

Permeate flux prediction

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In any membrane filtration, the prediction of permeate flux is critical to calculate the membrane surface required, which is an essential parameter for scaling-up, equipment sizing, and cost determination. Permeate flux prediction is an essential parameter in membrane performance evaluation and the projections for scaling-up from laboratory to the pilot plant or the industrial scale.

ultrafiltration

phenomenological models

non-phenomenological models

permeate flux prediction

1. Introduction

Membrane processes have become major techniques in the food industry over the last few decades, thanks to their ability to provide gentle treatment of products at low-to-moderate temperatures.

Membrane applications in the food industry have focused on separation, fractionation, purification, clarification, and concentration of several food products and by-products such as whey, milk, wine, beer, vinegar fruit, and vegetable juices [1]. Typical advantages over conventional separation systems include high separation precision, better selectivity, operation at room temperature, no chemical damage, high automation, easy operation, energy saving, reduced cost, comprehensive utilization of resources, and reduced pollution. For these reasons, membrane processes are often recognized as the best available technology (BAT) in the food industry [2][3][4]. Among pressure-driven membrane processes, ultrafiltration (UF) has been extensively applied in the treatment of industrial effluents [5][6][7][8][9][10], oil-based emulsions [11][12][13][14], biological macromolecules [15][16][17], milk [18][19][20], sugar cane [21][22], extracts of soybean flour [23], clay suspensions [24], black kraft liquor [25], and fruit juices [26][27][28][29][30][31][32][33][34][35][36][37] among others. Within the fruit juice industry, bergamot, kiwifruit, and pomegranate have great importance in the market, not only for their volume of production, but also because they are characterized by a high concentration of phytochemicals which are recognized to be associated with antioxidant activities within others. Bergamot (*Citrus bergamia*, Risso) is an evergreen tree almost exclusively grown on the Ionian and Tyrrhenian Coast of Reggio Calabria Province (South Italy) with a production of 18,750 tons in 2017 [38], representing a significant economic benefit. Bergamot has been mainly cultivated to extract essential oils with applications in food, cosmetic and pharmaceutical industries [39] because of their high content of phytochemicals such as flavanone glycosides, limonoids, and quaternary ammonium compounds, all health-beneficial biomolecules [40][41]. On the other hand, Bergamot juice is considered a residue for its bitter taste; however, this juice is characterized by a large

quantity and variety of nutraceuticals such as naringin, neoeriocitrin, neohesperidin, rutin, neodesmin, rhoifolin, and poncirus with demonstrated health implications [26]. Kiwifruit is another fruit with a high content of phytonutrients, including carotenoids, lutein, phenolics, flavonoids, vitamin C, and chlorophyll, all of them with strong antioxidant activity [31][32][33][42]; therefore, it offers benefits for specific health conditions and, consequently, it has a great potential for industrial exploitation. Italy, as the major producer worldwide, has a production of 330,000 tons/year (corresponding to 33% of the world production) principally in the regions of Latium, Emilia-Romagna, Piedmont, and Apulia [32]. Pomegranate (*Punica granatum* L.) is located in many different geographical regions, including tropical and subtropical regions. The leading producer locations include Mediterranean countries, India, Iran, and California [43]. Since several authors reported the therapeutic benefits of its consumption including antioxidant, antimicrobial, anti-carcinogenic, and anti-inflammatory properties, increased interest has been garnered for this fruit [44]. Polyphenolic compounds, including ellagotanins, anthocyanins, ellagic acid, and minerals, potassium, magnesium, and copper, are associated with a beneficial effect on health. The global pomegranate market was valued at USD 8.2 billion in 2018 and is expected to reach USD 23.14 billion by 2026 at a Compound Annual Growth Rate (CAGR) of 14.0 percent. Its widespread popularity drives increasing demand for pomegranate and its derivatives (such as pomegranate powder, pomegranate juice, functional beverages) as well as other pomegranate-derived products as a functional food and a source of nutraceuticals [43].

Regarding membrane processes, permeate flux in UF is one of the most critical parameters for evaluating membrane performance. Indeed, the evaluation of permeate flux, usually expressed as volume (or weight) per unit membrane area per unit time, is a critical issue in the projection of scaling-up from experience at the laboratory scale to pilot or industrial plants for a specific volume treatment requirement [45]. Thus, it is a crucial task to forecast permeate flux in long-term operations.

2. Theory

Regarding filtration, Carman [46][47] was the first one to propose a relationship for an aggregate cake, where the prediction of permeate flux is related to the structure parameters of the cake layer, including mean particle size and thickness [48]. This relationship is known as the Carman–Kozeny equation.

In a pressure-driven filtration process such as UF, the pure solvent flux (commonly water) through a porous membrane is directly proportional to the applied hydrostatic pressure, according to:

$$J_w = L_p \Delta P = \frac{\Delta P}{\mu_w R_m} \quad (1)$$

where J_w is the solvent permeate flux, ΔP is the transmembrane pressure, L_p is the membrane permeability, μ_w is the solvent viscosity, and R_m is the intrinsic membrane resistance.

Despite the importance of the critical flux theory as an operational parameter, the majority of the models developed for UF addressed in this review are focused on the prediction of permeate flux over operating time, but they are not focused on the determination of the limiting point or maximum

permeate flux. In this regard, from the Carman-Kozeny equation until these days, several models have been developed as a tool to both describe the reduction in flux and to understand different phenomena involved in membrane filtration, since the understanding of how these factors affect membrane performance is crucial for equipment design [79,80]. Ohanessian et al. [81] mentioned that membrane models available in the literature could be classified into two categories: the end-use, such as permeate flux prediction, and the understanding of the fouling phenomenon. Some authors [1,80,82,83,84] have said that the models applied in UF for flux prediction can be grouped into five categories: (i) concentration polarization models; (ii) osmotic pressure models; (iii) resistance-in-series models; (iv) fouling models, based on the classical film theory model; and (v) non-phenomenological models. [Figure 1](#) summarizes these categories, including models used to predict permeate flux in both MF and UF processes.

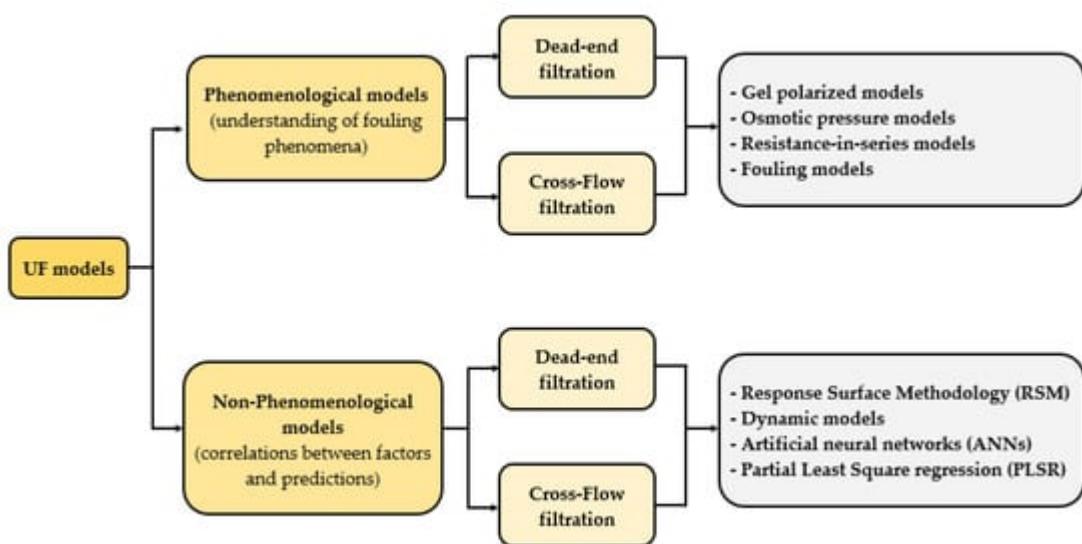


Figure 1. Classification of models developed for MF and UF processes.

3. Analysis of Model Goodness-of-Fit

The categories described previously comprised phenomenological, empirical, semi-empirical, and non-phenomenological models (e.g., statistical tools) developed between 1961 and 2019. In order to compare the capacity of permeate flux prediction, some models for each category were selected and tested with data of three fruit juices clarified by UF. The criteria used for the model selection include a series of items in the following order of importance:

- (i) Type of configuration: models tested or developed for cross-flow filtration of fruit juices were selected.
- (ii) Validation: models with more than one validation were considered.
- (iii) The number of citations: models with a high number of citations were selected in order to take into account the scientific impact of each model.

(iv) Membrane module: models tested or developed in fruit juice processing with hollow fiber and tubular membranes were selected.

(v) Mathematical complexity: Considering the easy application of the models, the most straightforward models were preferred.

Based on these criteria, the models selected were: Shear-induced diffusion by Davis [49] for concentration polarization category; models described by Keden and Katchalsky [50] and Wijmans et al. [51] were selected for osmotic pressure; Hagen-Poiseuille and Boundary gel law described by De et al. [52] were selected for the resistance-in-series category; models described by Ho and Zydny [53], Mondal et al. [54] and the dynamic model by Song [55] were chosen for the fouling category; and models described by Yee et al. [56] and Ruby-Figueroa et al. [57] were selected within the non-phenomenological category. Simulations were performed using experimental data obtained in the UF of three different fruit juices processed for 10 h, as reported by Ruby-Figueroa et al. [57]. In [Table 1](#), characteristics of the juices, membrane types, and operating conditions are reported. Variables such as viscosity, bulk concentration, permeate volume, osmotic pressure, the resistance of the polarized layer, gel concentration, and gel thickness were obtained using a series of correlations available in the literature.

Table 1. Description of the UF membrane, operating conditions, and physicochemical characteristics of the fruit juices analyzed in this work.

	Bergamot	Kiwi Fruit	Pomegranate	Reference
	DCQ II-006C	Koch Series-Cor TM HFM 251	FUC 1582	
Membrane characteristics and operation				
Membrane material	Polysulfone (PS)	Polyvinylidene fluoride (PVDF)	Triacetate cellulose (CTA)	-
Configuration	Hollow Fiber	Tubular	Hollow Fiber	-
Area (m ²)	0.16	0.23	0.26	-
MWCO (kDa)	100	100	150	-
ΔP (bar)	1	0.85	0.6	-
Temperature (°C)	20	25	25	-
Flow (Lh ⁻¹)	114	800	400	-
Porosity (dimensionless)	0.0057	1.1	0.0007	
Tortuosity (dimensionless)	3	3	0.03	-
Membrane thickness (m)	4.7×10^{-7}	2.0×10^{-6}	0.00023	[34]
Pore density, N	6.0×10^{12}	4.0×10^{16}	1.0×10^{13}	[58]

	Bergamot DCQ II-006C	Kiwi Fruit Koch Series-Cor TM HFM 251	Pomegranate FUC 1582	Reference
(number of pores m^{-1})				
Module length, L (mm)	330	406	136	[59]
Module diameter (m)	0.0021	0.025	0.0008	[30][58][60]
Hydraulic resistance (m^{-1})	3.6×10^{12}	1.6×10^{12}	2.1×10^{12}	-
Hydraulic permeability ($mPa^{-1}s^{-1}$)	2.7×10^{-10}	5.9×10^{-10}	4.6×10^{-10}	-
Fruit juices characteristics				
Total soluble solids (°Brix)	9.4	12.6	18.7	[30][38][43] [61]
Titratable Acidity	53.86 (gL^{-1})	-	1.04 (% citric acid)	[30][38][43] [61]
pH	2.40	3.19	3.61	[30][38][43] [61]
Total phenolic compounds	660 (mg/L)	421.6 (mg/L)	1930 (mg GAE/100 L)	[30][38][43] [61]
Turbidity (%)	33.67	-		[30][38][43] [61]
Feed density, ρ (kgm^{-3})	1091	1070	1131	[62][63]
Feed viscosity, μ (Pa s)	0.0019	0.0014	0.0017	[31][64]
Concentration in food (%)	12	10.08	4.9	[27][33][36]
				1990.

Z. Bial, W.F., Dravida, A., Michalek, A.J., Neisen, L. Solute polarization and cake formation in

Membrane Ultrafiltration: Causes, Consequences, and Control Techniques. Membr. Sci. Technol. 1970, 47–97.

The determination of the quality of fit for the selected models was performed using the root mean square error (RMSE), the mean absolute percentage error (MAPE) and the percentage of variability explained (R²) at 95% confidence level. In addition, a validation procedure was carried out using residual analysis. The analysis of

residuals, intended as the difference between the observed and predicted value, is fundamental for validating any model. The residuals represent the prediction error: they must have a random distribution and they must be

Integrated Membrane Operations in the Food Production; Cassano, A., Drioli, E., Eds.; Verlag Walter de Gruyter & Co.: Berlin, Germany, 2013; pp. 163–195. ISBN 3110284677.

unpredictable, which means that they must follow a normal distribution. In cases where the residuals do not have a

normal distribution, the constants and predictors included in the model are intended not to be enough to predict the

process. Salanilla, A.; Verardo, V.; Molina, J.; Segura-Carretero, A.; Martínez-Férez, A. Technical

optimization of an integrated UF/NF pilot plant for conjoint batch treatment of two-phase olives

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- models for the studied juices. In particular, phenomenological models present a capacity of prediction ranging from 25.21 to 79.73% (R-squared), while hybrid system (mathematical modeling and simulation) of aged juice juice to 51.69, and *Profiltration process for the treatment of drinking water*. *Desalination Water Treat.* 2016, 57, 505–517 of permeate flux with R-squares higher than 97% and lowered MAPE (0.25–2.03) and RMSE (3.74–28.91) in 9. Sousa, M.R.S.; Lora-García, J.; López-Pérez, M.F. Modelling approach to an ultrafiltration process comparison with phenomenological models. However, these models do not provide information related to the effect for the removal of dissolved and colloidal substances from treated wastewater for reuse in of different parameters on the permeate flux, a crucial point for the system scaling-up. On the contrary, 10. Torkamanzadeh, M.; Jahanshahi, M.; Peyravi, M.; Rad, A.S. Comparative experimental study on recycled paper manufacturing. *J. Water Process Eng.* 2018, 21, 96–106. phenomenological models are still a proper method for scaling-up purposes, mainly for research in the 10. Torkamanzadeh, M.; Jahanshahi, M.; Peyravi, M.; Rad, A.S. Comparative experimental study on understanding of the UF process. Therefore, the challenge herein is the development of new phenomenological models with assumptions that include the different phenomena occurring in the filtration of complex matrices in 10. Torkamanzadeh, M.; Jahanshahi, M.; Peyravi, M.; Rad, A.S. Comparative experimental study on Water Sci. Technol. 2016, 74, 2737–2750. order to improve the capacity of prediction of permeate flux in long-term operation.
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