Rheology and dynamics of multi-micro-/nanolayered polymer systems: A multi-scale investigation of interfaces/interphases and confinement effects

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Abstract. Interphase developed at the polymer–polymer interface critically determines macroscopic properties of multilayered polymer systems from multilayer coextrusion process. However, a deep understanding of the interfacial phenomena involving rheology and dynamics is still lacking. Herein, we present some fundamental studies on interface/interphase in multi micro-/nanolayered systems to better control the coextrusion process and final properties. Firstly, the interdiffusion kinetics and development of diffuse interphase were investigated by both rheological modeling and experiment based on a model miscible polymer pair (***Macromolecules*, 2013,** 46, 276-299). We demonstrated the effects of a robust diffuse layer–layer interphase on melt flow behaviors. Molecular rheology theories including tube model and convective constraint release (CCR) indicated a dilated tube diameter in the interphase, suggesting its weak entanglement intensity due to unfavorable interchain entanglements and its readiness to disentanglement under large deformations (***J. Rheol.*, 2016**, 60, 1–23, ***Soft Matter*,** 2017). Relating the interdiffusion, we further confirmed this finding by the existence of dynamic heterogeneity of the studied system in both terminal and segmental scales, together with locally structural heterogeneity (***Soft Matter*, 2016,** 12, 3252-3264). Secondly, multi-micro-/nanolayered films alternating of this model miscible polymer pair were obtained by forced-assembly coextrusion process (***ACS Appl. Mater. Interfaces*, 2018**, 10, 29019-29037). Influence of geometrical and macromolecular confinements on interfacial dynamics and rheology of multilayered systems was examined. We also revealed presence of multiple diffuse interphases and the corresponding formation mechanisms. Diffuse interphase even significantly altered the melt rheological behaviors of multilayered systems subjected to fast extensional flows, especially for nanolayered systems. Interdiffusion of fast-mode mechanism and the triggered interphases were responsible for those changes in extensional rheology. Such nonlinear rheological properties strongly dependent on interdiffusion are particularly important for industrially coextruded polymers that are subjected to processing involving extensional flows, such as coextrusion blowing. Our findings reveal the role of interdiffusion and diffused interphases in morphology/microstructure development, dielectric relaxations, dynamics and rheology of multilayered polymers. This work will offer some new enlightenment to improve the processability of multilayered polymers and target properties by controlling the interfacial diffusion, namely, the amount and length scale of interphases among layers.

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