Extending the scope of force field methods: reactivity and multiple scales

P. Raiteri[1] S. Piana[1] J. D. Gale[1] A. G. Stack[2]

[1]Nanochemistry Research Institute/Department of Chemistry, Curtin University, PO Box U1987, Perth, WA 6845, Australia

[2]Chemical Sciences Division, Oak Ridge National Laboratory, PO Box 2008, MS-6110, Oak Ridge, Tennessee 37831, United States

For complex dynamical systems, such as interfaces and surfaces in contact with solution, force field methods are often the only computationally feasible technique. However, even these methods face limitations for many important problems of interest relating to the description of the energy surface or the length- and time-scales involved. This talk will give examples of work within our group to address both of these challenges:

1) Reactivity: Here the proton conductivity of Y-doped $BaZrO_3$ will be examined. A comparison of two reactive force field approaches to describe proton diffusion will be made based on ReaxFF [1] and Empirical Valence Bond (EVB) theory [2] parameterised from first principles. This allows proton trapping by point defects and grain boundaries to be addressed, while connecting to experimental diffusion rates. Comments regarding some of the issues facing reactive force fields for interfacial systems involving water will be made.

2) Multiscale: The use of kinetic Monte Carlo methods makes it possible to overcome some of the problems associated with length- and time-scale associated with force fields. It has been shown that the crystallisation and evolution of particles in solvents can be simulated in this manner leading to agreement with direct experimental comparisons at the micron-scale [3]. However, the challenge of determining the rate constants remains a problem for many cases. Here the combined use of multiple rare-event theories to determine accurate rate constants for multistep pathways will be presented [4].

[1] A.C.T. van Duin, B.V. Merinov, S.S. Han, C.O. Dorso and W.A. Goddard III, *J. Phys. Chem. A* **112**, 11414 (2008).
[2] P. Raiteri, J.D. Gale and G. Bussi, *J. Phys. Cond. Matter* **23**, 334213 (2011).
[3] S. Piana, M. Reyhani and J.D. Gale, *Nature* **438**, 70 (2005).
[4] A.G. Stack, P. Raiteri and J.D. Gale, *J. Am. Chem. Soc.* **134**, 11 (2012).