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Permeate flux decline prediction in the ultrafiltration of macromolecules with empirical estimation of the gel layer concentration

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Abstract

Permeate flux decline with time was estimated by means of a model [1] that considers gel layer formation as the main fouling mechanism. Several experiments were performed with two different membranes. Both of them were monotubular ceramic membranes. One of them was made of ZrO_2–TiO_2 with a molecular weight cut off (MWCO) of 15 kDa (Orelis, France) and the other was made of Al_2O_3–TiO_2 with a MWCO of 5 kDa (Tami Industries, France). The feed was an aqueous solution of polyethylene glycol with a molecular weight of 35 kDa. All the experiments were carried out at the crossflow velocity of 1 m/s, when the cake layer is more likely to form. Transmembrane pressure (TMP) was varied between 0.1 and 0.5 MPa and the feed concentration was in the range of 5–15 g/L. The experimental results and those predicted by the model were compared for the experimental conditions tested. Model predictions were better for the Orelis membrane than in the case of the Tami Industries membrane under the same experimental conditions.

Keywords: Permeate flux decline; Gel layer concentration; Ultrafiltration; Macromolecules

1. Introduction

To achieve successful prediction of permeate flux decline in membrane processes is essential for the design and operation of membrane industrial processes. Research on modelling of ultrafiltration of macromolecules is being carried out; however models are still far away from explaining and predicting adequately the mechanisms that take part in membrane fouling. Polyethylene

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glycols (PEGs) have been used many times as model macromolecules for membrane modelling purposes [2,3]. In this way, many ultrafiltration models have been applied theoretically estimating all model parameters [4–9]. The best results were obtained with a model that considers cake formation as the main fouling mechanism [5] and a model based in the resistance in series model [8]. Both models make similar predictions for the experimental conditions tested. However the results were not completely satisfactory.

In this work, the model that considers cake formation as the main fouling mechanism was applied empirically estimating the concentration of the gel layer. The model predictions were compared with the experimental results obtained with two different membranes under several experimental conditions.

2. Materials and methods

Two types of monotubular ceramic membranes were used in the experiments: Carbosep M2 membranes supplied by Orelis, S.A. (France) and TiO_2–Al_2O_3 Tami MSKT membranes (Tami, S.A., France). The membrane material of Carbosep M2 membranes was ZrO_2–TiO_2 deposited over a carbon support. The molecular weight cut off (MWCO) of the membranes was 15 kg/mol, whereas for the Tami membranes it was 5 kDa.

These membranes had an internal diameter of 6 mm and an active surface of 35.5 cm^2.

The PEG used in the preparation of the feed aqueous solution was supplied by Merck-Schuchardt (Germany). Its molecular weight was 35,000 g/mol. The NaOH in pellets or flakes supplied by Panreac (Spain) was used to prepare the cleaning solutions for Carbosep M2 and Tami monotubular ceramic membranes. The NaClO supplied by Panreac was also used to clean Tami monotubular ceramic membranes.

Experiments were performed in the ultrafiltration pilot plant described elsewhere [5,7].

3. Theoretical

The model [1] analyzed was developed for crossflow UF, taking into account that it is a dynamic process that changes from a non equilibrium condition to an equilibrium condition in which the cake layer thickness remains constant. This model was described in detail by Vincent et al. [5].

According to the model, flux decline with time can be expressed by means of Eq. (1):

$$J_P = \frac{\Delta P - \Delta P_c}{\mu \cdot R_{bm}} \cdot \left(1 + 2 \cdot r_c \cdot \frac{\Delta P - \Delta P_c}{(\mu \cdot R_{bm})^2} \cdot \frac{C_0}{C_g} \cdot t\right)^{-\frac{1}{2}} \quad (1)$$

where J_P is the permeate flux, ΔP is the transmembrane pressure, ΔP_c is the critical pressure for cake formation, μ is the dynamic viscosity of the permeate, R_{bm} is the resistance of the blocked membrane, r_c is the specific resistance of the cake layer, C_0 is the feed concentration, C_g is the gel concentration and t is time.

The gel layer concentration was estimated by means of Eq. (2) [10]. This equation can be used in the case of tubular membranes and turbulent flow and it has the disadvantage that it is only valid when gel-forming occurs.

$$\frac{C_g}{C_0} = \exp\left(\frac{J_{Pss} \cdot D_{int}^{0.11} \cdot \upsilon^{0.56}}{0.023 \cdot \nu_{tang}^{0.89} \cdot D^{0.67}}\right) \quad (2)$$

In Eq. (2) J_{Pss} is the steady-state permeate flux, D_{int} is the internal membrane diameter, υ is the kinematic viscosity, ν_{tang} is the crossflow velocity and D is the diffusivity.

4. Experimental procedure

Experimental tests were performed at the crossflow velocity of 1 m/s, when the cake layer is more likely to form. Transmembrane pressure (TMP) and feed concentration were varied between 0.1 and 0.5 MPa and 5 and 15 g/L, respectively.

All the experiments were carried out at constant temperature (25°C) and lasted approximately 7 h until steady-state conditions were achieved. After each run the membrane was cleaned. Afterwards, the water permeability of the membrane was checked.

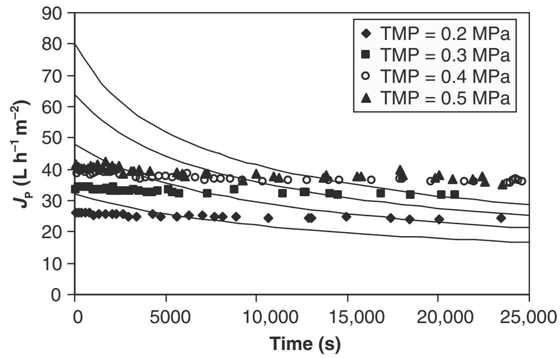

Fig. 2. Comparison between experimental (symbols) and predicted permeate flux (lines) for a feed concentration of 5 g/L for the Tami MSKT membrane.

5. Results

In Figs. 1–3 model predictions of permeate flux vs. time with empirical estimation of the gel layer and experimental results are compared. Previous research [5] demostrated that permeate flux predicted by the model considered in this work was not sensitive to crossflow velocity. Therefore, all the figures correspond to a crossflow velocity of 1 m/s. For this crossflow velocity the probability of gel layer formation is the highest. Consequently, the model is more appropriate for low crossflow velocities.

For the Carbosep M2 membrane (Fig. 1), model predictions when the gel layer concentration is empirically estimated are consistent with those obtained in previous work [5] when the gel layer concentration is theoretically estimated. This may be due to the fact that although the model considers the gel layer concentration as a model parameter, the model is not very sensitive to its variation. Steady-state permeate flux estimated by the model is very similar to the experimental results. For TMPs of 0.3 and 0.4 MPa predicted results are slightly higher than experimental results. For a TMP of 0.2 MPa experimental steady-state permeate flux is equal to that predicted by the model. However, for a TMP of 0.1 MPa steady-state permeate flux predicted by means of the model is slightly inferior to the experimental one. Moreover, the shape of the curve that corresponds to the predicted permeate flux is similar to that experimentally obtained. Nevertheless, for short time scales the model predicts less fouling than that experimentally observed.

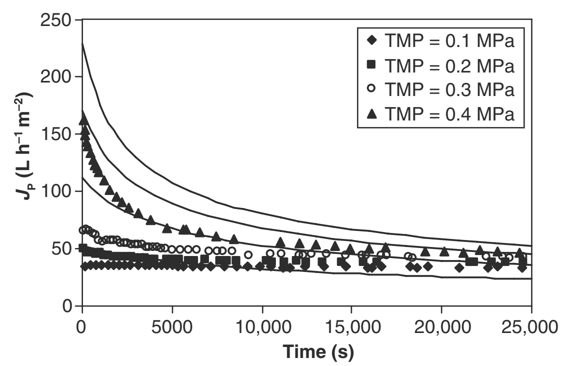

Fig. 1. Comparison between experimental (symbols) and predicted permeate flux (lines) for a feed concentration of 5 g/L for the Carbosep M2 membrane.

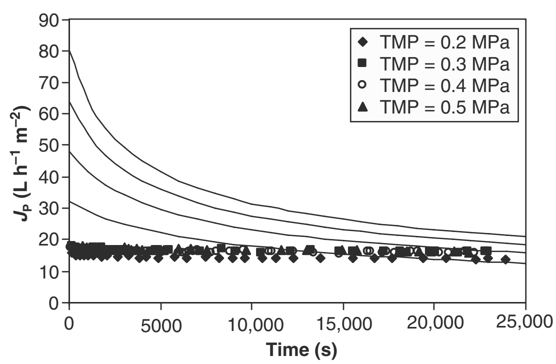

Fig. 3. Comparison between experimental (symbols) and predicted permeate flux (lines) for a feed concentration of 15 g/L for the Tami MSKT membrane.

In the case of the Tami MSKT membrane (Figs. 2 and 3), model predictions and experimental results are not very similar for all the feed concentrations tested. For that reason, the model is not appropriate when permeate flux decline with time is small. The model predicts a noticeable permeate flux decline with time for all the experimental conditions tested. However, experimental data show little variation on permeate flux with time for the Tami MSKT membrane.

For the Tami MSKT membrane and a feed concentration of 5 g/L (Fig. 2), the model predicts the transition from a pressure controlled ultrafiltration process to a mass transfer controlled process. Therefore, differences in permeate flux for different TMPs reduces as TMP increases. Nonetheless, the differences in permeate flux predicted by the model are inferior to those experimentally observed. For long time scales, the model predicts more severe fouling than the experimental one. However, for low time scales it occurs the opposite.

When comparing Figs. 2 and 3, it can be observed that the higher the feed concentration is, the more similar permeate flux predicted by the model for different TMPs is. This is in accordance with the experimental results. However, for the highest feed concentration tested (Fig. 3) experimental results are very similar for different TMPs. This cannot be observed for the results predicted by the model. As well as for the lowest feed concentration tested (5 g/L), the model predicts higher permeate fluxes than those experimentally observed for low time scales and the highest feed concentration tested (15 g/L). Nevertheless, for long time scales, experimental and predicted results are more similar in the case of high feed concentrations (Fig. 3), when the gel layer is more likely to form, than in the case of low feed concentrations (Fig. 2).

6. Conclusions

When the gel layer concentration is empirically estimated, permeate flux predicted by the model is similar to that obtained when the gel layer concentration is theoretically estimated. Therefore, the model does not precisely predict permeate flux, even when the gel layer concentration is empirically estimated.

The worst permeate flux predictions are obtained for short time scales. This can be due to the fact that steady-state conditions were considered to estimate the gel layer concentration. For short time scales, the model predicts less fouling than that experimentally observed for the experimental conditions tested.

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