Catching molecular motion at just the right time

September 21 2011

Marina G. Guenza and Ivan Lyubimov of the University of Oregon have developed a mathematical formula that returns pivotal information into course-grain simulations. Credit: University of Oregon

University of Oregon researchers have devised a mathematically rich analytic approach to account for often-missing thermodynamic and molecular parameters in molecular dynamic simulations.

The new approach, which returns atomistic-level data into the time frame of the macroscopic world, is detailed in a paper appearing online ahead of regular publication in the journal *Physical Review E*. The method is all about timing, says Marina G. Guenza, professor of theoretical physical chemistry, and may help reduce trial-and-error experimentation required in manufacturing when such information is

missing.

Molecular dynamic simulations are indispensable tools -- a natural partner of experiments and theory -- that help scientists understand the properties of new materials and processes by providing a view at the resolution of atoms. Simulations expedite the development of new materials by showing how those with a specific atomistic structure behave in various conditions, for example when they are strained or frozen.

Simulations of polymers and biological systems have been used since the 1990s. That effort has focused on the short-time motion of macromolecules described in atomistic detail, which, in addition to plastics and glasses, also applies to DNA and proteins, Guenza said.

However, modelers remove critical pieces of information, such as atom-level activity, to scale back simulations to cover only generic components and access longer times in an accessible simulation run. This technique provides helpful but incomplete data about behavioral responses, Guenza said. Simulations in which atomistic information is withheld are called coarse-grained models.

"These are big molecules," she said. "They move slowly. It is difficult to set up a simulation where the atomistic definition is included and still be able to see things happen on the long time scale, which can be really important. Coarse-graining allows one to simulate macromolecules for longer time, but, because some information is eliminated, the motion measured is unrealistically fast."

Entropy -- a loss of thermodynamic energy -- and surface friction are lost in these simulations, she said. Simulations at the atomistic level depict motion occurring in femtoseconds. (A femtosecond is a millionth of a nanosecond; a nanosecond, a billionth of a second.)

To understand what happens in macroscopic systems, you have to look at movement over longer periods of time -- over seconds, says Ivan Lyubimov, a UO doctoral student in chemistry and lead author. "When you try to simulate a second's worth of information at the atomistic level, with all the details included, it might take one or two years for the computer to run the simulation, and you'd still have errors due to numerical algorithms," he said.

Guenza and Lyubimov looked at simulations where thousands of macromolecules of polyethelene are represented as interacting soft particle, i.e. a coarse-grained model, and applied an original theory that refocuses the information missing in the simulations.

Guenza -- a member of the UO's Institute of Theoretical Science, Materials Science Institute and Institute of Molecular Biology – and Lyubimov first detailed the basics of their theoretical formula in 2010 in the *Journal of Chemical Physics*.

Their "first-principle" approach looks at the loss of energy, due to the change in entropy, caused by the coarse-graining of the molecule in simulations. Coarse-graining also affects the surface of molecules in simulation, so the formalism accounts for the loss of friction as well.

"We were able to show that if you run your simulation with less detail, we can correct for these factors, and you'll produce the correct motion -- the dynamics -- of the real system," Guenza said. "We have done a lot of tests with different experiments and simulations, and our method works pretty well. No one else has been able to do this with a theoretical solution."

The method, the authors wrote, is different from others currently in use, because it is analytical rather than numerical. It removes the need for separate, time-consuming atomistic simulations to account for missing

information obtained from coarse-grained simulations.

"Parameters can be varied for different systems, depending on the molecule size, density and temperature," Lyubimov said. "You can make realistic predictions for the type of material you want to study, at much less expense. You don't have to know all of the details, but you do need a certain number of parameters based on the chemical structure that you want to study."

Provided by University of Oregon