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Molecular Dynamics Simulation of Entangled Vitrimer Melts

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Vitrimers are a novel class of polymeric materials that combine the mechanical robustness of thermosets with the reprocessability of thermoplastics, owing to their dynamic covalent networks[1]. While their unique viscoelastic and self-healing properties have been explored experimentally[2], a molecular-level understanding of their behavior in entangled melts remains limited. This gap is largely due to the challenges of modeling both topological constraints and dynamic bond exchange reactions in molecular dynamics (MD) simulations.

In this work, we present large-scale MD simulations of entangled vitrimer melts, explicitly incorporating dynamic bond exchange mechanisms alongside entanglement effects. Our results demonstrate how the interplay between network topology and bond exchange kinetics governs the rheological response and relaxation dynamics of vitrimer melts. We systematically investigate how varying exchange reaction rates and entanglement density affect stress relaxation, chain mobility, and network structure.

Additionally, we examine the phase separation phenomena in vitrimeric solutions using MD simulations, comparing our results with predictions from the Flory-Huggins solution theory[3]. Our simulations aim to capture the phase separation driven by thermodynamic incompatibility between dynamic cross-links and backbone monomers, in agreement with experimental observations[4].

These findings provide key molecular insights into the dynamic behavior of entangled vitrimers, bridging the gap between experimental data and theoretical models. This work highlights the power of MD simulations in unraveling the complex physics of vitrimers and underscores the need for further computational studies in this emerging field.

References:

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