A Multi-Scale Approach to the Simulation of Lignocellulosic Biomass

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Project Goals: In concert with the imminent increase in the Department of Energy's leadership supercomputing power to the petaflop range, the objective of this project is to develop multiscale methods for extending the time- and length-scales accessible to biomolecular simulation on massively parallel supercomputers. This project also aims to apply the developed multiscale approaches to obtain an understanding of the structure, dynamics and degradation pathways of extended cellulosic and lignocellulosic materials. Information from multiscale simulation, when closely integrated with experiment, will provide fundamental understanding needed to overcome biomass recalcitrance to hydrolysis.

The multiscale simulation methods, ranging from highly accurate quantum mechanical (QM) methods to coarse-grained molecular dynamics (MD), have been used to obtain an understanding of the structure, dynamics and degradation pathways of extended cellulosic and lignocellulosic materials using capability high-performance simulation. Treating solvent implicitly is a critical multiscale concept, and to this end we have developed a parallel order-*N* Poisson-Boltzmann solver (1,2) and a treecode-based Generalized Born electrostatic solvation method (3). Furthermore, a statistical mechanical multiscale approach was derived that was found to describe the temperature dependence of cellulose fiber stability (7), and complementary Fragment Molecular Orbital (FMO) and all-atom MD simulations have been performed of cellulose crystal structures.

A range of coarse-grained models have been developed on various length scales (8). Based on the effective fragment potential, coarse-grained models have been developed for benzene and methanol and for glucose in solution. The coarse graining has also involved development and application of Boltzmann inversion techniques and of the "REACH" (Realistic Extension Algorithm via Covariance Hessian) methodology, which maps results obtained from atomistic MD simulations onto models for larger-scale, coarse-grained MD.

The physical properties of lignocellulosic biomass derived using the multiscale methodologies serve as a basis for interpreting an array of biophysical experiments (4-5), and, in particular, the simulation models derived will be used to calculate and interpret a variety of neutron-scattering properties. To aid in the interpretation we have developed "dynamical fingerprinting" as a means of reconciling the multiple time scales accessed by experiment and simulation (6). This combination of simulation and experiment will eventually lead to a description of the physicochemical mechanisms of biomass recalcitrance to hydrolysis, and thus will aid in developing a strategy as to how rationally to overcome the resistance.

References:

1. Lu B*, Cheng X*, Huang J, McCammon JA. AFMPB: An Adaptive Fast Multipole Poisson-Boltzmann Solver for Calculating Electrostatics in Biomolecular Systems. *Comput Phys Commun.* 2010, 181, 1150

2. Zhang B, Lu B, Cheng X, Huang J*, Pitsianis N, Sun X, and McCammon JA. Mathematical and Numerical Aspects of the Adaptive Fast Multipole Poisson-Boltzmann Solver. *Communications in Computational Physics* 2011, in press

3. Xu Z*, Cheng X* and Yang H. Treecode-based Generalized Born method. *J Chem Phys.* 2011 134(6):064107

4. Prinz, JH, Chodera, J, Pande V., Swope W, Smith JC and Noe F. Optimal use of data in parallel tempering simulations for the construction of discrete-state Markov models of biomolecular dynamics. *J. Chem. Phys.* 2011, 134, 244108

5. JH Prinz, M. Held, JC Smith & F. Noe. Efficient Computation, Sensitivity and Error Analysis of Committor Probabilities for Complex Dynamical Processes. *Multiscale Mod. & Sim*. **9**, 545-567 (2011).

6. F. Noe, S. Doose, I. Daidone, M. Lollmann, M. Sauer, J.D. Chodera & JC Smith. Dynamical Fingerprints: Probing individual relaxation processes in biomolecular dynamics with simulations and kinetic experiments. *Proc. Natl. Acad. Sci. U.S.A.* 2011, 108(12):4822-7

7. Klein H*, Cheng X*, Smith JC* and Shen T*. Transfer matrix approach to the hydrogen-bonding in cellulose Iα fibrils describes the recalcitrance to thermal deconstruction. *J Chem Phys.* 2011, 135, 085106

8. Srinivas G, Cheng X and Smith JC*. A Solvent-Free Coarse Grain Model for Crystalline and Amorphous Cellulose Fibrils. *J. Chem. Theo. Comp.* 2011, 7 (8), 2539–2548

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