Multi-scale modelling of CO_2 transport in flexible microporous materials

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The capture or sequestration of CO_2 is often envisaged using complex materials such as kerogen (geological sequestration) or activated carbon (capture), which have in common that they are mainly composed of a carbonaceous microporous phase. This phase, which is responsible for the high CO_2 retention capacity, is very often flexible, which facilitates the diffusion of CO_2 within it. However, the dynamic couplings between the fluid molecules and the carbonaceous matrix are poorly understood and can have a major influence on transport properties. The aim here is to develop spectroscopic measurement techniques coupled with multi-scale models based on molecular dynamics simulations to gain a better understanding of these properties and thus better predict the characteristic times associated with CO_2 capture/sequestration.

This thesis proposes a joint theoretical/experimental study under the supervision of Thierry Tassaing (molecular spectroscopy group) for the experimental part and Amaël Obliger (theoretical chemistry and modelling group) for the theoretical part. This project is part of Amaël Obliger's research into the properties of kerogen (transport, thermodynamics, poromechanics) on the nanometre scale using molecular simulations [Berthonneau_PNAS_18, Obliger_JPCB_19, Potier_LAN_23]. We propose here to enrich and compare them with in situ IR/Raman spectroscopy experiments developed by Thierry Tassaing [Champeau_JSF_14, Dubois_JSF_18, Ongkasin_IJP_20]. The objectives of this study are:

– To propose and solve a multi-scale transport model in which diffusion within the microporous kerogen matrix is dynamically coupled to the poromechanical relaxation of this matrix, taking into account the swelling induced by adsorption.

– To simultaneously measure the evolution of the CO_2 concentration in kerogen as well as that of the swelling of kerogen by in situ IR/Raman spectroscopy during adsorption/desorption experiments.

The student will implement an innovative method for measuring the thermodynamic and transport properties of kerogen-gas mixtures using in situ infrared and Raman microscopy. This method also provides information on adsorption and swelling kinetics on micrometre- to millimetre-sized samples [Champeau_JSF_14, Dubois_JSF_18, Ongkasin_IJP_20]. To enable comparison between the results of molecular simulations and spectroscopy, a multi-scale coupled diffusion/poromechanical transport model will be developed during the course of the thesis, which will be able to take existing results from molecular simulations as well as those obtained during the course of the thesis as input parameters. The thesis work would therefore be divided into two tasks, with the first involving experimental measurements, which would take up around 1/3 of the duration of the thesis, and the remainder of the course being dedicated to the theoretical work.

Candidates should preferably already have experience in molecular simulations and computational physics/chemistry. Funding for this PhD can be obtained from the University of Bordeaux via the Excellence Grants programme managed by the Chemical Sciences doctoral school.

Applications (CV, motivation letter, and transcripts of marks from Master's years) must be sent before **May 2nd** to thierry.tassaing@u-bordeaux.fr and amael.obliger@u-bordeaux.fr.

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