Artificial Neural Network Modeling of a Reactive Distillation Process for Biodiesel Production

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Abstract

The development of an artificial neural network model used to predict the mole fraction of biodiesel obtained from the reactive distillation process involving a reaction between palmitic acid and methanol has been carried out in this work. The data used for the neural network model development were generated with the aid of the parametric utility of Aspen HYSYS prototype of the process developed using Distillation Column Sub-Flowsheet and Wilson model as the fluid package. Six parameters (palmitic acid feed temperature, palmitic acid feed pressure, methanol feed temperature, methanol feed pressure, reboiler heat duty and reflux ratio) were used as the input variables to predict the biodiesel mole fraction. In the training of the neural network model, multiplicative lagged Fibonacci generator with the obtained optimized seed number of 47 was used. The results obtained from the training of the artificial neural network model developed revealed that the model was able to represent the process very well because the mean of squared error estimated from the simulation of the model was found to be approximately zero (that is, 3.93E-09). Furthermore, very good performance was recorded when the developed neural network model was used to predict the mole fraction of biodiesel produced from the bottom section of the reactive distillation column considered owing to the fact the mean of squared error was found, in this case, to be 6.92E-10. It has, thus, been discovered that the developed neural network model was able to perform very well in simulating and predicting the mole fraction of biodiesel produced from the reactive distillation process. Therefore, artificial neural network model has been shown to be a very good tool in representing a complex process like biodiesel reactive distillation.

Keywords: Biodiesel, Reactive Distillation, Aspen HYSYS, Parametric Utility, Artificial Neural Network.

Introduction

Biodiesel, being an alternative fuel, is currently receiving much attention owing to the limited availability of conventional petroleum diesel and, also, due to environmental concerns. It can be directly used to replace petroleum diesel without modifying diesel engines since their properties, for example, specific gravity, cetane number, viscosity, cloud point, and flash point, are similar (Simasatitkul *et al.*, 2011; Giwa *et al.*, 2014). It is a promising alternative or extender to conventional petroleum based diesel fuel. It has a number of advantages. For instance, it can be derived from a renewable domestic resource (vegetable oil), compared to diesel fuel on lifecycle basis, it reduces carbon dioxide emissions by about 78%, and it is nontoxic and biodegradable. All these benefits have made biodiesel a very good environmentally benign fuel (Wang *et al.*, 2004; Jaya and Ethirajulu, 2011; Giwa *et al.*, 2014).

Biodiesel is produced mostly from vegetable oils, which are edible oils, such as palm oil, sunflower, and soybean via transesetrification process using sodium hydroxide as a catalyst. However, the commercialization of biodiesel production from those vegetable oils still have drawback due to high cost of vegetable oil and the purification of the formed biodiesel product. Therefore, it is necessary to develop a process to produce biodiesel more efficiently and economically. One approach for this has been carried out by using Jatropha oil instead of edible oils as a feedstock. Actually, jatropha oil contains 20% saturated fatty acids and 80% unsaturated ones. In the unsaturated fatty acid, oleic acid is the most abundant (44.8%) followed by linoleic acid (34%), palmitic acid

(12.8%) and stearic acid (7.3%) (Shah *et al.*, 2004; Kusmiyati and Sugiharto, 2010; Giwa *et al.*, 2014). According to Kusmiyati and Sugiharto (2010), one kind of fatty acid derived from Jatropha oil, oleic acid, could be used as a raw material to produce biodiesel using esterification reaction involving alcohol such as methanol, ethanol, etc. Furthermore, Giwa *et al.* (2014) have confirmed that, in their work, and also discovered that biodiesel with high purity could be produced using palmitic acid via *esterification reaction.*

Normally, the esterification of fatty acid and alcohol into fatty acid ester is usually conducted in a batch reactor (Omota *et al.*, 2003). However, the production of biodiesel from esterification reaction in the conventional batch reactor has many problems because of its low conversion, heavy capital investments and high energy costs. So, this process using batch reactor has been found not to be economical (Gao *et al.*, 2007). To resolve these problems, an advanced technology for biodiesel production has been developed. This method is known as "reactive distillation" (Kusmiyati and Sugiharto, 2010; Giwa *et al.*, 2014)

Reactive distillation is a process that combines both separation and chemical reaction in a single unit. It is sometimes an excellent alternative to conventional flowsheets with separate reaction and separation sections (Al-Arfaj and Luyben, 2002a; Giwa and Karacan, 2012b; Giwa, 2013a; Giwa and Karacan, 2012c; Giwa and Karacan, 2012d; Giwa and Karacan, 2012f; Giwa, 2012; Giwa and Giwa, 2013a; Giwa, 2013a; Giwa *et al.*, 2013; Giwa and Giwa, 2013b; Giwa, 2014). It has been used in a small number of industrial applications for many years, but the last decade has shown an increase in both its research and applications (Al-Arfaj and Luyben, 2002b). In reactive distillation, the temperature levels for both reaction and vapor-liquid equilibrium must overlap (Al-Arfaj and Luyben, 2002a). By carrying out chemical reaction and separation in one process, the operating and investment costs can be minimized. Some additional benefits offered by reactive distillation technology include: (i) increased yield, because of overcoming chemical and thermodynamic equilibrium limitations, (ii) improved selectivity via suppression of side reactions, (iii) reduced energy consumption, due to effective utilization of reaction heat, in the case of exothermic reactions, (iv) avoidance of hot spots by simultaneous liquid evaporation, and (v) ability to separate close boiling components (Prakash *et al.*, 2011). Due to these advantages and with growing process understanding, the chemical process industry has developed an increasing number of processes based on reactive distillation (Bock *et al.*, 1997). However, reactive distillation is not extensively used in industry because it is perceived that its operation will always be more difficult and will pose higher requirements on the quality of the design than the conventional flow sheet in which a reactor is typically followed by a train of distillation columns. This behavior is mainly attributed to the complex interactions between the underlying physical phenomena taking place in reactive distillation processes, having a significant influence on the robust operation of the system (Giwa and Karacan, 2012b; Giwa, 2013b). These complexities have made the modelling of reactive distillation processes extremely difficult (Giwa and Karacan, 2012b; Giwa and Giwa, 2012). As such, a robust tool that can handle complex functions very well is needed to represent this complex process. One of these tools has been discovered to be "neural network", otherwise known as "artificial neural network" model because, according to Beale *et al.* (2010), artificial neural network can be trained to handle complex functions (Giwa and Giwa, 2013c).

Artificial neural network model can be viewed as a nonlinear empirical model that is especially useful in representing input-output data, in making predictions in time, and in classifying data (Himmelblau, 2000). It can be highly nonlinear, can learn easily, requires little or no a priori knowledge of model structure, is fault-tolerant and can handle complex problems that cannot be satisfactorily handled by the traditional methods (MacMurray and Himmelblau, 2000; Giwa and Karacan, 2012a).

Based on the information obtained from the literature, different types of neural network models have been employed to represent different Chemical Engineering processes. For instance, Giwa and Karacan (2012a) used three different types of delayed neural network (Nonlinear AutoRegressive (NAR), Nonlinear AutoRegressive with eXogenous inputs (NARX) and Nonlinear Input-Output (IO)) models to represent a reactive distillation column in predicting the temperatures of the top and the bottom sections of the reactive distillation column used for the production of ethyl acetate and they were able to obtain very good results from both NAR and NARX models while the results given by IO models were found not to be satisfactory. Also, Giwa and Karacan (2012e) developed two nonlinear blackbox (treepartition and sigmoid network NARX) models for the reactive distillation process used for the production of ethyl acetate from the esterification reaction between acetic acid and ethanol and found that sigmoid network NARX model was better than treepartition NARX model for the reactive distillation process studied in their work. Also, Giwa and Giwa (2013) carried out the modelling of a reactive distillation process using Layer-Recurrent Neural Network, and the simulated results they obtained were found to compare well with the measured ones. It has been observed that work on the use of artificial neural network to model this complex process of biodiesel production in a reactive distillation column is not common at all.

Therefore, this work has been carried out to model a reactive distillation process used for the production of biodiesel from the esterification reaction between palmitic acid and methanol using artificial neural network by

taking the mole fraction of biodiesel obtained from the bottom section of the reactive distillation column as the output variable and the temperatures and pressures of the feeds as well as the reboiler heat duty and reflux ratio of the column as the input variables of the model.

Methodology

The tool used for the artificial neural network modeling of the reactive distillation process used for the production of biodiesel (see Equation (1)) in this work was, actually, MATLAB ((Mathworks, 2013). However, the data used for the modeling were generated via the parametric utility of Aspen HYSYS (Aspen, 2012) model of the reactive distillation process developed. Shown in Figure (1) below is the Aspen HYSYS model of the reactive distillation process developed before the incorporation of the parametric unit operation that was containing the parametric utility of Aspen HYSYS. Meanwhile, before the addition of the parametric unit operation, the data given in Table 1 were used to run the developed Aspen HYSYS model, and it was ensured that the simulation converged. The addition of the parametric unit operation made the developed Aspen HYSYS model of the process to become as shown in Figure 2.

$$C_{16}H_{32}O_2 + CH_3OH \leftrightarrow C_{17}H_{34}O_2 + H_2O \quad (1)$$

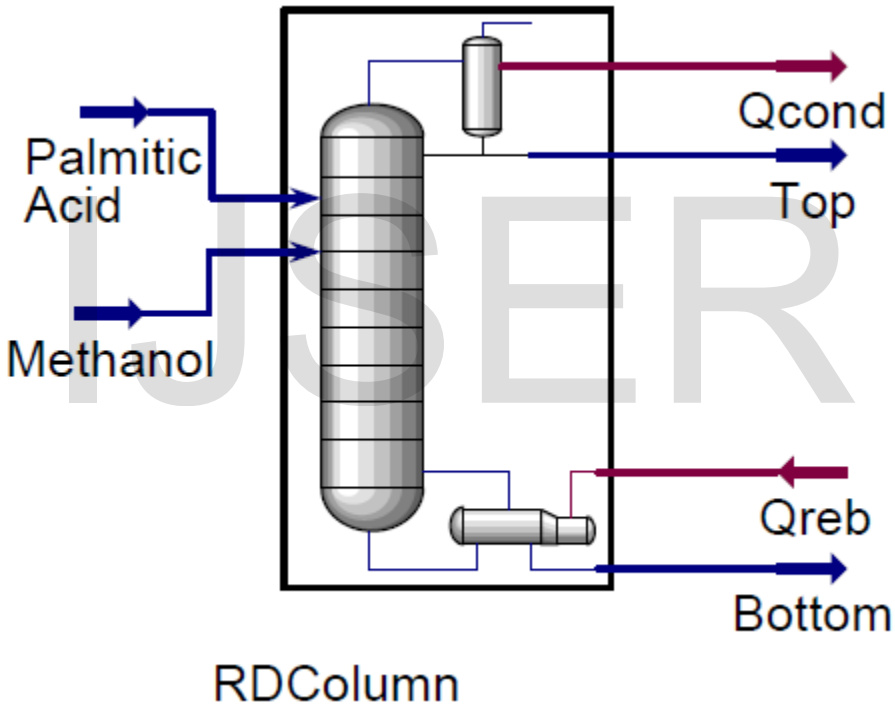

Figure 1. Aspen HYSYS model for biodiesel reactive distillation process

Table 1. Reactive distillation process simulation parameters

Parameter	Value
Palmitic Acid Feed	
Temperature (^oC)	350
Pressure (atm)	5
Molar flow (kgmole/hr)	100000
Mole fraction	1
Methanol Feed	

Temperature (^oC)	150
Pressure (atm)	1
Molar flow (kgmole/hr)	100000
Mole fraction	1

Property Package Wilson

Reaction

Type	Equilibrium
K_{eq} source	Gibbs Free Energy

Column

Type	Distillation Column Sub-Flowsheet
No of tray	30
Palmitic acid feed tray	8
Ethanol feed tray	19
Reflux ratio	3
Reboiler duty (kJ/s)	250000
Condenser type	Total
Reboiler type	Kettle
Condenser pressure (atm)	1
Condenser pressure drop (atm)	0
Reboiler pressure (atm)	1
Reboiler pressure drop (atm)	0

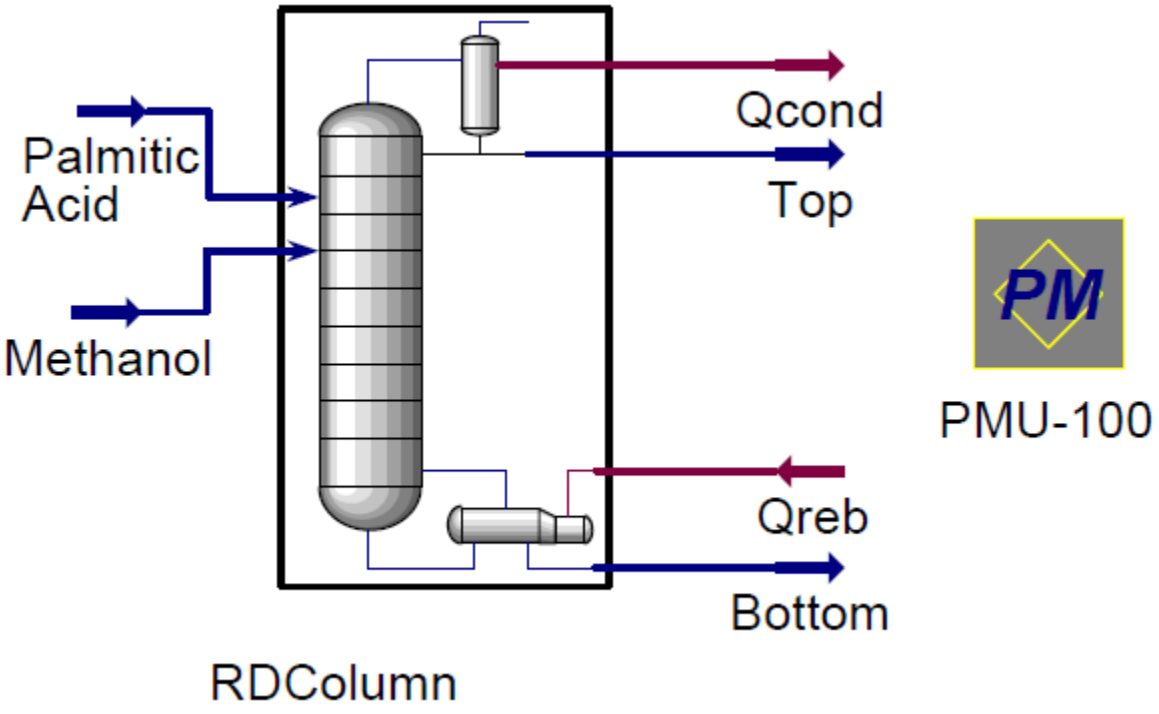

Figure 2. Aspen HYSYS model for the biodiesel reactive distillation process with parametric unit operation

The parametric unit operation could, actually, be used to simulate the developed model of the reactive distillation process directly using the neural network utility of the Aspen HYSYS, however, owing to the fact, in this research work, it was desired to use MATLAB to carry out the modeling of the artificial neural network of the process, the parametric utility was only used to generate the data required by MATLAB for the development of the artificial neural network model of the process.

The ranges of the input variables used to generate different data sets for training and prediction are as given in Tables 2 and 3, respectively, and the structure of the neural network employed is as given in Figure 3.

Table 2. Ranges of input variables for generating training data

Parameter	Initial value	Low limit	High limit
Palmitic acid feed temperature, ^{o}C	350	315	385
Palmitic acid feed pressure, atm	5	4.5	5.5
Methanol feed temperature, ^{o}C	150	135	165
Methanol feed pressure, atm	1	0.9	1.1
Reboiler heat duty, kJ/s	2.50E+05	2.25E+05	2.75E+05
Reflux ratio	3	2.7	3.3

Table 3. Ranges of input variables for generating prediction data

Parameter	Initial value	Low limit	High limit
Palmitic acid feed temperature, ^{o}C	350	325	380
Palmitic acid feed pressure, atm	5	4.7	5
Methanol feed temperature, ^{o}C	150	140	163
Methanol feed pressure, atm	1	0.91	1
Reboiler heat duty, kJ/s	2.50E+05	2.28E+05	2.70E+05
Reflux ratio	3	2.75	3.2

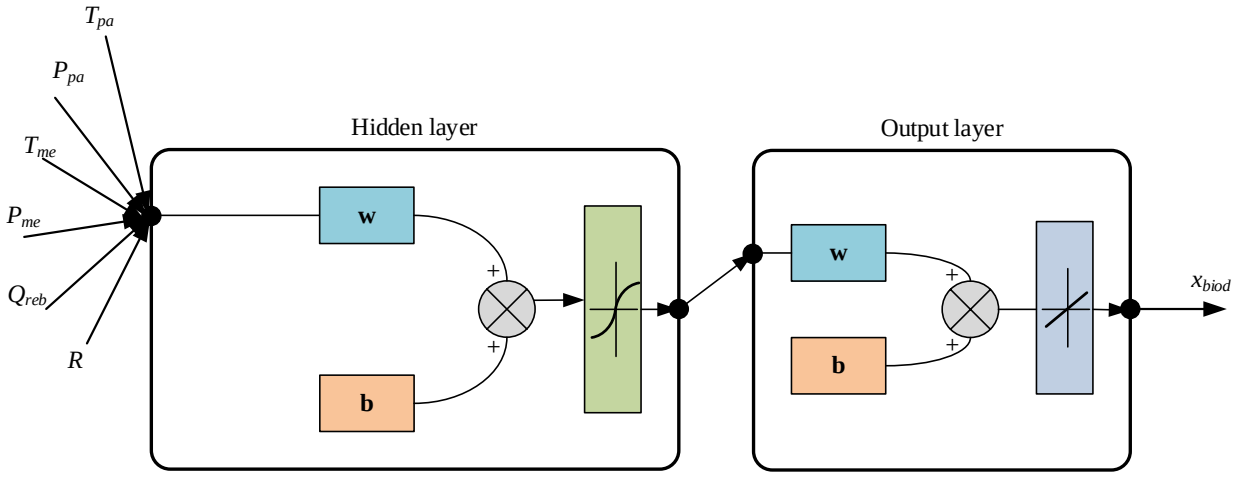

Figure 3. Structure of the feed-forward network of biodiesel reactive distillation process

After the data required had been generated, and the structure defined, the network was trained and the random number used for the initialization of the training of the network was obtained using multiplicative lagged Fibonacci generator, which is described in the work of Mascagni and Srinivasan (2004). Before using the algorithm for the training of the network, its seed number was optimized by varying it (the seed number) and recording the performance (taking to be the *mean of squared error*) of the artificial neural network.

Finally, after the training of the neural network model, using the input data generated for prediction purpose, it (the developed artificial neural network model) was used to predict the mole fraction of biodiesel obtained from the bottom section of the column in which the reactive distillation process was carried out.

Result and Discussion

Using the limits given Table 2, training data were generated for the input variables of the artificial neural network. Shown in Figures 4 – 9 below are the generated input variable data for palmitic acid feed temperature, palmitic acid feed pressure, methanol feed temperature, methanol feed pressure, reboiler heat duty and reflux ratio, respectively, for the training of the network. As can be noticed from the figures, the input data generated were random in nature. This was made to be like that so that the artificial neural network model being developed would be able to learn from different perspectives of the process. When the generated input variables were used to run the system, the results (mole fractions of biodiesel obtained from the bottom section of the column) given are as shown in Figure 10.

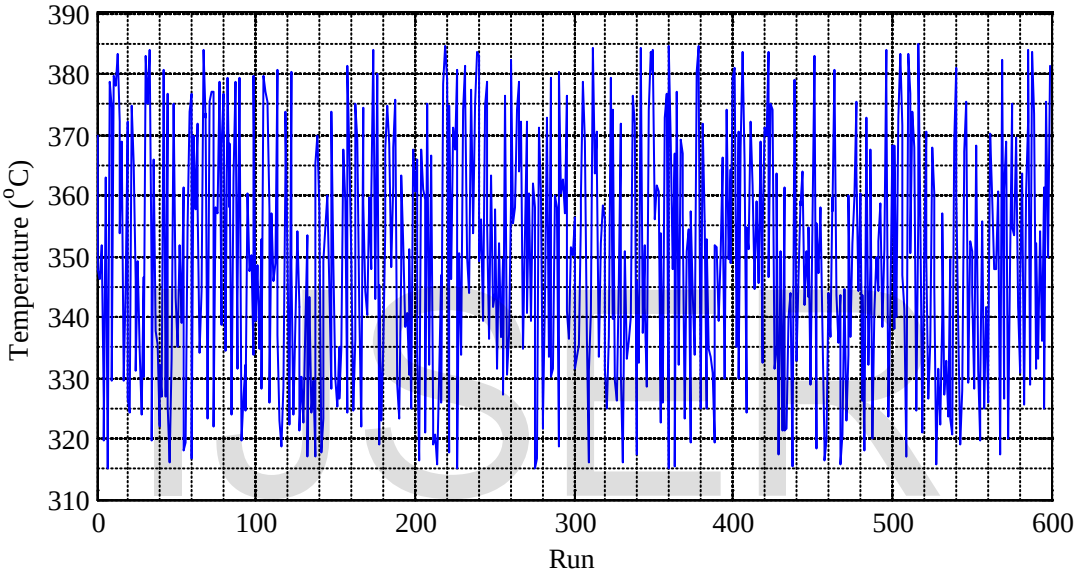

Figure 4. Palmitic acid feed temperature input used for generating the training data for the network

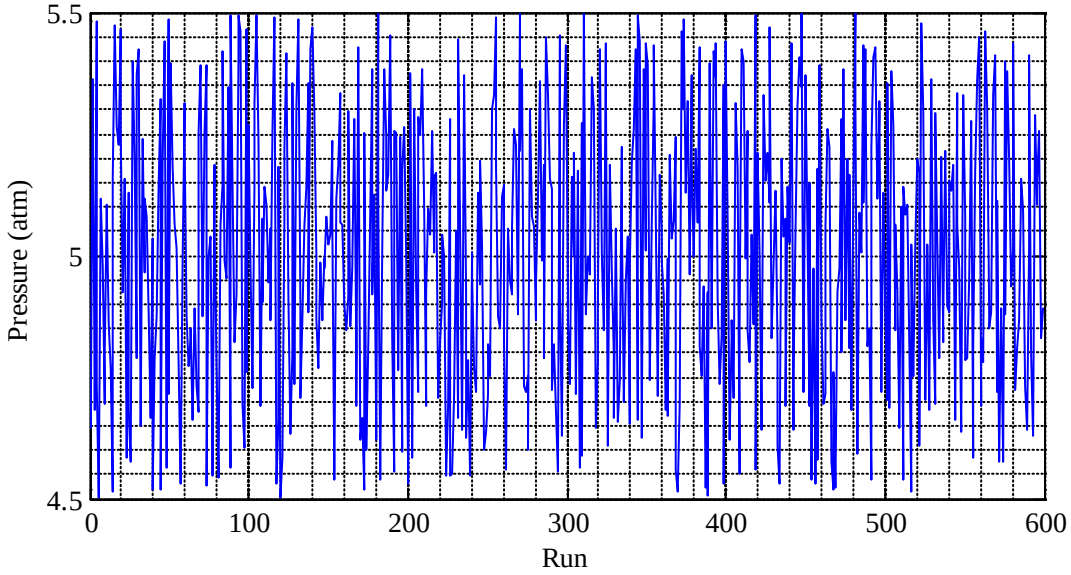

Figure 5. Palmitic acid feed pressure input used for generating the training data for the network

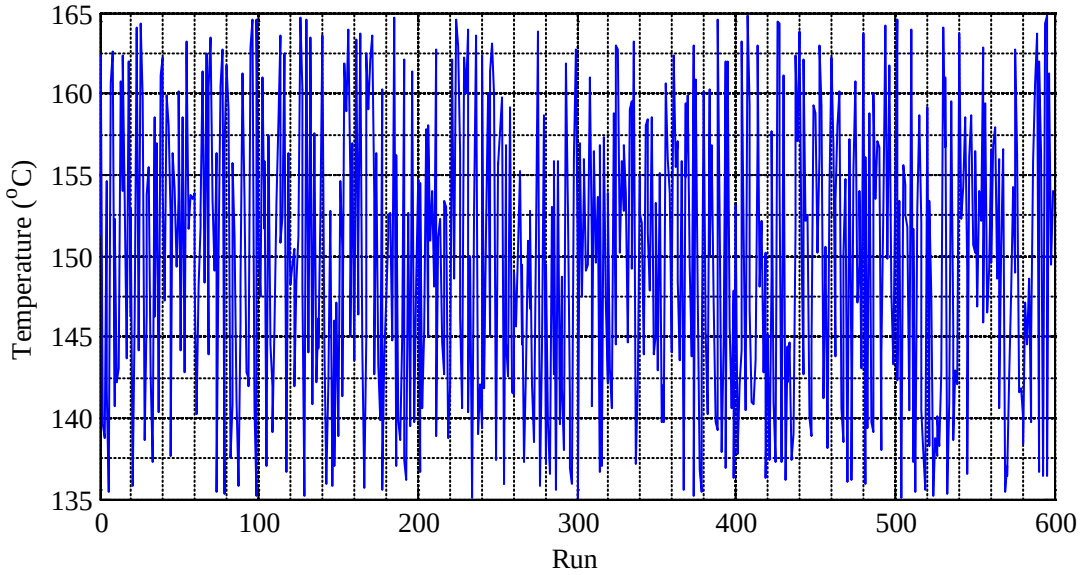

Figure 6. Methanol feed temperature input used for generating the training data for the network

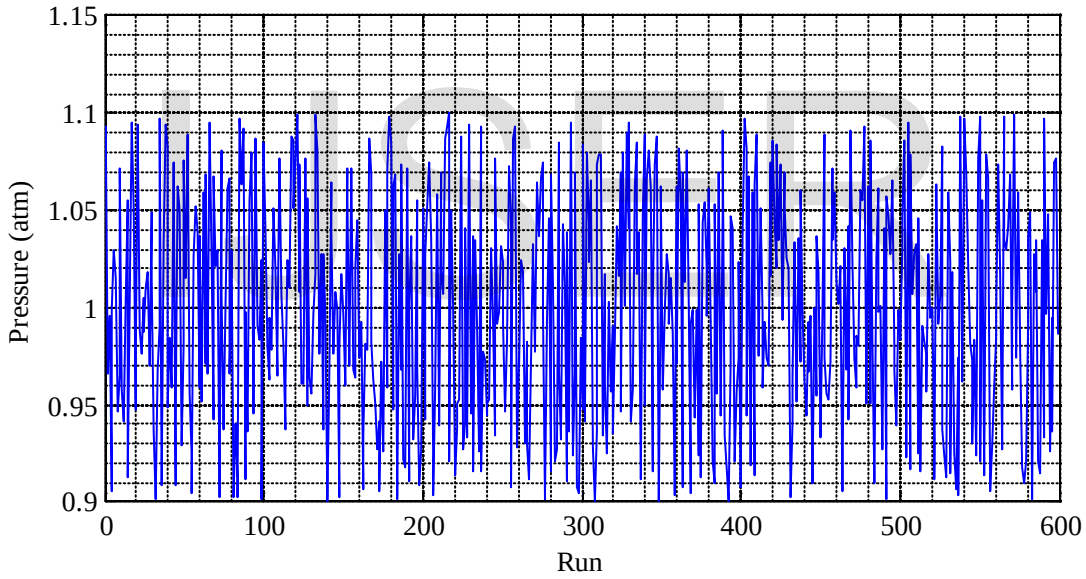

Figure 7. Methanol feed pressure input used for generating the training data for the network

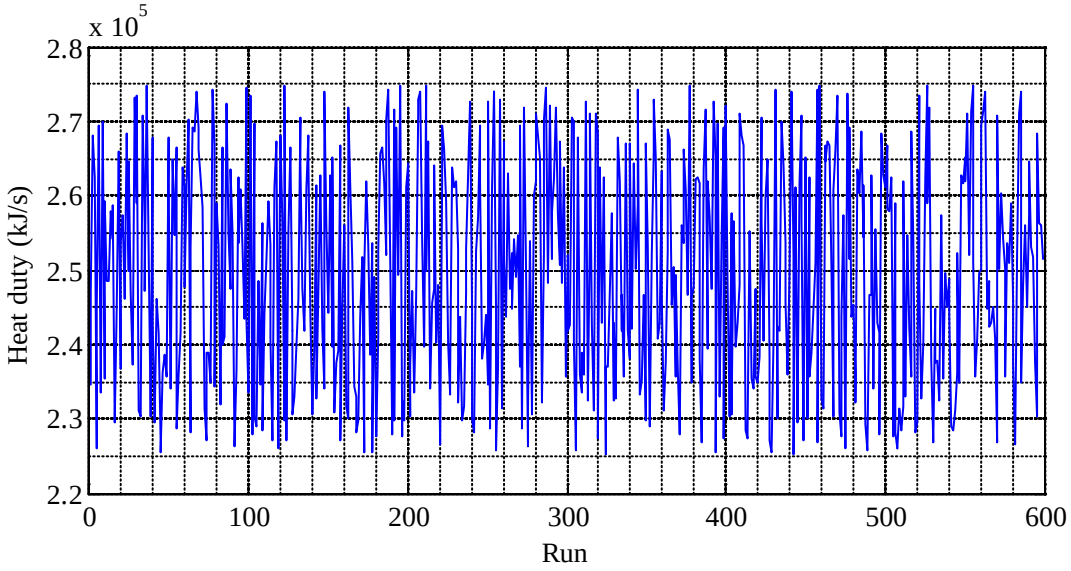

Figure 8. Reboiler heat duty input used for generating the training data for the network

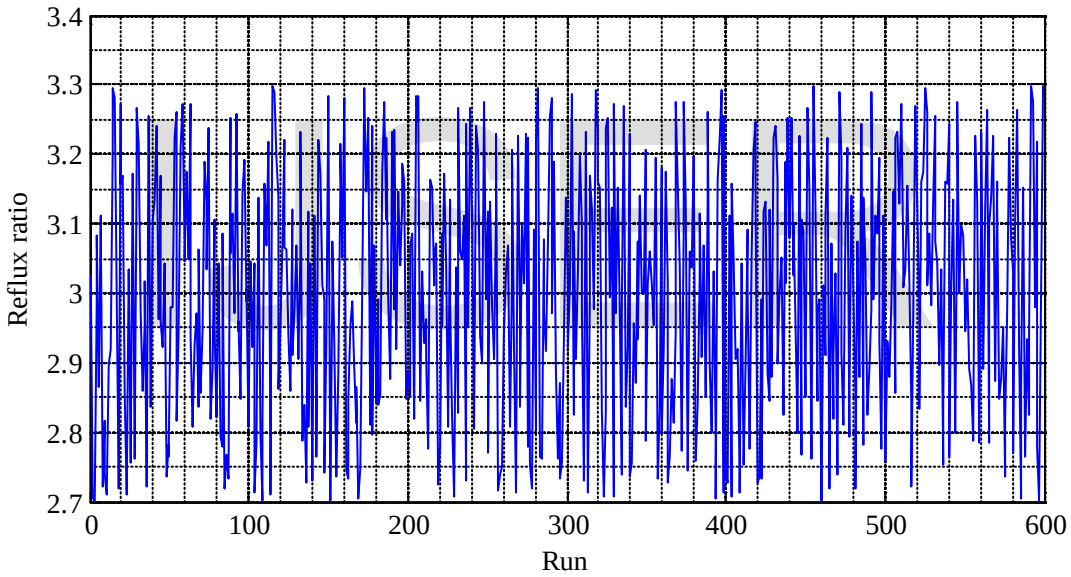

Figure 9. Reflux ratio input used for generating the training data for the network

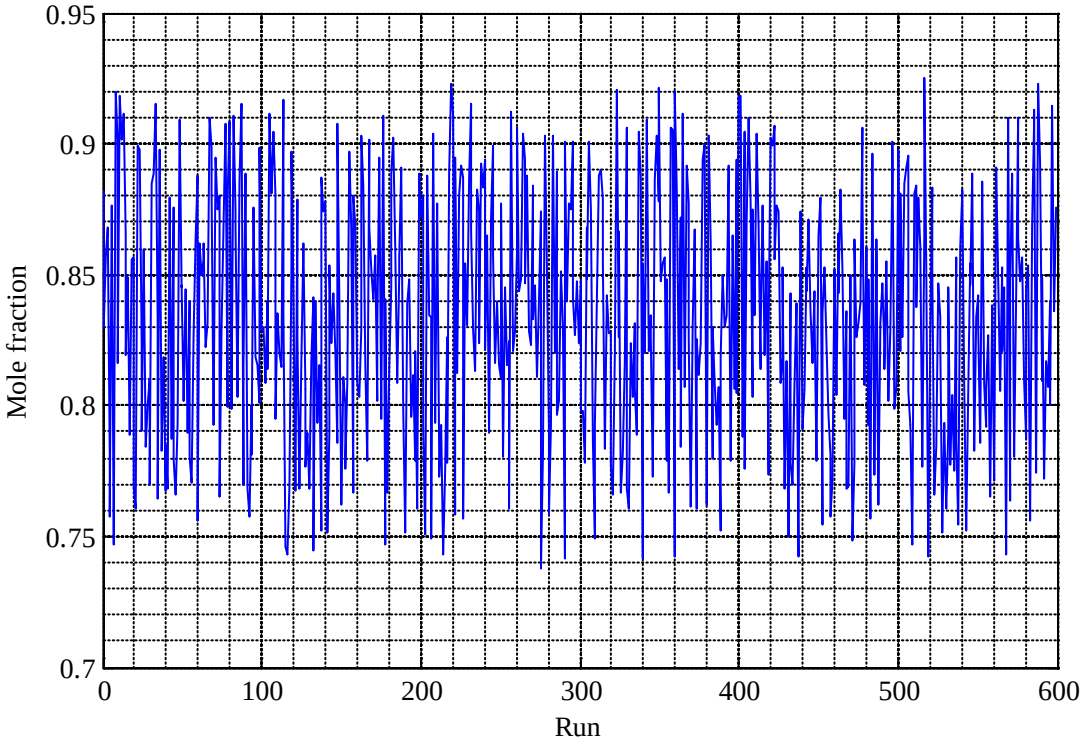

Figure 10. Measured biodiesel mole fraction data for network training

As can be observed from Figure 10, the mole fraction profile of biodiesel obtained from the system upon the application of the input variables, which were random values, was also found to be random in nature. This has demonstrated that the nature of the output variable (mole fraction of biodiesel) is very similar to the nature of the input variables of the process.

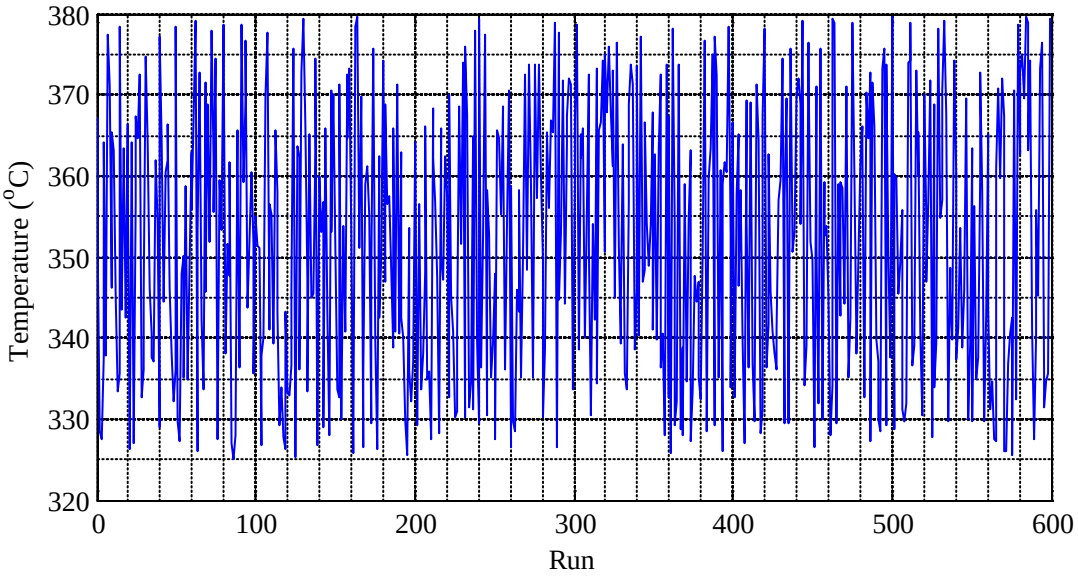

Figure 11. Palmitic acid feed temperature input used for generating the prediction data for the network

After obtaining the data for training the network, another set of data was generated, using another set of input variables, to test the prediction capacity of the network. The generated input variables used, for prediction purpose of the network, are as shown in Figures 11 – 16. The palmitic acid feed temperature and pressure input

variable data are shown in Figures 11 and 12, respectively. Given in Figures 13 and 14 are the generated input variable data of methanol feed temperature and pressure while the reboiler heat duty and the reflux ratio data are respectively given in Figures 15 and 16.

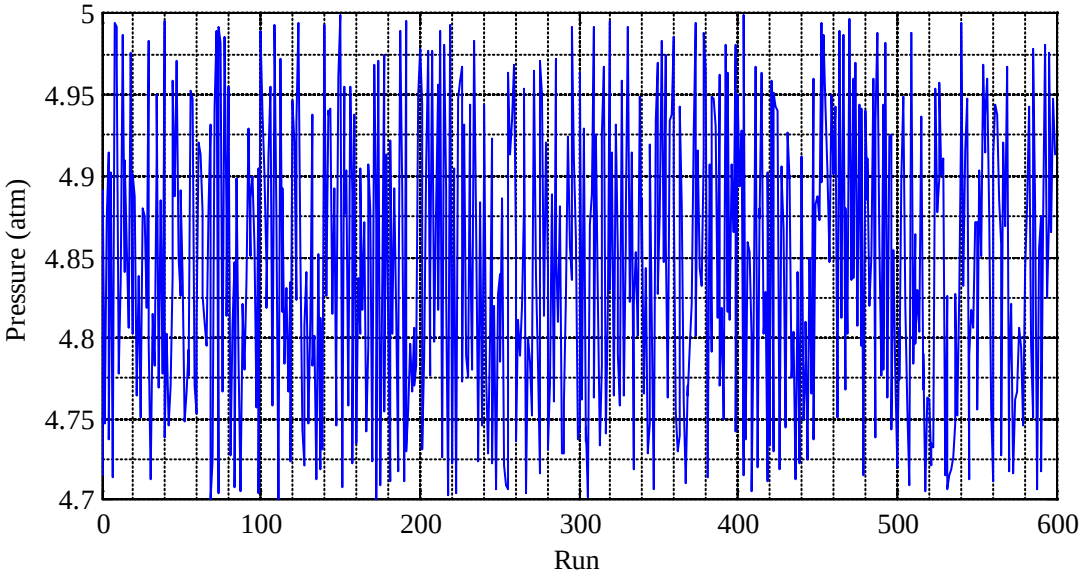

Figure 12. Palmitic acid feed pressure input used for generating the prediction data for the network

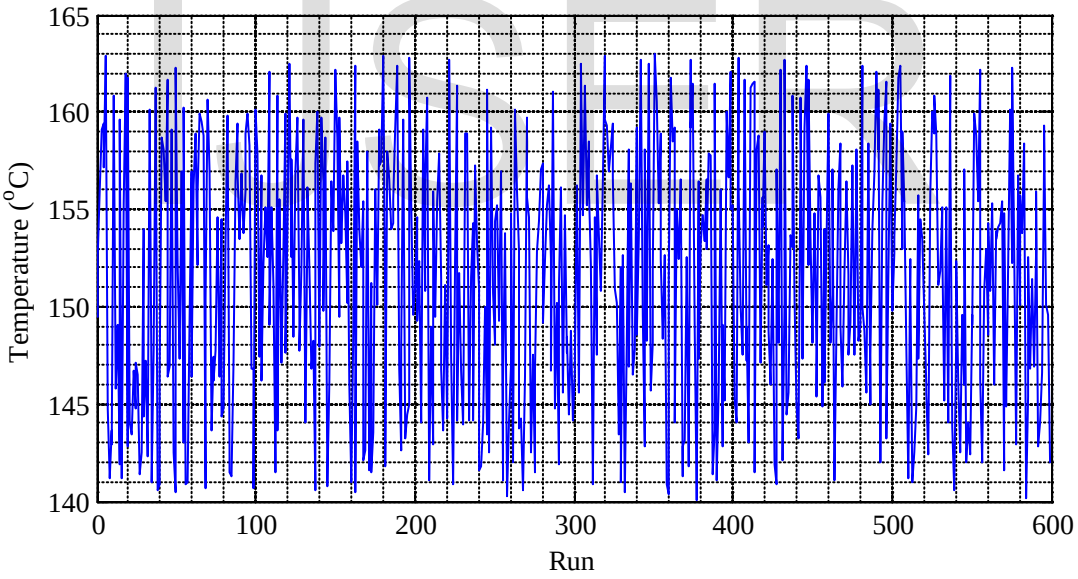

Figure 13. Methanol feed temperature input used for generating the prediction data for the network

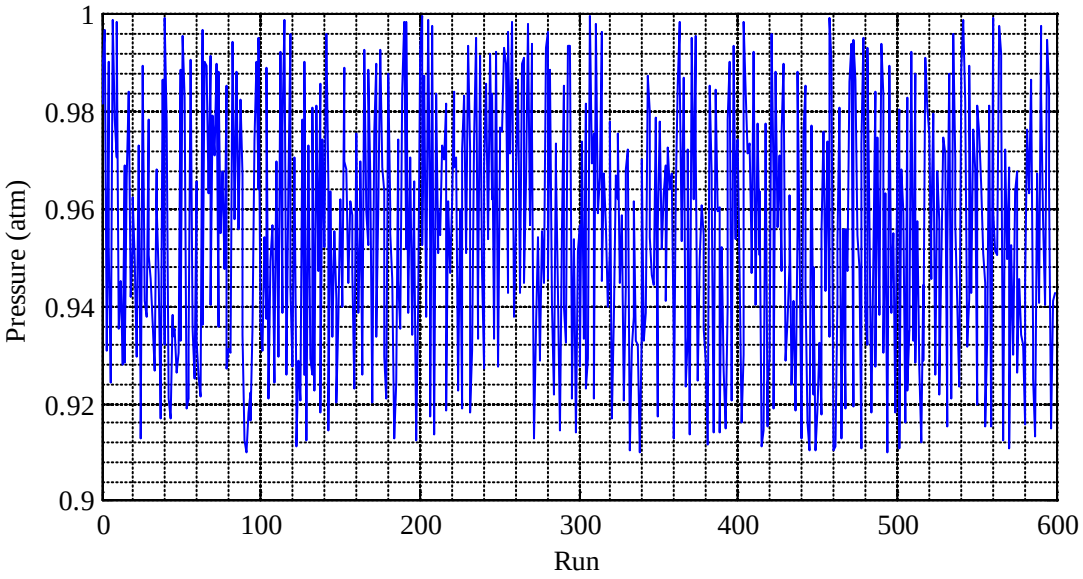

Figure 14. Methanol feed pressure input used for generating the prediction data for the network

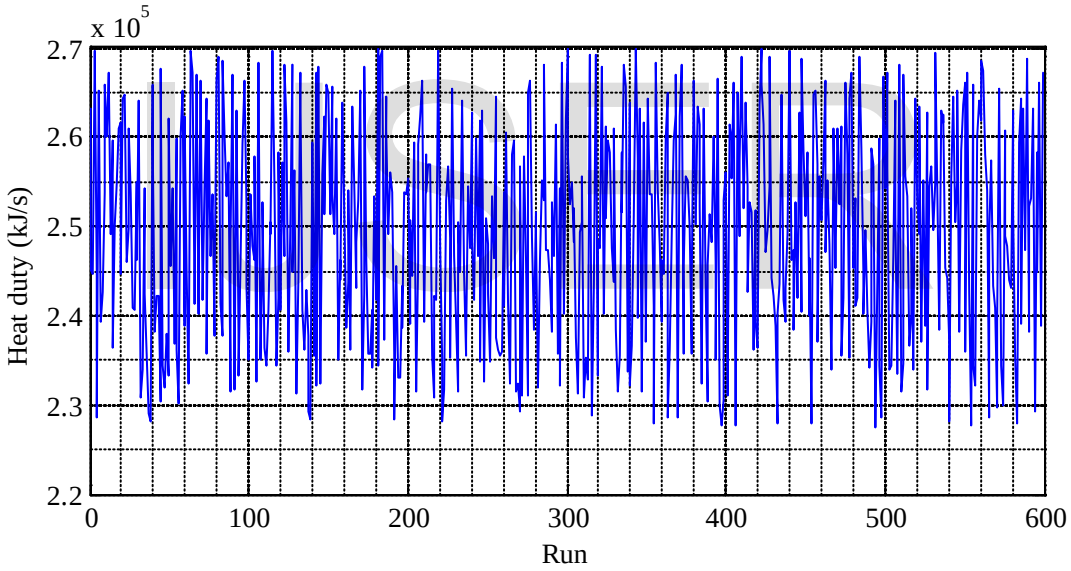

Figure 15. Reboiler heat duty input used for generating the prediction data for the network

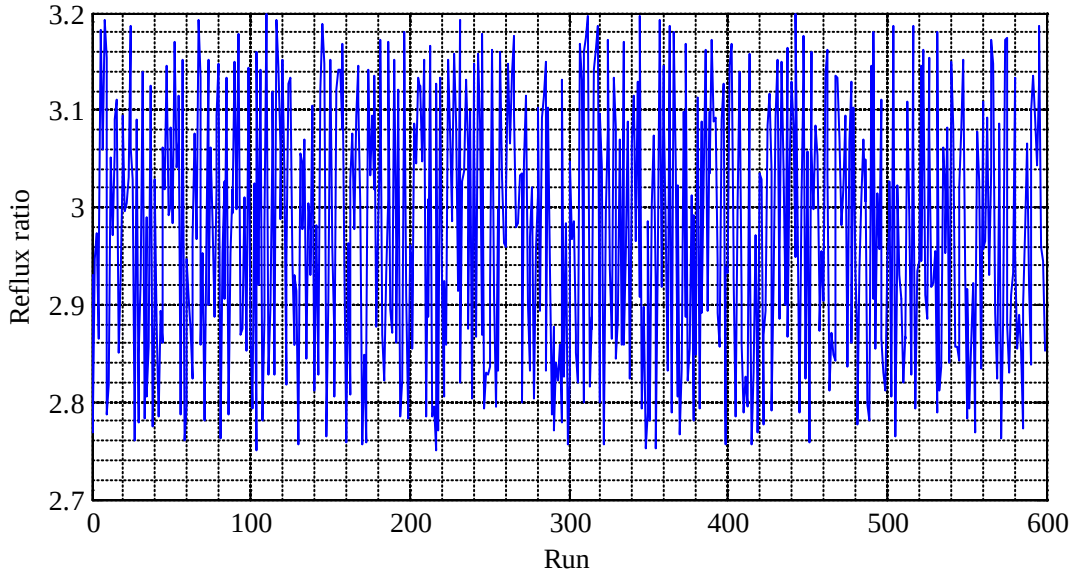

Figure 16. Reflux ratio input used for generating the prediction data for the network

When the generated input variable data of the prediction were used to run the system, the output (biodiesel mole fraction) obtained, as the prediction output data of the network, are given in Figure 17.

As can be observed from Figure 17, the mole fraction profile of biodiesel obtained from the bottom section of the reactive distillation column was found to be have a random nature similar, in nature, to those of its input variable data.

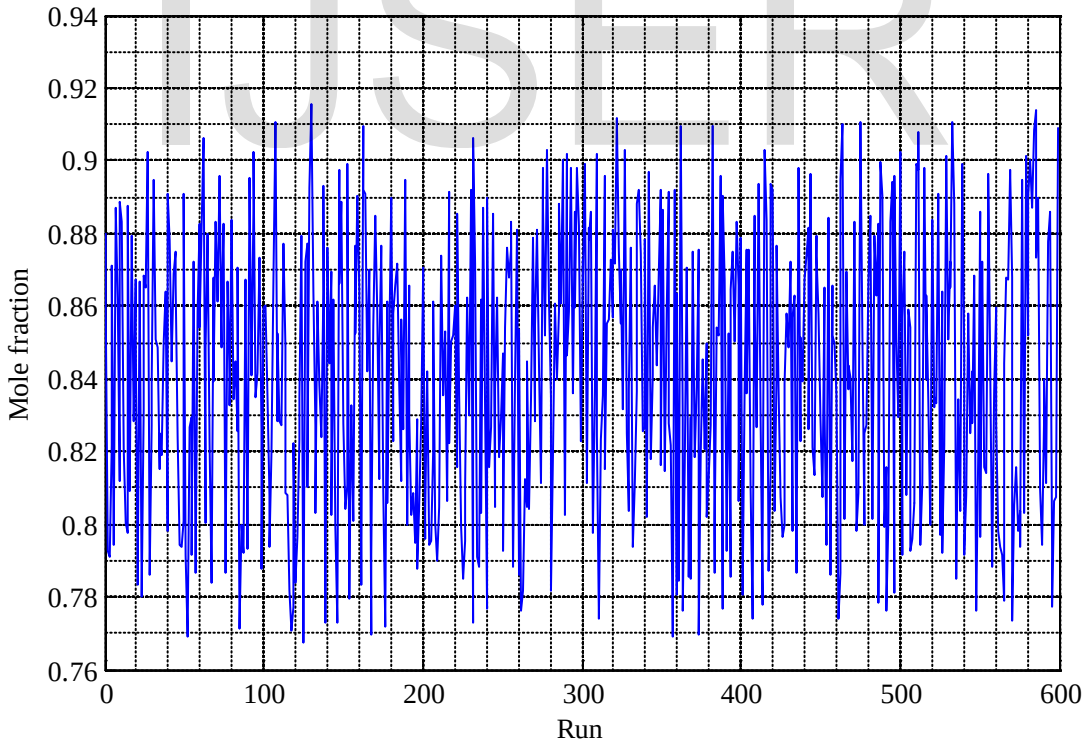

Figure 17. Measured biodiesel mole fraction data for prediction

After obtaining the data for the network training and prediction, it (the network) was set up, and there came the need to input an algorithm for generating the random number for training. The algorithm used in this work was

multiplicative lagged Fibonacci generator, and a parameter required by the algorithm in order to run is number of seed. In order to know the appropriate number of seed for the network, the algorithm was optimized by varying the number of seed and recording the performance of the network. The performance criterion used in this case was *mean of squared error*. As such, the values of means of squared errors obtained when the seed number of the algorithm was varied from 1 to 50 are as shown in Figure 18. According to the figure, for the seed number ranging from 1 to 50, the minimum value of the performance criterion, mean of squared error, was found to be 3.93E-09, and it occurred when the seed number was 47. Therefore, 47 seed number was used for the multiplicative lagged Fibonacci generator that produced the random number used for the training of the network, and the results obtained from the training are as given in Figure 19.

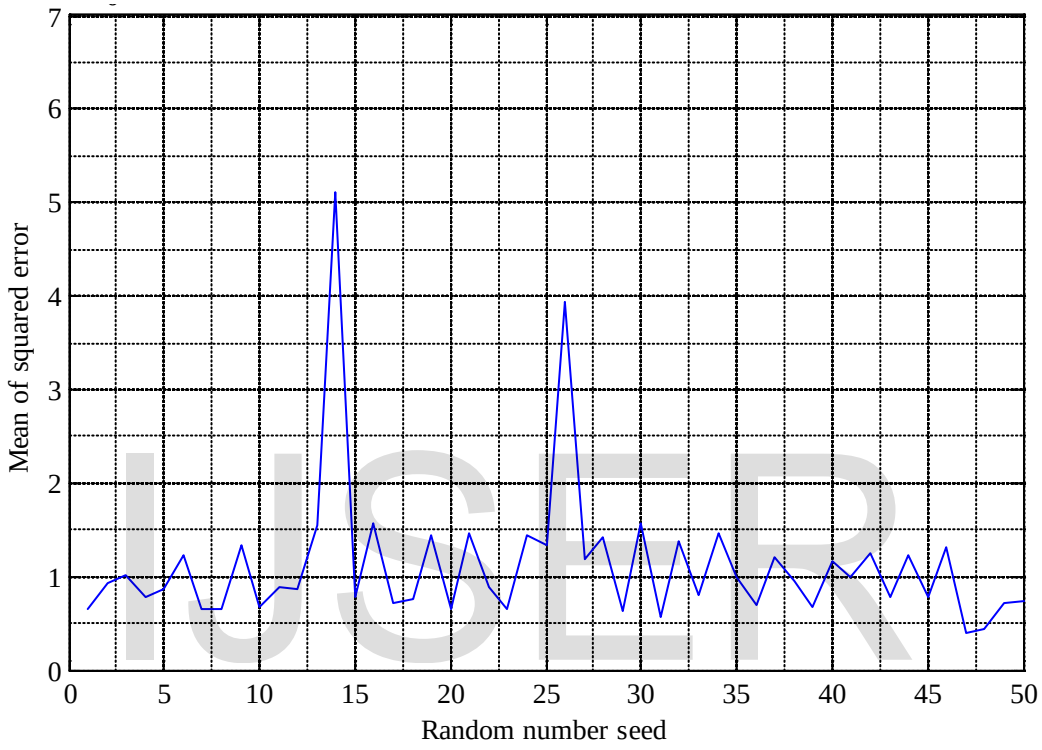

Figure 18. Variation of network performance (mean of squared error) with number of seed of random number generator

In Figure 19, the simulated and the measured mole fraction profiles of biodiesel are shown. The simulated values were obtained when the optimized seed number (47) was used for the algorithm (multiplicative lagged Fibonacci generator) used to generate the random number for the initialization of the training of the artificial neural network. As can be seen on the graph, there is a very good correlation between the simulated and the measured profiles of the biodiesel mole fraction. This is an indication that the developed artificial neural network model was able to represent the process very well. Of course, that was also revealed by its mean of squared error that has been found to be approximately zero (3.93E-09). Apart from the mean of squared error that was used to judge the performance of the developed artificial neural network in the training and simulation, sum of absolute error, mean of absolute error and sum of squared error were also estimated to be 0.016828, 2.80E-05, and 2.36E-06, respectively. These other performance criteria values were as well discovered to be low and appropriate enough for a good model. It was, therefore, discovered that the developed artificial neural network model has all the properties to be able to represent the process very well. Thus, it was expected that the model should be able to perform very well in predicting the mole fraction of biodiesel produced from the reactive distillation process being considered.

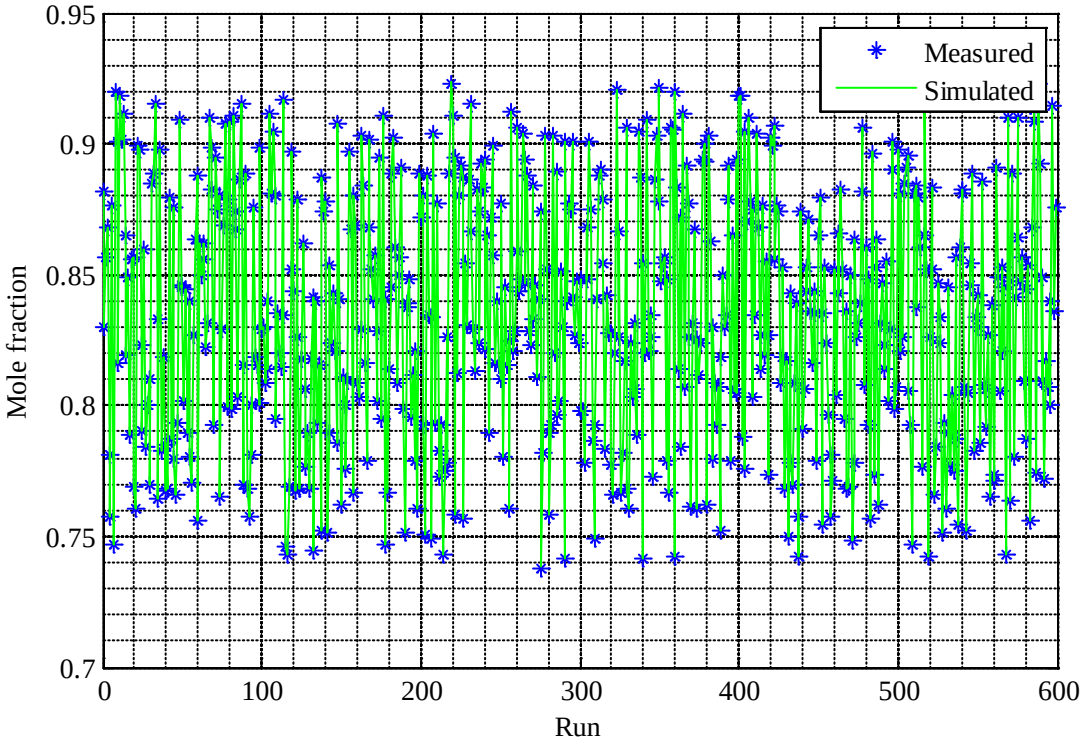

Figure 19. Simulated and measured mole fraction profiles of biodiesel

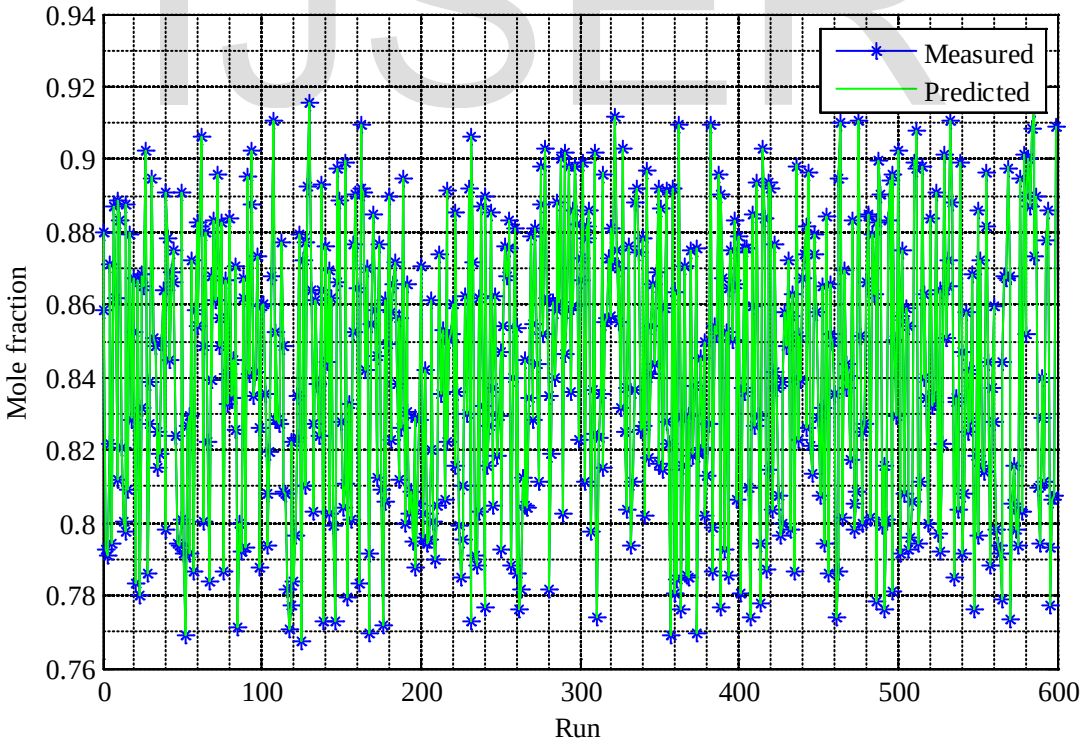

Figure 20. Predicted and measured mole fraction profiles of biodiesel

The results obtained when the developed artificial neural network was used to predict the mole fraction of biodiesel obtained from the reactive distillation process, by using the data (Figures 11 - 16) generated for prediction, are given in Figure 20. The good performance of the model can be seen on the figure as the good agreements between the predicted mole fractions of biodiesel and the measured ones can be clearly seen from the nature of the profiles. Also, the performance of the developed model in predicting the mole fraction of biodiesel obtained from the bottom section of the column was investigated by estimating the sum of absolute error, mean of absolute error, sum of squared error, and mean of squared error of the model in prediction to be 0.011804, 1.97E-05, 4.15E-07, and 6.92E-10, respectively. The results obtained from the estimation of the performance criteria have shown that the developed model has been able to perform very well in prediction too.

Comparing the performances of the model in simulation and prediction, it was discovered that the model was able to perform very well in prediction, even better than in simulation. This was found obvious from the values obtained for their performance criteria compared in Table 4.

Table 4. Comparison of the performance criteria of the developed model in training and prediction

Description	SAE	MAE	SSE	MSE
Training	0.016828	2.8E-05	2.36E-06	3.93E-09
Prediction	0.011804	1.97E-05	4.15E-07	6.92E-10

As can be observed from Table 4, it has been shown that all the performance criteria values of the model in prediction were less than those of its training. It then means that it is very possible for an artificial neural network model to have a better performance in prediction than in training.

Conclusion

The results obtained from the simulation of the artificial neural network model developed in this work have revealed that the developed model was able to represent the process very well because the mean of squared error of its performance estimated from the training of the network was found to be approximately zero (that is, 3.93E-09). Furthermore, very good performance was recorded when the model was used to predict the mole fraction of biodiesel produced from the bottom section of the reactive distillation column due to the fact the mean of squared error was found, in this case, to be 6.92E-10. It has, thus, been discovered that the developed neural network model was able to perform very well in predicting the mole fraction of biodiesel obtained from the reactive distillation process.

Nomenclature

MAE Mean of absolute error
MSE Mean of squared error
P_{me} Pressure of methanol feed, atm
P_{pa} Pressure of palmitic acid feed, atm
Q_{reb} Reboiler heat duty, kJ/s
R Reflux ratio of the column
SAE Sum of absolute error
SSE Sum of squared error
T_{me} Temperature of methanol feed, ^{o}C
T_{pa} Temperature of palmitic acid feed, ^{o}C
x_{biod} Mole fraction of biodiesel obtained from the bottom section of the column

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