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Editorial corner – a personal view

Multiscale modeling and simulations in elastomer materials: Opportunities and challenges

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Molecular dynamics (MD) simulation proves to be a powerful tool to observe and analyze the microscopic mechanism behind macroscopic phenomena at the atomic level (https://doi.org/10.3144/expresspolymlett.2015.54). The time step of numerical integration is so short that the simulation of a million steps can only reach nanosecond scale, limiting the prediction of long-term diffusion and relaxation behaviors (https://doi.org/10.1021/ma070843b1). To achieve the physical span in simulation time while maintaining atomic-level accuracy, many accelerated MD techniques have emerged.

A combination of atomistic and parameterized coarse-grained (CG) and slip-spring (SLSP) simulations have proven to be effective accelerated methods that can be applied to provide bottom-up predictions of diffusion and viscoelastic properties of long entangled polyisoprene chains, spanning from femtoseconds to seconds (https://doi.org/10.1021/acs.macromol.1c01376). Chemical specificity is preserved and critical data is transferred through all atom MD in CG models using iterative Boltzmann inversion (IBI), potentials of mean force (PMF) (https://doi.org/10.1002/adma.201403361). Simultaneously, larger length and time scales can be extended owing to several repeat units lump into a single interaction site. At highly CG SLSP level, multiple Kuhn segments of polymer chains are represented by one CG bead. The static and kinetic parameters of the SLSP model are determined and calibrated using matching the orientational autocorrelation of the end-to-end vector with the lower-level simulation models (https://doi.org/10.1021/acs.macromol.0c02583). This hierarchical simulation can be easily extended to other entangled pure systems. However, there remains a challenge in determining the interfacial interactions and quantifying the interfacial topological and dynamic properties resulting from high entangled polymer confinement and controlled by polymer-filler interaction in nanocomposites.

Alternative accelerated methodologies exploit the infrequent-event nature of the activated processes to predict state-to-state evolution that approximates what would result from an extremely long MD simulation (https://doi.org/10.1016/S1574-1400(09)00504-0). *e.g.* Parallel replica dynamics allows the simulation of strain rates as low as 10^3 s^{-1} (https://doi.org/10.1016/j.commatsci.2014.12.011). The long-term process of self-assembly and disassembly of nanostructures can be presented using metadynamics (https://doi.org/10.1073/pnas.1516652112). Umbrella sampling has been employed to study nanomechanics (https://doi.org/10.1021/jp303418a). These activated-event descriptions are widely applied to such varied processes as vacancy diffusion, pipe diffusion along a dislocation core, dislocation climb and impurity clustering, void growth, and surface morphology evolution (https://doi.org/10.1146/annurev.

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matsci.32.112601.141541). Whereas these methods will fail to provide significant acceleration when large numbers of states are connected by low barriers.

Furthermore, the efficiency of MD simulations is also based on the interplay of software and hardware that are nowadays moving to hybrid GPU-based technologies (https://doi.org/10.1016/j.cpc.2010.12.026). Multiple popular software packages include some form of GPU-acceleration support, with Gromacs performing optimally (https://doi.org/10.1002/wcms.1444). Massive parallelism on GPU is more efficient for solving spatial scale problems because it can be partitioned based on supercomputer architectures. However, the dynamic processes occurring sequentially in time series with the characteristics of serial arrangement are insensitive to simple parallelism.

The ultimate goal of MD simulation is to achieve quantum mechanical precision-massive scale-laboratory time scale. In conclusion, the combination of neural network potential function with a supercomputer, GPU support, and accelerated algorithm summarized above makes multiscale simulation feasible.

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