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# Analytical solutions for membrane wetting calculations based on log-normal and normal distribution functions for $CO_2$ absorption by a hollow fiber membrane contactor

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#### ABSTRACT

The analytical solutions for calculating the average wetting ratio,  $\langle x^* \rangle$ , for gas-liquid membrane contacting process based on log-normal and normal distribution functions have been developed. By using the membrane characteristics, liquid absorbent properties and operating conditions, the average wetting ratio is determined analytically. The overall mass transfer coefficient ( $K_G$ ) corresponding to the average wetting ratio is also calculated. Due to the pressure drop of liquid flow in the fiber, the operation mode would be changed along the fiber and six possible situations of operation cases are observed. In addition, the proposed calculation can predict the fiber lengths that are completely wetted, partially wetted and are completely dry. In order to verify the proposed calculation, the experiments of  $CO_2$  absorption from the gas mixture ( $CO_2$ - $N_2$ ) by polyvinylidenefluoride (PVDF) hollow fiber membrane contactor using water and monoethanolamine (MEA) as the absorbents were performed at various inlet liquid pressures  $(P_{IO})$  and liquid velocities. The results obtained from the experiments were reported in term of overall mass transfer coefficient and were validated with those obtained from the proposed calculations. It reveals that calculation based on log-normal distribution shows a better fit with the experiments compared to that based on normal distribution at all operating conditions indicating that the membrane pore structure can be described better using log-normal distribution function. In addition, the calculated average wetting ratio based on log-normal distribution function greed well with that obtained from the numerical model proposed in the literature.

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#### 1. Introduction

Gas-liquid membrane contactor is a mass transfer device for gas absorption in which the microporous hydrophobic hollow fiber membranes are generally employed as the phase barrier. The membrane provides the contact between gas and liquid phases making these two phases flow independently. Gas-liquid membrane contacting process offers the advantages over the conventional contactors, i.e., packed, plate and sprayed columns, that it can overcome flooding, foaming and channeling problems and provides very high contact area per unit volume [1,2]. In addition, scaling up is more straight forward. However, the membrane adds the additional resistance to the overall mass transfer, especially in case of the partially wetted mode where the membrane pores are partially penetrated by the liquid absorbent.

Membrane wetting is the main problem of gas-liquid membrane contacting processes. The negative effects of membrane wetting on absorption performance have been widely addressed in the literature [3–7]. It has been known that membrane wetting depends on many factors, i.e., membrane characteristics, absorbent properties and operating conditions. The effect of these parameters on membrane wetting is usually described by Laplace–Young Eqs. (2), (4), (6), (8) and (9) which provides the trans-membrane pressure difference that the liquid can penetrate the membrane pores. The average pore radius of the membrane is usually used to calculate the penetration pressure [8,9]. Due to the pore size distribution, the pores with size larger than the average pore radius are possible to be wetted. Therefore, the membrane pore size distribution should be considered in order to predict the pore wetting more accurately.

In general, the pore size distribution is described by different distribution functions such as log-normal and normal distribution functions, etc. Each distribution function presents the different shape of the distribution curve. The effects of pore size distribution on membrane transport were reported in literature [10–13]. Li et al. [14] reported that the membrane with higher geometric standard deviation exhibited higher membrane mass transfer coefficients. Derjani-Bayeh and Rodgers [15] investigated the effect of different standard probabilistic models including gamma, log-normal, normal, Weibel and Rayleigh distribution functions,

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on the area average flux and the area average membrane sieving coefficient. They concluded that an uncertainty in the choice of distribution in describing the membrane morphology could lead to a propagated uncertainty in predicting the overall membrane performance. Kong and Li [16] compared the use of log-normal and normal distribution functions to predict the membrane's coefficients. They found that the analysis based on log-normal distribution presented a better off in producing a realistic prediction of membrane's coefficients. Lu et al. [5] developed the mathematical model to calculate the membrane wetting accounting for the effect of pore size distribution described by log-normal distribution. Boributh et al. [2] presented the model considering the effect of pressure drop of liquid flow in lumen side, pore size distribution as well as operating conditions on the membrane wetting. The results indicated that the pore size distribution characteristics significantly affected the degree of membrane wetting and the wetting behavior.

The mathematical models investigating the degree of membrane wetting and the effect of membrane wetting on the absorption efficiency have been widely proposed [17-20]. Wang et al. [21] developed the two dimension model to predict CO<sub>2</sub> absorption and validated the model with the experimental results. Mavroudi et al. [22] proposed the first-order expression to describe the membrane resistance change with time for physical absorption of pure CO<sub>2</sub>. Khaisri et al. [23] developed the mathematical model for investigating mass and heat transport based on the resistance-in-series model to describe the influence of wetting on the process performance. Faiz and Al-Marzougi [17] presented the model considering pseudo wetting on the absorption performance for CO<sub>2</sub> absorption at high pressure. Although the modeling study on the absorption of CO<sub>2</sub> by gas-liquid membrane contacting process have been investigated widely for the past decade, however, several models presented the approximation of the membrane wetting as the fitting parameter. The calculations of membrane wetting did not include all important parameters affecting membrane wetting. To the best of our knowledge, there is no the published work on the analytical calculation of the membrane wetting. Basically, the analytical solution is more convenient compared to the iteration methods because the simulation tool is not needed.

The aim of this work is to present the analytical solutions for predicting average wetting ratio,  $\langle x^* \rangle$  by taking the membrane characteristics, liquid absorbent properties as well as operating conditions into consideration. The mass transfer equations together with the Laplace-Young equation considering the membrane pore size distribution based on log-normal and normal distribution functions were solved analytically. The proposed calculation is also able to predict the change of operation mode along the fiber length and determine the lengths of individual operation mode. In order to verify the proposed calculation, the experiments of CO<sub>2</sub> absorption by PVDF hollow fiber membrane contactor using water and MEA solution as the absorbents were performed at different liquid inlet pressures  $(P_{L0})$ , liquid velocities and MEA concentrations and were validated with the proposed calculation. Additionally, the calculated average wetting ratio based on log-normal and normal distribution functions were compared to that obtained from the numerical model proposed in our previous work [9].

#### 2. Theory

#### 2.1. Membrane pore size distribution

For the porous membranes with non-uniform pore sizes, the log-normal and normal distribution functions are usually

employed to describe the distribution of membrane pore sizes. Two important parameters including the average pore radius  $(r_m)$  and the geometric standard deviation  $(\sigma)$  of membrane pore size are used in the distribution function, f(r) [14]. The log-normal and normal distribution functions can be expressed by the following equations:

$$f_{L}(r) = \frac{1}{\sqrt{2\pi}r} \left( \ln(1+\sigma^{2}) \right)^{-0.5} \exp\left( -\frac{\left( \ln(r/r_{m}) \left( 1+\sigma^{2} \right)^{0.5} \right)^{2}}{2 \ln(1+\sigma^{2})} \right)$$
(1)  
$$f_{N}(r) = \frac{1}{\sqrt{2\pi}r_{m}\sigma} \exp\left( -\frac{\left( 1-r/r_{m} \right)^{2}}{2\sigma^{2}} \right)$$
(2)

where *r* is membrane pore radius.

#### 2.2. Mass transfer coefficients

The operation of gas–liquid membrane contactors can be classified into 3 modes including non-wetted (dry) mode, partially wetted mode, and wetted mode. The non-wetted mode is preferred because it provides the highest performance. However, the operations of membrane contactors are usually performed in a partially wetted mode as shown in Fig. 1 and the resistance-in-series model for partially wetted hollow fiber module can be written as

$$\frac{1}{K_G d_{Int}} = \frac{H}{Ek_L d_i} + \frac{H}{Ek_{ML} d_{InML}} + \frac{1}{k_{MG} d_{InMG}} + \frac{1}{k_G d_0}$$
(3)

where  $K_G$  is the overall mass transfer coefficient and  $k_L$ ,  $k_G$ ,  $k_{MG}$ , and  $k_{ML}$  are the mass transfer coefficients of liquid, gas, membrane for gas-filled pores, and membrane for liquid-filled pores, respectively. *H* represents Henry's constant, and  $d_o$ ,  $d_i$ ,  $d_{Int.}$ ,  $d_{InMG}$  and  $d_{InML}$  are the outer fiber diameter, inner fiber diameter, gas–liquid interfacial diameter and logarithmic mean diameters of the non-wetted and the wetted membranes, respectively. *E* is enhancement factor accounting for the effect of chemical reaction on the absorption [9,20,23]. By assuming that the reaction between CO<sub>2</sub> and MEA is a second-order irreversible reaction, the enhancement factor can be determined by the following equation:

$$E = \frac{-(Ha^*)^2}{2(E_{\infty}^* - 1)} + \sqrt{\left(\frac{(Ha^*)^2}{4(E_{\infty}^* - 1)^2} + \frac{E_{\infty}^*(Ha^*)^2}{(E_{\infty}^* - 1)} + 1\right)}$$
(4)



**Fig. 1.** Mass transfer regions and resistance-in-series in partially wetted mode for gas-liquid membrane contacting process.

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