Nonlinear shear rheology of entangled polymers diluted in oligomer matrix

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Abstract

The relaxation dynamics of molten monodisperse polymers are well described by tube-based theories in the linear viscoelastic regime. However, predicting their behavior under large shear rates is still a challenge today and many questions still need to be addressed. The objective of the present work is to investigate the role played by the molecular environment on the stretch and orientation state of linear polymer chains under strong shear flow. To this end, we diluted a long linear polystyrene (PS) of molar mass of 820 kg/mol into PS oligomers, at different concentrations varying from 5wt% to 50wt%. The transient shear behavior of these samples was then measured for a large range of shear rates (corresponding to Rouse-Weissenberg numbers from 0.09 to 90), using a strain controlled rotational rheometer equipped with a cone-partitioned plate geometry. From the comparison between the data and their analysis by means of the TMA tube-based model, we attempt at understanding and rationalizing the different scaling laws observed in the shear thinning regime, as well as the transient stress response and in particular the importance of the stress overshoot as function of the long chains concentration. We examine how shear flow affects the capability of the chains to create new entanglements and how this property depends on concentration. Our results provide a framework for developing a rigorous understanding the nonlinear shear rheology of entangled polymers.