



Membrane contactors: An alternative for de-aeration of salt solutions?

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ARTICLE INFO

Keywords:

Membrane contactors

De-aeration

Salt solution

Dissolved oxygen

Corrosion

ABSTRACT

Corrosion is a major issue in industry, and the oxygen concentration in the fluids processed influences corrosion rates. Membrane contactors, such as Liqui-Cel® modules, are suitably used for de-aerating ultrapure water to low oxygen levels. Liquid mass transfer correlations for water de-aeration have been determined by varying liquid flow rates at ambient conditions. Limited information is available on membrane de-aeration of concentrated salt solutions. In our work the feasibility of membrane contactors for de-aeration of salt solutions containing 0.1–25 wt% NaCl has been investigated and confirmed. Oxygen outlet concentrations as low as 70 ppb have been obtained at a temperature of 40 °C and a (vacuum) pressure of 70 mbar using the Liqui-Cel® 2.5 × 8 Extra-flow module. Mass transfer coefficients have been determined for a range of liquid velocities, viscosities, densities, and oxygen diffusion coefficients by changing the salt concentration and the temperature. This means that not only the Reynolds number has been varied (as commonly done), but the Schmidt number as well. Their effect on the Sherwood number has thus been established. The obtained correlation allows sufficiently accurate prediction of oxygen removal from NaCl solutions over the entire salt concentration range. A membrane area of 455–4105 m² (using Liqui-Cel® 14 × 40 modules) is required for de-aeration of a saturated sodium chloride solution to an oxygen concentration below 50 ppb using a total liquid flow of 100–900 m³/h, which is typical for salt production.

1. Introduction

Corrosion of metal piping and equipment related to the presence of oxygen in the system is a major problem in industry. It eventually leads to the need for replacing the equipment, which translates into additional production costs. Corrosion avoidance is especially challenging in environments where salt solutions, such as in seawater desalination, chlor/alkali production and crystalline salt production, are processed. Conventional technologies for the removal of oxygen as applied in industry include (bulky) vacuum tower de-gasifiers and forced draft de-gasifiers. However, it is very challenging to reach low levels of dissolved oxygen with these conventional methods. Dissolved oxygen levels lower than 50 ppb can be achieved using chemical methods (e.g. the addition of ions such as SO₃²⁻ that react with oxygen to form SO₄²⁻, or oxygen scavengers such as hydrazine and diethylhydroxylamine), but the need for adding toxic substances such as hydrazine or the increase in solid contents resulting from the addition of e.g. sodium sulfite make the use of chemical methods less attractive [1].

Membranes contactors may offer an alternative for de-aeration. These contactors, operated in either vacuum mode, sweep gas mode or combined (vacuum and sweep gas) mode, resemble vacuum membrane

distillation (VMD) and stripping gas membrane distillation (SGMD). VMD and SGMD are two of the most common membrane distillation (MD) configurations [2–5]. Scale-up of the membrane contactors requires suitable mass transfer models. The mechanism of mass transfer in membrane contactors has been studied for absorption and/or stripping of different gasses from aqueous streams [1,6–19]. Especially Sengupta et al. [16] and Zheng et al. [20] have generated a mass transfer model for membrane de-aeration based on oxygen removal from ultrapure water at ambient conditions, using various Liqui-Cel® modules with different membrane surface areas per module. This model, based on a Sherwood, Reynolds and Schmidt number relation, includes parameters such as liquid density, viscosity and the oxygen diffusion coefficient, although it has been determined through variation of the liquid velocity at constant liquid density, viscosity and oxygen diffusion coefficients only. The model has not been validated for de-aeration of other fluids such as salt solutions.

To the best of our knowledge, hardly any information is available for membrane de-aeration dealing with solutions containing salts, especially at high salt content. Only Criscuoli et al. [21] have presented limited information on lab-scale membrane de-aeration of salt solutions. Their study focused on the performance of the membrane

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Nomenclature

A_T	transfer area between the two phases in the contactor [m^2]
C	dissolved oxygen concentration [kg/m^3]
C_{out}	dissolved oxygen outlet concentration [kg/m^3]
C_{in}	dissolved oxygen inlet concentration [kg/m^3]
D	diffusion coefficient (determined from Wilke-Chang correlation) [m^2/s]
d_F	fibre outside diameter [m]
d_h	hydraulic diameter (Eq. (11)) [m]
d_m	module diameter ($d_m = 2 * R_{Co}$) [m]
d_t	liquid inlet tube diameter ($d_t = 2 * R_{Ci}$) [m]
E	removal efficiency [-]
f_p	packing fraction of the hollow fibres [-]
f_x	fractional open area for flow of liquid [-]
K_L	overall mass transfer coefficient [m/s]
k_L	liquid mass transfer coefficient [m/s]
L_C	effective length of the cartridge [m]
n_f	number of fibers [-]
$n_{modules}$	total number of modules required [-]

P_{vac}	(vacuum) pressure [bar]
Q_L	volumetric flow rate of the shell side liquid [m^3/s]
$Q_{L,target}$	volumetric flow rate of the shell side liquid per module giving the required oxygen outlet concentration of 50 ppb [m^3/s]
$Q_{L,total}$	total volumetric flow rate of the shell side liquid that needs to be de-aerated [m^3/s]
R_{Ci}	cartridge inside radius [m]
R_{Co}	cartridge outside radius [m]
Re	Reynolds number [-]
Sc	Schmidt number [-]
Sh	Sherwood number [-]
T	temperature [$^{\circ}\text{C}$]
v_e	linear liquid velocity (Eq. (12)) [m/s]
α	proportionality parameter in Sherwood correlation (Eq. (1)) [-]
β	Sherwood dependence on the Reynolds number [-]
γ	Sherwood dependence on the Schmidt number [-]
μ	liquid dynamic viscosity (determined from [34]) [$\text{kg}/\text{m}\cdot\text{s}$]
ρ	liquid density (determined from [34]) [kg/m^3]

contactor for oxygen concentration and pH control of the water (permeate) produced in a reverse osmosis desalination plant. However, they also performed some membrane de-aeration experiments with model solutions mimicking nanofiltration permeate (fed to the reverse osmosis unit) and reverse osmosis concentrate using sweep gas and PVDF, PTFE and acrylic based flat sheet membranes (samples of 40 cm^2 membrane area). Both solutions contained sodium chloride concentrations below 65 g/L . Despite the first interesting results presented by Criscuoli et al. [21], a systematic evaluation of the effect of salt concentration on oxygen mass transfer in membrane degassers is not available, whereas this is essential for scaling-up of membrane degassers and application of the technology in commercial practice.

Especially the removal of oxygen in membrane de-aeration of saturated salt solutions seems challenging, since (a minor amount of) water is evaporated simultaneously and crystallization of salt may be detrimental to the operability of the membrane degasser. The concentration of salt solutions using membrane distillation has been investigated for bulk water removal from seawater and brackish water up to saturation concentration [2,22–28]. Sanmartino et al. [26] reported the occurrence of crystallization when the solubility limit of NaCl was exceeded. This caused membrane wetting or blocking of the membrane pores due to salt crystals deposition and growth on the membrane surface and in the pores. However, simple washing with water appeared to be sufficient for partial recovery of the membrane properties [26]. The studies suggest that membrane contactors might be suitable for de-aeration of concentrated NaCl solutions as well, even though challenges associated to the formation of crystals, fiber break or membrane wetting might be expected.

In this study we evaluated the use of a Liqui-Cel® membrane contactor for oxygen removal from salt solutions. The main objective was to determine oxygen removal rates for de-aeration of salt solutions up to saturation (25.3 wt% NaCl), to generate mass transfer correlations through variation of the Reynolds and Schmidt numbers, and to use the obtained correlation to gain insights in the required membrane area for commercial scale saturated NaCl solution de-aeration units.

2. Theoretical background

2.1. Determining Sherwood correlations for mass transfer description

In open literature several mass transfer models have been proposed for membrane contactors. For mass transfer in gas–liquid hollow-fiber membrane contactors, Sengupta et al. [16], Zheng et al. [20], Fallanza

et al. [29], and Shen et al. [30] have reported Sherwood correlations for shell-side liquid flow. For tube-side liquid flow Sherwood correlations can be found in Wang et al. [31].

The reported Sherwood (Sh) correlations are generally described as a function of the Reynolds (Re) and Schmidt (Sc) numbers according to:

$$Sh = \alpha \cdot Re^{\beta} \cdot Sc^{\gamma} \quad (1)$$

where:

$$Sh = \frac{k_L \cdot d_h}{D} \quad (2)$$

$$Re = \frac{\rho_L \cdot v_e \cdot d_h}{\mu_L} \quad (3)$$

$$Sc = \frac{\mu_L}{\rho_L \cdot D} \quad (4)$$

In almost all correlations reported the exponent γ , describing the Sherwood number dependence on the Schmidt number, is assumed to be equal to $\gamma = 0.33$. Li et al. [32] have investigated the hydrodynamics for in-line fiber arrays numerically, and have concluded that the dependence of the Sherwood number on the Schmidt can be different from the commonly used exponent $\gamma = 0.33$. They concluded that the obtained exponent β for Reynolds is only slightly higher than the exponent γ for Schmidt, and that the Sherwood number is approximately proportional to the Peclet number (the product of the Reynolds and the Schmidt number) to the power n with n between 0.33 and 0.40 [32]. In an earlier review on mass transfer, Gekas and Hallström [33] concluded that the Sherwood dependence on Schmidt was theoretically and experimentally described by an exponent $0.33 \leq \gamma \leq 0.5$, while the Schmidt exponent was smaller than the Reynolds exponent ($\gamma < \beta$).

For de-aeration of water using Liqui-Cel® membrane contactors (e.g. the Liqui-Cel® 2.5 × 8 Extra-flow, which is evaluated in the current study as well), Sengupta et al. [16] and Zheng et al. [20] have described modeling approaches to obtain the liquid mass transfer coefficient and presented a Sherwood relation for liquid flow at the shell side of the hollow fibers.

For this membrane contactor the mass balance on the liquid side is described according to:

$$-Q_L \cdot dC = K_L \cdot C \cdot dA_T \quad (5)$$

Integrating Eq. (5) leads to:

$$K_L = \frac{Q_L}{A_T} \ln \left(\frac{1}{1-E} \right) \quad (6)$$

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