Cascade-Forward Neural Network Modelling of a Biodiesel Reactive Distillation Process

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Abstract - In this work, a cascade-forward neural network model has been developed to represent a reactive distillation process used for the production of biodiesel from an esterification reaction between palmitic acid and methanol. In order to obtain data for the network, the parametric utility of an Aspen HYSYS prototype plant of the process developed using Distillation Column Sub-Flowsheet and Wilson model as the fluid package was utilized. The neural network model developed had six input parameters (palmitic acid feed temperature, palmitic acid feed pressure, methanol feed temperature, methanol feed pressure, reboiler heat duty and reflux ratio), and the output parameter was the mole fraction of the biodiesel obtained from the bottom section of the reactive distillation column. For the training of the neural network model, six different random number generators (Mersenne twister, multiplicative congruential generator, multiplicative lagged Fibonacci generator, combined multiple recursive generator, shift-register generator summed with linear congruential generator, and modified subtract with borrow generator) were tried by varying their seed numbers from 0 to 70, and the one with best performance, together with the corresponding seed number, was selected for the development of the cascade-forward neural network. The results obtained from the training and simulation carried out for the developed model showed the good representation of the process by the developed model because the estimated sum of absolute error, mean of absolute error, sum of squared error and mean of squared error of the model, which were the performance criteria used, were found to be favourable and had values of 1.16E-02, 1.93E-05, 5.46E-07, and 9.10E-10, respectively. Also, the performance of the developed model in predicting the mole fractions of the produced biodiesel was found to be very good as the sum of absolute error, the mean of absolute error, the sum of squared error and the mean of squared error, in this case, were estimated to be 8.39E-03, 1.40E-05, 2.17E-07, and 3.62E-10, respectively. In conclusion, cascade-forward neural network has been demonstrated to be very good in modelling this complex reactive distillation process for the production of biodiesel.

Keywords: Biodiesel, Reactive Distillation, Aspen HYSYS, Parametric Utility, Cascade-Forward Neural Network, Random Number Generator.

1 INTRODUCTION

Recently, integrated reactive separation processes have attracted attentions in both academic research and industrial fields (Völker *et al.*, 2007). One of these processes is known as reactive distillation, and it is very attractive, especially whenever conversion is limited by reaction equilibrium (Giwa and Karacan, 2012a; Giwa and Karacan, 2012c).

Reactive distillation is a process that permits the occurrence of both separation and chemical reaction in a single unit (Giwa and Karacan, 2012d; Giwa, 2013). It makes use of the benefits of equilibrium reaction with distillation to enhance conversion, with the condition that the product of interest has the highest or the lowest boiling point (Taylor and Krishna, 2000; Giwa, 2012). This process has a lot of advantages which include reduced investment and operating costs due to increased yield of a reversible reaction as a result of the separation of the product of interest from the reaction mixture (Pérez-Correa *et al.*, 2008), high conversion, improved selectivity, low energy consumption, ability to carry out difficult separations and avoidance of azeotropes (Jana and Adari, 2009; Giwa, 2012; Giwa and Giwa, 2012). It has been applied in a few number of ways in industries for many years, but the last decade has shown an increase in both its research and applications (Agreda *et al.*, 1990), particularly in the area of its modelling and design.

The modelling and design of reactive distillation (RD) systems are considerably more complex than those involved for either conventional reactors or conventional distillation columns because the introduction of an in situ separation

within the reaction section of the column leads to complex interactions between the vapour-liquid equilibrium, the vapour-liquid mass transfer, the intra-catalyst diffusion (for heterogeneously catalysed processes) and the chemical kinetics of the reactive distillation process. Such interactions have been discovered to lead to phenomena of multiple steady states and complex dynamics (Baur *et al.*, 2000; Giwa and Karacan, 2012b) of the process.

In the design of a reactive distillation process, a model is required (Giwa and Karacan, 2012b). However, obtaining a robust model for a reactive distillation process is still a challenge to Chemical Engineers owing to the integration of reaction and separation that has made the process to exhibit complex behaviours (Khaledi and Young, 2005), as mentioned before, such as steady state multiplicity, process gain sign changes (bidirectionality) and strong interactions between process variables (Jana and Adari, 2009; 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 Karacan, 2012c; Giwa and Giwa, 2013; Giwa *et al.*, 2015).

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, 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; Giwa *et al.*, 2015).

Even though artificial neural network can be used, to a high level of accuracy, to represent this process due to its complex and economical (associated with its various advantages and benefits) natures, it should be noted that it (reactive distillation) cannot be applied to all Chemical Engineering processes, but a process that accommodates the application of reactive distillation is production of biodiesel.

Biodiesel can be produced mostly from oils, which are edible ones, such as palm oil, sunflower oil, and soybean oil via transesetrification process using a catalyst like sodium hydroxide. However, the commercialization of biodiesel production from those oil types still have drawbacks due to high cost of vegetable oil and the purification of the formed biodiesel product. Therefore, an alternative approach discovered for the production of biodiesel was the use of jatropha oil. 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 they also discovered that biodiesel with high purity could be produced using palmitic acid via *esterification reaction* (Giwa *et al.*, 2015).

Esterification of fatty acid and alcohol to produce biodiesel can, actually, be carried out in a batch reactor (Omota *et al.*, 2003). However, that method has been found to possess many problems because of its low conversion, heavy capital investments and high energy costs (Gao *et al.*, 2007; Giwa *et al.*, 2015). The attempt to resolve this problem was what necessitated the use of the reactive distillation process that is modelled using artificial neural network because of its complexities.

Actually, 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 networks (Nonlinear AutoRegressive (NAR), Nonlinear AutoRegressive with eXogenous inputs (NARX) and Nonlinear Input-Output (NIO)) models to predict the temperatures of the top and the bottom sections of a 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 NIO models were found not to be satisfactory. Also, Giwa and Karacan (2012e) developed two nonlinear black box (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. Recently,

Giwa *et al.* (2015) used a feed-forward artificial neural network to represent a reactive distillation process used for the production of biodiesel from the esterification reaction between palmitic acid and methanol, and their results showed good agreements between the experimental and the measured mole fractions of biodiesel obtained. As can be observed from the survey of the literature carried out so far, it has been revealed that work on the use of cascade-forward neural network to represent a reactive distillation process has not been reported.

So, in this work, the representation of a reactive distillation process using cascade-forward neural network modelling method has been carried out. The case study process used in the modelling of the reactive distillation is biodiesel production from the esterification reaction between palmitic acid and methanol, as it was used in the work of Giwa *et al.* (2015).

2 METHODOLOGY

The approach used in carrying out the cascade-forward neural network modelling of the reactive distillation process used for the production of biodiesel (see Equation (1)) in this work involved the use the Neural Network Toolbox of MATLAB (Mathworks, 2013). However, owing to the fact that the toolbox required data to be able to do the modelling, the results obtained by Giwa *et al.* (2015) were utilized. For clarity purposes, the method used for the data generation are still, and a little bit further, outlined in this work.

The data were generated through the use of the parametric utility of Aspen HYSYS (Aspen, 2012) model of the reactive distillation process developed and shown in Figures 1 and 2. Figure 1 shows the developed Aspen HYSYS model of the process before the incorporation of the unit operation having the parametric utility of Aspen HYSYS. Meanwhile, before the addition of the parametric unit operation, the data given in Table 1 were used to simulate the developed Aspen HYSYS model to convergence. The addition of the parametric unit operation made the developed Aspen HYSYS model of the process to look as that shown in Figure 2.

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

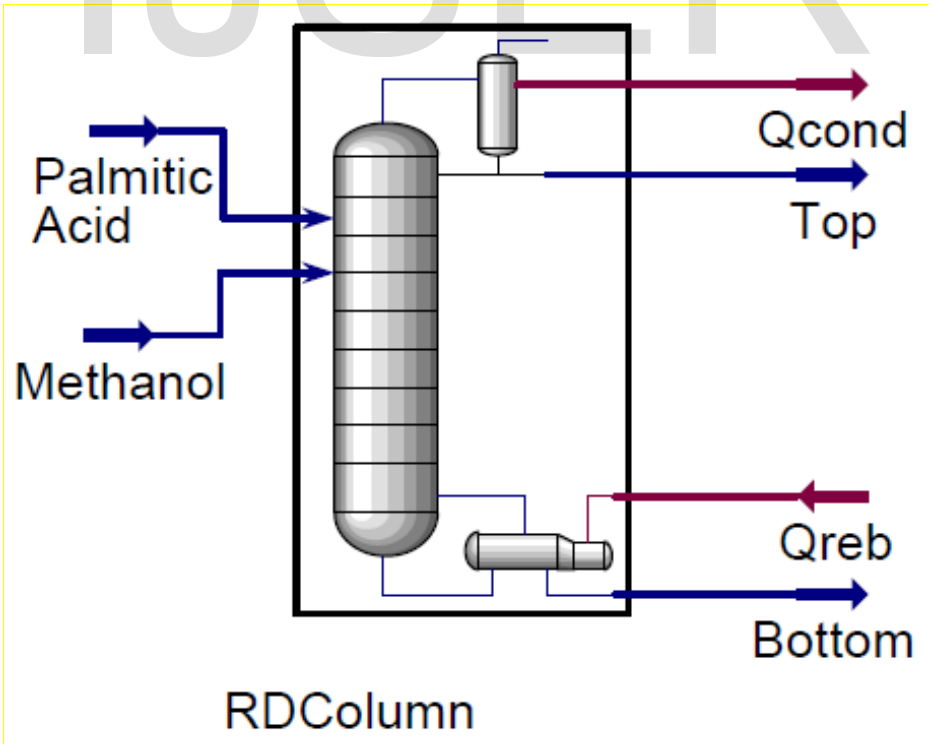

Figure 1. Aspen HYSYS model of biodiesel reactive distillation process

Table 1. Reactive distillation process modelling and simulation parameters

Parameter	Value
Palmitic Acid Feed	
Temperature (°C)	350
Pressure (atm)	5
Molar flow (kmol/hr)	100000
Mole fraction	1
Methanol Feed	
Temperature (°C)	150
Pressure (atm)	1
Molar flow (kmol/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

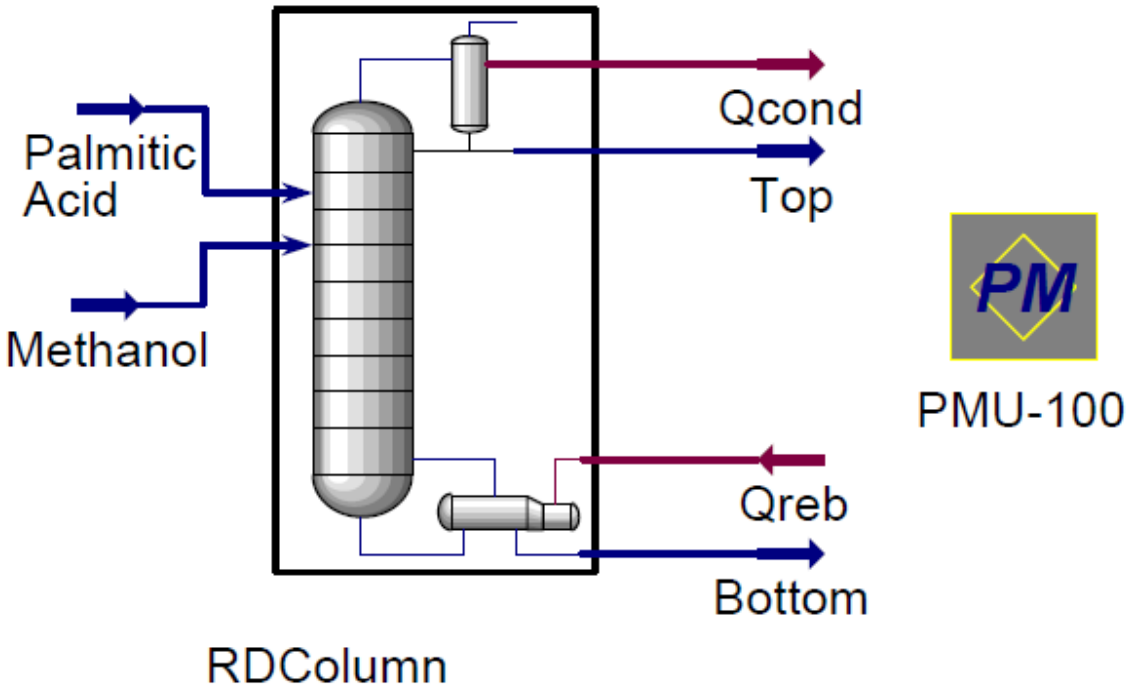

Figure 2. Aspen HYSYS model for the biodiesel reactive distillation process with parametric unit operation

The ranges of the input variables used for the generation of the different data sets for training and prediction are given in Tables 2 and 3, respectively, and the structure of the cascade-forward neural network employed is shown in Figure 3.

Table 2. Training data input ranges

Parameter	Initial value	Low limit	High limit
Palmitic acid feed temperature, °C	350	315	385
Palmitic acid feed pressure, atm	5	4.5	5.5
Methanol feed temperature, °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. Prediction data input ranges

Parameter	Initial value	Low limit	High limit
Palmitic acid feed temperature, °C	350	325	380
Palmitic acid feed pressure, atm	5	4.7	5
Methanol feed temperature, °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

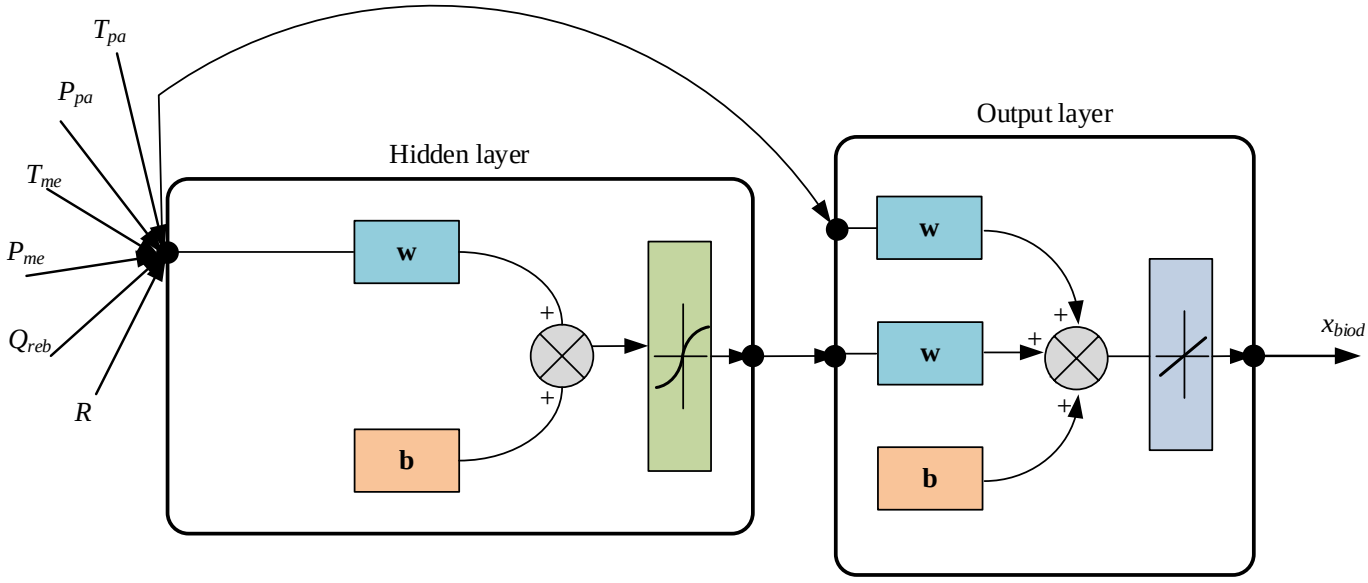

Figure 3. Structure of the cascade-forward network of biodiesel reactive distillation process

After the data required had been generated, and the structure was defined, the network was trained using the parameters given in Table 4, and different random number generators were used to generate the values for the initialization of the training. The random number generators considered, together with their codes in MATLAB, are those given in Table 5.

Before using the random number generator algorithms for the training of the network, their seed numbers were optimized by varying it from 0 to 70, simulating and recording their performances in the simulations. The performance criterion used in this case was "*mean of squared error*". Based on the performances recorded, one of the random number generators was selected and used to carry out the training of the developed model for the process.

Table 4. Neural Network model formulation parameters

Parameter	**Value**
Number of inputs	6
Number of outputs	1
Number of hidden layers	1
Number of neurons in hidden layer	5
Hidden layer transfer function	*tansig*
Output layer transfer function	*purelin*
Training algorithm	*Levenberg-Marquardt*

Table 5. Random number generators and their codes in MATLAB (Mathworks, 2013)

Generator	**Code**
Mersenne twister	*mt19937ar*
Multiplicative congruential generator	*mcg16807*
Multiplicative lagged Fibonacci generator	*mlfg6331_64*
Combined multiple recursive generator	*mrg32k3a*
Shift-register generator summed with linear congruential generator	*shr3cong*
Modified subtract with borrow generator	*swb2712*

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

3 RESULT AND DISCUSSION

Using the limits of the input variables given Tables 2 and 3, input random data were generated, and the graphical representation of the input data generated are already given in the work of Giwa *et al.* (2015).

When the generated input data were used to run the developed Aspen HYSYS model of the process, the mole fraction profile of biodiesel obtained as the bottom product are given in Figures 4 and 5 for training and prediction of the network, respectively.

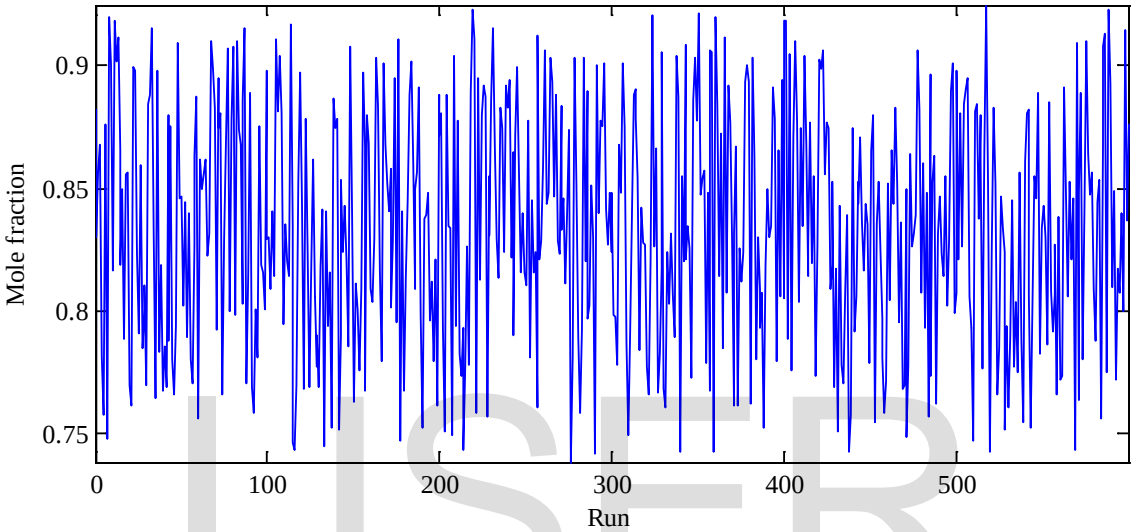

Figure 4 Biodiesel mole fraction data measured for cascade neural network training

Looking at Figures 4 and 5, it was discovered that the mole fraction profiles of biodiesel obtained from the system for training and prediction of the network were random in nature. This revealed that the outputs (mole fraction of biodiesel) of the process were similar in nature to the applied inputs because the inputs generated were also random in nature (cf. Giwa *et al.*, 2015).

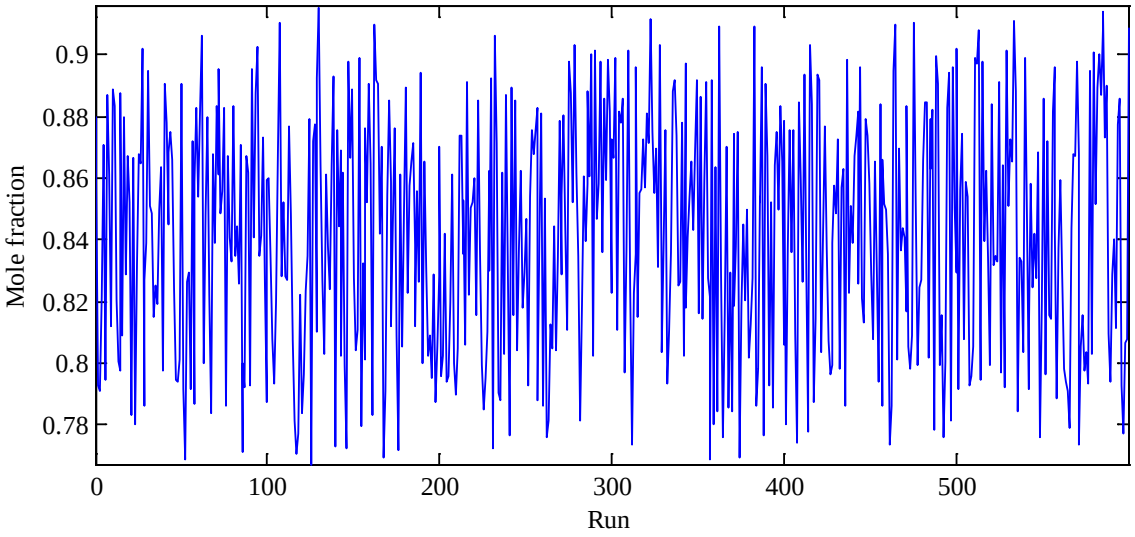

Figure 5. Biodiesel mole fraction data measured for cascade neural network prediction

Shown in Figure 6 is the variation of the performance of Mersenne twister random number generator as its seed number was varied. According to the figure, the best performance of the algorithm was observed when its seed number was 47. Furthermore, the seed number that gave the best performance when multiplicative congruential generator was used to obtain the random number of the training of the network was 56 (See Figure 7). From Figure 8 showing the network performance (mean of squared error) against seed number for multiplicative congruential generator, it was found that the best seed number was just 2, and combined multiple recursive generator, the plot of which is given in Figure 9 gave the best seed number to be 31. According to Figures 10 and 11, the best seed numbers for shift-register generator summed with linear congruential generator and modified subtract with borrow generator were obtained to be 45 and 6, respectively.

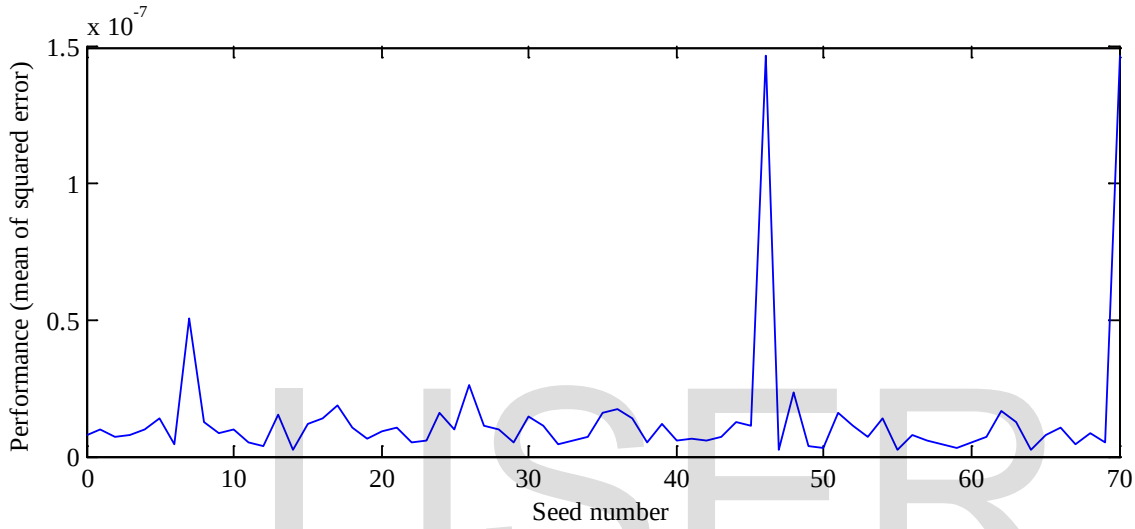

Figure 6. Network performance (mean of squared error) versus seed number of Mersenne twister random number generator

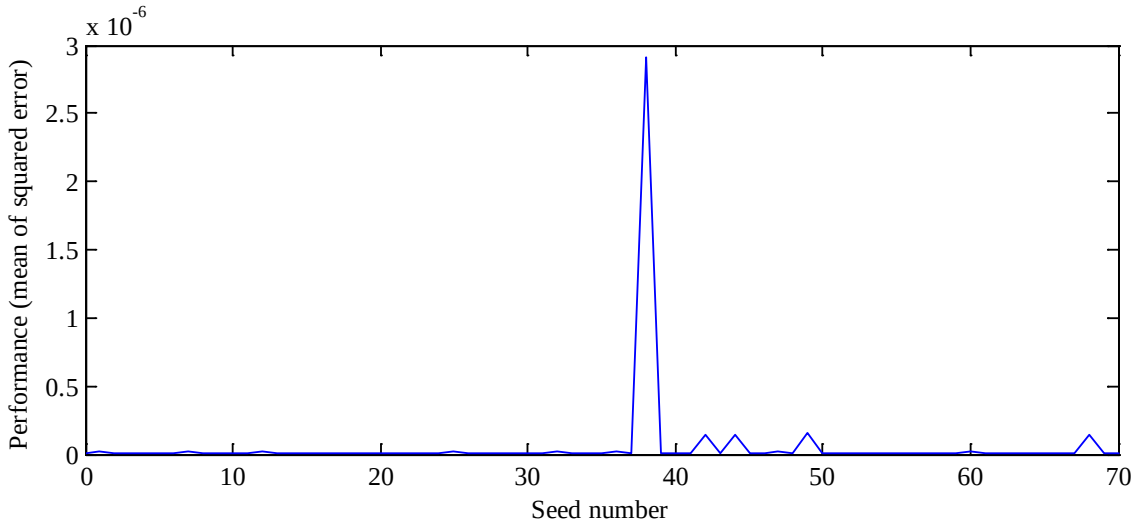

Figure 7. Network performance (mean of squared error) versus seed number of multiplicative congruential generator

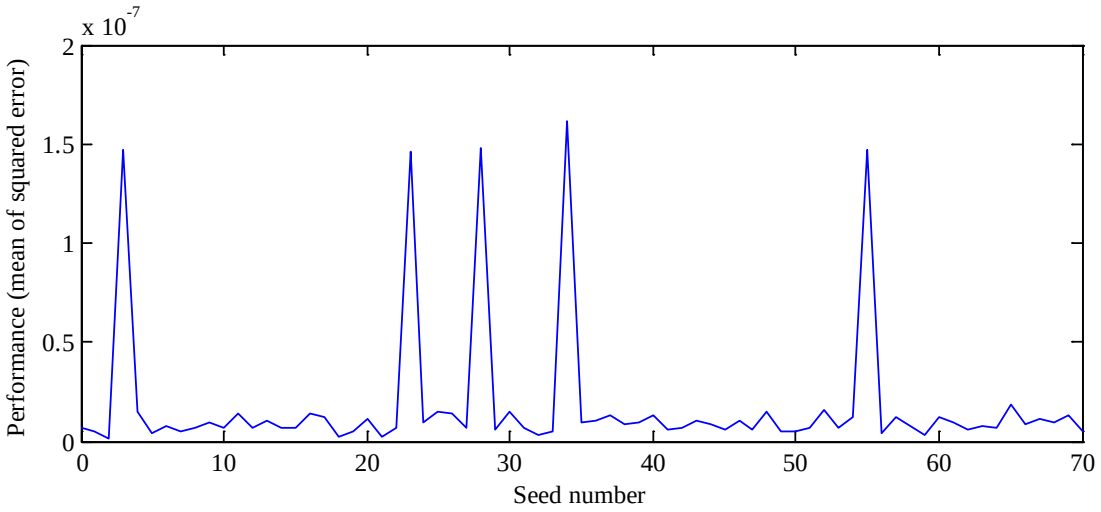

Figure 8. Network performance (mean of squared error) versus seed number of multiplicative lagged Fibonacci generator

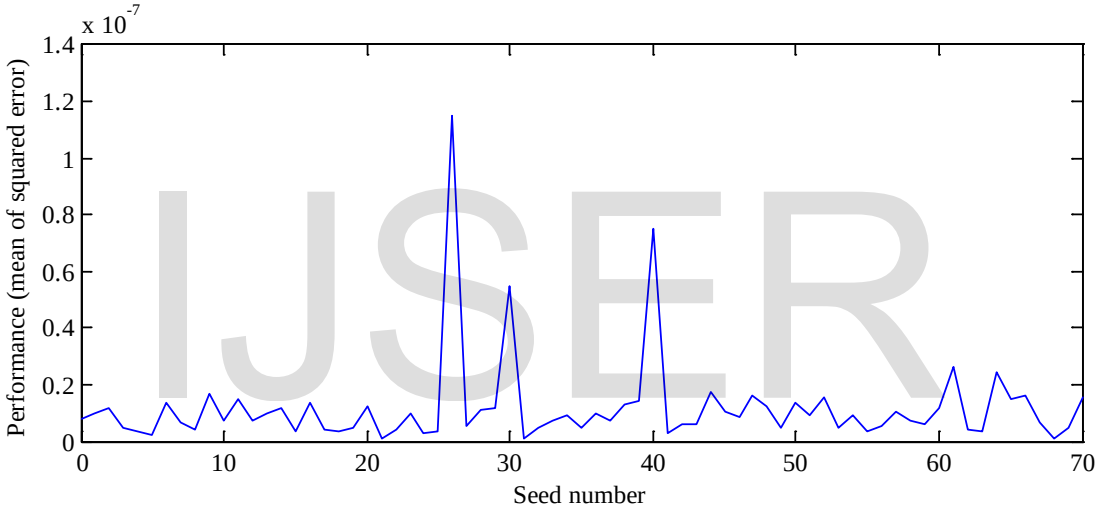

Figure 9. Network performance (mean of squared error) versus seed number of combined multiple recursive generator

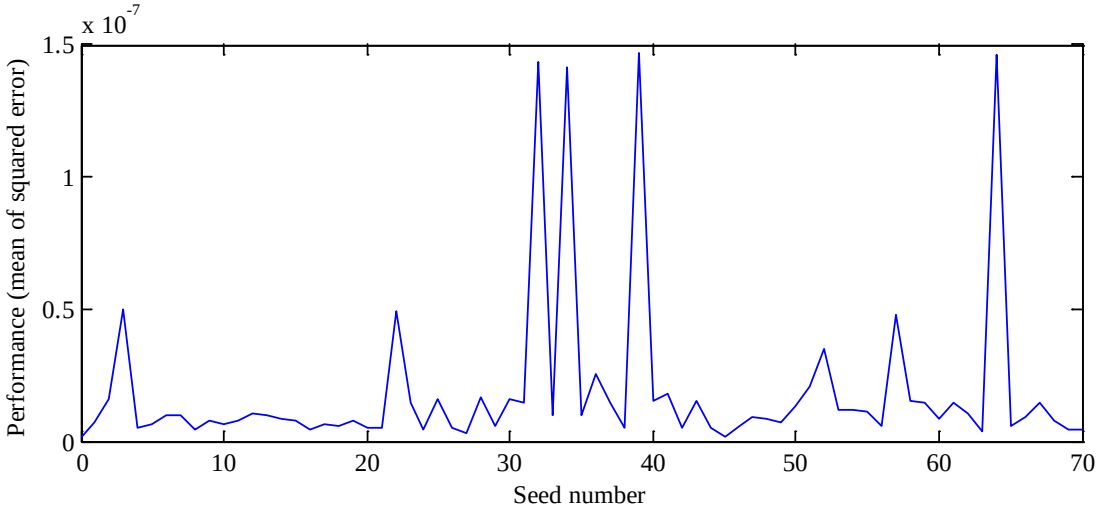

Figure 10. Network performance (mean of squared error) versus seed number of shift-register generator summed with linear congruential generator

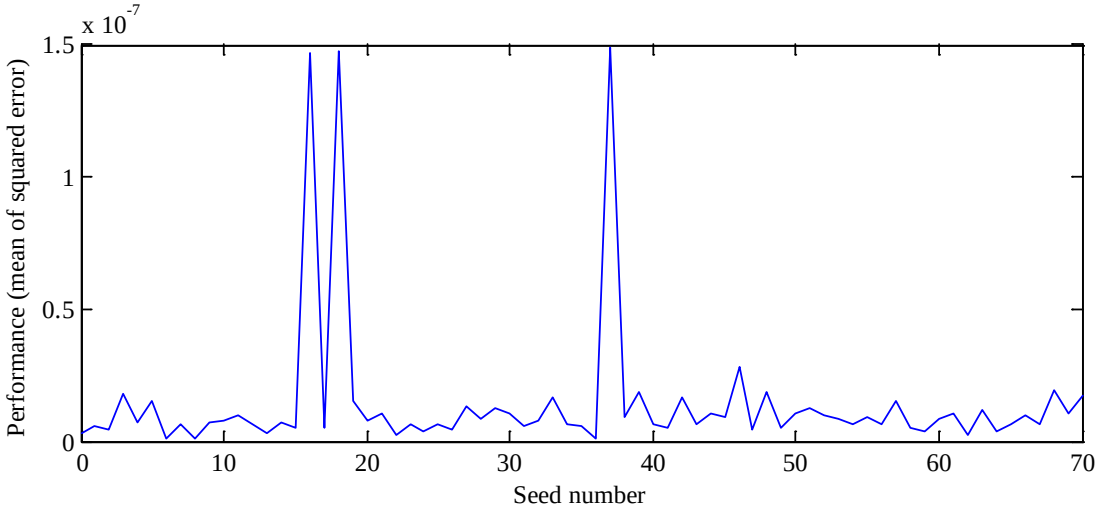

Figure 11. Network performance (mean of squared error) versus seed number of modified subtract with borrow generator

Based on the results obtained from the performances of the random number generators as their seed numbers were varied, the best performances of the algorithms were compared as given in Table 6, and it was discovered that the random number generator algorithm with the best performance, among all, was modified subtract with borrow generator because it was able to give the least mean of squared error (9.10E-10) when its seed number was 6. As such, this random number generator was selected as the one used to train the developed cascade-forward neural network, and the results obtained from the training were as given in Figure 12.

Table 6. Comparison of the performances of the random number generator algorithms

Algorithm	MSE	Seed number
Mersenne twister (default)	2.16E-09	47
Multiplicative congruential generator	1.03E-09	56
Multiplicative lagged Fibonacci generator	1.73E-09	2
Combined multiple recursive generator	9.37E-10	31
Shift-register generator summed with linear congruential generator	1.77E-09	45
Modified subtract with borrow generator	9.10E-10	6

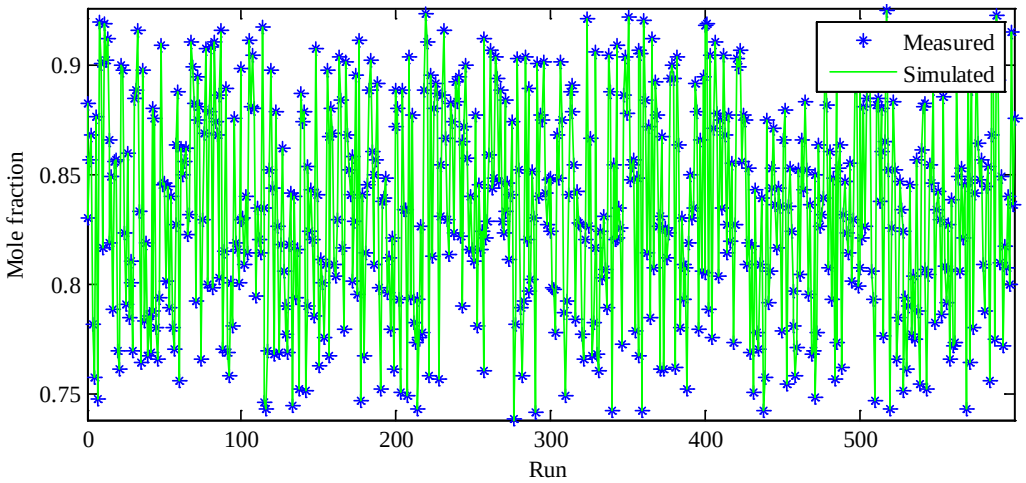

Figure 12. Simulated and measured mole fraction profiles of biodiesel

Figure 12 shows the simulated and the measured biodiesel mole fraction profiles. The simulated values of the profile were obtained using the modified subtract with borrow generator with the optimized seed number of 6 to generate the random number for the initialization of the training of the cascade-forward neural network developed. As can be observed from the figure, a very good correlation was found to exist between the simulated and the measured profiles of the biodiesel mole fraction obtained from the bottom section of the reactive distillation column used. This good correlation revealed that the developed cascade-forward neural network model was able to represent the process very well as this was also shown by the low value of the mean of squared error (performance value) of the model simulation that was estimated to be 9.10E-10 (a value that is approximately zero).

Moreover, the developed cascade-forward neural network model was used to predict the biodiesel mole fraction using the data generated for that purpose and given in the work of Giwa *et al.* (2015), and the results obtained as the predicted biodiesel mole fraction profile, together with the measured ones, are as shown in Figure 13. From the figure, it was seen that the model was able to perform well also in predicting the mole fraction of the biodiesel produced because good agreements were found to exist between the predicted values and the measured ones. This good performance was also revealed from the performance value (mean of squared error) of the prediction that was calculated to be 3.62E-10.

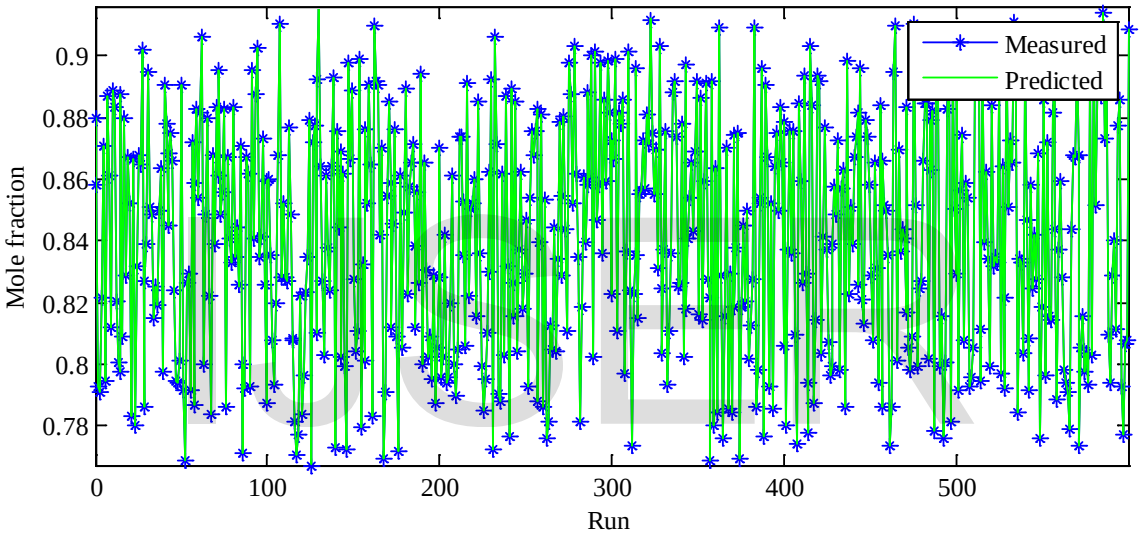

Figure 13. Predicted and measured mole fraction profiles of biodiesel

In addition to using the mean of squared error as the criterion for the determination of the performance of the developed cascade-forward neural network model in simulating and predicting the biodiesel mole fraction, some other criteria were used. The additional criteria used were sum of absolute error, mean of absolute error and sum of squared error of the model, and their values obtained from the simulation and the prediction carried out are given in Table 7.

Table 7. Comparison of the performance criteria of the developed model in training/simulation and prediction

Description	SAE	MAE	SSE	MSE
Training/simulation	1.16E-02	1.93E-05	5.46E-07	9.10E-10
Prediction	8.39E-03	1.40E-05	2.17E-07	3.62E-10

Comparisons of the values obtained for all the performance criteria (sum of absolute error, mean of absolute error, sum of squared error and mean of squared error of the model) indicated that the developed cascade-forward neural network model was able to perform very well both in simulation and in prediction. Also noticed from the performance of the model was that the performances of the model was better in prediction than in simulation, as can be seen from the performance values given in Table 7 in which all the error values of the prediction were found to be less than those of the

training/simulation. This observation was found to be in good agreement with one of the findings of Giwa *et al.* (2015) that developed a feed-forward artificial neural network model for a reactive distillation process used for biodiesel production and, also, obtained the performance of their developed model to be better in prediction than in simulation.

4 CONCLUSION

The results obtained from the training and simulation of the cascade-forward neural network model developed for the reactive distillation process used for the production of biodiesel revealed good representation of the process by the developed model because the estimated sum of absolute error, mean of absolute error, sum of squared error and mean of squared error of the model, which were the performance criteria used, were found to be favourable and have values of 1.16E-02, 1.93E-05, 5.46E-07, and 9.10E-10, respectively. Not only that, the developed model was also found to perform very well in predicting the mole fraction of the biodiesel obtained because the sum of absolute error, mean of absolute error, sum of squared error and mean of squared error of the model were estimated to be 8.39E-03, 1.40E-05, 2.17E-07, and 3.62E-10, respectively in prediction. Therefore, it has been demonstrated that cascade-forward neural network model is a good tool in representing this complex reactive distillation process for the production of biodiesel.

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NOMENCLATURE

b	Neural network bias
Bottom	Bottom products
MAE	Mean of absolute error
MSE	Mean of squared error
NAR	Nonlinear AutoRegressive,
NARX	Nonlinear AutoRegressive with eXogenous inputs
NIO	Nonlinear Input-Output
PM	ParaMetric
P_{me}	Pressure of methanol feed, atm
PMU	ParaMetric Utility
P_{pa}	Pressure of palmitic acid feed, atm
Q_{cond}	Condenser heat duty, kJ/s
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
Top	Top products
T_{pa}	Temperature of palmitic acid feed, ^{o}C
w	Neural network weight
x_{biod}	Mole fraction of biodiesel obtained from the bottom section of the column

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