Separation and Purification Technology 188 (2017) 38–51

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Intensification potential of hollow fiber membrane contactors for CO_2 chemical absorption and stripping using monoethanolamine solutions

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ARTICLE INFO

Article history:
Received 13 May 2017
Received in revised form 28 June 2017
Accepted 29 June 2017
Available online 6 July 2017

Keywords:
Carbon dioxide capture
Adiabatic modelling
Hollow fiber membrane contactor
Chemical absorption
Solvent regeneration
Intensification

ABSTRACT

In this work, the intensification potential of Hollow fiber membrane contactors (HFMC) for CO_2 capture by chemical absorption using amine solution have been evaluated by simulation, for both absorption and desorption steps. The simulations have been achieved considering typical industrial relevant conditions for post-combustion capture, based on CASTOR campaign at the Esbjerg pilot plant using packed column, operating at its energetic optimum. Rigorous adiabatic 1D simulations are achieved and revealed important temperature variation as well as significant water transmembrane fluxes in both absorber and desorber. Compared to packed column, a contactor volume reduction (i.e. intensification factor) of about 4 can be achieved in the stripping and absorption section using dry membranes corresponding to a k_m value of 10^{-3} m/s and external fiber radius of 200 μm. For significant absorber intensification factor, fibers should have an external radius less than 400 μm and membrane mass transfer coefficient should not be less than 5.10^{-4} m s^{-1}. HFMC implementation for high temperature stripping is promising providing that membranes resistant to high temperature (i.e. 120 °C) and equally resistant to wetting are available. Due to important water transfer in both absorber and desorber, in addition to wetting of porous membranes by liquid breakthrough, a new possible limiting phenomenon for HFMC technology is wetting by capillary condensation. Even though net solvent losses in the membrane contactor are smaller than those calculated for packed columns, a scrubbing section after the HFMC is still required for solvent recovery in order to meet solvent concentration standard in the CO_2 depleted gas stream. This issue represents an opportunity for the membrane contactor technology based on dense-film MEA selective composite membrane.

1. Introduction

CO_2 capture from large sources attracts considerable attention as a key strategy to mitigate greenhouse gas emissions. Energy production and industrial sectors are responsible of about 60% of the global CO_2 emissions. Among the different possibilities, post-combustion carbon capture and storage CCS) is particularly interesting because it offers retrofit possibilities. Among the different investigated capture processes, gas–liquid absorption using MEA chemical solvent in packed column is classically considered to be the best available technology and commonly taken as a references [32,42]. However, the following major challenges must be addressed to achieve the technical and economic targets:

(i) Decrease of the energy requirement of the solvent regeneration step, through novel solvent or heat integration approaches

(ii) Decrease of the size of the installation through process intensification.

Particularly, the treatment of large quantities of flue gases requires equipment of a large size. Hollow fiber membrane contactors (HFMC) are considered as one of the most promising strategies for the intensification of gas–liquid absorption processes, particularly for post combustion CO_2 capture by absorption in chemical solvents. The key concept of a membrane contactor is to make use of permeable membrane acting as a physical barrier between the gas and the liquid, allowing non-dispersive gas–liquid contact. While microporous hydrophobic membrane materials are mostly used for industrial applications, hydrophilic and non-polymeric materials can also be used for that purpose. Thus, membrane contactor permits to avoid liquid entrainment and flooding, which limit the operational range of packed columns. Moreover, membrane contactors present a special interest in offshore and zero gravity application (i.e. low equipment weight, no gravity-driven flow). The supplementary mass transfer resistance, due to the membrane, is expected to be overbalanced by the high interfacial

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http://dx.doi.org/10.1016/j.seppur.2017.06.074
1383-5866/

Nomenclature

Latin symbols

a	specific interfacial area ($m^2\ m^{-3}$)
a_d	dry specific area ($m^2\ m^{-3}$)
C	molar concentration ($mol\ m^{-3}$)
Cp	specific heat ($J\ mol^{-1}\ K^{-1}$)
d_h	hydraulic diameter (m)
D	diffusion coefficient ($m^2\ s^{-1}$)
E	enhancement factor (dimensionless)
G	molar flux of gas phase ($mol\ m^{-2}\ s^{-1}$)
Gz	Graetz number (dimensionless)
h	heat transfer coefficient ($W\ m^{-2}\ K^{-1}$)
ΔH_{abs}	enthalpy of absorption ($J\ mol^{-1}$)
ΔH_{vap}	enthalpy of vaporization ($J\ mol^{-1}$)
K_{eq}	chemical equilibrium constant (molar scale)
K	vapour liquid (VLE) equilibrium constant (–)
$\boldsymbol{K_{koz}}$	Kozeny constant (m^{-2})
k	mass-transfer coefficient ($m\ s^{-1}$)
k_{ov}	overal mass transfer coefficient ($m\ s^{-1}$)
L	molar flux of liquid phase ($mol\ m^{-2}\ s^{-1}$)
$\boldsymbol{k^{ref}_{M,CO2}}$	reference membrane transfer coefficient of CO_2 at 40 °C and 1 bar ($m\ s^{-1}$)
$\boldsymbol{D^{ref}_{G,CO2}}$	reference diffusion coefficient of CO_2 at 40 °C and 1 bar ($m^2\ s^{-1}$)
N	molar flux ($mol\ m^{-2}\ s^{-1}$)
P	pressure (Pa)
q	heat flux ($W\ m^{-2}$)
r_e	external fiber radius (m)
r_{CO2}	reaction rate relative to CO_2 ($mol\ m^{-3}\ s^{-1}$)
R	gas constant ($J\ mol^{-1}\ K^{-1}$)
T	temperature (K)
U	overall heat transfer coefficient ($W\ m^{-2}\ K^{-1}$)
u	superficial fluid velocity ($m\ s^{-1}$)
v	interstitial fluid velocity ($m\ s^{-1}$)
Z	effective contactor length (m)

Greek symbols

α	CO_2 solvent loading ($mol_{CO2}\ molMEA^{-1}$)
δ	fiber thickness (m)
ϕ	packing fraction (dimensionless)
λ	thermal conductivity ($W\ m^{-1}\ K^{-1}$)
μ	viscosity ($Pa\ s^{-1}$)
ρ	density ($kg\ m^{-3}$)
He	Henry constant (C_G/C_L) (–)

Subscripts

i	compound
G	relative to gas
L	relative to liquid
M	relative to the membrane
F	relative to fluid
int	internal surface of the fibers
ext	external surface of the fibers
Lean	lean liquid absorbent
Rich	rich liquid absorbent

area (a) provided by the membrane, which can be 2–10 times higher than in packed column leading potentially to large intensification factors [48,39,18,40].

A major limitation to membrane contactor development is related to porous membrane wetting issues and chemical stability on long term tests. Wetting is strongly related to solvent properties (viscosity and interfacial tension), membrane pore size, contact angle and transmembrane pressure through Laplace equation [43]. One of the most relevant performance parameter for industry is the average CO_2 specific transferred flux, defined as the quantity of CO_2 transferred per unit-volume of contactor. In order to evaluate effectively the intensification potential of the technology compared to packed column, a value around 1 $mol\ m^{-3}\ s^{-1}$ has been recommended to be taken as the base-line performances (performing packing) [17].

The main target of employing membrane contactors for this application is to provide a significant reduction in equipment size compared to the reference packed columns technology. No significant improvement in the specific energy requirement can be expected as the latter is principally governed by the nature of the solvent (mainly heat of reaction, loading capacity and kinetics), and is independent of the technology used be it packed column or membrane contactor.

In an industrial MEA plant for CO_2 post-combustion capture, the inlet and outlet solvent loadings of the absorption unit play a key role for the mass transfer performances and the recommended values are approximately 0.25 and 0.45 for the inlet and outlet loading, respectively, in order to minimize the energy requirement of the regeneration unit [28,30,47]. Generally, an industrial chemical absorption plant is characterized by nearly complete solvent conversion as well as a partially loaded solvent.

HFMC has been widely investigated both experimentally and theoretically and promising results have been reported in literature in terms of intensification potential at the laboratory scale and in pilot plant investigations (i.e. showing intensification factors ranging from 2 to 8). A literature review is given in Cui and deMontigny [10], Hillal and Ismail [22], Luis Van Gerven et al. [34] and Albarracin Zaidiza et al. [3]. The laboratory scale is characterized by mild operating conditions, corresponding to lab-gases (e.g. no ashes), short-term operation (e.g. no absorbent degradation), low reagent concentrations and conversions, i.e. high liquid-to-gas flowrate ratio and unloaded liquid absorbents. The pilot plant test using membrane contactor was mostly performed at low MEA conversion and relatively low CO_2 capture ratio, some of them using partially loaded liquid absorbents. These conditions do not apply to industrial framework. The only available data under industrial relevant conditions correspond to that obtained in the CASTOR campaign at the Esbjerg pilot plant using packed column.

Regarding HFMC modelling studies, process models of various complexities have been developed considering, almost exclusively, isothermal conditions and neglecting water transfer. Under mild conditions of laboratory scale and pilot plant experiments, the isothermal models have been shown to correctly describe the experimental data in HFMC [2]. However, under high solvent conversion and relatively high CO_2 solvent loading, typical of industrial conditions, the strongly exothermic and endothermic absorption and desorption operations respectively, are expected to lead to important temperature variation as well as significant solvent transmembrane fluxes [38].

The intensification potential of the technology needs to be estimated within industrial relevant operating conditions. Besides, temperature variation will influence physicochemical and thermodynamic properties, membrane-solvent interaction, such as contact angle or the pore shape as well as membrane structure stability. The transport of volatile component, such as water, through the membrane may lead to their condensation in the membrane due to local over-saturations and capillary effect [2].