



# Approximate models of concentration-polarization in Pd-membrane separators. Fast numerical analysis



Olga Nekhamkina\*, Moshe Sheintuch

Department of Chemical Engineering, Technion-Israel Institute of Technology, Haifa 32000, Israel

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## ABSTRACT

Several approximate solutions are derived to predict the concentration polarization effect in a shell-and-tube Pd-membrane empty separator with permeate flow either in the tube or in the shell. The first two approximations (Nekhamkina and Sheintuch (2015) [22]) are governed by ODEs describing the axial profiles of the average variables coupled with algebraic relations that describe the radial profiles of velocity components and species concentrations. The hydrogen membrane flux is approximated by a mass transfer coefficient  $k = D(\partial c_{H_2}/\partial r)_w / (c_{H_2} - c_{H_2,w})$  expressed via the Sherwood number  $Sh = kd/D$  which is a geometry-dependent parameter ( $D$  is hydrogen diffusivity,  $c_{H_2,w} = p_{H_2,w}^{ret}/RT$  is its membrane wall concentration and  $d$  a characteristic length). Here  $Sh$  is obtained by 2-D CFD simulations in a wide range of parameters and when plotted vs the separator length, it reaches an asymptotic value that matches the predicted values and is independent of operating conditions.

The third approximation reduces the polarization effect to an algebraic equation by approximating the effectiveness factor  $\eta = [(p_{H_2,w}^{ret})^{0.5} - (p_{H_2,w}^{per})^{0.5}] / [(p_{H_2}^{ret})^{0.5} - (p_{H_2}^{per})^{0.5}]$  which depends mainly on the ratio of the mass-transfer coefficient to the membrane permeance parameter  $\Gamma = \Gamma(Sh)$ . This expression can be used for fast design of separators that avoids or accounts for polarization effects. The three approximations offer a trade-off between accuracy and ease of application.

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## 1. Introduction

The permeance of a Pd or Pd-based membrane is the key parameter in the design of membrane separators and membranes. With the incorporation of membranes of better permeance and the integration of heat supply to the reactor through its wall, radial gradients in membrane reactors may become important. The separation of hydrogen at the membrane wall will cause, in a wide separator or reactor, its concentration to be different than that in the bulk, while that of the other species will be larger (concentration polarization). That will diminish the hydrogen gradient, and the effect may be compounded by increased permeance inhibition if one of the other species adsorbs well on the membrane. Many studies have attributed membrane permeance inhibition to either concentration polarization or to co-adsorption inhibition, without supporting their conclusion with concrete simulations, approximations or criteria.

Permeance measurements are usually conducted in empty (unpacked) shells and tube arrangements, with a hydrogen

mixture in inerts (or in reactants or products). Measurements with pure hydrogen feed yield directly the permeance, the constant in Sieverts' law [1]

$$J_{H_2}^{mem} = K_{H_2} \left( \sqrt{p_{H_2,w}^{ret}} - \sqrt{p_{H_2,w}^{per}} \right) \quad (1)$$

but such values usually differ from those made in a mixture either due to hydrodynamic effects on diffusive transport or due to co-adsorption inhibition. We ignore the latter effect here. The impact of concentration polarization on reducing the apparent constant of Sieverts' law, compared with that measured in hydrogen, or its effect on deviation from the Sieverts' law was measured experimentally in many separator systems [2–11].

To account for radial gradients in membrane separators or reactors with permeable walls several studies have employed 2-D numerical simulations of mass and momentum balances [2,9–15]. Such studies employ either special software like COMSOL or FLUENT, or original homemade codes but do not allow to derive a design criterion predicting when this effect can be ignored. Semi-empirical approximations using effective mass transfer resistance to approximate the flux were addressed in Refs. [8,9,11,12,16–21], expressing it via the Sherwood number ( $Sh = kd/D$ , where  $k$  is the mass transfer coefficient,  $d$  is the characteristic length,  $D$  – the

\* Corresponding author.

E-mail address: [aermwon@technion.ac.il](mailto:aermwon@technion.ac.il) (O. Nekhamkina).

diffusion coefficient). This approach was applied to experimental results [8] and was also intuitively suggested for model reduction based on the similarity between the mass and the heat transfer [11,21]. However, it is not obvious that such an analogy is valid, because of the different boundary conditions employed in the separator vs heat exchanger.

Approximate separator models accounting for the polarization effect were proposed in our recent study [22] for a tube and for an annular channels. These models are based on uncoupling the problems of describing the hydrodynamic field and the concentration field, assuming a constant total molar concentration (i.e., constant pressure). The axial velocity profile is approximated as a product of the averaged velocity  $\langle u \rangle$  and a known radial function  $f(r)$  defined by the channel geometry (ensuring the appropriate BCs). We used parabolic functions  $f(r)$  that ensures in the case of impermeable walls an exact solution for a tube (Poiseuille profile) which is also a good approximation for an annular channel. The radial velocity profile is constructed using the continuity equation and a prescribed  $u(r)$ -profile with the normal wall velocity ( $v_w$ ) as a parameter.

The approximate 2-D concentration profiles were constructed using a known velocity distribution and employing certain assumptions concerning averaging (see [22] for details). Thus, the approximate solution is governed by the set of ODE equations with respect to the average variables coupled with algebraic relations to define the radial profiles. The problem is closed by finding the wall normal velocity ( $v_w$ ) using the Sieverts' law.

The proposed approximations allow to define the following: (i) a mass-transfer coefficient ( $k^{app}$ ):

$$k^{app} = \frac{D_i(\partial c_i^{app}/\partial r)_w}{\langle c_i^{app}(r) \rangle - c_{iw}} \quad (2)$$

showing that the apparent Sherwood number ( $Sh^{app} = k^{app}d/D$ ) is only geometry dependent; (ii) a separation parameter  $\Gamma$  which represents the ratio of diffusive to permeating flux, which for membrane that follows Sieverts' law (1) takes a form:

$$\Gamma = \frac{DSh}{d\kappa\sqrt{p^{ret}}}, \quad (3)$$

where

$$\kappa = \frac{K_{H2}}{c_{tot}}, \quad (4)$$

and  $\kappa(p^{ret})^{0.5}$  is the characteristic wall normal velocity

Two approximate models were suggested in Ref. [22]: The first one allows to predict 2D concentration and velocity profiles, the second one presents its reduced version and allows to predict the 1-D average and the wall profiles of the state variables. These approximations were verified [22] against published CFD simulations [13,14] conducted with a COMSOL 4.0 version showing a good agreement; the agreement with experimental result [6] was poorer due to the erroneous diffusivity value (this will be commented in Section 3).

In the present paper we verify three models vs our own CFD data, allowing us to enlarge the domain of parameters at will. These include the two approximations above and a 3rd one that reduce the problem to an algebraic presentation by accounting for the polarization effect via the effectiveness factor  $\eta$ :

$$\eta = \frac{\sqrt{p_{H2w}^{ret}} - \sqrt{p_{H2w}^{per}}}{\sqrt{\langle p_{H2}^{ret} \rangle} - \sqrt{p_{H2w}^{per}}} = \eta(\Gamma) \quad (5)$$

expressing the ratio of the actual flux over the flux calculated from the average concentration. In turn, the effectiveness factor is

dependent on the separation factor  $\eta = \eta(\Gamma)$ , as suggested in [23]. In the present work we check its validity and refer to the appropriate model as approximation no. 3: Using this approximation coupled with relation (3) allows to find an analytical solution of the governing ODE equations and to express the axial average and the wall concentrations and the transmembrane flux profiles by algebraic functions.

We focus on the analysis of the  $Sh(z)$  and  $\eta(z)$  profiles obtained by CFD, comparing the former to its predicted value and the latter to its approximated profile. This offers a rigorous basis for the approximation by the mass-transfer resistance. The comparison of the approximated and the exact transmembrane flux profiles validates the proposed models.

The structure of this paper is the following: the exact and the approximate models are outlined in the next section. In Section 3 the approximate models are validated by comparison with the CFD simulations of (i) the  $Sh(z)$  and  $\eta(z)$  profiles, (ii) the 2-D velocity and the concentration fields, (iii) the transmembrane flux profiles. Finally, in this section the comparison with the experimental data [6] is presented. In concluding remarks we summarize the obtained results and discuss the proper separator design and the implementation of the proposed models for reactor simulations. The results are summarized in a Table.

## 2. Separator models

In this section we consider an exact and three approximate separator models of various degrees of complexity: (i) A 2-D model in a boundary layer approximation (Section 2.1) that is referred to below as an exact (CFD) one; (ii) A reduced 2-D model in which the hydrodynamic and the concentration fields are separated (Section 2.2) assuming that the total molar density is preserved ( $c_{tot} = \text{constant}$ ). This model was used below to derive in turn three approximate separator models. The first two models are briefly described in Section 2.3 where we address the approximations for Sh number and the separation parameter  $\Gamma$ . The third model is presented in Section 2.4, here we analyze the effectiveness factor  $\eta$ .

We addressed two axisymmetric geometries (i) A tube with transport at its wall and (ii) An annular cylinder with transport at an inner tube; approximate relations for a planar channel are presented as a limiting case of an annular domain of a finite width with  $R \rightarrow \infty$ . For all models the flow is assumed to be isothermal.

### 2.1. The exact 2-D axisymmetric channel flow model

We employ a 2-D separator model in a boundary layer approximation, i.e. assuming that (i) the transversal velocity component ( $v$ ) is much smaller than the axial component ( $u$ ), and that (ii) the gradients of the velocity components and species concentrations in the axial direction are much smaller than those in the radial direction. These assumptions lead to the governing equations, similar to those used in the boundary layer (BL) problem [24] and include the conservation laws of the total mass, the momentum and the mass fraction of the individual components coupled with the equation of state; the radial pressure gradient is negligible:

$$\frac{\partial(\rho u^{mas})}{\partial z} + \frac{1}{r} \frac{\partial(r\rho v^{mas})}{\partial r} = 0 \quad (6)$$

$$\frac{\partial[\rho(u^{mas})^2]}{\partial z} + \frac{1}{r} \frac{\partial(r\rho v^{mas}u^{mas})}{\partial r} = -\frac{\partial p}{\partial z} + \frac{\mu}{r} \frac{\partial}{\partial r} \left( r \frac{\partial u^{mas}}{\partial r} \right) \quad (7)$$

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