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Non-Gaussian polymer dynamics on the scale of an entanglement strand

The research on Non-Gaussian (NG) dynamics in polymer melts has been largely focused on the decaging of polymer segments near the glass transition and was quantified in terms of the NG parameter. Recently, based on simulations, theoretical investigations led to quantitative determination of the NG parameter for the linear entangled polymers. We present a study on diffusion of short poly(ethylene oxide) (PEO) in a strongly entangled PEO melt¹. Within the entanglement volume, the dynamics of the short PEO was found significantly Non-Gaussian. The COM MSD are sub-diffusive at short times until they have reached the size of the reptation tube d . Then, a crossover to Fickian diffusion takes place indicating cooperative chain motion within the entanglement volume d^3 . Thus, the host dynamics within the tube is not only cooperative but also significantly Non-Gaussian.

Quantitative comparison of the dynamic structure factors from unentangled and strongly entangled melts was performed for the poly(butylene oxide) (PBO)². The spectra from entangled PBO can be very well described by the dynamic structure factor based on the concept of local reptation, including the Rouse dynamics within the tube and allowing for NG corrections. Comparing quantitatively the spectra from both polymers leads to the surprising result that their spectra differ only by the contribution of classical Rouse diffusion for the low molecular weight melt. The sub diffusive component is common for both the low and high molecular weight PBO melts, indicating that in both melts the same interchain potential is active, thereby supporting the validity of the Generalized Langevin Equation approach.

1. M. Kruteva et al. *Macromolecules*, 54(24), 11384 (2021)
2. M. Kruteva et al. *ACS Macro Letters*, 13(3), 335 (2024)

Session

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