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Modeling and Prediction of Hydrogen Production During Ethanol Steam Reforming Using Artificial Neural Network

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ABSTRACT

The present study utilizes multilayer perceptron artificial neural network (ANN) to predict hydrogen yield (Y_{H_{2.Y}}), carbon monoxide yield (Y_{{CO.}_{Y}}), and ethanol conversion (Y_{{ETOH}_{Con.}}) during ethanol steam reforming. Reaction temperature, water-to-ethanol molar ratio (WEMR), and liquid hourly space velocity (LHSV) were used as input variables. Twenty experimental data, obtained by conducting ethanol steam reforming reaction in the lab., were used to build the ANN model. The experimental data were divided into 16 training data (80%) and 4 testing data (20%). Three network topologies [3 3 3], [3 4 3], and [3 5 3] were examined in this work. Based on obtained results, the [3 4 3] architecture was selected as the preferred configuration. Over all evaluated algorithms (Trainlm, Trainscg, and Traingdx), the trainlm algorithm and log-sigmoid transfer function were used in the selected model. The model performance was evaluated using mean squared error (MSE) and an efficiency coefficient. The testing stage results of the model reported that the efficiency coefficients were 96.45% for hydrogen yield, 93.70% for carbon monoxide yield, and 96.75% for ethanol conversion; whereas the corresponding MSE values were 0.00293, 0.00437, and 0.00278 respectively. The predictions of ANN model were also compared with a previously developed design of experiments (DOE) model using the same experimental data. A comparison of the ANN model with the statistical (DOE) model showed that the ANN model provides a slightly better fit to the experimental data.

النمذجة والتنبؤ بإنتاج الهيدروجين أثناء عملية إعادة التشكيل البخاري للإيثانول باستخدام الشبكات العصبية الاصطناعية

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الكلمات المفتاحية:
إنتاج الهيدروجين.
الشبكات العصبية الاصطناعية.
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التنبؤ بتفاعلات الإصلاح البخاري.

الملخص

تستخدم هذه الدراسة الشبكات العصبية الاصطناعية (ANN) كأداة لمحاكاة والتنبؤ بإنتاجية الهيدروجين (YH2)، وإنتاجية أول أكسيد الكربون (YCO)، وتحويل الإيثانول (YETOH) الناتجة عن عملية الإصلاح البخاري للإيثانول. وقد استخدمت درجة حرارة التفاعل، والنسبة المولية للماء الي الإيثانول، السرعة الفراغية للسائل بوصفها متغيرات ادخال. استخدمت في هذا العمل 20 نقطة من البيانات المعملية، تم الحصول عليها من إجراء تفاعل إعادة التشكيل البخاري للإيثانول في المختبر، لبناء نموذج الشبكة العصبية الاصطناعية. قسمت البيانات المعملية الي 16 نقطة بيانية للتدريب وتشكل 80% من النقاط الكلية و4 نقاط بيانية للاختبار وتشكل 20%. في هذه العمل تمت دراسة 3 تراكيب للشبكة، وهي [3 3 3] و [3 4 3] و[3 5 3]. واستنادا إلى النتائج المتحصل عليها، تم اختيار البنية [3 4 3] بوصفها التكوين المفضل.

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ومن بين خوارزميات التدريب التي جرى تقييمها، وهي Trainlmو Trainscgو Traingdx، استخدمت Trainlm مع دالة النقل Log-sigmoid في النموذج المختار. تم تقييم أداء النموذج باستخدام متوسط مربع الخطأ (MSE) ومعامل الكفاءة. وأظهرت نتائج مرحلة الاختبار أن معاملات الكفاءة بلغت 96.45% بالنسبة لي انتاج الهيدروجين، و93.70% بالنسبة لي انتاج اول أكسيد الكربون، و96.75% بالنسبة لي تحول الايثانول. في المقابل بلغت قيم متوسط مربع الخطأ 0.00293 و0.00437 و0.00278 على الترتيب. أيضا تمت مقارنة تنبؤات نموذج الشبكة العصبية الاصطناعية مع تنبؤات نموذج سابق تم تطويره باستخدام منهجية تصميم التجارب DOE وذلك بالاعتماد على نفس البيانات المعملية. الدراسة أظهرت افضلية بسيطة لنموذج الشبكة العصبية على نموذج DOE في عملية التنبؤ بالنتائج.

1. Introduction

Steam reforming method using different raw materials substrate is widely used to produce hydrogen [1-7]. Ethanol is a potential substrate material for hydrogen production. The performance of ethanol steam reforming process depends on catalytic system used and operating conditions. Developing reliable relationships between operating variables and process responses is therefore important for modeling, predicting, and optimizing hydrogen production. Earlier work by Bshish et al. reviewed catalytic systems used in hydrogen production through ethanol steam reforming [1]. They also performed experimental work to examine hydrogen yield over different nickel-based catalysts and different reaction conditions [2, 4].

Developing an active, selective, and stable catalytic system for producing hydrogen is a major challenge as far as ethanol steam reforming is concerned. Our group [1] performed an extensively reviewed various catalytic systems for hydrogen production through ethanol reforming.

The nonlinear relationships between reaction conditions and process responses can make empirical prediction challenging. Artificial neural networks (ANNs), particularly multilayer perceptron (MLP), can present nonlinear mappings between multiple inputs and outputs [8-9].

The use of neural networks in the area of catalysis was reviewed by several researchers. Omata et al. [10] developed Co-supported catalyst for the oxidation of CO using the ANN technique. The loading of Co and the preparation conditions of the Co-supported catalyst were optimized by using ANN. Three years later, Umegaki et al. [11] developed an alternative high-performance Cu-based catalyst for oxidative steam reforming of methanol using ANN. The trained ANN model predicts the performance of many catalysts. Based on the estimated data, they found the best conditions for catalyst composition and preparation conditions. The optimum Cu–Pr– Ti catalyst was Cu/Pr/Ti = 58/16/26, calcination temperature = 623 K, and total metal salt concentration in preparation step = 1 M. The basic principles of ANN were explained by Holeňa and Baerns [12]. They applied multilayer perceptrons to data on catalyst composition and catalytic results in the oxidative dehydrogenation of propane to propene. By using such networks, they established relationships between the composition and the performance of a catalyst. A method based on ANN was developed to simulate the relations between catalyst components and catalytic performance, as proposed in previous research [13]. A multi-trained design strategy was applied to the catalyst design. They concluded that the new method was highly efficient and universal.

Adib et al. [14] demonstrated the applicability of ANN models to predict the molar percentage of CH4, CO2, and CO in the Fischer–Tropsch process of natural gas in the presence of a Co/Al2O3 catalyst. Three input parameters were used, including temperature, pressure, and reaction time. Correlation coefficients for the CH4, CO2, and CO were 0.94, 0.93, and 0.96, respectively. The authors also argued that operation time has a significant effect on the molar percentage of CH4 as a desired product with respect to other operational parameters. They concluded that an ANN is a reliable and highly accurate estimation method that can be used to predict the output composition of the Fischer–Tropsch synthesis. Other successful implementations of the ANN technique have been reported in the field of catalysis [15-19].

The present study applies an ANN to develop an integrated model capable to predict hydrogen yield, ethanol conversion, and carbon monoxide yield based on reaction temperature, water to ethanol molar ratio, and LHSV. The ANN results compared with a previously developed DOE model based on the same experimental study [].

The aim of the present study is to develop an integrated system capable to predict hydrogen yield, ethanol conversion, and carbon monoxide yield using ANN models. Also, ANN models were compared with our previous published statistical models with the same experimental data used in the previous statistical approach

2. Methodology

2.1. Experimental Dataset and Variables

The ANN model was developed using 20 experimental data obtained from ethanol steam reforming reaction conducted at laboratory scale [2, 4]. In this model, the three inputs were reaction temperature, water-ethanol molar ratio, and liquid hourly space velocity. The experimental ranges of inputs were 200-500ºC for temperature, 0-1 h for LHSV, and 1-24 for water-ethanol molar ratio. The three examined outputs were hydrogen yield, carbon monoxide yield, and ethanol conversion. Table 1 shows the input and outputs variables used in training and testing the ANN model.

Table 1: Input and Output Variables used in Training and Testing ANN Model

Role	Variable	Symbol	Evaluation Range
Input	Reaction Temperature	T	200-500 ºC
Input	Water-Ethanol Molar Ratio	WEMR	1-24
Input	Liquid Hourly Space Velocity	LHSV	0-1 h
Output	Hydrogen Yield	Y_{H2}	As reported
Output	Carbon Monoxide Yield	Y_{CO}	As reported
Output	Ethanol Conversion	X_{EtOH}	As reported

2.2. ANN Architecture and Training

In this work, MATLAB mathematical software version 7.11.0 (R 2010b) was used to develop the seeked ANN model. The multi-layer perception (MLP) neural network based on back propagation (BP) algorithm is used as a predictor. The used type of MLP artificial neural network consists of three layers or groups, input layer, hidden layer, and the output layer as shown in Fig. 1. Each layer of these three layers has a number of neurons. The number of neurons in the input and output layers are equal to the number of input and output variables. Hidden layer can be one or more than one layer. In this work, the three candidate architectures were evaluated: [3 3 3], [3 4 3], and [3 5 3]. The middle number in these architectures denotes the number of neurons in the single hidden layer. The neurons activity in the hidden layer is determined by the activities of the input units and the weight on the connections between the input and the hidden units. The output performance depends on the activity of the hidden layer and the weights between the hidden and output layers [14].

First, the input data of each neuron (a1, a2, a3) are multiplied by the corresponding weight functions that assigned to them (wj1, wj2, wj3). Then the combined input cj, forms as a summation of products of neurons and weight functions as shown in equation (1).

$$cj = a_1 w_{j1} + a_2 w_{j2} + a_3 w_{j3} \quad (1)$$

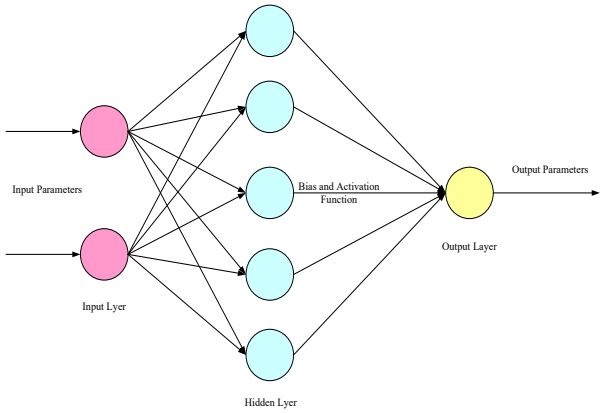


Fig. 1: Basic structure of MLP neural network[6]

Each neuron in one layer has direct connections to the neurons of the subsequent layer by means of activation function (commonly a sigmoid function). Since we have more than one input and output in this study, the feed forward neural network process was set (MIMO) network structure.

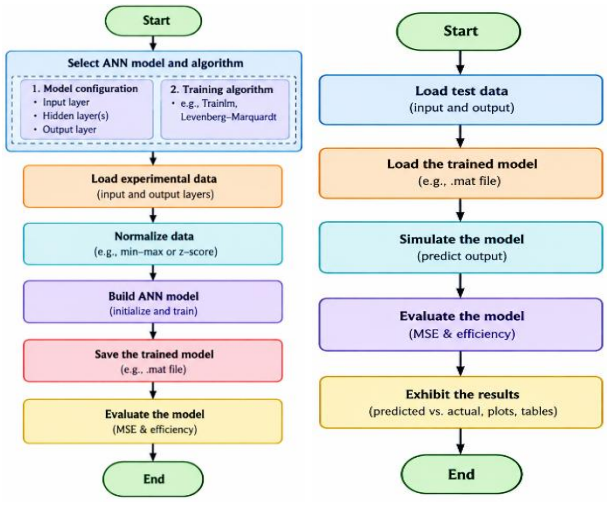
To construct neural networks, the log-sigmoid transfer function (logsig), the most common type of transfer function [21], was used. For all developed networks, the logsig function was used between the hidden layers and the output layers. The non-linear nature of this function enhances the performance of a neural network. The networks were trained using three different (algorithms 1) Levenberg Marquardt Learning Algorithm (Trainlm), 2) Gradient descent w/momentum & adaptive back propagation (Traingdx), 3) Scaled conjugate gradient back propagation (Trainscg). Trial and error search method was applied to select the optimum topology of the model. The information was propagated forward to the output layer where the output variable was calculated and compared to the actual value in order to generate a prediction error.

2.3. Data Partitioning and Normalization

In order to build ANN model, 20-observation dataset, collected from experimental work, were partitioned into two subsets [20]. The first set: 16 observations (80% from all dataset used) was used for training the model. The second set is consists of 4 observations (20%) was used to validate the model. Figure 2 a and b summarize the training and testing procedure, respectively.

Process inputs for neural network models were inconsistent based on their magnitudes. Thus, these input data were normalized within a consistent range [0 to 1] before introducing them to the input layer of the network to ensure that each data are given fair contribution in determining the network output. The data normalization method employed in this study is expressed in equation (2).

$$Nor_X = \frac{X_i}{max(X_i)} \qquad (2)$$



(a) (b)

Fig. 2: (a) Training Procedure (b) Testing Procedure for the Propsed ANN Model

Where, Nor_X is the normalized data of the input and the output, X_i is the actual input /output before scaling (normalization), $max(X_i)$ is the maximum data, respectively.

2.4. Training and Testing workflow

During training, the model configuration and training algorithm are selected, the training data are loaded and normalized, the ANN is trained, and the trained model is saved. Whereas in the testing stage, the saved model is loaded and applied to the 4 held out observations; predicted values are then compared with the corresponding experimental results.

2.5. ANN Model Evaluation

The criteria used to evaluate ANN model performance included mean square error (MSE), and the efficiency (*Eff.*). Equations (3) and (4) were used to calculate (MSE) and (*Eff.)*, respectively.

$$MSE = \frac{1}{n}\sum_{i=1}^{n}\left(y_{Exp} - y_{ANN}\right)^2 \qquad (3)$$

$$Eff = \frac{1 - MSE}{Var.} \qquad (4)$$

$$Var = SD\left(y_{Exp}\right)^2 \qquad (5)$$

where, y_{Exp} and y_{ANN} are vectors of an experimental and prediction value, respectively. *Var* and *SD* are the variance and standard deviation, respectively.

After appropriate input parameters for hydrogen prediction has been selected, the optimum number of neurons was searched in order to improve the predictive performance. Therefore, training algorithm and neuron number in the hidden layers were changed, while the transfer function, and number of neurons in the input and output layers were fixed.

Neurons numbers in the hidden layer were varied from [3 to 5] for the three training algorithms (TrianLM, TrainScg, and TrainGdx) tested. The optimal number of neurons in the hidden layer was selected based on the minimum mean square error (MSE) and maximum efficiency in the testing stage.

3.Result and discussion

3.1. Input & Output Data

In order to obtain good results as well as to fasten significantly the calculations, input and output data normalization is crucial. Figures 3 (a) and (b) show the input and output data respectively before normalization. Fig. 3 (a) explain the three input parameters. Input 1 refers to the temperature which varies from 200-500 ºC. Input 2 shows the molar ratio which vary from 1-24.while input 3 refer to the liquid hourly space velocity which is vary from 0-1h^{-1}. Outputs 1, 2, and 3 in Fig. 3 (b) refer to H_2 yield, CO yield, and ethanol conversion into gaseous product, respectively. It is clearly seen that the magnitudes of the input and output variables are inconsistent. Thus, these data were normalized within a consistent range (0 to 1) before introducing them to the input layer of the network to ensure that each data is given fair contribution in determining the network output and to reducing any errors. Data after normalization for the input and output variables are shown in Fig. 3 (c) and (d), respectively. The normalization process was made according to equation (2).

3.2. Training Stage and ANN Topology Selection

The training process for the neural network is stopped only when the mean squared error falls below 0.0001 and the neural network have architecture of [3 3 3], [3 4 3], and [3 5 3] with the three deferent algorithms (trainlm, traingdx, and trainscg) are examined at all the trials. Tables 1-6 show the performance of ANN design to model the three outputs. The criteria used to obtain the best ANN design based on efficiency and MSE of each model in the training and testing stage. As it seen, models were trained using different training algorithm with different number of nodes in the hidden layer. It was found that the [3 4 3] is the best selected architecture based on the model developed assessment reported in these tables, The trainlm, training algorithm was used for the selected network, providing a minimum average MSE and highest average efficiency forthe three outputs, over the three tested algorithms.

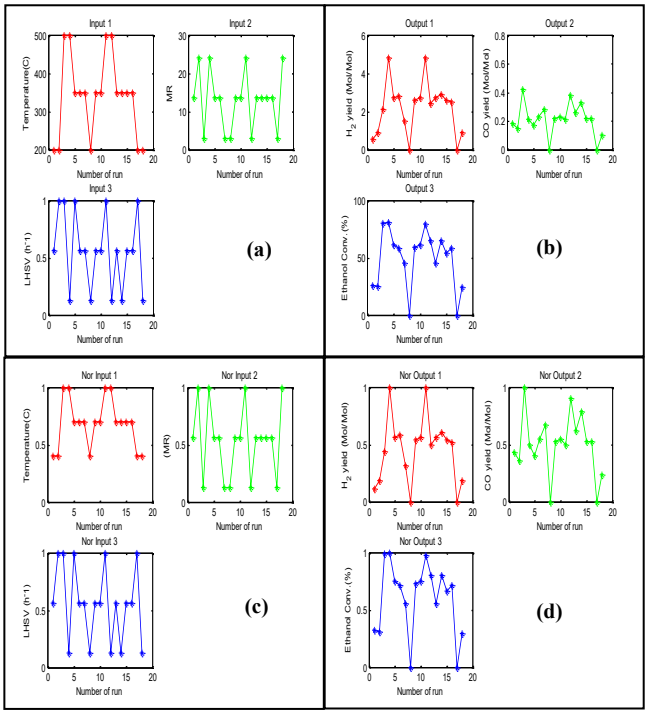


Fig. 3: a) Input data before normalization, b) Output data before normalization, c) Input data after normalization, d) Output data after normalization

Table 1: ANN Design using *Trianlm* algorithm in training stage

Neurons per layer	Training stage					
	Mean square error (MSE)			Efficiency		
	H_2	CO	ETOH	H_2	CO	ETOH
Trainlm [3 3 3]	0.00380	0.00046	0.00038	0.9953	0.9942	0.9953
Trainlm [3 4 3]	0.00047	0.00045	0.00470	0.9947	0.9944	0.9947
Trainlm [3 5 3]	0.00017	0.00075	0.00017	0.9979	0.9907	0.9979

Table 2:ANN Design using *Trianscg* algorithm in training stage

Neurons per layer	Training stage					
	Mean square error (MSE)			Efficiency		
	H_2	CO	ETOH	H_2	CO	ETOH
Trainscg [3 3 3]	0.00047	0.00063	0.00047	0.99417	0.9922	0.9941
Trainscg [3 4 3]	0.00052	0.00071	0.00052	0.99350	0.9911	0.9935
Trainscg [3 5 3]	0.00053	0.00058	0.00053	0.99340	0.9928	0.9934

Table 3: ANN Design using *Triangdx* algorithm in training stage

Neurons per layer	Training stage					
	Mean square error (MSE)			Efficiency		
	H_2	CO	ETOH	H_2	CO	ETOH
Traingdx [3 3 3]	0.0284	0.0354	0.02840	0.65166	0.5656	0.65166
Traingdx [3 4 3]	0.0251	0.0323	0.02518	0.69110	0.6035	0.69110
Traingdx [3 5 3]	0.0081	0.0082	0.00812	0.90020	0.8993	0.90029

Table 4: ANN Design using *Trianlm* algorithm in testing stage

Neurons per layer	Testing stage					
	Mean square error (MSE)			Efficiency		
	H_2	CO	ETOH	H_2	CO	ETOH
Trainlm [3 3 3]	0.01220	0.00920	0.0077	0.8518	0.8664	0.9091
Trainlm [3 4 3]	0.00293	0.00437	0.00278	0.9645	0.9370	0.9675
Trainlm [3 5 3]	0.01495	0.00408	0.00976	0.8189	0.9411	0.8859

Table 5:ANN Design using *Trianscg* algorithm in testing stage

Neurons per layer	Testing stage					
	Mean square error (MSE)			Efficiency		
	H_2	CO	ETOH	H_2	CO	ETOH
Trainscg [3 3 3]	0.01439	0.00907	0.00925	0.8257	0.8693	0.8918
Trainscg [3 4 3]	0.01340	0.00586	0.00790	0.8372	0.9155	0.9065
Trainscg [3 5 3]	0.01410	0.00327	0.00790	0.8292	0.9528	0.9076

Table 6:ANN Design using *Triangdx* algorithm in testing stage

Neurons per layer	Testing stage					
	Mean square error (MSE)			Efficiency		
	H_2	CO	ETOH	H_2	CO	ETOH
Traingdx [3 4 3]	0.0319	0.02919	0.03218	0.61307	0.5797	0.6239
Traingdx [3 3 3]	0.0306	0.0457	0.01995	0.6286	0.34107	0.7668
Traingdx [3 5 3]	0.0076	0.0173	0.00913	0.9070	0.7498	0.8932

Figuers 4 and 5 show experimental data distribution before and after training stage, resspectively. In these figuers, the blue colour specifies the target plot and the red colour specify the neural network. Figure 6 (a) shows that the training process of the network took 47 epoch to get the goal of the proposed error (10^{-3}). Furthermore, the linear regression (R = 0.99414) between the outputs and the corresponding targets, during the training stage, is clearly seen in this figure 6 (b).

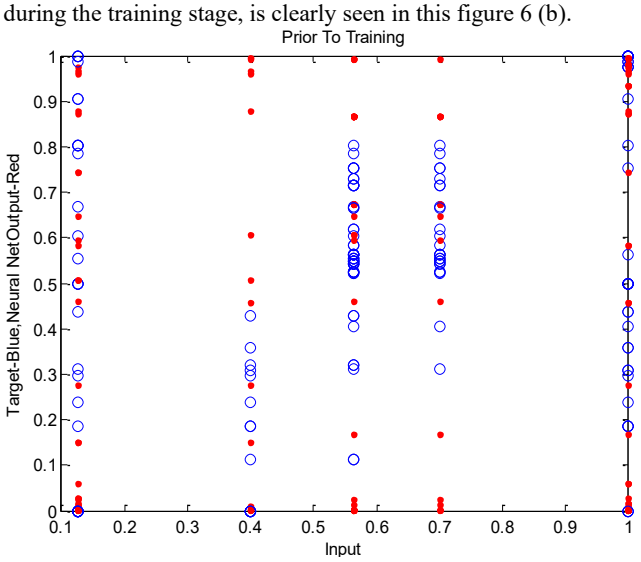


Fig. 4: Dataset distributions before training the ANN model

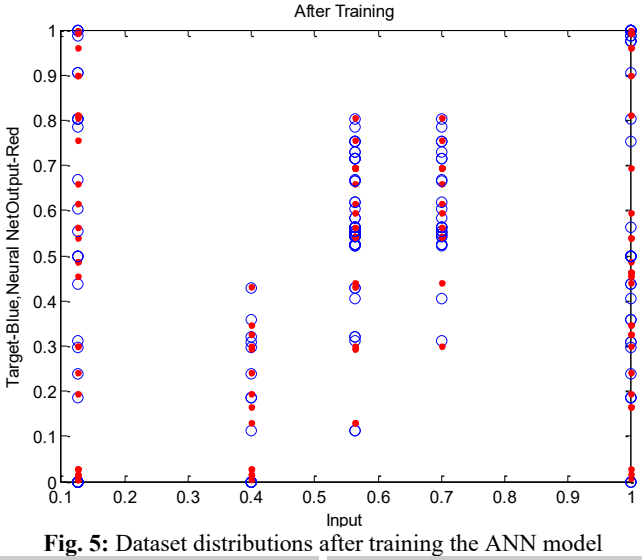
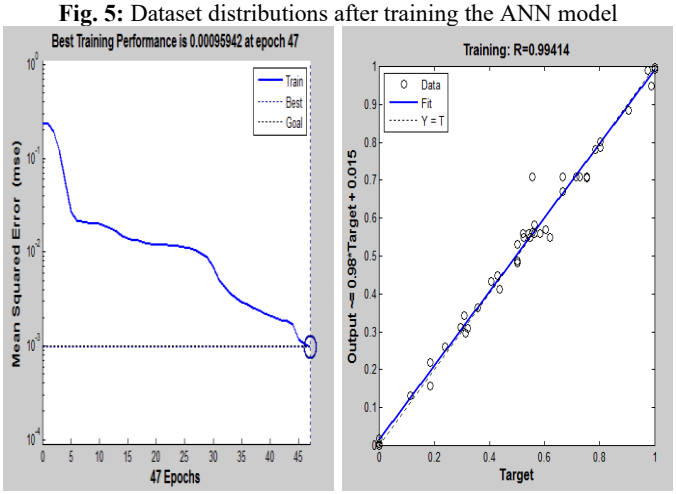


Fig. 5: Dataset distributions after training the ANN model



(a) (b)

Fig.6: (a) Iterations curve and **(b)** linear regression analysis between the network outputs and the corresponding targets during training stage

3.3. Testing Stage Performance

Table 7 reports the results of testing stage retained from table 4. The efficiency coefficients were 0.9645, 0.937, and 0.9675 for hydrogen yield, carbon monoxide yield, and ethanol conversion, respectively. The corresponding MSE values were 0.00293, 0.00437, and 0.00287, respectively.

Table 7 MSE and Efficiency values Obtained in Testing Stage

Outputs	MSE	Efficiency
H_2	0.00293	0.9645
CO	0.00437	0.9370
ETOH Conv.	0.00278	0.9675

3.4. Comparison of Experimental and ANN-Predicted Results

Fig. 7 shows the experimental value versus predicted values from neural network model developed. It is clearly shown that developed model can represent the process to predict the outputs (H_2, CO, and ethanol conversion) quite satisfactorily, with model efficiency higher than 0.90 and MSE lower than 0.0045 as shown in Table 7. MSE obtained in this study was lower comparing with similar study conducted to design an optimum Ni/Al_2O_3 catalyst for the production of hydrogen [22]. that is a sign for a good performance of the ANN in this study.

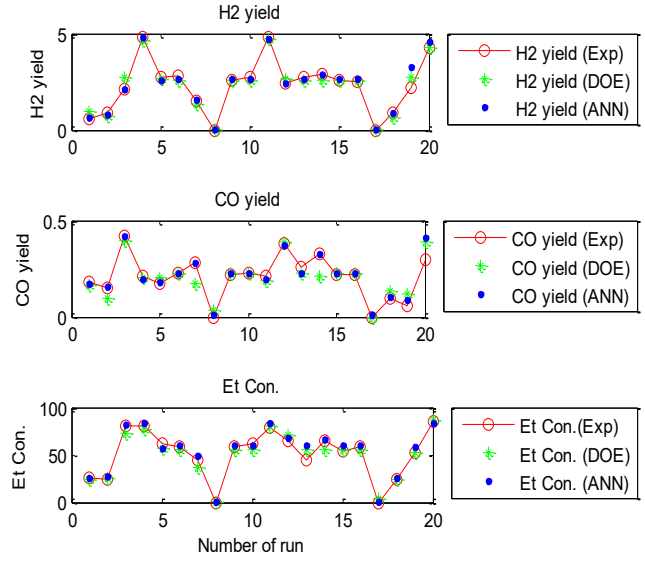


Fig. 7: ANN predicted and observed data for the three outputs

Experimental versus ANN and statistical (DOE) Models

Fig. 8 shows the experimental value versus predicted value from statistical model and ANN developed model. It is clearly shown that both developed models can represent the process quite satisfactorily because both model approaches, efficiency coefficients were high. However, the neural network efficiency for neural network was slightly higher than statistical model. It can be conclude that, statistical modelling and ANN modelling can be used to represent ethanol steam reforming reaction. However, ANN modelling is slightly more appropriate in predicting the output than empirical modelling. This result was in a good agreement with other published data to investigate carbon dioxide reforming of methane to syngas using response surface methodology and artificial neural network [23].

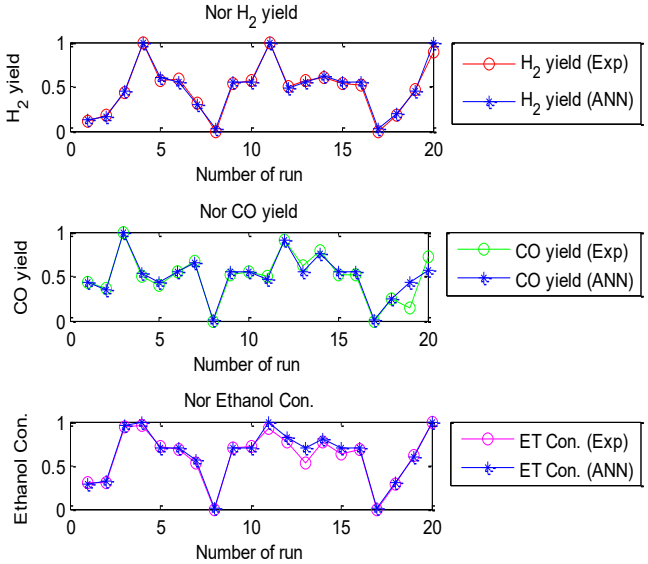


Fig. 8: Experimental values of the outputs versus both ANN and statistical (DOE) model predicted values

4.Conclusion

A multilayer perceptron ANN was developed to predict the performance of ethanol steam reforming reaction. The inputs of the reaction that used in building the ANN model were reaction temperature, water to ethanol molar ratio, and LHSV. The corresponding outputs were H_2 yield, CO yield, and ethanol conversion. Three ANN architectures [3 3 3], [3 4 3], and [3 5 3] were considered in this study. Based on obtained results, the [3 4 3] structure was selected as the preferred ANN model configuration. Among tested algorithms, the trainlm algorithm was found the best along with the logsig transfer function. In the testing stage, the ANN model exhibited a good prediction to the outputs variables with high efficiency values recorded. The efficiency values were 96.45%, 93.7%, and 96.75% for H_2 yield, CO yield, and ethanol conversion respectively. The corresponding MSE values of the ANN model were 0.00293, 0.00437, and 0.00278. These results indicate that the model highly capable to reproduce the testing output observations within the studied domain. The ANN was also compared with the prior DOE model showing that the ANN model provides a slightly better fit to the experimental data. Further experiments and independent validation are recommended before applying the model outside the investigated operating range.

5.References

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