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
Name: George Jackson
CID (IC only): 170163
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2nd supervisor
Name: Patricia Hunt
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3rd supervisor (optional)
Name: Mike Gillan
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4th supervisor (optional)
Name: Erich A. Müller Imperial College , Department of Chemical Engineering
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Please complete the following:

1. Project title: Understanding and Modelling CO_2 capture: From quantum mechanics to chemical and phase behaviour

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

The focus of this project is a multiscale description of the phase behaviour of systems of CO_2 and aqueous amines and/or ammonia. Absorption of CO_2 with aqueous solutions of amines and/or ammonia are seen as the most probable technology for large-scale separation of CO_2 from combustion flue streams for subsequent sequestration. The project thus concerns problems of great importance for the environment, and therefore for society. This project is at the forefront of current research because complex reactions, physical chemistry and

phase behaviour of CO_2 in amine and ammonia solutions are poorly understood. It is important that we understand the chemistry driving these systems as this will allow us to develop and control a robust capture process that can be viably implemented at an industrial scale.

The ultimate aim of this project is to develop a pseudo-empirical macroscopic model founded on quantum mechanics which will provide a fundamentally well grounded description of the phase behavior of CO_2/amine/ammonia systems. The model will be refined against experimental data available from pilot plants and collaborators in Chemical Engineering (Paul Fennel).

3. What is the multi-scale nature of the project? (≤ 100 words please)
The projects spans in a bottom-up fashion from the quantum level up to the macroscopic description. From QM we would like to explore the chemistry of the reactions, i.e., gain an understanding of the number and nature of the species formed. A detailed description of the intermolecular potentials will be obtained from these QM calculations. This information may be expressed in terms of simple effective pair potentials which can be further used in classical MD simulations and perturbation theories (SAFT) for a description of the phase behaviour of these systems. Ultimately, the theoretical framework can be used within a process modelling environment to describe the separation of CO_2 in amine solvents.

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 density functional theory and higher level), experience in the chemistry of such systems.
Gillan: : Extensive experience over past 15 years in DFT m.d. simulation on liquids and solids.
Muller: Experience in large-scale MD and MC atomistic simulations of gases and liquid mixtures.
Jackson: Perturbation theories and equation of state for fluids, continuum models

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 molecular chemical calculations on a pair (or more) of ammonia molecules with focus on self-association (hydrogen bonding). These results 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-vapor phase equilibria. (4 weeks) If time permits, the potential may be matched to an equation of state and compared to experimental data. (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.