2014 AIChE Poster Abstract

Title: Discovering chemistry with an *ab initio* nanoreactor

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Abstract: Heterogeneous catalysis, combustion chemistry, and many other essential processes are comprised of interconnected networks of individual reactions. Detailed models of the reaction network are valuable for understanding these processes and optimizing them for desired characteristics (e.g. selectivity for a specific product), but they require knowledge of the individual reaction pathways that are highly difficult to characterize experimentally. Here we show results from an *ab initio* nanoreactor, a new computational method to discover reaction pathways in these complex systems.[1]

The *ab initio* nanoreactor is a large-scale molecular dynamics simulation of chemical reactions that discovers new molecules and reaction mechanisms without preordained rules or hypotheses. The simulations are driven by quantum chemistry and accelerated with the *TeraChem* software running on graphics processing units (GPUs), which greatly expands the system size and time scale. The simulations are analyzed using a machine learning approach that automatically recognizes reaction events, performs accurate energy refinements to obtain thermochemical data and activation energies, and uses this to generate a kinetic model for computing reaction rates.

To further increase the reaction discovery by several orders of magnitude, we perform accelerated nanoreactor simulations driven by reactive force fields. The thermochemical data and activation energies from the *ab initio* nanoreactor simulations are used to parameterize the reactive force fields. The model parameterization is performed using ForceBalance, a method for automatically deriving highly accurate force field parameters[2, 3].

Here we show discovered reaction pathways in several diverse systems. The first simulation investigates the polymerization of acetylene at high density (Figure 1); the results show formation of long chain hydrocarbons and aromatic species including benzene. The second simulation is an idealization of the classic Urey-Miller experiment, and the results show new pathways for forming the amino acid glycine from primitive compounds proposed to exist on the early Earth. We also present new pathways for long chain hydrocarbon synthesis on Fischer-Tropsch catalysts and soot particle formation from polycyclic aromatic hydrocarbons in flames.

References:

[1] L.-P. Wang, A. Titov, R. McGibbon, F. Liu, V. S. Pande and T. J. Martínez. “Discovering chemistry with an ab initio nanoreactor”, *under review*.

[2] L.-P. Wang, V. S. Pande and T. J. Martínez. “Building force fields – an automatic, systematic and reproducible approach, *J. Phys. Chem. Lett.* **2014**, 4, 1885-1891.

[3] L.-P. Wang, T. Head-Gordon, J. W. Ponder, P. Ren, J. D. Chodera, P. K. Eastman, T. J. Martínez and V. S. Pande, “Systematic improvement of a classical molecular model of water”, *J. Phys. Chem. B* **2013**, 117, 16236-16248

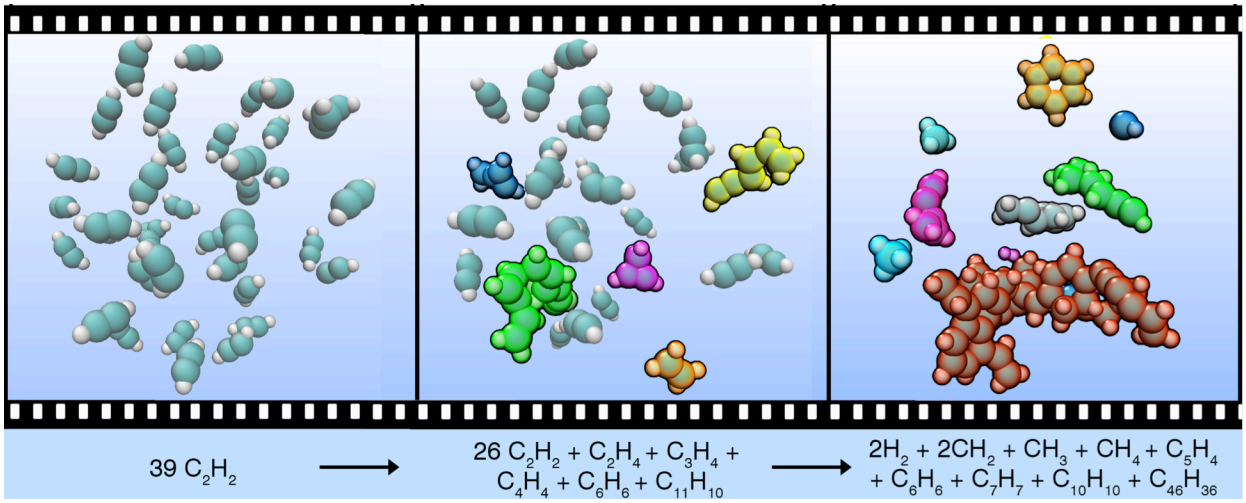


Figure 1: Timeline of a nanoreactor simulation trajectory. *Left*: Simulation begins with acetylene molecules (C = teal, H = white). New molecules are automatically highlighted with colors to indicate observed reactivity. *Middle*: Simple products appear first, including short polymeric species (green, yellow) as well as ethylene (orange) and cyclopropene (violet). *Right*: At longer simulation times the molecular size distribution becomes considerably wider; more than half of the atoms form a large molecule containing multiple aromatic rings (red, bottom right). A long-lived benzene molecule is also formed (gold, top right).