

Modeling and Simulation of Water Softening by Nanofiltration Using Artificial Neural Network

Mousavi, Mahmoud^{*†}; Avami, Akram

Department of Chemical Engineering, Faculty of Engineering, Ferdowsi University of Mashhad,
P.O. Box 91775-1111 Mashhad, I.R. IRAN

ABSTRACT: An artificial neural network has been used to determine the volume flux and rejections of Ca^{2+} , Na^+ and Cl^- , as a function of transmembrane pressure and concentrations of Ca^{2+} , polyethyleneimine, and polyacrylic acid in water softening by nanofiltration process in presence of polyelectrolytes. The feed-forward multi-layer perceptron artificial neural network including an eight-neuron hidden layer has the least error in modeling this non-linear process. The overall agreement between the artificial neural network results and experimental data is very good for both the volume flux and rejections, because the maximum values of normalized bias and error are -0.01122 and 1.0737 respectively.

KEY WORDS: Artificial neural network, Nanofiltration, Rejection, Volume flux, Water softening.

INTRODUCTION

Water hardness is due to presence of divalent ions like calcium and magnesium. The traditional processes for water softening include lime-soda and ion exchange processes. Membrane softening is becoming an alternative to these processes. Nanofiltration process with charged membranes can be used for this purpose [1,2].

The efficiency and cost of membrane processing is dependent on flux and rejection, which are the function of membrane type, processing parameters and fluid properties. Prediction of flux and rejection with respect to variations of these items is desired. The mathematical models are necessary for this purpose. Two series of models have been represented so far. The first is the models based on physical-chemical-structural descriptions, which include the models based on the extended

Nernst-Planck equation [3-12], frictional models [13,14] and capillary model [15,16]. The second is the black box models, which include the linear and non-linear regression models, irreversible thermodynamics models [17-21], and models based on artificial neural networks (ANNs).

ANNs have been applied to a limited extent to membrane processes. ANNs have been used to predict the evolution of membrane fouling during crossflow microfiltration and ultrafiltration of cane sugar and gum streams [22]. The effects of the ANN hidden structure and training data scatteration on the ANN predictions were studied. It was found that five or six experiments were required to establish a model under constant conditions. In general, satisfactory results were obtained

^{*} To whom correspondence should be addressed.

[†] E-mail: mmousavi@um.ac.ir

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from the ANN analysis. The permeate flux and rejection for reverse osmosis of ethanol and acetic acid and ultrafiltration of bleach plant effluent were predicted by ANNs [23]. ANNs predictions were compared with results from the finely porous mass transfer model. It was found that ANNs were easier to use, more efficient from the computational point of view and sufficiently accurate for industrial design. Recurrent ANNs and a hybrid model (combining a physical model with an ANN) were used to quantify the dynamics of baker's yeast crossflow microfiltration [24]. Although the hybrid model was more accurate, it was limited by the assumptions used in establishing the physical model. The time evolution of membrane fouling and transmembrane pressure in a ultrafiltration process for drinking water were also studied using ANNs [25,26]. It was shown that a simple variable such as turbidity was sufficient as a water quality parameter to model reversible fouling. Very good accuracy from the ANN application was obtained. ANNs were applied to predict the rate of ultrafiltration of proteins and the dynamic crossflow ultrafiltration rate of colloids [27,28]. The careful selection of input variables helped in optimizing the ANN training process and the appropriate selection of training points helped in achieving very accurate predictions of experimental results. Recurrent ANNs were used to study the evolution of flux and deposit thickness in bentonite suspension microfiltration [29]. The recurrent ANNs produced satisfactory predictions of flux and deposit thickness. Additionally, the ANNs were used to predict flux and deposition thickness beyond the experimental period in order to find the limiting values of flux and deposit thickness.

Recently, an ANN has been used to predict the rejections of single salts and mixtures of these salts at a nanofiltration membrane [30]. Predictions for mixtures with a monovalent cation and monovalent / divalent anions were comparable to those of physics-based approaches, and prediction for mixtures with monovalent/divalent cations and a monovalent anion were better than physics-based predictions. More recently, ANNs have been used and tested to dynamically model and simulate crossflow ultrafiltration of milk [31-33]. The aim of the research was the dynamic prediction of permeate flux, total hydraulic resistance and the solute rejection (protein, fat, lactose, ash and total solids) as a function of

transmembrane pressure, temperature, pH and fat percent. The agreement between the empirical data and the model results was excellent with maximum and average errors less than 3.61% and 1.06%, respectively.

Driving force of nanofiltration process is pressure drop. Moreover, the aim of this paper is investigation of water softening, therefore, concentration of Ca^{2+} in the influent is studied next. Polyelectrolytes react with cations in the influent to form complexes which are adsorbed on the inner surface of membrane or are rejected by the membrane that consequently remain in the upstream side of the membrane. Therefore, polyelectrolytes decrease the volume flux and increase the ions rejections. In order to investigate these effects two different types of polyelectrolytes are studied in this paper; polyethyleneimine, which is a weak base and polyacrylic acid, which is a weak acid.

In this paper, a model based on ANNs is represented for the prediction of flux and solute rejection vs. the transmembrane pressure, Ca^{2+} concentration (the criterion of water hardness), polyethyleneimine concentration, and polyacrylic acid concentration in water softening by nanofiltration process in the presence of the above polyelectrolytes.

ARTIFICIAL NEURAL NETWORKS

In recent years the application of ANN for modeling has been greatly increased. An ANN is a group of simple elements, known as neurons, arranged in parallel layers which are inter-connected. An ANN is made of input layer, hidden layers and output layer. Each layer consists of several neurons fully-connected to the other layer by weighted connections. The number of neurons in input and output layers is determined by the structure of the problem in study. The number of hidden layers or the number of neurons in each hidden layer depends on degree of desired accuracy and there is also no way to determine them systematically. ANNs can be classified according to connectivity patterns, number of layers, learning rule and transfer functions.

A feed-forward multi-layer perceptron ANN is defined by at least three layers through which information flows in the forward direction only. So far it has been in use more than other kinds and can provide good results [30,34] (see Fig.1).

Input layer transmit the inputs to the neurons in the

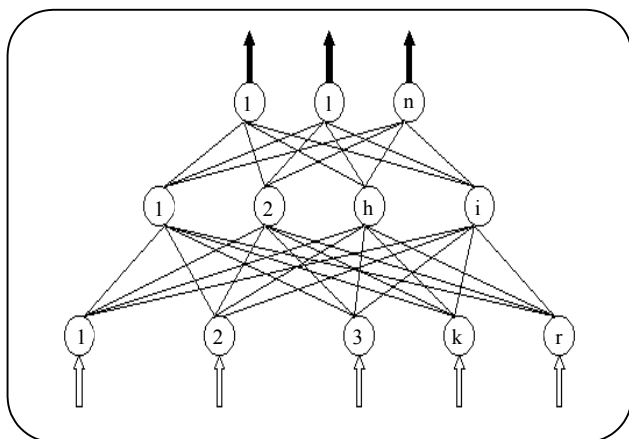


Fig. 1: Schematic structure of an ANN[34].

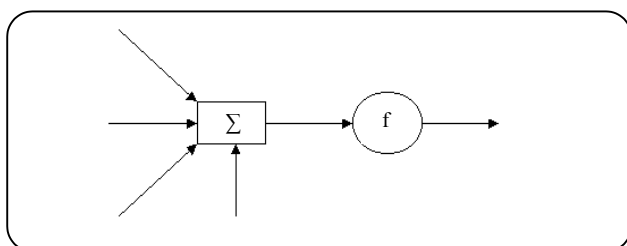


Fig. 2: Mathematical representation of a neuron.

hidden layer, thus there is no need for transfer function. All the outputs which has been received from input layer, are summed and then are transferred through a non-linear transfer function. This is the way, how the output of hidden layer is calculated.

Most useful transfer functions are as: logarithmic sigmoid, hyperbolic tangent, simple linear. Generally one neuron has more than one input. Fig. 2 shows a neuron with multiple inputs.

Net input (n) is calculated by:

$$a = f(w * p + b) \quad (1)$$

Finally output is:

$$n = \sum_{i=1}^m p_i * w_i + b = w * p + b \quad (2)$$

The specification of hidden layers is determined by trial and error method. The universal approximation theory suggests that a network with a single hidden layer and sufficient number of hidden neurons can map any input to any output with any arbitrary degree of accuracy [34].

In addition, there are bias neurons accepting no input and transmit constant output equal to one. These neurons preserve the universal approximation property of the network [35]. ANN can be trained and then it can predict reliable outputs with high accuracy.

EXPERIMENTAL DATA

Each ANN needs the data for training and testing. The necessary data are obtained using a recirculating membrane testing system with the bipolar flat membrane made in Nitto Denko Co. and the University of Tokyo [36]. These membranes contain a negatively charged layer of sulfonated polyethersulfone and a positively charged layer of either polyethyleneimine or quaternary ammonium polyelectrolyte. In order to investigate the effect of polyelectrolytes, a 50 wt% polyethyleneimine solution and polyacrylic acid with a molecular weight of 70000 were used. Both of these polyelectrolytes were obtained from Aldrich Chemical Co.

Volume flux through the membrane and ions concentrations were measured in each experiment. Atomic absorption (Pye Unicam, Model SP 191) was used for the measurement of Na^+ and Ca^{2+} concentrations. Chloride ion concentration was measured by titration with silver nitrate. Membrane characteristics including pore size and thickness were obtained by scanning electron microscopy (SEM). Surface charge density for the membrane used in this work was estimated from the limited data available in the literature. Values of these parameters are 11.6 nm for pore radius, 48 nm for thickness, and 0.065 C/m^2 for surface charge density. The values of ion radius and diffusivities were obtained from the literature [16].

Four sets of experimental data were obtained, each one considers the effect of one parameter on volume flux and rejection, while other parameters are constant (see table 1). The influent in any experiment was including water and CaCl_2 and NaCl salts.

NEURAL NETWORK MODELING DEVELOPMENT

The first stage for making ANN is to choose an optional structure according to inputs and outputs. Then the model must be trained and tested to get the desirable outputs.

As mentioned before, each ANN has input layer, output layer and hidden layers in between. The number of

Table 1: Number of data used for training and testing.

Sets	Training	Testing
Set 1 (effect of pressure drop)	11	10
Set 2 (effect of Ca^{2+} concentration)	16	15
Set 3 (effect of polyethyleneimine concentration)	11	10
Set 4 (effect of polyacrylic acid concentration)	11	10

neurons in input layer is the same as the number of input parameters. The effective parameters on volume flux and rejection are transmembrane pressure and concentrations of Ca^{2+} , Cl^- , Na^+ , polyethyleneimine, and polyacrylic acid. So the input layer has 6 neurons. The number of neurons in output layer is also determined by the case under consideration. The output parameters are: volume flux and rejections of Ca^{2+} , Cl^- , and Na^+ . So the output layer has 4 neurons. One or more hidden layer must be added for completing the network. According to universal approximation theory, single hidden layer was chosen.

In order to train the network and get suitable weights and biases for each set of data, the data is divided into two groups of training and testing. Table 1 shows the number of data used for training and testing for each case, separately.

For checking and selecting the best cases, one must calculate and compare errors. There are several types of standard error calculations among which the following one is used:

$$E = \sum (y'_k - y_k)^2 \quad (3)$$

Error is calculated and its minimum value provides the best weights and biases at the end of testing for each output.

The most important part of an ANN is the proper selection of transfer function. Logarithmic sigmoid function known as "logsig" for hidden layer and simple linear function known as "purelin" for output layer were determined as the best choices by trial and error. The general form of logarithmic sigmoid function is given as follow:

$$f(x) = \frac{1}{1 + e^{-x}} \quad (4)$$

And general form of linear function is:

$$f(x) = x \quad (5)$$

Incidentally, 8 neurons for hidden layer are proper to minimize the error[37].

RESULTS AND DISCUSSION

There are the four sets of experimental data to study the effect of pressure drop and concentrations of Ca^{2+} , polyethyleneimine, and polyacrylic acid on the volume flux and rejection of solutes in water softening by nanofiltration process.

Results of ANN model presented in this paper are compared with the experimental data. In the following sections some of these results are presented. More details can be found in [37].

Incidentally, the mean difference between the predicted results by model and experimental data divided by experimental data, is called normalized bias, is used for comparison of predicted results and experimental data in addition to error.

Effect of pressure drop

Pressure drop (the driving force for nanofiltration process) has a great effect on the volume flux and rejection of solutes. So the first set is about the variations of pressure drop while the other input parameters are constant and have the following values:

$[\text{Ca}^{2+}] = 150\text{ppm}$, $[\text{Na}^+] = 100\text{ppm}$, $[\text{Cl}^-] = 420.6\text{ppm}$, $[\text{polyelectrolytes}] = 0$

Figs. 3 and 4 represent the effect of pressure drop on the volume flux and rejection of Ca^{2+} , Cl^- , and Na^+ . The solid lines show the ANN results while the triangles are the experimental data.

The volume flux is proportional to driving force and increases with increasing dp according to Fig. 3. The increase of the rejections in Fig. 4 issues from less increase of the flux of solutes than the flux of water with increasing dp .

The values of normalized bias and error are represented in tables 2 and 3, respectively. The normalized bias values show that the model underpredicts slightly the volume flux and underpredicts very slightly the rejections. Also, the error values show that model has much little error.

Table 2: The normalized bias values.

Sets	Volume flux	$R_{Ca^{2+}}$	R_{Cl^-}	R_{Na^+}
Set 1	-0.01122	-0.00066	-0.00075	-0.00104
Set 2	-0.00065	-0.00317	-0.00344	-0.00411
Set 3	-0.00036	0.000267	5.80E-05	-1.29E-05
Set 4	-0.00166	0.000491	0.000551	0.00172

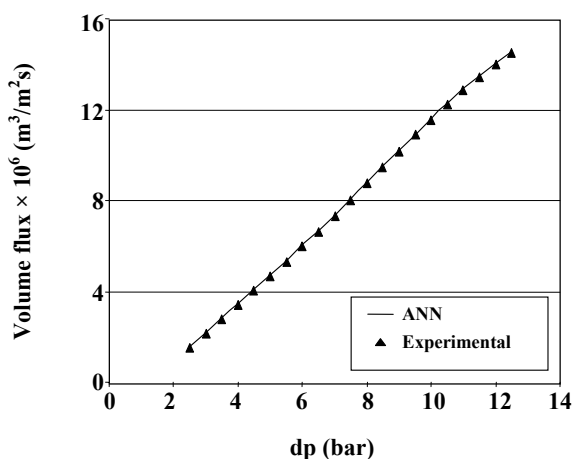


Fig. 3: The ANN and experimental results for volume flux versus pressure drop.

Effect of Ca^{2+} concentration

The concentration of Ca^{2+} is changed in order to study effect of water hardness. The values of other parameters are:

$dp=10\text{bar}$, $[Na^+]=100\text{ppm}$, and $[\text{polyelectrolytes}]=0$

Figs. 5 and 6 show the variations of volume flux and rejections versus variations of Ca^{2+} concentration. While Ca^{2+} concentration increases, osmotic pressure increases that results in decrease of effective driving force, so the volume flux and rejections will decrease.

The normalized bias values are available in table 2. It shows that the model underpredicts very slightly the empirical data. Incidentally, the model predicts volume flux with very much little error and rejections with little error (see table 3).

Effect of polyelectrolyte concentration

Adding polyelectrolytes to input water is an important factor. To determine the role of this factor, the effect of two different types of polyelectrolyte is investigated. Presence of polyelectrolytes results in their adsorption on the

Table 3: Error values.

Sets	Volume flux	$R_{Ca^{2+}}$	R_{Cl^-}	R_{Na^+}
Set 1	0.0727	0.0414	0.0433	0.0489
Set 2	0.0044	1.0737	1.0323	0.7985
Set 3	0.0003	0.0106	0.0008	0.0038
Set 4	0.0054	0.1177	0.0267	0.1955

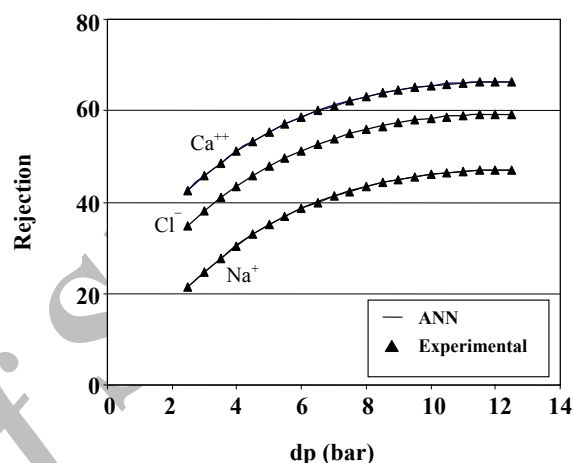


Fig. 4: The ANN and experimental results for the rejection of solutes versus pressure drop.

inner surface of membrane pores and also adsorption of ions by polyelectrolytes. Thus, as illustrated in Figs. 7-10, the volume flux is decreased and rejections are increased.

Polyethyleneimine

In the third set, the concentration of polyethyleneimine is changed and the values of other parameters are as below:

$[Ca^{2+}] = 131\text{ppm}$, $[Na^+] = 100\text{ppm}$, $[Cl^-] = 387\text{ppm}$, $dp=10\text{bar}$, $[\text{polyacrylic acid}]=0$

Figs. 7 and 8 illustrate the effect of this parameter. At higher concentrations the adsorption of polyethyleneimine on the surface of membrane pores is decreased and the formation of complex between the polyelectrolyte and ions controls the volume flux and rejections.

Tables 2 and 3 represent the normalized bias and error values. The model underpredicts very slightly the volume flux and overpredicts very slightly the rejection of Ca^{2+} and Cl^- while underpredicts very slightly the rejection of Na^+ . Also error values show that the model has very much little error.

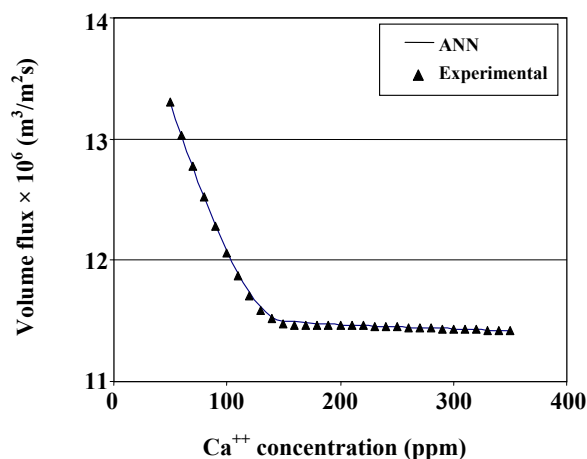


Fig. 5: The ANN and experimental results for volume flux versus Ca^{2+} concentration.

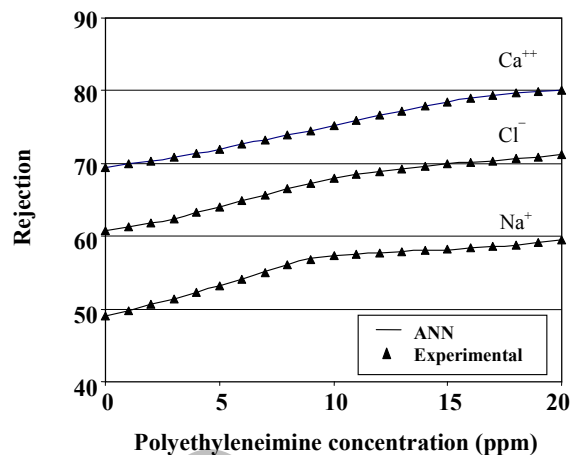


Fig. 8: The ANN and experimental results for the rejection of solutes versus polyethyleneimine concentration.

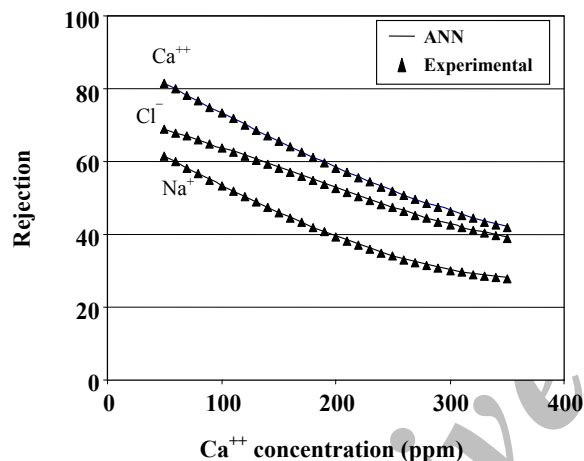


Fig. 6: The ANN and experimental results for the rejection of solutes versus Ca^{2+} concentration.

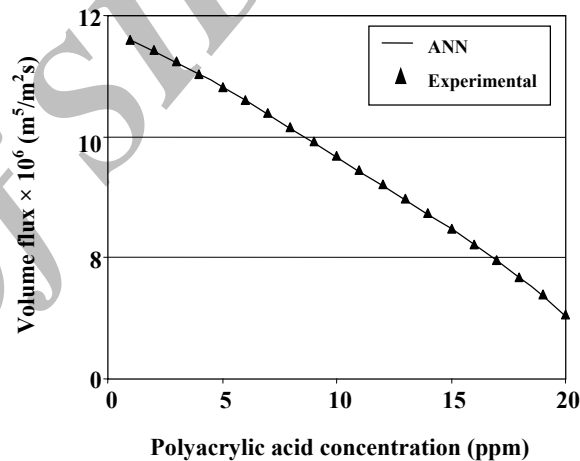


Fig. 9: The ANN and experimental results for volume flux versus polyacrylic acid concentration.

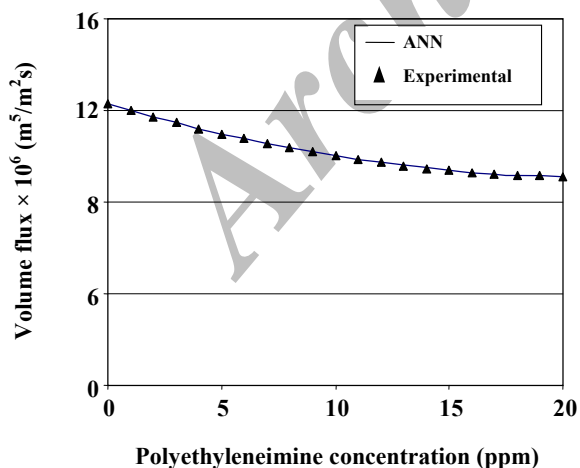


Fig. 7: The ANN and experimental results for volume flux versus polyethyleneimine concentration.

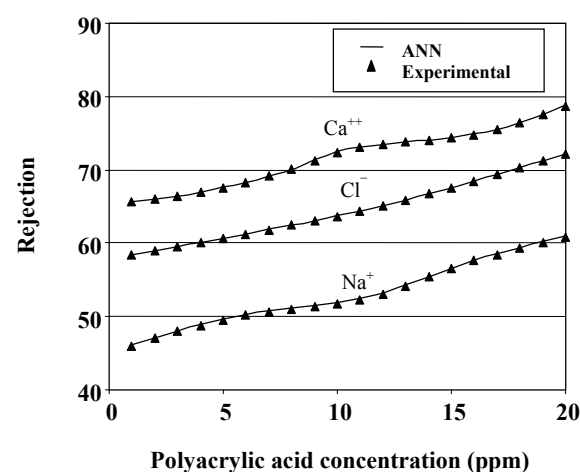


Fig. 10: The ANN and experimental results for the rejection of solutes versus polyacrylic acid concentration.

Polyacrylic acid

Finally, the effect of polyacrylic acid concentration is investigated. It must be noted that the other parameters are constant and their values are as below:

$[Ca^{2+}] = 150 \text{ ppm}$, $[Na^{+}] = 100 \text{ ppm}$, $[Cl^{-}] = 420.6 \text{ ppm}$, $dp = 10 \text{ bar}$, $[polyethylenetmin \ e] = 0$

Figs 9 and 10 represent the results. The model predicts the empirical data with acceptable accuracy. The normalized bias values show that the model underpredicts very slightly volume flux and overpredicts very slightly rejections (see table 2). It has very much little error for volume flux and rejection of Cl^{-} and little error for rejections of Ca^{2+} and Na^{+} (see table 3).

CONCLUSIONS

The values of normalized bias and error which is obtained by the model indicate that ANN model can be one of the best methods in modeling water softening using nanofiltration. According to the present paper, the feed-forward multi-layer perceptron ANN with 8 neurons must be used to determine the volume flux and rejection of solutes in presence of polyelectrolytes. Logarithmic sigmoid and simple linear transfer functions are obtained as the best choices for the hidden and output layers respectively.

The maximum and minimum errors that are obtained from the model are 1.0737 and 0.0003 respectively. The modeling results show that very slightly, the model in sets 1 and 2 underpredicts the experimental data, while in sets 3 and 4 underpredicts the volume flux and overpredicts major of the rejections. As a result, the overall agreement for each set was very good and the model has effective usage to predict the volume flux and rejections in water softening using nanofiltration process.

List of symbols

a	Output of a neuron
b	Bias
dp	Pressure drop (bar)
E	Error
f	Transfer function
m	Number of input elements
n	Net input
p	Input vector
R	Rejection

y	Experimental output
y'	Output of network
w	Network weight

Subscripts

i	Input
k	Output

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