Combined Monte Carlo and Molecular Dynamics Methods To Study The Long-Time-Scale Evolution of Particulate Matter and Molecular Structures Under Reactive Flow Conditions.

Alison Kubota, Leonard Harris, Christopher J. Mundy, William J. Pitz, Carl Melius and Charles K. Westbrook
Lawrence Livermore National Laboratory, Livermore, CA 94551, USA

Maria-Jose Caturla
Department de Fisica Aplicada Universitat d'Alicant, E-03690, Alicant, Spain

Introduction:

One of the largest challenges in computational modeling is the development of new computational methods and capabilities for studying molecular-scale structures over very large time-scales. In particular, there is great interest in understanding the nucleation and growth of carbon soot particles as well as their fate in the atmosphere. Gasoline and diesel burning engines as well as coal-burning power plants are some of the sources of atmospheric carbon soot. Carbon soot and particulate matter in general was found to be the leading factor responsible for cancer risk from toxic air pollution. Thus, it is important to understand the detailed physical and chemical processes that lead to particulate matter formation under reactive conditions. Previously, there have been no predictive tools available to model the heterogenous gas-surface processes with detail.

We have recently developed and implemented a new computational tool to time-integrate the detailed structure of atomistically resolved surfaces and nanostructures driven by chemical and physical kinetic rule-based rate expressions. Fundamental chemical and physical processes such as chemical reactions, surface adsorption and surface diffusion are performed using a non-lattice continuous-space kinetic Monte Carlo scheme and

driven by user-defined rule-based kinetic rate expressions, while atomic structure relaxations and processes very far from thermal and structural equilibrium are approached using classical atomistic dynamics methods. We demonstrate the sensitivity of particle evolution to chemical and physical kinetic mechanism using a parallel implementation of the combined Monte Carlo and molecular dynamics code. We demonstrate the strong role of molecular oxygen and oxygenated species in both the flame and atmospheric environments. Additionally, we apply this combined method to initiation and propagation of chemically-driven shockwaves, demonstrating the multi-time and multi-equilibrium scales that are spanned within a single integrated framework.