590d The Effect of Chain Architecture on Polyolefin Dynamics: Quasielastic Neutron Scattering and Simulation

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We present the results of a comparison between united atom simulation, explicit atom simulation and neutron scattering experiments for a series of polyolefins with differing chain architectures. Dynamic relaxation spectra for the polyolefins in consideration: atactic polypropylene (aPP), head-to-head polypropylene (hhPP), polyethylene-propylene (PEP) and polyethylene-butene (PEB) have been obtained with both time of flight and backscattering spectrometers. All four species are model polymers prepared using anionic polymerization. Although they exhibit similar energetic interactions, the variance in both the side group size and frequency causes a range of intermolecular packing characteristics. We draw correlations between this packing and the dynamic response as characterized by the self intermediate scattering function S(q,t). Simulations of these polymer melts allow a more detailed study of molecular packing and dynamics: both pair distribution functions and an isolation of various molecular motions that contribute to S(Q,t) become accessible. We address the origins of the fast and slow processes in the experimental relaxation spectra including contributions from methyl group rotations. Additionally we present the dependence of the simulation chain length and the impact of the united atom description.