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Cooperative tracer chain dynamics in highly entangled polymer melts

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Using neutron spin echo spectroscopy the dynamics of short unentangled tracer chains in the melt of a highly entangled polymer matrix has been investigated [1]. Independent of the tracer chain length the center of mass mean square displacement (MSD) is sub-diffusive at short times and crosses over to Fickian diffusion at longer times. The obtained diffusion coefficients on the nm scale agree very well with results from macroscopic methods but the dependence on the tracer chain length strongly deviates from the Rouse expectation. For all the different tracers the cross-over to normal diffusion always occurs at the same MSD, which corresponds to the tube diameter of the entangled host. This observation cannot be understood within the standard reptation model where within the entanglement volume simple Rouse motion is assumed, but might be explained by cooperative chain motions, where the tracer chains move cooperatively with the host chains to an extent limited by the tube size.

[1] M.Zamponi et al., Phys.Rev.Lett. 126, 187801 (2021).

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