ANN applied to the prediction of acetonitrile-water azeotrope separation

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Abstract

The purpose of this research is to simulate and validate a process of separation of the acetonitrile-water mixture by extractive distillation in DWSIM, which serves as a basis for the design of the artificial neural network (ANN) capable of predicting the mole fractions of acetonitrile, ethylene glycol and water. For the elaboration of the neural network, a database was generated from the simulation performed in DWSIM, and the database was composed of 100 pairs, with 4 inputs, feed flow temperature, acetonitrile mole fraction at feed, pre-concentration column pressure (C1) and ethylene glycol mole fraction. The artificial neural network was designed in MATLAB software using 9 hidden neurons and a Bayesian Regularization algorithm for training, with an MSE value of 3.0723e-05 and a total regression coefficient of 0.99996. The network was validated by developing a comparative statistical analysis, obtaining 95% reliability. The simulation allowed obtaining 0.9830 of acetonitrile in the distillate of column 2 (extraction column), in column 3 (recovery) 0.9016 of water in the distillate and 0.9737 of ethylene glycol in the bottom. In addition, it should be mentioned that there was no recirculation of the products from the bottom of column 3 to column 2. This proposal allowed obtaining results close to the reference research. It should be mentioned that the percentage of errors is less than 10%, giving efficient results in the investigation. It is recommended to normalize the input values to the ANN that are greater than 1.

Keywords: *<EXTRACTIVE DISTILLATION> <AZEOTROPE> <DWSIM> <ACETONITRYL> <ETHYLENGLICOL> <MATLAB> <ARTIFICIAL NEURON NETWORK> <NEURONS>.*

1. INTRODUCTION

Azeotropic compounds have posed a problem for chemical industries in achieving their separation, as they are mixtures with close boiling temperatures in which the compositions of liquid and vapor are equal under an arranged pressure (1). Azeotropes are caused by a strong deviation from ideal mixture behavior (described by Raoult's law) and pose great challenges for separation processes. Ideal or near-ideal mixtures can be clearly differentiated on the basis of their volatility, according to which separation in the VLE (Vapor - Liquid in Equilibrium) states can be efficiently achieved by simple distillation. However, this no longer applies to azeotropes where the volatilities of the components are equal. The design of an azeotrope separation process always starts with VLE data and a phase diagram, which can be obtained by experimentation, thermodynamic models and equations of state.

The mixture of acetonitrile and water can be found in some chemical industries, like Mallinckrodt Chemicals, that is a mixture of azeotropes as a waste stream. This company purifies seventy peptide drugs with acetonitrile (2). However, with technological advances, it has been possible to perform several studies to achieve its separation. Some require the addition of a third component known as a solvent. This changes the boiling temperatures of a component, and with extractive distillation, the decomposition of the azeotrope is achieved (3). The cost of removing this residual solvent and purchasing fresh acetonitrile increases as the capacity of the process expands. However, laboratory experiments in extractive distillation are time-consuming and expensive due to the many parameters involved. Therefore, it is convenient to predict experimental data with the help of available simulation programs (4).

Extractive distillation could be used to separate azeotropic or near-boiling-point homogeneous mixtures. A low-volatile liquid is added to the mixture as a carrier to increase the volatility throughout the concentration region by decreasing a component's partial pressure or volatility. The entraining agent has to meet many different properties. The boiling point of the trainer must be much higher than the boiling points of the other components, and it must be thermostable, inexpensive, and non-toxic, to mention only the main characteristics (5). If the mixture has a non-ideal vapor-liquid equilibrium behavior, it can form an azeotrope, a mixture of chemical components with identical compositions of liquid and vapor phases in equilibrium (6). For overcoming this difficulty, some special distillation processes have been applied, such as pressure swing distillation (PSD), extractive distillation (ED), and azeotropic distillation (AD) (7).

Neural network techniques are becoming more relevant due to the efficient performance and diversity of their applications in the chemical industry sectors, employing several resolution methods (8).

As a result of good modeling capabilities, neural networks have been widely used for several chemical engineering applications, such as sensor data analysis, fault detection, and identification of nonlinear processes. However, there are very few reported works evaluating the ability of artificial neural networks (ANNs) to model processes, including extractive distillation (ED) (9).

Azeotrope separation is an important and challenging problem in the chemical process industry. Several methods can be used for this purpose, including pressure swing distillation (PSD), heterogeneous azeotropic distillation (HAD), and extractive distillation (ED) (10).

A study of great interest for developing cell integration work is titled "A new model for predicting binary mixture of ionic liquids plus water density using artificial neural networks." The most important part of this study is the application of Artificial Neural Networks whose main purpose is predicting density values in a binary mixture of water and ionic liquids at different temperatures for different imidazolium-based ionic liquids. Two intelligent models, the multilayer perceptron and radial basis function models, were developed (11).

The development of predictive models in the properties of azeotropic mixtures places as a reference the following technological research topic "Prediction of thermodynamic properties of azeotropic mixtures of ANN." In this study, the coefficients of activity and relative volatility were used to determine the azeotropic properties proposing two different modeling approaches, such as predicting azeotropic variables and correlating experimental data. The first modeling calculates the azeotropic position in ANN mixtures using numerical methods using experimental variables such as relative volatility and activity coefficient. Then, knowing the NRTL and Wilson parameters in each temperature range, the azeotropic composition of the ANN mixtures and their binary systems can be evaluated by solving the nonlinear equations with Newton's method. If a root is between 0 and 1, this indicates the existence of the azeotrope in the mixture; otherwise, the mixture is azeotropic (12). Based on the work done by Zhao, its authors conclude that the ability to observe that each ANN mixture contains two azeotropic binary systems also makes the ANN mixture an azeotrope.

Artificial neural networks mimic the human brain in processing input signals and transform them into output signals. It provides a powerful nonlinear modeling algorithm between feature variables and output signals. ANN is a kind of nonparametric modeling technique that is suitable for complex phenomena whose underlying functions are not known to researchers. In other words, ANN can learn from the data without assumptions of specific functions. (13).

Artificial neural networks can be single or multi-layered and consist of processing units (nodes or neurons) interconnected by adjustable weights that allow signals to travel through the network consecutively. Generally, the ANN can be divided into three layers of neurons: input (receives information), hidden (responsible for extracting patterns and performing most of the internal processing) and output (produces and presents final network outputs). The transfer functions, learning algorithm and architecture determine the overall behavior of the artificial neural network (14). Artificial neural networks consist of highly interconnected layers of single nodes, where neurons act as nonlinear processing elements within the neural network (15).

The accuracy of both models was examined using different graphical and statistical methods. The input parameters were temperature, water/ionic liquid ratio, molecular weight, critical temperature, critical pressure and boiling point temperature of ionic liquids, and the model output was density.

Molecular weight, critical temperature, critical pressure, and boiling point temperature were used to discriminate between ionic liquids (16).

In another study for neural network construction, temperature, water ratio, molecular weight, boiling temperature, critical temperature and pressure of the ionic liquid were defined as input data, while the output parameter was the density of the ionic liquid. In this study, two commonly used smart ANN models, Multilayer Perceptron ANN (MLP-ANN) and Radial Basis Function ANN (RBF-ANN), were applied to predict the experimental density data based on input parameters. For determining the optimal performance of the ANN models, the data points were divided into three subsets called training, validation, and test data points. The train set, including 70% of the data points, was used to train and build the models. The validation data (15% of all data) was used to test the globalization and generalization capability of the ANN models during the training phase. The test data (15% of all data) were used to determine the quality of the ANN models for predicting the pattern of invisible data (17).

2. Materials and methods

2.1. Description of the process

Figure 1 illustrates the extractive distillation process for separating acetonitrile-water from Cui and Sun. The process comprises three columns: 1) the Pre-concentration column, 2) the Acetonitrile extraction column, and 3) the Ethylene glycol recovery column.

In the pre-concentration column, the bottom product has a molar flux of 99.43% water. In the extraction column, ethylene glycol is fed as a solvent to obtain as head product acetonitrile in greater quantity and the recovery column, water and ethylene glycol are obtained in the head and bottom products, respectively.

For the analysis and simulation of the process, the feed basis is 500 Kmol/h with a molar composition of 80% water and 20% acetonitrile at a temperature of 50 °C.

2.1.1. General information about the process

Table 1 summarizes the composition of the process feed taken from Cui and Sun that enters the pre-concentration column. Following tables list the operating conditions of the pre-concentration, extraction and recovery column.

Unlike the process used by Cui and Sun, no recirculation of the bottom product from the recovery column to the extraction column was used. In the extraction column, 116.72 Kmol/h of ethylene glycol was fed to obtain acetonitrile in the head product with a molar concentration of 98.39% and in the bottom products, a mixture of ethylene glycol, water and traces of acetonitrile. In the recovery column, water was obtained in the head product with a molar concentration of 90.16% and ethylene glycol was recovered in the bottom product.

Figure 1. Simulation of the distillation system for separating the acetonitrile-water in DWSIM.

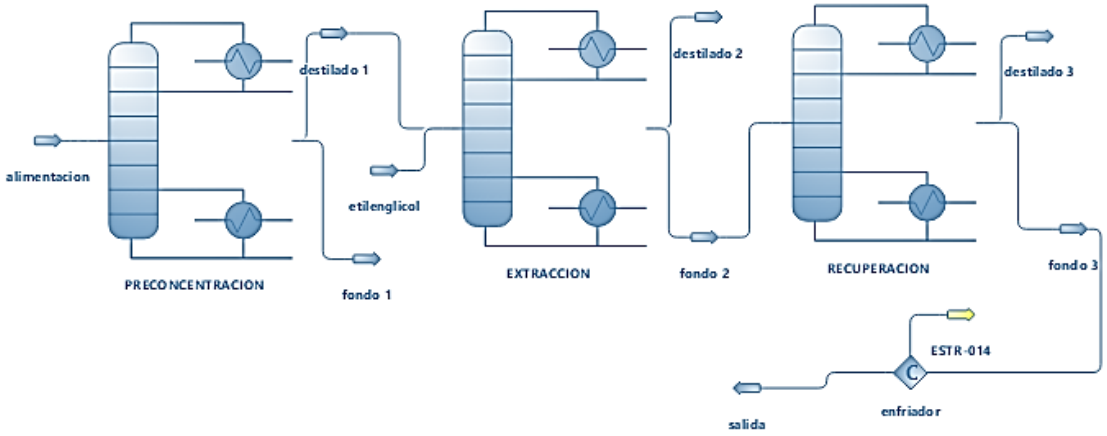


Table 1. Feeding conditions

Parameter	Quantity	Unit
Absolute pressure	1	atm
Temperature	50	°C
Molar feed flow	500	Kmol/h
Initial molar composition of acetonitrile	0,2	---
Initial molar composition of water	0,8	---

Table 2. Operating conditions of the pre-concentration column.

Parameter	Quantity	Unit
Absolute pressure	0,83	atm
Number of total plates in the column	16	--
Number of inlet feed plate	11	--
Acetonitrile production (distillate)	65,3	%
Water production (distillate)	34,7	%
Distillate molar flux	152,97	Kmol/h
Bottom molar flux	347,02	Kmol/h
Thermal load on the condenser	1577	kW
Thermal load on the reboiler	1963	kW

Table 3. Operating conditions of the extraction column

Parameter	Quantity	Unit
Absolute pressure	0,38	atm
Number of column plates	42	--
Number of feed plate	31	--
Solvent inlet plate number.	9	--
Solvent flow	116,72	Kmol/h
Acetonitrile Production (distillate)	99,99	%

Water production (distillate)	0,01	%
Ethylene glycol production (distillate)	Traces	%
Distillate molar flux	99,97	Kmol/h
Background molar flow	169,72	Kmol/h
Thermal load on the condenser	1291	kW
Thermal load on the reboiler	1495	kW

Table 4. Operating conditions of the recovery column

Parameter	Quantity	Unit
Absolute pressure	0,1	atm
Number of column plates	12	--
Number of feed plate	6	--
Water production (distillate)	99,99	%
Ethylene glycol production (background)	99,99	%
Distillate molar flux	53,01	Kmol/h
Bottom molar flux	116,72	Kmol/h
Thermal load on the condenser	714	kW
Thermal load on the rebolier	798	kW

2.2. Simulation in DWSIM

DWSIM is an open-source CAPE-OPEN-compliant chemical process simulator for Windows, Linux and macOS systems. DWSIM features a complete set of unit operations, advanced thermodynamic models, support for reaction systems, oil characterization tools and a full-featured graphical interface.

The distillation columns used for the simulation in Figure 1 correspond to the "ChemSep Colum" model. In addition, a thermodynamic NRTL model was used based on the research conducted by Cui and Sun (18). The tables above show the operating conditions for the process simulation. ChemSep columns require the specification of two operating parameters and the pressure value.

The mathematical method used by the simulator in this process corresponds to Newton's method, and the number of iterations can be modified according to the system's operation.

2.3. Sensitivity analysis

Table 5 shows 4 inputs for the Artificial Neural Network, which are considered significant in the process: Feed temperature, Fraction of acetonitrile, Pressure of the pre-concentration column (Column 1) and the fraction of ethylene glycol.

Table 5. Sensitivity analysis results.

Tickets		Exits		
Variable	**Range**	**Acetonitrile Fraction**	**Water Fraction**	**Ethylene glycol fraction**

Supply temperature (°C)	40-60	0,983043-0,983018	0,901602-0,901606	0,973719-0,973718
Water Fraction (Feed)	0,7-0,9	1-0,902538	0,754301-0,911440	0,986079-0,963424
Acetonitrile Fraction (Feed)	0,1-0,3	0,57281-1	0,947599-0,631066	0,921754-0,994762
Column 1 pressure (atm)	0,1-1	0,998885-0,978195	0,895595-0,903170	0,973716-0,972718
Ethylene glycol temperature (°C)	50-80	0,983280-0,982713	0,902030-0,901062	0,973719-0,973719
Ethylene glycol fraction	0,7-0,99	0,999980-0,983030	0,715818-0,901604	0,748254-0,973719

• Of the 6 variables analyzed, 4 were taken because they have the greatest significance in the acetonitrile separation process.

• The ethylene glycol fraction was chosen because recirculation was eliminated, and the composition at the end of the process was not pure.

2.4. Artificial Neural Network (ANN) design and training

The ANN design (Figure 2) is based on four input parameters: pre-concentration column pressure, feed stream temperature, molar composition of acetonitrile in the feed stream and molar composition of the ethylene glycol solvent. With 3 output parameters corresponding to the molar concentrations of water, acetonitrile and ethylene glycol.

The ANN design employs 9 hidden neurons; the present value was chosen by comparing the different numbers of neurons, as shown in Table 6.

Table 6. Error values with different numbers of neurons

Artificial neural network validation values							
		Levenberg-Marquardt		Bayesian Regularization		Scaled Conjugate Gradient	
NUMBER OF NEURONS	**SAMPLES**	**MSE**	**R**	**MSE**	**R**	**MSE**	**R**
5 Training	70	2,69e-5	9,99e-1	9,71e-6	9,99e-1	3,38e-3	9,85e-1

	Validation	5	5,03e-5	9,99e-1	0,00e-0	0,00e-0	1,90e-3	9,91e-1
	Testing	25	4,47e-5	9,99e-1	7,75e-6	9,99e-1	3,47e-3	9,84e-1
6	**Training**	70	2,06e-5	9,99e-1	3,88e-6	9,99e-1	6,39e-4	9,97e-1
	Validation	5	2,98e-5	9,99e-1	0,00e-0	0,00e-0	8,02e-4	9,96e-1
	Testing	25	1,58e-5	9,99e-1	2,47e-5	9,99e-1	1,04e-3	9,95e-1
7	**Training**	70	1,22e-4	9,99e-1	8,24e-6	9,99e-1	3,61e-4	9,98e-1
	Validation	5	1,46e-4	9,99e-1	0,00e-0	0,00e-0	2,21e-4	9,99e-1
	Testing	25	4,97e-4	9,98e-1	5,38e-6	9,99e-1	5,27e-4	9,97e-1
8	**Training**	70	1,36e-4	9,99e-1	6,95e-6	9,99e-1	3,64e-4	9,98e-1
	Validation	5	2,78e-4	9,98e-1	0,00e-0	0,00e-0	4,49e-4	9,97e-1
	Testing	25	3,48e-4	9,98e-1	9,33e-6	9,99e-1	4,64e-4	9,98e-1
9	**Training**	70	4,81e-5	9,99e-1	6,91e-6	9,99e-1	1,77e-3	9,92e-1
	Validation	5	5,27e-5	9,99e-1	0,00e-0	0,00e-0	2,86e-3	9,85e-1
	Testing	25	1,61e-4	9,99e-1	6,28e-6	9,99e-1	2,16e-3	9,90e-1
10	**Training**	70	3,40e-5	9,99e-1	7,67e-7	9,99e-1	5,98e-3	9,74e-1
	Validation	5	2,88e-5	9,99e-1	0,00e-0	0,00e-0	5,58e-3	9,78e-1
	Testing	25	7,67e-5	9,99e-1	2,15e-5	9,99e-1	8,02e-3	9,64e-1
15	**Training**	70	2,02e-4	9,99e-1	1,97e-6	9,99e-1	2,87e-3	9,86e-1
	Validation	5	1,40e-3	9,92e-1	0,00e-0	0,00e-0	3,34e-3	9,90e-1
	Testing	25	6,02e-4	9,97e-1	3,22e-5	9,99e-1	8,06e-3	9,70e-1
20	**Training**	70	1,22e-4	9,99e-1	6,00e-7	9,99e-1	1,09e-2	9,50e-1
	Validation	5	5,33e-4	9,94e-1	0,00e-0	0,00e-0	8,49e-3	9,68e-1
	Testing	25	1,32e-3	9,95e-1	1,19e-4	9,99e-1	1,88e-2	9,19e-1
25	**Training**	70	1,82e-6	9,99e-1	1,44e-7	9,99e-1	7,11e-2	7,55e-1
	Validation	5	6,59e-4	9,97e-1	0,00e-0	0,00e-0	5,56e-2	7,99e-1
	Testing	25	1,07e-3	9,95e-1	1,59e-4	9,99e-1	1,01e-1	6,7911e-1
30	**Training**	70	4,71e-4	9,98e-1	4,66e-9	9,99e-1	5,98e-3	9,72e-1
	Validation	5	2,97e-3	9,87e-1	0,00e-0	0,00e-0	5,22e-3	9,82e-1
	Testing	25	3,31e-3	9,79e-1	1,80e-4	9,99e-1	2,01e-3	9,17e-1
40	**Training**	70	1,49e-5	9,99e-1	8,54e-13	9,99e-1	6,91e-4	9,96e-1
	Validation	5	2,08e-3	9,84e-1	0,00e-0	0,00e-0	7,78e-3	9,77e-1
	Testing	25	5,74e-3	9,74e-1	1,12e-4	9,99e-1	1,71e-2	9,35e-1
50	**Training**	70	2,12e-3	9,90e-1	4,24e-10	9,99e-1	3,55e-3	9,83e-1
	Validation	5	1,41e-2	9,26e-1	0,00e-0	0,00e-0	3,28e-2	8,81e-1
	Testing	25	1,21e-2	9,59e-1	2,59e-4	9,98e-1	4,60e-2	8,39e-1
60	**Training**	70	2,86e-4	9,98e-1	3,62e-10	9,99e-1	1,05e-2	9,55e-1
	Validation	5	2,60e-2	8,72e-1	0,00e-0	0,00e-0	3,96e-2	9,25e-1
	Testing	25	2,40e-2	8,85e-1	7,22e-5	9,99e-1	5,16e-2	7,87e-1
70	**Training**	70	3,68e-5	9,99e-1	1,30e-13	9,99e-1	5,79e-2	7,96e-1
	Validation	5	2,68e-2	9,21e-1	0,00e-0	0,00e-0	1,70e-1	5,17e-1
	Testing	25	2,97e-2	8,92e-1	2,03e-1	9,99e-1	2,53e-1	4,03e-1
80	**Training**	70	1,32e-4	9,99e-1	1,83e-12	9,99e-1	9,15e-4	9,96e-1
	Validation	5	5,26e-2	8,55e-1	0,00e-0	0,00e-0	9,55e-2	8,20e-1
	Testing	25	6,45e-2	7,69e-1	3,77e-4	9,98e-1	1,00e-1	7,26e-1
90	**Training**	70	1,08e-5	9,99e-1	4,72e-13	4,72e-13	6,75e-4	9,97e-1
	Validation	5	5,05e-2	8,89e-1	0,00e-0	0,00e-0	8,23e-2	8,85e-1
	Testing	25	8,66e-2	7,80e-1	8,03e-5	9,99e-1	1,37e-1	6,39e-1
100	**Training**	70	5,32e-4	9,97e-1	2,28e-12	9,99e-1	1,90e-2	9,24e-1
	Validation	5	5,47e-2	8,07e-1	0,00e-0	0,00e-0	1,46e-1	8,13e-1
	Testing	25	1,81e-1	6,68e-1	8,41e-4	9,97e-1	1,63e-1	6,14e-1

The Bayesian Regularization model omits the validation of the values, and it should be emphasized that the mean square errors must be close to zero for the proposed artificial neural network to be effective.

The Bayesian Regularization training algorithm performs better than the Levenberg-Marquardt algorithm in a predictive capacity. The study found that the network training execution time is longer than both algorithms (Levenberg-Marquardt and Scaled Conjugate Gradient); the greater the number of hidden neurons, the longer the training time.

Once the simulation was validated in DWSIM, a database of 120 data sets (with 4 inputs and 3 outputs) was generated from the variation of operating parameters. Table 7 shows the range of variation of the inputs.

Table 7. Restrictions for ANN entries

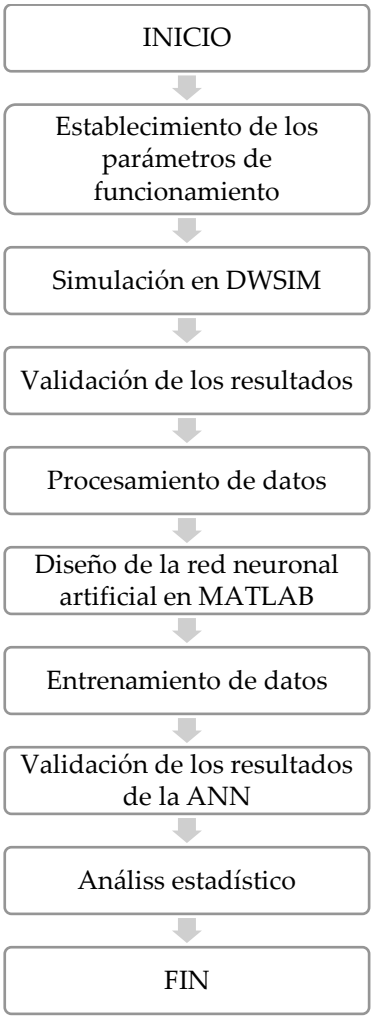
	Restrictions			
Parameter	Pressure	Temperature	Feeding fraction	Solvent fraction
Details	Atm [in the pre-concentration column] [in the pre-concentration column].	°C [Feed molar flow] [Feed molar flow] [Feed molar flow] [Feed molar flow] [Feed molar flow	Adimensional [Molar composition of acetonitrile].	Adimensional [Molar composition of ethylene glycol].
Range	0,1-1	40-60	0,1-0,3	0,7-0,99

From the total set of data generated, the first 100 are selected for the design of the ANN in MATLAB, while 20 different pairs are used to perform additional validation through the corresponding statistical analysis to corroborate the performance of the ANN and evaluate its level of learning.

2.5. Description of the methodology

The first part of this study consists of simulating the process of Figure 1, considering the operating conditions of the distillation columns. The next step is to validate the results obtained and proceed with the neural network design considering the inputs and outputs determined for the process prediction and the restrictions imposed by the simulation. Subsequently, training and validation of the ANN are performed by means of a statistical graphical analysis to evaluate the predictive capacity of the neural network. Figure 3 illustrates the flow diagram of the methodology.

Figure 2. Schematic of the designed ANN



3. Results and Discussion

3.1. Simulation validation

Before the design of the ANN, it is essential to compare the simulation developed in DWSIM with results from the literature. The study developed by (18) was used for validation. The process in DWSIM was adapted according to the simulation developed in ASPEN PLUS. Table 8 details the comparison of the results. As can be seen, the percentage errors of the mole fractions of interest in their respective distillation columns do not exceed 10%.

Table 8. Simulation validation

UNIT	PARAMETER	ASPEN PLUS [Cui and Sun., 2019]	DWSIM	ERROR (%)
Pre-concentration (1)	X - background$_{H20}$	0,999	0,994	0.56%
	X_{ACN} - distilled	0,653	0,642	1.57%
Extraction (2)	X_{ACN} - distilled	0,999	0,983	1.69%

	X - $background_{EG}$	0,687	0,666	3.02%
Recovery (C3)	X -$distilled_{H2O}$	0,999	0,901	9.83%
	X - $background_{EG}$	0,999	0,973	2.62%

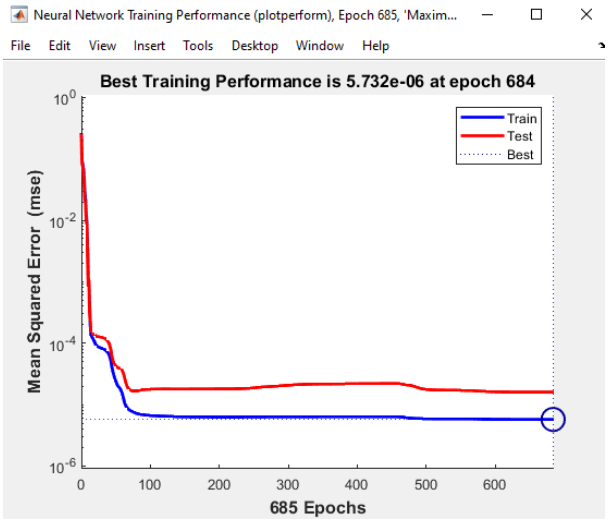
Table 9. Simulation results in DWSIM

	Pre-concentration		**Extraction**		**Recovery**		
Objects	Distillate C1	Fund C1	Distillate C2	Fund C2	Distillate C3	Fund C3	Units
Temperature	95,042	82,084	100,763	53,220	155,199	42,484	°C
Pressure	0,851	0,851	0,385	0,385	0,385	0,101	Atm
Mass flow	6305,15	5006,16	8419,35	3831,36	7301,35	1118	Kg/h
Mole fraction of acetonitrile	0,0056	0,6429	0,0310	0,9830	1,51E-05	0,0983	-
Water mole fraction	0,9943	0,3570	0,3019	0,0169	0,0262	0,9016	-
Mole fraction of ethylene glycol	-	-	0,6669	7,39E-21	0,9737	2,59E-12	-

3.2. Artificial Neural Network

The ANN designed has 9 hidden neurons and was trained with a base of 100 data sets with 4 inputs and 3 outputs using the Bayesian Regularization algorithm. The mean square error and Pearson's correlation coefficient were used to validate the model. Table 10 shows the mean squared error (MSE) values for the training phase and test phase of the ANN. The Bayesian Regularization algorithm is a robust model that eliminates the need for validation and leverages the data in training and learning the ANN. The MSE values for the training and validation phase are 1.08e-6 and 3.07e-5, respectively. Figure 4 shows the evolution of the mean square error during the training phase, with a final MSE of 5.732e-6.

Figure 3. ANN training performance



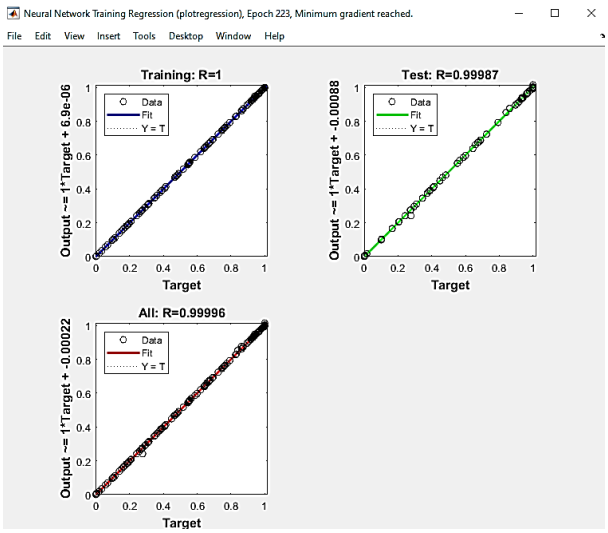
As shown in Figure 5, there is a moderate dispersion between the output and targets of the neural network in the training phase and test phase. In addition, the R values for the training phase are 1 and 0.99, respectively.

Therefore, the overall R-value of 0.99 indicates that the outputs and targets are effectively correlated.

Table 10. Mean square error of each ANN design phase.

PHASE	MSE
TrainPerformance (Training)	1,0834e-06
TestPerformance (Test)	3,0723e-05
ValPerformance (Validation)	NaN

Figure 4. Coefficients of the training regression and ANN test.



3.2.1. Prediction model for acetonitrile, water and ethylene glycol

Figures 6 and 7-8 compare the predictions and the values defined by the simulation in the extraction and recovery column. It can be seen that the comparisons obtained in the following cases are equal. The neural network prediction is efficient since there is a good correlation between input and output values. The developed model resembles the simulation data, proving that the neural network is efficient and adequate for predicting acetonitrile and water concentration.

Based on the analysis of Figures 6-8, the average percentage error (%E) of the predictions is 0.02% (Ethylene glycol in the background) and 0.20% (Water in the distillate) in the recovery column; 0.09% (Acetonitrile in the distillate) in the extraction column.

Figure 5. Comparison of ANN and DWSIM results in the extraction column distillate (acetonitrile mole fraction).

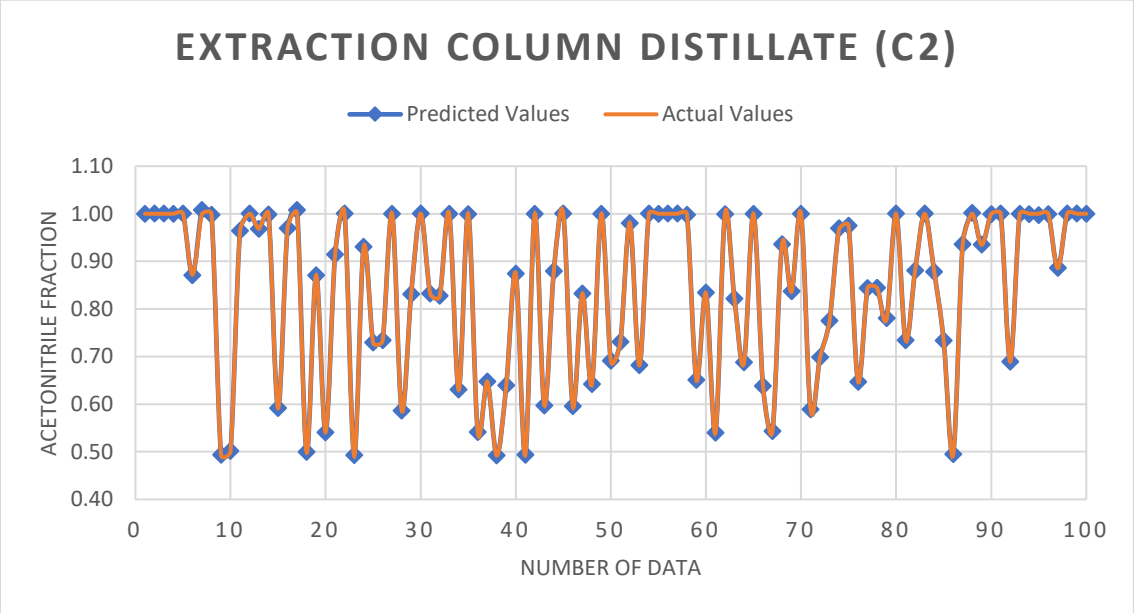


Figure 6. Comparison of ANN and DWSIM results in the recovery column distillate (ethylene glycol mole fraction).

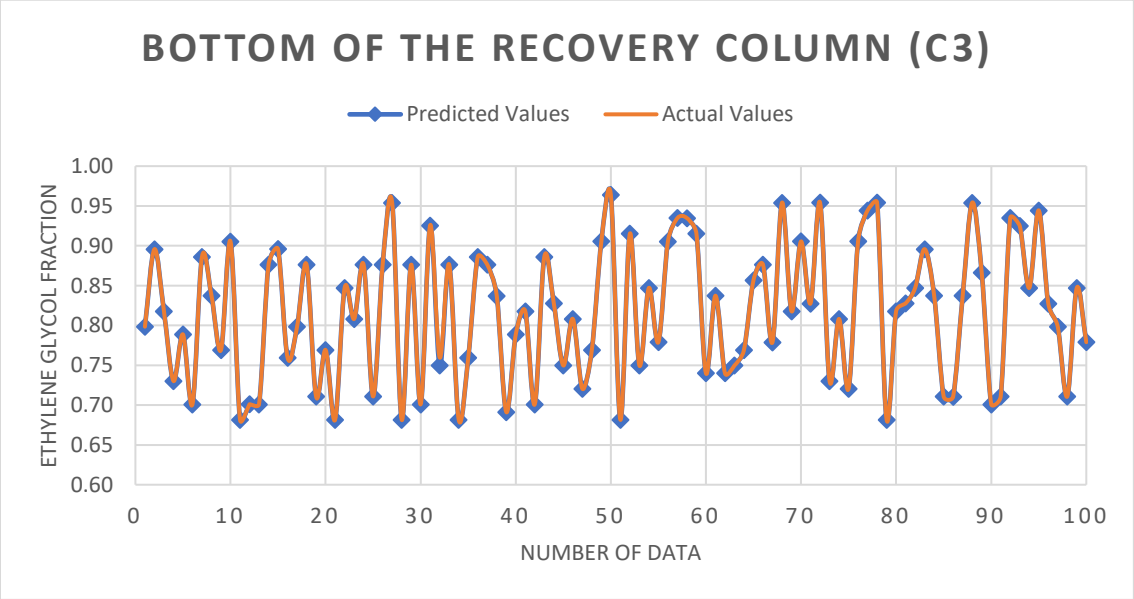
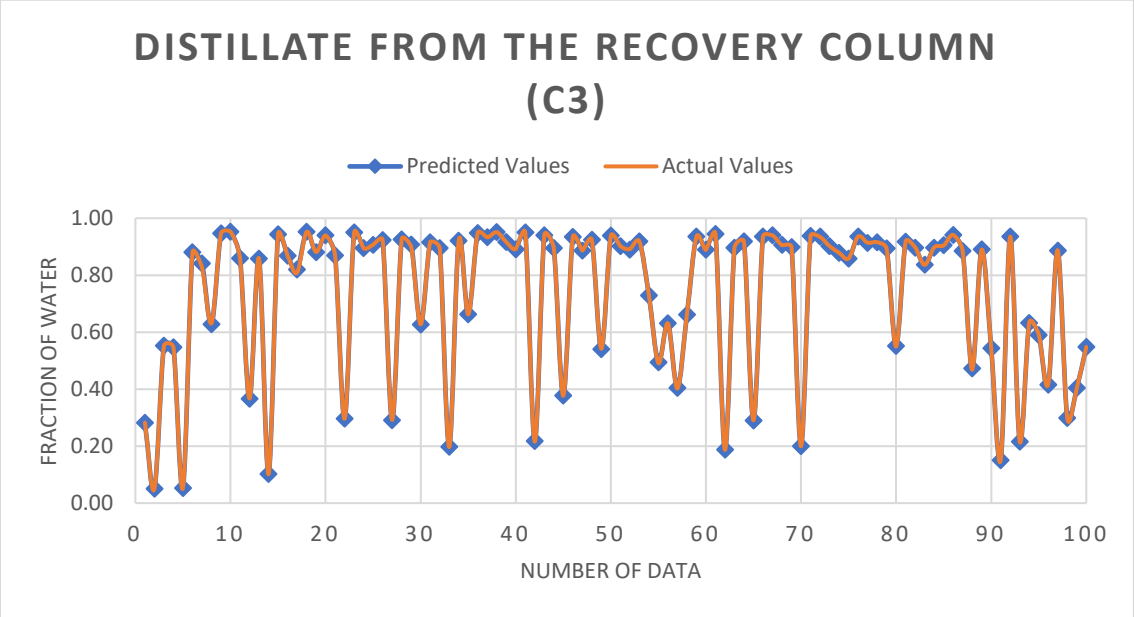


Figure 7. Comparison of ANN and DWSIM results in the recovery column distillate (water mole fraction).



3.2.2. ANN model verification

The performance of the designed neural network during training and testing is necessary to evaluate the functionality with another set of data not known to the neural network and to test its predictive capability. For this, a set of 20 pairs of data (feed temperature, acetonitrile molar composition, ethylene glycol molar composition and pressure in the pre-concentration column) simulated in DWSIM was generated to collect new operating conditions. The ANN was designed to predict acetonitrile's molar composition in the extraction column; the molar composition of water and ethylene glycol in the recovery column was used to verify the prediction capability.

The comparison between predicted values and actual values is shown in Figures 9-11. Again, the results show an overlap between these values, so it can be said that the designed ANN has a good predictive ability for predicting the molar compositions of acetonitrile, ethylene glycol and water.

Figure 8. Comparison between predicted and actual values of acetonitrile molar composition in the extraction column.

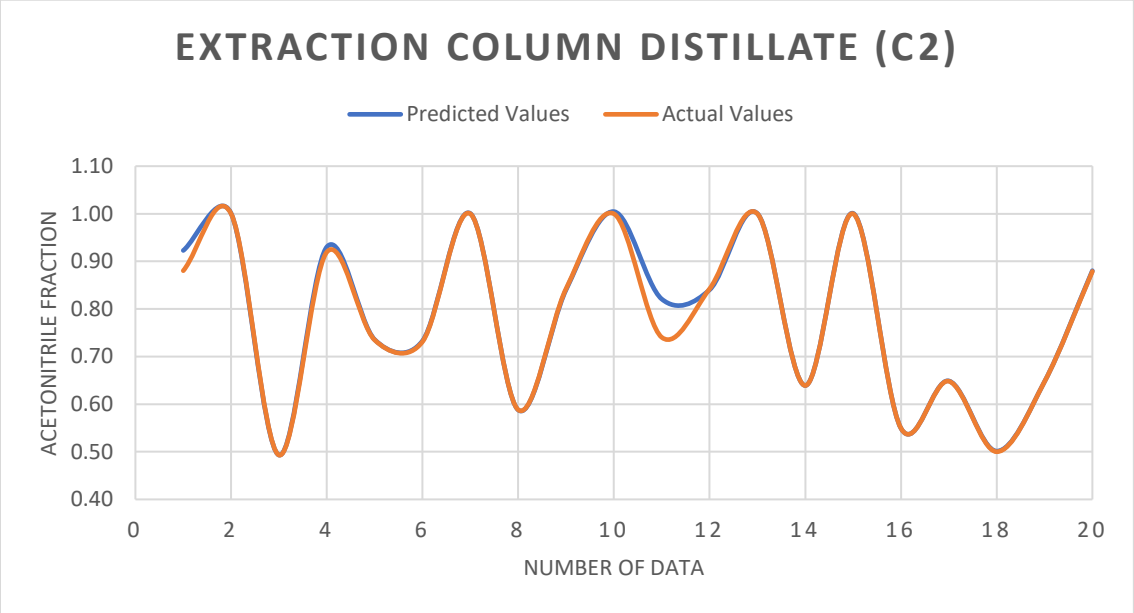


Figure 9. Comparison between predicted and actual values of ethylene glycol molar composition in the recovery column.

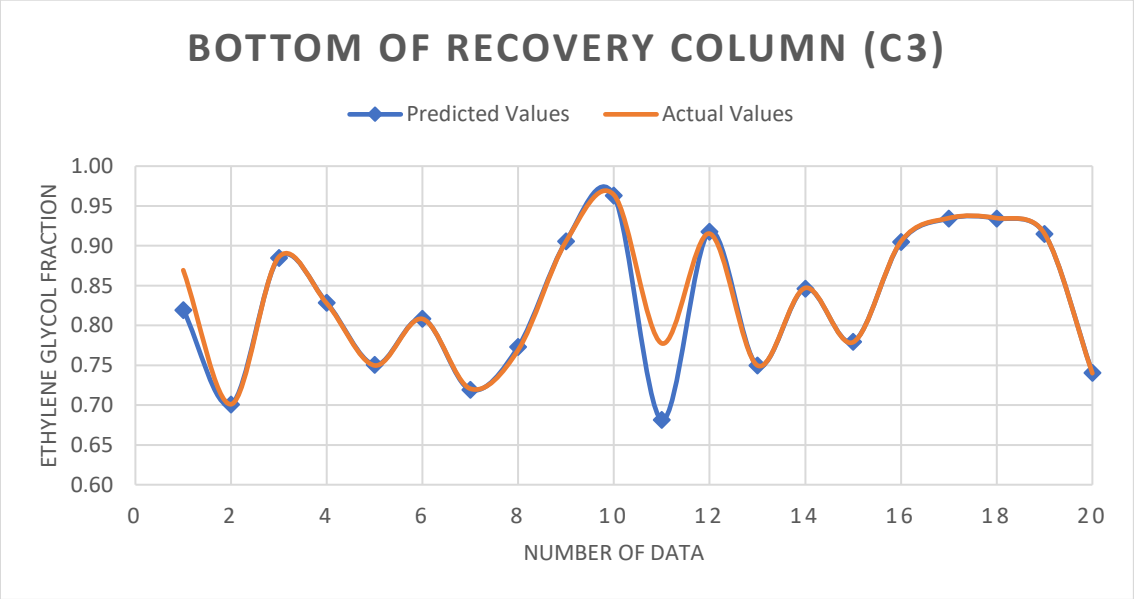
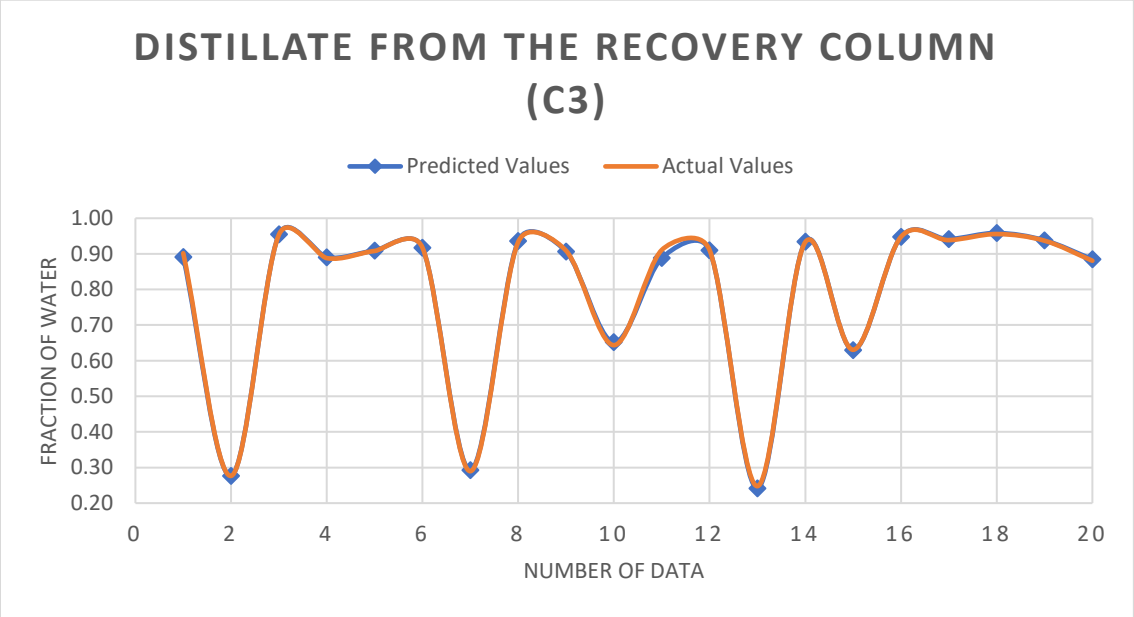


Figure 10. Comparison between predicted and actual values of the molar composition of water in the recovery column.



3.2.3. ANOVA analysis

The basic operation of an ANOVA consists of calculating the mean of each group and then comparing the variance of these means (intervariance) versus the average variance within the groups (intravariance). Under the null hypothesis that the observations in the different groups all come from the same population (they have the same mean and variance) (19).

The P-value for each output is determined to know whether or not the null hypothesis is discarded. If this value is less than 0.05, the null hypothesis is discarded; if it is more significant, it is accepted.

Consequently, as can be seen in Table 11, all the P-values of the outputs are more significant than 0.05, so the null hypotheses are accepted, and it is concluded that there is no significant difference between the means of any variable with a confidence level of 95%.

Table 11. ANOVA analysis

Source	Sum of Squares	Gl	Medium Square	F-Ratio	P-Value
Molar fraction of acetonitrile in the distillate of the extraction column.					
Between groups	5,0645E-04	1	5,0645E-04	0,02	0,9005
Intra groups	1,21478	38	0,0319679		
Total (Corr.)	1,21529	39			
Molar fraction of ethylene glycol at the bottom of the recovery column					
Between groups	5,0091E-04	1	5,0091E-04	0,07	0,7945
Intra groups	0,276505	38	7,27646E-03		
Total (Corr.)	0,277006	39			

Molar fraction of water in the recovery column distillate					
Between groups	4,0704E-06	1	4,0704E-06	0	0,9934
Intra groups	2,24216	38	5,90043E-02		
Total (Corr.)	2,24217	39			

The ANOVA table decomposes the variance of the data into two components: a between-group component and a within-group component. The F-ratio for the mole fraction of acetonitrile in distillate in column 2 is 0.0158423, for the mole fraction of ethylene glycol in the bottom of column 3 is 0.0688398, and for the mole fraction of water in distillate in column three is 6.89855E-05, which is the ratio of the between-group estimate to the within-group estimate. Since the P-value of the F-ratio is greater than or equal to 0.05, there is no statistically significant difference between the means of the 2 variables at the 95.0% confidence level.

4. Conclusions

In the study, an ANN was designed to predict products in the separation process of acetonitrile-water azeotropes by extractive distillation, using a set of 120 data obtained by simulation in DWSIM. The inputs and outputs of the ANN were determined from a sensitivity analysis resulting in 4 inputs and 3 outputs.

The ANN was trained with the Bayesian Regularization algorithm and several hidden neurons of 9. The calculated MSE and R2 were 1.0834e-06 and 0.99996, respectively. In addition, the ANN was validated by ANOVA analysis comparing the simulated and predicted values; there is no significance between the simulated and predicted values. Thus, the ANN created is accepted, and its use is recommended for acetonitrile-water azeotrope separation processes by extractive distillation.

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