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On the need of Force Field parametrization for a correct description of the host-guest interactions in supramolecular structures that capture and storage CO2

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Introduction
Fast rising of sea level due to ice melting at the Earth's pole or radical changes in terrestrial ecosystems are some of the disastrous consequences that may devastate the globe if human beings are not concerned about contamination and global warming. Excessive amounts of pollutants in the atmosphere is at the root of environmental issues being carbon dioxide (CO2) one of the main greenhouse gases present in the atmosphere [1]. The concentration of this gas in the air has increased by 30% since the 19th century mainly due to intensive use of fossil fuels like coal, oil or natural gas (80% CO2 emissions worldwide) and human activity. Moreover, the International Panel on Climate Change (IPCC) has predicted that CO2 levels in the atmosphere could reach up to 590 ppm by 2100 entailing the rising of almost 2 ºC in the globe [2]. These high values can be obtained in part due to the economic and industrial growth of developing nations, where energy sources with a strong carbon footprint are extensively used. For this reason humans have two challenging issues to address: to find and use a sustainable source of energy and also to reduce greenhouse emissions to achieve environmental protection [3].

At present, CO2 is collected from the atmosphere using absorption processes in which chemical reactions may or may not be involved. The most common procedures are based on the use of basic aqueous solutions of NaOH and KOH or amine scrubbing. The main drawback is the energy that requires, which translates into high economic and environmental cost. Other limiting factors of this technology are the corrosive and environmentally unfriendly character of the reagents involved, the engineering of large and complex absorption units and also the thermal degradation of solvents such amines. The urgency for materials that can be used within CO2 capture has prompted the study of several technologies such as: boron nitride nanotubes, ionic liquids, porous inorganic membranes (PIMs), polymers with light organic functional groups, inorganic-organic interface composites, covalent organic framewoks (COFs), metal organic frameworks (MOFs) and zeolitic imidazolate frameworks (ZIFs) among others tested recently [4].

We use of molecular simulations with the aim of obtaining new compounds that have improved properties over the existing ones such as: high capture capacity, high recyclability, good thermal stability and easy desorption protocols of captured gases if necessary, as well as a high selectivity in the adsoption of certain pollutants. With the aid of computer-aided simulation it is possible to study prior to the synthesis static and dynamical properties such as: the affinity for different gases, capture capacity, isotherms that govern the process, dynamics of the adsoption and time scale and also the ability to identify the degree of occupation

of the structure among other properties [5-6]. Bulky solutions of soluble compounds in a specific solvent or extended solids with periodic boundary conditions are often used.

Many of the problems that would desirable to tackle with molecular modeling (interesting properties in carbon capture and storage materials) are, unfortunately too large to be studied by means of quantum mechanics since they imply prohibitive computational cost. To deal with this problem, molecular mechanics (MM) is thus invariably used to perform calculations on systems containing a large number of atoms and to simulate systems which the smallest spatial dimension is higher than nanometres [7]. In spite of this, it is necessary to delve deeper into the potentials that are being used in force fields and reparametrize them, if necessary, to obtain a correct description of complex systems such as MOFs, ZIFs, COFs among others.

Communication Main Body

With the aim of testing the general performance of force fields, DREIDING, AMBER and UFF were selected and compared with high level ab-initio calculations. Among the force fields, we were not able to use the AMBER force field since it does not have parametrized the cumulene carbon of CO2, thus, both UFF and DREIDING have been used to perform calculations using a conventional parametrization and an extra one with charge equilibration on all atoms. The charge equilibration method (QEq) proposed by Rappé et al. That uses as input data experimental atomic ionization potentials, electron affinities, and also atomic radii was used to assign charges to all atoms in both DREIDING and UFF. Some CO2-five-membered-ring heterocycles complexes have been considered for the analysis of the intermolecular interaction potential. Electron-Donor-Acceptor (EDA) complexes between CO2-and heterocycles have been used to analyse the binding energy prediction ability of force fields.

Once the inability of the force fields to correctly describe the interactions between CO2 and heterocycles (which are the fundamental links of the structures of the carbon dioxide capture compounds) has been demonstrated, then the study and parametrization of the interaction potentials computed with high-level ab-initio methods is carried out. Two major groups of study will be considered: interaction with the main structure of the heterocycle (carbon skeleton, π-electron density, interaction with the main heteroatom etc.) in different possible configurations and the study of hydrogen bond interactions between CO2 and X-H fragments of the heterocyclic structures. All interaction curves have been parametrized according a Lennard-Jones potential.

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