

# A flexible and robust model for low temperature catalytic desorption of CO<sub>2</sub> from CO<sub>2</sub>-loaded amines over solid acid catalysts

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## ABSTRACT

A numerical model for catalyst-aided carbon dioxide (CO<sub>2</sub>) desorption from CO<sub>2</sub>-loaded aqueous amines solution has been developed. The model includes a hot water-heater and considers phase separation at the top of the desorption column. The model was validated with experimental data obtained from an integrated CO<sub>2</sub> capture pilot plant which used 5 M monoethanolamine (MEA) solution with two industrial catalysts, namely, HZSM-5 and  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. The model considers the presence of electrolytes and multi-component mass transfer as well as both the physical and chemical contribution of the catalyst in aiding the process. The data obtained from model simulation were in good agreement with the experimental data in terms of CO<sub>2</sub> production rates with an absolute average deviation of approximately  $\pm 8.9\%$ . The simulation slightly over-predicted the CO<sub>2</sub> production rate at the low temperature regime (75 °C) and under-predicted the CO<sub>2</sub> production rate at the high temperature regime (95 °C). The developed model provides a range of capabilities for further studies of the catalytic CO<sub>2</sub> desorption process.

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## 1. Introduction

The capture and storage or utilization of carbon dioxide (CO<sub>2</sub>) from combustion gases is a key technological option to tackle the problem of global warming and climate change. Among the various options for CO<sub>2</sub> capture, post-combustion using reactive solvents is considered to be the most mature and viable technology technically as it can easily be retrofitted into existing plants. Consequently, it has even seen its first commercial scale deployment in Boundary Dam, near Estevan, Saskatchewan, Canada with quite a number of projects underway in other parts of the world. However, this technology suffers a major drawback involving its high-energy requirements for desorption of CO<sub>2</sub> from the amine after absorption. The energy usage of the desorption step in the capture process represents about 80% of the energy required for the post combustion CO<sub>2</sub> capture plant (Oyenekan, 2007). It is known that a capture plant utilizing 5 M MEA with 90% CO<sub>2</sub> removal efficiency can reduce the thermal efficiency of a coal-fired power plant from about 40% to 30% (Davison et al., 2014).

Until recently, the CO<sub>2</sub> desorption process has received less attention, and efforts to alleviate this problem has been focused on the combination of process integration and solvent improve-

ment. One of the new innovative approaches with the potential to improve the efficiency of the desorber is the replacement of the packing material in the desorber with solid acid catalyst (Idem et al., 2011). The presence of the catalyst in the desorber can serve as both a high surface area packing material and a chemical facilitator for faster and easier reversion of the ions to CO<sub>2</sub>, thereby allowing CO<sub>2</sub> to be desorbed at temperatures below 100 °C (Idem et al., 2011; Liang et al., 2016; Shi et al., 2014). In order to study the performance of this technology in a power plant integrated with a CO<sub>2</sub> capture plant as well as to achieve energy requirements reduction using this technology, the desorber should be a primary focus. As a tool, a thoroughly validated model is essential. Only a few studies have been reported in terms of modeling of desorption of CO<sub>2</sub> from CO<sub>2</sub>-loaded amine solutions. A few are as outlined in the work of Tobiesen et al. (2008). However, work is yet to be reported on modeling with respect to catalyst-aided CO<sub>2</sub> desorption.

The idea of catalyst-aided desorption process is to utilize hot water as heating medium instead of steam because of the ability for desorption to occur at temperatures below 100 °C. However, at a temperature below 100 °C and pressure of 1 atm, the reboiler is unable to produce enough vapour for heat and mass transfer in the desorber column, and therefore becomes redundant. Consequently, the conventional CO<sub>2</sub> desorption process is modified to exclude a reboiler but to include a water heater as shown in

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## Nomenclature

$a$	effective interfacial area, $m^{-1}$	$\phi$	catalyst volume fraction bed
$A$	area of column, $m^{-2}$	$\varphi$	volumetric holdup, $m^3/m^3$
$c$	molar concentration, $kmol/m^3$	$\rho$	bulk density of catalyst mixture, $kg/m^3$
$dz$	differential change in height of column, $m$	$\sigma$	electrical potential, $C/mol$
$D_{ij}$	binary diffusivity, $m^2/s$	$\delta$	film thickness, $m$
$E_a$	activation energy, $J/mol$	$\lambda$	thermal conductivity
$F$	Faraday's constant ( $9.65 \times 10^4$ ), $C/mol$	$\eta$	dimensionless film coordinate
$L, G$	liquid and gas molar flows respectively, $kmol/s$		
$H$	molar enthalpy, $J/mol$		
$K_i$	phase equilibrium constant, dimensionless	<b>Superscripts</b>	
$k_o$	rate constant, $m^3/kmol \cdot s$ for non-catalytic reaction and $m^3/kmol \cdot s \cdot g \text{ cat}$ for catalytic reaction	$B$	bulk
$N_i$	molar mass flux, $kmol/m^2 \cdot s$	$I$	interface
$Q$	molar heat flux, $J/m^2 \cdot s$	$BL$	bulk liquid
$R_i$	reaction rate, $kmol/m^3 \cdot s$		
$R_G$	gas constant (8.414), $J/mol \cdot K$	<b>Subscripts</b>	
$T$	temperature, $K$	$i, j$	component indices
$x_i, y_i$	liquid and gas mole fractions	$G$	gas phase
$z_i$	ionic charge	$L$	liquid phase
		$LG$	liquid-gas
		$e, k, c$	equilibrium, kinetic and catalytic reactions
		<i>mixture</i>	mixture of 6 mm inert marble and catalyst
<b>Greek letters</b>			
$\mu_i$	chemical potential, $J/mol$		
$\nu_i$	stoichiometric coefficient of component $i$		

Fig. 1. The consequence of this modification is that none of the commercial process simulators used in modeling post combustion  $CO_2$  capture (such as Aspen Plus (Freguia and Rochelle, 2003; Zhang et al., 2009), Promax (Ahmadi, 2012; Luo et al., 2009), and Protreat (Cousins et al., 2011)) – is able to model the process. This is because they have been configured to handle at least either two inlets and outlets (without reboiler, which is usually the case of absorption or some types of reactive stripping) or 1 inlet and 2 outlets (with reboiler, which is usually the case of stripping or desorption) or are simply not customizable. Also, the commercial

simulators have not been well programmed to handle very small flows, especially gas or vapour flows in the desorber column, and therefore encounter difficulties in converging.

The purpose of this work is to develop a validated model that is capable of predicting the performance of the catalytic desorber as well as the conventional desorber, and subsequently use it to understand how the design variables affect the operation of the catalytic  $CO_2$  desorber. The model is validated with pilot plant data utilizing 5 M MEA with varying weights of HZSM-5 and  $\gamma-Al_2O_3$  catalysts in the desorber.

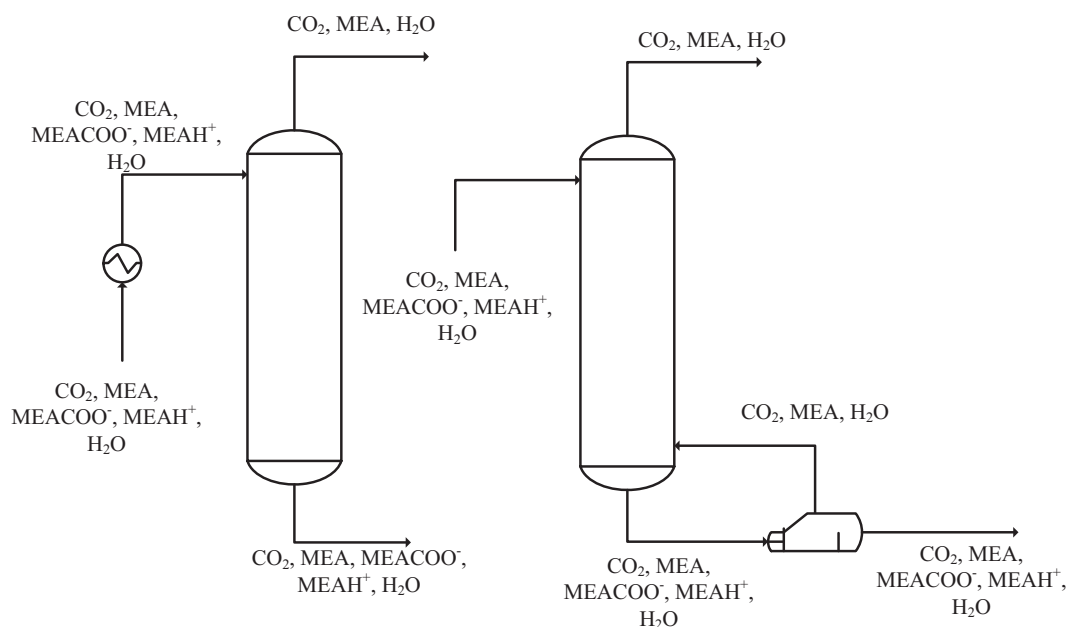


Fig. 1. Process configuration of the desorption process (Left: Low temperature catalytic process; Right: Conventional (commercial installation) desorption process with reboiler).

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