doi.org/10.52568/000561/JCSP/43.02.2021 104

Prediction of Succinic Acid Extraction Efficiency in the Emulsion Liquid Membrane by using Machine Learning Techniques

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(Received on 21st April 2020, accepted in revised form 3rd December 2020)

Summary: This research aims to predict succinic acid concentration in the external phase during the emulsion liquid membrane process by using artificial neural networks along with a popular alternative method: k-nearest neighbor technique. The solute concentration values can be predicted by the proposed method without performing a great number of emulsion liquid membrane experiments. Several computer simulations were performed to demonstrate the success of the system. Simulation results showed that the estimated solute concentration values are very close to the achieved experimental results. The optimal conditions for emulsion liquid membrane were found to be: solvent kerosene, TOPO concentration (1%w/w), Amberlite LA-2 concentration (4%w/w), surfactant concentration (5%w/w), Na_2CO_3 concentration (5%w/v), modifier (decanol) concentration (2%w/w), mixing speed 300 rpm. The average accuracy percentages achieved by artificial neural network and k-nearest neighbor approaches were 88.75±1.94% and 90.2±1.2%, respectively.

Keywords: Extraction; Emulsion liquid membrane; Artificial neural network; k-nearest neighbors; Simulation.

Introduction

Monocarboxylic acids are byproducts of multiple industries including food, textile, pharmaceutics, andpetro chemistry. Succinic acid has received a great attention in many fieldsowing to its high production value and the ability to be used in numerous applications.Succinic acid was produced by the petrochemical process, before the development of fermentation processes for its production. However, there are certain restrictions, such as having higher purification costs and limited stock. Thus, the necessity of developing an efficient and cheaper method for succinic acid extraction and purification escalates significantly.Generally, precipitation; electro dialysis, distillation, and reactive extraction remain the basic method for carboxylic acid separation from aqueous solution involving fermentation broth [1]. Besides, the emulsion liquid membrane (ELM) process has yielded promising results and can be potentially used for applications. The ELM process can be referred as a new separation technique, which is superior to the conventional processes. Liquid membranes have great potential for applications in biotechnology and recovery from fermentation products. A liquid membrane is an insoluble liquid, usually, an organic solvent, which is selective for a solute. Also, a liquid membrane separates two aqueous phases, a feed phase, initially containing the desired solute, and a recovery or stripping phase into which the solute is extracted. ELMs combine simultaneous extraction and the stripping processes in a single operation, therefore reduce the solute concentration in the aqueous solution to very low levels [2-5].

ELM consists of an organophosphorus extractant tri-octyl phosphine oxide (TOPO) or secondary amine Amberlite LA-2 (the carriers), *n*-heptane, kerosene, toluene, and Escaid 100 (the solvents), Span 80 (the surfactant), decanol (the modifier), and Na_2CO_3 (the stripping agent). The extraction of solute using an ELM is determined by solute-extractant complex-forming ability. Extraction also depends on the type of extractant and diluent. Stripping phase concentration, mixing speed, extractant concentration, surfactant concentration, and modifier concentration are the crucial parameters ofthe ELM process.

The study aims to estimate succinic acid concentration in aqueous solutions during the ELM process by using two popular machine-learning methods. The early-developed mathematical models depicting the ELM procedure have some certain limitations. Thus, utilizing experimental models independent of hypothesizes and presumptions offers a breakthrough concept. Among these models, the most preferred one was the artificial neural network (ANN) approach, which has been extensively used in chemical engineering and modeling [6-21]. In a recent study, ANN was applied to the external phase during the ELM extraction process to predict nickel concentration. The training, validation, and test procedures of the back-propagation ANN model were achieved through batch laboratory analysis. The simulation results revealed that the ANN model provided promising results for predicting external phase concentration automatically [14]. In a study conducted by Kim et al., the k-nearest

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neighbor (k-NN) method was utilized to estimate the wastewater treatment plant (WWTP) that includes the influent flow rate and four different water qualities. The results of the study demonstrated that the k-NN method was favorable for estimating WWTP influent characteristics [22].

In the proposed study, ANN and k-NN techniques were investigated to determine external phase concentration during the ELM extraction process. Also, an s-fold cross-validation technique was developed to improve the performance of both methods. One of the goals of the study is to apply two of the most popular machine learning techniques to facilitate a chemical process. The ANN and k-NN methods make possible to predict solute concentration in the ELM experiments without performing several experiments, the main contribution of the proposed study. The most important contribution of the study is to provide the use of KNN method in chemical applications [22-27].

Experimental

Materials

The surfactant was commercially known as Span 80 (Fluka). Amberlite LA-2 and TOPO were the carriers (Sigma Aldrich). Diluents used a membrane phase were kerosene, toluene, *n*-heptane, octanol and Escaid 100 (ExxonMobil). Decanol was used as modifier (Sigma Aldrich). Na_2CO_3 was internal aqueous phases (Merck). Succinic acid (Merck) was used to prepare the feed solution.

Preparation of ELM

The liquid membrane phase (organic phase) contains a surfactant, an extractant, and a diluent. Water in oil (W/O) type emulsion was prepared, mixing with the organic membrane phase (50 mL) and internal aqueous phases (50 mL) at 2000 rpm by using a mechanical overhead stirrer in 250 mL beaker. It was made by drop wise addition of Na_2CO_3solutions to organic membrane phase. The solution is stirred continuously for 20 min to obtain a stable ELM. The external phase (feed phase) was a succinic acid solution. Water in oil in water (W/O/W) double emulsions were formed by mixing the feed phase with the (W/O) phase. The dispersed globules were typically about 1 mm in size. The mixing process was stopped at different periods, and the samples were collected from the aqueous phase. The extraction was measured by analyzing the succinic acid solution. The acid concentration was determined by using High-Performance Liquid Chromatography (HPLC, Shimadzu LC-20AD, and Hypersil C18 ODS Column) [28-30]

Experimental design

In this study, experimental parameters were diluent, extractant type, mixing speed, surfactant concentration, stripping phase concentration, and modifier concentration. The experimental parameters were given in Table-1.

The effect of mixing speed was studied in the range of 300-500 rpm. To form a stable liquid membrane, a sufficient amount of surfactant was required. An increase in surfactant concentration leads to larger diffusional resistance at the interface and increases viscosity of the membrane solution. Experiments were performed with the surfactant concentrations ranging from 3% to 8%. The extraction of acid was determined by the formation of acid amine complex. The combination of amines (Amberlite LA-2) and solvation extractant (TOPO) was studied for extraction. TOPO variation studies in range of 1-4% w/w, while Amberlite LA-2 in 1-5%w/w. As the extraction step occurs in the interface between the feed solution and the liquid membrane, the transport of acids requires a simultaneous back-extraction or stripping step at the opposite side of the membrane. In the stripping process, the extractant was regenerated, and the organic acid was stripped. Na_2CO_3 was used as the stripping solution, andconcentration was varied 5 to 15%w/v.

Table-1: Experimental cases.

Experimental Parameters							
1	2	3	4	5	6	7	8
Time (minute)	Extractant type (TOPO,%)	Extractant type (Amberlite LA-2,%)	Diluent Type	Surfactant concentration (Span80,%)	Mixing speed (rpm)	Modifier concentration (Decanol,%)	Stripping phase concentration (Na_2CO_3, %)
0	0	0	Kerosene	3	300	1	5
2	1	1	Toluene	5	400	2	10
5	2	3	*n*-Heptane	8	500	4	15
10	3	4	Octanol				
15	4	5	Escaid				
20			100				
30							

The optimum conditions obtained from the experiments were: membrane phase with surfactant Span 80 (5% w/w), the carrier Amberlite LA-2 (4% w/w), and TOPO (1%w/w), modifier decanol (2%w/w), the diluent kerosene, (88% w/w), Na_2CO_3 (5% w/v), the mixing speed 300 rpm and the extraction efficiency at these parameter settings would be 87.7% in 30 min. Extraction efficiency in optimum conditions was shown in Table-2.

Table-2: Extraction efficiency in optimum conditions.

Time	2	5	10	15	20	30
% Extraction	49.1	61.4	70.6	78.5	81.2	87.7

The 280 data points were obtained during the experiments. The extraction efficiency of succinic acid should ideally converge to its best value "1" and it can be expressed as follows:

$$\eta = 1 - \frac{c}{c_o} \quad (1)$$

Where c and c_o are the concentrations of succinic acid in the feed phase at elapsed time and time zero.

According to the equation, the extraction efficiency (η) must converge to one. In other words, C/C_o proportion must converge to zero to achieve the desired experimental performance.

A wide range of time-consuming and expensive experiments should be performed to achieve the ideal C/C_o level. Our system concentrated on predicting succinic acid extraction efficiency during the external phase by exploiting ANN and k-NN techniques' decision ability without performing ELM experiments. For this purpose, a comprehensive system that utilized signal processes and machine learning techniques was designed. In the preprocessing level, adaptive quantization and labeling processes were applied to C/C_o proportions to assign the class labels. Then, an s-fold cross-validation technique was performed to improve the performance of ANN and k-NN techniques. The flow chart of the study was depicted in Fig.1.The main steps of the system were described in the following subsections.

Database Construction

The database was constructed by performing 280 ELM laboratory experiments. During these experiments, various types and proportions of constituents were evaluated. Some of these experiments were summarized in Table-3 to introduce the case. The obtained eight parameters were the inputs of the proposed system. The output of the system is the predicted C/C_o values. The size of the database matrix was 280x8.

Preprocessing

In the preprocessing step, measurements that are obtained by laboratory experiments were predisposed for consecutive steps having illustrated in Fig. 1. In this step, quantization and labeling procedures were applied to C/C_o values.

As discussed before, primary goal of the study was to predict C/C_o proportion of the ELM process, automatically. The C/C_o proportion must approach to zeroto achieve the desired extraction efficiency. Previously performed experiments revealed that the lower C/C_o percentages were more significant than the higher percentages. Thus, [0, 0.25) interval was emphasized and consequently [0, 0.25) interval was divided into four partitions as {0-0.1, 0.1-0.15, 0.15-0.2, and 0.2-0.25} for labeling. The remaining part, [0.25-1], was not divided into subintervals. This process can be considered as an adaptive quantization process in which the spans of significant subintervals are smaller than the others.

Table-3: The sample ELM experiments.

TrialNo	Solvent	Surfactant	Extractant		Feed phase concentration	Stripping phase	Mixing speed	Time	Results
		Span 80	TOPO	AmberliteLA2	(Succinic acid)	(Na_2CO)	(rpm)	(min)	C/Co(-)
1	Kerosene	0.05	0.01	0.04	0.02	0.05	300	0	1
12	Kerosene	0.05	0.01	0.04	0.02	0.05	400	15	0.230
30	Kerosene	0.05	0.00	0.05	0.02	0.05	300	2	0.564
108	Kerosene	0.05	0.04	0.00	0.02	0.05	300	5	0.875
217	Toluene	0.05	0.05	0.00	0.02	0.05	400	30	1
225	*n*-Heptane	0.05	0.05	0.00	0.02	0.05	400	0	1
233	Octanol	0.05	0.05	0.00	0.02	0.05	400	2	1
242	Escaid 100	0.05	0.05	0.00	0.02	0.05	400	10	0.783
275	Kerosene	0.05	0.05	0.00	0.02	0.05	500	2	0.661
291	Kerosene	0.05	0.05	0.00	0.02	0.10	400	10	0.207
306	Kerosene	0.03	0.05	0.00	0.02	0.05	400	15	0.570
317	Kerosene	0.08	0.05	0.00	0.02	0.05	400	2	0.836

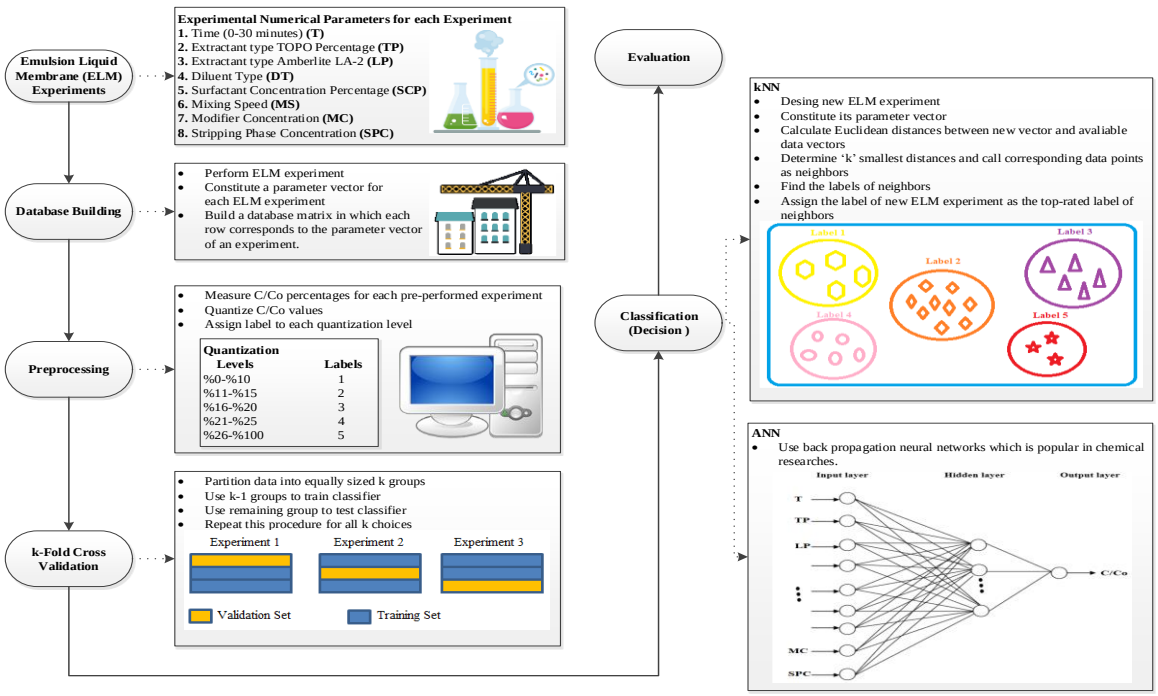

Fig. 1: The flow chart of the proposed study.

After quantization, all experimental C/C_o proportions were assigned to the five subintervals as {0-0.1, 0.1-0.15, 0.15-0.2, 0.2-0.25, and 0.25-1}, and a class label to the corresponding subinterval was given. At the end of the labeling process, five class labels were attained, as described in Table-4.

Table-4: Labeling process of the C/C_o values.

C/Co Percentages (%)	SubintervalLabel	
0-10	[0, 0.1)	1
10-15	[0.1, 0.15)	2
15-20	[0.15, 0.2)	3
20-25	[0.2, 0.25)	4
25-100	[0.25, 1]	5

Note that, at the output of the proposed study we aim to predict the labels given above. For example, if the predicted label were 1, the C/C_o proportion would be in [0-0.1) interval. The smaller C/C_o proportion denotes the more successful succinic acid extraction efficiency. The successes of the proposed system were evaluated by comparing the experimental results with the predicted results that areobtained bythe proposed system.

S-fold cross-validation

The *s*-fold cross-validation method has been extensively used in many classification studies, recently. The most important advantage of the cross-validation method is that it provides an increase in the performance of the classifiers, especially for relatively small databases.

Principally, in the *s*-fold process, available data is partitioned into s equally sized groups. Then,(*s-1*) of the groups are used to train a set of models. The remaining group is evaluated to test the classifier. This procedure is repeated for all *s* choices, as given in Fig. 2 with yellow blocks. The final performance of the classifier is the average performance scores of the *s* runs (experiments). If the number of runs is equal to the number of data, the resulted cross-validation process is called the leave-one-out technique. Increasing the number of runs makes the cross-validation method computationally complex [22].

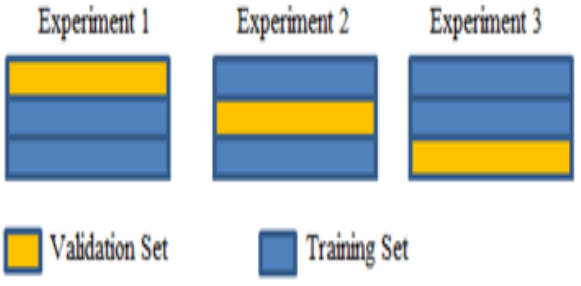

Fig. 2: S-fold cross-validation technique for *s*=3.

In our study, 3-fold cross-validation process was utilized to provide a performance augmentation

for ANN and k-NN classifiers. The 187 data were used for training, and the remaining 93 data were used for testing in each fold. Performance scores of each run were averaged to calculate the final performance of the classifier.

Decision/ Classification

In this step, the target was to reach a final decision about the C/C_o label. For this purpose, ANN and k-NN classifiers were preferred. In the following subsections, information about ANN and k-NN techniques was provided to better understand the study.

Artificial neural networks

The brain is a highly complex information-processing system. A neural network (NN) isa machine that imitates the human brain. NNs employ a massive interconnection of simple computing cells known as "neurons" or "processing units". NN performs two main tasks similar to the brain: 1) through a learning process, knowledge is attained from the network's environment, 2) to store the attained knowledge, synaptic weights that are connected to the neurons are utilized. During the learning process, synaptic weights of the network are updated adaptively to perform the given task.

A neuron is the fundamental element of NN, having themain task of information processing. The neuron model is illustrated in Fig. 3. The basic elements of a neural model can be summarized as follows:

a) A set of synapses or connection links: In Fig. 3, the input of synapse j is denoted as x_j. The synaptic weight of k.th neuron w_{kj} is multiplied with corresponding input and connected to neuron k. Unlike the weight of a synapse in the brain, synaptic weight of an artificial neuron can both take positive and negative values.
b) Adder: The weighted input signals are summed with an adder. The output of the adder is denoted by the u_k that is the linear combination of input signals.
c) Activation function: This function squashes the output amplitude of a neuron. The output of a neuron must be in acceptable limits.

Also, b_k shows the externally applied bias that can take negative or positive values. The sign of b_k has the effect of increasing or decreasing the net input of the activation function. With the above discussion, the output of neuron k can be expressed with Equations 2 and 3.

$$y_k = \vartheta(u_k + b_k) \quad (2)$$

where,

$$u_k = \sum_{j=1}^{m} w_{kj} x_j \quad (3)$$

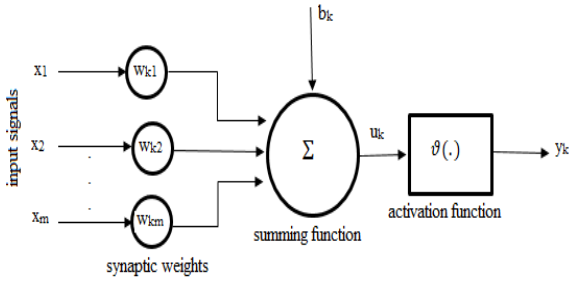

Fig. 3: The nonlinear model of neuron k.

In this study, an efficient back-propagation algorithm was applied to the given problem. This algorithm is based on computing the first-order derivatives of the cost function that is expressed as a function of synaptic weights and a bias term. The learning process of network targets minimizing error function, which is denoted as the difference between the desired output and the actual network [27].

k- neares tneighbor (k-NN) method

K-NN method is a supervised classifying algorithm that is widely preferred in many pattern recognition applications. In this method, learning is implemented by processing training data points. The crux of the algorithm is to assign a class label to new data point by calculating the distance between training data points and the new data point. Euclidean, Minkowski, Manhattan, and Dilca distances are the most preferred metrics.

In the k-NN method, the k value determines the number of neighbors required to decide the class of new data. k is generally chosen from odd numbers. In our study,the Euclidean distance was used. The main steps of k-NN can be summarized as follows:

a) Determine the k value.
b) By using Equation (4), calculate the Euclidean distances between new data point and cluster points;
$$\sqrt{\sum_{i=1}^{n}(X_i - Y_i)^2} \quad (4)$$
where X is the point in the cluster, and Y is the new data point.

c) According to the distance values obtained in (b), the smallest *k* distance is determined, and corresponding points are referred to as neighbors.
d) The class labels of neighbors are noted. The top-rated label is assigned as the label of the new data point.

In Fig. 4 the fundamental context of k-NN was illustrated. The k-NN algorithm is simple to implement and gives acceptable results in many applications. Furthermore, it provides reliable results even the data is distorted with noise [26, 27].

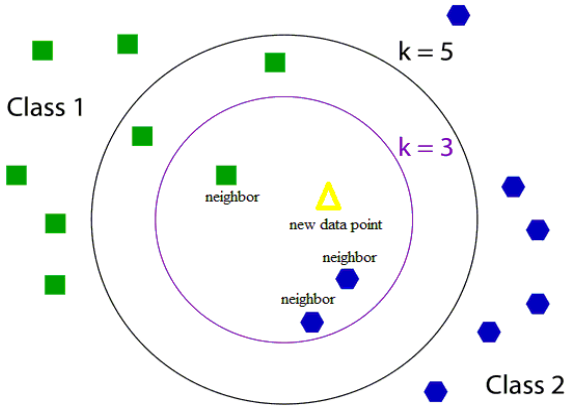

Fig. 4: A simple k-NN method illustration for only two classes.

Results and Discussion

As discussed before, the goal of the proposed study was to predict C/C_o labels without performing batch ELM experiments. In this section, several computer simulations were carried out to evaluate the performance of the study. The *accuracy* measurement was calculated to verify the efficiency of the proposed system that was illustrated in Fig. 1. Accuracy can be defined as the ratio of correct predictions to the number of all predictions. Since 3-fold cross-validation was used, three accuracy values were calculated for each validation set.

In Table-5, the accuracy percentages of both ANN and k-NN classifiers were listed. The number of neighbors in the k-NN method was changed as 3,5, and 7 to determine the best *k* value. Furthermore, the accuracy percentages for each run of 3-fold cross-validation processes were remarked as 1st run, 2nd run, and 3rd run, respectively. The last row of the table denotes the average accuracy percentages of the corresponding column.

Table-5: Accuracy percentages of k-NN and ANN.

	k-NN			
Classifiers	**k=3**	**k=5**	**k=7**	**ANN**
1. Run	**80.64**	**89.09**	**84.95**	**90.32**
2. Run	**85.10**	**90.29**	**85.11**	**87.76**
3. Run	**89.24**	**91.24**	**92.47**	**88.17**
Average Accuracy	**84.99**	**90.20**	**87.51**	**88.75**

According to the table, it is clear that the higher accuracy was achieved by k-NN technique (*k*=5). Consequently, average accuracy values of k-NN and ANN techniques were very close to each other; 90.20±1.20% and 88.75±1.94%, respectively. Thus, the k-NN method can be used to predict the extraction efficiency of ELM experiments with low computational complexity.

Furthermore, Table-6 was prepared to demonstrate the number of true and false predictions for each classifier to provide a comprehensive understanding of the classifier performance. In the table, *k* denotes the number of neighbors for the k-NN method. True prediction (TP) was calculated by multiplying the number of test data (93) by average accuracy percentage, and false prediction (FP) was 93-TP.

Table-6: True and false prediction numbers for classifiers.

Number of Neighbors:	**k-NN**	**ANN**
k=3	**TP: 79 / FP:14**	**TP:80 / FP:13**
k=5	**TP: 84/ FP:9**	**TP: 83 / FP:10**
k=7	**TP: 81 / FP: 12**	**TP:78 / FP:15**

*TP: Number of true predictions, FP: Number of false predictions

The performance of the proposed system can be illustrated through an alternative approach. In this manner, the Receiver Operating Characteristic (ROC) curves of classifiers were demonstrated. A ROC curve displays the true prediction rate against the false prediction rate for a varying decision threshold when a single output is used. In Fig.5, the ROC curve of the system was given. To plotthe ROC curve, labels 1, 2, 3, and 4 were assumed as "success," and label 5 was assumed as “fail” regardingthe extraction efficiencies.

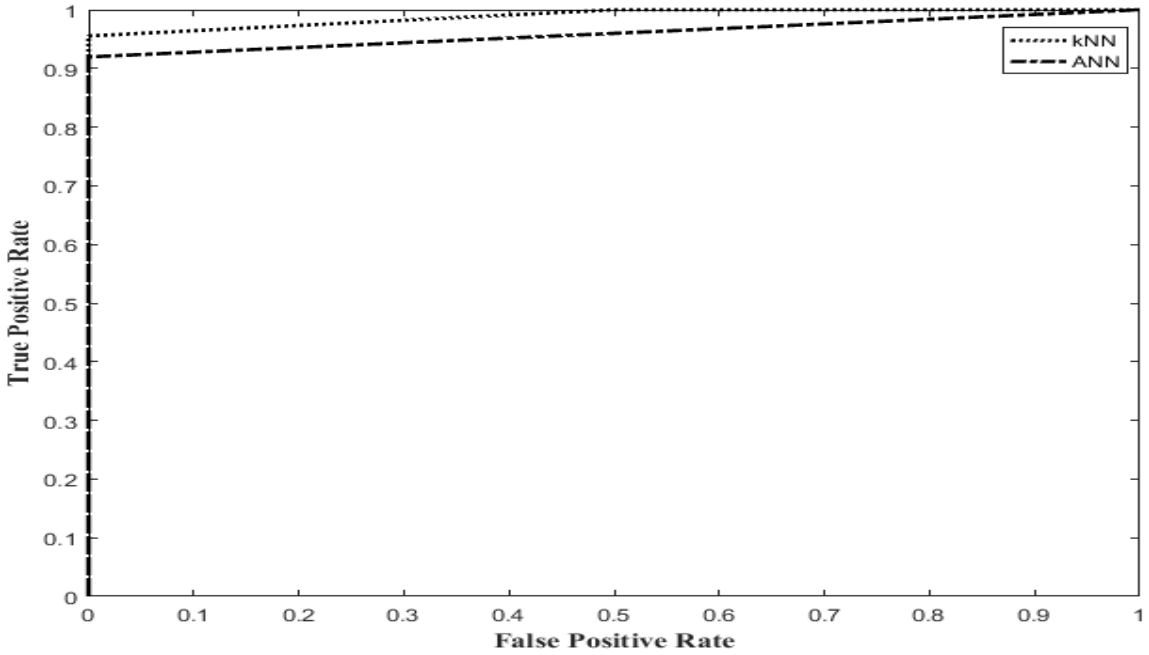

Fig. 5: ROC curve for used classifiers.

The ROC curve, which is close to, the upper left corner, indicates that the true predictions rate is higher than the false positives rate. According to the ROC curves, k-NN and ANN methods provided reliable results for the given classification problem.

In addition to the ROC curves, the learning curve of NN was illustrated in Fig. 6 to interpret the performance of the used network. Learning curve plots the Mean Square Error (MSE) variation against the number of epochs, which is considered as an iteration number to process all input data. A NN learns in every epoch. When the same inputs are applied to network several times, networks train themselves to provide a smaller MSE. The number of epochs should be increased until the accomplishment of complete network training [31]. The learning curve is expected to decrease monotonically when the epoch number increases. In Fig. 6, it was observed that the MSE value decreased with the increasing epoch number. In addition, the error rates of training and validation sets were very close to each other. So, the learning algorithm had a good fitting, and the used network performed the given classification task successfully.

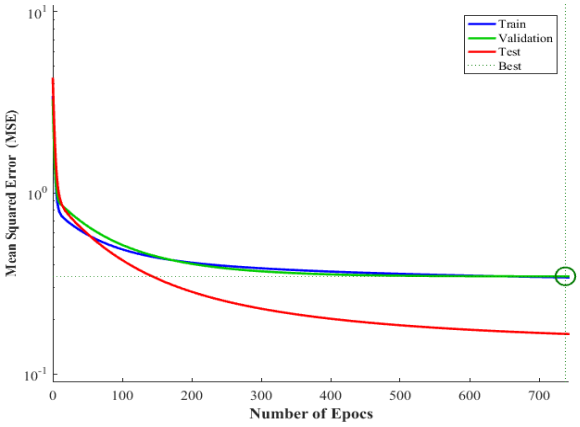

Fig. 6: The learning curve of the neural network.

Finally, in Fig. 7, predicted C/C_0 values were compared with the actual values that were obtained in the experiments. According to the figure, the results achieved by the proposed machine learning and signal processing based systems were very close to that of ELM experiments. Thus, the presented method can be used to predict C/C_0 values in the ELM process without performing the experiments several times. As far as we know, there is no published study focused on calculating the accuracy percentages of ANN or k-NN methods to predict C/C_o values in the ELM process.

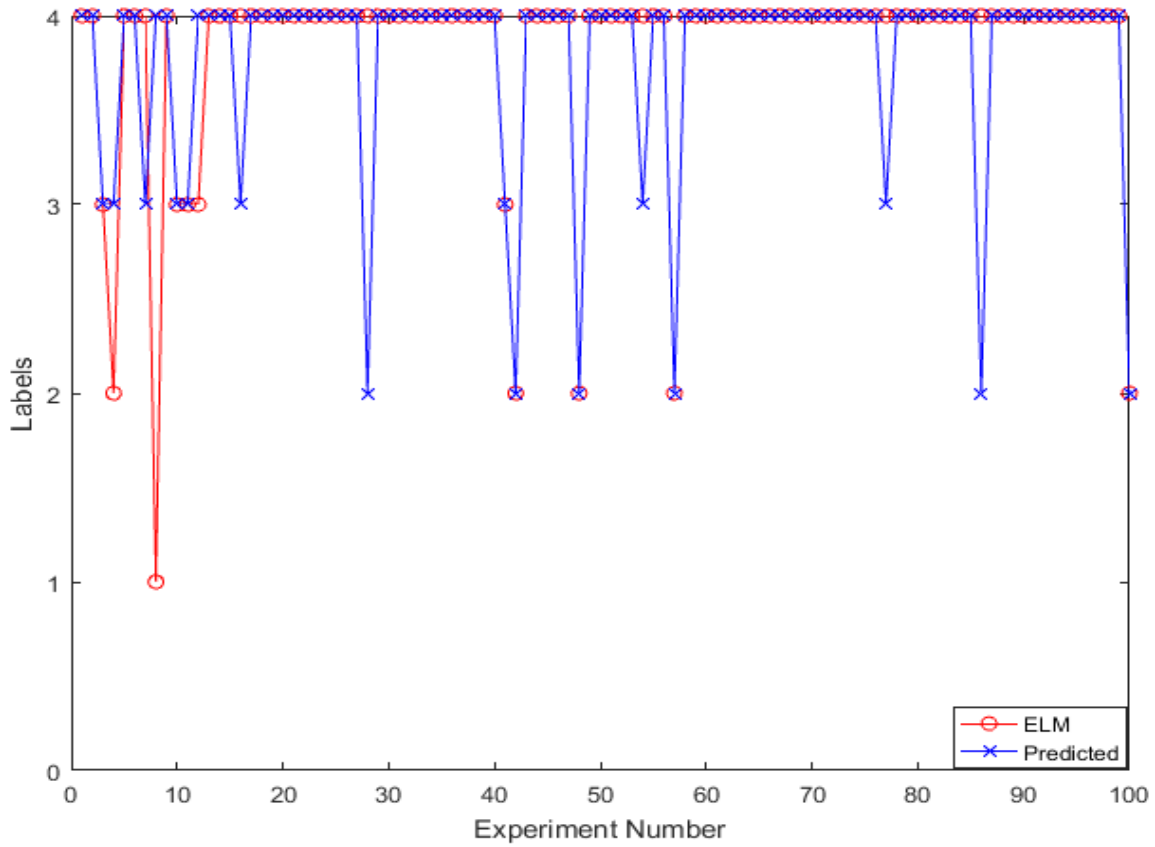

Fig. 7: The comparison of predicted and actual C/C_0 values.

Conclusions

The study aimed to predict solute concentration in the external phase during ELM experiments. The time, extractant type, extractant concentration, diluent type, surfactant concentration, mixing speed, stripping phase concentration, and modifier concentration were the input parameters that affect the system performance. The C/C_o predictions were outputs of the proposed system. C/C_o proportion is critical for the ELM efficiency. If this proportion converges to zero, the efficiency approaches to the maximum. Hence, it was intended to predict C/C_o proportion during the ELM process.

At first, the preprocessing step that included the labeling and quantization procedures was applied to the experimentally measured C/C_o values. By these two procedures, experimental C/C_o values were assigned to five subintervals, and the class labels of each C/C_o value were obtained. After preprocessing, 3-fold cross-validation was utilized to enhance the decision ability of ANN and k-NN techniques. The number of folds was chosen as 3 to decrease the computational complexity. The 3-fold cross-validation was followed by the classification/decision step in which the objective was to estimate the labels of C/C_o proportions as accurate as possible. In this final step, two of the most popular and convenient machine learning methods, ANN and k-NN, were preferred to perform the given task. At the output of the classification/decision step, the estimated labels of C/C_o proportions were obtained. The estimated C/C_o labels were compared with the actual C/C_o labels to evaluate the performance of system. The highest average accuracy percentages were 90.20 and 88.75 for k-NN (for k=5) and ANN, respectively. Hence, we can say that the predicted values of succinic acid concentration values in the external phase during the ELM process were very close to experimental results.

This study holds the significance of using machine learning and data mining techniques for chemical experiments. The main advantage of the study is to reduce the number of experiments to achieve C/C_o values. According to the obtained results, an expert can accurately predict C/C_o level by utilizing the proposed system.

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