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Modification of the extended Spiegler–Kedem model for simulation of multiple solute systems in nanofiltration process

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ABSTRACT

In the present work the extended Spiegler–Kedem model has been modified to predict the performance of the multiple solutes systems of nanofiltration with high concentration solutes using osmotic coefficient for calculation of osmotic pressure on the membrane surface in non-ideal solutions. Furthermore, a new method has been proposed for solving the model by simulating the multiple solutes nanofiltration systems using genetic algorithm (GA). The new method is independent of the number of solutes and data points and it is capable to predict the performance of multiple solutes systems with high precision.

Keywords: Extended Spiegler–Kedem; Multiple solutes system; Nanofiltration; Genetic algorithm (GA)

1. Introduction

The transport of the solute through the membranes could be explained by irreversible thermodynamics. In this model, the membrane is considered as a black box and solute and solvent fluxes are proportional to the chemical potential gradient between two membrane sides. Chemical potential gradient, as the only driving force here, is generated by pressure or concentration gradient. Model parameters include salt permeability, P_s, and the reflection coefficient, σ. Kedem and Katchalsky [1] introduced the relation between the volumetric flux, J_v, and the solute flux, J_s, through a membrane by the following equations:

$$J_v = L_p \cdot (\Delta P - \sigma \Delta \pi) \tag{1}$$

$$J_s = P_s \cdot \Delta C + (1 - \sigma) C J_v \tag{2}$$

where σ, P_s and L_p stand for the reflection coefficient, solute permeability and pure water permeability, respectively. Eq. (2) shows that the solute flux is the sum of diffusive and convective terms. Convective transport takes place because of applied pressure gradient across the membrane and the diffusive transport is due to the concentration difference on both sides of the membrane. When high concentration differences exist between the rejection and the permeate, Spiegler and Kedem [2] used the above equations and presented the following equations:

$$R = \sigma \frac{(1 - F)}{(1 - \sigma F)} \tag{3}$$

$$F = \exp\left(- \frac{1 - \sigma}{P_s} J_v \right) \tag{4}$$

where R stands for rejection. The parameters σ and P_s could be determined by the experimental data of rejection (R) as a function of volume flux (J_v) using the best-fit method. Salt rejection is determined by the following equation:

$$R = 1 - \left(\frac{C_p}{C_m} \right) \tag{5}$$

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where, C_p is the permeate concentration and C_m is the concentration at the membrane surface. C_m can be calculated using the concentration polarization equation:

$$\left(\frac{C_m - C_p}{C_b - C_p}\right) = \exp\left(\frac{J_v}{k}\right) \quad (6)$$

In this equation C_b is the bulk concentration and k is the mass transfer coefficient in the boundary layer.

The Spiegler–Kedem model has been extensively used for predicting the transport of single solute and solvent through the membrane in the reverse osmosis and nanofiltration systems [3–5]. Bhattacharya and Ghosh used this model and Fukuda et al. used the Kedem–Katchalsky model to predict the performance of binary solute system when one of the ions is impermeable to the membrane [6]. Ahmad et al. [7] extended the Spiegler–Kedem model for multiple solutes systems by considering solute–solute interactions. In this model, the solution on the membrane surface is assumed to be dilute and osmotic pressure is calculated using Vant Hoff equation. Extended Spiegler–Kedem model by considering concentration polarization (Film theory) is as follows:

$$J_v = \overline{L_p}\left(\frac{dP}{dx} - \sum_{s=1}^{n} \sigma_s \frac{d\pi_s}{dx}\right) \quad (7)$$

$$F_s = \exp\left[\frac{-J_v(1-\sigma_s)}{P_{ss}}\left(1 + \sum_{i=1}^{n} A_i\right)\right] \quad (8)$$

$$A_i = \frac{P_{si}(C_{pi} - C_{bi})\exp\left(\frac{J_v}{k_i}\right)}{J_v\left[C_{ps} - (1-\sigma_s)C_s\right]} \quad (9)$$

$$\frac{R_{os}}{1-R_{os}} = \frac{\sigma_s(1-F_s)}{1-\sigma_s}\exp\frac{-J_v}{k_s} \quad (10)$$

$$R_o = 1 - \left(\frac{C_p}{C_b}\right) \quad (11)$$

Ahmad et al. [7] estimated the parameters of the extended Spiegler–Kedem model using Levenberg–Marquardt method and Gauss–Newton algorithm based on the experimental data for binary and ternary solutes systems.

Based on the extended Spiegler–Kedem model, the objectives of the present work are as follows:

(1) Modification of the extended Spiegler–Kedem model using osmotic coefficient for obtaining the osmotic pressure of the solutes in case of the solute concentration on the membrane surface is more than 1 mol/l, and comparing the new results with the Ahmad's results and experimental data.
(2) Parameters estimation of the extended Spiegler–Kedem model for systems with three solutes using Genetic Algorithm (GA) and comparing the new results with the published results [7], and experimental data.
(3) Parameters estimating of the Extended Spiegler-Kedem model for systems containing 4 and 8 solutes using GA and comparing the new results with the experimental data and simulations available in the literature.

2. Modification of the extended Spiegler–Kedem model

2.1. Calculation of the osmotic pressure

The extended Spiegler–Kedem model should be modified with considering the osmotic factor for non-ideal solutions (in which the concentration on the membrane surface is more than 1 molar), using the following equation:

$$\Delta\pi_m = \varphi_m \kappa (c_m - c_p) R_g T \quad (12)$$

where φ_m stands for osmotic factor for non-ideal solutions and is calculated based on solute concentration on the membrane surface and κ is the total number of constituent ions in the salt [8]. Substituting Eq. (12) in Eq. (7) and integrating over the membrane thickness, permeate flux in new model is calculated by Eq. (13)

$$J_v = L_p\left[\Delta P - \sum_{s=1}^{n} \varphi_m \kappa \sigma_s a_s R_s C_{ms}\right] \quad (13)$$

where $a = R_g T/M$ is defined as the osmotic constant.

2.2. Calculation of osmotic coefficient

There are various methods for calculation of the osmotic coefficient. One of the most well-known models is the Pitzer model. The Pitzer model [9] is used to calculate the thermodynamic properties of mixed electrolyte solutions. This model requires parameters estimated from common-ion solutions in order to characterize binary interactions between different ions of the same sign and ternary interactions between different ions with equal or unequal sign in mixed electrolyte solutions. The parameters of this model have no clear physical significance and vary with changing temperature. Lin and Lee [10] proposed a three-characteristic-parameter correlation (TCPC) model attributing ion–solvent molecule interaction to the solvation effect, but the parameters provided could

only be applied in the concentration below 6 mol/kg. The mean spherical approximation model (MSA) acts as a powerful tool for calculating the thermodynamic properties of electrolytes [11], but the calculations made in this model are very complicated.

The model used here is a three parameter one, proposed by Ge et al. [12]. The parameters are ion–ion distance parameter, ion–solvent parameter, and solvation parameter. In this model, the ion–ion and ion–solvent molecule interaction are independent of the temperature and solvent. This new model was applied to correlate the experimental data from literatures for 208 electrolytes aqueous solution at T = 298.15 K in a wide range of concentration. Another advantage of this model is the clear physical meaning of these three parameters. In this model osmotic coefficient for aqueous solutions is calculated as follows:

$$\varphi = 1 - \frac{A\left|z_+ z_-\right|}{B^2 T^{1/2} a^2} \cdot \left[\frac{2}{I^{1/2}} - \frac{1}{(Ba)^{-1}T^{1/2} + I^{1/2}} - \frac{2\ln(1 + BaT^{-1/2}I^{1/2})}{BaT^{-1/2}I}\right] + \frac{S}{T} \cdot \frac{2n'}{2n' + 1} \cdot \frac{I^{2n'}}{v'_+ + v'_-} \quad (14)$$

φ_m stands for osmotic factor, A = 6064.613 $kg^{1/2}$ $mol^{-1/2}$ $K^{3/2}$, B = 56.827 $kg^{1/2}mol^{-1/2}$ $K^{1/2}$, z is the ion charge and, v' is stoichiometric number of solute as:

$$v'_+ z_+ = \left|v'_- z_-\right| \quad (15)$$

T, a, n' and S stand for temperature (K), the ion–ion distance parameter, the ion–solvent parameter and the salvation parameter, respectively. I is the ionic strength and can be calculated as:

$$I = \frac{1}{2}\sum_i m_i \cdot z_i^2 \quad (16)$$

where m is molality in mol/kg. According to the parameters a, S and n', it is possible to calculate the osmotic coefficients of electrolytes in aqueous solution.

3. New method for solving the extended Spiegler–Kedem model equations using GA

There are n solutes in solvent in the multiple solute nanofiltration systems. Concentration of solutes in feed (C_b) and experimental data of observed rejection (R_o) for each solute versus flux (J_v) are taken at different ΔP_s and constant feed rate, and concentrations for each set of experiment, in the steady state are obtained. The hydraulic permeability coefficient (L_p) can be determined using the linear relationship of J_v and ΔP with L_p which is the slope of the plot of pure water flux (J_v) versus ΔP.

In this part, the objective is the calculation of the parameters σ_s, k_s, P_{ss}, P_{si}. The term P_{ss} is the solute permeability coefficient of solute s with the consideration of the interaction of solute s and the P_{si} is the solute permeability coefficient of solute s with the consideration of the interaction of solute i, σ_s is reflection coefficient and k_s is the mass transfer coefficient.

Do the steps 1–5 for each solute:

1. Obtain C_p versus R_o from Eq. (11).
2. Substitute the above C_ps in Eq. (9).
3. Substitute the product from step 2 in Eq. (8).
4. Substitute the product from step 3 in Eq. (10).

In these four steps we will obtain n main equations.

5. Substitute the experimental points (J_v, R_o) in n main equations (from step 4)
6. Solve the equations with genetic algorithm simultaneously (for this purpose, we used multi objective optimization method in genetic algorithm by using Matlab 7.6.1 (R2008a) the software package).

The product from step 6 is numerical values for the parameters (P_{ss}, P_{si}, k_s, σ_s).

7. Substitute the parameters in the n main equations of step 4.
8. Solve the n above equations simultaneously and obtain the rejection of each solute for a given flux (for this purpose, we used nonlinear least squares method with Levenberg–Marquardt algorithm by using Matlab 7.6.1 (R2008a) the software package).

3.1. Genetic algorithm

Genetic algorithms (GAs) are search algorithms which are a model of machine learning that derive their behavior from a metaphor of processes of evolution in nature. GAs use optimization strategies inspired by Darwin's theory of evolution and have direct application in mathematical optimization to find the global minimum or maximum in a search space. An initial population of individuals is generated randomly, and newer individuals are created iteratively until acceptable solutions are found. Each iteration is called a generation. The main advantages of the GAs are their robustness and their ability to provide a balance between efficiency and effectiveness in different environments which cover a variety of applications.

4. Results and discussion

4.1. System with two solutes

The experimental data of Wadley et al. [13] were obtained on the laboratory scale using the MPT-31 nanofiltration membrane at 35 °C and in the range of pressures (2.0–3.5 MPa) to separate the sodium chloride with the initial feed concentration of 81.20 kg/m^3 from the organic matters of sugarcane in the waste water stream.

The molecular weight of organic substances was in 5000–20,000 g/mol range. The concentration of the organic substances in the feed was 3.20 kg/m^3. Simulation results of system using extended Spiegler–Kedem model are shown in Figs. 1–3 and the results of new model (this work) are shown in Figs. 4–6.

Sodium chloride shows negative rejection in the presence of organic substances. The mechanism of this effect is explained by Donnan effect that is caused due to the negative charge on the surface of the membrane [14–19]. As it is shown in Figs. 4 and 5, the new model can show the negative rejection of NaCl in the presence of organic substances. The concentration of NaCl in this system is about 1.4 mol/kg, and it will be more than this value on the membrane surface. In addition, the con centration of the solutes on the membrane surface increases with increasing the permeate flux. Under these conditions, there is error due to using Vant Hoff equation for calculation of osmotic pressure (Fig. 3)

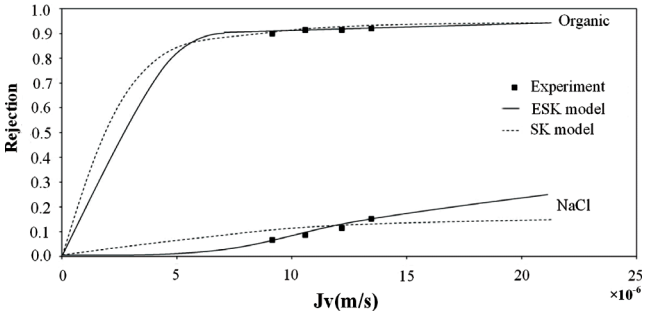

Fig. 1. Solutes rejection of binary solutes system plotted against volumetric flux using the experimental data and simulation results from the extended Spiegler–Kedem model [7].

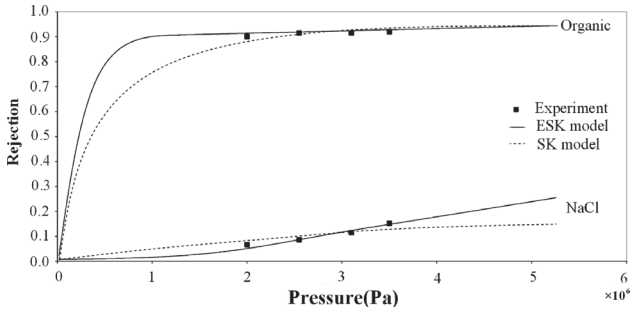

Fig. 2. Solutes rejection of binary solutes system plotted against pressure using the experimental data and simulation results from the extended Spiegler–Kedem model [7].

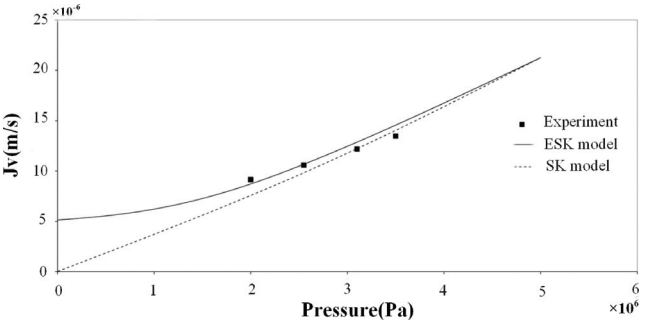

Fig. 3. Volumetric flux of binary solutes system of NaCl and organic matters of sugarcane plotted against pressure using the experimental data [13] and simulation results from the extended Spiegler–Kedem model [7].

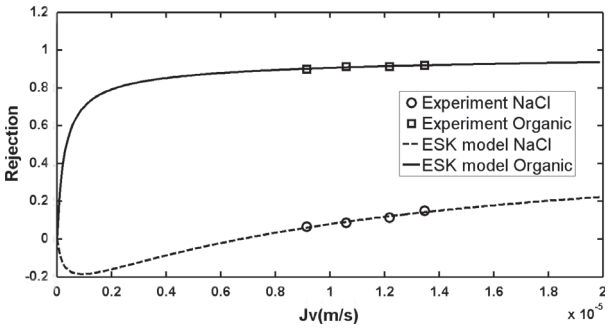

Fig. 4. Solutes rejection of binary solutes system plotted against volumetric flux from the simulation results of the extended Spiegler–Kedem model (new method).

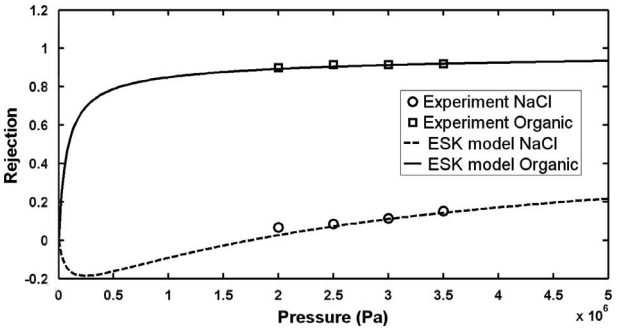

Fig. 5. Solutes rejection of binary solutes system plotted against pressure using the experimental data and simulation results from the extended Spiegler–Kedem model by incorporating osmotic coefficient (new method).

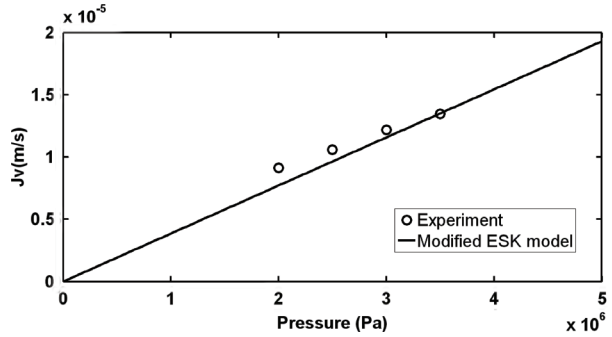

Fig. 6. Volumetric flux of binary solutes system of NaCl and organic matters of sugarcane plotted against pressure using the experimental data [13] and simulation results from the extended Spiegler–Kedem model by incorporating osmotic coefficient (new method).

(this error can be seen in nonlinear relationship between flux and pressure in Fig. 3 which is in the approximate range of 1–5 m/s). Fig. 6 shows that using the new method, this error will be decreased substantially (the relationship between flux and pressure is linear for the commercial membranes). Regarding to this point, and comparison of Fig. 3 with Fig. 6, it is evident that the new model compared with Ahmad et al. [7] model has better prediction of the behavior of nanofiltration systems with high solute concentrations.

4.2. System with three solutes

The experimental data of ternary solute system was taken from the published data of a filtration system using a negatively charged Nanomax 50 membrane for nanofiltration to separate mixed electrolyte solutions from the water [20]. The mixed electrolyte solutions studied were copper chloride and sodium chloride ($CuCl_2$, NaCl) mixture.

Filtration experiments were performed with a Millipore laboratory tangential filtration system equipped with a spiral-wound polymeric membrane (Nanomax 50, Millipore USA), having a filtration area of 0.37 m^2 and a pure water permeability of 22.7×10^{-2} m s^{-1} pa^{-1}. The Nanomax 50 is a composite membrane having a negatively charged thin skin layer made of polyamide arylene on a polysulfone support layer.

Experiments were performed for 1 h in batch circulation mode. Both permeate and retentate were returned to the feed vessel in order to keep constant concentration.The temperature of the recirculating feed solution was maintained at 20×0.5 °C. Copper salt were $CuC1_2 \cdot 2H_20$ and NaCl. Solution was prepared in demineralized water (pH = 5.70).

The mixtures were assumed completely ionized to form individual ions and thus there is a ternary solutes system. The mixture contains solutes of Cu^{2+}, Cl^- and Na^+ with the feed concentrations $[Cu^{2+}] = 0.0318$ kg/m^3, $[Cl^-] = 0.2127$ kg/m^3, $[Na^+] = 0.1150$ kg/m^3.

The results of the performance of the system using extended Spiegler–Kedem model with Levenberg–Marquardt method of solving the equations are shown in Fig. 7 and the results of new method (GA) are shown in Figs. 8–10. The parameters of extended Spiegler–Kedem model for ($CuCl_2$, NaCl) mixture, estimated

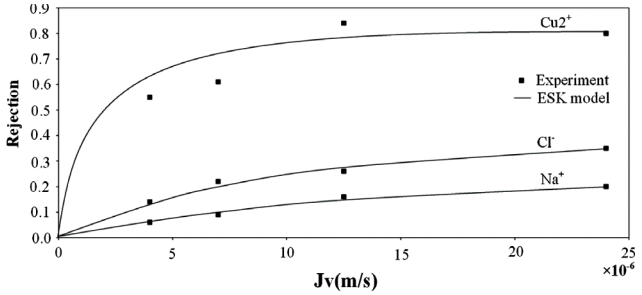

Fig. 7. Ions rejection of $CuCl_2$, NaCl system versus volumetric flux from the simulation results of the extended Spiegler-Kedem model [7].

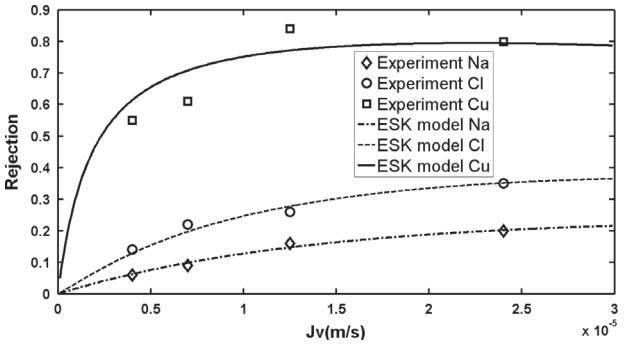

Fig. 8. Ions rejection of $CuCl_2$, NaCl system versus volumetric flux from the simulation results of the extended Spiegler-Kedem model (new method).

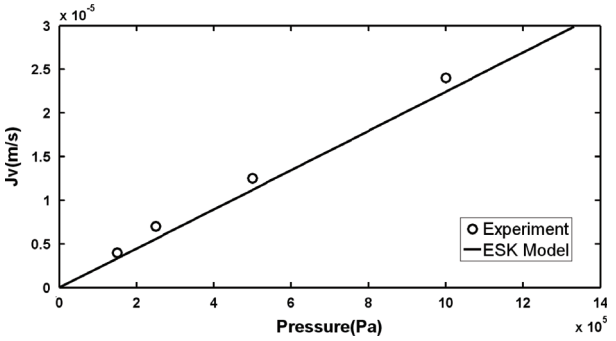

Fig. 9. Volumetric flux of $CuCl_2$, NaCl system plotted against pressure using the experimental data (circles) and simulation results from the extended Spiegler–Kedem new model (line).

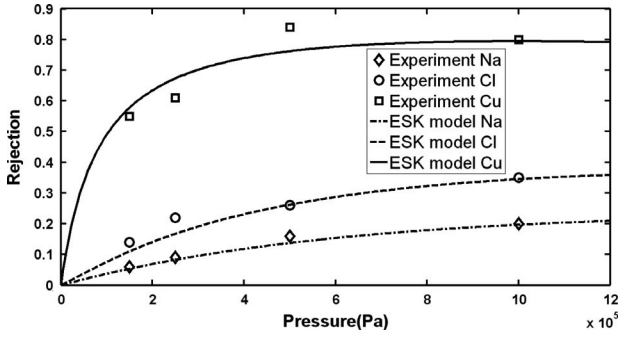

Fig. 10. Solutes rejection of $CuCl_2$, NaCl system plotted against pressure using the experimental data and simulation results from the extended Spiegler–Kedem model (new method).

using both the Levenberg–Marquardt method and new method, are listed in Table 1.

4.3. System with four solutes

The existing experimental data as well as NanoFlux simulations [21] are given in Fig. 11. Published experimental data and extended Spiegler–Kedem model with GA method simulations for the NF200 membrane for 1/1/1 ternary mixtures of $NaNO_3/NaCl/CaC1_2$ at total concentration of feed = 0.015 kg/m^3 are presented in Fig. 12. In this system there is Na^+ with the concentration of 0.69 kg/m^3, NO_3^- with the concentration of 0.93 kg/m^3, Cl^- with the concentration of 1.5957 kg/m^3 and Ca^{2+} with the concentration of 0.6 kg/m^3. The parameters of extended Spiegler–Kedem model for $NaNO_3/NaCl/CaC1_2$ mixture estimated using extended Spiegler–Kedem model with GA method are listed in Table 2.

4.4. System with eight solutes

Here the nanofiltration system was NF200 membrane which consisted of an aqueous solution of several electrolytes and lactose. In this system it has been reported that whey was ultrafiltered using a polyethersulfone membrane with a molecular weight cut-off of 10,000 Da to avoid fouling problems in the nanofiltration process [22,23]. The published results of the analytical characterization of UF–whey are shown in Table 3 (it was free of fat, and protein concentration was low).

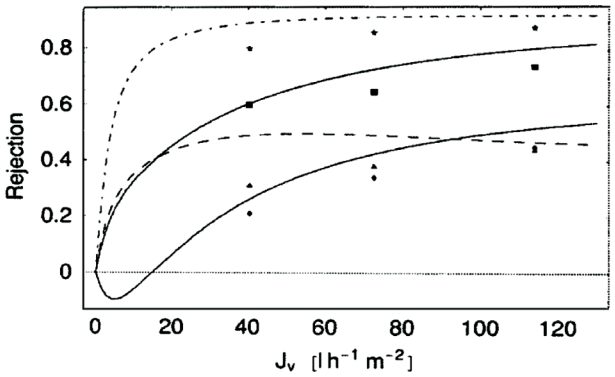

Fig. 11. Ions rejection of $NaNO_3/NaCl/CaC1_2$ system versus volumetric flux. Experimental data and Nanoflux simulations [21]. Nanoflux predictions (data): dot-dashed curve (stars), Ca^{2+}; upper solid curve (squares), Cl^-; dashed curve (triangles), NO_3^-; lower solid curve (diamonds), Na+.

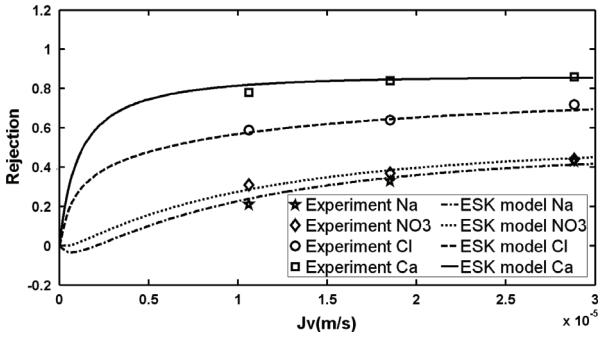

Fig. 12. Ions rejection of $NaNO_3/NaCl/CaC1_2$ system versus volumetric flux. Experimental data and extended Spiegler–Kedem model using GA method simulations.

Table 1
Parameters estimated using Levenberg–Marquardt method and new method (GA) for ternary solutes system

Method	Ahmad's method			New method		
Parameter/s	Na^+(1)	Cl^-(2)	Cu^{2+}(3)	Na^+(1)	Cl^-(2)	Cu^{2+}(3)
σ_s	0.9936	0.6501	0.9951	0.9927	0.6524	0.9977
k_s	4.12e–5	6.60e–5	2.50e–5	4.27e–5	6.32e–5	2.25e–5
P_{s1}	5.24e–5	9.59e–8	1.93e–9	5.27e–5	9.59e–8	2.10e–9
P_{s2}	4.62e–8	1.35e–5	1.52e–8	4.88e–8	1.31e–5	1.78e–8
P_{s3}	3.72e–7	3.38e–6	1.97e–6	3.92e–7	3.52e–6	2.07e–6
R^2 (coefficient of determination)	0.9787	0.9515	0.6307	0.9794	0.9577	0.6943

Table 2
Parameters estimated from Extended Spiegler-Kedem model using new method (GA) for system with 4 solutes

Parameter/s	Na^+(1)	NO3-(2)	Cl^-(3)	Ca^{2+}(4)
σ_s	0.9886	0.7499	0.80877	0.8597
k_s	4.12e–05	7.26e–5	4.44e–3	3.62e–3
P_{s1}	1.54e–5	9.22e–8	8.77e–8	4.19e–8
P_{s2}	1.17e–7	1.20e–5	1.02e–5	6.81e–8
P_{s3}	1.96e–6	1.21e–6	1.79e–6	5.60e–8
P_{s4}	7.51e–7	6.29e–8	6.63e–8	9.56e–7
R^2 (coefficient of determination)	0.9423	0.9030	0.8779	0.5374

Table 3
Analytical characterization of the UF-whey [23]

Solute	Concentration in UF-whey (NF-Feed) kg/m^3
Cl^-	1.29
Na^+	0.37
K^+	1.48
PO_4^{3-}	0.6
Mg^{2+}	0.072
SO_4^{2-}	0.093
lactose	41.9
Ca^{2+}	0.289

Membrane experiments were performed in a pilot plant equipped with both flat and spiral wound membrane modules.

The transmembrane pressure was varied between 0.5 and 2.5 MPa from low to high pressure and the temperature was set constant to 20 °C. The pH of the feed solutions was 6.7 and the crossflow velocity was 1.22 and 0.7 m/s, respectively.

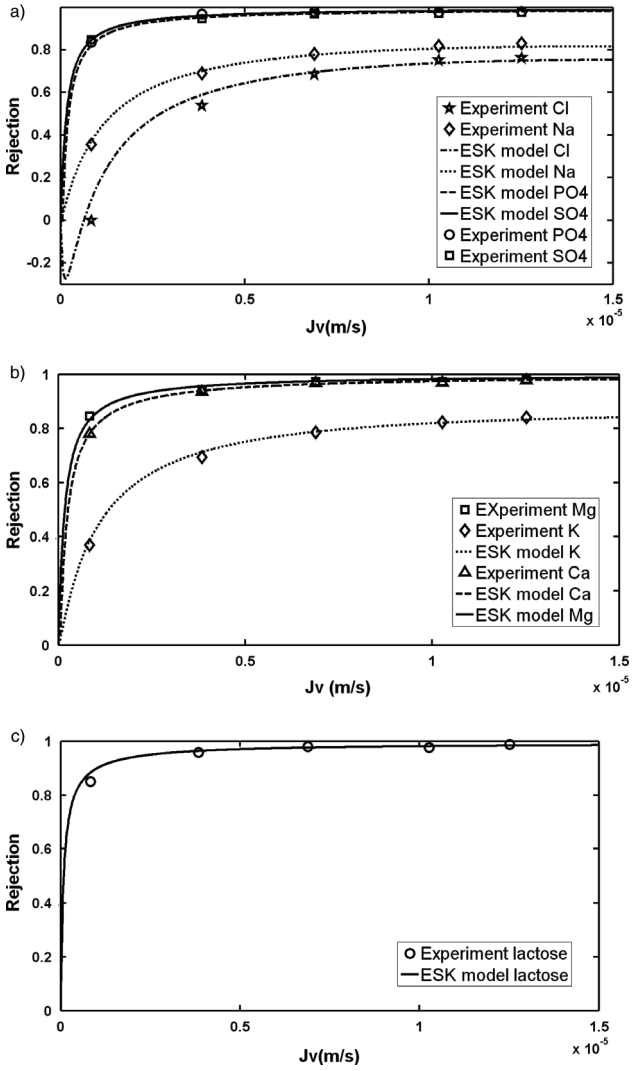

Fig. 13. Ions rejection of UF–whey system versus volumetric flux. Experimental results and extended Spiegler–Kedem model by using GA method simulations. (a) Cl^-, Na^+, PO_4^{3-}, SO_4^{2-}; (b) Mg^{2+}, K^+, Ca^{2+}; (c) lactose.

Table 4
Parameters estimated from extended Spiegler–Kedem model by using new method (GA) for system with eight solutes

Parameter/s	Cl^- (1)	Na^+ (2)	K^+ (3)	PO_4^{3-} (4)	Mg^+ (5)	SO_4^{2-} (6)	Lactose (7)	Ca^{2+} (8)
σ	0.8099	0.9994	0.85978	0.9908	0.9990	0.9950	0.9900	1.0000
k_s	5.81e–5	1.51e–5	9.90e–4	9.00e–4	9.00e–4	9.50e–4	8.51e–4	9.50e–4
P_{s1}	6.86e–7	9.04e–11	2.01e–9	2.00e–9	3.00 e–10	5.00e–11	1.01e–6	8.01e–9
P_{s2}	1.01e–7	1.20e–6	2.01e–9	2.00e–9	8.91e–10	5.01e–11	2.00e–9	9.00e–9
P_{s3}	1.26e–8	1.00e–9	9.55e–7	2.01e–9	7.61e–10	5.01e–11	2.00e–9	2.00e–9
P_{s4}	1.27e–8	1.01e–9	2.01e–9	2.07e–8	5.51e–10	5.00e–11	2.01e–9	2.01e–9
P_{s5}	3.58e–6	1.06e–10	2.00e–9	2.01e–9	9.00e–8	9.00e–10	2.00e–9	2.01e–9
P_{s6}	1.40e–8	1.03e–10	2.01 e–9	2.01e–9	6.90e–10	7.01e–8	4.01e–7	2.01e–9
P_{s7}	1.20e–8	5.06e–10	2.01e–9	2.01e–9	1.01e–10	1.51e–10	9.99e–8	1.00e–9
P_{s8}	1.29e–8	5.01e–10	2.01e–9	2.00e–9	3.01e–10	2.51e–9	2.01e–8	5.06e–8
R^2	0.9711	0.9950	0.9958	0.9702	0.9603	0.9635	0.9089	0.9977

The results of the performance of system using extended Spiegler–Kedem model using new method simulations are shown in Fig. 13a–c. Simulation of UF–whey system with extended Spiegler–Kedem model using new method consists of 80 parameters. The numerical values of the parameters are listed in Table 4.

5. Conclusions

Comparisons made between the methods for solving the equations of extended Spiegler–Kedem model (Levenberg–Marquardt and GA methods), showed that in GA method, the value of any number of parameters can be estimated, regardless of the number of solutes and experimental data points, with higher precision than Levenberg–Marquardt method. Levenberg–Marquardt method is dependent on the number of solutes and data points. In addition, modified method of osmotic pressure is suitable for predicting the performance of high concentration systems. The extended Spiegler–Kedem model, which its method for calculation of osmotic pressure was modified in this study, is suitable for predicting the performance of the systems with high (and low) concentration solutes. Moreover, using GA method of solution, it will be suitable for systems with any number of solutes and experimental data points with wide range of concentrations.

Symbols

A	—	Osmotic constant (m^3 Pa/g)
C	—	Concentration (kg/m^3)
F	—	Driving forces (kWs/m mol)
J_s	—	Flux of solute (kg/m^2 s)
J_v	—	Total volumetric flux (m^3/m^2 s)
J_w	—	Flux of solvent (m^3/m^2 s)
K	—	mass transfer coefficient (m/s)
$\overline{L}_p$	—	Specific hydraulic permeability constant (m/Pa m s)
L_p	—	Hydraulic permeability constant (m/Pa s)
m	—	Molality (mol/kg)
M	—	Solute molar mass (g/mol)
P	—	Pressure (Pa)
ΔP	—	Transmembrane pressure (Pa)
P_{si}, P_{ss}	—	Solute permeability constant (m/s)
R	—	True rejection
R_g	—	Ideal gas constant (8.314 m^3 Pa/mol K)
R_o	—	Observe rejection
T	—	Operating temperature (K)
X	—	Coordinate vertical to the membrane surface

Greeks

σ	—	Reflection coefficient
φ	—	Smotic coefficient
π	—	Osmotic pressure (Pa)
μ	—	Chemical potential (J/mol)

Subscripts:

b	—	Bulk
m	—	Membrane wall
p	—	Permeate
s,	—	*i* solute
w	—	Water

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