Comparative Study of Artificial Neural Networks (ANN) and Statistical Methods for Predicting the Performance of Ultrafiltration Process in the Milk Industry

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ABSTRACT: *Milk ultrafiltration is a membrane process, which is highly complex innature. The cost effectiveness of the process depends heavily on the flux permeate and the total hydraulic resistance of the membrane. In this work, a comparative study for the prediction of the performance of milk ultrafiltration with ANN and statistical method has been carried out. The result reveals that both methods carry out the prediction with a high degree of accuracy. However, the statistical method, contrary to neural nets, is both costly and time consuming and the accuracy of the data are also in doubt, as the operating conditions are not consistent throughout each of the test runs. The result also reveals that there is a good agreement between the predicted fluxes permeates and the total resistances of this work with the actual values. The findings of this study also shows that the artificial neural nets technique can be applied as a powerful tool and a cost and time effective way in predicting and assessing the performance of milk ultrafiltration process.*

KEY WORDS: *Milk ultrafiltration, Artificial neural networks, Statistical methods, Permeate flux, Hydraulic resistances.*

INTRODUCTION

Ultrafiltration process is widely used in the food, pharmaceutical, polymer, biotechnology industries and purification plants. It is the process of separation of heavy molecules solutes in a light solvent or a suspension of colloidal substance into two streams of different concentration. This is accomplished using a porous membrane subjected to a hydrodynamic pressure difference as a driving force. The separation are carried

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out according to the sizes and the two most widely used apparatus in the field are spiral wound module and the flat plate [1, 2]. The flow of feed stream in the process is crossways and alongside the membrane. Although the process is simple, it does not need a high pressure or temperature and low cost of its energy consumption [3], different factors causes the process to be less compatible with the rival techniques (i.e., the reduction of the fluxes). The efficiency and the cost of the process are highly dependent upon the movements of the permeate through the membrane and the total membrane resistances [4], which in turn depends on different factors. The type of the membrane, process parameters (i.e., pressure, temperature, and the feed flow rate) and physical and chemical properties of the fluid are the main decisive factors affecting stream fluxes, concentrations and the total membrane resistances [5]. The main limitation on the practical aspects of the UF process usage is the drop in efficiency of the membrane due to the gel polarization and fouling phenomena. UF milk fouling is a complex phenomenon and still posing problems and research is still being carried out in this field. Owing to sedimentation of elements on the surface and inside the membrane, the effective usage period of membrane will be reduced and consequently the cost of the cleaning will raise accordingly [6]. In fact, through the exclusion of the membrane fouling, one can increase the efficiency of the UF process. The cleaning operation takes about 2-3 hours daily in the industry. Therefore, the understanding of the phenomenon requires modeling and the simulation of the membrane process for optimization purposes, which can have a great advantage both from economical and practical point of view.

In this research, two methods that are proper substitute for the physical and phenomenological modeling of the process and in addition a statistical method have been chosen to predict the efficiency of the UF process. This method is both time consuming and expensive. Meanwhile this leads to a simple calculation of the membrane area though the reliability of the result is questionable, because the conditions are not the same in all stages of the process. A precise estimation of the membrane area can be accomplished by using the equations describing the dependence of permeate flux and fouling on the process variables. In fact, there have been some theoretical approaches to predict the ultrafiltration performance of colloidal solutions (e.g. milk). These are based on some models such as mass transfer model (film theory), gel-polarization model, osmotic pressure model, boundary layer-adsorption model, Brownian diffusion model, shear-induced diffusion model, inertial lift model and surface transport model [4]. In addition to the complexity of mathematical equations involved, each of these models has a number of limitations:

- They demand some experimental data for determining the input parameters. Perhaps this is always possible in practice, but the equipment required are especially sensitive instruments that might not be readily available.
- None of these can describe the full flux-time behavior of process; they often predict the steady or pseudo-steady-state flux.
- Each one has been shown to be valid for certain feeds under special conditions.

Therefore, there is always a need for alternative methods of predicting membrane process performance. One of these methods is Artificial Neural Networks (ANNs). Neural networking involves algorithms under which information is accumulated in programmed objects that are capable of learning through much iteration using simulated or real data. This form of artificial intelligence can handle problems for which relationships are less known compared with relatively highly structured expert systems or equation-based approaches. Neural net models cannot linear, polynomial and interactive terms without requiring the researcher to model them, but can include available theoretical or empirical knowledge about the process. Therefore, it is capable of signal processing, modeling, time series forecasting, classification and recognition [7]. The neural network model has also been used for obtaining an estimation of the permeate flux and resistance during reverse osmosis of the ethanol and acetic acid and ultrafiltration of the pulp bleach plant effluent [8]. The ANNs predictions compared with the finely porous mass transfer model [9]. In colloidal systems such as milk, physical and chemical properties (i.e., pH and fat milk) have a great effect on the behavior and the efficiency of the UF process owing to molecular interactions. Furthermore, the hydrodynamic factors such as transient membrane pressure (TMP), temperature and the pore size of the membrane (MWCO)(1) have a

(1) Molecular Weight Cut Off

substantial effect on the process efficiency. Therefore, in this work parameters such as temperature, TMP, fat milk, pH, time and MWCO as an inputs and the permeate flux and the total hydraulic resistance of the membrane as an outputs in predicting and assessing the performance of milk ultrafiltration process, using the statistical and ANN methods.

THEORETICAL

A neural network is by definition: a system of simple processing elements, called neurons, which are connected to a network by a set of weights (Fig. 1). The network is determined by the architecture of the network, the magnitude of the weights and the processing element's mode of operation. The neuron is a processing element that takes a number of inputs (p), weights them (w), sums them up, adds a bias (b) and uses the result as the argument for a singular valued function, the transfer function (f), which results in the neurons output (a).

The most common networks are constructed by ordering the neurons in layers, letting each neuron in a layer take as input only the outputs of neurons in the previous layer or external inputs. To determine the weight values, a set of examples is needed of the output relation to the inputs. Therefore, a set of data was produced describing the whole operating range of the system. The knowledge of the neural network is encoded in the values of its weights. The task of determining the weights from these examples is called training and is basically a conventional estimation problem. For this purpose, the back-propagation strategy has become the most frequently, and here, used method that tends to give reasonable answers. Standard back-propagation is a gradient descent in which the following relation modifies the network weights:

$$w_{ij}(n+1) = w_{ij}(n) + \eta \cdot \delta_i(n) \cdot x_i(n) \quad (1)$$

where $w_{ij}(n+1)$ is the weight of i to j element in (n+1)th step and $w_{ij}(n)$ are same as weight in nth step. Local error $\delta_i(n)$ is evaluated from $e_i(n)$ where n is step size and η is the learning rate and is equal to one [10]. The local error can be evaluated from the following relation:

$$e_i(n) = d_i(n) - y_i(n) \quad (2)$$

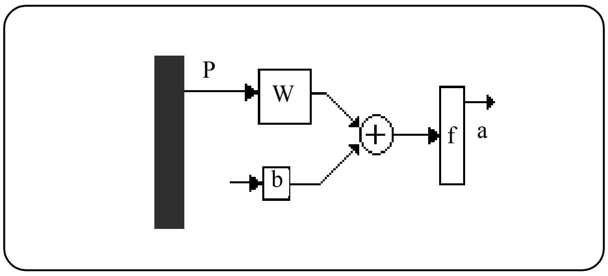

Fig. 1: The architecture of the neural network.

The term back-propagation refers to the manner in which the gradient is computed for non-linear multiple-layer networks. The typical performance function that is used for training feed forward neural networks is the mean sum of squares of the network errors between the network outputs and the target outputs [11]. In this work the batch gradient decent with momentum algorithm [12] was used as the training function. The momentum algorithm is development state of the gradient decent that weights learning that are obtained from the following relation:

$$w_{ij}(n+1) = w_{ij}(n) + \eta\delta_i(n) \cdot x_i(n) + \alpha \cdot \left(w_{ij}(n) - w_{ij}(n-1)\right) \quad (3)$$

where α is the momentum coefficient which its value ranges between 0.1 to 0.9. Equation (3) and other training functions usually give good results in neural network modeling of milk ultrafiltration process. The performance of the neural network model evaluated with the root mean square error (RMSE) and determination coefficient (R^2) between the modeled output and measures of the training data set, can be computed from the following relations:

$$R^2 = 1 - \frac{\sum_P \left(X_{obs} - X_{est}\right)^2}{\sum_P \left(X_{pred} - \overline{X}_{obs}\right)^2} \quad (4)$$

$$RMSE = \sqrt{\frac{\sum_P \left(X_{obs} - X_{est}\right)^2}{N}} \quad (5)$$

where X_{obs}, X_{est} are experimental and estimated values, respectively, and N is the number of data.

When the RMSE is at the minimum and R^2 is high (i.e., ≥ 0.8), a model can be judged with a high degree of accuracy [13]. Secondly, the comparison between the modeled output and the measured output must heuristically be reviewed. These methods were occasionally used in neural network model validation [14, 15]. Before the best model was found, a trial and error process was followed where different inputs and inputs-combinations were tested and the best input combination was selected. Hereby all reasonable combinations of input parameters were validated. Finally, the architecture of the neural network model was optimized by applying different amounts (1–10) of hidden neurons. When the increase of hidden neurons did not improve the model to any further extent, the model with the smallest amount and maximum performance was chosen as the best model. The choice of a specific class of networks for the simulation of a non-linear and complex map depends on a variety of factors such as the accuracy desired and the prior information concerning the input–output pairs. The most popular ANN is the feed forward multi-layer perceptron, where the neurons are arranged into an input layer, one or more hidden layers, and an output layer. Only one hidden layer was used in this study because of the proven non-linear approximation capabilities of multi-layered feed forward perceptron network for an arbitrary degree of accuracy [16]. 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. Generally, the transfer functions are sigmoidal function, hyperbolic tangent and linear function, of which the most widely used for non-linear relationship is the sigmoidal function [17]. The general form of this function is given as follows:

$$y_J = f(x_J) = \frac{1}{1+e^{-x_J}} \tag{6}$$

This sigmoid function maps input into output in a range between 0 and 1, distributed as an S-shaped curve, so the input and output data should be scaled to the same range as the transfer function used. Normalization of inputs leads to avoidance of numerical overflows due to very large or very small weights [17]. Therefore, data are normalized by the following relationship:

$$V_n = (1-\Delta_U-\Delta_L)\frac{V-V_{min}}{V_{max}-V_{min}}+\Delta_L \tag{7}$$

where V_n is the normalized value of V. The V_{max} and V_{min} are the minimum and maximum values of V, respectively. From experience, the authors have found that a better fit will be achieved if Δ_U and Δ_L (small margins) are kept a value of 0.05 [9]. In this work, the software that was adopted for the ANNs modeling was Matlab Toolbox version 7.0.

Total hydraulic resistance and permeate flux

By assuming that the osmotic pressures are small, the total hydraulic resistance [Rt] can be expressed by Darcy's law [3]:

$$Rt = \frac{TMP}{\mu_P \cdot J_P} \tag{8}$$

and the permeate flux by [18]:

$$J_p = \frac{TMP}{\mu_P \cdot Rt} \tag{9}$$

where μ_P is the permeate viscosity, Jp the permeate flux, and TMP the transmembrane pressure that can be calculated from the following equation:

$$TMP = \frac{P_I + P_o}{2} - P_P \tag{10}$$

where P_I , P_o and P_P are inlet, outlet and permeate pressures, respectively.

EXPERIMENTAL SET-UPS AND PROCEDURE

In this research, a membrane pilot plant from Biocon Company has been utilized for the data acquisition purposes. This apparatus is comprised from a feed tank, centrifugal pump, flow meter, membrane module, two pressure indicator, tubular heat exchanger, digital thermometer and a control valve (Fig. 2) which its specification are highlighted in table 1. In order to regulate the operating conditions for each test run, some distilled water was first introduced to the system for 10 minutes to check some parameters for the accuracy of the data. Then to run the experiment, the sample milk powder (with a constant milk fat of 8.5%) and hot water at 50 ^oC was mixed and added to the 12 liters feed tank at a constant rate of 15 lit/min throughout the experiment and letting the operation to be continued for 30 minutes. At the end, cleaning operation is carried out according to the instruction manual (i.e., cleaning-in-place or CIP). Care must be taken to cease the washing cycle when the difference between the exit and inlet

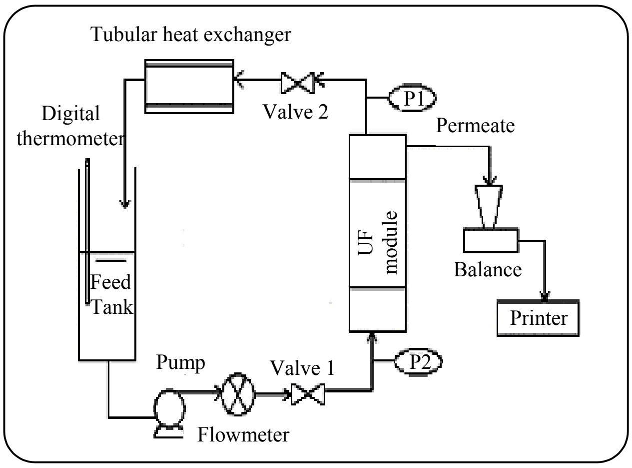

Fig. 2: Schematic flow diagram of a UF pilot plant.

Table 1: Specification of UF pilot plant.

Membrane material	Poly sulfone amid
Membrane module	Spiral wound
Membrane effective surface area	0.33 m^2
MWCO	20 kilo Dalton
Permissible pressure range	0.5-3 atm
Permissible temperature range	5-55 °C
Permissible pH range	2-11

water fluxes are less than 2-3 percent. Otherwise, the fouling has not been removed and the washing process must be repeated.

In this work, the affect of different parameters such as the transient membrane pressure (TMP), operating temperature, fat milk and pH on the flux permeate in m/s (J_p) and the total resistances in m^{-1} (R_t) has been analyzed (table 2). To obtain this objective, a total of sixteen-test run have been carried out and during each run the feed flow rate and concentration of the sample milk fat are regulated and kept constant using pasteurized and homogenized cream (28-30% fat). In order to measure the fat milk, a device called Lactostar from Funko Gerber Company was used and the measurement was repeated for three times at 25 °C. Furthermore, for regulating the milk pH an amount of 1% normal lactic acid have been used. To measure the sample milk and washing solution pH, a pH meter called Jenway (model 3010) was also adopted in this work.

STATISTICAL MODEL

Although some research has been carried out on theapplication of the milk UF [19, 20], but they have only dedicated their work on the hydrodynamics, the membrane type, configuration and its efficiency. In this work experimental data have been analyzed using two different software (i.e., Excel and Matlab) and the permeate flux and the total resistances have been predicted for different temperatures and plotted versus the time (Figs. 3 and 4).

The accuracy of the prediction with respect to correlation coefficient index (R^2), standard deviation, statistical mean have been presented in table 3. As the table 3 demonstrates, the accuracy of the predictions is quite high.

NEURAL NETWORKS MODELING

In this research, the effectiveness of UF milk modeling has been assessed using a multi-layer perceptron (MLP) and neural nets software (i.e., Matlab), for learning purposes different structures have been adopted, and the results achieved are compared together. The structure of neural nets is constructed in a way that a weak prediction and the time learning process expenditure could substantially be reduced by lowering the number of hidden layers [16]. Furthermore, neural nets with different structures can also reduce the learning procedure and to converge the results in the lowest number of iteration and to obtain a better prediction for the new data. As a typical example, the results obtained from feed forward back propagation neural nets (or multi-layer perceptron) have been presented in Fig. 5. In this work, the MLP have made with 3 layers that 1 neurons in first layer, 10 neurons in second layer and 1 neurons in last layer. The momentum coefficient and learning rate are 0.7, 1, respectively.

The total numbers of data utilized in this work are about 2500. From this about 30 percent of the input-output data was selected in a random manner for training (number of data 860) and the rest of the data were used to test (number of data 1360) and cross validate (number of data 280) the outcome and assess the error resulted from it. The results reveal that the prediction accuracy from the

Table 2: Parameter range and operating conditions in this work.

Variables	Parameter range			
	Press. diff. (KPa)	Temp. (^oC)	Fat%	pH
TMP	51	40	0.090	6.53
	101.33	40	0.095	6.54
	152	40	0.090	6.54
	203	40	0.095	6.54
	253	40	0.090	6.54
Temperature	152	30	0.110	6.57
	152	40	0.090	6.54
	152	50	0.120	6.60
Fat%	152	40	0.090	6.54
	152	40	1.190	6.56
	152	40	2.400	6.6
	152	40	3.260	6.53
PH	152	30	2.400	6.67
	152	30	2.380	6.43
	152	30	2.390	6.25
	152	30	2.420	5.97
MWCO	101.33	40	0.095	6.62
	101.33	40	0.100	6.55
	101.33	40	0.090	6.59

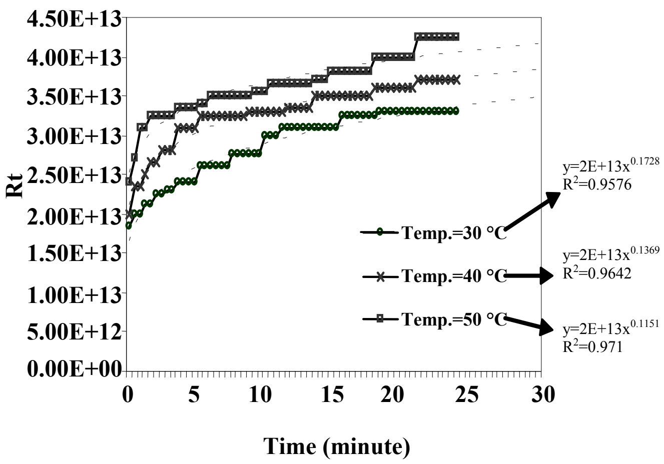

Fig. 3: R_t vs. time (TMP=152, Fat=0.1%, MWCO=20 and T=30 ^oC)

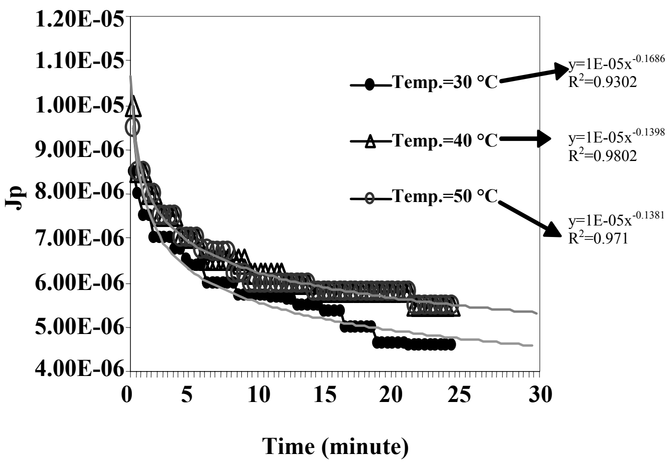

Fig. 4: J_p vs. time (TMP=152, Fat=0.1%, MWCO=20 and T=30 ^oC)

Table 3: Prediction accuracy from the statistical method.

		Jp (observed)	Jp (statistically)	Rt (observed)	Rt (statistically)
T=40 °C	Mean	5.6857E-06	6.35937E-06	3.61429E+13	3.10434E+13
	R^2	0.9802	0.9961	0.9442	0.8965
	RMSE	7.031E-07		5.10647E+12	
TMP=150 (Kpa)	Mean	5.7095E-06	6.21804E-06	5.40714E+13	5.01762E+13
	R^2	0.9739	0.9961	0.9766	0.9979
	RMSE	5.1513E-07		3.98463E+12	
FAT=3.3%	Mean	0.0000055	6.1303E-06	3.8181E+13	2.89843E+13
	R^2	0.9273	0.9963	0.9378	0.9977
	RMSE	6.3666E-07		9.23284E+12	
MWCO=20 (Kilo Dalton)	Mean	3.6286E-06	4.26825E-06	3.02952E+13	2.85866E+13
	R^2	0.969	0.996	0.891	0.9976
	RMSE	6.4242E-07		1.95407E+12	
pH=6.43	Mean	5.0667E-06	5.18966E-06	3.23333E+13	2.74965E+13
	R^2	0.9322	0.9964	0.9322	0.9976
	RMSE	1.3485E-07		4.84049E+12	

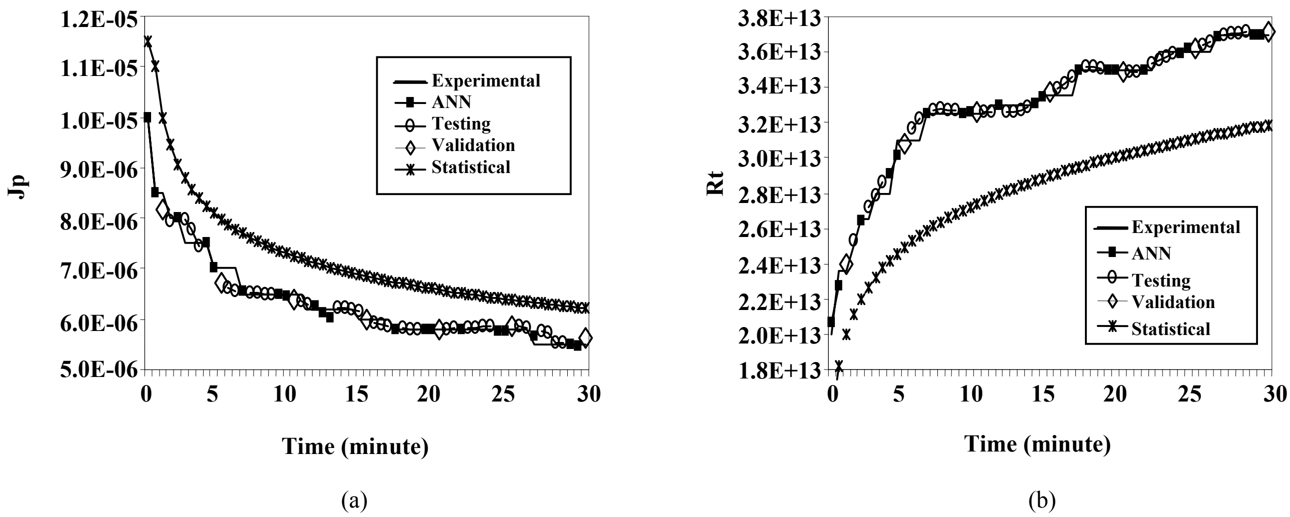

Fig. 5: Comparison of actual: (a) J_p and (b) R_t values with statistical and ANN methods at 40 °C (Training: Test: Validation = 20: 34: 7)

ANN method were quite high and a correlation coefficient of about one were obtained which shows an acceptable fitness through an appropriate training and test of the nets. The results obtained from nets and the experimental values at 40 °C show a high compatibility, as shown in Fig. 5. In addition, the correlation coefficient, the error resulted from the experimental and predicted values of the two methods are presented in table 4.

Comparative studies have been made between the findings of the present research and other workers in table 5. This comparison clearly shows that the extent of prediction of neural modeling are exceptionally well with respect to other parameters such as R-square and RMSE. In addition, even though it needs less data for learning processes in contrast to other models, it has a high ability for modeling the ultrafiltration process. Furthermore, comparison between the ANN, physical and statistical methods for modeling of ultrafiltration process reveals that ANN modeling (apart from its high degree of precision for prediction) has also a high ability in simulating the dynamic fluxes and total hydraulic resistances in the modeling of ultrafiltration process.

CONCLUSIONS

In colloidal systems such as milk, the physical and chemical properties such as pH and fat percentage have an immense influence on the system due to molecular interactions and consequently on the efficiency of the UF process. In addition, parameters such as temperature, transient membrane pressure and the extent of the pore sizes of the membrane has a huge affect on the hydrodynamics of the membrane and the effectiveness of the process.

In this research, two methods that are a proper substitute for the physical and phenomenological modeling of the process and in addition a statistical method have been chosen to predict the efficiency of the UF process. In addition, the permeate flux and the total hydraulic resistances obtained from the statistical method and the artificial neural nets technique were compared with the actual values. In the training of the nets, about 70 percent of the input-output data was selected in a random manner and the rest of the data were used to test and validate the outcome and assess the error resulted from it. The result also reveals that one can predict the efficiency of the UF process using ANN method with a high degree of accuracy that shows an acceptable fitness through an appropriate training and test of the nets. In addition, the results demonstrate that a simple alteration in the architecture of the nets can increase the scope of the vulnerability of the solution. On the other hand, the statistical method contrary to neural nets, is both costly and time consuming and the accuracy of the data are in doubt as the operating conditions are not consistent throughout each of the test runs. Therefore, the findings of this study reveal that the artificial neural nets technique can be applied as a powerful tool and a cost and time effective way in predicting and assessing the performance of milk ultrafiltration process. Furthermore, the modeling results exhibited that the dynamic behavior of permeate flux, total hydraulic resistance could be well-predicted using temperature and time as the input parameters. In addition, excellent agreement between experimental data and predicted values can be achieved by single hidden layer network and limited number of training points. As a result, the number of experimental tests that needed to be carried out on a pilot or large-scale plant is quite limited.

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Nomenclature

CR	Concentration ratio
FR	Filtration rate
I	Ionic strength
J	Permeate flux, ($m.sec^{-1}$)
J_{ini}	Initial flux value
Max	Maximum (logic operator)
Min	Minimum (logic operator)
MRD	Mean of relative deviations
MWCO	Molecular weight cut off, (kDa)
N_E	The number of experimental value
N_T	The number of training data points
N_Q	The number of querying data points
N_V	The number of validating data points
P	Pressure, (kPa)
PE	Prediction error
R	Total hydraulic resistance, (m^{-1}), rejection, (%)

Table 4: Correlation coefficient and error resulted for experimental values, statistical and ANN methods at 40 °C.

		Actual	Statistical	ANN
J_p	R^2	0.9774	1	0.9875
	RMSE		7.7E-7	7.9E-8
	Std	7.659E-7	9.11E-7	7.338E-7
R_t	R^2	0.9677	1	0.9864
	RMSE		4.19E+12	4.37E+11

Table 5: Comparative studies between the findings of present research and other workers.

Application	Input	Output	N_E	N_T	N_V	N_Q	N_T/N_E (%)	Accuracy	Ref.
RO of ethanol solution	P,T	R, J	60	24	36	-	40	MRD(J)=0.011 MRD(R)=0.075	[8]
UF of bleach plant effluent	P, V, CR	R, J	25	24	1	-	96	PE=0.10 MRD=0.02	[8]
MF of cane sugar syrup	TMP, V, t	R_T	-	6	-	-	-	R^2=0.98 γ =0.054	[21]
UF of BSA solution	pH, I	FR	254	24	230	-	9.45	Emax=0.1081 Eave=0.0213	[9]
UF of silica suspensions	I, TMP, t, Z	J	391	46	92	253	11.76	Emax=0.0302 Eave=0.056 RMSE=3.2E-6	[9]
UF of waste water	T, J_{ini}	J	541	-	-	-	-	Emax=0.0289 Eave=0.0137 RMSE=8.9E-6	[22]
UF of milk	TMP, pH, Fat, MWCO, t, T	J, R_T	2500	860	280	1360	34.4	Emax=0.0193 Eave=0.0043 RMSE(J)=7.9E-8 R^2(J)=o.9875 Std(J)=7.338E-7 RMSE(R_T)=4.37E11 R^2(R_T)=0.9864	This study

RMSE	Root mean square error
t	Time
T	Temperature
Temp	Temperature, (°C)
TMP	Transmembrane pressure, (kPa)
R^2	Determinate coefficient
Std	Standard deviations
V	Flow rate
Z	Zeta potential

Greek Letters

γ	The variation coefficient

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