



Modeling of suspension fouling in nanofiltration



F. Faridirad^a, Z. Zourmand^a, N. Kasiri^{a,*}, M. Kazemi Moghaddam^b, T. Mohammadi^b

^a Computer Aided Process Engineering (CAPE) Lab, School of Chemical Engineering, Iran University of Science and Technology (IUST), Narmak, Tehran 1684613114, Iran

^b Research Centre for Membrane Separation Processes, School of Chemical Engineering, Iran University of Science and Technology (IUST), Narmak, Tehran 1684613114, Iran

HIGHLIGHTS

- Dissolution and sedimentation of solute particles during NF were considered.
- Considering the fouling of solutes during NF of suspensions
- Derive a new mathematical model which calculates permeate flux
- Verifying the new model with experimental data
- Comparing the performance of the new model with other existing models

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ABSTRACT

Prediction of membrane process performance using experimental and mathematical models facilitates optimization of membrane processes and their operating conditions. On the other hand investigation of nanofiltration process performance over time and estimation of the time for recovery are very essential in the membrane industry. In this work a mathematical model considering membrane resistance changes and also dissolution of deposited particles in nanofiltration feed, for a cross flow nanofiltration system was developed. The validation of this model with experimental data demonstrated a good agreement. At a constant concentration of 0.2 g/L error of the present models varied from 3% (at the pressure of 1 bar) up to about 3.8% (at the pressure of 2.9 bar) while at a constant pressure of 2 bar error varied from 2.45% (at the concentration of 0.1 g/L) up to about 4.2% (at the concentration of 0.4 g/L). A comparison between this model and three other previous models proved a better performance for the present model with an average error of 3.25%.

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1. Introduction

Nanofiltration is a membrane process for liquid separation which has been extended recently [1]. Reverse osmosis is replaced by nanofiltration in many applications, due to the fact that NF requires less energy comparatively. Going by its specifications, nanofiltration lays between the ultrafiltration membrane and RO. Membrane filtration processes, such as nanofiltration (NF) and reverse osmosis, play an important role in the production of high quality reclaimed water when small organic compounds, (e.g., pesticides, endocrine disruptors and pharmaceutically active compounds) are to be removed from polluted water [2]. The important application of NF is for water treatment, waste water treatment and desalination. NF is a pressure driven separation process which can separate particles based on their size and electrostatic interactions between them. For the uncharged molecules, the dominant mechanism for separation is molecular size while for the separation of ions with similar size, electrostatic forces

play a more important role [3]. NF membranes have pores of around 1 nm [4], therefore they got higher water flux at lower pressures compared to RO. The main problem in using NF technology is capacity reduction of industrial units caused by membrane fouling, particularly during inorganic separation [5,6], leading to membrane life reduction [7]. Inorganic fouling induced by concentration polarization and scale formation is one of the major limitations of NF as in other membrane processes used in water treatment [6,8]. This necessitates fouling control in membrane processing [9]. Suspension fouling is of particular interest in the membrane field. Suspensions are defined as fine particles whose characteristic size is roughly in the range of 1 nm–1 μm [10]. In these processes flux decline is dependent on many factors such as membrane characteristics & module geometry, feed condition, solute type and also operating conditions [11]. Concentration polarization refers to the reversible accumulation of solute within a thin boundary layer adjacent to the membrane surface. Membrane fouling can be irreversible with solute adsorption on or in the membrane pore walls, leading to complete or partial pore blocking [12]. Formation of cake layer over the membrane surface can determine rejection properties of the system since the deposited layer will act as a “secondary”

* Corresponding author.

E-mail address: capepub@cape.iust.ac.ir (N. Kasiri).

membrane prior to the “real” membrane or support material [13]. Fouling importance has initiated many efforts for modeling flux decline profile during NF [14,15]. To improve fouling models many dynamic models have been derived [4,11,16]. Theoretical models are not capable of predicting the filtration flux precisely, due to the simplifying assumptions required to enable their analytical solution [17]. Various flux decline mechanisms are studied which are generally governed by cake or Gel layer [18–20], resistance in series and pore blocking [21,22]. Ballet et al. studied experimentally the effect of feed pressure, ionic strength, concentration, and pH on the retention of phosphate anions [23]. Li et al. investigated the effects of pressure, flow rate on flux, and retention in seawater desalination process [24]. Darvishmanesh et al. developed a new semi-empirical model based on the traditional solution diffusion with imperfection model for solvent resistant NF [25]. This model demonstrated a good prediction for the flux of solvent through the membrane. Fadaei et al. developed a mass transfer model to predict ion transport through the NF membrane to account for the concentration polarization phenomenon and its influence on ion separation [26]. They used CFD techniques and successfully predicted the local concentration of ions; permeate flux, and rejection of ions in a rectangular cross flow. In the field of CFD many other works have been done such as modeling the impacts of feed spacer geometry on NF processes by Guillen and Hoek [27] and CFD modeling of porous membranes by Pak et.al [28]. Field et al. introduced the concept of critical flux for microfiltration, stating that there is a permeate flux below which fouling is not observed. It was a very powerful optimization tool for this kind of separation operation. It was possible then to identify a critical flux for UF and NF membranes [29].

The purpose of this research is to introduce a new fouling model based on cake formation to describe the flux decline caused by inorganic solutes. Fouling based models are appropriate for predicting the filtration and clean up time duration. In model development the effect of many parameters such as operating conditions, cake filtration characteristics, membrane permeability, cross flow velocity, specific cake resistance and other experimental parameters on flux reduction has been investigated. The developed model was then validated against experimental data obtained from water–oil solution NF. Finally model performance was compared with three other models developed by Lihan Huang [30] and Wu [31], as well as the cake or Gel Layer Filtration model [32]. In these models the fact of dissolution of solutes, transmission of particles through the membrane, membrane resistance changes, and also the effect of operating parameters such as cross flow velocity and temperature were neglected. Considering the solute dissolution, membrane resistance changes and also the effect of operating conditions lead to a model with better performance and better agreement with experimental data.

2. Model development

In the NF process feed enters from one side of the cell, having passed over the membrane surface, leaves the cell from the other side with permeate passing through the membrane and exiting from a second exit of the chamber. Cross flow arrangement reduces solute particle

accumulation on the membrane leading to reduced cake formation and fouling [33]. Membrane fouling is related to cross flow nanofiltration velocity with larger flow velocities causing more turbulence hence less fouling [34].

One of the theories for explaining membrane fouling is cake filtration model which was developed by Hoek et al. to evaluate reverse osmosis and nanofiltration processes [35,36]. According to this model flux can be explained as:

$$J(t) = \frac{\Delta p}{R_m + R_{cp} + R_c(t)} \tag{1}$$

In which $J(t)$ is solvent (permeate) flux which is a function of effective pressure applied on the membrane surface (Δp), hydraulic membrane resistance (R_m) and cake layer resistance (R_c) and also the resistance related to concentration polarization layer (R_{cp}). Although concentration polarization layer effect is significant at the very early stages of the separation process, this gradually fades away when pore blockage takes over and due to which concentration polarization has been neglected in the model. Eq. (1) is therefore simplified to:

$$J(t) = \frac{\Delta p}{R_m + R_c(t)} \tag{2}$$

Cake layer resistance is calculated through Eq. (3) using specific cake resistance (α).

$$R_c = \alpha M_d \tag{3}$$

In Eq. (3), M_d is accumulated mass per surface unit. The constant α is related to membrane porosity through Carman–Kozeny's equation [36]:

$$\alpha = \frac{45\mu(1-\varepsilon)^2}{\rho a_p^2 \varepsilon^3} \tag{4}$$

Here μ is water (solvent) viscosity, ε is cake porosity, ρ is cake density and a_p is particle radius. If temperature is fixed, viscosity would remain constant. Despite this, Arrhenius equation can be used to take into consideration the effect of temperature on permeate flux.

$$\mu = \mu_0 \times \exp\left(-\frac{E}{RT}\right) \tag{5}$$

Pressure difference is calculated through Eq. (6) by using pure water permeate.

$$\Delta p = J_0 R_m \tag{6}$$

In this model membrane fouling is modeled as a dynamic process. The model is based on the following assumptions:

- 1- Due to the small membrane length, a uniform cake layer of a uniform resistance will be formed through the membrane surface.

Table 1
Estimated parameters of the developed model, obtained from GA, using 70% of experimental data.

	Pressure	ρ	α	b	a	M_1	M_2	M_3
$c = 0.2 \text{ g/L}$	$P = 1 \text{ bar}$	2300	10^{11}	0.00027	3.42×10^9	0.03	0.00004	1000
	$P = 2 \text{ bar}$	2400	10^{12}			0.07	0.00002	1100
	$P = 2.4 \text{ bar}$	2450	1.4×10^{12}			0.1	0.00001	1180
	$P = 2.9 \text{ bar}$	2500	10^{13}			0.1	0.000001	1200
	Concentration	ρ	α	b	a	M_1	M_2	M_3
$P = 2 \text{ bar}$	$c = 0.1 \text{ g/L}$	2350	10^{11}	0.000087	3.34×10^9	1000	0.000006	0.06
	$c = 0.2 \text{ g/L}$	2400	1.4×10^{11}			1070	0.00003	0.06
	$c = 0.3 \text{ g/L}$	2400	1.6×10^{11}			1100	0.00003	0.05
	$c = 0.4 \text{ g/L}$	2450	1.8×10^{11}			1130	0.00002	0.4

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