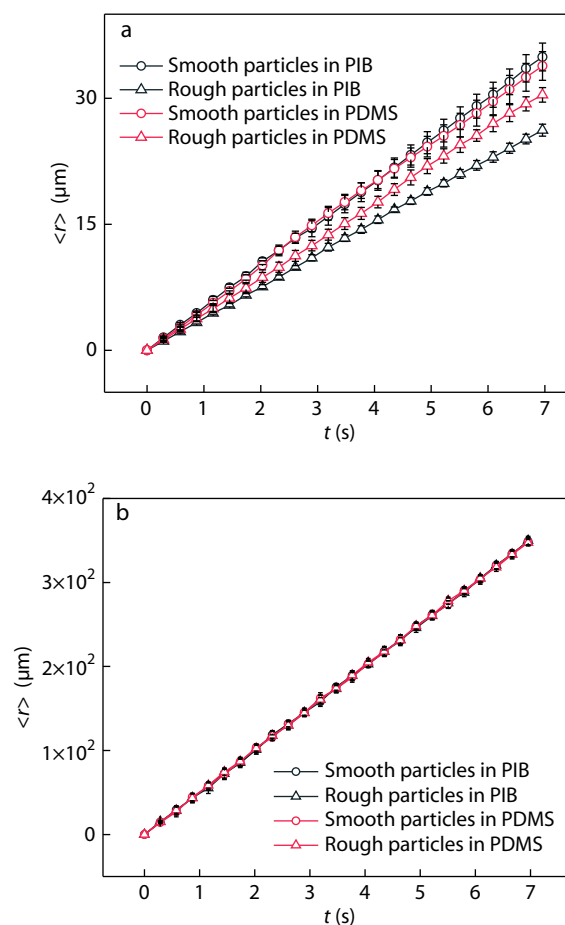


Fig. 6 Displacement $\langle r \rangle$ of individual smooth and rough particles in PIB or PDMS at different fluid rates: (a) $5.74 \mu\text{m/s}$ and (b) $52.0 \mu\text{m/s}$.

dominate over the contribution of the difference in particle roughness. The above mentioned results show that the size and spatial arrangement of droplets in the presence of particles with different roughnesses are inextricably related to the shear rate, and the fundamental reason may be ascribed to the shear-rate dependent diffusion behavior of particles.

PIB/PDMS (10/90) Blends

At the thermodynamic equilibrium, the discrepancy in the affinity of particles towards the different components of a blend will play a decisive role in determining the uneven distribution of particles in the blend. Therefore, it is expected that the influence of smooth and rough particles on the morphological evolution of PIB/PDMS blends will exhibit a compositional dependence. To explore the combinational effect of asymmetric particle affinity and roughness towards the flow-induced morphology of blends, the inversed PIB/PDMS (10/90) blends are further explored in this study.

Fig. 7 shows that the change in the blend composition alters the distribution of particles. Smooth particles tend to reside at the PIB/PDMS interface at low particle contents (such as 0.1 wt%). These interfacial particles should protrude towards droplets as they accumulate gradually inside the PIB droplets after interfacial saturation when adding more smooth particles (Figs. 7a–7c). As in the PIB/PDMS (90/10)

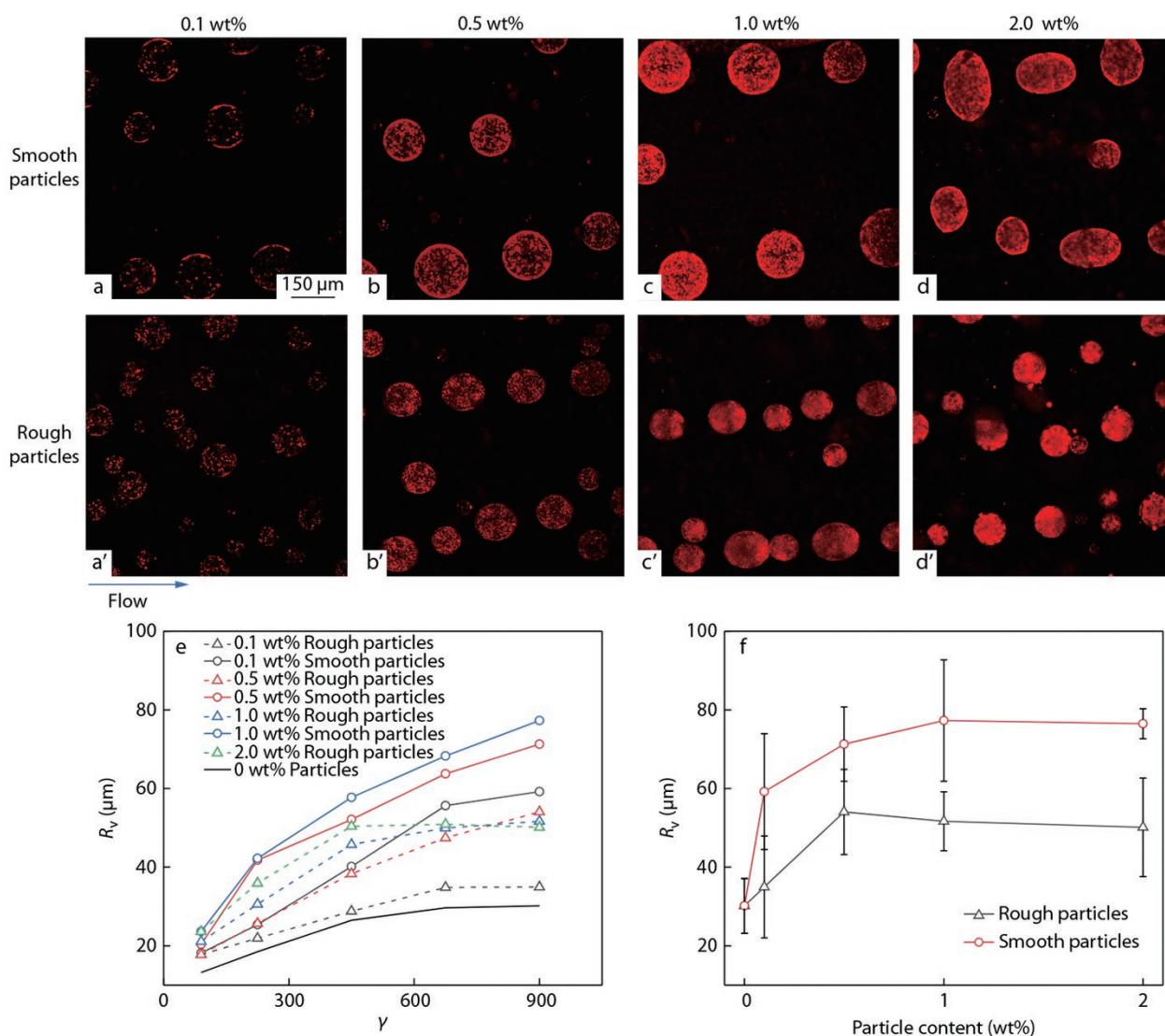


Fig. 7 Morphology of PIB/PDMS (10/90) blends with (a–d) smooth particles and (a'–d') rough particles after shearing at 0.05 s⁻¹ for 1.8 × 10⁴ s (γ = 900): (a, a') 0.1 wt%; (b, b') 0.5 wt%; (c, c') 1.0 wt%; (d, d') 2.0 wt%. The scale bar in (a) is 150 μm and the blue arrow denotes the flow direction. The gap space of the shear cell is 200 μm. (e) The growth of droplet radius (R_v) in PIB/PDMS (10/90) blends with different particle concentrations at 0.05 s⁻¹ as a function of shear strain. (f) Dependence of R_v of PIB droplets in PIB/PDMS (10/90) blends on particle content after shearing at 0.05 s⁻¹ for 1.8 × 10⁴ s.

blends, the addition of smooth particles in the PIB/PDMS (10/90) blends also leads to significantly promoted droplet coalescence (Fig. 7e) at a low shear rate (0.05 s⁻¹) possibly via a "bridging-dewetting" mechanism.^[9,22] When the particles wet the droplet phase preferentially, the matrix film formed between two approaching droplets under the flow will be unstable and dewet at the three-phase contact lines to cause droplet coalescence. With increasing particle concentration (0.5 wt%–1.0 wt%), the droplets gradually present a single-layer pearl necklace like arrangement along the flow direction (Fig. 7) due to the confinement effect of shear walls.^[29] It should be noted that as the particle concentration reaches 2.0 wt%, large droplets with irregular shapes appear, suggesting the interfacial solidification of droplets due to particle jamming.^[30–33] Moreover, most of these solidified droplets align in the vorticity direction under slow flow, and this phe-

nomenon has been reported previously in other particle-laden blends by us and explained by using Jeffery's orbit theory.^[33] According to this theory, the rigidized droplets need to adopt a vorticity arrangement during flowing to reduce the energy dissipation.^[34] Unlike the PIB/PDMS (90/10) blends, no droplet bridging phenomenon is found in the PIB/PDMS (10/90) blends filled with smooth particles. It is because that the droplet bridging needs the interfacial particles to protrude considerably out of the droplet phase.^[23]

In contrast, rough particles tend to be distributed uniformly within the PIB droplets and they rarely aggregate at the interface regardless of their concentration. The addition of rough particles also promotes the flow-induced coalescence of the blends although it is not as profound as in the case of blends filled with smooth ones due to the lacking of interfacial particles. The pearl necklace like structure is more

regular than that in the blend filled with smooth particles and there is a strong confinement effect of shear walls.^[27] Even at a high concentration of 2.0 wt%, rough particles cannot "rigidize" the droplets and generate vorticity orientation. Our previous work^[20] has shown that only when the particle concentration reaches 4.0 wt%, a particle network structure can be formed by rough particles, which will significantly enhance the viscoelasticity of the droplets and "rigidize" the droplets.

Interestingly, the addition of particles still leads to promoted droplet coalescence in the PIB/PDMS (10/90) blends at a higher shear rate of 0.5 s^{-1} (Fig. 8), which differs significantly from the case of PIB/PDMS (90/10) blends (Fig. 4). As the concentration of smooth particles increases from 0.1 wt% to 2 wt%, the droplet radius grows gradually from $18.5 \text{ }\mu\text{m}$ to

$28.1 \text{ }\mu\text{m}$, which is much larger than that of neat (10/90) blends ($10.8 \text{ }\mu\text{m}$). The blends with rough particles demonstrate a less profound growth in the droplet size. The droplet size in the presence of 2.0 wt% rough particles increases only to $23.9 \text{ }\mu\text{m}$. The promoted coalescence at high shear rates can be explained by the enriching of particles in PIB droplets, which increases the viscosity of PIB droplets^[20] and may shift the breakup-coalescence equilibrium of droplets to higher shear rates.^[35]

In terms of the shape and spatial arrangement of droplets, unlike the results obtained at low shear rates (Fig. 7), the droplets in the PIB/PDMS (10/90) blends at a higher shear rate do not display any vorticity orientation or ordered structure (Fig. 8) even at high particle concentrations. This could be understood reasonably by the smaller droplet size generated at

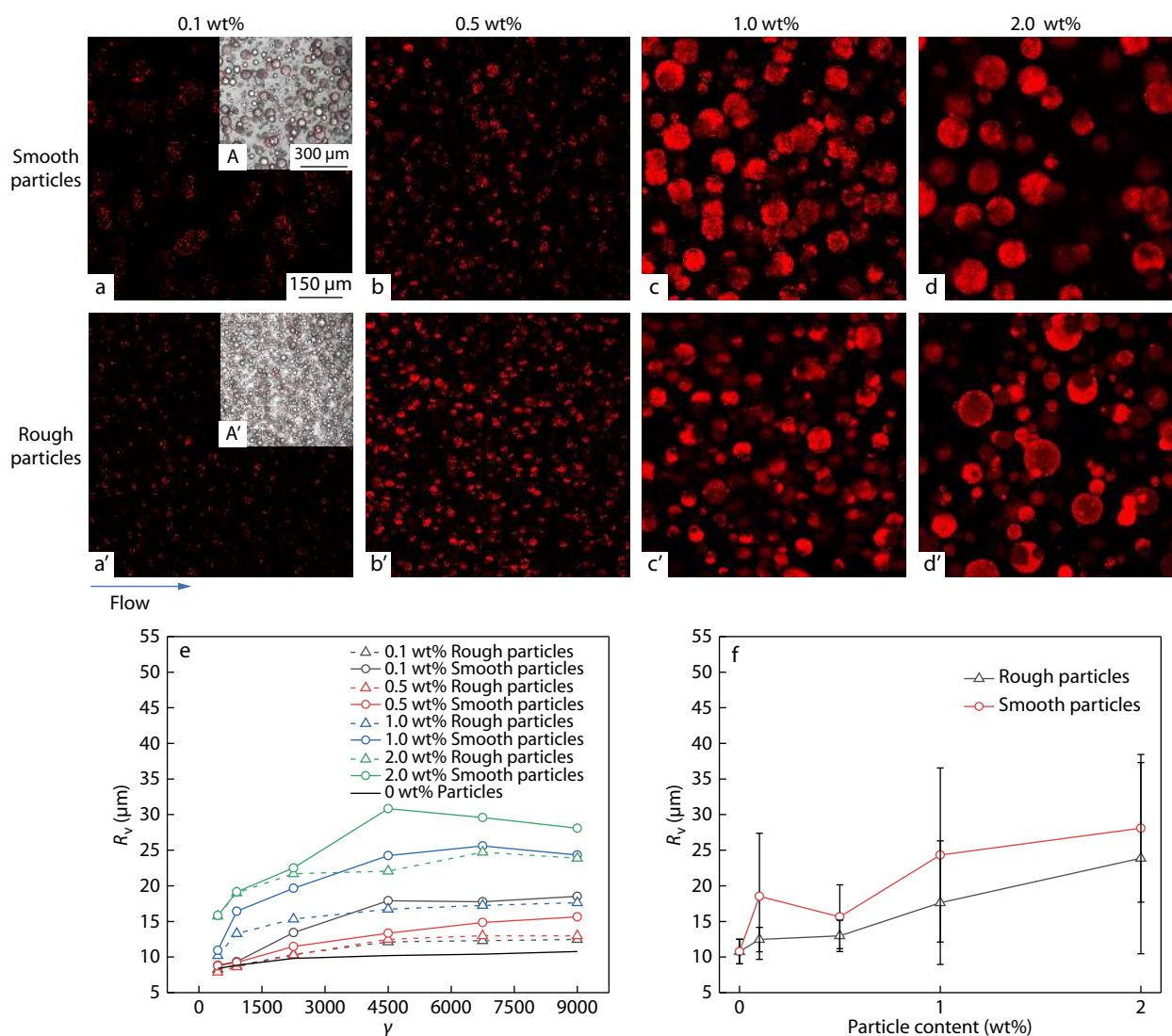


Fig. 8 Morphology of PIB/PDMS (10/90) blends with (a–d) smooth particles and (a'–d') rough particles after shearing at 0.5 s^{-1} for $1.8 \times 10^4 \text{ s}$ ($\gamma=9000$): (a, a') 0.1 wt%; (b, b') 0.5 wt%; (c, c') 1.0 wt%; (d, d') 2.0 wt%. Insets A and A' are corresponding overlay images. I–IV and I'–IV' are droplet size distribution corresponding to (a–d) and (a'–d'). The scale bar in (a) is $150 \text{ }\mu\text{m}$ and the blue arrow denotes the flow direction. The gap space of the shear cell is $200 \text{ }\mu\text{m}$. (e) Growth of droplet radius (R_v) in PIB/PDMS (10/90) blends with different rough or smooth PS particle concentrations as a function of shear strain at 0.5 s^{-1} . (f) Dependence of R_v of PIB droplets in PIB/PDMS (10/90) blends on particle content after shearing at 0.5 s^{-1} for $1.8 \times 10^4 \text{ s}$.

higher shear rates, which prevents most droplets from entering a confined state, so they cannot “fell” the confined effect of the shear wall. In the case of (90/10) blends, the preferential distribution of particles in the matrix phase does not show such viscosity enhancement and coalescence hindrance effect, and thus the blends show a strong dependence of droplet size on the change of shear rate.

CONCLUSIONS

The nanoscale surface roughness of particles plays an important role in structuring the immiscible polymer blends under the shear flow. The roughness effect of PS particles leads to slower diffusion dynamics and makes them cannot diffuse easily toward the interface of PIB/PDMS blends. This effect depends not only on the shear rate but also on the blend ratio. For (90/10) blends, rough particles result in less profound droplet coalescence and the absence of particle-bridging droplet clusters. The contribution of particle roughness diminishes under strong shear flow, thereby resulting in a similar size and spatial arrangement of droplets while slightly refining the morphology. For (10/90) blends, both types of particles promote the coalescence of preferentially wetted droplets at low and high shear rates. However, the blends with rough particles display less profound coalescence and do not present any vorticity alignment even under slow flow as observed in the blends with smooth particles. Therefore, the size and spatial arrangement of droplets in polymer blends can be controlled by adding particles with different diffusion and distribution behaviors, which can be realized by changing the roughness of particles together with shear conditions.

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