



# Gas-liquid absorption in industrial cross-flow membrane contactors: Experimental and numerical investigation of the influence of transmembrane pressure on partial wetting



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## HIGHLIGHTS

- Membrane wetting is found to be independent of transmembrane pressure.
- An original model is developed to simulate gas absorption in cross-flow HFMMs.
- Liquid shellside hydrodynamics is modeled as flowing through a porous medium.
- Chilton-Colburn equation is optimized to estimate Sherwood number at local scale.

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## ABSTRACT

Membrane wetting is a limitation to mass transfer in industrial Hollow-Fiber Membrane Modules (HFMM). The influence of transmembrane pressure on wetting is investigated through both experimental and numerical works. On the one hand, experiments for CO<sub>2</sub> absorption in softened water are carried out at pilot scale with Liqui-Cel<sup>®</sup> Extra-Flow modules operating in a cross-flow mode. After several hours of operation (50–300 min), the membrane partially wetted mode is established at steady state and decreases the absorbed CO<sub>2</sub> flowrates up to 10.5% compared to membrane dry mode. Interestingly, CO<sub>2</sub> absorption is found to be independent on different transmembrane pressure (0.5 and 2.5 bar), both in the dry and partially wetted mode. On the other hand, a 2D predictive tool is developed to help in the design of HFMM gas absorption systems. The model depicts the complex hydrodynamics in commercial cross-flow modules. The liquid in the shellside is flowing through the fiber bundle considered as a porous medium. The liquid mass-transfer coefficient is non-uniform over the membrane module and is estimated at a local scale. The parameters from the empirical Chilton-Colburn correlation are optimized with experimental results in the dry mode through a Particle Swarm Optimization (PSO) algorithm. The simulation results are in good agreement with the experiments (−13/+17%). In the partially wetted mode at steady state, wetting is then described by considering a log-normal pore size distribution and the Laplace-Young equation. The transmembrane pressure decreases the simulated absorbed CO<sub>2</sub> flowrate. By comparing to experimental data, the numerical relative error is increasing with transmembrane pressure. Therefore, a suggestion is made to adapt the description of wetting in HFMM gas-liquid models.

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## 1. Introduction

Nowadays, membrane processes are found in more and more industrial applications. In the field of separation techniques, they indeed offer several advantages: energy savings, ease of operation, flexibility in design, production of high-purity compounds and

lower environmental impacts (Pabby and Sastre, 2013). In particular, HFMMs find applications in liquid-liquid processes such as extraction of aroma compounds (Baudot et al., 2001; Bocquet et al., 2006). They were also investigated for gas-liquid applications: blood oxygenation, liquid degasification, membrane distillation, acid gas removal, gas humidification (Sirkar, 2008). In addition, they were proven to offer significant size savings compared to conventional packed columns in post-combustion gas capture (Hoff and Svendsen, 2013; Zhao et al., 2016). Recent works

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## Nomenclature

### Latin symbols

$a$	specific area ( $\text{m}^2/\text{m}^3$ )
$C_A^l$	dissolved concentration of specie A in the liquid phase ( $\text{mol}/\text{m}^3$ )
$C_A^g$	concentration of specie A in the gas phase ( $\text{mol}/\text{m}^3$ )
$c$	exponent parameter in Chilton-Colburn equation
$D$	diffusion coefficient ( $\text{m}^2/\text{s}$ )
$d_H$	hydraulic diameter (m)
$d_{fi}$	inner diameter of a hollow fiber (m)
$d_{fo}$	external diameter of a hollow fiber (m)
$d_{ln}$	logarithmic mean diameter of a hollow fiber (m)
$Gz$	Graetz number
$g(r)$	pore size distribution ( $\text{m}^{-1}$ )
$H^{cc}$	dimensionless Henry's coefficient
$k_H^{px}$	Henry's coefficient (Pa)
$k_g$	gas phase mass-transfer coefficient (m/s)
$k_l$	liquid phase mass-transfer coefficient (m/s)
$k_m$	membrane phase mass-transfer coefficient (m/s)
$k_{tot}$	overall mass transfer coefficient (m/s)
$l_{eff}$	effective contact fiber length in membrane contactor (m)
$M_A$	molar mass of component A (g/mol)
$N_f$	total number of fibers in membrane contactor
$\vec{n}$	normal vector
$P_g$	pressure in the gas phase (bar g)
$P_l$	pressure in the liquid phase (bar g)
$\Delta P$	transmembrane pressure (bar)
$Q_g$	total gas flowrate ( $\text{N m}^3/\text{s}$ )
$Q_l$	total absorbent flowrate ( $\text{m}^3/\text{s}$ )
$Q_{\text{CO}_2}$	$\text{CO}_2$ flowrate ( $\text{N m}^3/\text{s}$ )
$Pe$	Peclet number
$Re$	Reynolds number
$R_{\text{CO}_2}$	source term in $\text{CO}_2$ mass balance equation ( $\text{mol m}^{-3} \text{s}^{-1}$ )
$R_g$	gas mass transfer resistance (s/m)
$R_m$	membrane mass transfer resistance (s/m)
$R_l$	liquid mass transfer resistance (s/m)
$R_{tot}$	total mass transfer resistance (s/m)
$R_i$	inner diameter of the hollow fiber bundle (m)
$R_o$	outer diameter of the hollow fiber bundle (m)
$r$	pore radius (m)
$r_c$	Laplace-Young critical pore radius (m)
$Sh$	Sherwood number
$Sc$	Schmidt number
$\Delta t$	characteristic time (s)
$T$	temperature (K)

$u$	velocity (m/s)
$x$	molar fraction in the liquid phase
$y$	molar fraction in the gas phase

### Greek symbols

$\Phi$	packing fraction in the bundle of fibers (%)
$\varepsilon$	porosity (%)
$\delta$	membrane thickness (m)
$\tau$	tortuosity
$\psi_{\text{CO}_2}$	$\text{CO}_2$ loading rate (%)
$\eta$	wetting ratio (%)
$\gamma$	surface tension (N/m)
$\theta$	contact angle ( $^\circ$ )
$\sigma$	pore distribution geometric standard deviation
$\nu$	cinematic viscosity ( $\text{m}^2/\text{s}$ )
$\rho$	density ( $\text{kg}/\text{m}^3$ )
$\kappa$	permeability of porous medium ( $\text{m}^2$ )
$\mu$	dynamic viscosity (Pa s)
$\Omega$	particle hard-shell diameter
$\lambda$	molecule mean free path
$\omega$	factor parameter in Chilton-Colburn equation

### Subscripts/superscripts

<i>in</i>	inlet of the membrane contactor
<i>out</i>	outlet of the membrane contactor
<i>abs</i>	absorbed in the membrane contactor
<i>eq</i>	at equilibrium
<i>eff</i>	effective parameter
<i>wet</i>	membrane wetted mode
<i>dry</i>	totally dry membrane mode
<i>p</i>	relative to the membrane pores
<i>i</i>	relative to the phase <i>i</i>
<i>g</i>	relative to the gas phase
<i>m</i>	relative to the membrane phase
<i>l</i>	relative to the liquid phase
<i>mol</i>	molecular diffusion
<i>Kn</i>	Knudsen diffusion
$\text{CO}_2$	carbon dioxide
<i>bed</i>	relative to the fiber bundle assimilated to a porous medium
<i>exp</i>	relative to the experiment
<i>mod</i>	relative to the model
<i>res</i>	residence time
<i>shell</i>	relative to shellside

(McLeod et al., 2013) investigated the purification of biogas into biomethane by HFMMs.

The membrane is an interface for mass transfer. It can be passive (without selectivity) or contain a thin selective layer depending on the application. However, the presence of the membrane introduces an additional resistance to mass transfer. Regarding gas-liquid applications, this membrane resistance is often negligible compared to the diffusion layer resistance in the liquid phase while operating in the non-wetted mode. However, (Kreulen et al., 1993) stated that mass transfer resistance increases in the wetted mode when the membrane pores are filled with liquid. This phenomenon was studied by Mavroudi et al. (2006) for  $\text{CO}_2$  absorption in water: the permanent regime in the wetted mode was established after three hours of operation and the total mass transfer resistance increased by 21–53% compared to the dry mode. These observations require further investigation in a process optimization perspective.

The wetting phenomenon is described as dependent on many parameters: pore size, surface tension, contact angle and operating transmembrane pressure. The choice of the solvent and the membrane material is of the utmost importance to avoid membrane wetting (Mosadegh-Sedghi et al., 2014). The influence of most of these parameters have been investigated. Lv et al. (2010) noticed a variation up to 20–30° in the contact angle when the hollow fiber was immersed in different solvents. Scanning Electron Microscope (SEM) observations confirmed the apparition of structure modifications in the membrane after wetting. Bougie and Iliuta (2013) performed breakthrough pressure measurements and observed a decrease in the maximal pore size after wetting.

Modelling works also explored the influence of operating pressures (Boributh et al., 2011; Goyal et al., 2015) and membrane characteristics (Goyal et al., 2015; Yan et al., 2014). (Faiz and Al-Marzouqi, 2009; Keshavarz et al., 2008) developed models for simultaneous  $\text{CO}_2$  and  $\text{H}_2\text{S}$  absorption in MEA and DEA solutions

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