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Breakthrough in a flat channel membrane microcontactor

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ABSTRACT

In literature three different models describing the breakthrough pressure were identified: the Young–Laplace model, the Kim–Hairrott–Zha model and the Franken et al. model. However, large differences between experimental results and model are often reported and a comparative study is lacking. In the present study, the different models were experimentally validated by measuring breakthrough pressures in liquid/liquid systems. The average differences between model and experimental results for the three models were respectively 37.5%, 26.7% and 32.2%. The Kim–Harriott–Zha model obtained the best results, however for the model it is advised that a solvent system with a low contact angle is searched to determine the maximum correction angle. Therefore a fourth model was proposed omitting this step and yielding an average deviation of 25.0%.

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1. Introduction

Membrane contactors are interesting tools in separation processes, which have evolved the last two decades into well-accepted unit operations (Pabby and Sastre, 2013). The membrane immobilizes the interface between two phases, liquid–liquid or gas–liquid, thereby allowing operation in parallel flow without mixing or disrupting the flows. In this way liquid–liquid extraction can for instance be performed, hence omitting a compulsory often slow phase separation step. A disadvantage of classical liquid–liquid extraction is indeed the formation of stable emulsions, but also foaming, unloading and flooding often poses problems (Gabelman and Hwang, 1999). Combining a membrane with shallow channels, high mass transfer kinetics can be easily reached (Hereijgers et al., 2013), requiring only a few minutes to obtain thermodynamic

equilibrium. Kiani et al. (1984) and D’Ella et al. (1986) demonstrated this in different membrane configurations, using both a flat membrane and hollow fibers. Several applications have been reported, such as the removal of phenols out aqueous waste streams (González-Muñoz et al., 2003). Using 1-decanol as solvent and a concentrated aqueous NaOH solution as stripping solvent in a second membrane contactor, 99% of the phenol could be removed. Bocquet et al. (2006) extracted aroma compounds from aqueous solvents using n-hexane and Dupuy et al. (2011a) extracted terpenes from lemon essential oil. For the recovery of metal ions Maruyama et al. (2004) built a microfluidic channel with intermittent partition walls instead of a membrane to pin the interface between the pillars, creating a stabilizing effect.

Supported liquid membrane (SLM) extraction is a special case of membrane extraction (Audunsson, 1986; Jönsson et al.,

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1993). In SLM the pores are filled with a wetting organic solvent and the channels at both sides of the membrane contain water-based solvents at a different pH. Chargeable solutes enter the contactor at a pH value at which they are uncharged, enabling transport through the organic solvent. Once entered in the aqueous acceptor phase, the solutes become charged due to a different pH, preventing them from back-diffusion. SLM is a technique primarily used in sample preparation, where the acceptor phase often stands still, pre-concentrating the sample prior to analysis. Additionally an electric field can be applied across the membrane to accelerate mass transfer. Pedersen-Bjergaard and Rasmussen (2006) demonstrated this, by immersing a polypropylene hollow fiber housing the acceptor solution and holding the organic solvent (typically 2-nitrophenyl octyl ether) in the pores into the donor solution. The hollow fiber was mechanically closed at one end preventing leakage of the acceptor solution into the donor solution. Placing an electrode in each phase a recovery of 70–80% was reached in typically 5 min only, extracting various basic drug substances such as pethidine, nortriptyline, methadone, haloperidol and loperamide.

Besides liquid–liquid extraction, membranes are also used as interface stabilizer in membrane distillation modules or gas absorption modules. Using a hydrophobic membrane, the aqueous liquid feed is fixed at the pore mouths, preventing it from entering or wetting the pores. Only vapor is hence able to migrate through the pores. A cold aqueous liquid or surface at the other side of the membrane can subsequently condense the vapor inside the module. Alternatively a sweeping gas or vacuum can be used requiring however a large external condenser (Alkhubdhiri et al., 2012). Membrane distillation is applied for several applications such as desalination (Banat and Simandl, 1998), removal of heavy metals (Zolotarev et al., 1994) and treatment of organic polluted aqueous waste streams (El-Abbassi et al., 2013). For gas absorption one side of the membrane is contacted with liquid and the other side with gas. Mavroudi et al. (2006) examined in this way the mass transfer resistance of CO₂ absorption using a hollow fiber.

Critical in all these membrane operations is the stability of the parallel flow profile, which is the purpose of the membrane. By capillary action the interface between both phases should be pinned preferably at the pore mouth. If the interface is pinned inside the pores, the membrane is partially wetted, and has as a consequence a negative impact on the mass transfer in gas–liquid absorption. Wang et al. (2005) reported a reduction of as much as 20% of the global mass transfer coefficient even if the pores are only 5% wetted. When the operation conditions are not carefully selected, the pressure difference across the membrane can become excessive. At that moment one of the phases will flow through the pores and disperse in the other phase, making phase separation again necessary and the membrane irrelevant. This phenomenon is called breakthrough. However, the operation conditions or pressure difference across the membrane at which this occurs are often differently defined in the literature and no comparative study is performed.

In the present paper, the validity of different models describing the breakthrough pressure is examined by experimentally measuring breakthrough pressures, focusing on liquid–liquid extraction systems.

2. Breakthrough pressure models

Based on the pressure profile of both phases two situations can be defined. In the first situation the pressure of the non-wetting phase is higher than the pressure of the wetting phase. In this classical situation the non-wetting phase will not enter the pores by capillary forces, holding the interface at the pore mouth. At the breakthrough pressure, also called the critical entry pressure, the non-wetting phase will penetrate through the pores and disperse in the wetting phase. To calculate this breakthrough pressure the Young–Laplace law (Eq. (1)) is most often used in the literature (Alkhubdhiri et al., 2012; García-Payo et al., 2000; Bougie and Illiuta, 2013; Saffarini et al., 2013; Dupuy et al., 2011b; Lawson and Lloyd, 1997):

$$\Delta P_b = \frac{-2\gamma\beta \cos \theta}{r} \quad (1)$$

with γ the interfacial tension between the two immiscible liquids in liquid–liquid operation. In gas–liquid operations γ is replaced by the surface tension σ . The influence of the pore shape is represented by β . The Young–Laplace law is defined for cylindrical pores and therefore β is often lacking, as a value of 1 is assumed in that case. For irregular shapes a value between 0 and 1 is found. θ is the contact angle between liquid 1, liquid 2 or gas phase and the membrane material and r is the maximum pore radius.

Instead of correcting the Young–Laplace equation using the β -coefficient, Kim and Harriott (1987) proposed a different model (Eq. (2)). They noticed that some membranes have a fibrous structure like paper or a mat of glass wool and that the pores are the irregular spaces between the adjacent fibers. Therefore Kim and Harriott (1987) assumed it is more accurate to describe the pores as a donut-like geometry, resulting in the following model (Eq. (2)) to describe the breakthrough pressure.

$$\Delta P_b = -\frac{2\gamma \cos(\theta - \alpha)}{r(1 + (R/r)(1 - \cos \alpha))} \quad (2)$$

with γ the interfacial or surface tension, θ the contact angle, r the smallest pore radius along the pore passage holding the largest pore bottleneck, R the radius of the fibers of the membrane forming the pores (Fig. 2) and α the correction angle for the pore shape which can be calculated from the contact angle. Setting the derivative of (Eq. (2)) to zero yields the following equation (Eq. (4)), from which α can be calculated:

$$\frac{d(\Delta P_b)}{d\alpha} = 0 \quad (3)$$

$$\sin(\theta - \alpha) = \frac{(R/r) \sin \theta}{1 + (R/r)} \quad (4)$$

For air–liquid systems the model yields adequate correlations but for liquid–liquid system deviations are large and unsatisfactory (Kim and Harriott, 1987), unfortunately no explanation for these observations is given. Zha et al. (1992) proposed a similar model as (Eq. (2)) by studying SLM systems and introduced a restriction for the correction angle α . Its absolute value can never be higher than the maximum correction angle α_m . An estimate of α_m is obtained by measuring the breakthrough pressure with a solvent system with a low contact angle. Zha et al. (1992) used a 25% aqueous ethanol solution and air as the second phase to measure this and

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