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Aneesur Rahman Prize Talk: Dynamics of Entangled Polymer Melts: Perceptive from Molecular Dynamics Simulations[1]

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Twenty years ago at the APS March Meeting, Kurt Kremer and I presented the first numerical evidence from computer simulations that the reptation model of Edwards and de Gennes correctly describes the dynamics of entangled linear polymer melts. For chains longer than the entanglement length N_e, the monomers of a chain move predominantly along their own contour. The distinctive signature of reptation dynamics, which we observed, was that on intermediate time scales, the mean squared displacement of a monomer increases with time as $t^{1/4}$. Though these early simulations were limited to chains of a few N_e, they demonstrated the potential of computer simulations to contribute to our understanding of polymer dynamics. Here I will review the progress over the past twenty years and present an outlook for the future in modeling entangled polymer melts and networks. With present day computers coupled with efficient parallel molecular dynamics codes, it is now possible to follow the equilibrium dynamics of chains of length $10 - 20N_e$ from the early Rouse regime to the long time diffusive regime. Result of these simulations support the earlier results obtained on chains of only a few N_e. Further evidence for the tube models of polymer dynamics has been obtained by identifying the primitive path mesh that characterizes the microscopic topological state of the computer- generated conformations of the chains. In particular, the plateau moduli derived on the basis of this analysis quantitatively reproduce experimental data for a wide spectrum of entangled polymer liquids including semi-dilute theta solutions of synthetic polymers, the corresponding dense melts, and solutions of semi-flexible (bio)polymers such as f-actin or suspensions of rodlike viruses. We also find that in agreement with the reptation model, the stress, end-to-end distance and entanglement length of an entangled melt subjected to uniaxial elongation, all relax on the same time scale.

[1]Work in collaboration with K. Kremer, R. Everaers, C. Svaneborg, S. K. Sukumaran and N. Uchida