We have studied thermal- and shock-induced decomposition and subsequent reactions in various energetic materials (PETN, RDX, HMX) and high explosives (HE) as well as composite double base propellants (Al/Al₂O₃ nanoparticles embedded into HE matrix such as RDX or PETN) using nonequilibrium molecular dynamics (MD) with the first principles-based reactive force field ReaxFF. Studies of initiation of reactions in energetic materials in condensed phase and their sensitivity to heat or impact stimuli are of interest for developing efficient rocket propellants, munitions, etc. with increased safety and survivability while retaining or improving performance. Such simulations are essential to understand atomistic mechanisms of condensed phase exothermic chemistry and detonation. Development of solid composite propellants and prediction of their performance depending on composition and nanostructure requires understanding of detailed kinetics and combustion mechanism of nanoparticles as well as their melting and burning rates at the gas/surface interface. The key concepts that enable ReaxFF to describe such processes with chemical reactions are: 1) the use of partial bond orders (a many body contribution based purely on atomic positions) to describe covalent interactions, 2) environment dependent atomic charges (updated at every step during the dynamics) to describe electrostatics, and 3) extensive validation by quantum mechanics calculations. We developed simulation techniques:

- to investigate shock sensitivity of energetic materials and mechanisms of their decomposition under different conditions of shock or impact loading,
- to predict specific impulse and combustion kinetics of nitramine/aluminum composites from atomistic-scale simulations,
- to determine the effects of crystal structure, composition, and molecule conformation on the sensitivity, decomposition rates, and combustion kinetics,
- to study the nonlinear elastic response of HE crystals under highly anisotropic (e.g. uniaxial) compression at the shock front and validate the ReaxFF for such high pressure conditions.

Here we test this methodology in simulation of different energetic materials (RDX vs. PETN) and propellants (RDX/Al₂O₃/Al vs. RDX RDX/Al₂O₃/Al) as well as different initiation processes (heat-up vs. shock compression).