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Investigation and optimization of olefin purification in methanol-to-olefin process based on machine learning approach coupled with genetic algorithm

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Abstract−In this study, the goal was to develop a robust model for prediction of the performance of a purification section of methanol to olefin (MTO) process based on machine learning approach. The optimum operating conditions were determined by performing a genetic algorithm as an optimization technique. Finding the optimum conditions caused a considerable decrease both in fixed capital investment (FCI) and working capital investment (WCI). To do this, the separation section of MTO process was investigated and modelled through artificial intelligence (AI). This separation section of MTO process is comprised of three columns: C2-stripper, deethanizer, and C3-strippmer. For each column, three operative parameters (number of stages, reflux ratio, and pressure) were selected and investigated. The performance of columns was assessed through monitoring the purity of products in top stream and energy consumption. The experimental layout was designed using response surface methodology (RSM). And the obtained data were used to develop artificial neural network (ANN) models for each column. Several structures of ANNs were investigated to select the optimum model parameters. The observations show good agreement between the real and predicted data.

Keywords: Separation, Olefin, Artificial Neural Network, Genetic Algorithm

INTRODUCTION

Light olefins are key materials in chemical industries with several products with various usages being made from olefins and polyolefins [1,2]. Widespread demand for light olefins motivates the petrochemical industries to establish and develop new production routes. Olefins can be produced from naphtha pyrolysis in steam cracker furnace. In this route, high temperature of furnace breaks hydrocarbon bonds and causes formation of small and unsaturated molecules such as olefins [3]. Methanol to olefin (MTO) process is another route that can be used as an alternative to hydrocracking and other high energy consuming methods. Since methanol is produced from natural gas and coal, the relatively low price of these feedstocks is an important advantage of MTO. Catalyzer lifetime and olefin selectivity are two main issues in MTO [4]. To improve the olefin production yield, the by-products of C_4^+ can be separated and entered into a cracking reactor [5]. Recently, a new route was developed based on coal to olefin (CTO) [6]. This route would be beneficial in the regions where coal is available.

As mentioned, selecting an appropriate catalyst in MTO process is critical. Zeolite and zeotype catalysts show acceptable performance in MTO process [7]. Rostamizadeh et al. assessed the performance of high silica H-ZSM-5 nanocatalyst in the MTO process. They reported methanol conversion and propylene selectivity of 100% and 43%, respectively [4]. In developing optimum MTO catalysts, high olefin production rate, high selectivity, catalyst stability, minimum side products, minimum catalyst cost, minimum waste emission, and ease of scaling up should be considered [5]. Shirbakht et al. investigated α-olefin polymerization using a conventional Ziegler-Natta catalyst. They reported that by increasing the monomer length from C4 to C10 and introduction external donor led to decrease of catalyst activity [8].

In chemical industries, distillation columns are used widely as a separator. In distillation columns light products are evaporated and removed as condensate on the top of the column. Heavy products are collected at the bottom of column. The cost of distillation columns is the main part of the total costs of the separation section of any process. Energy consumption in a distillation column is high. Due to high energy consumption of multicomponent separation in a distillation column, energy integration is a critical aspect. The operative conditions of these columns influence the economic aspects of the projects. It is clear that the aim of each design procedure is to minimize the total cost of the production process. Generally, optimum design and operating the separation units is of interest in chemical engineering.

In this study, the focus is on determining the optimum operative condition of a distillation column series used to split a multicomponent mixture into pure ethylene and propylene. In previous work, the distillation columns in MTO process were simulated and optimized using ASPEN-HYSYS coupled with statistical approach [9].

In a conventional method, to investigate the effect of operative parameters, one factor is changed while the other factors are fixed. And the change of response vs. this parameter is tracked and investigated. There are several bottlenecks to this technique that inhibit using it in practical studies. However, this technique is not able to consider interaction between parameters. Further, the number of experiments in this method is high, which is time and cost consum-

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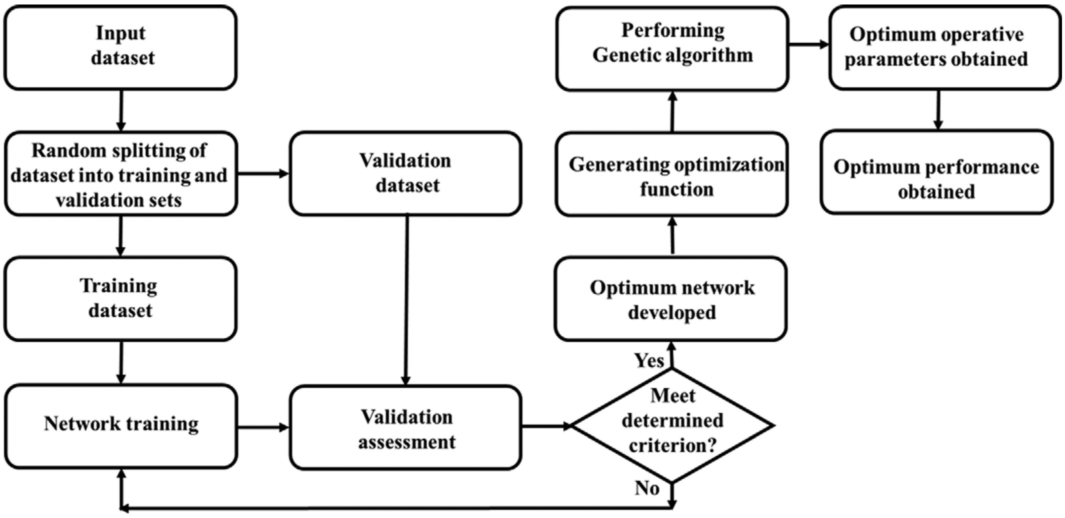

Fig. 1. Implemented algorithm to find column optimum performance.

ing. To face these limitations, design of experiment (DoE) techniques, such as response surface methodology (RSM), are used. In RSM, a polynomial model is used. In this way the factors of a polynomial equation are set to fit the data.

In this work, the DoE layout was performed using RSM. This technique is used in several experimental works due its ability in screening the unimportant factors and decreasing the number of runs in experiments [10-13].

Although this technique is able to consider the interactions between factors and conducting the experiment with lower runs, it has several limitations. Usually, the relation between input parameters (inputs) and responses (outputs) is complicated and explaining this relation with polynomial equation is hard and sometimes impossible. Artificial intelligence (AI) provides a useful tool to build a model based on a set of neurons, their weights, and connections with the ability of training to find the complicated relation between input and output that could not be done by other methods.

In Fig. 1 the procedure used in this work to find the optimum performance of separation columns has been shown. In first step, data of previous work are used to develop an artificial neural network (ANN) model with the ability to predict the composition of top streams and power consumption of each distillation column. To do this, input data are split into training and validation dataset. The training dataset is used to tune the network parameters. Then, the validation dataset is used to test the model prediction ability. After the optimum model parameters are selected, the developed model is used to generate an optimization function. Finally optimization procedure is performed through genetic algorithm based on the optimization function. Accordingly, optimum performance condition is obtained.

METHODOLOGY AND PROCEDURE

1. MTO Process

The catalytic mechanism of the MTO process is complicated. There is an intermediate substance named dimethyl ether (DME) that is converted to desirable products through a series of chemical reactions. The overall reaction is exothermic and operates at temperature and pressure of 350-550 °C and 2-3 bar, respectively. The aim of this work is not to focus on the chemical reaction of MTO process. Further, deep explanations about the reaction mechanisms have been addressed in previous work [9]. Table 1 shows ten elementary reactions introduced by Taheri et al. [14].

Table 1. The reactions involved in MTO process [14]

	Reaction	Order of reaction
1	$2CH_3OH \xleftrightarrow{K_1} CH_3OCH_3 + H_2O$	1
2	$2CH_3OCH_3 \xrightarrow{k_2} C_2H_4 + 2CH_3OH$	1
3	$2CH_3OH \xrightarrow{k_3} C_2H_4 + 2H_2O$	elementary
4	$C_2H_4 + CH_3OCH_3 \xrightarrow{k_4} C_3H_6 + CH_3OH$	elementary
5	$C_3H_6 + CH_3OCH_3 \xrightarrow{k_5} C_4H_8 + CH_3OH$	elementary
6	$C_4H_8 + CH_3OCH_3 \xrightarrow{k_6} C_5H_{10} + CH_3OH$	elementary
7	$CH_3OH \xrightarrow{k_7} CO + 2H_2$	elementary
8	$CO + H_2O \xrightarrow{k_8} CO_2 + H_2$	elementary
9	$CH_3O + H_2 \xrightarrow{k_9} CH_4 + H_2O$	elementary
10	$C_2H_4 + H_2 \xrightarrow{k_{10}} C_2H_6$	elementary

Methanol is fed into the MTO reactor, a fluidized bed type. The products from this reactor are comprised of ethylene, propylene, untreated methanol, ethane, propane, catalyst particles, carboxylic acids, heavy hydrocarbons, and water. In the next step, the main portion of the olefins is separated from the others. After that, the olefin stream is split by passing through two separation sections. In the first section, ethylene, ethane, propylene, and propane are separated from the others and fed into the second separation section. This section comprised of three distillation columns is aimed to produce pure ethylene and pure propylene. In previous work, the performance of these columns was assessed, simulated and modelled by ASPEN-HYSYS coupled with statistical approach, and the optimum conditions for each columns were found out [9]. In this work, these columns are modelled using neural networks and the optimum operational conditions are determined by genetic algorithm based on the developed models. These three columns were named C2 stripper, deethanizer, and C3 stripper. In each column, three operative parameters (number of stages, value of reflux ratio, and column pressure) were selected for investigation and experimental layout was designed by using RSM.

2. Neural Networks

ANN is a method of processing information that is inspired by the biological nervous system—a tool that processes data like the human brain. These networks are made up of a large number of interconnected processing elements called neurons. An artificial neuron is made up of inputs, outputs, weights, biases, and an activation function. Weights and biases are randomly assigned. Weights will change during the network training process. Weight change is made in such a way that the neuron outputs approach the actual values. These neurons systematically work together to solve the complicated non-linear problems. ANNs learn how to figure out the input-output relations by a series of existing data, just like humans. The ability to learn is the most important feature of an intelligent system. A system with learning ability is more flexible and easier to program. In this methodology, the network is trained by creating a series of connections between the neurons and applying a training algorithm to it. Overall, with the use of neural network-based learning methods, the analysis of complex data progresses step by step. Finally, we will achieve more flexible capabilities in ANN-based models and their application in different field of science and engineering [15-18]. There are two types of neural networks: feedforward and feedback. In this work, a feedforward neural network is developed. In feedforward neural networks the response path is always forward and there is no return to the neurons of its previous layers. In this type of network, signals are only allowed to pass through a one-way path (from input to output). Thus, the output of each layer only affects the properties of next layer and does not change its own layer. So, there is no feedback. In Fig. 2, the structure of this type of network with three inputs, two outputs, and two hidden layers is shown.

In this work, the network was trained using supervised learning methods. In this method, the learning algorithm is given a set of data pairs. Each data contains input to the network and the target. After the data are entered into the network, the values of network outputs are compared with the targets. Then the learning error is calculated and used to adjust the network parameters (weights). This process is repeated while the learning error approaches a determined value. In this method, if the same input is given to the network next time, the network output will be closer to the target output. In Fig. 3, the block flow diagram (BFD) of training process is shown.

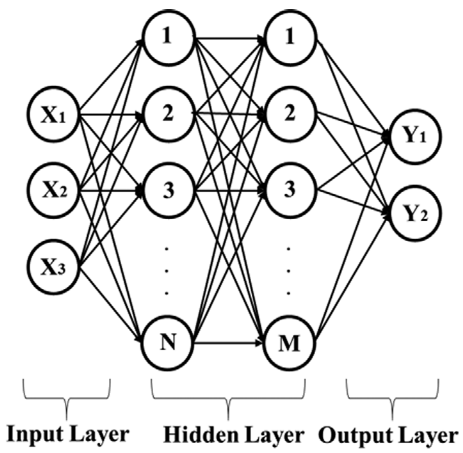

Fig. 2. Structure of multilayer perceptron.

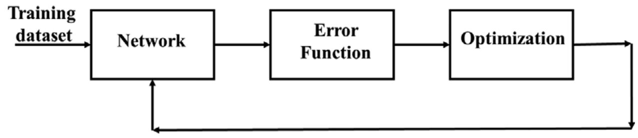

Fig. 3. Block flow diagram (BFD) of training process.

In the current study, the Levenberg-Marquardt (LM) optimization algorithm was used to update the values of bias and weight during the training procedure. The number of hidden layer was one and maximum epoch number was fixed at 100.

The Leave-one-out method as a cross-validation technique was used to prevent over-fitting during training the model. In this approach, one of the observations is removed from the dataset and is selected as validation, while the other observations are labelled as training dataset and the model is trained using it. Next, the sec-

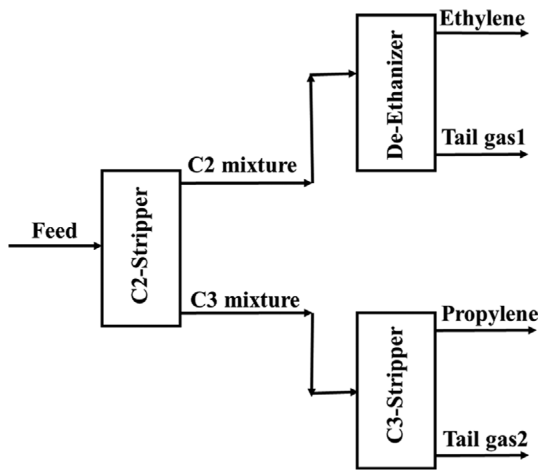

Fig. 4. BFD of second separation section.

Table 2. The range of operative variables in different distillation columns

Column	Parameter	Range
C2-Stripper	Column stage	9-48
	Reflux ratio	0.26-2.44
	Column pressure (kPa)	991-3010
De-Ethanizer	Column stage	20-70
	Reflux ratio	0.26-2.44
	Column pressure (kPa)	991-3010
C3-Stripper	Column stage	50-200
	Reflux ratio	0.26-2.44
	Column pressure (kPa)	991-3010

ond observation is selected as the validation one and this procedure is repeated N times where N is the number of observations (dataset size). Finally, the average of R^2 values is introduced as validation-R^2.

RESULTS AND DISCUSSION

In previous work, the second separation section of MTO process was investigated and simulated by ASPEN-HYSYS coupled with DoE methodology [9]. In Fig. 4 the BFD of second separation section is shown. There, the separation section includes three columns: C2-stripper column, deethanizer, and C3-stripper column. The conceptual design of this work, as well as the data of feed stream entered into C2-stripper column, was obtained from the work done by Yu and Chien [19]. The ranges of the operational variables for different distillation columns are presented in Table 2.

1. C2-Stripper Column

In a C2/C3 distillation column the sum of absolute values of condenser and reboiler consumption power as well as the mole fraction of C2 (ethylene and ethane) in top stream were selected as the indexes of separation performance. The distillation column was simulated using ASPEN-HYSYS. To assess the effects of operative variables on separation yield, RSM was selected for designing the experimental layout.

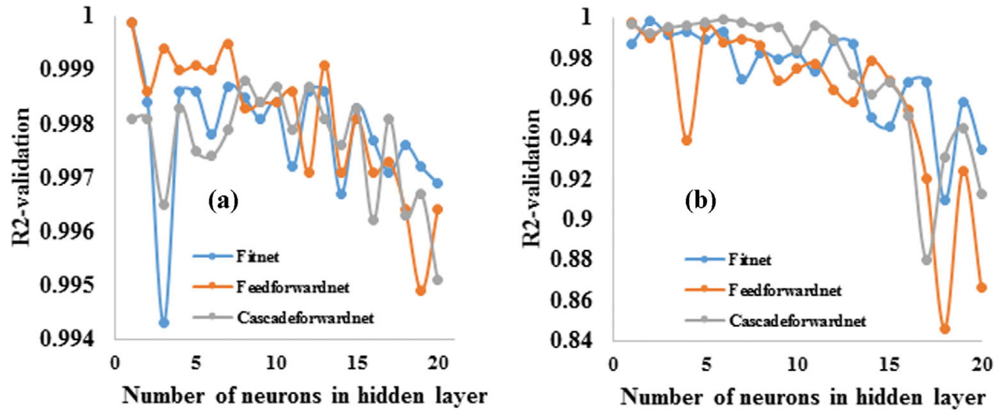

Fig. 5. R^2-validation values vs. number of neurons in hidden layer for (a) C2 fraction and (b) power consumption.

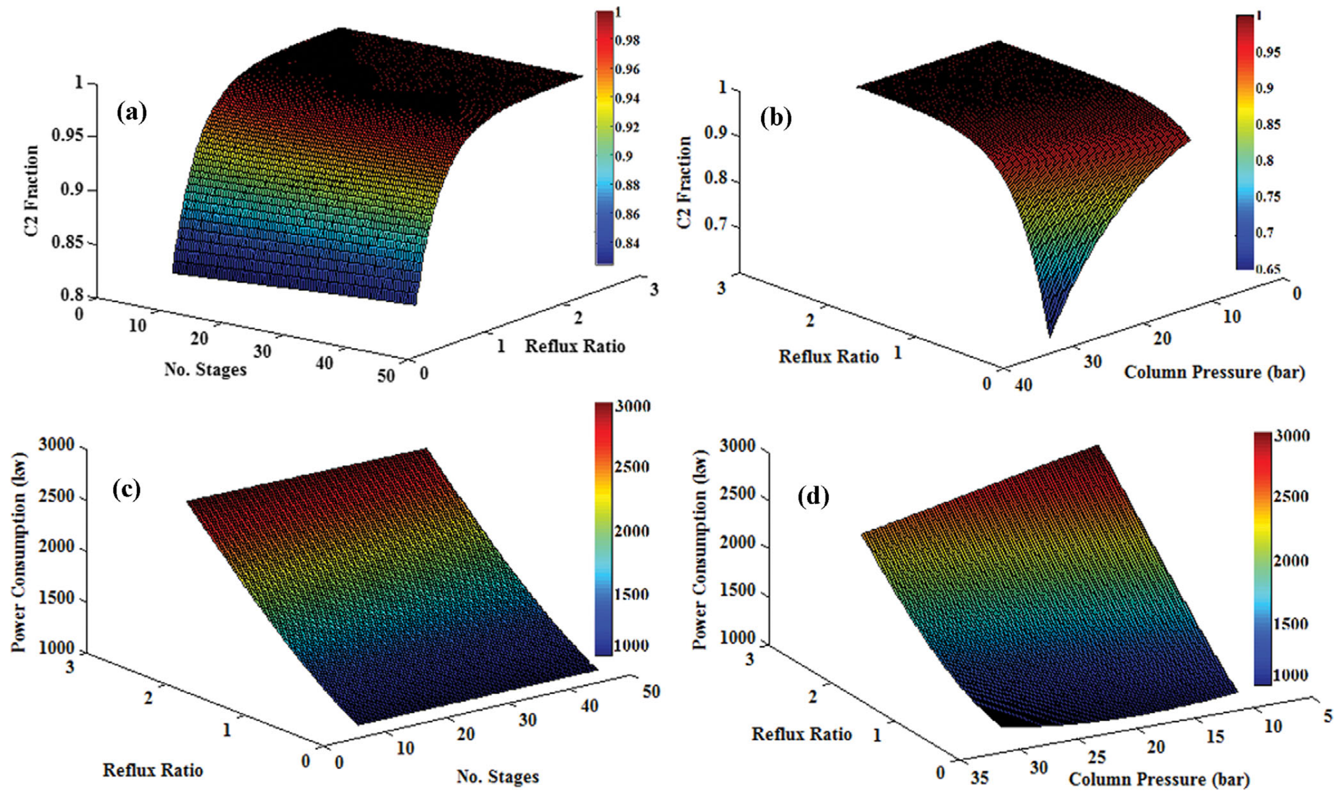

Fig. 6. 3D plot of the predicted values as a function of No. stages, reflux ratio, and pressure of the distillation column.

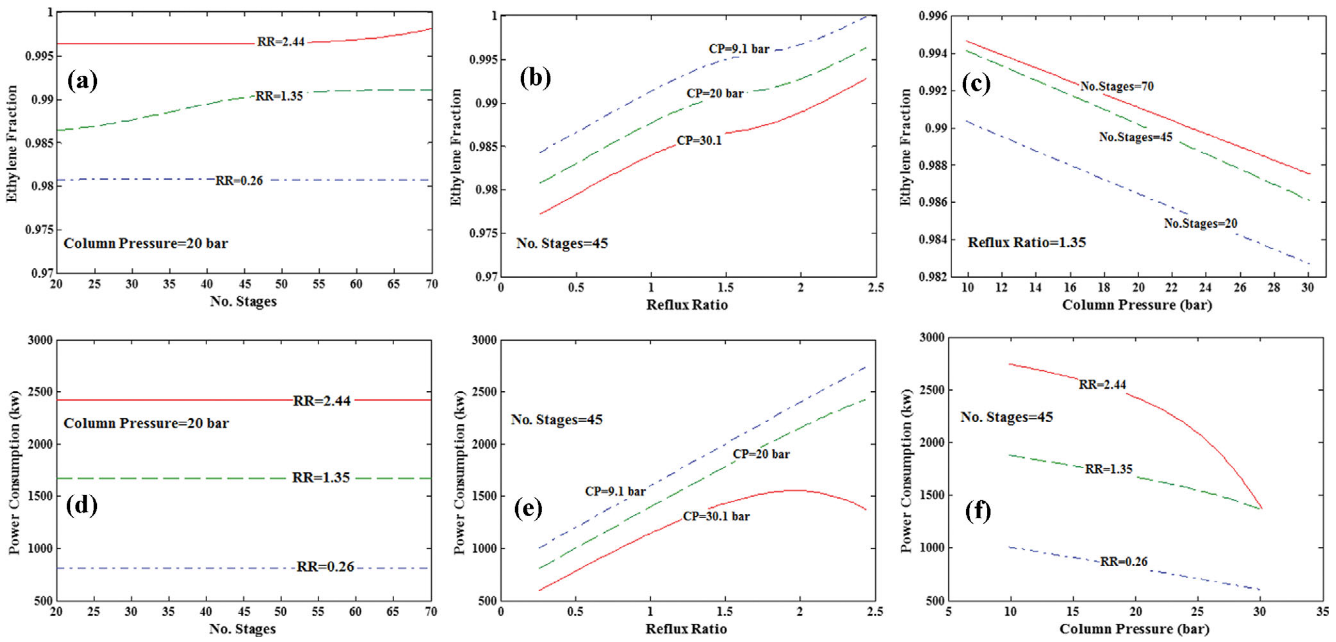

Fig. 7. 2D plot of the predicted ethylene fraction in top stream and power consumption of deethanizer as a function of No. stages, reflux ratio, and column pressure.

In Fig. 5, the values of validation-R^2 for developed models based on networks with different structures are shown. According to the results presented in Fig. 5, for C2 fraction in top stream a feedforwardnet and for energy consumption a cascadeforwardnet with one neuron in hidden layer were selected to develop the optimum model. The value of R2-validation for these two networks is approximately 1.

The 3D plot of C2 fraction and power consumption of C2/C3 distillation column in terms of number of stages, reflux ratio and column pressure is presented in Fig. 6. According to Fig. 6(a), the effect of the number of stages on C2 fraction is negligible. A similar trend is observed for power consumption in Fig. 6(c). Fig. 6(a) and (b) reveal that reflux ration has distinctive effect on C2 fraction and power consumption, respectively. Increase in reflux ratio leads to increase in C2 fraction and power consumption. For C2 fraction, this trend is more severe at low values of reflux ratio. The effect of column pressure on power consumption is mild. But at low values of reflux ratio, an increase in column pressure leads to huge decrease of C2 fraction in the top stream.

2. Deethanizer Column

The top stream of the C2-stripper column was transferred into the next distillation column termed "deethanizer". There are some limitations on the pressure of the deethanizer that the pressure needs to be equal to or less than the pressure of the previous column. The range of operative parameters is presented in Table 2. In this column the range of number of stages is 20 to 70. For each response, networks with one neuron in hidden layer and three different functions were developed. It is figured out that the type of function has no obvious effect on model performance for prediction of ethylene purity. On the other hand, the results show that the cascadeforwardnet function had the best prediction ability in power consumption. So, this function was selected for tuning the model in this column, and subsequent discussions could be done on the basis of this optimum structure of neural network model.

In Fig. 7, 2D plots of the power consumption and ethylene fraction in top stream of deethanizer are shown in terms of the number of stages, reflux ratio, and column pressure. As shown in Fig. 7(d), the number of trays has no distinctive effect on the ethylene purity in the top stream. Only little change is observed for reflux ratio of 1.35. Further, the number of stages absolutely has no effect on power consumption in this column. This is a worthy conclusion and leads to design of column with lower number of stages, which leads to lower fixed capital cost (FCI). It is important to note that these results were observed when the column pressure was fixed at 20 bar. Fig. 7(b) and (e) show that any increase in reflux ratio results in increase in ethylene purity and power consumption. According to the importance of ethylene purity and energy price, optimum condition will be determined. Fig. 7(c) and (f) reveal that the power consumption and ethylene purity decrease as the column pressure increases. So, optimum condition is determined according to the importance of criteria.

3. C3-Stripper Column

The heavy products of the C2 stripper were transferred to the propane/propylene separation unit, termed C3-stripper column. The purity of propylene in light stream and the required energy for condenser and reboiler were selected as the indexes of separation performance. The range of operative parameters is shown in Table 2. A cascadeforwardnet with one neuron in hidden layer was selected and trained.

In Fig. 8 the contour plots of propylene fraction and power consumption are depicted in terms of operative parameters. Fig. 8(a) and (c) show that any increase in reflux ratio leads to increase in propylene purity and power consumption. Fig. 8(b) shows a mild increase in propylene fraction, while the number of trays increases.

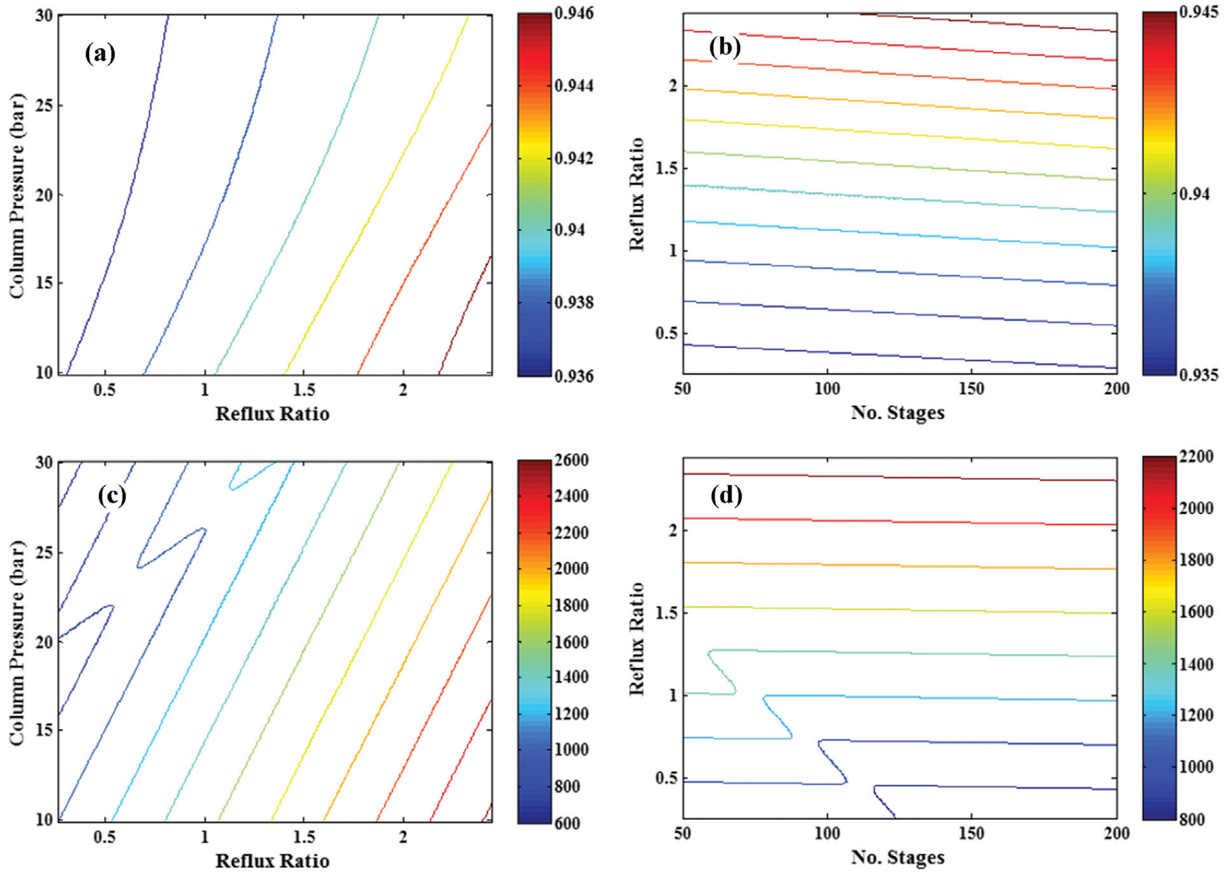

Fig. 8. Contour plot of the predicted propylene fraction in top stream and power consumption of propylene/propane separation column as a function of No. stages, reflux ratio, and pressure.

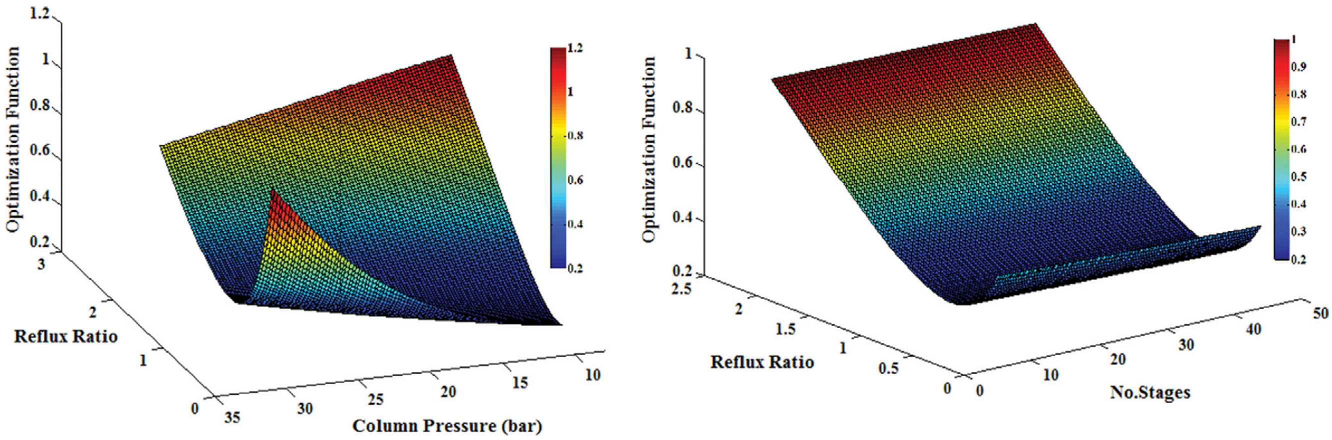

Fig. 9. Sensitivity of optimization function of C2-stripper vs. No. stages, reflux ratio, and column pressure.

But Fig. 8(d) reveals that changes in the number of trays has no effect on energy consumption in reboiler and evaporator.

4. Optimization

In this work, optimum conditions were found by genetic algorithm. In this optimization process, two different goals were set as follows:

- I Maximizing the purity in top stream of column
- II Minimizing the energy consumption in column

Increasing the number of trays leads to increase in fixed capital investment. And increase in energy consumption leads to increase in working capital. In this approach, each response has a degree of importance that is determined with an assigned weight. To perform the optimization through genetic algorithm, an optimization function is defined as follows:

$$\text{Optimization function}=(1-\text{purity})\times w_p+(\text{power consumption})\times w_{pc} \quad (1)$$

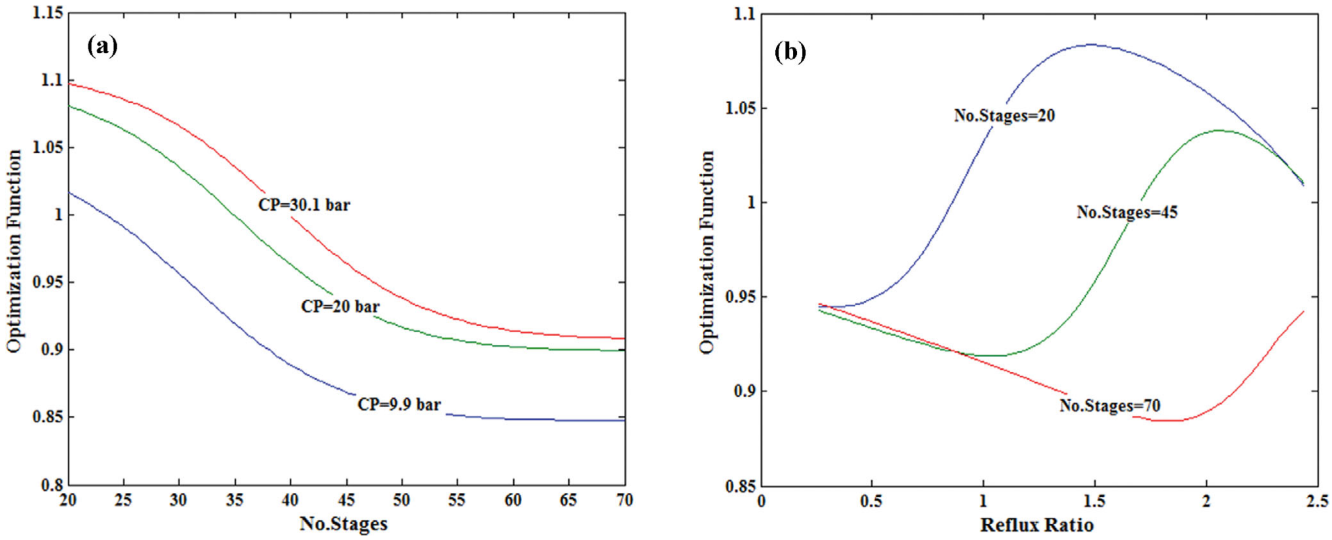

Fig. 10. Optimization function of the deethanizer column as a function of No. stages, reflux ratio, and column pressure.

Table 3. Optimized conditions

Unit	Degree of importance (purity-power consumption)	No. stages	Reflux ratio	Column pressure (bar)	Purity (%)	Power consumption (kw)
C2 Stripper	100-0	48	2.4398	9.911	0.99978	2,747.3546
	0-100	48	0.26044	18.1242	0.86755	997.6963
	50-50	48	0.34871	9.9106	0.95336	1,109.2035
	60-40	48	0.44851	9.9196	0.9659	1,176.5583
	70-30	48	0.57855	9.9911	0.97722	1,266.5227
De-ethanizer	100-0	70	2.44	9.91	1	2,744.1
	0-100	69	0.26012	30.0891	0.97707	600.2251
	50-50	70	2.44	30.0798	0.99385	1,378.3864
	60-40	70	2.1529	9.91	1	2,520.0491
	70-30	70	2.3115	9.91	1	2,644.2554
C3 Stripper	100-0	200	2.44	9.9106	0.94749	2,651.8036
	0-100	200	0.26	29.4528	0.9343	352.4278
	50-50	200	0.35281	30.09	0.9346	394.0747
	60-40	200	2.4322	11.9458	0.94721	2,576.2105
	70-30	196	2.44	9.9107	0.94747	2,650.9372

where w_p and w_{pc} are the degree of importance of purity and power consumption, respectively. It is clear that the aim of the genetic algorithm in this case is to find the condition in which the optimization function gets minimum values. Since the difference in the order of magnitude of purity and power consumption may lead to erroneous evaluation of results, the responses are normalized. As mentioned, two goals are set. Each response is re-evaluated by assigned weights. These determined weights represent the importance of each performance index. Fig. 9 presents the sensitivity of optimization function of C2-stripper column to operative parameters. As shown, the number of stages has no important effect on optimization function. Fig. 10 shows the optimization function of the deethanizer column. Fig. 10(a) shows that by increasing the number of trays while the reflux ratio is constant, the optimization function decreases. On the other hand, by the increasing the column pressure the optimization function increases. In Table 3, the optimum conditions for each column according to the degree of importance of each response are presented.

CONCLUSION

The results of this work show that artificial intelligence (AI) is able to develop robust models for prediction of the separation performance of three columns in the second separation section of a methanol to olefin (MTO) process. Assessing various structures of neural networks revealed that a network with one hidden layer and one neuron in hidden layer could be an optimum choice. The R^2-validation of these models approaches 1, implying excellent agreement between real and predicted values. Optimization process based on selected developed model for various columns by different strategies showed that the number of trays should be set at maximum value, while the optimum values of reflux ratio and column pressure change according to the optimization strategy.

REFERENCES

1. N. Bahri-Laleh, M. Nekoomanesh-Haghighi, S. Sadjadi and A. Pajouhan, *Polyolefins J.*, **3**, 11 (2016).
2. A. H. Saeedi Dehaghani, V. Pirouzfar and A. Alihosseini, *Polym. Bull.*, **77**, 6467 (2020).
3. M. H. B. Zakria, M. G. Mohd Nawawi and M. R. Abdul Rahman, *Polyolefins J.*, **9**, 15 (2022).
4. M. Rostamizadeh, F. Yaripour and H. Hazrati, *Polyolefins J.*, **5**, 59 (2018).
5. M. Yang, D. Fan, Y. Wei, P. Tian and Z. Liu, *Adv. Mater.*, **31**, 1902181 (2019).
6. Y.-Q. Zhuang, X. Gao, Y.-p. Zhu and Z.-h. Luo, *Powder Technol.*, **221**, 419 (2012).
7. W. Zhang, J. Chen, S. Xu, Y. Chu, Y. Wei, Y. Zhi, J. Huang, A. Zheng, X. Wu, X. Meng, F. Xiao, F. Deng and Z. Liu, *ACS Catal.*, **8**, 10950 (2018).
8. S. Shirbakht, S.A. Mirmohammadi, K. Didehban, S. Sadjadi and N. Bahri-Laleh, *Adv. Polym. Technol.*, **37**, 2588 (2018).
9. A. Hedayati Moghaddam, *Chemical Papers*, **76**, 4787 (2022).
10. V. Zeynali, J. Sargolzaei and A. Hedayati Moghaddam, *Desalination and Water Treatment*, **57**, 24240 (2016).
11. A. Rahbar, M. Nekoomanesh-Haghighi, N. Bahri-Laleh and H. Abedini, *Catal. Lett.*, **145**, 1186 (2015).
12. S. M. A. Masoudi, A. Hedayati Moghaddam, J. Sargolzaei, A. Darroudi and V. Zeynali, *Environ. Prog. Sust. Energy*, **37**, 1638 (2018).
13. A. Hedayati Moghaddam, H. Hazrati, J. Sargolzaei and J. Shayegan, *Appl. Water Sci.*, **7**, 2753 (2017).
14. A. Taheri Najafabadi, S. Fatemi, M. Sohrabi and M. Salmasi, *J. Ind. Eng. Chem.*, **18**, 29 (2012).
15. A. H. Moghaddam, J. Shayegan and J. Sargolzaei, *J. Taiwan Inst. Chem. Eng.*, **62**, 150 (2016).
16. H. R. Tashaouie, G. B. Gholikandi and H. Hazrati, *Desalination and Water Treatment*, **39**, 192 (2012).
17. D. M. Himmelblau, *Ind. Eng. Chem. Res.*, **47**, 5782 (2008).
18. A. H. Moghaddam, J. Sargolzaei, M. H. Asl and F. Derakhshanfard, *Polym.-Plast. Technol. Eng.*, **51**, 480 (2012).
19. B. Y. Yu and I. L. Chien, *Chem. Eng. Technol.*, **39**, 2293 (2016).