



Contribution ID: 46

Type: Oral

Cooperative tracer chain dynamics in highly entangled polymer melts

The dynamics of short unentangled tracer chains in the melt of a highly entangled polymer matrix has been investigated using neutron spin echo spectroscopy [Zamponi et al., Phys. Rev. Lett. 126, 187801 (2021)]. Irrespective of the tracer chain length, the center of mass mean square displacement is subdiffusive at short times and crosses over to Fickian diffusion at longer times. The diffusion coefficients obtained on the molecular length scale are in very good agreement 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 mean square displacement, 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.

Session

Soft Condensed Matter

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Session Classification: Soft Condensed Matter