Application for a TSM-DTC funded PhD studentship

Please complete this form electronically
and submit to Lilian Wanjohi (l.wanjohi@imperial.ac.uk)
by Friday January 8, 2010

1st supervisor
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Please complete the following:

1. Project title: Droplets: from molecular nanoclusters to the atmospheric aerosols

2. Project abstract (≤ 200 words please and please add 1 or 2 key references)

The focus of this project is to obtain an understanding of nanoscale liquid droplet formation at a molecular level. Despite the importance of nucleation for climate processes, particularly air pollution and cloud formation (which have key effects on the global radiative balance of earth) *little information exists on the precise molecular-level mechanisms*. This is a significant issue for climate modeling which must take nucleation effects into account.

We will focus on nanoscopic droplets of sulfuric acid and water, the release of sulfur dioxide into the atmosphere (industrial plumes, coal fired power plants) produces sulfuric acid, one of the most important species for new-particle formation in the atmosphere. There is an impact via acid rain on vegetation, buildings, water sources, air quality and human health. However, sulfuric acid has also been suggested as a tool for future geo-engineering of the earths atmosphere where sulfur dioxide injections into the stratosphere would produce a veil of particulates that would scatter solar radiation back into space hence enabling real climate control.

In this project we will gain an understanding of the processes by which droplets form from gas phase molecules, this will impact on climate models and potential geo-engineering schemes. The droplets studied will span a wide range in size; starting from just a few acid molecules and a less than complete water coordination shell, they can grow to become large nanoscale atmospheric particulates. Detailed information will be extracted from quantum chemical calculations, and used to undertake large-scale atomistic MD simulations from which we will obtain macroscopic scale insights.
T. Kurten and H. Vehkamaki, Advances in Quantum Chemistry, Vol 55, pp. 407-427, 2008
P. Crutzen, Climatic Change (2006) 77: 211–219

3. What is the multi-scale nature of the project? (≤ 100 words please)
This projects spans a quantum mechanical (QM) to macroscopic level description. Using QM we will explore the fundamental chemistry and speciation of the system. A detailed description of the intermolecular potentials will be obtained, this information will be used in classical molecular dynamics (MD) simulations to study phase behavior and in particular dynamics of the droplets, analyzing fluctuations and interfacial tensions. Properties obtained from MD will be incorporated in free energy calculations for the prediction of the critical nucleus, stability of drops, and nucleation rates. Ultimately, this information will allow us to explore and understand the free energy landscape of these systems, and thus the stability of aggregates and nucleation kinetics of the system.

4. How do the expertises of the supervisors complement each other? (≤ 100 words please)
All four supervisors have a specialization at different levels of the multi-scale approach.
Hunt: Quantum mechanics (electronic structure theory: density functional and higher levels) and experience with ab-initio MD particularly of ion solvated aqueous clusters and ions solvated in the condensed phase.
Ford: MC, MD and continuum free energy modeling of clusters; aerosol physics.
Muller: Large scale MD atomistic simulations, including fluid-solid interfaces.
Jackson: Fluctuation analysis and interfacial property estimation from simulation, classical nucleation theory and surface free energies.

5. Is there a self-contained 12-week MSc project that would usefully initiate this PhD project? (If the answer is no the project will not be offered as an MSc project)
Yes! The student will be given a "taster" from each stage of the project, applying the relevant techniques to a single very simple model system and comparing their results to those in the established literature. Perform high level quantum chemical calculations on different isomers of a sulfuric acid-water pair. These will then be used to benchmark a selection of DFT methods, the best of which will be used to study larger clusters. (4 weeks)

The best method will be selected to generate an effective semi-empirical intermolecular potential (e.g. a Mie(n,m) potential with either point charges or discrete association sites) via tabulation or a fit. (2 weeks) The developed potential will be compared to other published effective force fields (OPLS-type potentials) using classical MD to explore the gas phase thermodynamics and liquid-vapour phase equilibria.(4 weeks) If time permits, the MD data will be analysed for dynamic fluctuations, surface tensions and data to be input into more course grained models. (2 weeks) The literature evaluation will give the student an appreciation of the current state-of-the-art in this field. The full project implements this process for a larger range of species involving more complex interactions.