cecam Centre Européen de Calcul Atomique et Moléculaire	**Workshop Scientific Report** Please do not repeat the program (unless there were last-minute changes) or the initial description - we already have this material.
Title	Computational Carbon Capture
Organizers	Berend Smit Sofia Calero Thijs J.H. Vlugt

Scope of the workshop (one-two paragraphs)

The scope of the workshop was to explore how computational methods can help the discovery of new materials for carbon capture and sequestration. At present several groups are working on the development of techniques to study the properties of different materials in the context of carbon dioxide capture. Important material properties to study are the adsorption isotherm, the adsorption and desorption enthalpy, the maximum loading of carbon dioxide, the diffusion coefficient of carbon dioxide in the medium, and (if applicable) the viscosity of the medium. Examples include carbon dioxide adsorption in ionic liquids or in crystalline porous materials such as zeolites, zeolitic imidazolate frameworks (ZIFs) or metal organic frameworks (MOFs). However, the number of possible materials is very large, this hold both for ionic liquids and porous structures. Therefore, it is difficult, if not impossible experimentally synthesize all possible structures. With this workshop we wanted to bring together these experimental and theoretical groups to discuss how computational methods can help in these screening efforts. The following questions are of particular importance:

1. Are the computational techniques sufficiently advanced that we can compute the properties that are relevant for CO2 capture? Where are new methods required?

2. Are the intermolecular potentials sufficiently accurate that we can predict the properties of new materials?

3. How should the ideal capture material look like? What is the target property we should screen for?

Main **outcomes** of key presentations (one page)

The workshop started with the welcome talk by Prof. Thijs J.H. Vlugt, chair of the first session, who introduced the scope of the workshop. The invited papers were categorized into five sessions, were the main capture technologies, the development of computational methods and force fields and the different techniques for screening were addressed. In particular, the workshop sessions focused on:

- Main capture technologies:
a) absorbent based technologies in which a solvent captures the carbon dioxide and the energy costs are associated with the regeneration of the solvent,
b) adsorbed based technologies in which the solid adsorbent is used in a given process and the costs for it is the regeneration of the adsorbed by, for example, increasing the temperature or decreasing the pressure,
c) post-combustion carbon dioxide capture by carbonation-calcination cycle using calcium oxide as adsorbent, and
d) membrane based technologies in which a membrane is used to separate the gas molecules.

- Recent progress on computational techniques:
a) techniques able to compute the properties that are relevant for CO2 capture,
b) development of new simulation methods when required,
c) development intermolecular potentials accurate enough to predict the properties of new materials.
d) development of QM techniques to obtain transferable classical force fields for MOFs
e) development of more coarse grained methods to predict solubilities, for example the use of the PC-SAFT equation of state in the simultaneous design of the process and the optimal solvent

- Screening and design of optimal materials for carbon capture:
a) determining of target properties for screening of materials,
b) development of pre-screening methods, especially developing methods to relate target properties to microscopic properties of the host material
c) defining the main features of the "ideal" material for carbon capture.

-Interplay between computer simulations and experiments
a) pitfalls in experimental determination of adsorption and absorption properties
b) pitfalls when obtaining force fields used in molecular simulation from experimental data

Report on selected discussions (one page)
eg. Were there interesting hints for new research? for new developments? for collaborations?

The papers presented in the workshop provided interesting hints for new research, leading to the developments on new methods and force fields and fostered new collaborations between experimental and simulation groups. A few examples are given below:

(1) new strategies for obtaining force field parameters in molecular simulations from experiments. In her presentation, Conchi Ania showed many examples of (systematic) errors in adsorption experiments, obviously leading to inaccurate force fields. This leads to discussions on how to obtain force fields from experimental adsorption data. Another issue here is that the introduction of a somewhat artificial reference state (i.e. excess adsorption) is really needed in experiments, as this complicates systematics comparisons between simulations and experiments. It would be very nice if a solutions for this could be found. For sure this will require collaboration between simulators and experimentalists.

(2) the use of ab initio methods to obtain classical force field parameters for MOFs. Presently used force fields describing the adsorption and diffusion of guest molecules in MOFs are not transferable, i.e they work for one type of MOF but not for another. The use of ab inito techniques may solve this problem. The presentations of Laura Gagliardi and Allison Dzubak initiated discussions on how to do this in the best way, leading to possible future collaborations with research groups focussing on classical molecular simulations.

(3) to determine the optimal host material for carbon dioxide capture, it is important to not only investigate the host material, but the whole process of carbon capture and storage. In his presentation, Andre Bardow showed how a physically based equation of state could be use to perform the optimization of both the host material and the process simultaneously. This works very well for adsorption, i.e using a liquid like ionic liquids or other novel solvents. It is very interesting how to extend this to adsorption, using solid hosts like zeolites and metal organic frameworks.

(4) methods for database screening. Maciej Haranczyk showed how computational approaches can be used to screen zeolites for their applications for carbon capture and storage. A quite different approach was put forward by William Schneider for ionic liquids. This initiated discussions on the optimal design strategy, clearly needing close collaborations between experimentalists and simulators.

(5) Both liquids (e.g. ionic liquids) and solids (e.g. zeolites, MOFs, ZIFs) are considered for capture and storage og carbon dioxide. Presently, itis unclear which of the two will win. A serious drawback of the use of ionic liquids is their very high viscosity and their sensitivity for water, while stability issues can become crucial for metal organic framework. A more final conclusion on which way to go would involve collaborations between bith the ionic liquid community and the porous solids community. It was very nice that representatives of both were present at the workshop.

To what extent were the **objectives** of the workshop achieved (strong points, weak points)? (one paragraph at least)

The objectives of the workshop were:

(1) To bring together the different computational groups working on carbon dioxide capture with the aim to arrive at a comparison of computational methods used in the various sub-fields.
(2) To intensify the synergy between experimental and simulation groups.
(3) To identify the targets and material properties that the host materials should have.
(4) To identify the most promising strategies for designing the optimal material for carbon dioxide capture and storage.
(5) To investigate the possibilities for applying the presently used techniques for reactive solvents.
(6) To investigate how and if a pre-screening tool can be used to find a host material with optimal properties.

We strongly feel that we achieved the defined objectives of the workshop. The following issues are also important for developing new carbon capture and storage technology:

(1) cost aspects, in particular a comparison between the costs of solid adsorption processes and liquid adsorption (amines, ionic liquids).

(2) experimental synthesis of host materials that were selected using computational techniques

Do you have suggestions for new workshops/tutorials/conferences on the topic?

A new important development is the consideration of structural transitions in the host material. As an example, the MIL-53 metal organic framework changes structure upon the adsorption of carbon dioxide, therefore leading to a large storage capacity and possibilities for an easy mechanical release of carbon dioxide. Liquid crystals could also be considered for this. The difficulty that it is very difficult to describe and predict these transitions, as well as their effects on the adsorption, desorption and diffusion of carbon dioxide in these materials. We propose to organize a separate workshop on this topic in the future.