New developments of fast computational methods for first principles geochemical and geophysical simulations

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Conventional methods of directly simulating the behavior of complex strongly interacting atomic systems (molecular dynamics, Monte Carlo) have provided important insight into the behavior of gases, fluids, and solids of geochemical and geophysical interest. The even wider application of these methods is limited by the difficulty of developing molecular level representations of potential interactions to capture complex chemistry commonly encountered in these systems (reactions, polarization, etc.). Static quantum chemistry methods have provided a means to calculate reactive mechanisms in cluster approximations to mineral systems. These methods are limited to small atomic sizes and generally cannot be applied to problems in which dynamics play a role. In this talk new developments in the implementations of methods to simultaneously simulate the electronic structure and molecular dynamics of nanoscale materials will be described (*ab-initio* molecular dynamics, AIMD). These methods, implemented into NWChem software, calculate of inter-atomic forces directly from the fast solution of DFT equations for very large systems and, therefore, avoid problems of force development limiting the application of MD. This talk will focus on the development and application in three areas:

(1) the implementation the PAW method an all-electron plane-wave method; *application* to the solvation structure of transition metal ions in solution;

(2) the development of a QM/MM method for simulations of large systems; *application*: simulation of enzyme reactions;

(3) the development of a plane wave implementation of exact exchange; *application:* the localization of charge in hematite.

The talk will focus on the fundamentals of these methods and the realities in terms of system size, computational requirements and simulation times that are required for their application.