



# On individual resistances of selective skin, porous support and diffusion boundary layer in water vapor permeation



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## ABSTRACT

The efficiency of gas permeation processes is typically determined by two main parameters: selectivity and permeance. While selectivity depends to a large extent on chemical nature of the selective polymer layer, permeance is often dominated by fluid dynamics next to the membrane interface. A well-known phenomenon in terms of diffusive transport limitation is concentration polarization. It occurs in both the stagnant boundary layer and the porous support. The latter is called internal concentration polarization. The more selective and permeable a material is, the more severe the impact of concentration polarization can be. As a result process optimization is an issue far beyond the mere optimization of the selective membrane layer. The accurate identification and quantification of the limiting resistances other than the selective layer is a major challenge. Hence we introduce a systematic characterization approach to deconvolute the total mass transfer resistance. By combining single and mixed gas measurements, we quantify contributions of the selective skin, the porous support and the stagnant boundary layer. All tests were performed with a system comprising water vapor and air, with water vapor as the preferentially permeating component. Depending on process parameters, boundary layer resistance was found to be larger or in the same order of magnitude as the one of the selective layer. The influence of the porous substrate varied with the materials used. The methodology developed is important for humidification as well as dehydration processes, in particular for enthalpy exchangers in building ventilation systems.

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## 1. Introduction

In contrast to pressure driven liquid membrane processes, most gas permeation processes are driven by a concentration gradient. The latter is established by a difference in gas phase partial pressure. Transport through a dense polymeric gas permeation membrane is typically described by the well known solution–diffusion model [1] given in the following equation:

$$P = D \cdot S \quad (1)$$

Here permeability  $P$  equals the product of solubility  $S$  and diffusivity  $D$ . It is assumed that gas molecules dissolve on the feed side of the selective skin layer, diffuse through the polymer and desorb on the permeate side. A separation of two or more species is obtained by a difference in either solubility or diffusion coefficients. Even though the use of solution and diffusion coefficients is more fundamental, many publications report permeances instead. The main reason is

that permeance data is much easier to obtain. However permeance has to be handled with care if used for process design. Often single gas permeances do not hold for mixed gas applications [2,3]. Effects like membrane swelling, plasticization, competitive sorption and counter diffusion can have a tremendous impact on permeance. Additionally mass transfer is lowered due to boundary layers next to the membrane surface. Concentration profiles evolve due to the imbalance of high fluxes through the membrane and lower fluxes within this boundary layer. The phenomenon is well known as concentration polarization [4]. While many publications have addressed concentration polarization in liquid systems [5–9] only few publications have focused on gaseous systems [10–13]. A common assumption is that gas–gas diffusion coefficients are large enough to immediately compensate polarization effects.

Motivated by contradictory statements given in literature we report an experimental approach to deconvolute the total mass transfer resistance in terms of water vapor transport. Most polymers are highly water vapor permeable and show a significant selectivity of H<sub>2</sub>O over N<sub>2</sub>, O<sub>2</sub> and CO<sub>2</sub> [13]. This combination of high permeabilities and selectivities is a prerequisite for severe concentration polarization [14]. By focusing on water vapor transport it is possible

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to apply water vapor analytics instead of complex gas chromatography. A system which potentially suffers from concentration polarization issues is what we call a membrane-based enthalpy exchanger. Enthalpy exchangers are typically operated with a sweep gas, which doubles the potential impact of boundary layer effects.

## 2. Theory

Two concentration profiles of a solution–diffusion membrane are given in Fig. 1. Concentration at the membrane interface is described with gas–liquid equilibrium. According to Fick's law the concentration profile of the selective layer is linear. Differences between ideal behavior (Fig. 1a) and real gas behavior (Fig. 1b) are caused by two major issues. Firstly a change of intrinsic membrane properties due to plasticization, swelling, counter diffusion and competitive sorption [2,3]. Secondly concentration polarization effects within the adjacent flow [5,12] and the membrane support [15,16] respectively. While the change of membrane properties is beyond the scope of this work, the impact of polarization effects is visualized in Fig. 1b. In a system comprising air and water vapor the latter is preferentially transported. Thus components different from water vapor, i.e. nitrogen and oxygen, are retained by the membrane surface. A similar effect can be observed in the permeate. Water vapor is enriched at the membrane surface. Its transport into the bulk is controlled by diffusion. According to Fig. 1b boundary layer effects are additionally intensified by internal concentration polarization within the mechanical support. The depletion of water vapor in the feed and its enrichment in the permeate causes a lowering of the transmembrane concentration gradient. As a consequence water flux decreases compared to the ideal case of Fig. 1a.

### 2.1. Resistances in series

The mole flux of component  $i$  through a membrane with a surface area  $A$  is given by

$$\dot{n}_i = k_{tot} \cdot A \cdot (c_{i,f} - c_{i,p}). \quad (2)$$

Here  $c_{i,f}$  and  $c_{i,p}$  equal the bulk concentration of component  $i$  in the feed and permeate respectively.  $k_{tot}$  is the total transport coefficient summarizing all resistances in between the bulk phases.  $k_{tot}$  is typically calculated by a resistance in series model according to Fig. 1b

$$\frac{1}{k_{tot}} = \frac{1}{k_{bl,f}} + \frac{1}{k_{mem}} + \frac{1}{k_{sup}} + \frac{1}{k_{bl,p}}. \quad (3)$$

While it is assumed that transport coefficients of the membrane  $k_{mem}$  and the support  $k_{sup}$  are independent of fluid dynamics, the corresponding parameters of feed and permeate boundary layer ( $k_{bl,f}$ ,  $k_{bl,p}$ ) are known to be a function of cross flow velocity. With a sweep ratio (feed flow rate/sweep flow rate) of one and equal feed and sweep channel geometry equation (3) simplifies to

$$\frac{1}{k_{tot}} = \frac{1}{k_{mem}} + \frac{1}{k_{sup}} + \frac{2}{k_{bl}}. \quad (4)$$

For non-supported, symmetric membranes  $k_{sup}$  does not need to be considered, which finally results in

$$\frac{1}{k_{tot}} = \frac{1}{k_{mem}} + \frac{2}{k_{bl}}. \quad (5)$$

### 2.2. Deconvolution approach

Fig. 2 suggests a deconvolution approach requiring a systematic combination of multiple experiments. In a first step membrane

resistance  $1/k_{mem}$  is obtained via constant-volume variable-pressure measurements [17]. We assume that this resistance also holds in the presence of nitrogen and oxygen. For most polymers solubility of  $N_2$  and  $O_2$  is low compared to the one of water vapor. Competitive sorption effects can be neglected. Swelling due to water vapor is already considered within single gas data. Counter diffusion is neglected, since  $N_2$  and  $O_2$  concentrations are almost equal on both sides of the membrane. The second parameter, which is obtained experimentally, is the total transport resistance  $1/k_{tot}$ . This parameter is a direct outcome of mixed gas measurements. By testing a symmetric membrane first, the boundary layer resistance  $1/k_{bl}$  can be calculated according to Eq. (5). As a result  $1/k_{bl}$  is given as a function of cross flow velocity and fluid properties. Repeating the same measurements with an asymmetric membrane leads to the quantification of the support resistance  $1/k_{sup}$ . This time equation (4) is used to calculate  $1/k_{sup}$  by replacing  $1/k_{bl}$  with the data of the symmetric membrane.

It needs to be emphasized that membrane morphology could in principle change fluid dynamics of the adjacent flow. In turbulent flow surface roughness has a strong impact on pressure loss [18] and thus on boundary layer correlations [19]. However this is not the case for laminar flow. Here roughness typically changes the Reynolds number at which laminar flow becomes turbulent. For macro-pipes ( $d_h > 3$  mm) with roughness heights below  $\epsilon = 0.05 d_h$  the critical Reynolds number is  $Re_c = 2300$  [20]. Experiments in the course of this study were performed at  $Re < 1000$ , while all samples showed surface roughnesses of  $\epsilon < 0.05 d_h$ . Thus in our case boundary layer resistance should be independent of membrane sample.

### 2.3. Overall transport coefficient

A simplified sketch of gas flow along the membrane is shown in Fig. 3. As the membrane is operated in counter current flow, feed inlet is at  $x=0$  while permeate inlet is at  $x=1$  respectively.

With a sweep ratio of one (equal flow rates  $\dot{V}$  on both sides of the membrane), the water vapor transfer can be calculated by means of inlet ( $c_{f,0}$ ,  $c_{p,1}$ ) and outlet concentrations ( $c_{f,1}$ ,  $c_{p,0}$ ) according to

$$\dot{n} = \dot{V} \cdot \frac{(c_{f,0} - c_{f,1}) + (c_{p,1} - c_{p,0})}{2}. \quad (6)$$

By combining Eqs. (6) and (2) transport coefficient  $k_{tot}$  is expressed as

$$k_{tot} = \frac{\dot{n}}{A_{mem} \cdot (c_f - c_p)}. \quad (7)$$

Bulk concentrations  $c_f$  and  $c_p$  are calculated using logarithmic mean values

$$c_f = \frac{(c_{f,0} - c_{f,1})}{\ln c_{f,0} - \ln c_{f,1}} \quad (8)$$

$$c_p = \frac{(c_{p,1} - c_{p,0})}{\ln c_{p,1} - \ln c_{p,0}}. \quad (9)$$

### 2.4. Membrane transport coefficient

Results of single gas measurements are most often reported in gas permeation unit (GPU<sup>1</sup>), whereas transport coefficients  $k$  are given in m/s. Consistent units require conversion. An appropriate term comprises gas constant  $R$ , temperature  $T$  and the molar volume of an ideal gas (at STP)  $V_{m,ig}$

<sup>1</sup> 1 GPU =  $2.7 \cdot 10^{-3} \text{ m}^3/(\text{m}^2 \text{ hbar})$ .

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