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APPLICATION OF ARTIFICIAL NEURAL NETWORK AND RESPONSE SURFACE METHODOLOGY FOR MODELLING OF HYDROGEN PRODUCTION USING NICKEL LOADED ZEOLITE

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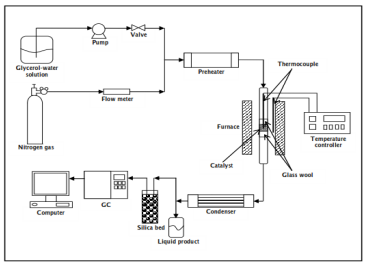
Graphical abstract



Abstract

Hydrogen gas production via glycerol steam reforming using nickel (Ni) loaded zeolite (HZSM-5) catalyst was focused on this research. 15 wt % Ni(HZSM-5) catalyst loading has been investigated based on the parameter of different range of catalyst weight (0.3-0.5g) and glycerol flow rate (0.2-0.4mL/min) at 600 °C and atmospheric pressure. The products were analyzed by using gas-chromatography with thermal conductivity detector (GC-TCD), where it used to identify the yield of hydrogen. The data of the experiment were analyzed by using Response Surface Methodology (RSM) and Artificial Neural Network (ANN) in order to predict the production of hydrogen. The results show that the condition for maximum hydrogen yield was obtained at 0.4 ml/min of glycerol flow rate and 0.3 g of catalyst weight resulting in 88.35 % hydrogen yield. 100 % glycerol conversion was achieved at 0.4 of glycerol flow rates and 0.3 g catalyst weight. After predicting the model using RSM and ANN, both models provided good quality predictions. The ANN showed a clear superiority with R^2 was almost to 1 compared to the RSM model.

Keywords: Hydrogen gas, glycerol steam reforming, Ni-HZSM-5, response surface methodology, artificial neural network

Abstrak

Penukaran gliserol terhadap gas hidrogen yang menggunakan HZSM-5 yang telah diubah suai dengan nikel merupakan fokus utama dalam kajian ini. 15 % nikel(HZSM-5) digunakan untuk menjalankan eksperimen pada perbezaan berat pemangkin (0.3-0.5 g) dan kadar aliran gliserol (0.2-0.4 mL/min) pada tekanan atmosfera dan suhu 600 ° C. Produk ini akan dianalisis dengan menggunakan gas kromatograf (GC-TCD) untuk mengkaji peratus hydrogen terhasil. Kemudian, eksperimen data dianalisa menggunakan kaedah gerak balas permukaan (RSM) dan rangkaian neural tiruan (ANN) untuk menjangka hidrogen

yang terhasil. Keputusan eksperimen menunjukkan 88.35 % penghasilan optimum gas hidrogen telah terhasil pada kadar aliran gliserol = 0.4 mL/min and berat pemangkin = 0.3 g dengan 100% penggunaan gliserol pada kadar aliran gliserol = 0.4 mL/min dan berat pemangkin = 0.3 g. Selepas membuat perbandingan menggunakan RSM dan ANN, kedua-dua model menunjukkan kualiti yang baik. Namun, ANN memberikan keputusan yang lebih jelas dengan mempunyai R^2 hampir 1 berbanding dengan RSM.

Kata kunci: Gas hidrogen, pembaharuan wap gliserol, Ni-HZSM-5, kaedah gerak balas permukaan, rangkaian neural tiruan

1.0 INTRODUCTION

Glycerol is a byproduct during manufacturing of several chemicals such as petroleum, soap and biodiesel. Biodiesel is an alternative fuel for diesel engines that produced from vegetable oil or animal fat with a simple monohydric alcohol such as methanol. The reaction requires a catalyst, usually a strong base, such as sodium hydroxide, and produces new chemical compounds called methyl esters that also known as biodiesel. According to Gerpen [1] and Shawn and Conley [2], biodiesel is the mono alkyl esters of long fatty acids derived from renewable lipid feedstock such as animal fats or vegetable oil that can be used in compression ignition diesel engines.

Generally, biodiesel was produced through a process known as transesterification, which involves altering the chemical properties of the oil by using methanol. It is a simple process that gives high conversions of yields with only glycerol as a byproduct. Glycerol production increased as the production of biodiesel increased due to the crude glycerol as a byproduct of biodiesel production. The generation of crude glycerol gives a yield at about 10% (wt/wt) of biodiesel during the process of biodiesel production. In that case, the global biodiesel market was estimated to reach 37 billion gallons by 2016 with an average annual growth of 42%, in which about 4 billion gallons of crude glycerol will be produced [3].

Therefore, it is imperative to find alternative uses for glycerol. Glycerol has many uses in different industries, such as food, paint, pharmaceutical, cosmetic, soap, toothpaste and others. Besides, glycerol also can produce value-added chemicals by a conversion process such as citric acid, lactic acid, hydrogen, ethanol, etc [3]. The simplest and most abundant element is hydrogen, which is growing from time to time due to the technological advancements in fuel cell industry. Nowadays, almost 95% of hydrogen is produced from fossil fuel. Fossil fuels are not renewable resources. So, renewable resources based on the technologies for hydrogen production are attractive options for the future due to the carbon neutral nature of these technologies give a minor effect to the environment. Last few years, a great interest in utilizing glycerol for the hydrogen production [4].

Hydrogen can be produced from glycerol by undergoing some processes such as steam reforming, autothermal reforming, aqueous-phase reforming and supercritical water reforming [4] and they found that the best conditions are at a temperature above 627 °C, 9:1 of the molar ratio of water/glycerol and at atmospheric pressure. While, Adhikari *et al*. [5] found that the nickel loaded on cerium(IV) oxide (Ni/CeO_2) was the best performing catalyst compared to the nickel loaded on magnesium oxide (Ni/MgO) and nickel loaded on titanium dioxide (Ni/TiO_2) in terms of H_2 selectivity and glycerol conversion under the experimental conditions investigated. Ni/CeO_2 gave 74.7 % of H_2 selectivity (maximum H_2 selectivity) compared to the Ni/MgO (38.6%) and Ni/TiO_2 (28.3%) at a water/glycerol molar ratio (WGMR) of 12:1, 600 °C and 0.5 ml/min of feed flow rate.

Besides, Iriondo *et al*. [6] studied that the noble metals were active and stable for steam reforming, but the cost was quite high. For that reason, the non-noble metals such as nickel (Ni) and copper (Cu) were decided to be used as metal catalysts in this study due to availability and lower cost than noble metals [7]. In addition, Nichele *et al*. [7] has investigated that the best result for hydrogen production via glycerol steam reforming was achieved with nickel loaded zirconium dioxide (Ni/ZrO_2) at 650 °C, where the hydrogen yield and glycerol conversion was 65% and 72%, respectively. Another study by Buhari [8] reported that 15wt% of Ni-ZSM-5 has the best performance on hydrogen composition in a product with 58.37% at temperature 600 °C. Therefore, nickel loaded HZSM-5 catalyst was chosen in this study.

The representative of reaction using the application of experimental design and mathematical technique is essential. Response Surface Methodology (RSM) is a mathematical and statistical method that can be used to analyze the problems and optimize the response, in which several independent variables influence a dependent variable (known as a response) [9-11]. Many researchers have successfully applied the optimization of enzyme production from microorganism using RSM [12-14] and also optimizing hydrogen production from methane [15]. However, the Artificial Neural Network (ANN) is more interesting in chemical industries compared to the RSM. ANN is a powerful

modeling technique that offers several advantages compared to the conventional modeling techniques [16-18]. ANN is an accuracy, flexibility and efficiency since it has an attractive feature and also has the ability to learn linear and nonlinear relationships between variables directly from a set of examples [7, 16-19]. The ANN also able to train new data accurately ANN has a universal approximation capability where it can approximate almost non-linear functions including quadratic functions, while RSM only can use for quadratic approximation. In other words, the ANN can be concluded as higher accuracy modelling technique compared to the RSM as it represents a much better way for non-linearities [21].

The objectives of this study are to determine the relationship of catalyst weight (range = 0.3-0.5g) and glycerol flow rate (range = 0.2-0.4mL/min) at temperature 600 °C on 15wt% of the nickel loading and to select the best model from RSM and ANN.

2.0 METHODS

2.1 Catalyst Preparation

10.2 g of HZSM-5 and 8.92 g of nickel(II) nitrate hexahydrate loading were dissolved in 100 ml of distilled water (15.0 wt% Ni, 85.0 wt% ZSM-5). The mixed solution was stirred continuously overnight and dried in an oven for overnight at 105 °C. Then, the calcination process occurs where calcined in the furnace for 5 hours at 500°C. Finally, the sample was put in the vial and stored in the desiccator.

2.2 Reactivity Test

The performance of the catalyst for glycerol conversion was tested using the quartz tube in a fixed bed reactor as shown in Figure 1. The modified catalyst was placed in the core of the reactor that supported by quartz wool. The modified catalyst was activated by packing between glass wool inside the reactor at 600 °C of study temperature. Nitrogen gas was emerged in the reactor for about 10-50 ml/min. WGMR was introduced in the process at the range of study flow rate (0.2-0.4ml/min). Then, the heating process of glycerol was carried out in the preheater at a temperature about 250 °C. The reactor was activated for 30 minutes at 600 °C of study temperature. The production flows through the condenser where the liquid product was collected while gas products continuously flow into a silica bed to trap any moisture in the product. The production of gaseous was analyzed by GC (Agilent Technologies, 6890 System) with thermal conductivity detector (TCD) and being illustrated using computer.

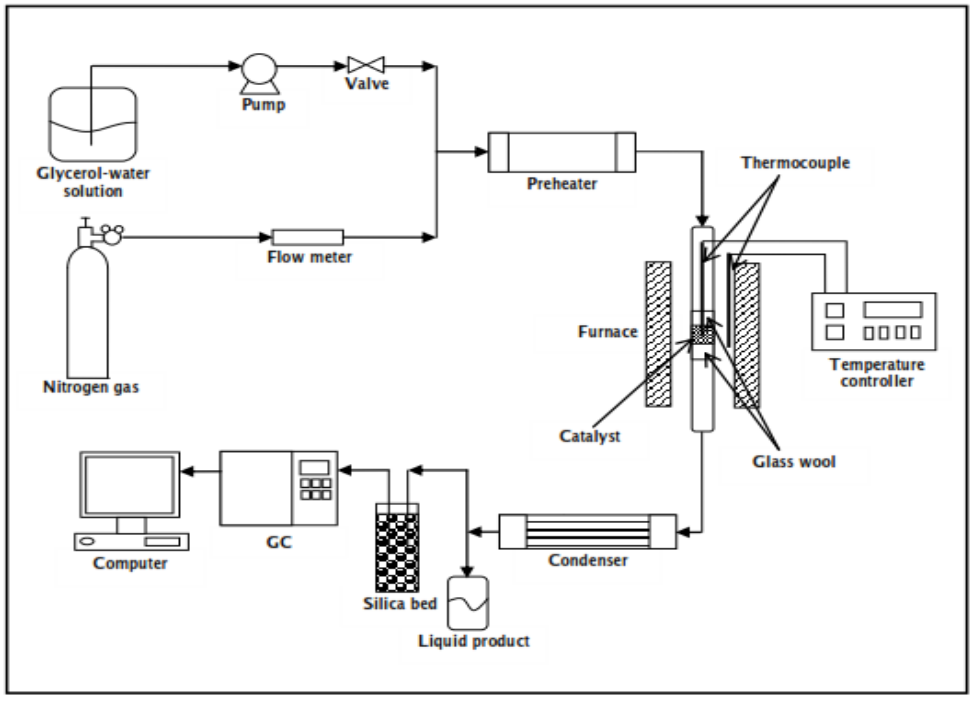


Figure 1 Schematic layout of the rig of the glycerol conversion

2.3 Experimental Design

The experimental conditions were defined using central composite design (CCD) techniques in RSM that developed by Statsoft Statistica Release 7.0 software. This software was chosen due to the friendly user. The central composite design (CCD) was used to predict the condition of optimum process for hydrogen yield and to study the interaction of process variables. The range and the coded level of the independent

process variables studied are listed in Table 1. The process variables studied were catalyst weight (x_1) and glycerol flow rate (x_2). Each variable consisted of three different levels from low (-1) to medium (0) and to high (1).

Table 1 Experimental range and level coded of independent variables

Factors	Symbol	Range and levels		
		-1	0	+1
Catalyst weight, g	X_1	0.3	0.4	0.5
Glycerol flow rate, ml/min	X_2	0.2	0.3	0.4

The central composite design (CCD) that used in this research was 22 factorial designs, two central points and four star points. The total number of the experiment was 10 where 8 experiments with two replications at the central point to identify the errors. The method of least squares was employed to establish the full quadratic model for glycerol conversion and hydrogen yield. RSM model was used to determine the relationship of the glycerol flow rate and catalyst weight and also to optimize the reaction conditions.

$$Yu = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_{12}X_1X_2 + \beta_{11}X_1^2 + \beta_{22}X_2^2$$

where, β_0 is the intercept coefficient (offset), β_1 and β_2 the linear terms, β_{11} and β_{22} the quadratic terms, β_{12} the interaction terms and X_1 and X_2 the encoded independent variables. The statistical analysis of this model was performed in the form of analysis of variance (ANOVA) in order to test for significant differences between means. The conversion of glycerol was the percentage of glycerol convert to the product and it was defined by the following expansion:

$$Conversion = \frac{Amount\ of\ initial\ WGMR - amount\ of\ WGMR\ after\ reaction}{Amount\ of\ initial\ WGMR}\ x\ 100\%$$

The hydrogen yield is calculated based on the following equation:

$$Yield = \frac{Mole\ of\ hydrogen\ produced}{Mole\ of\ glycerol\ reacted} \times 100\%$$

In chemical engineering application, most commonly used of ANN is the multi-layer perceptron, which is a back propagation feed forward neural network due to the fast computation capacity and generalization ability of data even chemical engineering problems composed by complex systems [22]. In this study, multi-layer feed forward network generated by JMP version 11 for simulation because the application of ANN in JMP 11 is flexible and easy to use [18] compared to the Statsoft Statistica Release 7.0 software. The same experimental data that had been used in RSM design were applied in the ANN design. Catalyst weight and glycerol flow rate as an independent variable (input variables) while, hydrogen yield and glycerol conversion were used as dependent variables (output variables). The training of the network was performed using Levenberg Marquardt Learning Algorithm that can provide a numerical solution to the problem of minimizing a function. The best performance of the models was described based on the correlation of determination (R^2) and mean square error (MSE). The R^2 is mathematically described as based on Ahmad and Daniel [23].

$$R^2 = 1 - \frac{\sum_{i=1}^{1-n}[(y_i - \bar{y}_i)\hat{}2]}{\sum_{i=1}^{1-n}[(y_i - \bar{y})\hat{}2]}$$

where yi is the actual output value from observation, ŷi is the output value predicted from observation i, y is the mean of y values, and n is the total number of data. A perfect fit would result in R^2 should be at least 0.80 for the best fit of a model [24]. The MSE is calculated based on Demuth *et al*. [25] by following equation:

$$MSE = \frac{1}{Q}\sum_{k=1}^{Q}(t_k - a_k)^2$$

Where, Q is the number of data, t is the output target and a is the output network. Figure 2 showed the feed forward hierarchical neural network architecture for the prediction of hydrogen yield and glycerol conversion.

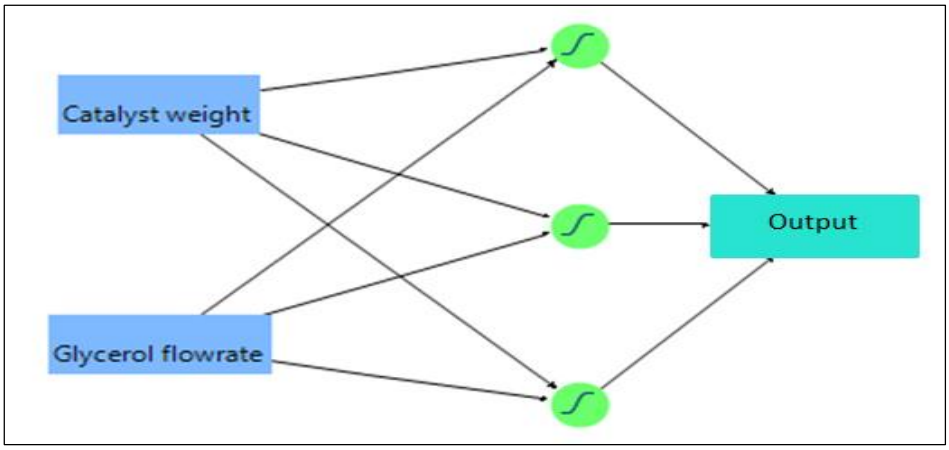


Figure 2 Architecture of feed forward hierarchical neural network

3.0 RESULTS AND DISCUSSION

3.1 Polynomial Empirical Model

In this study, the RSM models were developed using Statsoft Statistica Release 7.0 software. The experimental value and predicted response of hydrogen yield from statistical model for 10 runs were presented in Table 2. The relationship between the independent variables and response variables was estimated using regression analysis of the experimental data. Equation 3.1 presented a quadratic model for predicting the optimal point for the hydrogen yield.

$$Y_1 = 94.6077 + 11.0625X_1 + 22.1120X_2 - 28.7500X_1X_2 - 3.3750X_1^2 - 18.6250X_2^2 \quad \text{-------(3.1)}$$

The regression equation (Eq. 3.2) obtained for the glycerol conversion.

$$Y_2 = -183.35 + 418.73X_1 + 987.34X_2 - 1334.74X_1X_2 + 66.49X_1^2 - 581.21X_2^2 \quad \text{--------(3.2)}$$

Where, Y_1 and Y_2 are the response variables corresponding to the hydrogen yield and glycerol conversion respectively, while X_1 and X_2 represent the catalyst weight and the glycerol flow rate respectively, as independent variables.

3.1.1 Analysis of Variance (ANOVA) Responses

The analysis of variance (ANOVA) with a 5 % level of significance was used to test the empirical model for hydrogen yield and glycerol conversion as well as demonstrated the total, error and regression of sum of squares. The total, error and regression of sum of squares for hydrogen yield and glycerol conversion as shown in Table 3.

Table 3 Analysis of variance (ANOVA) for quadratic model

Sources	Sum of squares (SS)	Degree of freedom (d.f)	Mean squares (MS)	F-value	$F_{0.05}$
Hydrogen yield model					
Regression (SSR)	2209.119	5	441.824	6.282	6.26
Residual	281.322	4	70.331		
Total (SST)	2490.441	9			
Glycerol conversion model					
Regression (SSR)	0.532	5	0.106	10.113	6.26
Residual	0.042	4	0.011		
Total (SST)	0.574	9			

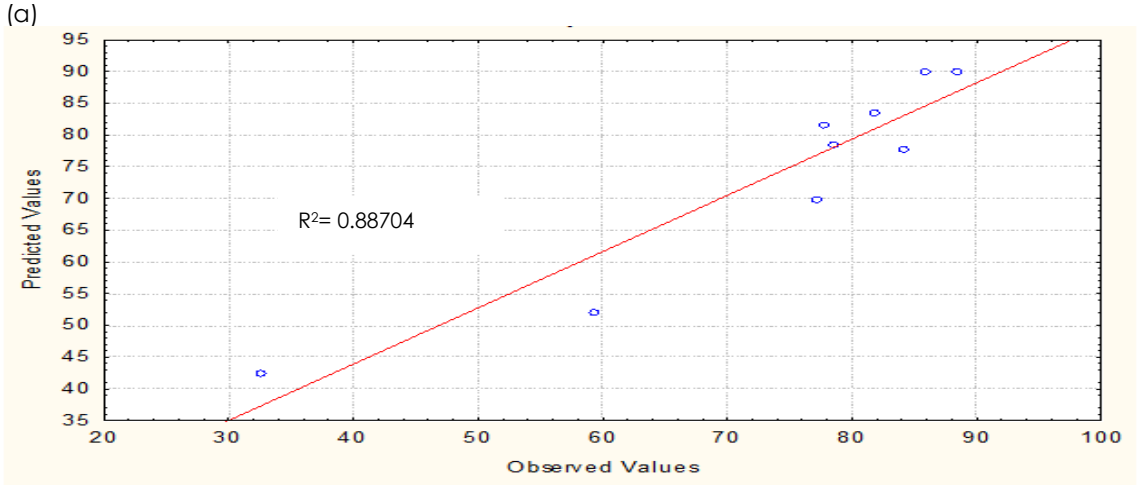
Table 2 Experimental value and predicted response of hydrogen yield and glycerol conversion

Run	Variables		Hydrogen yield, Y_1		Glycerol conversion, Y_2	
	Catalyst weight, X_1 (g)	Glycerol flow rate, X_2 (ml/min)	Experimental	Predicted	Experimental	Predicted
1	0.258579	0.300000	77.08704	69.72072	100.00	99.9696
2	0.500000	0.200000	81.79643	83.38060	100.00	100.0976
3	0.541421	0.300000	85.84106	89.94400	100.00	99.8954
4	0.400000	0.300000	78.50265	78.50265	100.00	100.0000
5	0.400000	0.300000	78.50265	78.50265	100.00	100.0000
6	0.400000	0.441421	77.65270	81.68843	99.61	99.5479
7	0.500000	0.400000	84.06105	77.63027	99.32	99.4099
8	0.300000	0.400000	88.34577	90.02499	100.00	100.0374
9	0.400000	0.158579	59.36768	52.06857	99.78	99.7071
10	0.300000	0.200000	32.69169	42.38585	99.53	99.5751

From Table 3, the F-value indicates a ratio between the mean square of regression and mean square of error. Generally, the model is the best predictor of the experimental results with high confidence level of 95 % if the calculated F value greater than the tabulated F value. Therefore, the statistical model of the hydrogen yield in this research was the F value of 6.282, greater than $F_{0.05}$ = 6.26. While, for the glycerol conversion, the calculated F value was 10.113, greater than $F_{0.05}$ = 6.26. Both of the statistical models of hydrogen yield and glycerol conversion showed a good prediction model.

Besides, the comparison between the observed value and predicted value of hydrogen yield and glycerol conversion were shown in the Figure 3. The result indicates that the R^2 value for hydrogen yield was 0.88704 (88.70 %) of the variation. Meanwhile, the R^2 value of the glycerol conversion was 0.92670 which 92.67 %, indicating of the variation of data can be accounted to the model. The empirical model should be at least 0.75 to adequately explain most of the variability in the assay reading [26].

(a)



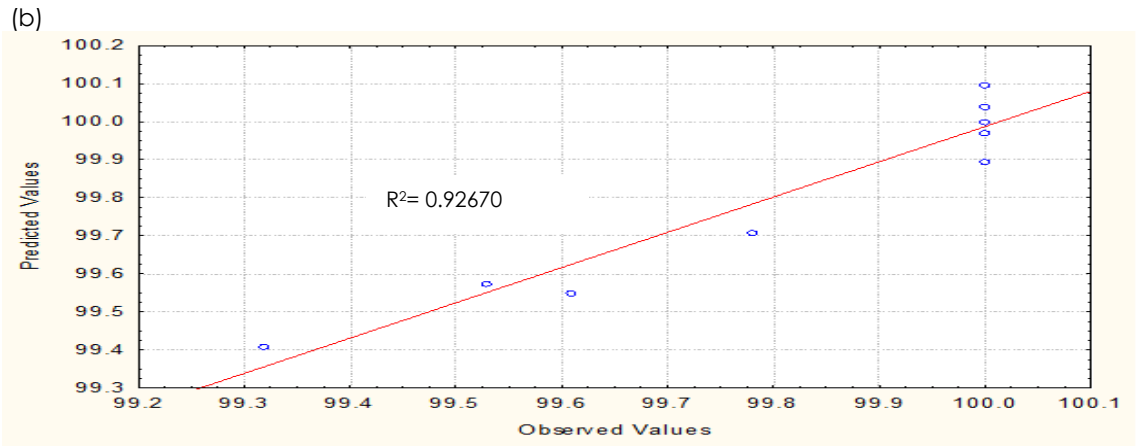


Figure 3 Parity plot for a) hydrogen yield and b) conversion

3.1.2 Interactive Effects of Variables on Hydrogen Yield and Glycerol Conversion

The interaction between variables in this study that gave impact to the hydrogen yield and glycerol flow rate was illustrated by using an empirical model which is plotted as three dimensional surfaces. Figure 4 depicts the effect of glycerol flow rate and catalyst weight interaction in the presence of 15 wt% of nickel loading and 600 °C on hydrogen yield and glycerol conversion, respectively. Based on the contour plot, the interaction between glycerol flow rate and catalyst weight gives the biggest impact to the hydrogen yield and glycerol conversion.

Previous researcher, Yun [27] found that the hydrogen yield was increased at 0.25 ml/min to 0.50 ml/min but then, the value of hydrogen yield starting to decrease with the increasing of feed flow rate. It had been proven by Slinn *et al*. [28], which stated that the conditions for water gas-shift, a reactant molecule needs to be met at available active site. It was contact with a water molecule for a certain period of time and the enough energy to overcome the activation energy barrier. Hence, the faster flow rates would decrease the hydrogen yield because of the conditions for water-gas-shift was not being met. However, if the flow rate was too slow, it could also reduce the hydrogen yield due to the increasing of by-products.

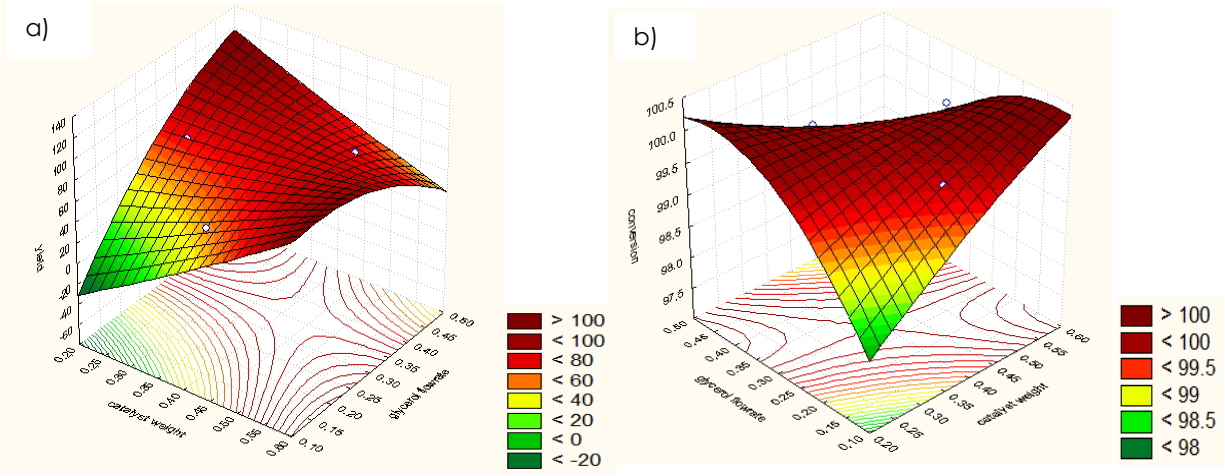


Figure 4 Response surface plot of the combination between glycerol flow rate and catalyst weight on a) hydrogen yield and b) glycerol conversion

3.2 Feed Forward Neural Network Model

In this study, two neural network models were developed for predicting the hydrogen yield and glycerol conversion. All the ANN models were trained by using Levenberg Marquardt Learning Algorithm. Table 4 shows the experimental and predicted values for all conditions of the reaction.

In overall, the percentages of hydrogen yield obtained were high as well as glycerol conversion based on Table Almost all of the experiment shows 100% conversion of glycerol obtained. Table 5 summarizes the statistical parameters of selected ANNs: Number of hidden nodes, R^2, MSE and sum frequency.

Table 4 Experimental value and predicted response of hydrogen yield and glycerol conversion

Run	Variables		Hydrogen yield, Y_1		Glycerol conversion, Y_2	
	Catalyst weight, X_1 (g)	Glycerol flow rate, X_2 (ml/min)	Experimental	Predicted	Experimental	Predicted
1	0.258579	0.300000	77.087040	77.610693	100.00	99.78
2	0.500000	0.200000	81.796430	83.172634	100.00	100.00
3	0.541421	0.300000	85.841060	85.808672	100.00	100.00
4	0.400000	0.300000	78.502654	78.417956	100.00	100.00
5	0.400000	0.300000	78.502654	78.417956	100.00	100.00
6	0.400000	0.441421	77.652700	77.610659	99.61	100.00
7	0.500000	0.400000	84.06105	83.963236	99.32	99.32
8	0.300000	0.400000	88.345770	84.030456	100.00	100.00
9	0.400000	0.158579	59.367680	56.382686	99.78	99.78
10	0.300000	0.200000	32.691690	34.586376	99.53	99.61

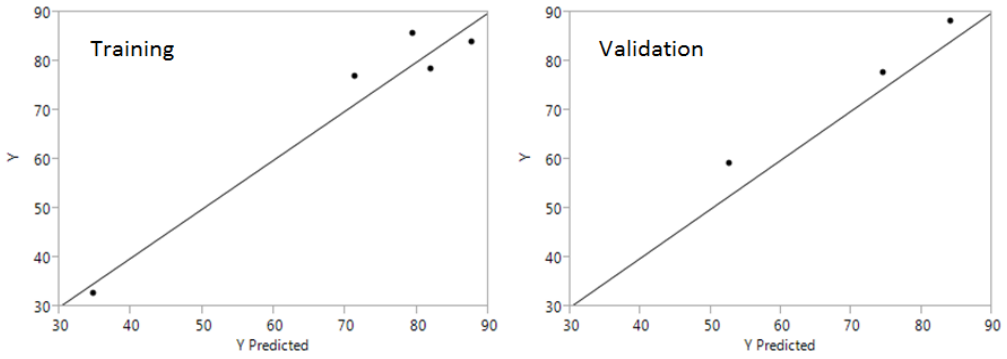
Table 5 The parameters of selected ANNs

	$ANNH_2$		ANNG	
	Training	Validation	Training	Validation
Number of hidden nodes	3	3	3	3
R^2	0.94215	0.59076	0.99935	0.53991
MSE	4.37916	6.88171	0.00638	0.14709
Sum Freq	6	4	6	4

*$ANN-H_2$: ANN for hydrogen yield
*ANNG: ANN for glycerol conversion

Selection of the network for prediction of hydrogen yield and glycerol conversion was carried out with a trial and error method on the optimum number of nodes and the R^2 and MSE obtained. Two variables were used as input (catalyst weight and glycerol flow rate) into the ANN network with three hidden nodes. The best network model was chosen based on the highest R^2 and lowest MSE [16-18, 29].

In this study, the highest R^2 and MSE values in the training phase for $ANN-H_2$ were 0.94215 and 4.37916, respectively. For the validation phase, the R^2 and MSE values were 0.59076 and 6.88171, respectively. The highest R^2 and MSE values for ANN-G in the training phase, were 0.99935 and 0.00638, respectively. For the validation phase, the highest R^2 and MSE values were 0.53991 and 0.14709, respectively. Figure 5 shows the plot diagram for training and validation phase for hydrogen yield and glycerol conversion.



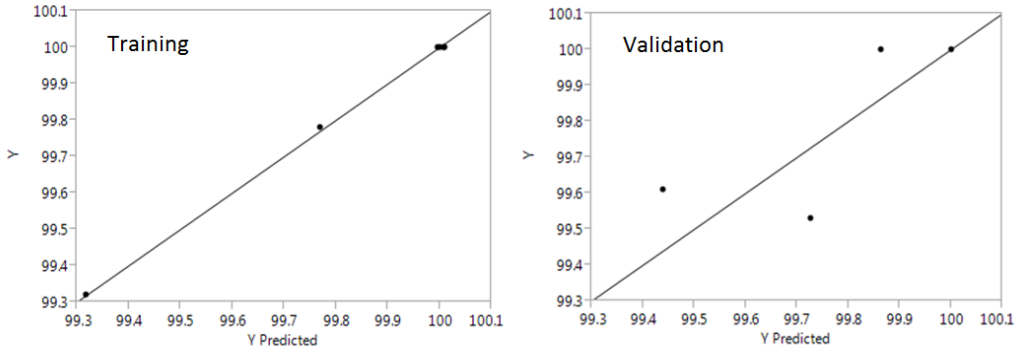


Figure 5 Training and validation phase for a) hydrogen yield and b) glycerol conversion

3.3 Comparison of RSM and ANN Models

The estimation of the capability of the RSM and ANN techniques was also evaluated in this study in order to predict the responses at 10 experiment points. The comparative values of R^2 and MSE was given in Table 6.

Table 6 Comparison between RSM and ANN

Statistical parameters	Hydrogen yield		Glycerol conversion	
	RSM	ANN	RSM	ANN
R^2	0.88704	0.88987	0.92670	0.93091
MSE	70.331	8.28052	0.011	0.09958

Based on Table 6, the R^2 values of hydrogen yield for RSM and ANN were 0.88704 and 0.88987, respectively, and the MSE values of hydrogen yield for RSM and ANN were 70.331 and 8.28052, respectively. For glycerol conversion, the R^2 values for RSM and ANN were 0.92670 and 0.93091, respectively, and the MSE values for RSM and ANN were 0.011 and 0.09958, respectively.

By comparing the R^2 and MSE values, the ANN technique is more precise compared to the RSM even with the fewer number of the experiment. Geyikci *et al.* [30] stated that the ANN technique may require a total number of data compared to the RSM technique. However, despite the ANN model developed using a fewer number of data, but it still gives a good correlation between observed and predicted value for training value [21, 31, 32].

4.0 CONCLUSIONS

This study compares the performance of RSM and ANN technique with their modelling using experimental data in the hydrogen gas production using nickel loaded zeolite. Both models provide reliable result. However, the ANN gives high accuracy compared to the RSM even with a limited number of the data

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References

[1] Gerpen, J. V. 2005. Biodiesel Processing and Production. *Fuel Processing Technology* 86: 1097-1107.
[2] Shawn, P. and Conley, B. T. 2006. What is Biodiesel? *BioEnergy*. ID337.
[3] Xiaohu, F. R. B. a. Y. Z. 2010. Glycerol (Byproduct of Biodiesel Production) as a Source of Fuels and Chemicals-Mini Review. *The Open Fuels and Energy Science Journal*. 3: 17-22.
[4] Adhikari, S., Fernando, S. D. and Agus Haryanto. 2009. Hydrogen Production from Glycerol: An update. *Energy Conversion and Management.* 50: 2600-2604.
[5] Adhikari, S., Fernando, S., To, S. D. F., Bricka, R. M., Steele, P. H. and Haryanto, A. 2008. Conversion of Glycerol to Hydrogen via a Steam Reforming Process over Nickel Catalysts. *Energy Fuel.* 22(2): 1220-1226.
[6] Iriondo, A., Barrio, V. L., Cambra, J. F., Arias, P. L., Guemez, M. B., Navarro, R. M., Sanchez-Sanchez, M. C. and Fierro, J. L. G. 2009. Influence of La2O3 Modified Support and Ni and Pt Active Phases on Glycerol Steam Reforming to Produce Hydrogen. *Catalysis Communications*. 10: 1275-1278.
[7] Nichele, V., Signoretto, M., Menegazzo, F., Gallo, A., Santo, V. D., Cruciani, G. and Cerrato, G. 2012. Glycerol Steam Reforming for Hydrogen Production: Design of Ni Supported Catalysts. *Applied Catalysis B: Environment*. 111: 225-232.
[8] Buhari, J. 2013. Hydrogen Production from Glycerol using Nickel Loaded Zeolite Catalyst. Degree Thesis. Universiti Teknologi Malaysia.
[9] Myers, R. H. and Montgomery, D. H. 2002. *Response Surface Methodology: Process and Product Optimization Using Designed Experiment*. A Wiley-Inter-science Publication.
[10] Maran, J. P., Sivakumar, V., Sridhar, R. and Prince Immanuel, V. 2013. Development of Model for Mechanical Properties of Tapioca Starch Based Edible films. *Ind. Crop. Prod*. 42: 159-168.
[11] Maran, J. P., Sivakumar, V., Sridhar, R. and Thirgananasambandham, K. 2013. Development of Model for Barrier and Optical Properties of Tapioca Starch Based Edible films. *Carbohyd. Polym*. 92: 1335-1347.
[12] Banerjee, R. and Bhattacharyya, B. C. 1992. Optimization of Multiple Inducers Effect on Protease Biosynthesis by Rhizopus oryzae. *Bioprocess Eng.* 7: 225-228.

[13] De Coninck, J., Bouquelet, S., Dumortier, V., Duyme, F. and Denantes, V. 2000. Industrial Media and Fermentation Process for Improved Growth and Protease Production by Tetrahymena Thermophile BIII. *J Ind Microbiol Biotechnol*. 24: 285-290.
[14] Murthy, M. S. R. C., Swaminathan, T., Rakshit and Kosugi, Y. 2000. Statistical Optimization of Lipase Catalyzed Hydrolysis of Methyloleate by Response Surface Methodology. *Bioprocess Eng*. 22: 35-39.
[15] Kusworo, T. D., Songip, A. R. and Amin, N. A. S. 2010. Optimization of Partial Oxidation of Methane for Hydrogen Production on NiO-CoO/MgO Catalyst using Design of Experiment. *IJET-IJENS*. 10(1): 1-8.
[16] Azid, A., Juahir, H., Latif, M. T., Zain, S. M. and Osman, M. R. 2013. Feed-Forward Artificial Neural Network Model for Air Pollutant Index Prediction in the Southern Region of Peninsular Malaysia. *J. Environ. Prot.* 4: 1-10. doi: 10.4236/jep.2013.412A001.
[17] Mutalib, S. N. S. A., Juahir, H., Azid, A., Sharif, S. M., Latif, M. T., Aris, A. Z., Zain, S. M. and Dominick, D. 2013. Spatial and Temporal Air Quality Pattern Recognition Using Environmetric Techniques: A Case Study in Malaysia. *Environ. Sci. Processes Impacts*. 15: 1717-1728. doi: 10.1039/c3em00161j.
[18] Azid, A., Juahir, H., Toriman, M. E., Kamarudin, M. K. A., Saudi, A. S. M., Hasnam, C. N. C., Aziz, N. A. A., Azaman, F., Latif, M. T., Zainuddin, S. F. M., Osman, M. R. and Yamin, M. 2014. Prediction of the Level of Air Pollution Using Principal Component Analysis and Artificial Neural Network Techniques: A Case Study in Malaysia. *Water Air Soil Pollut.* 225: 2063. doi: 10.1007/s11270-014-2063-1.
[19] Serra, J. M., Corma, A., Argente, E., Valero, S. and Botti, V. 2003. Neural Network for Modelling of Kinetic Reaction Data Applicable to Catalyst Scale Up and Process Control and Optimisation in a Frame of Combinatorial Catalysis. *Applied Catalysis A: General*. 254: 133-145.
[20] Nagata, Y. and Chu, K.H. 2003. Optimization of a Fermentation Medium Using Neural Networks and Genetic Algorithms. *Biotechnol. Lett*. 25: 183-1842.
[21] Desai, K. M., Survase, S. A., Saudagar, P. S., Lele, S. S., and Singhal, R. S. 2008. Comparison of Artificial Neural Network (ANN) and Response Surface Methodology (RSM) in Fermentation Media Optimization: Case study of Fermentative Production of scleroglucan. *Biochemical Engineering Journal*. 41: 266-273. doi:10.1016/j.bej.2008.05.009.
[22] Singh, A., Tatewar, D., Shastri, P. N., and Pandharipande, S. L. 2008. Application of ANN for Prediction of Cellulase and Xylanase Production by Trichoderma Reesei Under SSF Condition. *Indian Journal of Chemical Technology*. 15: 53-58.
[23] Ahmed, G. E.-D. and Daniel, W. S. 2001. A Neural Network Model to Predict the Wastewater Inflow Incorporating Rainfall Events. *Water Research*. 36: 1115-1126.
[24] Guan, X. and Yao, H. 2008. Optimization of Viscozyme L-Assisted Extraction of Oat Bran Protein Using Response Surface Methodology. *Food Chemistry*. 106: 345-351.
[25] Demuth, H., Beale, M. and Hagan, M. 2009. *Neural Network Toolbox™ 6 User's Guide*. The MathWorks Inc.
[26] Haaland, P. D. 1989. *Experimental Design in Biotechnology*. New York: Marcel Dekker Inc.
[27] Yun, H. B. A. H. 2013. Hydrogen Production from Glycerol Using Copper and Nickel Loaded Zeolite Based Catalyst. Master Thesis. Universiti Teknologi Malaysia.
[28] Slinn, M., Kendall, K., Mallon, C. and Andrews, J. 2008. Steam Reforming of Biodiesel By-Product to make Renewable Hydrogen. *Bioresource Technology*. 99: 5851-5858.
[29] Daliakopoulos, I. N., Coulibaly, P. and Tsanis, I. K. 2005. Groundwater Level Forecasting Using Artificial Neural Networks. *Journal of Hydrology*. 309: 229-240.
[30] Geyikci, F., Kılıc, E., Coruh, S. and Elevli, S. 2012. Modelling of Lead Adsorption from Industrial Sludge Leachate on Red Mud by Using RSM and ANN. *Chemical Engineering Journal*. 183: 53-59.
[31] Saidina Amin, N. A., Mohd Yusof, K., and Isha, R. 2005. Carbon Dioxide Reforming of Methane to Syngas: Modeling Using Response Surface Methodology and Artificial Neural Network. *Jurnal Teknologi*. 43: 15-30.
[32] Mourabet, M., Rhilassi, A. El, Bennani-Ziatni, M., and Taitai, A. 2014. Comparative Study of Artificial Neural Network and Response Surface Methodology for Modelling and Optimization the Adsorption Capacity of Fluoride onto Apatitic Tricalcium Phosphate. *Universal Journal of Applied Mathematics*. 2(2): 84-91. doi:10.13189/ujam.2014.020202.