Fluctuating Hydrodynamics of Reactive Liquid Mixtures

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We discuss a low Mach number formulation of the equations of isothermal fluctuating hydrodynamics for multi-species reactive mixtures of miscible liquids, as well as associated computational algorithms and applications. We use a general Maxwell-Stefan-based formulation of multi-species diffusion, and include a quasi-incompressible constraint that allows the density to change with composition. For chemical reactions, we show that the traditional diffusion process description of thermal fluctuations via the chemical Langevin equation does not correctly reproduce the large deviation theory of the chemical master equation (CME) [J. Chem. Phys. 142, 224107 (2015)]. However, a description based on Poisson noise, which can be efficiently simulated using tau leaping, is consistent and no harder to simulate than the Langevin description. While the CME is well known for ideal dilute solutions of reactants in a solvent, we discuss how to generalize it to more general non-ideal mixtures. For spatially-extended systems, we combine a fluctuating hydrodynamics description and numerical model for diffusion with a stochastic description of reactions, and discuss some inherent difficulties associated with a purely local description of reactions [J. Chem. Phys. 146, 124110 (2017)]. We also account for advection and the coupling between velocity and concentration fluctuations, and discuss some difficulties in handling the fact that reactions typically change the local density of the fluid. We present numerical studies of equilibrium fluctuations, giant nonequilibrium fluctuations, and reactive gravitational instabilities in reactive liquid mixtures