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Platform Molecule Removal from Aqueous Mixture with Organophilic Pervaporation: Experiments and Modelling

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Abstract

The work is motivated by a separation problem, which is ethanol removal from aqueous mixtures with membranes. Ethanol can be considered as promising biomass based platform molecule. The platform molecule includes several building-block chemicals grouped together, resulting in a range of downstream chemical products. To solve the target, organophilic pervaporation system is investigated using benchmarked Sulzer PERVAP™ 4060 membranes. Separation factors, total permeation fluxes, permeances and selectivities are experimentally determined. The target of this work is to parameter estimation for semi-empirical pervaporation model. The measured data are evaluated with improved pervaporation model by Valentinyi et al. [1]. Three different polymeric flat sheet membranes are investigated, PERVAP™ 4060, PERVAP™ 1060 and CELFA-CMG-OG010. It is found that the model can be applied also for each organophilic separation case.

Keywords

organophilic pervaporation, ethanol removal, platform molecule, mathematical modelling, parameter estimation

1 Introduction

As the era of fossil oil nears its end and environmental pertains describing to the extraction of non-sustainable fossil feedstocks use pressure to the petrol sector, new sustainable feedstocks for chemicals and materials will be appropriate. The conventional chemical industry relies mainly on a small set of base chemical building blocks that are fabricated globally on a mighty quantity [2-5].

Farmer and Mascal [2] have defined these resources as platform molecules with the followings: “A bio-based (or bio-derived) platform molecule is a chemical compound whose constituent elements originate wholly from biomass (material of biological origin, excluding fossil carbon sources), and that can be utilized as a building block for the production of other chemicals”. Ethanol (EtOH) is mentioned as platform molecule, it is determined the constituent of this biomass molecule is derived from sugars [2]. Fig. 1 shows more ethanol-based organic molecules [6].

Bozel and Petersen [7] have classified ethanol into the group of revisited platform molecules [8]. They have also summarized the main criteria for the inclusion and resulting technology needs of top 10 platform molecules. In the case of ethanol the most important recommendations are the optimization of fermentation organisms, development of biochemical production of alcohols from biomass and alcohol-water separations techniques [7].

This research focuses on the third recommendation, which is the alcohol removal from water mixtures. Pervaporation (PV) is selected for investigation of ethanol from aqueous mixtures. Pervaporation has more green specialties against to other traditional processes, such as distillation [9, 10]. There are simply actualization, energy-saving and no-pollution effects, furthermore no need for extra material to add for the separation [11]. Pervaporation method applied for organic-organic

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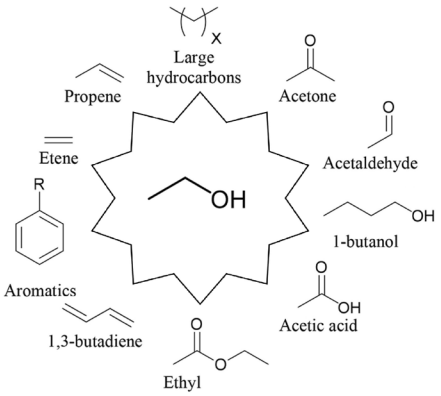

Fig. 1 Ethanol as a platform molecule [6]

separations, removal of low concentration organic from its aqueous mixtures and dehydration of organics [12].

The separated liquid mixture is vaporized at vacuum on the permeate side of dense membranes and the transport process can be described as sorption and diffusion phenomena. Depending on the affinity of permeating component, this method can be specified into main areas: hydrophilic PV and organophilic PV. Removal of organic, for our cases ethanol, organophilic membranes are applied. PV can be described by certain equations. The partial flux is calculated applying the following formula [13]:

$$J_i = P_i / (\Delta t \cdot A) \tag{1}$$

where P_i is the partial weight of component i in the permeate product, Δt is the time of duration of separation process and A is the effective membrane area.

Separation factor is determined by Eq. (2) [13]:

$$\alpha = \left(y_i (1 - x_i)\right) / \left(x_i (1 - y_i)\right) \tag{2}$$

where α is separation factor (dimensionless), x_i is weight fraction of water in feed and y_i is weight fraction of water of permeate. Pervaporation Separation Index (PSI) is specified [13]:

$$PSI = J \cdot (\alpha - 1) \tag{3}$$

The efficiency of pervaporation membranes can be determined by the permeance as partial flux normalized for driving force the pressure difference-normalized flux [13-15]:

$$P_i / \delta = J_i / \left((\gamma_{i0} \cdot x_{i0} \cdot p_{i0}) - (y_i \cdot p_3) \right) \tag{4}$$

The membrane selectivity β is calculated as the ratio of permeances [13-15]:

$$\beta = (P_i / \delta) / (P_j / \delta) \tag{5}$$

The ethanol removal is the actual task. As for the literature survey some papers were published in the separation of ethanol-water mixture by organophilic pervaporation. Table 1 summarizes a comparison of experimental data for the organophilic pervaporation of the ethanol-water mixture with polydimethylsiloxane (PDMS) membranes. The most widespread benchmarked material of organophilic PV is PDMS and the research focuses on the investment of this membranes.

The focus of this research is to represent the emerging method of organophilic PV and provide adequate understanding of the process for successful explanation of experimental data. The aim of this work is to investigate polymeric PV membranes and to fit parameters for semi-empirical transport model.

Table 1 Comparison of experimental data with composite PDMS membranes with different supports and without fillers for organophilic pervaporation of ethanol-water mixture

Membranes	T [°C]	F_{EtOH} [m/m%]	J_{total} [kg/(m²h)]	α [-]	PSI [kg/(m²h)]	Reference
PDMS - PTFE s.	30	2	0.10	10.0	0.9	Zhang et al. [16]
PDMS - CA s.	40	5	1.14	9.3	9.5	Luo et al. [17]
PDMS - PA s.	45	4	1.85	8.5	13.9	Shi et al. [18]
PDMS - CA s.	40	5	1.30	8.5	9.8	Li et al. [19]
PDMS - PS s.	42	5	1.44	6.7	8.2	Zhang et al. [20]
PDMS - PS s.	50	8	0.26	6.4	1.4	Guo et al. [21]
PDMS - PS s.	45	4	1.60	5.0	6.4	Shi et al. [18]
PDMS - PVDF s.	40	10	8.00	2.2	9.6	Chang et al. [22]
PDMS - CA s.	50	0.3	2.80	3.0	5.6	Mohammadi et al. [23]
PDMS	40	6	0.10	8.7	0.8	Naik et al. [24]
PDMS	30	5	0.05	8.0	0.3	Slater et al. [25]
PDMS graft copol.	48	6.6	0.03	6.6	0.2	Kashiwagi et al. [26]
PDMS	50	5	0.08	4.2	0.3	Lazarova et al. [27]

2 Materials and methods

The laboratory apparatus is P-28 membrane unit from CM-Celfa Membrantechnik AG (see Fig. 2). The equipment has 28 cm^2 effective area (A). Cross-flow circulation is achieved at constant value of ~182 L/h and the size of the feed tank is 500 mL [13].

The isotherm conditions are adjusted with an ultrathermostat. The vacuum on the permeate side is maintained with VACUUMBRAND PC2003 VARIO vacuum pump and kept up at 2 Torr (3 mbar). The permeate is gathered in two traps connected in series and cooled with liquid nitrogen to hinder loss of the permeate [13]. The ethanol concentration of the feed (F), permeate (P) and retentate (R) are measured with Shimadzu GC2010Plus+AOC-20 autosampler gas chromatograph with a CP-SIL-5CB column connected to a flame ionization detector, EGB HS 600 headspace apparatus is used for sample preparation [13, 29].

Composite PDMS flat sheet membranes are used in organophilic laboratory experiments. The pervaporation measurements are carried out at six different feed concentrations and three temperatures with Sulzer PERVAP™ 4060, as follows: 1, 3, 5, 10, 15 and 20 weight percent (m/m%) ethanol in feed and 50, 60 and 70°C.

The procedure of Valentinyi et al. [1] is elected for modelling of pervaporation, which is a development of basic Rautenbach model [30]. The concentration dependencies of the transport coefficient $\bar{D}_i$ and the temperature dependencies of the pervaporation mean the improvements [13, 31]. Eq. (6) shows the basic equation of the improvement PV model:

$$J_i = 1\Big/\left(1+\left\{\left[\bar{D}_i\cdot\exp\left(B\cdot x_{i1}\right)\right]\Big/\left(p_{i0}\cdot\bar{\gamma}_i\right)\right\}\right) \cdot\left[\bar{D}_i\cdot\exp\left(B\cdot x_{i1}\right)\right]\Big/\bar{\gamma}_i\left(\left(p_{i1}-p_{i3}\right)\Big/p_{i0}\right) \tag{6}$$

The fundamental Rautenbach model (Model I) and the advanced one (Model II) are applied for modelling of organophilic pervaporation experiments. $\bar{D}_i$ and activation energies E_i and in the case of improved model B parameters are estimated based on experimental data [13]. In Eq. (6) the exponential B parameter represents the concentration dependencies of the transport coefficients. Nonlinear estimation is applied by determining a regression custom loss function (Eq. (7)) in STATISTICA® program environment. The model verification can be taken with objective function (OF), which is minimized the difference of the measured and the modelled values.

$$OF=\sum_{i=1}^{n}\left(\left(J_{i,\bar{m}easured}-J_{i,\bar{m}odelled}\right)\Big/J_{i,\bar{m}easured}\right)^2 \tag{7}$$

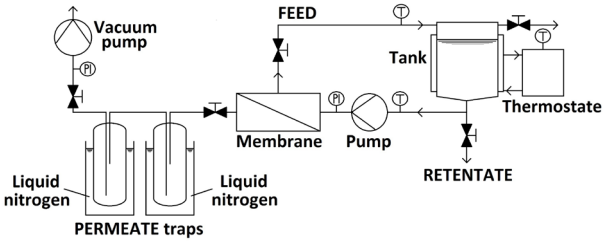

Fig. 2 Flowsheet of CM-Celfa P-28 Membrantechnik AG apparatus in PV mode [13, 28]

Further membranes are investigated for parameter estimation: Sulzer PERVAP™ 1060 and CELFA-CMG-OG010. The preliminary pervaporation experiments are carried out by Molina et al. [32] and conditions of measurements are described in detail in PhD Thesis of Molina [33].

3 Results and discussion

Fig. 3 presents the effect of feed concentration on the pervaporation achievement of the PERVAP™ 4060 organophilic membranes at different operating temperatures. It can be seen that increasing ethanol feed concentration and operating temperature increase the total fluxes. However, increasing the ethanol concentration decreases the separation factors. The maximal total flux of 4.03 kg/(m^{2}h) can be reached at 70°C and at the feed ethanol concentration of 18.4 m/m%. Compared with other literature results (see Table 1), it can be seen PERVAP™ 4060 has the highest total flux value and PSI and it follows the tendency of the total flux. Studying Table 1, it can be determined the maximum separator factor of 8.1 is also a high value. Furthermore, selectivity follows the trend of the separator factor.

It can be stated at higher ethanol concentration the separation effectiveness of the organophilic pervaporation membranes are decreased, similar tendency have been already published by Slater et al. [25], Lazarova et al. [27], Vane [34], Chai et al. [35] and Fu et al. [36].

Table 2, Table 3 and Table 4 summarize the estimated values of transport coefficients, activation energies, exponential parameters and minimized objective functions of the two models.

Comparison of the measured and calculated partial fluxes are presented in Fig. 4, Fig. 5 and Fig. 6.

Fig. 4, Fig. 5, Fig. 6 and Table 2, Table 3, Table 4 show that Model II is much more applicable for description of organophilic pervaporation than Model I, because the basic model presumes constant $\bar{D}_i$. Many authors have

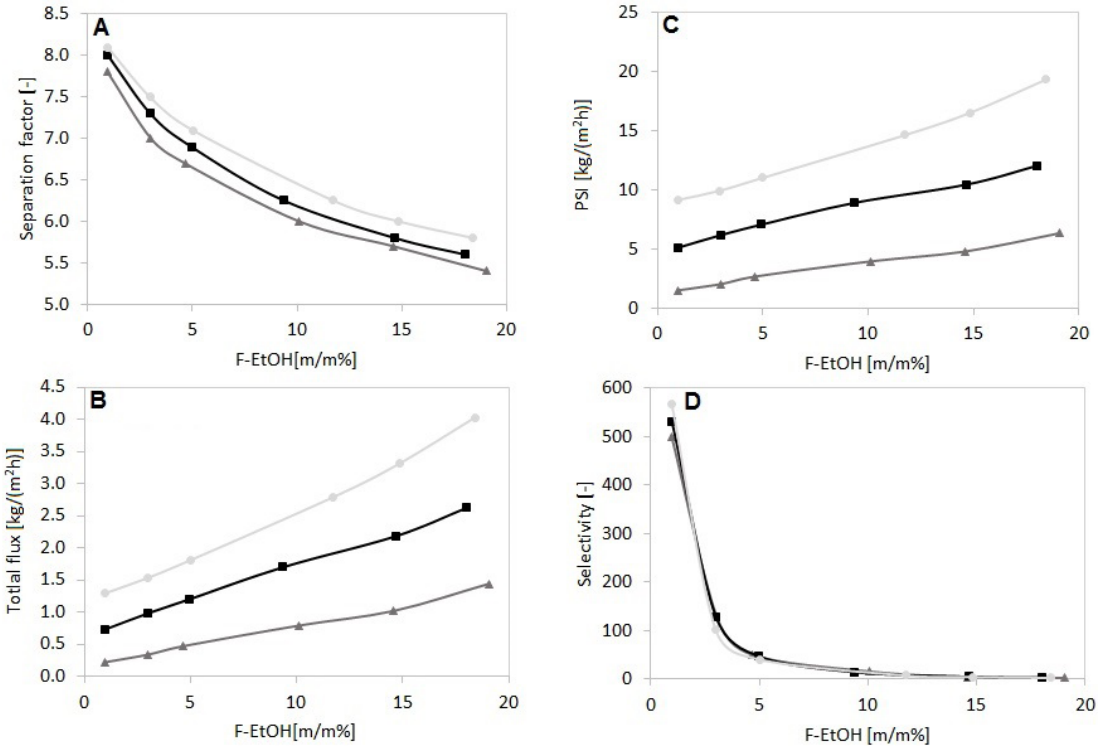

Fig. 3 Pervaporation performance as a function of feed concentration at different operating temperatures for PERVAP™ 4060 membrane (50°C: ▲; 60°C: ■; 70°C: ●)

Table 2 Estimated parameters and minimized objective functions for ethanol–water mixture in the case of PERVAP™ 4060

PERVAP™ 4060	Model I		Model II	
	Water	EtOH	Water	EtOH
$\bar{D}_i$ [kmol/m²h]	0.015	0.076	0.026	0.077
E_i [kJ/kmol]	31386	33082	31363	33090
B [-]			-0.73	-0.04
OF [-]	0.0038	0.1580	0.0001	0.1610

Table 3 Estimated parameters and minimized objective functions for ethanol–water mixture in the case of PERVAP™ 1060

PERVAP™ 1060	Model I		Model II	
	Water	EtOH	Water	EtOH
$\bar{D}_i$ [kmol/m²h]	0.003	0.045	0.006	0.072
E_i [kJ/kmol]	62806	33283	62801	35892
B [-]			-0.77	-10.60
OF [-]	0.0088	1.1475	0.0020	0.1109

Table 4 Estimated parameters and minimized objective functions for ethanol–water mixture in the case of CELFA-CMG-OG010

CELFA-CMG-OG010	Model I		Model II	
	Water	EtOH	Water	EtOH
$\bar{D}_i$ [kmol/m²h]	0.003	0.024	0.006	0.038
E_i [kJ/kmol]	63017	46412	63019	47534
B [-]			-0.77	-9.14
OF [-]	0.0089	0.9359	0.0020	0.2027

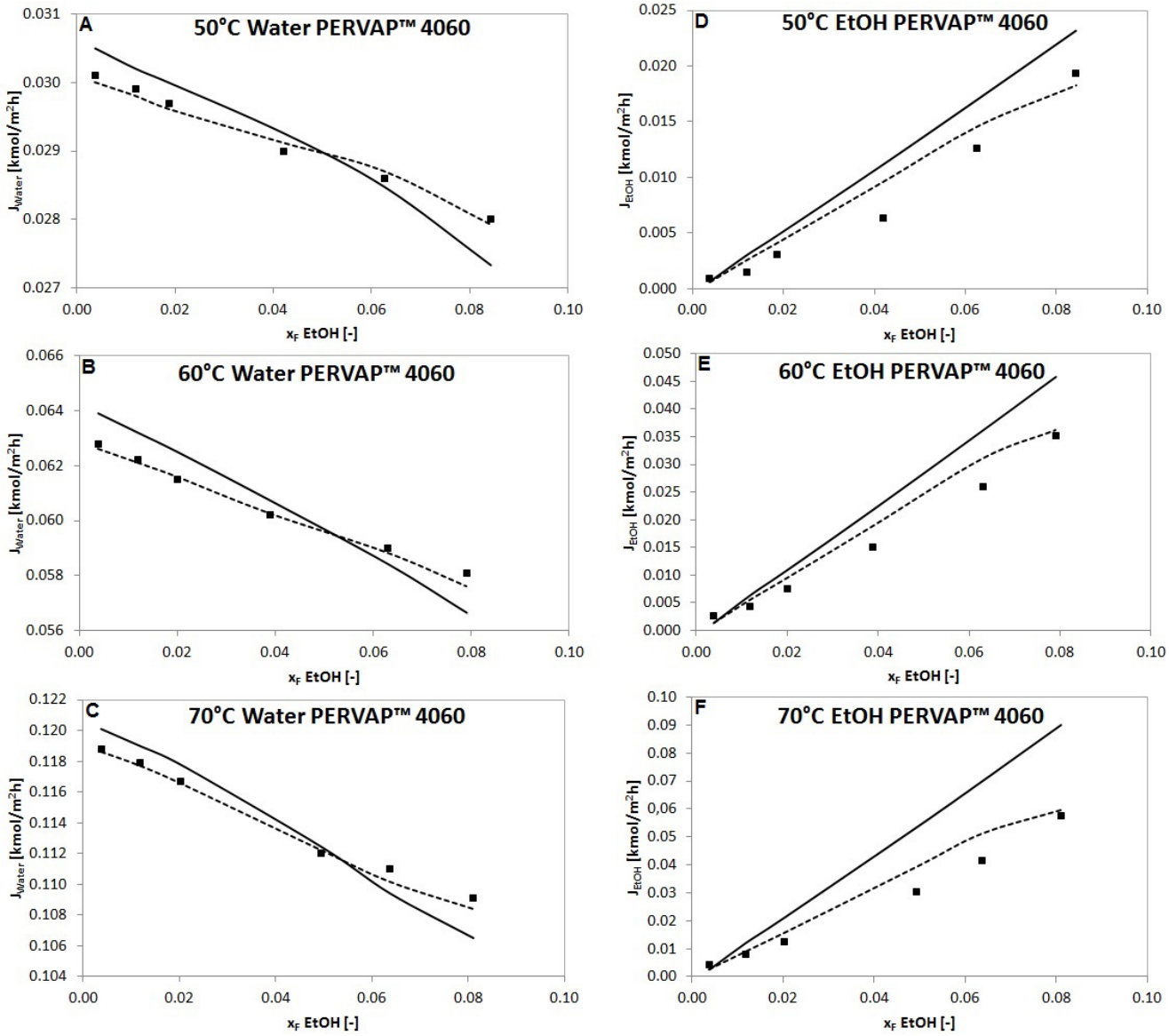

Fig. 4 Measured partial fluxes (■) of water and ethanol compared to fluxes calculated with Model I (——) and Model II (– – –) in a function of feed ethanol content in molar fraction with PERVAP™ 4060 organophilic membrane

suggested an exponential relationship between feed concentration and diffusion coefficient [1, 13, 37, 38] and our investigations can be also confirmed that the dependency of $\bar{D}_i$ between concentration in this organophilic case.

4 Conclusions

The flux of the investigated Sulzer PERVAP 4060 membrane is found to vary from 0.22 to 4.03 kg/(m²h) over the feed ethanol concentration range of 1.0–20.0 m/m% at 50–70. The highest PSI of 19.3 kg/(m²h) and it is measured with flat sheet, benchmarked PDMS membrane. The separation factor is reached between 5.4 and 8.1. The figures show that the tendency of separation factor and selectivity are similar, which is in agreement with former researches.

Semi-empirical model is applied, where parameter estimation from laboratory experiments are required to determine the parameters of the pervaporation model. Thereafter, the verification of the determined parameters is carried out by comparing the modelled and measured data. The results of parameter estimation and modelling of the organophilic pervaporation show that the model of Valentinyi et al. [1] (Model II) is able to the modelling of pervaporation and also results in a better fit to the experimental data.

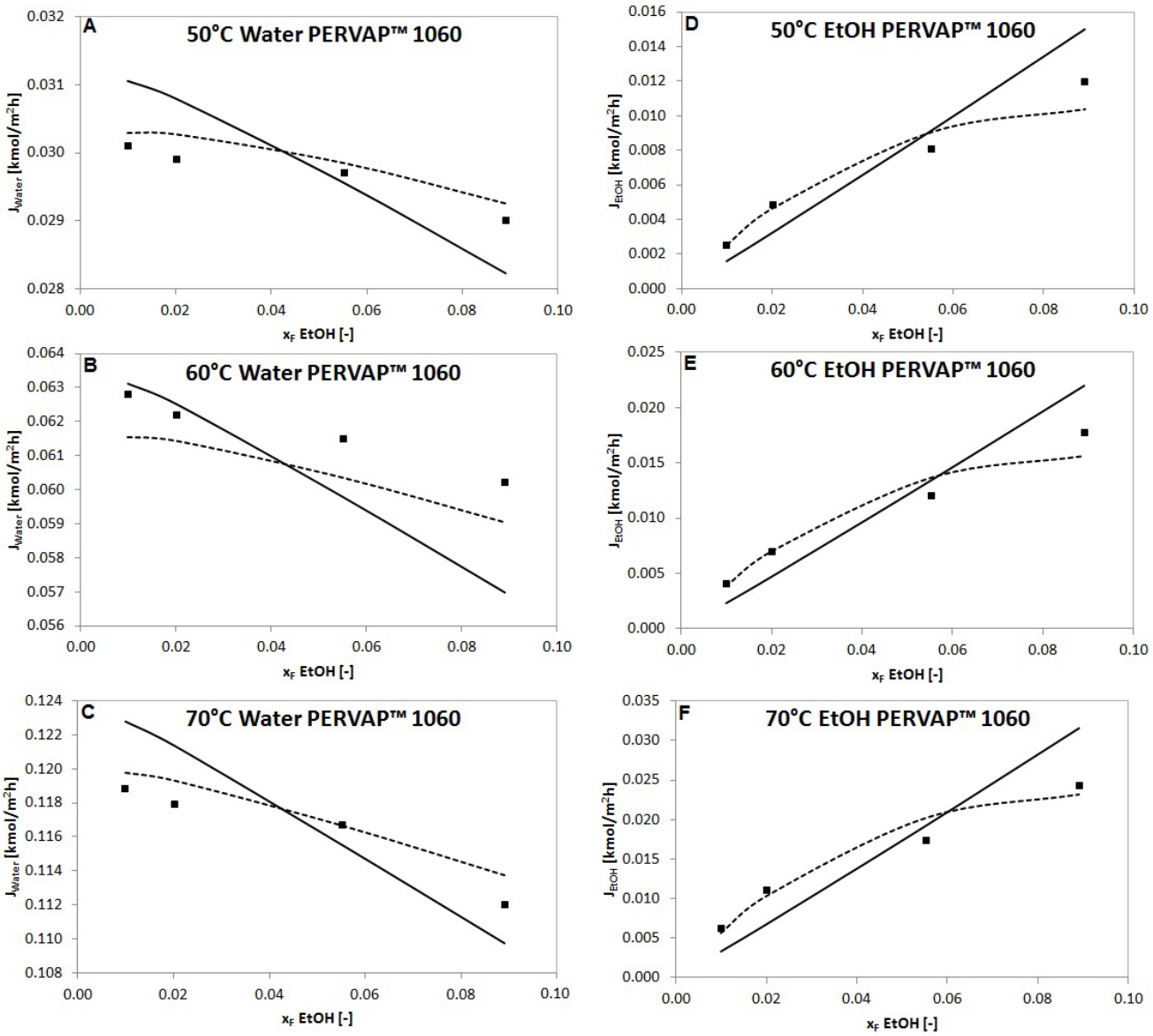

Fig. 5 Measured partial fluxes (■) of water and ethanol compared to fluxes calculated with Model I (——) and Model II (– – –) in a function of feed ethanol content in molar fraction with PERVAP™ 1060 organophilic membrane

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Nomenclature

A	Membrane transfer area [m^2]
B	Constant in Model II [–]
$\bar{D}_i$	Transport coefficient of component i [kmol/(m^{2}h)]
F	Feed
i	Component number
j	Component number
J_{total}	Total flux [kg/(m^{2}h)]
J_i	Partial flux [kg/(m^{2}h)]
P	Permeate
p_{i0}	Pure component vapour pressure [bar]

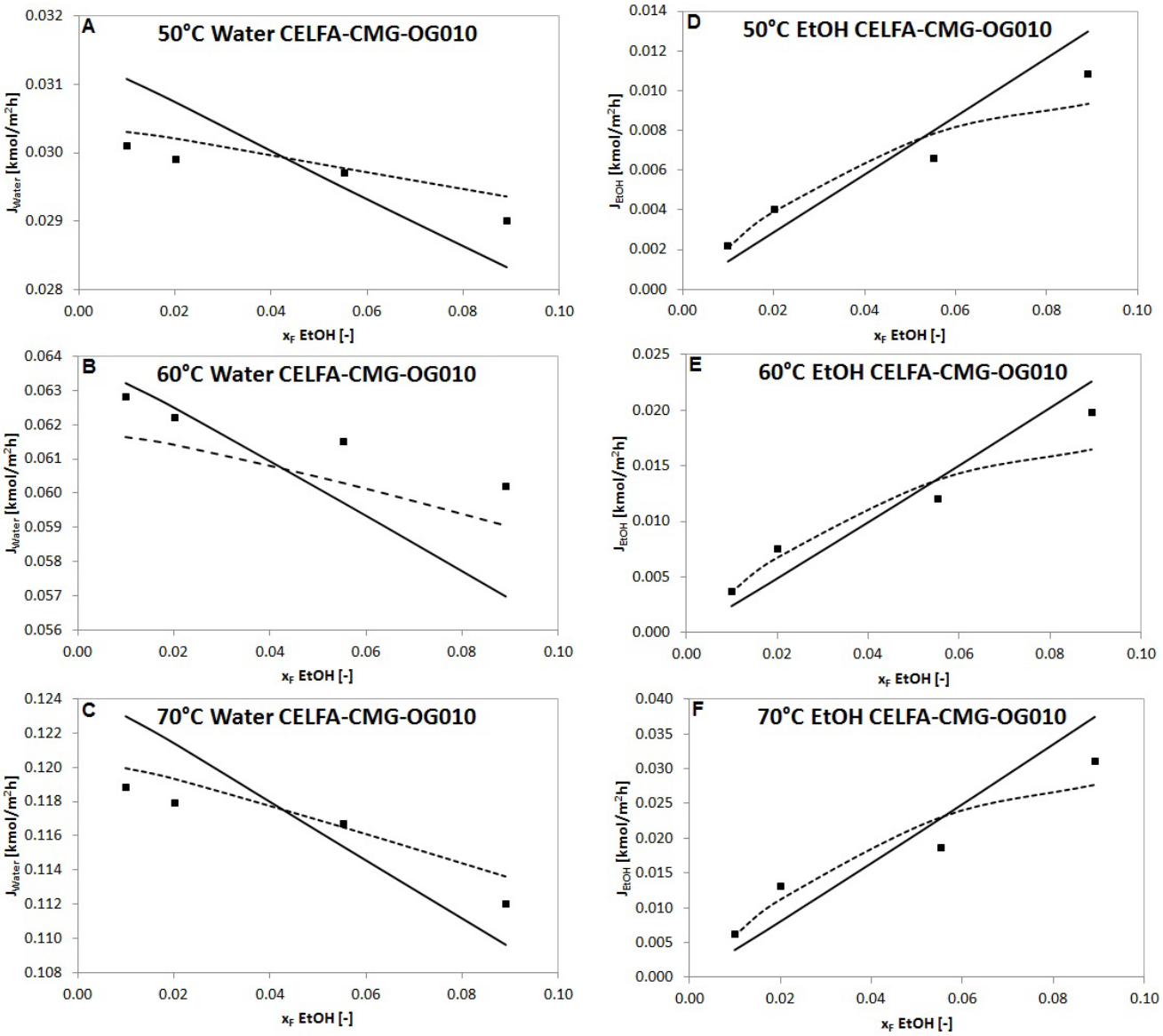

Fig. 6 Measured partial fluxes (■) of water and ethanol compared to fluxes calculated with Model I (——) and Model II (– – –) in a function of feed ethanol content in molar fraction with CELFA-CMG-OG010 organophilic membrane

p_{i1}	Partial pressure of component *i* on the liquid phase membrane side [bar]
p_{i3}	Partial pressure of component *i* on the vapor phase membrane side [bar]
p_3	Pressure on the permeate side [bar]
P_i/δ	Permeance of component *i* [kg/(m²h bar)]
R	Retentate
s	support
t	Time [h]
T	Temperature [°C]
x_{i1}	Concentration of component *i* in the feed [m/m%]

Abbreviations

CA	Cellulose acetate
copol.	copolymer
EtOH	Ethanol
OF	Objective function
org	organophilic
PA	Polyamide
PDMS	Polydimethylsiloxane
PS	Phosphatidylserine
PSI	Pervaporation Separation Index [kg/(m²h)]
PTFE	Polytetrafluoroethylene
PV	Pervaporation
PVDF	Polyvinylidene fluoride

VLE Vapor-Liquid Equilibrium

Greek letters

α Separation factor
β Selectivity
$\overline{\gamma}_i$ Average activity coefficient of component i
γ_{i1} Activity coefficient of component i in the feed
δ Membrane thickness [µm]

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