diffusion-fundamentals.org

The Open-Access Journal for the Basic Principles of Diffusion Theory, Experiment and Application

Simulation of N_2/CH_4 Counter-Diffusion in Composite Membranes of the Type Silicalite-1- α-Alumina

Vlastimil Fíla[1], Milan Kočiřík[2], Pavel Hrabánek[2], Arlette Zikánová [2], Libor Brabec[2], Bohumil Bernauer[1]

[1]Institute of Chemical Technology, Technická 3, 166 28 Prague 6-Dejvice, Czech Republic

[2]J. Heyrovský Institute of Physical Chemistry of ASCR, v.v.i., Dolejškova 3, 182 23 Prague 8-Libeň, Czech Republic, E-mail: milan.kocirik@jh-inst.cas.cz

1. Introduction

The membrane model used describes (i) mass transport in macroporous support comprising diffusion in transition region between Knudsen and bulk diffusion superimposed on convective flow and (ii) mass transport in zeolitic phase accompanied by a parallel transport in defects of zeolitic layer. The mass transport in the support is described by Dusty Gas Model (DGM) and that in the zeolitic phase by Generalized Maxwell Stefan (GMS) approach. Parallel transport in defects of zeolitic layer is also described by DGM. Modelling is exemplified on dynamics of a semi-open Wicke-Kallenbach (W-K) cell (cf. Fig. 1) operated with a flat silicalite-1-α-Al_2O_3 membrane using a N_2/CH_4 mixture. It appeared that measurement of dynamic responses of the semi-open W-K cell represents a rapid method to characterize composite membrane quality.

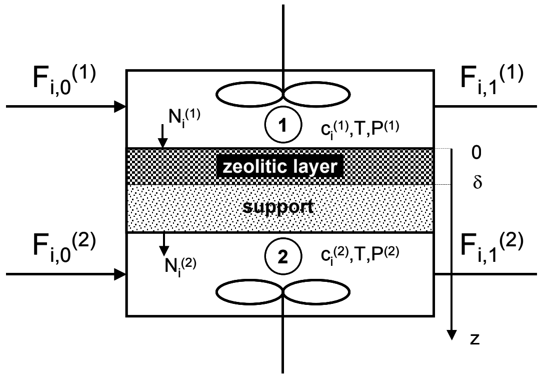

Fig. 1: Scheme of W-K cell

Semi-open W-K cell works with closed compartment (2). Dynamic responses of the cell evaluated with the system CH_4/N_2 are (i): $P^{(2)}_{N2}(\tau)$ and (ii) $P^{(2)}_{CH4}(\tau)$ i.e. single component responses to step change in $P^{(1)}_{I}$ at $\tau \rightarrow 0+$; (iii): $P^{(2)}(\tau) \equiv P_{tot} = P^{(2)}_{N2}(\tau) + P^{(2)}_{CH4}(\tau)$ started from the state $P^{(1)}_{CH4}(0) = P^{(2)}_{CH4}(0) = 0$ and $P^{(1)}_{N2}(0) = P^{(2)}_{N2}(0) = 100$ kPa by the step change to $P^{(1)}_{N2}(0+) = 80$ kPa, $P^{(1)}_{CH4}(0+) = 20$ kPa; (iv): $P^{(2)}_{CH4}(\tau)$ as the response to the same disturbance that defined in the case (iii).

2. Mathematical model

Mathematical model of the W-K cell which contains the above composite membrane involves the following relations:

(i) Mass balance equations of mixture components in individual compartments
(ii) Constitutive equations for components transport in silicalite-1 layer
(iii) Mass balance equations of components in silicalite-1 layer

(iv) Constitutive equations of components in defects of the silicalite-1 layer
(v) Mass balance equations of components in the defects of the silicalite-1 layer
(vi) Constitutive equations of components in the pore system of the support
(vii) Mass balance equations of components in the pore system of the support
(viii) Boundary conditions at silicalite-1 layer inlet and defect pore inlet
(ix) Matching conditions at silicalite-1 layer - support layer interface
(x) Boundary conditions at support outlet
(xi) Matching conditions at the interface silicalite-1 matrix– defect pore

3. Examples of the simulation

Figure 2 shows the results of calculations as the normalized responses $X(\tau) = \left(P^{(2)}(\tau) - P^{(2)}(0)\right) / \left(P^{(1)}_{CH_4}(0+) - P^{(1)}_{CH_4}(0)\right)$ of the total pressure in compartment (2) for a silicalite-1 membrane of the thickness 50 μm, free of the defects and with negligible mass transport resistance of the support. The total gas flow rate at the inlet of the compartment (1) i.e. $F_{N2,0}^{(1)} + F_{CH4,0}^{(1)} = 360$ ml.min^{-1}.

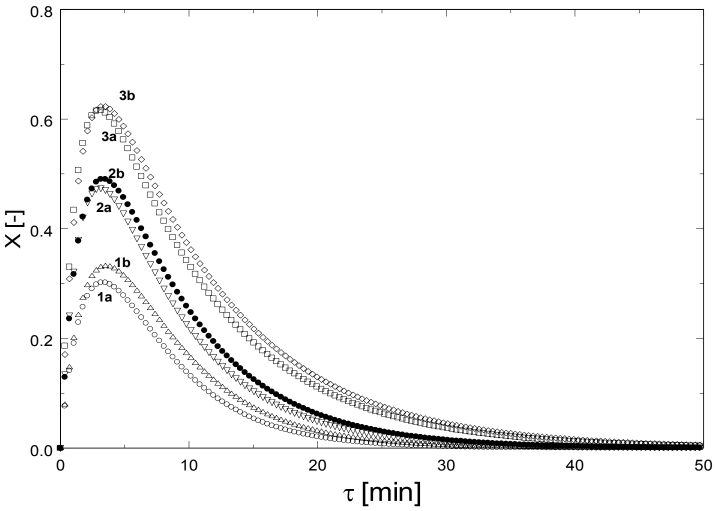

Fig. 2: **Effect of Maxwell Stefan cross-diffusion coefficients $Đ_{N2CH4}$ on X vs. τ responses,** Curves 1: $Đ_{N2} = 4.2 \times 10^{-9}$ m^2.s^{-1}, $Đ_{CH4} = 2.4 \times 10^{-9}$ m^2.s^{-1}; Curves 2: $Đ_{N2} = Đ_{CH4} = 3.3 \times 10^{-9}$ m^2.s^{-1}; Curves 2: $Đ_{N2} = 2.4 \times 10^{-9}$ m^2.s^{-1}, $Đ_{N2} = 4.2 \times 10^{-9}$ m^2.s^{-1}; Curves a: effect of $Đ_{N2CH4}$ is disregarded; Curves b: effect of $Đ_{N2CH4}$ is considered

4. Conclusion

Simulator of dynamic responses in W-K cell for composite membranes of the type silicalite-1-α-alumina was elaborated as a tool to evaluate membrane defects.

Acknowledgement The research was supported by GA ASCR via grant No IAA 400400909 and Ministry of Education of Czech Republic via grant No MSM 6046137301.