



# Particle-scale CO<sub>2</sub> adsorption kinetics modeling considering three reaction mechanisms



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## ABSTRACT

In the presence of water (H<sub>2</sub>O), dry and wet adsorptions of carbon dioxide (CO<sub>2</sub>) and physical adsorption of H<sub>2</sub>O happen concurrently in a sorbent particle. The three reactions depend on each other and have a complicated, but important, effect on CO<sub>2</sub> capturing via a solid sorbent. In this study, transport phenomena in the sorbent were modeled, including the three reactions, and a numerical solving procedure for the model also was explained. The reaction variable distribution in the sorbent and their average values were calculated, and simulation results were compared with experimental data to validate the proposed model. Some differences, caused by thermodynamic parameters, were observed between them. However, the developed model reasonably simulated the adsorption behaviors of a sorbent.

The weight gained by each adsorbed species, CO<sub>2</sub> and H<sub>2</sub>O, is difficult to determine experimentally. It is known that more CO<sub>2</sub> can be captured in the presence of water. Still, it is not yet known quantitatively how much more CO<sub>2</sub> the sorbent can capture, nor is it known how much dry and wet adsorptions separately account for CO<sub>2</sub> capture. This study addresses those questions by modeling CO<sub>2</sub> adsorption in a particle and simulating the adsorption process using the model. As adsorption temperature changed into several values, the adsorbed amount of each species was calculated. The captured CO<sub>2</sub> in the sorbent particle was compared quantitatively between dry and wet conditions. As the adsorption temperature decreased, wet adsorption increased. However, dry adsorption was reduced.

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## 1. Introduction

Anthropogenic emissions of carbon dioxide (CO<sub>2</sub>), one of the most critical species in greenhouse gases (Pachauri and Reisinger, 2007), is considered as the main culprit to global climate change (Rogner et al., 2007). In the United States, about 83% of greenhouse gases are produced from the burning of fossil fuels (EIA, 2006a), which emits about 2 billion metric tons of CO<sub>2</sub> annually (EIA, 2006b) and amounts to roughly two-thirds of the total U.S. power sector CO<sub>2</sub> emissions (Ciferno et al., 2009). To mitigate the emission problem, various government agencies and industries have been developing carbon capture, transport, and long term sequestration technologies (Figueroa et al., 2008).

Generally speaking, three types of approaches are being examined for CO<sub>2</sub> capture from fossil-fuel-based power plants: (1) pre-combustion, (2) post-combustion, and (3) oxy-combustion. In pre-combustion capture, CO<sub>2</sub> is removed from the fuel stream before it is burned, and H<sub>2</sub> is sent to a combustion turbine. For example, a polymer-based membrane with high H<sub>2</sub>/CO<sub>2</sub> selectivity was reported by Berchtold (2006). Supported ionic liquid

membranes (Ilconich et al., 2007) and lithium silicate-based sorbent (Li et al., 2006) were also developed to select CO<sub>2</sub> from coal-derived syngas. In oxy-combustion, fuel is burned with a mixture of nearly pure O<sub>2</sub> and recycled flue gas, and its flue gas contains mainly CO<sub>2</sub> and water. After removing water by condensation, highly concentrated CO<sub>2</sub> can be easily collected. The drawback of this approach is extracting O<sub>2</sub> from air, which is a costly process. Meanwhile, in post-combustion capture, CO<sub>2</sub> is captured from flue gas produced by the combustion process. Notably, existing power plants can be easily retrofitted with the equipment required for this process (Figueroa et al., 2008). Thus, it is considered the most feasible approach to implement in the near future.

As a post-combustion capture process, aqueous amine-based solvent capture is commercially available, but it is a highly energy-intensive and corrosive process. When the process was applied to a power plant, it led to an 80% increase in the cost of electricity (Rao and Rubin, 2002). Unlike the costly aqueous amine-based capture, CO<sub>2</sub> capture using amine-impregnated solid sorbent is attracting researchers' attention with the potential to reduce energy consumption, improve regeneration, enable greater capacity, offer selectivity, and provide ease of handling (Samanta et al., 2012). The calcium looping process is another post-combustion technology, which exploits the reversible gas–solid reaction between calcium oxide (CaO) and CO<sub>2</sub> to produce calcium carbonate (CaCO<sub>3</sub>)

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## Nomenclature

$A$	amplitude of reaction
$Am_0$	initial concentration of amine in sorbent
$C$	mass concentration of species ( $\text{mol m}^{-3}$ )
$c_p$	specific heat ( $\text{J kg}^{-1} \text{K}^{-1}$ )
$D$	diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ )
$d_{\text{pore}}$	pore size (cm)
$d_p$	diameter of particle (m)
$\Delta H$	enthalpy of reaction ( $\text{J mol}^{-1}$ )
$\Delta S$	enthalpy of reaction ( $\text{J mol}^{-1} \text{K}^{-1}$ )
$E$	activation energy of reaction ( $\text{J mol}^{-1}$ )
$G$	generation/consumption rate ( $\text{mol m}^{-3} \text{s}^{-1}$ )
$K$	equilibrium constant of reaction
$R$	universal gas constant ( $8.314 \text{ J mol}^{-1} \text{K}^{-1}$ )
$R$	reaction rate ( $\text{mol m}^{-3} \text{s}^{-1}$ )
$r$	radial direction (m)
$h_i$	convection mass transfer coefficient of species $i$ ( $\text{m s}^{-1}$ )
$h_T$	convection heat transfer coefficient ( $\text{W m}^{-2} \text{K}^{-1}$ )
$i$	species index
$M$	molecular weight ( $\text{kg mol}^{-1}$ )
$Nu$	Nusselt number
$P$	ambient pressure (Pa)
$Pr$	Prandtl number
$Re$	Reynolds number
$Sc$	Schmidt number
$Sh$	Sherwood number
$T$	Kelvin temperature (K)
$t$	time (s)

### Superscripts and subscripts

$d$	dry adsorption of $\text{CO}_2$
$i$	species
$j$	reaction
$k$	space index of particle
$p$	water adsorption
$S$	on outer surface
$w$	wet adsorption of $\text{CO}_2$

### Greek

$\alpha$	coefficient of finite difference equation in mass transfer
$\beta$	coefficient of finite difference equation in heat transfer
$\varepsilon$	porosity
$\kappa$	conductivity ( $\text{W m}^{-1} \text{K}^{-1}$ )
$\rho$	density ( $\text{kg m}^{-3}$ )
$\mu$	viscosity ( $\text{Ns m}^{-2}$ )
$\sigma$	conductivity ( $\text{S m}^{-1}$ )
$\nu$	stoichiometry

(Abanades et al., 2004). The technology also shows a good prospect for capturing  $\text{CO}_2$  effectively (Manovic and Anthony, 2010; Broda and Muller, 2012).

Modeling and simulation studies have been performed on several carbon capture systems for improved performance at different scales. For example, the  $\text{CO}_2$  absorption/stripping process with aqueous monoethanolamine (MEA) was modeled based on thermodynamics and reaction rates (Freguia and Rochelle, 2003). The researchers analyzed process variables to find operating conditions at a low-steam requirement. To find the best configuration of the  $\text{CO}_2$  stripper, the performance of several alternative designs was simulated and compared (Oyenekan and Rochelle, 2007).  $\text{CO}_2$

absorption using a membrane contactor also was studied via modeling and experiment (Hoff et al., 2004). The absorption rate was calculated with changing temperature, liquid velocity, and  $\text{CO}_2$  partial pressure. The two-dimensional diffusion-reaction model closely predicted the experimental results. A computational fluid dynamics (CFD)-based model for  $\text{CO}_2$  stripping also was developed (Ghadiri et al., 2013). Using this model, the effect of liquid velocity and temperature on stripping performance was investigated. Studies using CFD-based simulation also examined the feasibility of converting existing coal-fired power plants to oxy-combustion (Tigges et al., 2009).

In addition, various modeling and simulation studies regarding  $\text{CO}_2$  capture by solid sorbents have been performed. The  $\text{CO}_2$  adsorption behavior of amine-functionalized mesoporous silica was modeled using chemical interaction with the amine group and physisorption on the adsorbent's surface (Serna-Gurerrero et al., 2010). The model was applied to several sorbents, and the  $\text{CO}_2$  adsorption isotherm of each adsorbent was predicted. In zeolite imidazolate frameworks, the effect of functional group structure on  $\text{CO}_2$  adsorption was investigated using molecular modeling (Morris et al., 2010). It showed that polarizability and structural symmetry were the key factors for high  $\text{CO}_2$  uptake. Modeling was also used to evaluate the performance and cost of amine-based  $\text{CO}_2$  capture systems (Rao and Rubin, 2006). The most cost-effective strategies and key factors in controlling  $\text{CO}_2$  levels were proposed, with a fluidized bed reactor considered as the most promising design for  $\text{CO}_2$  capture by solid sorbents (Yang and Hoffman, 2009). In this design, heat and mass transfer and chemical reactions in the reactor depend on the interactions of gas and solid phases in the bed (Smolders and Baeyens, 2001). To calculate the two-phase flow in the fluidized bed, several models have been developed (Kato and Wen, 1969; Kunii and Levenspiel, 1990; de Souza-Santos, 2007). When fluidized-bed hydrodynamics is combined with a  $\text{CO}_2$  adsorption reaction, device-scale simulations as such can provide more detailed information for improving the reactor design. However, the CFD-based device-scale  $\text{CO}_2$  capture predictions depend on accurate models in both the particle-scale reaction kinetics as well as in multi-phase flow simulations.

At the particle scale,  $\text{CO}_2$  adsorption by an amine sorbent was simulated using a shrinking core model to elucidate the relative importance of pore diffusion and surface chemical reaction in controlling the rate of reaction (Monazam et al., 2011). Lee et al. (2011) proposed a phenomenological model for the adsorption kinetics of amine-impregnated mesoporous sorbent in the presence of water, and the model was fitted to experimental data obtained from thermogravimetric analyses. Recently, Xu et al. (2012) proposed a generalized kinetic model for heterogeneous gas–solid reactions with the introduction of a distinctive layer between reacted and not-yet-reacted regions.

In this study, adsorption phenomena are modeled on the particle scale based on three fundamental reactions occurring in the amine-impregnated mesoporous sorbent in a  $\text{CO}_2$  rich environment. The model calculates on the particle scale (1) how different adsorption reactions progress; (2) how the reactions depend on each other; and (3) the amount of each species adsorbed on the sorbent particle resulting from the reactions. Similar particle-based models have been developed and used in several other research areas, i.e., metal sorption with resins (Juang and Lin, 1995), olefin polymerization (McKenna and Soares, 2001), and ozone uptake (Smith et al., 2002), but only for single reaction. In the current study, three different reactions occur simultaneously in the amine-based sorbent particle, assisting each other in one way and hindering in another, which makes particle-scale  $\text{CO}_2$  adsorption prediction more difficult. With the successfully developed particle scale reaction kinetics model and the numerical solution algorithm presented here, mass transfer and adsorption phenomena resulted via different reaction

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