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Membrane contactor with hydrophobic metallic membranes: 1. Modeling of coupled mass and heat transfers in membrane evaporation

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ABSTRACT

The concentration of thermo-sensitive solutions can be successfully carried out by low temperature evaporation in membrane contactors. This process of membrane evaporation which is approaching to a sweep gas membrane distillation is based on the concentration of an aqueous solution by its continuous evaporation through a metallic hydrophobic membrane. The driving force of the process is the difference of water vapor pressure between the aqueous feed solution and a dry gas. This work is the first part of a study about membrane evaporation process carried out with flat hydrophobic metallic membranes. In this first part we present a detailed research on membrane characterization, describe the basic principles of the process and develop a complete mass and heat transport model which is validated with some experimental results. Simulations have been carried out in order to study theoretically the influence of different operating parameters and membrane structure on process effectiveness.

1. Introduction

Membrane contactors have been developed as an attractive alternative to satisfy the requirement of process intensification strategy and to overcome the limitations of traditional membrane operations (reverse osmosis, electrodialysis, micro- ultra- and nanofiltration). The key concept of this membrane operation is the use of a porous hydrophobic membrane in order to create an interface for mass transfer between two phases. Indeed, the membrane acts as a physical barrier without significant effect in terms of selectivity [1]. Besides, in contactor processes the membrane is unable to separate entities in terms of size as in classical pressure-driven membrane processes, but structural parameters of membranes as porosity and tortuosity can keep on playing an important role in global mass transport. Various advantages can be underlined for these processes: emulsion problems are avoided, large and constant exchanged area is involved and independent fluid dynamics allows an easily controlled operation.

Membrane contactors have been applied for many concentration and distillation processes generally called under the generic name of membrane distillation (MD). In such thermal driven processes an aqueous heated solution (feed side) is in contact with a porous hydrophobic membrane whereas an extractive phase is circulated in the other side [2,3]. The hydrophobic nature of the membrane prevents the penetration of liquid phase and creates a liquid vapor interface at the entrance of each pore. Volatile compounds diffuse across the membrane and are condensed (in a liquid phase or under a fresh wall), removed by vacuum (vacuum membrane distillation or VMD [4,5]) or by a gas phase located in the permeate side (sweep gas membrane distillation or SGMD [6,7]). These processes are generally carried out at temperatures much higher than room temperature and then, are not well adapted for the concentration of thermal sensitive solutions. In that view we developed a new type of SGMD process which has been called membrane evaporation (ME) [8]. This process was carried out at very soft conditions (low transmembrane pressures and temperatures) avoiding any risk of thermal degradation of compounds. Therefore, it is well adapted for the concentration of solutions containing thermo-sensitive products such as biological, pharmaceutical or food solutions. In this earlier work [8] the membrane contactor was built with metallic membranes which have a dual function: first they acted as classical contactors creating an interface between an aqueous and a gas phase and secondly they could be heated by an infrared lamp in order to decrease the thermal polarization effect. However, this heating method resulted in many problems of heat loss and water phase heating. In addition, water evaporation fluxes were measured by a weight loss method [2,3,8] which is not very precise and requiring a high number of experiences.

The modeling of membrane contactors has been generally carried out with classical resistances-in-series models taking into account the resistances of different boundary layers and the mass transport mechanism through membrane pores. We have already

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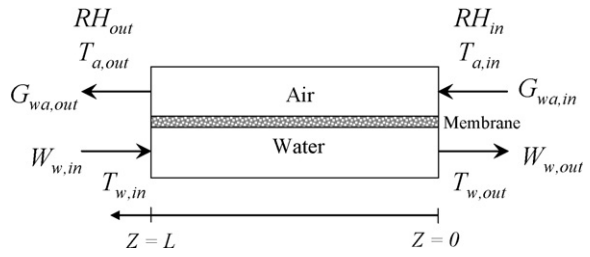

Fig. 1. Schematic representation of EM process module for a counter-current configuration with air and water flowing in the *Z*-axis direction (membrane length). $T_{a,in}$, $T_{a,out}$ and $T_{w,in}$, $T_{w,out}$ are the input and output temperatures of air and water, respectively, RH_{in} and RH_{out} are the input and output relative humidity of air. The input and output water flow are $W_{w,in}$ and $W_{w,out}$ and $G_{wa,in}$ and $G_{wa,out}$ are the input and output air flow.

reported some works concerning the modeling of membrane contactors of different geometries for osmotic evaporation (OE) [9,10], aroma compounds and metals extraction [1,11,12]. The SGMD process has been modeled using the same type of resistances-in-series models named above; Bansini et al. [7] have developed a model considering the coupling of Knudsen and molecular diffusion for the vapor transport through the membrane porosity, they gave a first approach of the thermal polarization phenomenon for this kind of evaporation process. The coupling of mass and heat transfers by convection and conduction in a global model for the SGMD has been reported by Khayet et al. [13,6]; they considered the viscous flow in addition to the Knudsen and molecular diffusion for the mass transport through the membrane porosity. These authors demonstrated that during the evaporation process the temperature decreased in the neighbouring of the membrane surface in the aqueous phase side of the contactor. Other approach using Stefan–Maxwell equations has been also reported by Rivier et al. [14] and Boi et al. [15].

This work is the first part of a study on membrane evaporation process carried out with flat hydrophobic metallic membranes. In this present work a complete model using the classical in-series-resistances approach and coupling heat and mass transfer has been developed and validated with experimental results obtained from membrane module with different metallic membranes. Then simulations have been carried out in order to study theoretically the influence of different structural characteristics of the membranes as well as operating parameters on evaporating fluxes.

2. Theoretical considerations

The objective of the modeling of this type of membrane process is to optimize it by determining the main resistances to the transfer and estimating the evaporating flux evolution with the operating conditions and membrane structure. We have already presented similar models to describe the mass transfer across porous hydrophobic membranes considering the gas transport in a porous media, like Knudsen or molecular diffusion as well as convection. These models of gas transport have been coupled to classical in-series-resistances models in order to characterize globally the system and then calculate water fluxes [9,10]. In a preliminary work on ME process we developed a first simplified model for the mass transfer but neglecting thermal effects [8]. Indeed, in this work we complete this previous model and consider also the coupling of mass transfer with heat transfer effects. In this type of process, heat transfer should be taken into account because during evaporation process, the temperature of the water–air interface decreases and then gas physical properties, water vapor pressure and the efficiency of the process will be influenced.

Fig. 1 gives a schematic representation of the membrane module considered for the model establishment in the case of a counter-current configuration.

The development of the model for the case of water solution concentration was carried out taking into consideration the following assumptions:

(1) Steady state operation.
(2) Ideal gas behavior.
(3) The total pressure is constant in the whole gas compartment and across the membrane.
(4) The proprieties of the membrane are perfectly homogeneous.
(5) No mass boundary layer in the liquid side is considered since it is pure water.
(6) Air solubility in water is negligible.
(7) The water–air interface is at thermodynamic equilibrium.

2.1. Mass transfer

As it was explained above, the model is based on resistances-in-series. They correspond respectively to the boundary layer which is formed in the gas phase at the surface of the hydrophobic porous layer and to the hydrophobic porous layer which is filled with air and water vapor. We considered the solution to be treated as only pure water; thus the resistance to the mass transfer due to this boundary layer could be ignored. In Fig. 2 are shown schematically the different resistance layers considered for modeling purposes in the case of counter-current configuration. In spite of the hydrophobic coating treatment, we took the assumption that when membranes present an asymmetric structure constituted of a separative layer and a support, this last which generally presents very large pores, is filled with liquid water. In this work three different metallic membranes have been used: membrane A which presents an homogenous structure and membranes B and C which are constituted by a wide mesh support and a thin porous separative layer. The thickness considered for the mass transfer equations through the metallic membrane concerns only the separative layer for membranes B and C and the total membrane thickness for self-standing membrane A.

As explained above, the resistances-in-series model has been successfully used to describe the mass transfer in similar membrane operation processes [1,9]. This model considers a mass transfer resistance in each layer. So, the global mass transfer coefficient and mass water vapor flux can be written as

$$k_T = \left(\frac{1}{k_{bl}} + \frac{1}{k_m}\right)^{-1} \tag{1}$$

$$N_w = k_T(P_w^{Sat} - p_{w,3}) \tag{2}$$

For this calculation we have to determine k_T and water partial pressures at liquid/gas interface ($p_{w,1}$), membrane/gas interface ($p_{w,2}$) and gas bulk phase ($p_{w,3}$) (for instance see Fig. 2).

2.1.1. Mass transfer through membrane pores

As far as all of the membranes have a macroporous structure (see Section 3) we consider that the molecular diffusion is probably the main mass transport mechanism through the porosity [1,9,16]. This assumption can be verified by calculating the Knudsen number, which in our operation conditions takes values ranged between 0.04 and 0.06 (for pressures between 0.9×10^5 and 1.013×10^5 Pa and temperatures varying from 298 to 318 K). Then, the molar flux of water vapor across the membrane is given by Eq. (3) which concerns a mixture of air and water vapor [17]:

$$n_w = -cD_{w,air}^e \frac{\partial y_w}{\partial X} + y_w(N_w + N_{air}) \tag{3}$$