

# A study of mass transfer resistance in membrane gas–liquid contacting processes

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## Abstract

The change of mass transfer resistance with time has been examined for membrane-based carbon dioxide absorption in water. A commercial polypropylene hollow fiber membrane module was used with gas flow through the lumen side and liquid cross-flow on the shell side. Experiments were carried out for a prolonged period of time to evaluate absorption performance. Absorption flux decreased significantly with time due to a developing resistance to mass transfer. However, the initial flux value was restored after membrane drying, indicating that the additional resistance was reversible. A theoretical model was developed to analyze and predict flux deterioration with time in terms of partial resistances in series. The resistance against pure gas mass transfer was considered to be the sum of the liquid phase resistance and the resistance of the membrane; the latter arises from membrane pore filling by gas or liquid or both. A shell side mass transfer correlation was used for the prediction of the liquid phase resistance. The experimental absorption flux decline with time was attributed to gradual partial pore filling by liquid, and it was used to estimate the magnitude of the membrane resistance and its temporal variation. Model predictions, in good agreement with experimental results, permitted the estimation of liquid penetration into the membrane matrix. Although this was relatively low (~13%), the resulting resistance of the liquid-filled pores accounted for over 98% of the membrane resistance and for 20–50% of the total resistance to absorption.

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## 1. Introduction

In recent years, membrane-based absorption processes are investigated for the recovery/removal of acid gases from flue gases, natural gas and various industrial process gas streams. The membrane used in the contacting process is often considered passive, simply providing a support for the interface of two immiscible fluids and not introducing any selectivity. However, the membrane is a major component of the system; it constitutes one of the resistances to mass transfer, and any pore wetting affects considerably the efficiency of the process, compared with a conventional gas absorption system [1,2].

The pores of the membrane can be filled with either gas or liquid. These two types of operation are referred to as non-wetted mode and wetted mode, respectively. Several studies have addressed possible partial wetting of the membrane, which affects the performance of the process [3–10]. Pore wetting depends on the structural characteristics of the porous material, the operating pressure and the nature of the liquid phase in contact with the membrane [11–13]. When partial wetting occurs, the length of liquid penetration in the pores may vary with time, while the remaining pore volume is filled with gas. The gas–liquid interface is shifted with time from the pore mouth to some (variable) location inside the membrane pores.

For a given membrane material and structure, the life history of the membrane can alter its hydrophobic character, due to morphological changes by the initial intrusion of liq-

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uid into the membrane [10,12,14,15]. In addition, although the pressure of the gas and the liquid across the membrane is important to prevent wetting [11], pressure drop over the fiber length, in the case of hollow fiber membranes, can result in their wetting. When the liquid flows on the lumen side, the significant pressure drop inside the lumen leads to membrane wetting in the initial fiber length due to a higher trans-membrane pressure in this section than the breakthrough pressure [5,10]. Finally, the nature of liquid in contact with the membrane may cause wetting. The presence of ionic species, complexes, microorganisms or impurities even in trace amounts, can change the wetting characteristics of the membrane–liquid system [8,12,16]. This situation becomes more pronounced when chemical reactions take place between the gas and the liquid phase.

Since the diffusion of a gaseous species in the liquid phase is much slower than in the gas phase, the membrane resistance can increase significantly if it operates in the wetted or partially wetted mode. Membrane wetting or partial wetting has been considered to explain experimental results and gradual degradation of performance with time [3–7,9,10]. Capillary condensation of water vapor in the pores [3,6], modification of system wetting characteristics [4] or pressure difference between the shell side and the lumen side [5,10] have been considered as possible reasons for liquid intrusion into the membrane pores and consequently, for an additional mass transfer resistance and absorption flux decline. In some cases, the pore length blocked by the liquid phase was used as an adjustable parameter to fit the predicted flux values to experimental ones [4,7,9]. It is obvious that a more detailed theoretical and experimental study is required for understanding the partially wetted operating mode in a gas–liquid membrane contactor.

In the present work, the change of mass transfer resistance with time has been examined for membrane-based carbon dioxide absorption in water using a commercial polypropylene hollow fiber membrane module. Experiments were carried out for a period of time up to 750 min to study the stability of the system, and hence, the wettability of the membrane by the water. During an experimental run, the absorption flux was changing with time. Initially, there was a short-time period during which the absorption flux remained constant, and thereafter, the flux decreased rapidly from its initial value to a final one, where it seemed to stabilize. Thus, the mass transfer resistance was changing with time and the resistance profile versus time was distinguished into two regions. In the initial period, the total mass transfer resistance and the gas flux remained constant with time. At longer experimental times, apart from the initial total resistance, an additional resistance to mass transport was developed, resulting in flux decline. Once the experimental run was over, the initial flux value was restored after membrane drying, indicating that this additional resistance is reversible. A theoretical model was developed to analyze and predict flux deterioration with time in terms of partial resistances in series. The resistance against pure gas mass transfer was considered to be the liq-

uid phase resistance and the resistance of the membrane; the latter arises from membrane pore filling by gas or liquid or both. A shell side mass transfer correlation was used for the prediction of the liquid phase resistance. The experimental flux decline with time was attributed to gradual pore filling by liquid, and it was used to estimate the magnitude of the membrane resistance and its temporal variation.

## 2. Theory

Flux decline is a major problem in several membrane processes, such as reverse osmosis, ultrafiltration, and classical filtration of liquids. In these processes, flux decreases when the resistance to transport increases above that of the virgin membrane. This is caused by accumulation of retained material near, on or within the membrane, which blocks or constricts the pores and forms a layer of additional resistance to flow across the membrane. Resistance-in-series models have been developed to analyze flux decline due to fouling of the membrane (reversible and/or irreversible blocking) [17]. The similarity of flux profile over time in membrane gas absorption process with that of reversible pore blocking in filtration led us to consider the adaptation of a resistance-in-series model to analyze and predict the flux behavior in a membrane gas–liquid contacting process.

For the non-wetted mode of a gas–liquid membrane contactor, mass transfer is determined by the consecutive steps in the three phases shown schematically in Fig. 1, i.e. diffusion of gaseous component  $i$  from the bulk gas to the membrane wall, diffusion through the totally gas-filled pores of the membrane to the membrane–liquid interface, and dissolution into the liquid absorbent, followed by liquid phase diffusion and/or chemical reaction. Hence, a resistance-in-series model can express the total resistance,  $R_{\text{total}}$ , defined as the reciprocal of the overall mass transfer coefficient,  $K_{\text{ol}}$ :

$$R_{\text{total}} = \frac{1}{K_{\text{ol}}} = \frac{H}{k_g} + \frac{H}{k_m} + \frac{1}{k_l} = R_g + R_m + R_l \quad (1)$$

where  $R_g$  is the gas phase resistance,  $R_m$  the membrane resistance and  $R_l$  the liquid phase resistance. For a non-wetted membrane,  $R_m$  expresses the resistance to mass transfer in the membrane pores, which are filled by gas.

For gas absorption, the resistance to gas diffusion from the bulk gas to the membrane external surface,  $R_g$ , can be ignored compared to other resistances [1,4,10]. In the case of physical absorption (such as carbon dioxide in water), the resistance connected with the liquid phase should depend on experimental hydrodynamics and it should remain unchanged for a constant liquid flow rate. In order to characterize the mass transfer coefficient in the shell side,  $k_l$ , and consequently, the liquid phase resistance,  $R_l$ , the following correlation is used:

$$Sh_s = aRe_s^b Sc_s^c \quad (2)$$

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