Estimation of Membrane Fouling Parameters for Concentrating Lactose using Nanofiltration

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Abstract

The research deals with parameter estimation of permeate flux model with fouling for the nanofiltration process. We propose a new technique towards fouling estimation with fouling model being an explicit function of concentration. The objective is to experimentally concentrate lactose in a lactose-salt solution at constant temperature and pressure using cross-flow nanofiltration. The experimental results show a decrease in the permeate flux over time, as the concentration of lactose increases. The limiting flux model is used to model the experimental permeate flux data without fouling. This limiting flux model parameters and the fouling parameters are then estimated via least-squares method using the experimental flux data.

Process Description

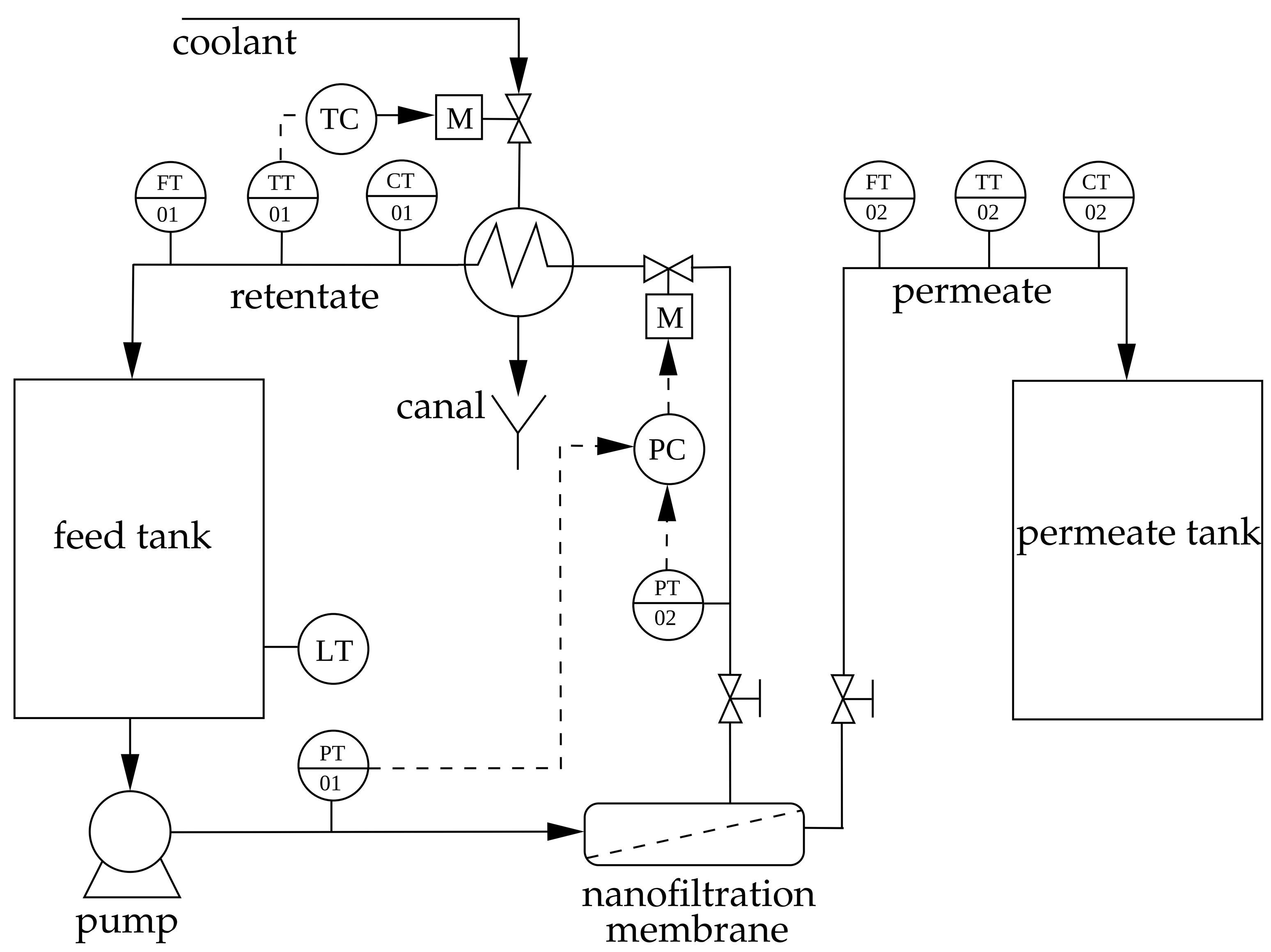

Figure 1: Nanofiltration process scheme.

The nanofiltration experiments were conducted in cross-flow mode at the trans-membrane pressure of 25 bar and temperature of 25 °C.

Materials

- Lactose monohydrate and sodium chloride – Centralchem (Slovakia),
- Reverse osmosis water – in-house production,
- NFW-1812F nanofilter membrane – Synder Filtration, U.S.A

Process Model

The mathematical model of the process is given by material balances of solutes and the overall material balance as:

$$\frac{\mathrm{d}c_i}{\mathrm{d}t} = \frac{c_i}{V} A\, J\, R_i, \qquad c_i(0) = c_{i,0}, \quad i = 1, 2,$$

$$\frac{\mathrm{d}V}{\mathrm{d}t} = -A\, J, \qquad V(0) = V_0,$$

where c_i is the concentration of ith solute, A is membrane area and V is the volume of processed solution. R_i is the rejection coefficient of ith solute defined as $R_i = 1 - c_i/c_{\mathrm{p},i}$, where $c_{\mathrm{p},i}$ is the concentration of ith solute in permeate. As the rejection of lactose is complete ($R_1 = 1$), it does not leave the system, thus at any time

$$c_1 V = c_{1,0} V_0,$$

where c_1 is the concentration of lactose, and $c_{1,0}$, V_0 are the initial conditions.
J is the permeate flux with fouling and can be classified into following models:

- cake filtration model ($n = 0$),
- intermediate blocking model ($n = 1$),
- internal/standard blocking model ($n = 1.5$),
- complete pore blocking model ($n = 2$)

The first three flux models can be described by the following equation:

$$J = J_0 \left(1 + K\,(2-n)(AJ_0)^{2-n} t\right)^{\left(1/(n-2)\right)},$$

while the complete pore blocking model can be expressed as $J = J_0\,\mathrm{e}^{-K\,t}$.

Several experiments with different concentrations of lactose and salt revealed that the flux does not depend on the amount of salt, and hence limiting flux model is used to define the flux of unfouled membrane, i.e. ,

$$J_0(c_1) = k \ln \frac{c_{\mathrm{lim}}}{c_1},$$

with k as the mass transfer coefficient, while c_{lim} as the limiting concentration of lactose.

Optimization Problem

The minimization of the sum of squared differences between experimental flux data (J_{exp}), and estimated flux model (J_{est}) can be formulated as:

$$f = \min_{K,k,c_{\mathrm{lim}}} \sum_{j=1}^{m} (J_{j,\mathrm{exp}} - J_{j,\mathrm{est}})^2$$

s.t.

$$\frac{\mathrm{d}c_1}{\mathrm{d}t} = \frac{c_1^2}{c_{1,0}V_0} A\, J, \qquad c_1(0) = 40\,[\mathrm{g/L}],$$

$$J_0 = k \ln \frac{c_{\mathrm{lim}}}{c_1}, \qquad J = J(J_0, K, n, t), \qquad J_{j,\mathrm{est}} = J(J_0, K, n, t_j),$$

- non-linear estimation
- linear least-squares method for comparison

Results

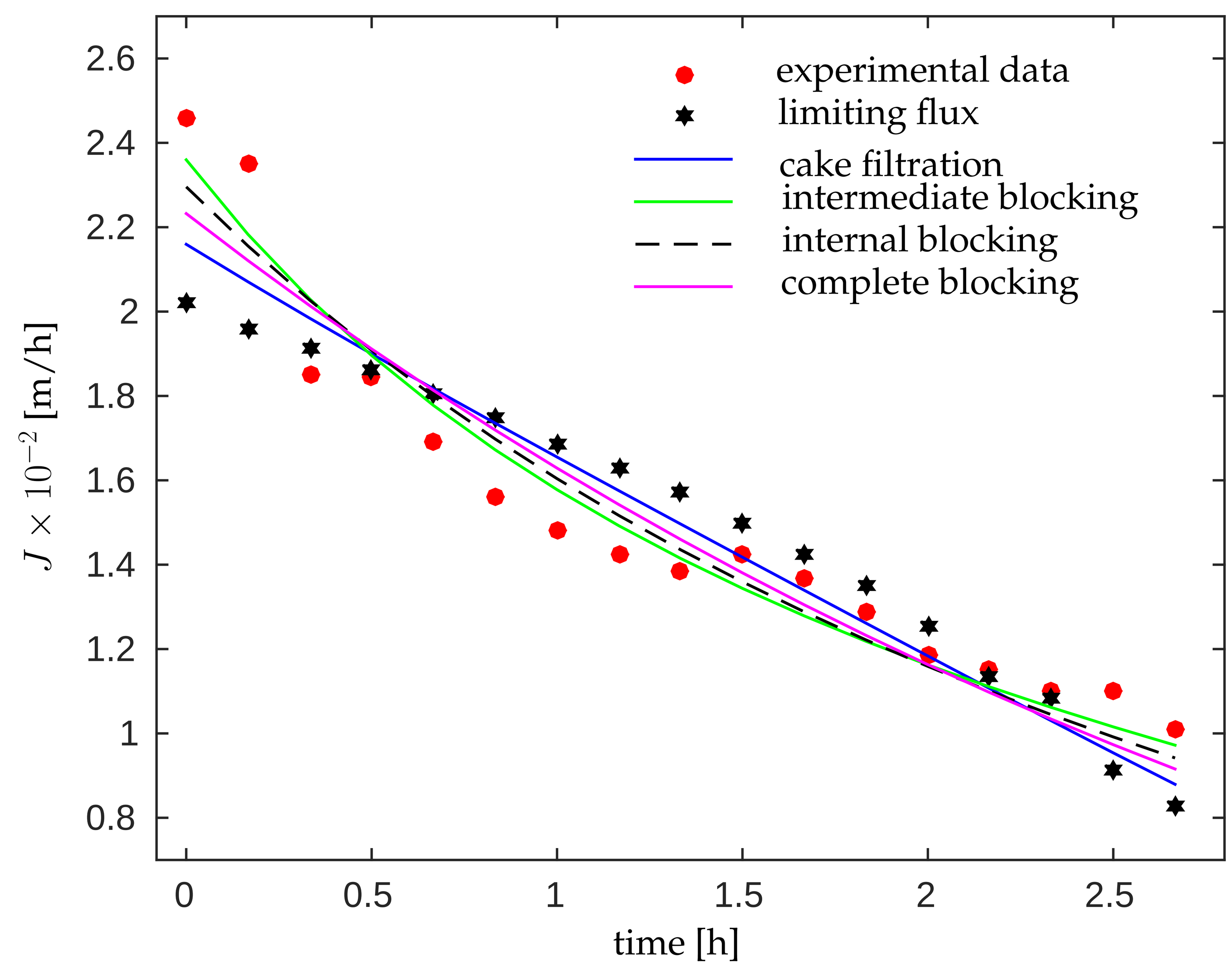

Figure 2: Comparison of experimental data and estimated models.

model (n)	K	$k \times 10^{-2}$ [m/h]	c_{lim} [g/L]	$f \times 10^{-5}$ [m/h]
cake filtration (0)	494.14 [s/m^2]	1.30	210.43	3.43
intermediate blocking (1)	33.47 [1/m]	0.76	880.89	1.36
internal blocking (1.5)	2.59 [1/$\sqrt{s}$]	0.74	880.98	1.91
complete blocking (2)	0.19 [1/s]	0.72	880.97	2.55
limiting flux (–)	–	0.66	880.97	5.98

Table provides estimated values of all parameters. The value of the objective function qualifies the intermediate fouling model as the best fit for the experimental case studied. The limiting flux parameters (k, c_{lim}) estimated for the three fouling models other then the cake filtration model are in a very close proximity of linearly and non-linearly estimated limiting flux model without fouling.

Conclusions

We studied the parameter estimation of membrane flux models with fouling, by using the experimentally obtained data of permeate flux for concentrating lactose using nanofiltration. This estimation was conducted by non-linear least squares method. The results of parameter estimation of limiting flux model showed that the mass transfer coefficient (k) and limiting concentration (c_{lim}) for this experiment were quite high, and lactose could be concentrated with even higher factor. The estimation of fouling parameters resulted in internal/standard blocking ($n = 1.5$), intermediate blocking ($n = 1$) and complete blocking ($n = 2$) models as the better fits, while intermediate blocking model fits the best. The obtained model will be used in the future for experimental evaluation of optimal control theory developed for membrane processes.

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