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Computationally based analysis of the energy efficiency of a CO_2 capture process

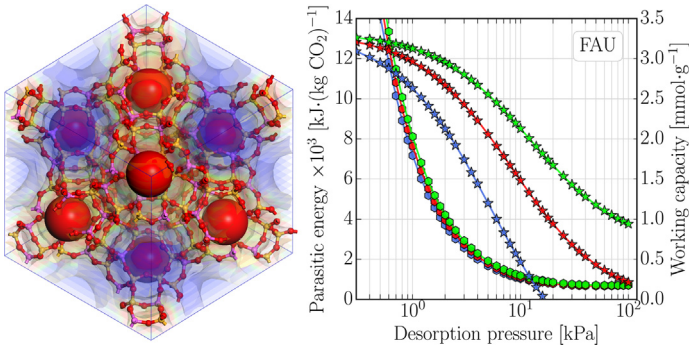
Bojan Vujic, Alexander P. Lyubartsev *

Stockholms University, Department of Materials and Environmental Chemistry, Division of Physical Chemistry, Stockholm University, SE 106 91 Stockholm, Sweden

HIGHLIGHTS

- A model for thermodynamic evaluation of the energy efficiency of CO_2 capture.
- Accounting for the working capacity, regenerability and purity of the captured CO_2.
- Determination of the most efficient desorption conditions for CO_2 capture.
- Grand Canonical Monte Carlo simulations for more than 1000 zeolite structures.

GRAPHICAL ABSTRACT



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ABSTRACT

We propose a model for thermodynamic evaluation of the energy efficiency of a CO_2 capture in a temperature-pressure swing adsorption. Major parts of this model are computational prediction of the adsorbed gas loading as a function of temperature and partial CO_2 pressure, evaluation of the energy expenses under specified conditions for the working capacity, regenerability of the sorbent and purity of the captured CO_2, as well as determination of the most optimal desorption conditions in terms of desorption pressure and temperature. The proposed model can be applied for fast evaluation of the energy costs of the CO_2 capture process with the use of both experimental or simulation adsorption data with respect to pressure and temperature. We tested this model analyzing data obtained from Grand Canonical Monte Carlo simulations for more than thousand different zeolite structures. Within our approach it is possible to evaluate a theoretical limit of the energy expenses for each specific material and to use the proposed method in screening different structures for the most efficient sorbent material from the energy efficiency point of view under specified requirements for the working capacity of the process, regenerability and purity of captured CO_2. We show that setting realistic from the industrial point of view parameters of the CO_2 capture cycle leads to substantial reduction of the number of suitable zeolite structures, and to increase of the energy penalty of the CO_2 capture compared to evaluations based on minimization of the parasitic energy only.

1. Introduction

One of the major problems the modern era is facing is a rapid increase of greenhouse gases concentration in the atmosphere. A special emphasis is placed particularly on carbon dioxide, as the main greenhouse gas. Increasing concentration of CO_2 in the atmosphere leads to global warming, and may also have a negative impact on human health (Patz et al., 2005; Milly et al., 2005; McMichael et al., 2006). Because of this reason, science and technology are facing a problem of finding new ways to fast and yet efficiently reduce CO_2 emission. Carbon capture and storage

* Corresponding author.
E-mail address: alexander.lyubartsev@mmk.su.se (A.P. Lyubartsev).

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(CCS) technology (Bacsik et al., 2016; Hedin et al., 2013; Xu and Hedin, 2016) is expected to play an important role in the reduction of greenhouse gases emissions into the atmosphere (Miles and Kapos, 2008; Paustian et al., 1998; Choi et al., 2009; Ming et al., 2016; Songolzadeh et al., 2012; Smit et al., 1986).

CCS can be defined as the separation and capture of CO_2 produced at stationary sources, followed by transport and storage in geological reservoirs or in the ocean (Damen et al., 2006). Carbon dioxide can be captured directly from industrial sources by one of three main methods: pre-combustion capture (via oxygen-blown gasification), post-combustion capture, and oxyfuel process (Songolzadeh et al., 2012; D'Alessandro et al., 2010; Benson and Orr, 2008). Post-combustion capture in power plants involves the separation of CO_2 from flue gas mixtures, followed by its compression and sequestration in geological formations (Metz et al., 2005; Massood et al., 2007).

There are several techniques for separation of carbon dioxide from flue gas mixture, such as adsorption (Martin-Calvo et al., 2014; Sethia et al., 2010; Maurin et al., 2005; Hedin et al., 2013; Akhtar and Bergström, 2011), absorption (Yan et al., 2008; Conway et al., 2015), cryogenic membrane (Pfaff and Kather, 2009; Ye et al., 2014) and micro algal bio-fixation (Pellerano et al., 2009). Practical choices can vary, but in general, adsorption can be a very good choice in terms of efficiency, possibility of large-scale applications and recycling of the adsorbent material. Considering the overall cost of CO_2 capture, adsorption process in comparison with other methods is the most efficient one (Pellerano et al., 2009; Liu et al., 2010).

Adsorption relates to uptake of sorbate molecules onto the surface or in pours of a solid adsorbent, to which they adhere via two types of forces, weaker van der Waals and dipole forces in the case of physisorption, or stronger covalent bonding in the case of chemisorption (Hedin et al., 2013). Absorption in contrasts with adsorption is the process where sorbate molecules dissolve into the bulk of the material itself (Bhown and Freeman, 2011). The most used physisorbents, such as active carbon (Ribeiro et al., 2008; Berlier, 1997), metal-organic frameworks (MOFs) (Bastin et al., 2008; Bao et al., 2015; Lee et al., 2015; McDonald et al., 2015; Verdegaal et al., 2016) and zeolites (Liu et al., 2010; Cheung et al., 2012; Siriwardane et al., 2005; García-Pérez et al., 2007; García-Sánchez et al., 2009), are better options than chemisorbents, such as amine-modified mesoporous silica (Bacsik et al., 2010; Hedin et al., 2010) due to relatively easy regeneration of the sorbent material by the change of temperature or pressure.

Whatever technique is used for CO_2 capture, it inevitably requires certain amount of energy, which exert an additional load on the power plant and which is called for this reason *parasitic energy*. Within existing techniques parasitic energy can account for about 30% of the total energy produced by a power plant (Bhown and Freeman, 2011; Lin et al., 2012) and reducing this energy cost is of primary importance for further development of CCS technology. Not surprisingly, much effort in recent years, in both experimental (Jensen et al., 2012; Harlick and Tezel, 2004) and computational (Gomez-Alvarez et al., 2016; Babarao and Jiang, 2008; Deroche et al., 2008; Krishna and Long, 2011; Yazaydin et al., 2009; Kim et al., 2012; Hasan et al., 2013; Matito-Martos et al., 2014; Matito-Martos et al., 2015) studies have been devoted to investigation of the adsorption properties of various materials like zeolites and MOFs, as well as the energy costs and efficiency of CCS.

In recent computational studies (Lin et al., 2012; Berger and Bhown, 2013; Huck et al., 2014) a model for computation of parasitic energy in a typical temperature–pressure swing process has been suggested which included two main contributions: thermal energy to desorb CO_2 and thus regenerate the adsorbent, and compression energy to compress captured CO_2 to the transportation pressure (conventionally 150 bar). These energy contributions can be evaluated from the adsorption isotherms determined in computer simulations of considered materials. By varying thermodynamic conditions of the desorption state, a minimum of the parasitic energy (per mol or per gram of the captured CO_2) can be found. The most optimal desorption conditions are defined by the minimum of the parasitic energy. Computations can be done for any material with known atomic structure, thus the method becomes suitable for screening many possible candidates for efficient CO_2 adsorbents.

The model described in works (Lin et al., 2012; Huck et al., 2014) predicted the theoretical minimum of the parasitic energy from the basic relationships of thermodynamics. However, that methodology does not consider several important parameters of the adsorption-desorption cycle which are important from the industrial applications point of view: working capacity (which is defined as the difference between adsorption and desorption loadings and equals to amount of CO_2 captured in a single cycle), purity of the captured CO_2 and regenerability (percent of CO_2 remaining in the sorbent). Our computations show that in many cases working conditions defined by the minimum of the parasitic energy alone correspond to a very small working capacity (which would require many adsorption/desorption cycles for the same amount of CO_2 captured and thus make the whole capturing process slow and expensive), to a low regenerability of the sorbent framework and to poor purity of the captured CO_2. Amount of the parasitic energy related to the realistic working conditions can thus be highly underestimated. We therefore propose in this work to complement the search of the optimal conditions for the minimum of the parasitic energy by additional conditions for the minimal acceptable working capacity, purity and regenerability.

Another problem we address to is how to make the computer search for the most suitable adsorbents more efficient. For precise computation of the parasitic energy, it is required to have a large number of fugacity steps (desorption points) for each adsorption isotherm, as well as a large number of adsorption isotherms in a reasonable temperature range. Such computations require hundreds of simulations for each considered material which makes the screening process expensive in terms of CPU time. Here we propose a thermodynamical model (based on the Langmuir adsorption model), which expresses the adsorption load of CO_2 and N_2 as a function of partial gas pressures and temperature. Parameters of this function can be fitted by running simulations for about ten temperature–pressure points, which allows reducing the number of computations (relative to the computation of the whole set of adsorption isotherms) by more than one order of magnitude. Furthermore, within the model the parasitic energy itself is expressed as an analytical function of the parameters which facilitates evaluation of the parasitic energy, and makes it possible to use the model for experimental data. Using this methodology we are able to fast and yet accurately evaluate the minimum of the parasitic energy and thus find the most optimal desorption conditions (in terms of desorption temperature and pressure) for working capacity, regenerability and purity of captured CO_2 specified in advance.

2. Materials and computational methods

2.1. Zeolites as CO_2 adsorbents

Among many materials which can be used as adsorbents (Hedin et al., 2013; Ribeiro et al., 2008; Cheung et al., 2012; Hedin et al., 2010; Xu and Hedin, 2016), zeolites have shown promising results for separating carbon dioxide from flue gas mixtures in the pressure swing adsorption (PSA) (Chue et al., 1995), temperature swing adsorption (TSA) (Merel et al., 2008; Ntiamoah et al., 2016) as well