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ANN IN NATURAL GAS TREATMENT PROCESS: CARBON DIOXIDE (CO2) - ETHANE (C2H6) AZEOTROPE SEPARATION

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Abstract: The aim in this study was to develop an artificial neural network (ANN) to forecast the mole fractions of the CO2- C2H6 azeotropic separation during the natural gas treatment process. The ANN was designed with experimental data (150 data pairs) obtained in DWSIM from a previously process described in bibliography. The sample used to train the ANN is structured by three inputs: pressure, temperature and solvent / feed ratio, and six outputs: the mole fractions of distilled CO2 and residual ethane in the extractive column, the mole fractions of distilled ethane and residual propane in the solvent recovery column and the mole fractions of distilled ethane and residual ethane in the concentrator column. The neural network was designed using 80 hidden neurons in its architecture and the Bayesian regularization algorithm for training (MSE=0.0036 and R=0.9554). The ANN's prediction was validated using statistical parameters (ANOVA and Kruskal Wallis) which indicate that the designed ANN is statistically valid and can be used to predict the mole fractions of the CO2- C2H6 azeotropic separation and can be used in the improvement of the processes of sweetening of natural gas after its demethanization.

Keywords: Azeotrope; Extractive Distillation; Natural Gas; DWSIM; Carbon Dioxide; MATLAB; Artificial Neural Networks (ANN)

1. Introduction

For decades, oil has been the basis of the energy structure worldwide; due to its high level of consumption (90% only in transportation) and extensive development (production of derivative products), it has become the main engine of both developed and developing countries [1]. However, despite the prevalence of oil as the main energy source, natural gas (NG) is currently transforming and changing this trend. NG is known as a mixture of gaseous hydrocarbons that is frequently found in fossil deposits in various forms, such as: non-associated (alone), dissolved or associated (accompany-ing oil) and in coal deposits [2], and constitutes the third energy source worldwide after oil and coal.

The composition of NG can change according to the type of reservoir, depth, loca-tion and geological conditions. However, for the gas to be used commercially or indus-trially, it must undergo a conditioning pretreatment, which is mainly based on the re-moval of water content and acid gases characteristics of its composition [3]. NG treat-ment generally involves four stages: sweetening, dehydration, oil removal and lique-faction [4].

There are several methods for NG sweetening: chemical absorption with amines, physical absorption, membrane permeation, and low-temperature distillation [5]. The low-temperature extractive distillation process (conventional process) separates CO2 from hydrocarbons through a sequence of two distillation columns and is one of the most widely used because it minimizes operational and safety problems [6]. However, despite its widespread industrial use and its major benefits, its high energy demand commonly accounts for more than 50% of plant operating costs [6,7].

In this regard, several studies to optimize the CO2-C2H6 separation process stand out. For example, Torres et al., [8] analyzed by simulation the effect of different fractions of liquefied hydrocarbons as a drag agent, the thermodynamic efficiency and the gen-eration of greenhouse gases in conventional process to remove CO2.This alternative presented better overall performances compared to the conventional chemical absorp-tion system. Lastari et al. [9] analyzed the effect of the solvent/feed composition and the location of the feed trays on the total energy requirement, establishing that the optimum solvent/feed ratio is in the range of 1.053 - 1.064 for the treatment of the mixture (CO2- C2H6) and that the stage where the feed and solvent enter has significant effects on the total energy demand of the column.

On the other hand, Tavan et al. [10] established through simulation in ASPEN HYSYS that there is a significant reduction in operating costs (total energy demand) using reactive absorption (RA) configurations making use of diethylamine as solvent, compared to conventional extractive processes. They then proposed, by means of rig-orous simulation methods, an extractive dividing-wall column (EDWC) taking into ac-count environmental (CO2 emission) and energy parameters and confirmed that this technology reduces the total energy demand by approximately 51.6% [11]. In addition, they proposed the feed-splitting technique, to separate the feed prior to entering the heat exchanger, in order to maintain a fraction of the feed at its original temperature. By means of this technique, the energy reduction in contrast to the conventional process is 56% [12]. With the aforementioned, EDWC is one of the most efficient and operative techniques, which allows an approximate saving of 17% in total annual costs [7]. It is important to mention hybrid technologies to minimize energy costs in distillation processes, this technology reports the advantages of a profitable process units [13–15].

Ebrahimzadeh et al., [5] suggest a three-column extractive distillation configuration for CO2-C2H6 separation (ASPEN PLUS simulation). The process described leads to a 10% reduction in annual cost without compromising the desired purification, and also significantly minimizes the energy required for liquefaction.

1.1. ANN as a prediction tool in chemical processes

ANNs are able of solving linear and nonlinear multivariate regression problems, hence permitting the study of the relationship between the input and output variables of the method employing a constrained number of test runs. In addition, ANNs can be effortlessly created by applying an appropriate plan of tests [16,17].

ANN models have been applied in several chemical processes, such as nonlinear multi-variable predictive controllers for distillation columns, where the controller uses an on-line optimization routine, which determines the future control variables that minimize the deviations between the predicted control variables and their set points [18]. Zamprogna et al. [19] designed a virtual sensor (recurrent ANN) that estimates product compositions, using measurements of secondary parameters such as temper-ature and molar flow, in a batch distillation column of intermediate vessels. The work showed that the estimated compositions match the actual values. Li et al. [20] combined ANNs with genetic algorithms (GA) to model the azeotropic distillation system of isopropanol-water mixture with complex mechanisms and optimize the energy.

Liau et al. [21] designed an ANN capable of predicting the quality of the crude oil as a function of the input parameters, thus optimizing and maximizing the production rate and the process. The developed system provides the optimal operating conditions of the fractionation unit considering the operating variables. Fernandez et al. [22] built an identification and control tool for a laboratory-scale distillation column based on neural networks using LABVIEW. They demonstrated that ANNs are a potential tool for their functionality when interacting with instruments, sensors and actuators.

Motlaghi et al. [23] created an ANN model of a crude oil distillation systems (CDU), to forecast the unspecified values of the desired product flow and temperature at the specified characteristics of the inpud feed, being able to minimizing the model output error and maximizing the required oil generation rate with respect to control parameter values (product quality).

On the other hand, Vafae et al. [24] used a multilayer perceptron (MLP) network as a new and effective method to simulate recoveries from 16 oil data sets using as input variables: API degrees, viscosity, characterization factor and steam distillation factor, to predict distillation performance. This study showed that MLP is more effective than the EOS equation of state method and Holland - Welch correlation.

In addition, Ochoa et al. [25] suggest a methodology to optimize heat-integrated in CDU, which considers an ANN model to represent the distillation column. The ANN is incorporated into the process in an optimization system to efficiently decide the working conditions that progress the overall economy of the process.

Leng et al. [26] used a back propagation ANN to build a model between the phys-ical properties and Terahertz spectra -THz- (technique used to realize the identification and determination of the principal gas components distilled from oil shale) with the input of THz-FDS over the range from 0.2 to 1.5 THz to quantitatively characterize the component and total pressure of the distillation process. The results indicate that the THz (Terahertz) technique combined with ANN is an effective tool for gas detection and can be used in industrial unconventional gas plants.

Osuolale and Zhang [27] presented a neural network-based strategy to model and optimize energy efficiency in distillation columns. They used ASPEN HYSYS for dis-tillation system simulation and ANN models to obtain optimal operating conditions that can maximize energy efficiency, quality, and product yield. The application of the proposed methodology improved 32.4% of exergetic efficiency.

There are several investigations that employ neural network models applied to chemical processes, however, the alternative processes of extractive distillation and separation of azeotropes have not been exploited within the ANN area. The advantage over simulation is that ANN is able to learn directly from a process and give shorter response times, allowing systems to be modeled in a more complex and realistic manner [28,29]. Other advantages are its simplicity, versatility, accurate approximation of complex nonlinear processes and "black box" approach; it does not require detailed knowledge of the system being analyzed[30,31].

In the present work we propose to generate an ANN, from Chemical Process Simulator open source (DWSIM), of the process proposed in [5] to predict the mole fractions of the main components obtained in the extractive distillation, both in the distillate and in the residues of each extractive column, as a function of the operating conditions of the process. In the future, ANN can be incorporated into industrial plants for prediction, optimization, and continuous improvement of processes.

2. Materials and Methods

2.1. Process Description

Figure 1 illustrates the alternative extractive distillation process for the separation of azeotrope CO2-C2H6, adapted from [5]. The process is composed of three columns: C1 - CO2 extractive column, C2 - solvent recovery column and C3 - concentrator column.

Column (C1) receives a free methane stream from a natural gas demethanizer (not considered in the process) containing CO2, C2H6 and hydrocarbons, which is mixed with the recirculation from the distillate of column (C3) containing CO2 and C2H6. In addition, the bottom product (hydrocarbons) from the column (C2) also enters in column (C1).

During the process, in C1 the head product is not entirely CO2 and the bottom product contains 10% CO2 together with C2H6 and heavy hydrocarbons.. In C2, the high purity solvent from the bottom stream of the first column is recovered at the base of the second column and the distillate from this column is fed to the third column (C3), which produces C2H6 (bottom) and azeotropic mixture (distillate).

Table 1 summarizes the composition of the feed entering the extractive column C1. While Tables 2-4 detail the operating conditions of the extractive, solvent recovery and concentrator columns, respectively. Unlike the conventional process, the distillate from the extractive column remains in a liquid state and does not require a liquefaction pro-cess, thus minimizing the total energy demand. The heavy hydrocarbons leaving the solvent recovery column as a bottom product are divided into NGL product and solvent, which is recycled back to the extractive column (recirculation 1). In C3 the bottom product is C2H6 with high purity (99.7 mole %), while distillate is CO2-C2H6 mixture (1.35 kmol/s, 46.2 mole % CO2). After heat recovery, it is recycled to C1 (recirculation 2 – Feed Stage = 5).

The thermal power source used for all heat exchangers is saturated steam at 6.9x105 Pa. In addition, as opposed to the conventional design using a total condenser, partial condensers are used in the solvent recovery column and concentrator.

Table 1. Feed Conditions to C1

Parameter	Quantity	Unit
Pressure	2.43e6	Pa (abs)
Temperature	320.15	K
Feed base (molar flow)	4	kmol/s
Initial composition of CO_2*	0.3225	-
Initial composition of C_2H_6*	0.4623	-
Initial composition of propane C_3 *	0.0753	-
Initial composition of isobutane i-C_4 *	0.0753	-
Initial composition of butane n-C_4 *	0.0323	-
Initial composition of isopentane i-C_5 *	0.0215	-
Initial composition of pentane n-C_5 *	0.0108	-

Source: Ebrahimzadeh et al., [5]

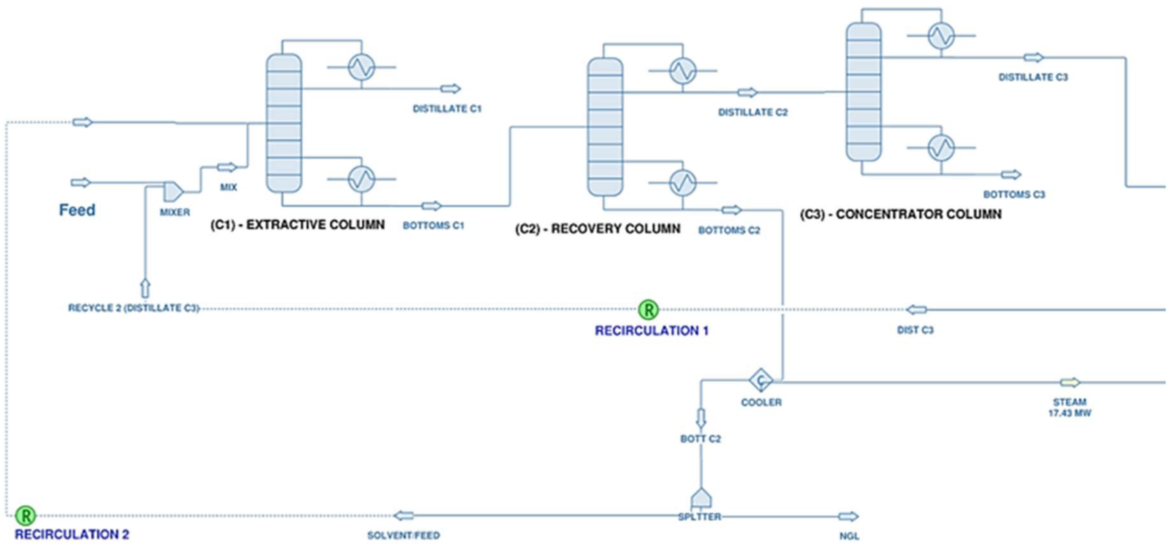

Figure 1. Simulation of the alternative distillation system to separate CO2- C2H6 azeotropes in DWSIM

Table 2. Extractive column operating conditions

Parameter	Quantity	Unit
Pressure	2.3e6	Pa (abs)
# Column stages*	39	-
# Feed stage*	36	-
# Solvent inlet stage*	5	-
Solvent/feed ratio	0.6	-
Reflux ratio (RR)	4.61	-
Solvent molar flow (recycle stream from C2)	2.4	kmol/s
Feed molar flow	4	kmol/s
Molar flow of recycle stream from C3	1.35	kmol/s
Condenser duty	87.86	MW
Reboiler duty	15.12	MW

Source: Ebrahimzadeh et al., [5]

Table 3. Solvent recovery column operating conditions

Parameter	Quantity	Unit
Pressure	2.3e6	Pa (abs)
# Column stages*	37	-
# Feed stage*	15	-
Reflux ratio (RR)	1.08	-

Condenser duty	32.1	MW
Reboiler duty	92.6	MW

* Numbered from the top of the distillation tower.
Source: Ebrahimzadeh et al., [5]

Table 4. Concentrator column operating conditions

Parameter	Quantity	Unit
Pressure	2.3e6	Pa (abs)
# Column stages*	43	-
# Feed stage*	10	-
Reflux ratio (RR)	2.9	-
Condenser duty	37.1	MW
Reboiler duty	20.9	MW

Source: Ebrahimzadeh et al., [5]

2.2. Metodology

Fig. 5 details the methodological procedure for the development of the ANN. As a first point, the simulation and validation of the process detailed in Fig. 1 is carried out based on the operating conditions of Tables 1-5. Subsequently, the ANN is designed and validated considering the inputs and outputs described in Figs. 3-5 and the constraints defined by the simulation. Finally, the functionality and predictive capacity of the ANN is evaluated through a graphic and statistical analysis.

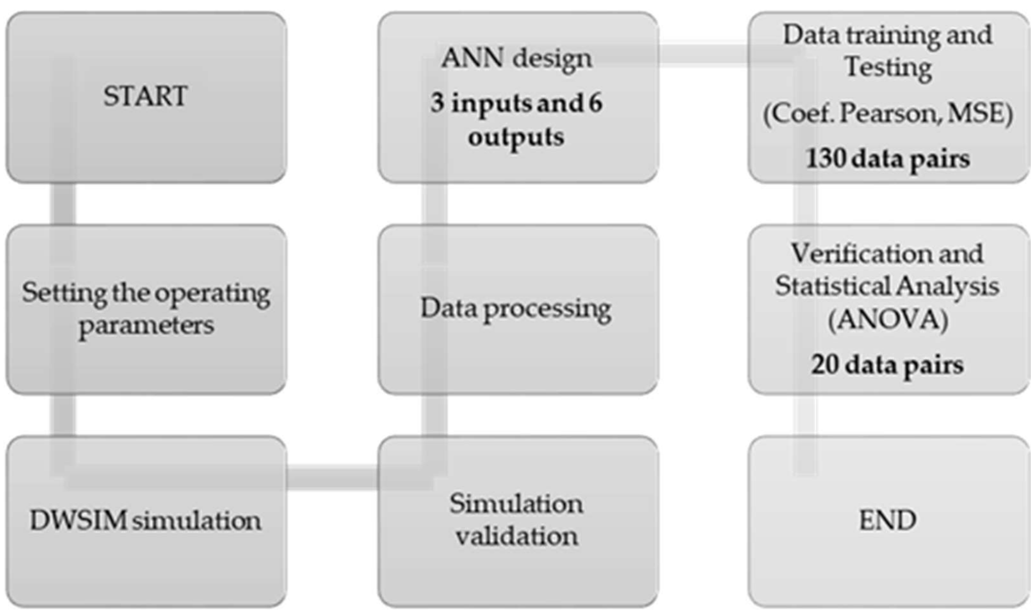

Figure 2. Methodological scheme of the designed ANN

2.3. DWSIM simulation

DWSIM (open-source chemical process simulator) is available for Windows, Linux, Android, macOS, and iOS and allows engineers to model process plants by using rig-orous thermodynamic and unit operations [32].

The distillation towers used for the simulation in Fig. 1 correspond to the "Chem-Sep Column" model. All flow streams operate with the Peng-Robinson (PR) properties package, while for the distillation towers the EOS / Predictive PR 78 thermodynamic models are adapted, which is one of the most widely used packages in hydrocarbon modeling and in principle are able to estimate the phase equilibrium and other ther-modynamic properties of a wide assortment of frameworks [33–35]. The conditions established in Tables 2-4 correspond to the operating conditions under which the pro-cess simulation was carried out. It is important to note that the ChempSep columns require the specification of only two operating parameters in addition to the pressure value.

The mathematical method, we used to find the convergence of the simulation process was Newton's Method for which a maximum of 100 iterations was established.

2.4. Design and training of the ANN

The ANN design (Figure 3) is based on three input parameters: pressure (C1, C2 and C3), temperature (inlet temperature to column C1) and solvent/feed ratio to column C1.These input variables were chosen because of the importance they represent in the quality of the final products and in optimization processes [9,10,36–38]. While six output parameters were considered: mole fractions of CO2 distillate and C2H6 bottom in C1 , C2H6 distillate and C3 bottom in (C2), and C2H6 both distillate and bottom in (C3).

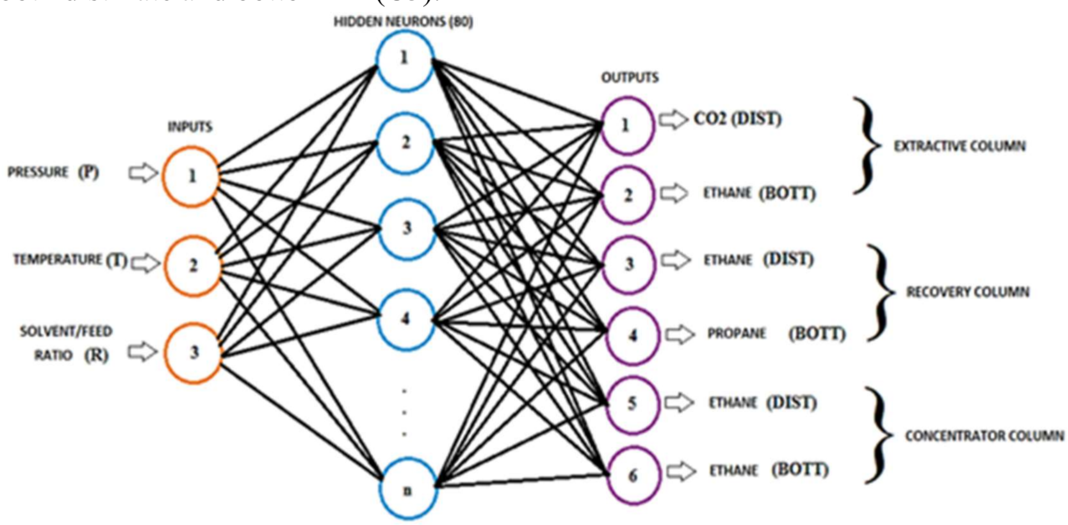

Figure 3. Schematic of the designed ANN

Based on the guidelines of Chen et al., [39], for network learning and training 70% of the total data pairs (90 data sets) were used, while 30% (40 data sets) were utilized to perform a testing to assess its level of learning. Figures 4 and 5 describe the inputs and outputs used in the ANN designed (Appendix A).

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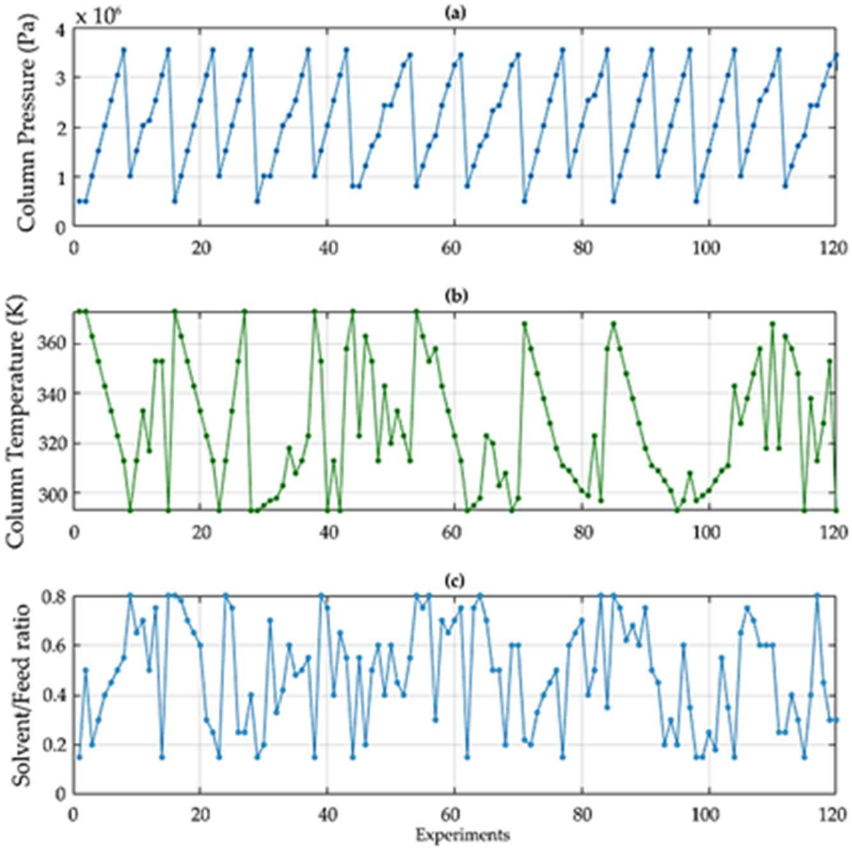

Figure 4. Inputs: (a) Columns pressure (Pa); (b) Columns temperature (K); (c) Solvent/feed ratio

The ANN training adjusts the weights of the connections between neurons for the ANN to make adequate predictions regarding the targeted output data. Validation measures the ANN's prediction errors to assess its performance. Testing process eval-uates the prediction of ANN using pairs of data that were not used in the training process [40].

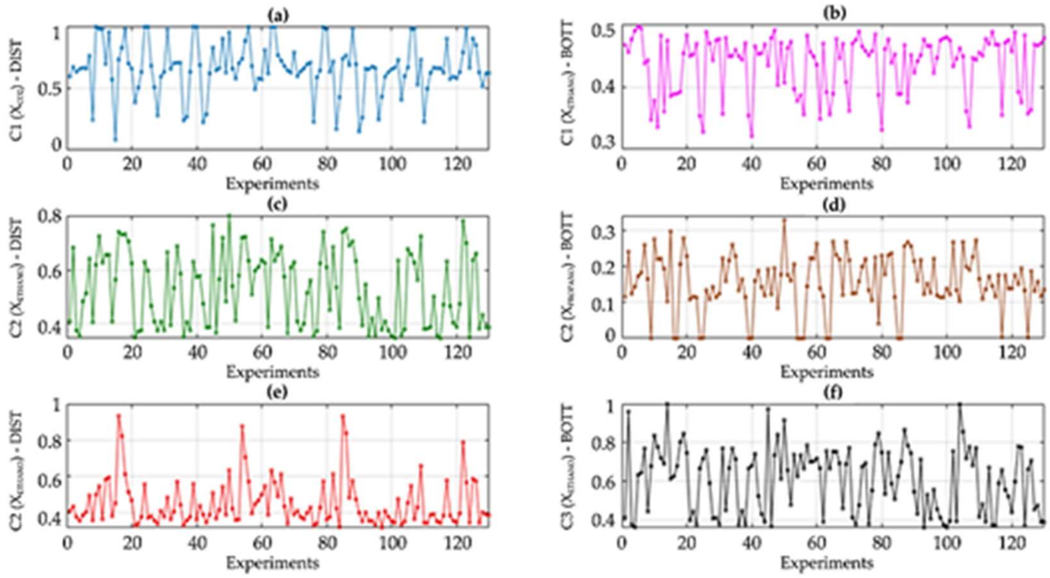

Figure 5. Outputs: (a) Mole fraction of CO2 distillate in C1; (b) Mole fraction of C2H6 bottom in C1; (c) Mole fraction of C2H6 distillate in C2; (d) Mole fraction of C3 bottom in C2; (e) Mole fraction of C2H6 distillate in C3; (F) Mole fraction of C2H6 bottom in C3.

The specific bibliography in ANN, suggests at least 50 points to predict quantities with regression algorithms [41–43]. In this respect, 130 pairs of data (with 3 inputs and 6 outputs) was generated, coming from the random variation of the operating parameters and/or performances selected for the study. Table 5 shows the range of variation of the inputs, that were chosen based on typical and extremes ranges of operation [9,10,36–38] . To ANN validation, we used the following performance indicators: mean square error (MSE) and regression coefficient (R) by means of Eq. (1) and Eq. (2) and additionally an ANOVA. In addition, the ANN performance was optimized through a trial-and-error procedure in order to minimize the MSE and maximize the correlation coefficients in the training, validation and testing stages.

$$MSE = \frac{1}{\mathrm{n}}\sum_{\mathrm{t}=1}^{\mathrm{n}}\left(y - y'\right)^2 \tag{1}$$

$$\mathrm{R} = \frac{n\sum_{i=1}^{n}(y'y) - [\sum_{i=1}^{n}y'][\sum_{i=1}^{n}y]}{\sqrt{[n\sum_{i=1}^{n}y^2 - [\sum_{i=1}^{n}y^2]\,[n\sum_{i=1}^{n}y'^2 - [\sum_{i=1}^{n}y'^2]}} \tag{2}$$

Where: n is the number of observations; y are the actual results (simulation out-puts); y ' are the predicted targets (ANN outputs).

Parameter	Pressure (Pa) C1, C2, C3	Temperature (K) inlet stream to C1	solvent/feed ratio (-) regulated in the splitter
*Range	506625 – 3.54e6	2.93.15 – 373.15	0.15 – 0.8

* Less or greater than the established ranges, the simulation does not run.

3. Results and discussion

This section presents the analysis and discussion of the process simulation and the design, training and validation of the ANN.

3.1. Simulation validation

The validation of the simulation process in DWSIM was carried out with the comparison of the study developed in ASPEN PLUS by Ebrahimzadeh et al. [5]. Table 6 summarizes the percentage errors of the mole fractions of interest in their respective distillation columns, which do not exceed 3%. This percentage of error is justified by the presence of very small traces of other constituents in the distillate and the bottom cur-rents, which are considered negligible.

3.2. ANN topology

This section describes the design and structuring of the ANN by analyzing the correlation coefficient (R) and the mean square error (MSE).

3.2.1. Selection of ANN Training Algorithm

For the design and structure of the ANN, three training algorithms were used: Levenberg - Marquardt (LM), Bayesian regularization (BR) and scaled conjugate gradi-ent backpropagation (SCG). According to specialized bibliography, these algorithms minimize the MSE to a greater extent compared to other algorithms available in the literature [44–46]. As in other prediction studies [47,48], the R and MSE values (Table 7) were evaluated for the 3 algorithms studied by varying the number of neurons in the hidden layer.

Table 6. Simulation validation (mole fraction)

Column	Parameter	Aspen Plus [Ebrahimzadeh et al., 2016]	DWSIM	Error (%)
Extractive (C1)	CO_2 distillate	0.956	0.953	0.24
	C_2H_6 bottom	0.396	0.406	2.60
Recovery (C2)	C_2H_6 distillate	0.799	0.801	0.29
	C_3 bottom	0.337	0.328	2.59
Concentrator (C3)	C_2H_6 distillate	0.538	0.554	2.98
	C_2H_6 bottom	0.997	0.975	2.25

Table 7. Pearson's correlation coefficient (R) and root mean square error (RMSE) values for trial and error using Levenberg–Marquardt (LM), Bayesian regularization (BR) and scaled conjugate gradient backpropagation (SCG) algorithms.

# hidden	LM		BR		SCG	
neurons	R Global	MSE	R Global	MSE	R Global	MSE
20	0.930	0.0074	0.959	0.00187	0.901	0.0129
40	0.926	0.0076	0.948	0.00181	0.899	0.0095
60	0.916	0.0065	0.873	5.81 E-07	0.895	0.0072
80	0.889	0.0096	0.955	0.00160	0.864	0.0078
100	0.961	0.0068	0.901	1.29 E-11	0.843	0.0033

After the training process, the results detailed in Table 8 conclude that the most suitable and robust model to predict the output targets is BR (MSE min = 1.29E-11, R max = 0.955). The advantage of a BR algorithm is its ability to predict complex relationships and its ability to make decisions less biased [44,49,50].

The computational cost of the BR algorithm is higher, compared to other training algorithms, however, it gives rise to good generalizations for difficult, small or noisy data sets. Furthermore, it shows a better performance of the predictive capacity in contrast to the Levenberg-Marquardt algorithm. The advantage lies in its ability to handle potentially complex relationships, which means that it can be used in quantita-tive studies to provide a robust model[44].

3.2.2. Selection of the number of neurons in the hidden layer

The determination to the optimal neuron's number it's useful to conduct trials in determining required local minimum in the error surface, and oscillations in R [2].

According to the analysis in Table 8, when 20 and 80 neurons are used in the hidden layer, the best R values are obtained in the testing and global phase. For 20 neurons the R value in testing is 0.846 and for 80 neurons 0.841, while the MSE values for 20 and 80 neurons are: 0.0018765 and 0.0001609. The results would indicate that the optimal number of neurons is 80. As seen in Table 9, the lowest percentage error (< 9 %) occurs when 80 neurons are used in the hidden layer.

Table 8. R and MSE values for determining the optimal number of neurons in the hidden layer

# hidden neurons	R Training	R Testing	R Global	MSE
20	0.979	0.846	0.959	0.00187
40	0.979	0.802	0.948	0.00181
60	0.999	0.640	0.873	5.81 E-07
80	0.978	0.841	0.955	0.000160
100	0.999	0.625	0.901	1.29 E-11

Table 9. Percentage error (%E) values for determining the optimal number of neurons in the hidden layer

# hidden neurons	%E CO_2 Distillate C1	%E C_2H_6 Bottoms C1	%E C_2H_6 Distillate C2	%E C_3 Bottoms C2	%E C_2H_6 Distillate C3	%E C_2H_6 Bottoms C3
20	66.75	12.15	24.63	16.72	42.95	39.01
40	54.63	11.26	29.05	39.84	54.56	27.58
60	77.54	11.29	28.07	41.68	53.86	35.70
80	8.03	2.54	4.18	8.53	5.90	6.09
100	62.90	13.27	39.75	26.46	85.03	50.69

The ANN (perceptron type) was designed with MATLAB NNTOOL version R2018a and from the analysis developed, it is defined that the structure of the ANN is composed of: three (3) input neurons, a hidden layer with 80 neurons and six (6) output neurons. According to the study developed by Abiodum et al.,[51] a hidden layer may be sufficient for prediction in most ANN applications.

3.2.3. ANN training and testing

Table 10 shows the MSE values for the training phase and testing phase of the ANN. For the validation phase there are no results. This is justified because ANNs using the BR algorithm are more robust models and can reduce or eliminate the need for validating, taking advantage of these data during training. The MSE values for the training and testing phase are: 0.0036 and 0.0222 respectively, indicating that the ANN performs adequately and that the predictions are made with sufficient accuracy. Figure 6 shows the evolution of the mean square error (MSE) during the training phase, with a final MSE of 0.0036. The MSE performance function for the training data (train) is very close to zero, indicating that the predictive capability of the network is very good.

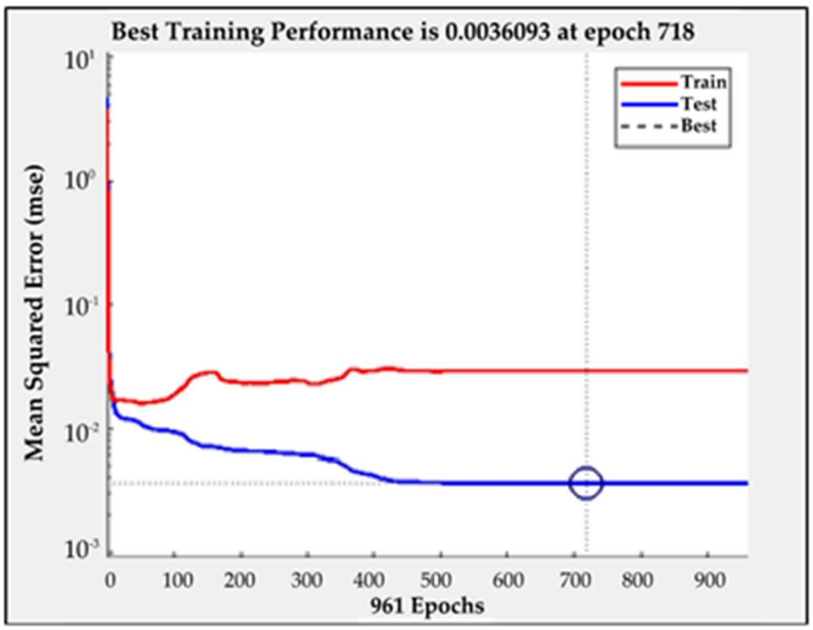

Figure 6. ANN training performance (MSE)

On the other hand, as seen in Fig.7, there is a moderate dispersion between the outputs and targets of the ANN in both the training and test phases. However, the R values for the training and testing phase are 0.98 and 0.84 respectively, and the overall R value of 0.96 which indicates that the outputs and targets have an acceptable correlation. The closer the R value is to 1, the better the performance of the ANN. To validate the ANN, the decision was made that the R value should be in the range of 0.95 to 1 and the MSE lower than 0.025.

Table 10. Mean square error of ANN designed

PHASE	MSE
trainPerfomance (training)	0.0036
testPerfomance (testing)	0.0222

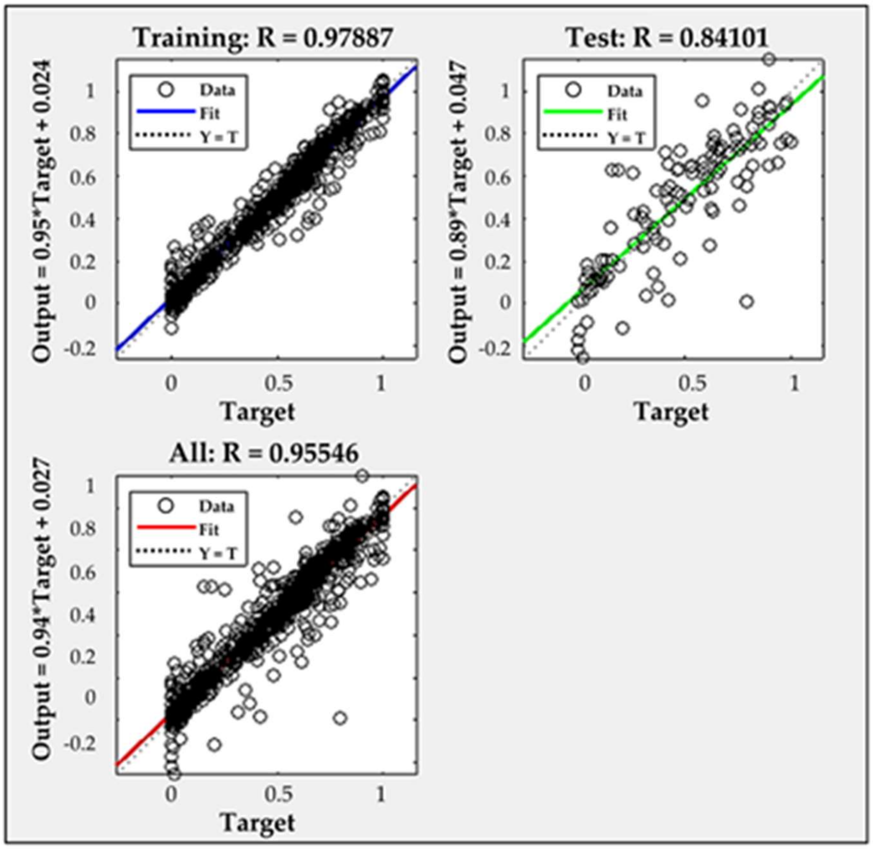

Figure 7. Regression coefficient R for ANN training and testing

3.3. Model prediction of CO2, C2H6 and C3 mole fractions in the extractive, solvent recovery and concentrator columns.

Figures 8 -9, 10-11 and 12-13 show the overlap between the experimental values (obtained by simulation) and the predictions (obtained by ANN) in the extractive column, solvent recovery and concentrator, respectively. It can be seen that the com-parisons obtained in the three columns are relatively equal. The developed model clearly approximates the observational data (simulation) proving in this way that the ANN constitutes a robust and adequate model for the prediction of C2H6 and CO2 con-centrations and that it can be applied in CO2- C2H6 azeotrope separation process in en-hanced oil recovery processes.

Based on the analysis of Figures 8-13, the average percent error (%E) of the pre-dictions are: 5.036% (CO2 in the distillate) and 2.55 % (C2H6 in the residue) in the ex-tractive column (C1); 4.19% (C2H6 in the distillate) and 4.18% (C3 in the residue) in the solvent recovery column (C2); 5.91% (C2H6 in the distillate) and 6.09% (C2H6 in the res-idue) in the concentrator column (C3).

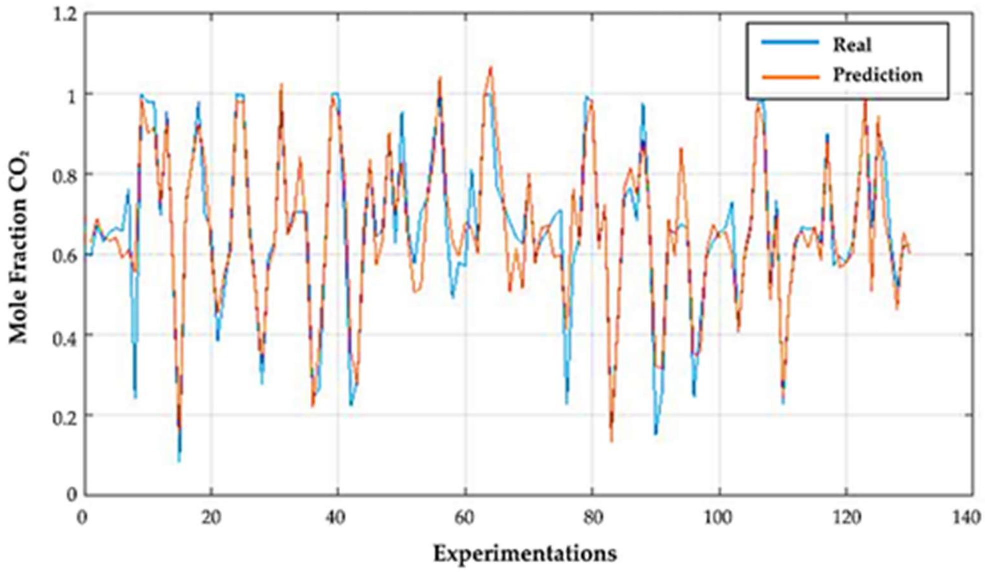

Figure 8. Comparison of DWSIM (real) and ANN (predicted) results in extractive column distillate (CO2 mole fraction)

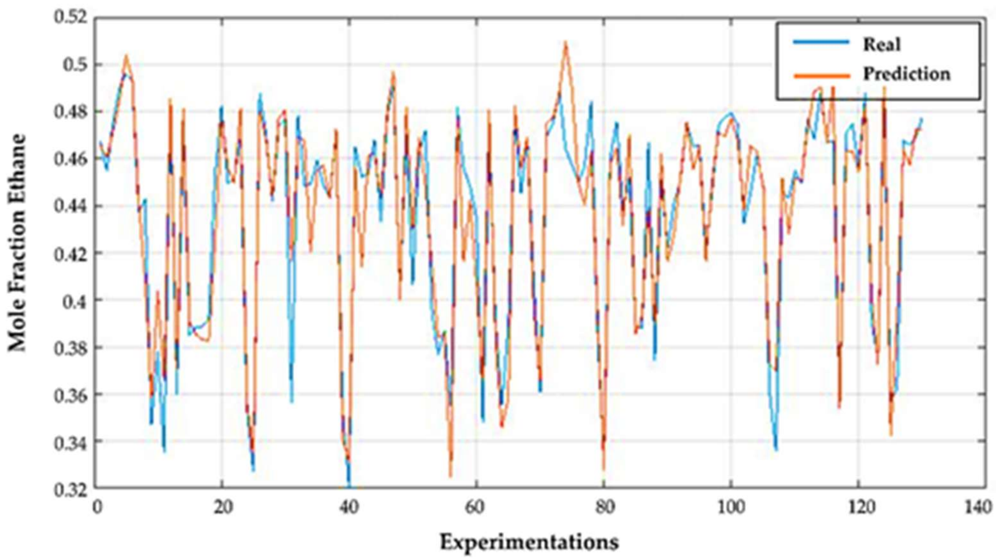

Figure 9. Comparison of DWSIM (real) and ANN (predicted) results on the extractive column bottoms (C2H6 mole fraction)

3.4. ANN model verification

The ANN predictive capacity of the concentration of CO2, C2H6 and C3 in the ex-tractive, solvent recovery and concentrator column was tested with a set of 20 random input data (P, T and solvent / feed ratio) unknown by the ANN. The results show an overlap between the experimental data and the predictions.Tthis indicates that ANN has a good predictive capacity of the mole fractions of distillates and residues of the distillation columns (Fig.14).

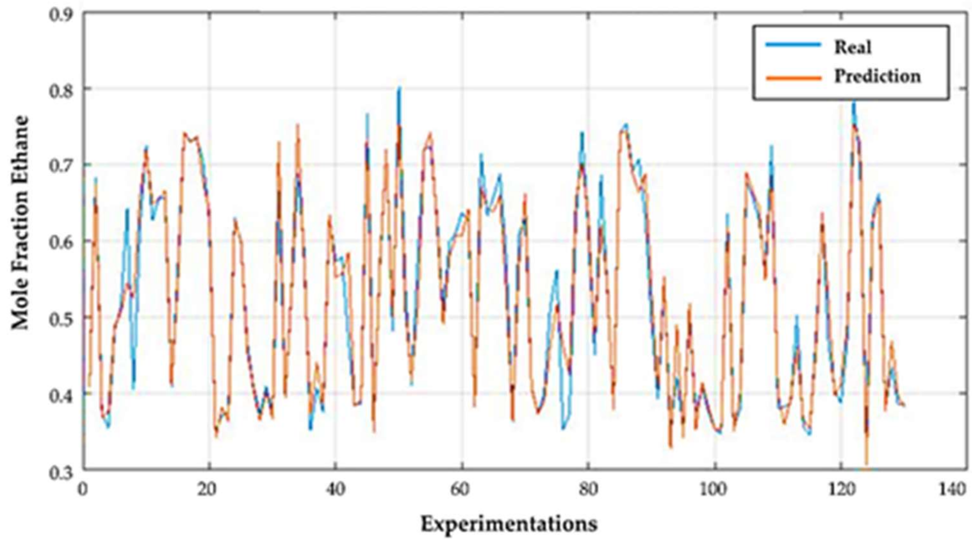

Figure 10. Comparison of DWSIM (real) and ANN (prediction) results in solvent recovery column distillate (C2H6 mole fraction)

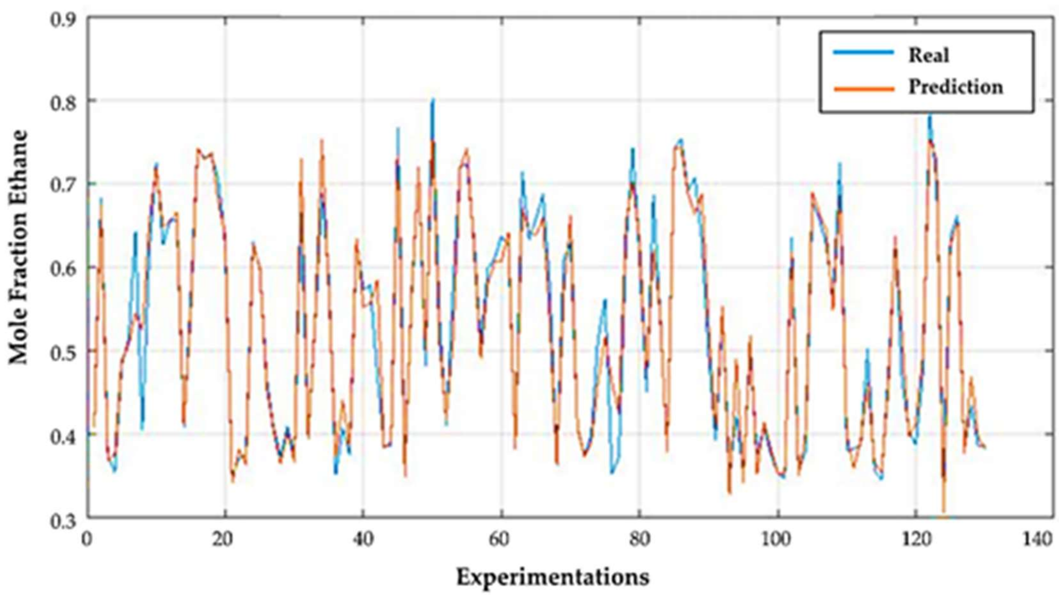

Figure 11. Comparison of DWSIM (real) and ANN (prediction) results on solvent recovery column bottoms (C3 mole fraction)

In this research we used the functions ANOVA and Kruskal-Wallis [52] using SPSS 22.0, to statistically validate the ANN. Table 11 shows the results from ANOVA and, for all cases, P-values (probability value in statistical significance tests) is greater than 0.05, while Table 12 summarizes the results of the Kruskall-Wallis test that was performed to validate of outliers of the predictions. This test also verifies that the P-value is greater than 0.05 in all cases, indicating that there is no statistically significant difference be-tween the means of the observations and the predictions. These statistical tests reveal that the ANN constructed is statistically valid for the prediction of the mole fractions of $CO2$, $C2H6$ and C3 in columns C1, C2 and C3, with a confidence level of 95%.

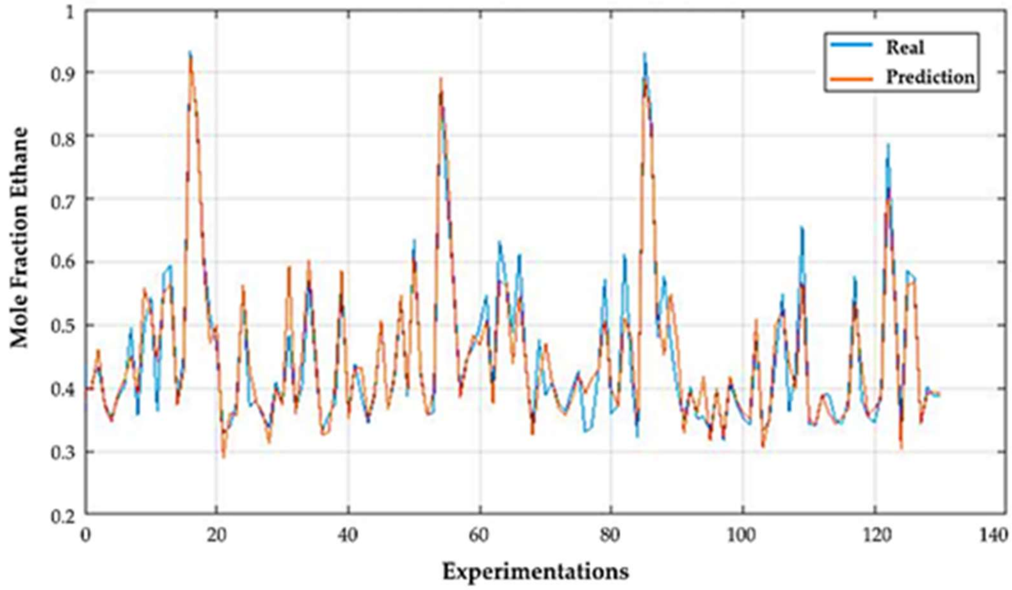

Figure 12. Comparison of DWSIM (real) and ANN (predicted) results in the concentrator column distillate (mole fraction of C2H6)

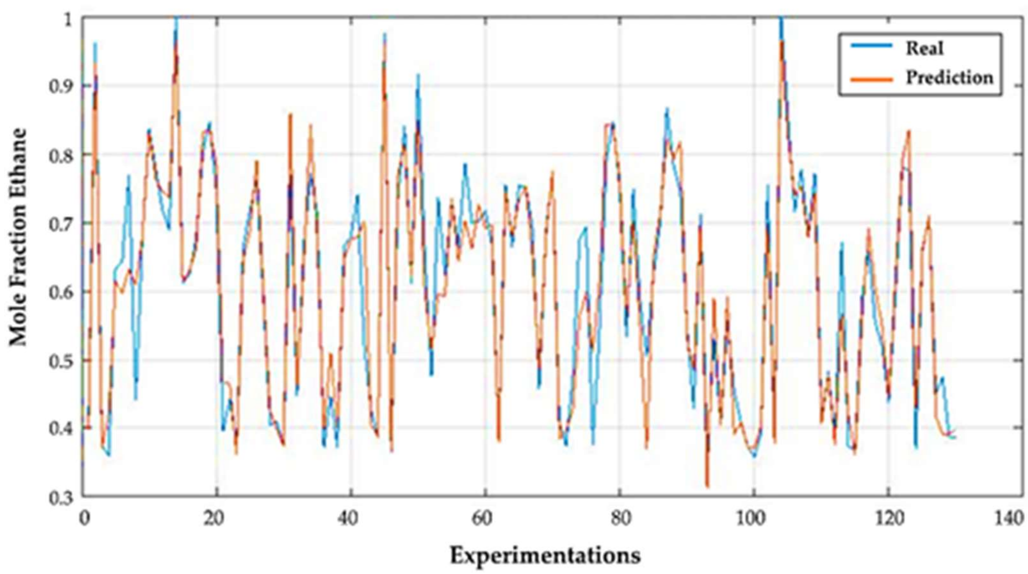

Figure 13. Comparison of DWSIM (real) and ANN (prediction) results on the concentrator column residue (mole fraction of C2H6)

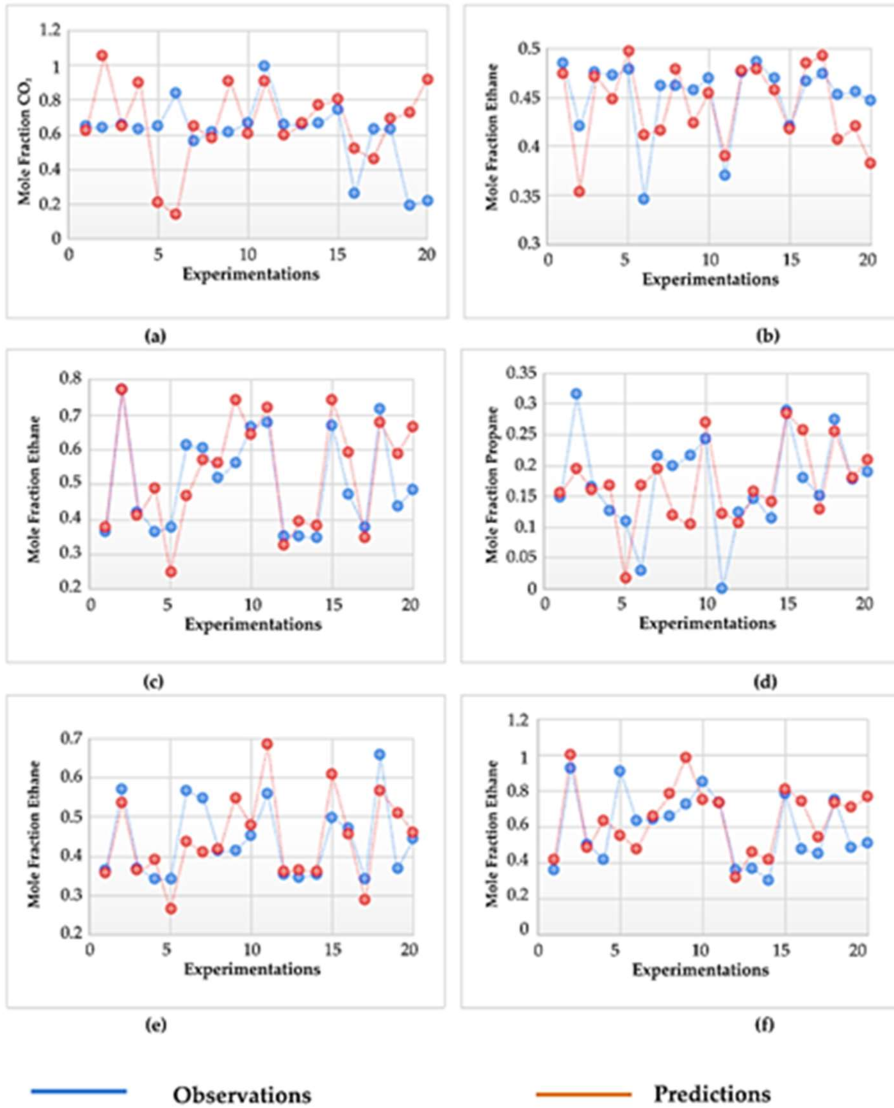

Figure 14. Comparison between observations and predictions. Extractive column: a) CO2 mole fraction (distillate), b) C2H6 (bottoms); Solvent recovery column: c) C2H6 (distillate), d) C3 (bot-toms); Concentrator column: e) C2H6 (distillate), f) C2H6 (bottoms).

Table 11. ANOVA

Source	Sum of squares	DOF	Mean square	F-Value	P-value
Mole fraction of CO_2 in the extractive column distillate					
Inter groups	0.0343	1	0.0343	0.78	0.383
Intra groups	1.6778	38	0.0441		
Total (Corr.)	1.7122	39			
Mole fraction of C_2H_6 in the extractive column bottoms.					
Inter groups	0.0010	1	0.0010	0.70	0.407
Intra groups	0.0572	38	0.0015		
Total (Corr.)	0.0583	39			

Mole fraction of C_2H_6 in solvent recovery column distillate.					
Inter groups	0.0083	1	0.0083	0.37	0.549
Intra groups	0.8684	38	0.0228		
Total (Corr.)	0.8767	39			
Mole fraction of C_3 in the solvent recovery column bottoms.					
Inter groups	2.38E-05	1	2.38E-05	0.00	0.946
Intra groups	0.1983	38	5.21E-03		
Total (Corr.)	0.1983	39			
Mole fraction of C_2H_6 in the concentrator column distillate.					
Inter groups	1.79E-04	1	1.79E-04	0.02	0.897
Intra groups	0.4027	38	0.0105		
Total (Corr.)	0.4028	39			
Mole fraction of C_2H_6 in the concentrator column bottoms.					
Inter groups	0.0321	1	0.0321	0.88	0.353
Intra groups	1.3843	38	0.0364		
Total (Corr.)	1.4164	39			

Table 12. Kruskal-Wallis Test

	Average Range	Statistical	P-value
Extractive column			
CO_2 Observation (Distillate)	19.05	0.615	0.433
CO_2 Prediction (Distillate)	21.95		
C_2H_6 Observation (Bottoms)	21.75	0.457	0.499
C_2H_6 Prediction (Bottoms)	19.25		
Solvent recovery column			
C_2H_6 Observation (Bottoms)	19.25	0.457	0.499
C_2H_6 Prediction (Bottoms)	21.75		
C_3 Observation (Bottoms)	20.85	0.035	0.849
C_3 Prediction (Bottoms)	20.15		
Concentrator column			
C_2H_6 Observation (Bottoms)	19.95	0.088	0.767
C_2H_6 Prediction (Bottoms)	21.05		
C_2H_6 Observation (Bottoms)	18.75	0.896	0.344
C_2H_6 Prediction (Bottoms)	22.25		

4. Conclusions

The mole fractions of an alternative extractive distillation system for the separation of CO2-C2H6 azeotropes in enhanced oil recovery processes were predicted using an ANN based on the process simulation in DWSIM in this study. The ANN developed has 80 hidden neurons and was trained with a base of 130 pairs of data with 3 input variables (neurons): pressure (P), temperature (T), and solvent/feed ratio. Itis capable of predict-ing 6 output variables (neurons): the mole fraction of CO2 in the distillate and C2H6 in the bottoms of the extractive column (C1), the mole fraction of C2H6 in the distillate and C3 in the bottoms of the solvent recovery column (C2), and the mole fractions of C2H6 in the distillate and in the bottoms of the concentrator column (C3).

The Bayesian regularization approach was used to train the ANN, which has an MSE of 0.0036 and a total R of 0.95546. A comparison statistical analysis (ANOVA and Kruskall-Wallis) between the data (DWSIM) and the values predicted by the neural network was also used to validate the ANN. Statistical tests show that the ANN accu-rately predicts the mole fractions at the outputs with a 95% significance level.

According to the findings, the ANN developed in this work can be used as a pre-diction tool for improving natural gas sweetening operations. For instance, real oper-ating parameters of the described process must be used as input, apply them in situ and verify the predictions at the control points (outputs of the ANN). Subsequently, vali-dated in the plant and coupled to the existing control process, the energy optimization of the process can be promoted by coupling genetic optimization algorithms to the network (hybrid technologies). Optimization studies in a real plant will be the subject of future research.

Supplementary Materials: The following are available online at www.mdpi.com/xxx/s1, Figure S1: title, Table S1: title, Video S1: title.

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Appendix A

Database generated for simulation parameters, training and validation of the ANN

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