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Modeling of waste brine nanofiltration process using artificial neural network and adaptive neuro-fuzzy inference system

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ABSTRACT

In this study, artificial neural network (ANN) and adaptive neuro-fuzzy inference system (ANFIS) models were used to predict the average permeate fluxes and sodium chloride rejection of waste brine nanofiltration process. The ANFIS and ANN models were fed with three inputs: feed concentration (40, 60, 80, and 100 g/l), pressure (1.0, 1.25, 1.5, 1.75, and 2.0 MPa), and temperature (30, 40, and 50°C). Both models were trained with 30% of total experimental data. Thirty percent of the experimental data were used to test the prediction ability of ANFIS and ANN models. Independent permeate flux and NaCl rejection predictions were calculated for the remaining of total data (40%). The results revealed that ANN predictions agreed well with variety of experimental data. It was found that ANN with 1 hidden layer comprising 8 neurons gives the best fitting quality, which made it possible to predict flux and rejection with acceptable correlation coefficients ($r = 0.90$ and $r = 0.87$, respectively). A hybrid method (the combination of least squares and back propagation algorithms) was used as the training method of the ANFIS. The overall agreement between ANFIS predictions and experimental data was excellent for both permeate flux and salt rejection ($r = 0.96$ and $r = 0.94$, respectively).

Keywords: Membrane; Effluent; Fuzzy inference system; Neural network; Simulation; Sodium chloride

1. Introduction

The ion-exchange resin process is currently considered as one of the most efficient sugar liquor decolorizers. High molecular weight sugar liquor colorants such as melanins, melanoidins, products of alkaline degradation of sucrose, caramels, and polyphenols are first adsorbed onto the resins, and finally desorbed from the exhausted resins using an alkaline 100 g/l sodium chloride solution at approximately pH 12. Typical molar mass of colorants ranges from 500 to 20,000 D. Effluents resulting from this regeneration contain mostly sodium chloride (up to 100 g/l) and important amounts of colored organic matter, and, therefore, constitute a major pollution source [1–3].

Nanofiltration (NF) process was often applied at the end of the treatment procedure, accompanied by other separation techniques. The NF process benefits from ease of operation, reliability and comparatively

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low energy consumption as well as high efficiency of pollutant removal [4]. NF has now been widely used in salt removal in water treatment and fractionation of small molecules in many other industries [5]. NF membranes have the advantages of providing a high water flux at low operating pressure and maintaining a high salt and organic matter rejection [6]. The rejection mechanism of NF involves size sieving (steric hindrance), Donnan exclusion (electrostatic interaction of charged solutes with charges attached to the membrane matrix) and dielectric exclusion (interaction of ions with a polarized charge) [7]. NF process allowed the achievement of 74% reduction in salt consumption and 89% reduction in water consumption while reducing the volume of toxic waste discharged from sugar refineries [2].

Predicting the performance of NF process in terms of permeate flux and components rejection is necessary for the operation analysis and optimization of present process and design of a new membrane separation [4,8]. In the last two decades, researchers explored the potential of artificial neural networks (ANNs) and adaptive neuro-fuzzy inference system (ANFIS) as an analytical alternative to conventional modeling techniques, which are often limited by strict assumptions of normality, linearity, homogeneity, and variable independence. Fuzzy inference systems (FIS) and ANNs are both model-free numerical estimators. They share the ability to improve the predictive capability of a system working in uncertain, imprecise, and noisy environments [9].

ANNs are information processing networks constituting a set of highly interconnected neurons arranged in multiple layers that can be trained to fit one or more dependent variables to any degree of accuracy using a set of independent variables as inputs [10]. During the last 15 years, ANNs have been at the focus of much attention, largely due to their wide range of applicability and ease by which they handle complex and highly nonlinear problems. ANNs were successfully applied to problems from various areas including business, medical and industrial fields [11]. Once the ANN is trained using experimental data, it can be used in a purely predictive mode to calculate the dependent variable(s) for any values of input variables. Process modeling is an area where ANNs of various configurations and structures have been considered as alternative modeling techniques, particularly in cases where reliable mechanistic models cannot be obtained. In the field of membrane separation processes, ANNs have successfully been applied to different types of membranes including microfiltration, ultrafiltration, and NF [8,12–15]. For example, ANN simulation used to predict the rejection of two salts (NaCl and $MgCl_2$) at typical seawater concentrations in a crossflow NF membrane process. ANN model successfully tracked the nonlinear behavior of rejections vs. pressure and flux [4]. Salehi et al. [12] found that ANN with one hidden layer comprising nine neurons gives the best fitting with the experimental data, which made it possible to predict flux and total hydraulic resistance with high correlation coefficients (0.96 and 0.98, respectively).

FIS and ANN are complementary technologies. In order to utilize the strengths of both, FISs and ANNs may be combined into an integrated system called ANFIS; the integrated system then has the advantages of both ANNs (e.g. learning abilities, optimization abilities, and connectionist structure) and FISs (e.g. humanlike if-then rules, and ease of incorporating the expert knowledge available in linguistic terms) [9]. The ANN and ANFIS models have the unique advantage that no clear relationship between the input and output variables needs to exist before the model is applied since the relationship is identified through a self-learning process. By utilizing data samples from experiments, both ANN and ANFIS models can be applied to solve problems with no (or with too complex) algorithmic solutions, or in cases where the input information is incomplete or uncertain [16].

The concepts of ANFIS were firstly proposed in 1993 by Jang; in general, an ANFIS is a feed-forward neural network of six layers [17]. ANFIS has an excellent ability of approximation and generalization. Sugeno fuzzy model is the popular fuzzy model applied in ANFIS. ANFIS with zero-order Sugeno model has been proved to have universal approximation ability under certain circumstances. Because of its good characteristics, ANFIS has been widely used in many fields, such as system identification, fuzzy control, and data processing [18]. For example, Saghatoleslami et al. used ANFIS to dynamically model the crossflow ultrafiltration of milk to predict permeate flux and total hydraulic resistance as a function of transmembrane pressure, pH, temperature, fat, molecular weight cut off, and processing time [19]. In another study, fuzzy inference system has been applied in modeling of crossflow milk ultrafiltration [20].

Recently, a few studies have considered the application of ANNs and ANFIS for modeling and controlling of desalination systems and membrane processes. In this study, the efficiency of ANN and ANFIS models for prediction of permeate flux and NaCl rejection of NF treatment of waste brine obtained from sugar decolorizing resin regeneration was investigated at different conditions of feed concentration, pressure, and temperature.

2. Materials and methods

2.1. Experimental setup

The experimental data employed for modeling was collected using a NF pilot plant system. The used polymeric tubular AFC80 membrane was purchased from PCI membrane systems, USA, made of polyamide film. The MICRO 240 tubular module had 0.024 m^2 membrane area and 30 cm length, which accepts two 12.7 mm diameter membrane tubes. The duration of each experimental run was 60 min. A permeate collection vessel, located on a digital mass balance (±0.05 g), was used to collect permeate and measured permeate flux (kg/m^2 s) during the experiments.

For all experiments, the operating pressure was in the range of 1.0–2.0 MPa (1.0, 1.25, 1.5, 1.75, and 2.0 MPa) and the temperature varied from 30 to 50˚C (30, 40, and 50˚C). In order to investigate the effect of feed concentration on the average flux and NaCl rejection, feeds were prepared at four concentration levels of 40, 60, 80, and 100 g/l.

The salt concentration in the retentate and permeate was determined based on the conductivity of samples which were measured using a conductivity meter (Jenway 4010, Bibby Scientific Limited, UK) at 20˚C [1]. NaCl rejection was also calculated using the following equation:

$$\mathrm{R} = \left(1 - \frac{C_\mathrm{p}}{C_\mathrm{b}}\right) \times 100 \tag{1}$$

where C_p and C_b are the concentrations of salt in permeate and retentate sample, respectively.

2.2. ANN simulation

The most popular ANN is the multilayer feed-forward neural network, where the neurons are arranged into layers of input, hidden, and output. A schematic description of the three-layered network structure used in this study is shown in Fig. 1. Feed-forward neural network usually has one or more hidden layers, which enable the network to model nonlinear and complex functions [12]. In this type of ANNs, information flows in the forward direction only. The number of input neurons corresponds to the number of input variables into the neural network, and the number of output neurons is similar to the number of desired output variables. In between the input and the output layers, there is at least one hidden layer that can have any number of neurons. The number of neurons in the hidden layer(s) depends on the application of the network. In the hidden and output layers, the net input (X_j) to node j is of the form [21]:

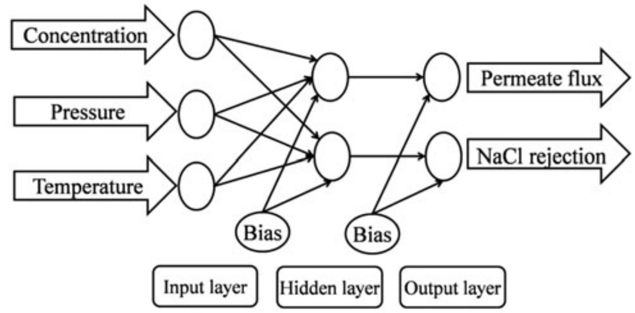

Fig. 1. Multilayer feed-forward perceptron network architecture with one hidden layer for prediction of NaCl rejection and average flux of regeneration waste brine.

$$X_j = \sum_{i=1}^{n} f(W_{ij}y_i) + b_j \tag{2}$$

where y_i are the inputs, W_{ij} are the weights associated with each input/node connection, n is the number of nodes, and b_j is the bias associated with node j. Additionally, bias is an extra input added to neurons. The reason for adding the bias term is that it allows a representation of phenomena having thresholds [22]. Each neuron consists of a transfer function expressing internal activation level. Output from a neuron is determined by transforming its input using a suitable transfer function. The transfer function can be linear or nonlinear (commonly sigmoidal and hyperbolic tangent) functions depending on the network topology [12].

In this work, the operational variables of NF treatment of waste brine (feed concentration, pressure, and temperature) were used as inputs and permeate flux and NaCl rejection as output. A hyperbolic tangent activation function (Eq. (3)) was chosen to be used as the transfer function in the hidden and output layers, due to lower obtained mean-squared error (MSE) values comparing to the respective sigmoid function and linear function:

$$\tanh = \frac{e^{x} - e^{-x}}{e^{x} + e^{-x}} \tag{3}$$

The network architecture refers to the number of layers in the network and the number of neurons in each layer. The universal approximation theory suggests that a network with a single hidden layer with sufficient number of hidden neurons is able to map any input to any output to any degree of accuracy

[23,24]. The ANN used in the present work featured a one hidden layer and bias nodes in the input and hidden layers (Fig. 1). On the other hand, to find the best architecture, different networks were built with different hidden neurons. In total, 120 data were collected from experiments at different feed concentrations, pressures, and temperatures. The data order was first randomized and then all data were separated into three partitions. Thirty percent of data were used to train the network, thirty percent of data were used to test the prediction quality of the network during the training, and the remaining forty percent of data was used to validate the performance of the trained network for prediction of unseen data. In this study, the fast Levenberg–Marquardt (LM) optimization technique was used to train the network [25]. The most widely employed algorithm for training ANNs is the back propagation approach [26]. It is based on searching an error surface (error as a function of ANN weights) using gradient descent for point(s) with minimum error. Each iteration in back propagation constitutes two sweeps: forward activation to produce a solution and the backward propagation of the computed error to modify the neurons' weights [27]. Testing step was carried out with the best weights stored during the training. Each predicted value was compared against the experimental value to test of network performance. For this purpose, 3 statistical parameters including the correlation coefficient (r), Eq. (4), MSE, Eq. (6), and mean absolute percentage error (MAPE), Eq. (7), were used as follows:

$$r = \sqrt{1 - \frac{\sum_{i=1}^{N}[\mathrm{O}_i - \mathrm{T}_i]^2}{\sum_{i=1}^{N}[\mathrm{O}_i - \mathrm{T}_m]^2}} \tag{4}$$

$$\mathrm{T}_m = \frac{\sum_{i=1}^{N}\mathrm{O}_i}{N} \tag{5}$$

$$\mathrm{MSE} = \frac{\sum_{i=1}^{N}(\mathrm{O}_i - \mathrm{T}_i)^2}{N} \tag{6}$$

$$\mathrm{MAPE} = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{\mathrm{O}_i - \mathrm{T}_i}{\mathrm{O}_i}\right| \times 100 \tag{7}$$

where O_i is the i^{th} actual value, T_i is the i^{th} predicted value, and N is the number of data.

The NeuroSolutions software (Excel software release 6.0) was used for designing the neural networks and simulation of NF treatment of waste brine. This software incorporates various types of ANN presented by NeuroDimension, Inc., USA.

2.3. ANFIS simulation

Fuzzy logic is widely used in the field of intelligent control, classification, pattern matching, image processing, etc. [28–32]. Neuro-fuzzy uses neural network learning functions to refine each part of the fuzzy knowledge separately. One approach to the derivation of a fuzzy rule base is to use the self-learning features of ANNs, to define the membership function (MF) based on input–output data [28].

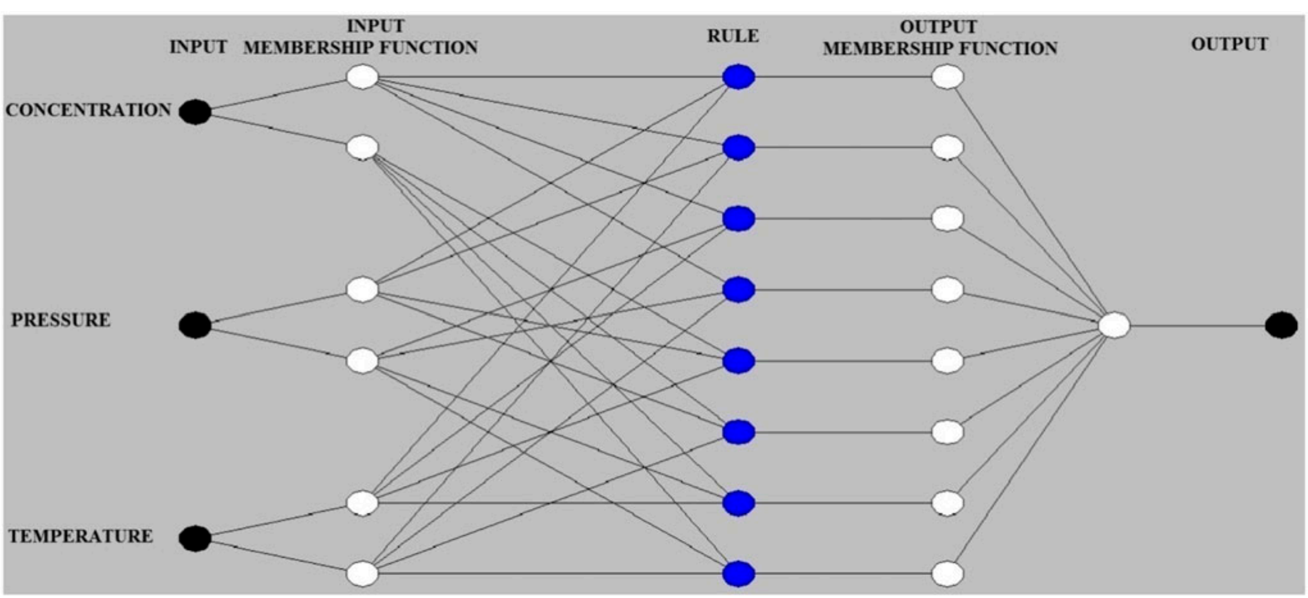

Fig. 2. The General structure of the ANFIS for the NF model with three inputs.

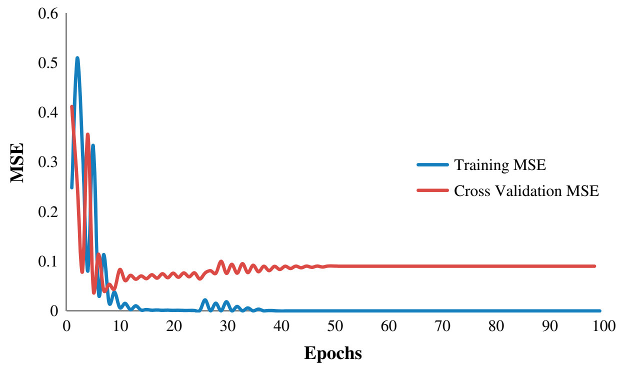

Fig. 3. MSE as a function of the number of iterations (epochs) of ANN training process.

In this study, the architecture of ANFIS had six layers to accomplish the tuning process of the fuzzy modeling system (Fig. 2). ANFIS modeling was started by obtaining a data-set (input–output data points). The data order was first randomized and then all data were separated into three partitions. Thirty and forty percent of total data were used to training, testing, and validating (unseen data) the network, respectively. Each input/output pair contained three inputs (feed concentration, pressure, and temperature) and one output (average permeate flux or NaCl rejection). The training data-set was used to find the initial premise parameters for the MFs by equally spacing each of the MFs. The number of MFs assigned to each input variable is chosen by trial and error.

The ANFIS toolbox of Matlab 7.6 was used to obtain the results, and to build a neuro-fuzzy model for predicting the permeate flux and NaCl rejection of NF treatment.

3. Results and discussion

The experimental results of NF treatment of waste brine obtained from sugar decolorizing resin regeneration have been reported previously [1]. Totally, the permeate flux was increased by increasing the temperature and pressure; however, it decreased with the increase in feed concentration. The sodium chloride rejection was decreased by increasing the feed concentration and temperature, whereas it increased with an increase in pressure.

3.1. ANN results

Initially, the network was trained by 18 data points. The training process was carried out for 1,000 epochs or until the cross-validation data's MSE did not improve for 100 epochs to avoid overfitting of the network. A plot of the MSE and the number of epochs is shown in Fig. 3. A sharp drop was observed for MSE in the first little iteration (fast training) and training was completed after 56 epochs. This is a well-known characteristic of the LM optimization method [26]. LM algorithm provided faster convergence and the lower MSE than other training algorithms such as standard back propagation (BP), resilient BP, conjugate gradient, and quasi-Newton methods.

The results showed that the ANN with eight hidden neurons had the minimum MSE values (0.9 and 3.4) for permeate flux and NaCl rejection prediction, respectively. Table 1 illustrates the weight and bias values of the optimum ANN, which could be used to predict NaCl rejection and average flux of NF treatment of waste brine. Figs. 4 and 5 show the results of ANN model for the training/testing data. The correlation coefficients (r) are around 0.999, meaning that there is high linear correlations between the

Table 1
Corresponding weight and bias values of each neuron for optimum ANN configuration selected to predict NaCl rejection and permeate flux of regeneration waste brine

Hidden neurons	Bias	Input neurons			Output neurons	
		Concentration (g/l)	Pressure (MPa)	Temperature (°C)	Flux (kg/m^2 s)	Rejection (%)
1	−0.061	0.6446	0.1857	−0.3148	−0.2482	0.1618
2	–	−1.4839	0.6599	−0.5733	−0.6466	0.1405
3	0.588	0.3619	−0.2244	−0.435	0.6093	−0.4239
4	0.104	0.0912	0.5956	1.2611	−0.2616	−0.0757
5	0.053	0.1332	0.5894	−0.0684	0.0995	1.0431
6	0.216	−0.0039	−0.0107	0.0114	0.4479	−0.8753
7	–	0.6156	0.2857	−0.2153	0.842	−0.0685
8	–	−0.5552	0.6818	−1.8216	0.1452	−0.8955
Bias					−0.0345	0.1383

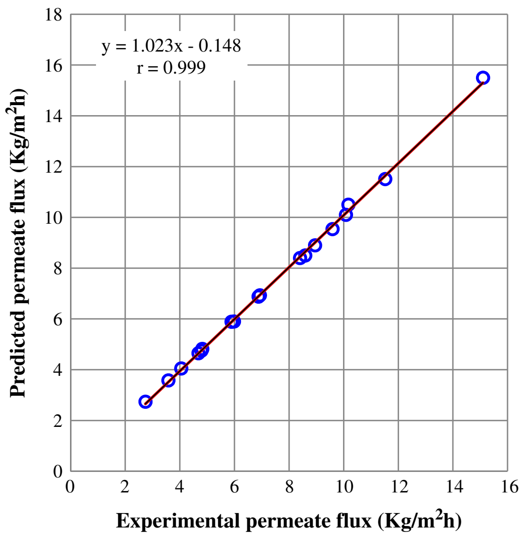

Fig. 4. Permeate flux measurements vs. ANN predictions for the training/testing data-set.

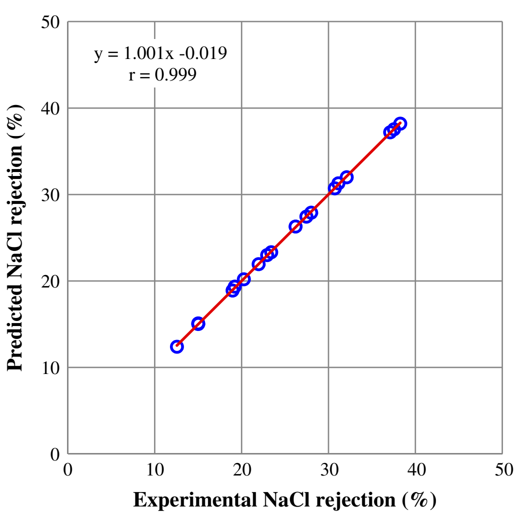

Fig. 5. NaCl rejection (%) measurements vs. ANN predictions for the training/testing data-set.

experimental permeate flux and NaCl rejection data and the predicted values.

The prediction efficiency of the selected ANN model for unseen data is presented in Figs. 6 and 7. It can be seen that the average flux and NaCl rejection values predicted by the best ANN configuration (3/8/2) are plotted against their experimentally measured values. The calculated correlation coefficient values for estimation of flux and NaCl rejection were 0.90 and 0.87, respectively, which show high correlation between predicted and experimental values. Abbas and Al-Bastaki used ANN to predict independent

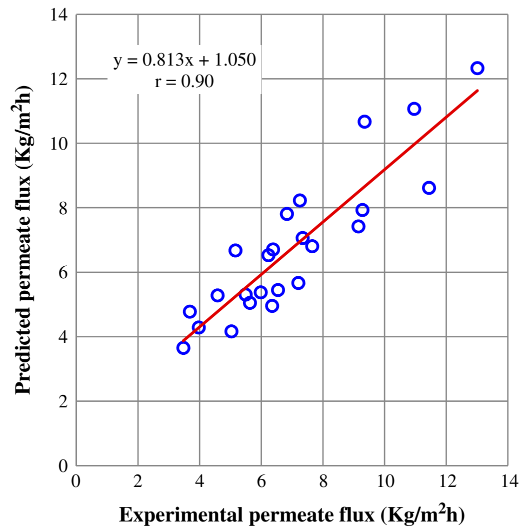

Fig. 6. Experimental (validating data-set) vs. predicted values for permeate flux of NF treatment of waste brine by optimum ANN configuration.

rejection values and compared to the measured rejections and an excellent correlation was found [26]. Bowen et al. applied ANN to predict the rejections of single salts (NaCl, Na_2SO_4, $MgC1_2$, and $MgSO_4$) and mixtures of these salts at a NF membrane. The overall agreement between ANN predictions and experimental data was very good for both single salts and mixtures [8]. Salehi et al. [12] used ANN to dynamic modeling of flux and total hydraulic resistance in NF treatment. They reported that the ANN with nine hidden neurons

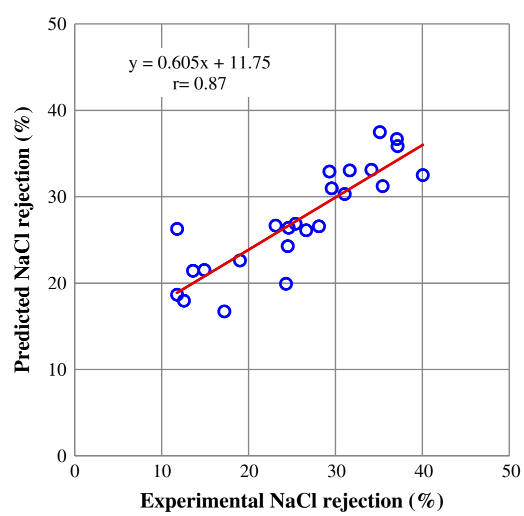

Fig. 7. Experimental (validating data-set) vs. predicted values for NaCl rejection (%) of NF treatment of waste brine by optimum ANN configuration.

had the minimum MAPE values (3.27) for dynamic flux prediction. ANN models were used to predict the total acceptance of ice cream by Bahramparvar et al. [10].Thirty, ten, and sixty percent of the sensory attributes data were used to train, validate, and test the ANN model, respectively. It was found that ANN with one hidden layer comprising 10 neurons gives the best fitting with the experimental data, which made it possible to predict total acceptance with acceptable MAPE (27) and correlation coefficients (0.96). Furthermore, high prediction accuracy ($R = 0.97$) was obtained by applying ANN models in predicting the rejection of neutral organic compounds by polyamide NF and reverse osmosis (RO) membranes [15].

3.2. ANFIS results

The ANFIS network parameters such as the type and number of MF and epochs have been varied to obtain best results in terms of model validation. Eighteen data points were used for training the system to predict the permeate flux and NaCl rejection (%). One-hundred neural nets learning epochs were used to get a low error of training. A plot of the training error and the number of epochs is shown in Fig. 8. ANFIS training was completed after two epochs (very fast training). The final fuzzy inference system that predicts the permeate flux and NaCl rejection (%) is shown in Fig. 9. Two Gaussian type MFs for each input (three inputs) resulted in high accurate prediction results.

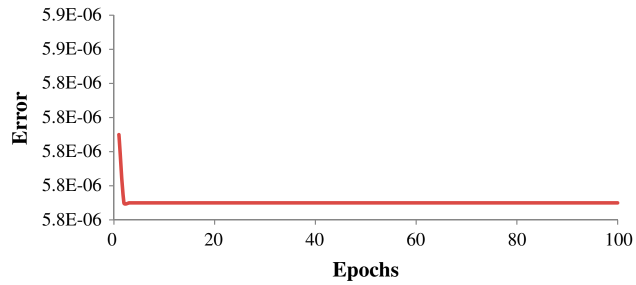

Fig. 8. ANFIS training error as a function of the number of epochs.

The permeate flux and NaCl rejection (%) values vs. ANFIS predictions for train data points are shown in Figs. 10 and 11. It can be seen that an excellent agreement between the predicted and experimental data was achieved. A comparison between the experimental and ANFIS predicted permeate flux and NaCl rejection (%) after training for unseen data is shown in Figs. 12 and 13. It can be seen that the system was well trained to model the permeate flux and NaCl rejection (%) of waste brine NF. The calculated correlation coefficient values for estimation of permeate flux and NaCl rejection (%) were 0.96 and 094, respectively, which show high correlation between predicted and experimental values. To quantify the agreement between the actual and predicted permeate flux

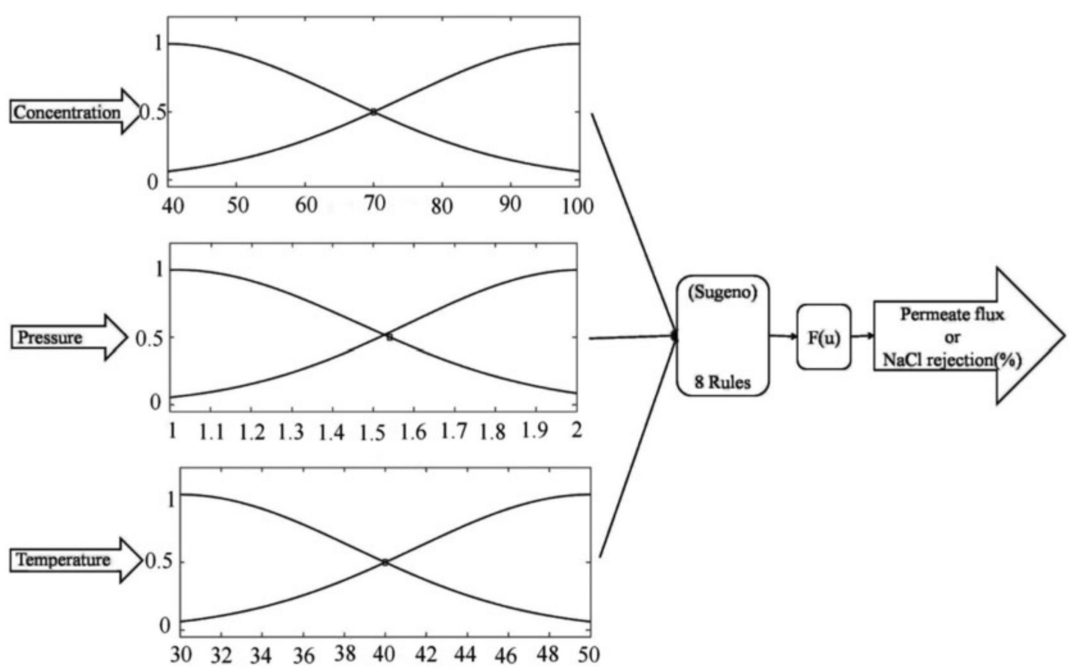

Fig. 9. The final ANFIS architecture for predicting the average permeate flux and NaCl rejection (%) of NF, with three inputs, two MFs for each input, constructed eight rules and one output for each model.

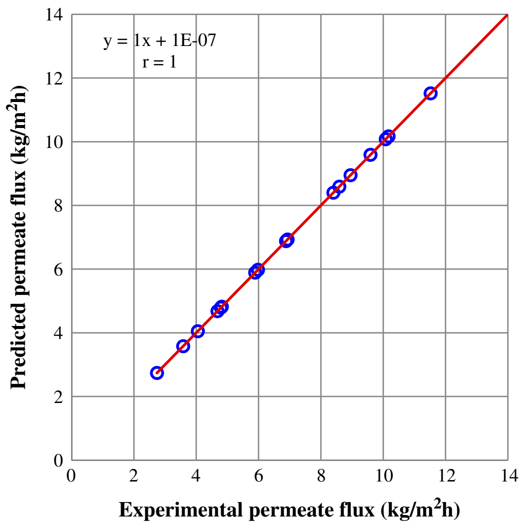

Fig. 10. Permeate flux measurements vs. ANFIS predictions for the training/testing data-set.

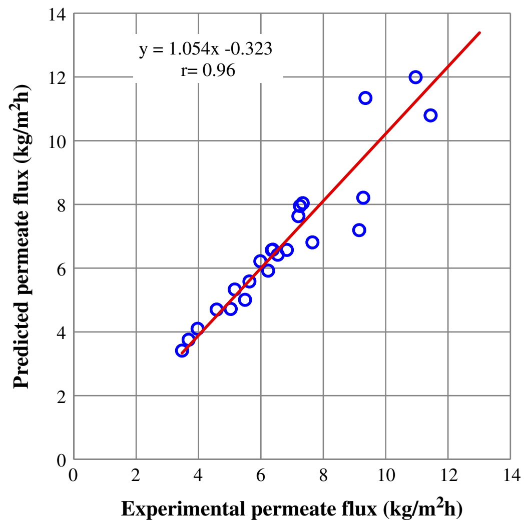

Fig. 12. Experimental (validating data-set) vs. predicted values for permeate flux of NF treatment of waste brine by optimum ANFIS configuration.

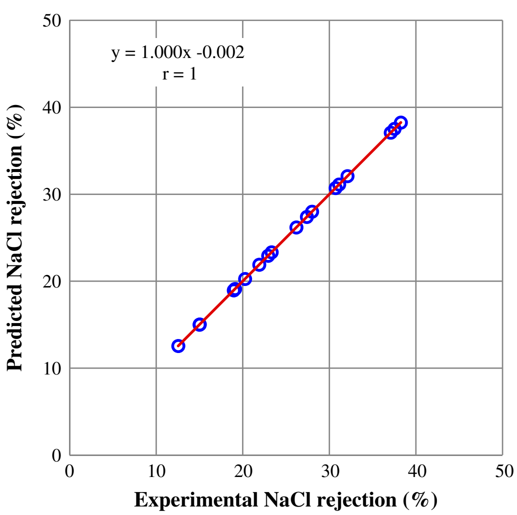

Fig. 11. NaCl rejection measurements vs. ANFIS predictions for the training/testing data-set.

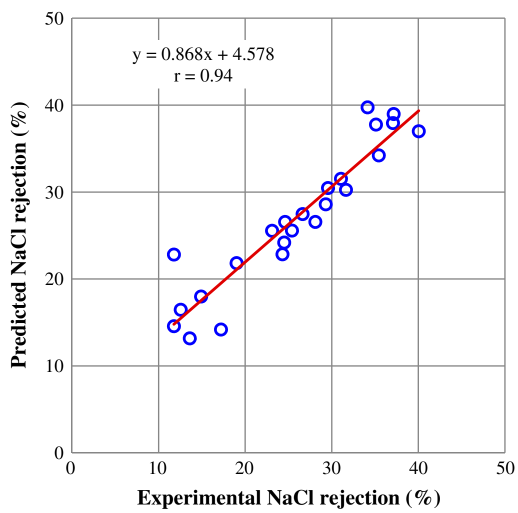

Fig. 13. Experimental (validating data-set) vs. predicted values for NaCl rejection (%) of NF treatment of waste brine by optimum ANFIS configuration.

values, linear regression was used to fit a line to the predicted target data-set. It can be seen that the obtained best line has a slope of 1.054 and an intercept of −0.323, which is very close to the perfect prediction.

The ANN and ANFIS models can be applied either separately as stand-alone modules or as an addition to the existing conventional mathematical models. In comparison to the conventional models, the ANFIS model will able to predict and optimize system performance faster and deliver better results in many instances [9,16].

The results obtained by ANN and ANFIS models are compared with experimental values to assess the efficiency of these models. Table 2 shows the data points used for system's validation along with the experimental and predicted values of permeate flux and NaCl rejection (%). However, in the case of developing a model, ANFIS model performs better than ANN model in predicting target output because ANFIS can deal with the values of a variable beyond the data range using MFs [9,16,29,31].

ANN modeling offers great advantage on improving the performance of ultrafiltration process by accounting the effects of different variables, i.e. feed

Table 2
Comparison of predicted and experimental values (validating data) for average permeate flux and NaCl rejection (%) of waste brine NF by optimum ANN and ANFIS configurations

Concentration (g/l)	Pressure (MPa)	Temperature (°C)	Experimental permeate flux (kg/m^2 h)	Predicted permeate flux (kg/m^2 h)		Experimental NaCl rejection (%)	Predicted NaCl rejection (%)	
				ANN	ANFIS		ANN	ANFIS
40	1.00	30	3.97	4.29	4.10	40.04	32.51	37.01
40	1.00	40	5.16	6.68	5.33	29.30	32.92	28.60
40	1.25	40	6.83	7.81	6.57	31.63	33.05	30.27
40	1.50	30	5.98	5.38	6.22	37.06	36.68	37.95
40	1.50	50	10.96	11.07	11.99	26.62	26.12	27.48
40	1.75	30	7.20	5.67	7.63	35.10	37.49	37.78
40	1.75	50	13.01	12.33	14.37	29.56	30.98	30.47
40	2.00	40	11.44	8.62	10.80	31.06	30.32	31.53
60	1.00	40	5.49	5.31	5.01	25.40	26.88	25.58
60	1.50	30	5.63	5.05	5.58	34.13	33.13	39.76
60	1.50	40	9.15	7.42	7.19	24.60	26.41	26.57
60	1.75	30	6.54	5.45	6.42	37.14	35.87	38.99
60	1.75	40	9.28	7.93	8.21	28.10	26.58	26.58
80	1.00	40	3.68	4.78	3.75	11.76	26.28	22.82
80	1.25	30	3.47	3.65	3.41	14.90	21.53	17.99
80	1.50	30	5.03	4.17	4.72	24.50	24.29	24.20
80	1.75	40	7.34	7.06	8.04	13.59	21.44	13.18
80	1.75	50	9.35	10.67	11.34	11.76	18.66	14.58
80	2.00	30	6.35	4.96	6.57	35.42	31.23	34.23
100	1.25	40	4.58	5.28	4.70	23.10	26.66	25.56
100	1.25	50	6.23	6.53	5.92	17.20	16.73	14.20
100	1.50	50	7.25	8.23	7.95	12.55	17.97	16.47
100	1.75	40	6.38	6.71	6.58	19.00	22.62	21.83
100	2.00	40	7.65	6.81	6.81	24.30	19.93	22.85

properties, transmembrane pressure, and membrane pore size on filtrate volume as the main output of the filtration process. ANN modeling of ultrafiltration may be an alternative to previously proposed empirical and semiempirical models [33].

4. Conclusion

ANN and ANFIS models do not require the prior knowledge of the relationship between the input and output variables because they can discover the relationship through successive training. Moreover, ANN models can predict several output variables at the same time, which is difficult in general regression methods. The application of ANN and ANFIS to the simulation of crossflow NF of waste brine from resin regeneration was investigated to predict the NaCl rejection and average flux (as outputs) vs. pressure, temperature, and concentration (as inputs). The ANN results suggested an optimum ANN model with 3/8/2 configuration could potentially be used to predict permeate flux and NaCl rejection with acceptable correlation coefficients (0.90 and 0.87, respectively). It was also found that ANFIS models with two Gaussian type MFs (gussmf) for all input variables and linear for output gives the best fitting with the experimental data, which made it possible to predict average permeate flux and NaCl rejection with low mean absolute percentage error (6.9 and 11.4, respectively) and high correlation coefficients (0.96 and 0.94, respectively). The results indicate that both ANN and ANFIS models can give good predictions of flux and NaCl rejection (%). However, the ANFIS model performs better than ANN model. Therefore, this method can be applied to relevant NF projects with satisfactory results.

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