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LECTURE

"Assessing the Rheo-mechanical Properties of Polymer/Solid Interphases via Atomistic Simulations"

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Assessing the Rheo-mechanical Properties of Polymer/Solid Interphases via Atomistic Simulations

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The resistance of polymer-matrix nanocomposites to failure depends on several factors such as the elastic constants of the filler and matrix phases, the strength of the filler-matrix interface, and the geometry of the filler particles. We will discuss a generic, rare event-based simulation strategy, for predicting and understanding failure in materials.[1] We will first investigate failure in graphene-based polymer-matrix nanocomposites under shear stresses. The resistance to failure is quantified by conducting a series of shear deformation experiments with molecular dynamics (MD) for various temperatures (T) and shear stresses (σ_{zx}), as a function of the chemical constitution of the filler (i.e., pure graphene, graphene with vacancies and graphene oxide). The (T, σ_{zx}) measurements are fitted to an extended Boltzmann-Arrhenius-Zhurkov kinetic equation[2] which allows obtaining the macroscopic material constants for each matrix/filler combination; the latter can be fed as input to mesoscale finite element models. The aforementioned methodology will be then applied for predicting the buckling kinetics of 2D materials under compressive forces.[3–6]

Subsequently, we will change gears, and strafe our attention to the effect of surface nanopatterning on the flow of high molar mass polymers past solid surfaces.[7,8] Most constitutive laws for describing the rheology of polymer fluids invoke the no-slip boundary condition; that is, the velocity of the fluid near the surface is zero relatively to the frame of reference of the latter. In many situations, this approximation has been deemed inadequate, especially when considering the flow on non-Newtonian fluids.[9,10] Slip depends of several molecular factors such as the thickness of the cavity,[11] the strength of adhesion of the polymer to the interface in relation to the cohesive energy of the polymer,[12] the geometry/roughness of the interface,[13] and more. The atomistic model for Couette flow of PE past Au surfaces agrees with the experimental measurements by Hatzikiriakos and Deakym.[9] It is demonstrated that the orientation and thickness of the nanopatterned surfaces play a key role regarding the crystallization and reorganization tendencies of the chains at the interface, slip promotion/suppression, and flow conditions. By quantifying slip as a function of pattern orientation, thickness, and distribution one can obtain insights for designing interfaces with tailor-made rheological properties.

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