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Adiabatic modelling of CO_2 capture by amine solvents using membrane contactors

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ABSTRACT

The modelling of CO_2 chemical absorption using hollow fiber membrane contactors is addressed. A one-dimensional, multicomponent adiabatic model for the CO_2 absorption using an aqueous solution of monoethanolamine is established. The model is validated using both, laboratory and pilot-scale experiments. The simulation results are compared to those from an isothermal model in order to investigate the influence of heat release on contactor performance. When industrial relevant operating conditions are applied, the adiabatic simulations show significant axial temperatures peaks, up to 30 °C. Correspondingly, local gas-phase vapor molar fractions values of up to 0.4 are attained. If compared to simulations from an isothermal model, deviations of about 60% were obtained, thus clearly demonstrating the necessity of adiabatic modelling under industrial conditions. Intensification factors comprised of between 2 and 10, for external fiber radiuses in the range of 300–100 μm are attained. The mass transfer coefficient is varied from 10^{-4} to 10^{-3} m s^{-1} which corresponds to experimentally observed values of microporous membranes that are presumably resistant to liquid breakthrough. However, wetting remains a major problem in microporous as well as composite membranes, as capillary condensation is likely to occur.

1. Introduction

Post-combustion carbon dioxide capture is a key issue for achieving greenhouse gas emission reductions. Gas–liquid absorption using aqueous monoethanolamine (MEA) solutions in packed columns is currently considered as the most adequate technology for post-combustion applications. Two major challenging targets are aimed in order to meet both, technical and economic constraints: the decrease of process energy requirement through novel solvent or heat integration approaches, and the decrease of equipment size through process intensification. An alternative technology, the hollow fiber membrane contactor (HFMC), shows promising intensification potential, because its specific contact area of 1500–3000 m^{-1}, values far above those encountered in conventional packed columns (about 200 m^{-1} [1]).

Experimental and theoretical researches have been published on HFMC for CO_2 capture using amine solutions, addressing material, mass transfer and process design issues. However, in these studies the performance of the process has been essentially evaluated considering fresh amine solvent (non-loaded), and low amine conversion leading to isothermal behavior. Moreover water transfer has been neglected. These conditions are convenient in laboratory research but are not relevant in an industrial context. The present work is intended to fill the gap: a multicomponent adiabatic model for CO_2 capture in a membrane contactor under industrial relevant conditions is developed.

The paper is structured as follows: published isothermal and adiabatic studies are discussed in Section 1. In Section 2, the overall system characteristics are presented in detail. Section 3 exposes the modelling approach which comprises of physico-chemical, thermodynamic, kinetic and mass- and heat transfer aspects. Simulation results are presented and discussed in Section 4. Finally, the model results are compared to available, non-isothermal experimental data.

1.1. Literature overview

General literature reviews concerning the use of HFMC in CO_2 reactive absorption are presented in [2,3]. An increase of the specific CO_2 flux, when using HFMC has been obtained by numerous authors. For microporous membranes, wetting was considered to be the main problem of this technology.

A literature review focusing on modelling issues is given in [3] and [4]. Process models of various complexities have been

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developed considering isothermal conditions and neglecting water transfer. Simple models based on an average overall mass transfer coefficient have been suggested [5]. The principal assumption of these models is the invariance in conductance for each of the gas-side, membrane- and liquid-side, over the entire fiber length. They cannot be applied to operating conditions in which the liquid-side conductance varies widely over the reactor length [6]. For this case, one dimensional models based on series resistance approaches were proposed, considering varying gas- and liquid-side conductances [6]. Simultaneously, supported by the rapid development of CFD facilities, two dimensional models were published [4,7,8]. These models are based on the resolution of the convective diffusion equations coupled with the equations of the chemical reactions, using cylindrical geometry coordinates, and applied to a binary feed gas mixture of CO_2 and N_2.

In a previous paper [4], a comparison of one and two dimensional approaches assuming isothermal behavior and neglecting water transfer was performed for CO_2 capture with amine solution using membrane contactors. The aim was to identify the most efficient model strategy. The results showed comparable simulation results, with a maximum relative deviation of 2.2%, thus indicating the quality of the 1D approach.

To our knowledge, few research papers tackle the adiabatic multi-component modelling of HFMC for chemical absorption of CO_2 [9–13]. Hoff et al. [9,10] developed and validated a two-dimensional diffusion-reaction model in a relatively large range of operation conditions. It is however not clear whether industrially relevant conditions were attained. Moreover, no temperature profiles were shown. The authors considered that in HFMC with reversible chemical reactions, the liquid-phase product diffusion is the limiting factor. For scaling purposes, they pointed out that 2D models are too complicated and that 1D models are preferable, provided that they have been validated with more rigorous approaches.

A similar 2D model, but including the membrane wetting, has been proposed by Iliuta et al. [12]. They concluded that non-isothermal simulations reveal that the hollow-fiber membrane module operation can be considered as quasi-isothermal. However, the operating and simulation conditions were far from industrial concern (i.e. very high liquid-to-gas flow ratio and MEA conversions lower than 0.2.).

The solvent evaporation in CO_2 absorption using HFMC has been addressed by Ghasem et al. [13]. In their model, free convection of water inside the membrane pores was considered to evaluate the mass flux across the membrane. Since they used aqueous NaOH solution as solvent, the solvent evaporation is mainly due to the use of dry inlet gas. This situation does not correspond to that in CO_2 postcombustion capture by amine solvents.

Finally, Rongwong et al. [11] developed a one-dimensional model including membrane wetting. The model was validated using experimental data from literature. Again, the operating and simulation conditions were far from industrial concern, the corresponding temperature variations were almost linear and lower than 2 K.

In summary, the influence of water transfer and of CO_2 reactive absorption heat, on temperature has been scarcely studied in HFMC. The intensification potential of the technology needs to be estimated within industrial relevant operating conditions. The aim of the present paper is to fill this knowledge gap. A one-dimensional, adiabatic, multi-component model for reactive CO_2 absorption using HFMC is developed. The simulation results are compared with available pilote-scale experimental data as well as with isothermal modelling.

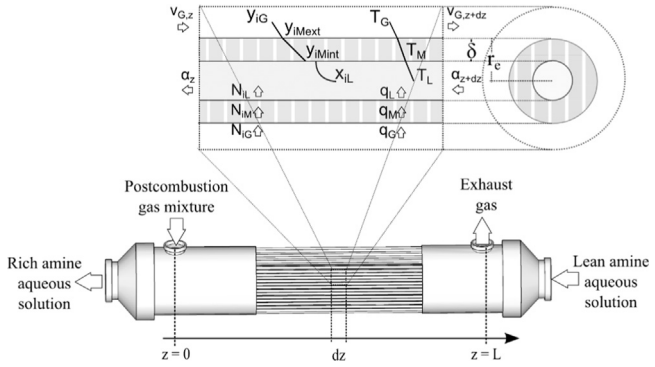

Fig. 1. Schematic representation of the hollow membrane contactor for CO_2 capture in MEA 30 wt aqueous solution.

2. Overall system characteristic

The mass and heat transfer in a membrane module with a hydrophobic microporous membrane is described as a three step process, as illustrated in Fig. 1. Mass transfer is considered for four species (i=CO_2, H_2O, N_2 or MEA). The illustrated reactive species (i=CO_2) is transferred from the gas phase to the external face of the membrane, it then diffuses through the membrane pores, and is finally absorbed by the liquid solution where it reacts.

The membrane contactor consists of a bundle of hydrophobic cylindrical hollow fibers. The contactor has an effective length L, an external radius r_e, varied from 100–500 μm, a fiber volume fraction φ of 0.6 and a relative thickness δ/r_e, set to 0.2. Since content of particles (e.g. ashes) in the postcombustion gases is significant, the amine solvent flows in the lumen to prevent fiber closure. The gas mixture (saturated air and carbon dioxide) flows in the shell surrounding the hollow fibers in a counter-current flow arrangement.

2.1. Operational domain and constraints

The operational parameters and constraints that were applied to all simulations are reported in Table 1. They were chosen in order to represent industrially relevant operating conditions for carbon dioxide capture. A mixture of 0.15 volumetric CO_2, on a dry

Table 1
Operating conditions and geometrical characteristics of the HFMC used in the simulations.

Parameter	Value	Units
Gas		
CO_2 molar fraction	$y_{CO_2}^{in}=0.14$	–
H_2O molar fraction	$y_{H_2O}^{in}=0.07$	–
Inlet temperature	$T_G^{in}=313$	K
Inlet pressure	$P_G^{in}=1.05\times 105$	Pa
Pressure drop	$\Delta P_G=5\times 103$	Pa
CO_2 capture ratio	$\Theta=0.9$	–
Liquid		
MEA total mass fraction	$w=0.3$	$kg_{MEA}\,kg_L^{-1}$
CO_2 loading of lean solvent	$\alpha^{in}=0.242$	$mol_{CO2}\,mol_{MEA}^{-1}$
CO_2 loading of rich solvent	$\alpha^{out}=0.484$	$mol_{CO2}\,mol_{MEA}^{-1}$
Resulting MEA conversion	$\xi=0.932$	–
Outlet pressure	$P_L^{out}=1.05\times 105$	Pa
Inlet temperature	$T_L^{in}=313$	K
Contactor		
External fiber radius	$r_e=1-5\times 10^{-4}$	m
Relative fiber thickness	$\delta/r_e=0.2$	–
Packing fraction	$\varphi=0.6$	–
CO_2 mass transfer coefficient in membrane	$k_{M,CO2}^{ref}=10^{-4}$–10^{-2}	$m\,s^{-1}$