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DAFTAR ISI:

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04 Feb. 19

Dear Dr. Asep Bayu Nandiyanto
[Chief Editor IJoST]

Greeting,

I wish to submit an original research article entitled "*Application of Artificial Neural Network to Predict Bio-diesel Yield from Waste Frying Oil Transesterification*" for consideration by **Indonesian Journal of Science & Technology**.
I confirm that this work is original and has not been published elsewhere, nor is it currently under consideration for publication elsewhere.

In this paper, I report that artificial neural network (ANN) is a powerful method to predict biodiesel yield from a transesterification of waste frying oil. This is significant because the prediction using ANN model is better than that of kinetic approach.
We believe that this manuscript is appropriate for publication by **Indonesian Journal of Science & Technology** because it complies with journal's Focus & Scope especially for especially in the field of Chemistry and Chemical Engineering and also Environment Science and Engineering.

Biodiesel has become one of promissing energy source in the future. Reaction yield is main target in every biodiesel industry and the prediction of it is important. Our finding reveals that ANN model can be efficiently explored to predict biodiesel yield. Yield prediction using ANN models with activation function of logsig-purelin-logsig and purelin-logsig-tansig is excellent with *RRMSE* (Relative Root Mean Squared Error) of 2.41% and 2.44%, respectively, and R^2 (determination coefficient) of 0.9355 and 0.9391%, respectively.

We have no conflicts of interest to disclose.

Please address all correspondence concerning this manuscript to me at agus.haryanto@fp.unila.ac.id.

Thank you for your consideration of this manuscript.

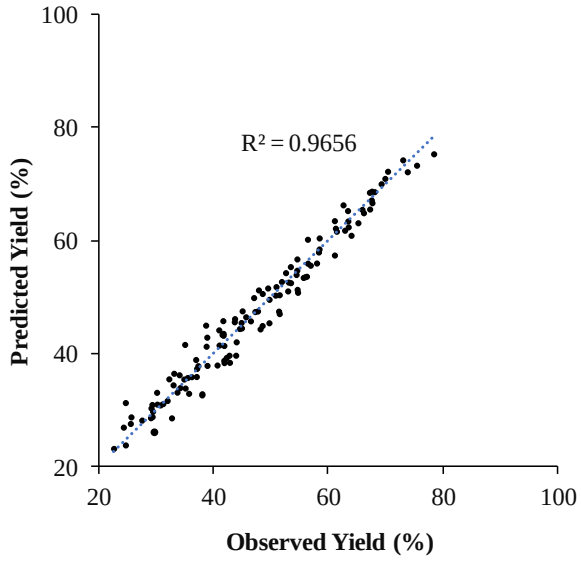
Sincerely,

[Agus Haryanto]

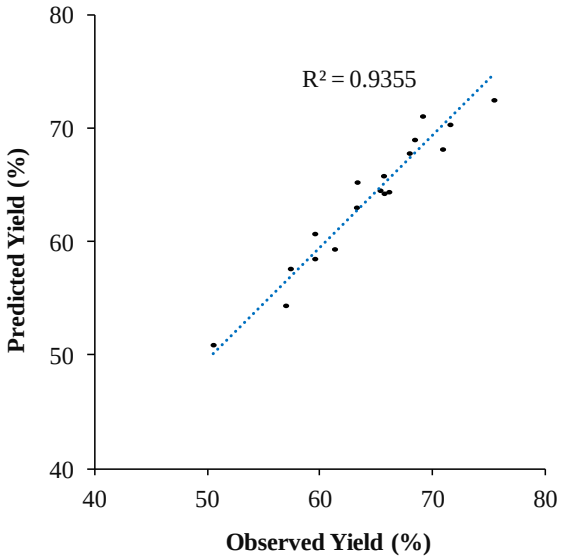
HIGHLIGHT

- ANN model provides a powerful method to predict biodiesel yield produced from transesterification of waste frying oil.
- Twenty seven ANN models from combinations of tansig, logsig, purelin activation function were trained using 116 data set of biodiesel yield observed at three different molar ratios, six different temperatures and seven points reaction time.
- ANN models with activation function of logsig-purelin-logsig and purelin-logsig-tansig be the best with *RRMSE* of 2.41% and 2.44%, respectively, and R^2 of 0.9355 and 0.9391%, respectively.

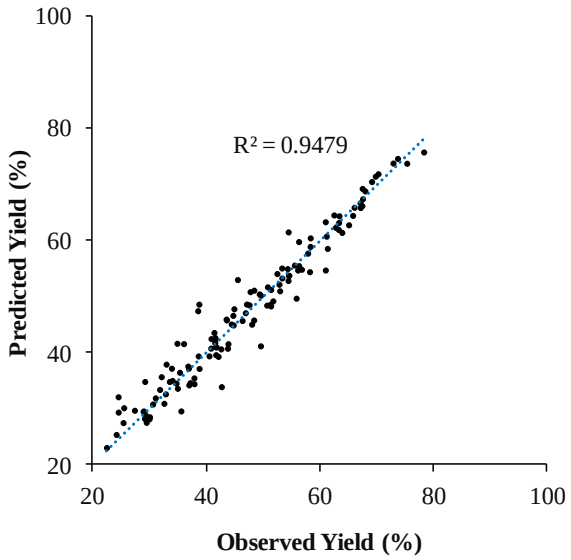
GRAPHICAL ABSTRACT



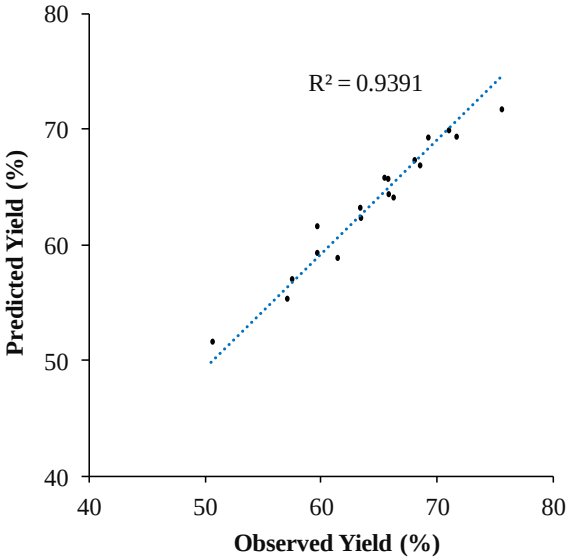
logsig-purelin-logsig (training)



logsig-purelin-logsig (validation)



purelin-logsig-tansig (training)



purelin-logsig-tansig (validation)

Indonesian Journal of Science & Technology

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Application of Artificial Neural Network to Predict Bio-diesel Yield from Waste Frying Oil Transesterification

Authors

ABSTRACTS

Used frying oil (UFO) has great potential as feedstock for biodiesel production. This study aims to develop an artificial neural network (ANN) model to predict biodiesel yield produced from base-catalyzed transesterification reaction of UFO. The experiment was performed with 100 ml of UFO at three different molar ratios (oil:methanol), namely 1:4, 1:5, 1:6, reaction temperatures of 30 to 55 °C (raised by 5 °C) and reaction time of 0.25, 0.5, 1, 2, 3, 6, and 10 min. Prediction model was based on ANN back propagation type with supervised learning method. Validation model was carried out using data set measured at reaction duration of 8 min. The results showed that network architecture of the ANN model consists of four layers, namely input layer, first hidden layer, second hidden layer, and output layer. The training used in this work is the Levenberg-Marquardt train type with learning rate of 0.001 and activation function of purelin-logsig-purelin. The model validation revealed that predicted yield is excellently accurate with %RMSE value of 1.51% and the coefficient of determination (R^2) of 0.946. Prediction values resulted from the ANN model were better than those of first-order kinetics.

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yield.

1. INTRODUCTION

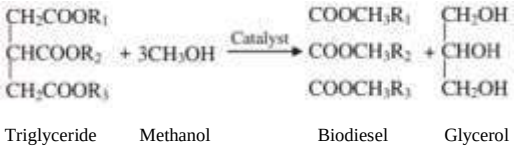
Biodiesel is an alternative energy source for diesel fuel offering several advantages. First, biodiesel is made from vegetable oils or animal fats so it is classified as a renewable, biodegradable, and nontoxic energy source (Khan *et al*., 2013). Biodiesel, therefore, is ecologically friendly because the production and application of biodiesel results in lower greenhouse gas (GHG) emissions than petroleum fuels do. Studies in the United States (Sheehan *et al*., 1998) revealed that based on life cycle analysis biodiesel is able to decrease GHG emission 78% as compared to petroleum diesel fuel. Other works using varous feedstock in different countries also reported that

biodiesel producing lower GHG such as in India with jatropha curcas (Kumar *et al*., 2012; Achten *et al*., 2010), China with various oil (Hou *et al*., 2011; Guo *et al*., 2010), Southeast Asia (Indonesia, Malaysia, Thailand) with palm oil (Siregar *et al*., 2015; Harsono *et al*., 2012; Hassan *et al*., 2011; Silalertruksa & Gheewala, 2012), Brazilia with soybean (Oliveira *et al*., 2017), and Europe with rapeseed oil (Malça & Freire, 2011). Second, unlike fossil fuels which are bestowed to a few countries, oil-plants producing biodiesel feedstock are spread throughout the world, so that geopolitically biodiesel can be one that can increase energy security (Paltsev, 2016).

Utilization of vegetable oils as feedstock for biodiesel production, however, is more expensive because the cost of raw materials can reach 80 to 85% of operational costs (Canakci & Sanli, 2008; Hindryawati *et al*., 2014). One potential cheaper raw material is used frying oil (UFO), which is not alowable to be dumped directly because it has a high COD value. In addition, UFO contains toxic compound formed during high temperature heating like hydroperoxides and aldehydes. When these compounds ingested through consumed food they may be responsible for increasing blood pressure (hypertension) and attributable to cardiovascular diseases and diabetes (Leong *et al*., 2015; Jaarin *et al*., 2016). Repeatedly heated cooking oils even produce carcinogenic compounds such as polycyclic aromatic hydrocarbons that relate to the incidence of tumor and cancer diseases (Ganesan *et al*., 2017). Therefore, using UFO repeatedly to fry food or to make food-related ingredients, such as chili sauce, may endanger human health.

The potential of used cooking oil in Indonesia is quite large because the consumption of cooking oil shows a tendency to increase from 0.198 L/capita per week in 2007 to 0.205 in 2012 and 0.221 in 2017 (BPS, 2018). This figure is very close with the study in Bogor (Fujita *et al*., 2013), which found cooking oil consumption of one family is 3L/month or 36 L/year. With a proper management, UFO has great potential to be used as energy sources in the form of biodiesel to replace diesel oil (Chhetri *et al*., 2008). The development of biodiesel from UFO will provide a healthier choice of using UFO.

The reaction usually chosen to produce biodiesel from oil is transesterification with methanol and a base catalyst (Fajardo *et al*., 2011). For every mol of triglyceride or vegetable oil, the process required stoichiometrically three mols of methanol to produce three mols biodiesel or FAME (fatty acid methyl ester) and one mol glycerol, as presented in Equation (1).



Triglyceride Methanol Biodiesel Glycerol (1)

Important factors in biodiesel synthesis, among other, include molar ratio (MR) of oil to methanol, reaction temperature (*T*), and reaction duration (*t*). Transesterification is a reversible and equilibrium reaction so that to obtain maximum yield should be carried out with excess of methanol (Ejikeme *et al*., 2010), rather than 1:3 as in Equation 1. The disadvantage of this reaction is that there may be a side reaction in the form of safonification which will consume the catalyst so that reducing biodiesel yield, especially if the FFA content in UFO is quite high (Narasimharao *et al*., 2007). The relation of biodiesel yield and dependent variables is unlinear and so complex that require a robust model to accurately predict the yield. Conventional approaches such as kinetics analysis can be applied successfully in certain constrained environments, but it is not flexible in different conditions. The complexity of these relationships can be

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analyzed by using ANN (artificial neural network) models so as to produce high accuracy biodiesel yield prediction (Lavalle *et al*., 2012; Seo, 2013).

ANN model has been an important tool to solve many problems in a wide range of areas such as pattern recognition, function approximation, categorization, prediction, optimization, associative memory, and control (Jain *et al*., 1996). Recently, ANN model is explored in biodiesel production yield from the transesterification reaction (Yuste & Dorado, 2006). The ANN model has been used, for instance, to predict biodiesel viscosity from vegetable oils and animal fats (Jahirul *et al*., 2014).

The general objective of this study is to predict biodiesel yield resulted from UFO transesterification reaction which is broke down into three specific objectives. First, examine relationship between molar ratio, reaction temperature and reaction time as independent variables on biodiesel yield as the dependent variable. Second, develope ANN models for UFO transesterification reaction. Third, validate ANN models that have been made to obtain the most accurate one to predict biodiesel yield.

2. MATERIALS AND METHODS

2.1. Biodiesel synthesis

Biodiesel was sinthesized using UFO collected from fried food pedlars in the vicinity of the University of Lampung. Important variables including molar ratio, reaction temperature, and reaction time were varied to obtain biodiesel yield. Detil of materials and processing method have been recently reported elsewhere (Authors, 2019).

2.2. ANN model development

Figure 1 showed step-by-step ANN model development to validation. The ANN model to be developed is back-propagation type with supervised learning method.

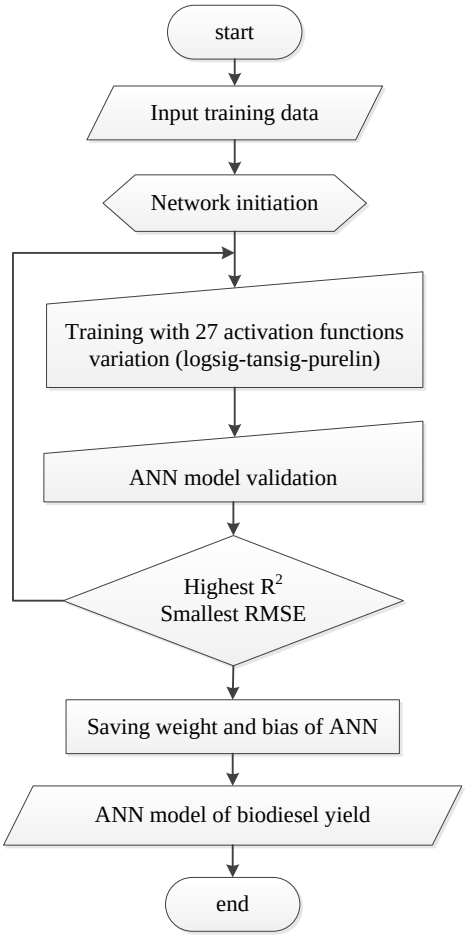

Figure 1. Flowchart fot ANN model development and validation

Figure 2 presented form of ANN model consisting of three layers for biodiesel yield prediction based on molar ratio, reaction temperature and reaction time. The model consits of three layers, namely input layer, hidden layer, and output layer. Due to complexity of involved variables, two hidden layers are used, each with five neurons or nodes. Karsoliya (2012) guided that neuron number of hidden layer is less than twice of the number of neurons in input layer.

The ANN model works in three steps, namely feed forward, back-propagation, and weight adjustments which are calculated based on the established equations (Widodo *et al*., 2013). At the first step, each input node receives an input value, x_i (i = 1, 2, 3, ..., n) and forwards the signal to all nodes in

the hidden layer. Each hidden layer node will add all the weighted input signals (z_in_j), which is x_i multiplied by the weight (v_{ij}) and added by received bias, b_j (j = 1,2,3, ..., p) as in Equation 2. By using activation function as in Equation 3, the signal coming out from the hidden layer node is then calculated.

$$z\_in_j = b_j + \sum_{i=1}^{n} x_i v_{ij} \quad (2)$$

$$z_j = f(z\_in_j) \quad (3)$$

In this work, three activation functions (logsig, tansig, and purelin) were selected that making 27 combinations need to be validated. These three functions are defined and presented graphically in Figure 3.

Each output layer node, y_k (k = 1,2,3, ..., m), will add up all signals from the hidden layer nodes (multiplied by weight, w_{jk} and added bias, b_k) as in Equation 4. The signal coming out of the output node is calculated by using activation function (Equation 5).

$$y\_in_k = b_k + \sum_{j=1}^{p} z_i w_{jk} \quad (4)$$

$$y_k = f(y\_in_k) \quad (5)$$

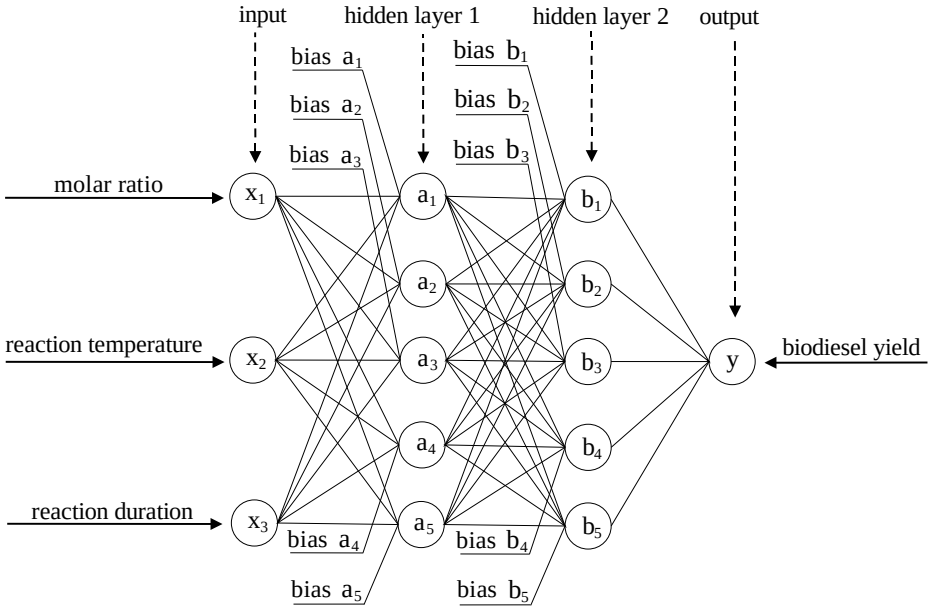

Figure 2. Form of ANN model consisting of three layers for biodiesel yield prediction

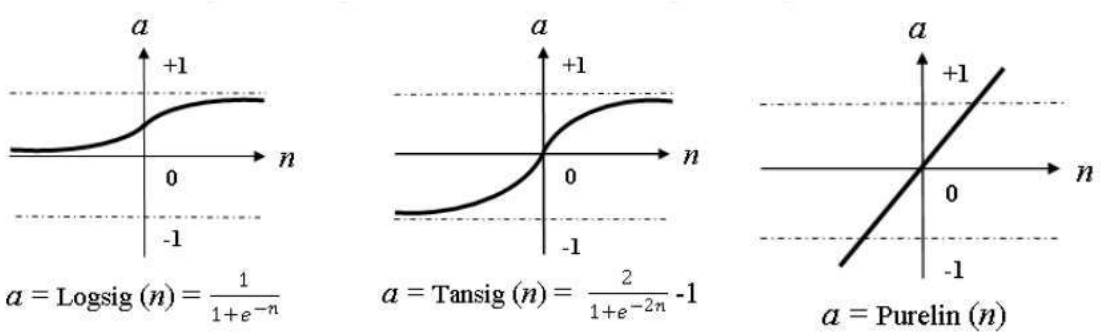

Figure 3. Definition and graphical representation of logsig, tansig, and purelin activation functions

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The second step (back-propagation) starts by calculating the error information (δ_k) between each output node (y_k) with target value (t_k) associated with data for learning as in Equation 6. In order to correct the weight and bias values, correction for weight (Δw_{jk}) and bias (Δb_k) were calculated using the predetermined learning rate (α) as given in Equation 7 and Equation 8.

$$\delta_k = (t_k - y_k) f'(y\_in_k) \quad (6)$$

$$\Delta w_{jk} = \alpha \delta_k z_j \quad (7)$$

$$\Delta b_k = \alpha \delta_k \quad (8)$$

The third step (weight adjustment) starts by calculating the error information (δ_j) between each hidden layer node (z_j, j = 1,2,3, ..., p) with the nodes in the input layer as in Eq. 9. Then calculate the correction weight (Δv_{ij}) and correction of bias (Δb_j) to correct the weight value (v_{ij}) and bias value (b_j) as in Equation 10 and Equation 11 by using learning rate (α).

$$\delta_j = \left(\sum_{k=1}^{m} \delta_k w_{jk} \right) f'(z\_in_j) \quad (9)$$

$$\Delta v_{ij} = \alpha \delta_j x_j \quad (10)$$

$$\Delta b_j = \alpha \delta_j \quad (11)$$

In the weight adjustment step, each output unit (y_k) is fixed by the new value of its weight and bias as in Equation 12 and Equation 13 (Widodo *et al*., 2013). Similarly, Equation 14 and Equation 15 was used to fix each hidden unit (z_j).

$$w_{jk}\,(\text{new}) = w_{jk}(\text{old}) + \Delta w_{jk} \quad (12)$$

$$b_k\,(\text{new}) = b_k\,(\text{old}) + \Delta b_k \quad (13)$$

$$v_{ij}\,(\text{new}) = v_{ij}\,(\text{old}) + \Delta v_{ij} \quad (14)$$

$$b_j\,(\text{new}) = b_j\,(\text{old}) + \Delta b_j \quad (15)$$

2.3. Model training

The ANN training process is preceded by network initialization. Network initialization is to determine the initial network architecture so that the network training process can be carried out. The learning rate (α) used to train ANN models is 0.001 (Amini, 2008). The type of training used is the trainlm (Levenberg-Marquardt) training type (Anandhi *et al*., 2012). The maximum number of iterations is set at 1000 and the smallest Mean Square Error (MSE) is 0.00001 (Kusuma & Abadi, 2011). Variations in activation functions used by 27 variations are combinations of logsig-tansig-purelin (Dorofki *et al*., 2012). Table 1 was reproduced from our previous work (Authors, 2019) for the training purpose. All data were used for training set data except values collected at reaction time of eight minutes.

2.4. Model validation

Model validation was performed to assess the accuracy between predicted and observed values. In this case, validation is evaluated by using relative root mean square error (*RRMSE*) and coefficient of determination (R^2) with a target to obtain the smallest *RRMSE* and the highest R^2 values.

$$RRMSE = \frac{\sqrt{\frac{1}{n}\sum_{i=1}^{n}(O_i - P_i)^2}}{\overline{O}} \times 100 \quad (16)$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(O_i - P_i)^2}{\sum_{i=1}^{n}(O_i - \overline{O})^2} \quad (17)$$

where n is number of data, O_i is observed value of i^{th}, P_i is predicted value of i^{th}, and $\overline{o}$ is average observed value. The predicted yield is classified as excellent with *RRMSE* < 10%, good (10-20%), fair (20-30%), and poor (> 30%) (Li *et al*., 2013).

The R^2 is used to assess the closeness of calculated values to the measurement data. The prediction values are considered excellent if $R^2 \approx 1$ (Despotovic *et al*., 2016).

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Tabel 1. Biodiesel yield at different *MR*, *T*, and *t* used as training set data

Time (min)	Biodiesel yield (%)					
	30°C	35°C	40°C	45°C	50°C	55°C
MR 1:4						
0.25	24.27	25.49	29.38	35.67	24.63	29.26
0.5	29.10	31.16	35.06	37.28	29.30	37.07
1	34.83	37.01	42.23	40.93	36.15	41.64
2	38.72	40.98	48.14	47.02	38.84	55.98
3	41.69	46.46	51.37	53.08	45.63	61.12
6	47.69	52.96	56.43	61.47	54.57	65.93
8	50.52	56.97	61.32	63.33	65.37	69.15
10	53.43	58.44	65.22	67.23	69.27	73.04
MR 1:5						
0.25	30.14	25.60	32.68	30.70	27.49	29.57
0.5	32.95	37.99	37.97	32.21	34.14	31.92
1	36.90	41.81	41.85	34.98	43.97	43.88
2	41.69	44.89	44.83	45.01	50.79	47.23
3	43.64	51.49	49.71	50.92	54.55	54.61
6	53.46	55.67	58.00	56.41	63.41	63.53
8	57.39	59.56	59.56	66.15	68.44	70.93
10	61.29	63.46	67.27	68.09	70.40	73.84
MR 1:6						
0.25	38.88	35.44	33.64	24.62	30.08	22.57
0.5	41.82	42.72	40.63	34.01	42.80	29.02
1	43.69	48.50	44.54	41.50	49.69	33.07
2	47.86	51.49	48.52	49.47	51.87	38.64
3	52.59	56.24	54.43	56.91	58.34	54.69
6	58.46	64.02	62.90	61.11	62.62	67.56
8	63.27	65.73	65.68	67.97	71.59	75.50
10	66.19	67.67	67.61	69.92	75.46	78.44

3. RESULTS AND DISCUSSION

3.1. ANN model for yield prediction

Table 2 shows a summary of the results from training and validation of 27 architectural combination models of logsig-tansig-purelin activation functions. Model accuracy is evaluated from the *RRMSE* and R^2 values of the models. We can see that 24 of 27 combination models demonstrate excellent accuracy with *RRMSE* values less than 10% (between 3.32 and 6.90%) and R^2 more than 0.90 (between 0.9364 and 0.9860). The remaining three models can still be grouped as good models with *RRMSE* values between 11 and 12.5% and R^2 values between 0.7589 and 0.8307. During the training session, the best architecture is tansig-tansig-purelin with *RRMSE* value of 3.32% and R^2 of 0.9860. However, this model fails as the best model during validation session. The *RRMSE* value of this model rises to 4.53% and its R^2 decreases significantly to 0.8382.

During the validation session, 25 of the 27 models meet the excelent criteria with *RRMSE* values of less than 10%. Two models fall into fair group with *RRMSE* values of 23.71% and 27.95%. Even the three lowest models previously mentioned (purelin-

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purelin-purelin, purelin-purelin-tansig, and purelin-purelin-logsig) which have *RRMSE* values greater than 10% in the training session, exhibit excelent *RRMSE* (< 10%) during validation. However, if the models are examinated from its determination coefficients, some of the best models have very low R^2 values, five models have even negative R^2. This means that the accuracy of the results during the training session does not always reflect that the validation results will also be accurate. The mentioned three lowest models in the training session still showed the worst performance with a negative R^2 values. Nine architectural models defend themselves as excelent models with R^2 values greater than 0.90 and *RRMSE* values during validation session are lower than those of training session.

Based on the discussion above, the selection of the best activation function network architecture that in turn will be used as a prediction model must be based on both the smallest *RRMSE* value and the highest R^2 value in the validation session. A low *RRMSE* value indicates a small deviation from all data so that the prediction model built successfully achieves a high level of accuracy. On the other side, high R^2 value indicates a very close relationship between calculated biodiesel yields and observed yield values.

Table 2. Result from training and validation of ANN models

Activation function	Training		Validation	
	***RRMSE* (%)**	**R^2**	***RRMSE* (%)**	**R^2**
logsig-logsig-logsig	4.22	0.9772	5.15	0.8049
logsig-logsig-tansig	3.79	0.9817	3.83	0.7978
logsig-tansig-logsig	5.20	0.9648	6.34	-0.8360
logsig-tansig-tansig	3.87	0.9809	4.86	0.7865
tansig-logsig-logsig	3.99	0.9793	27.95	-0.2730
tansig-tansig-logsig	3.66	0.9829	4.04	0.8332
tansig-tansig-tansig	3.49	0.9843	2.77	0.9110
tansig-logsig-tansig	5.14	0.9658	4.98	0.3850
logsig-tansig-purelin	5.25	0.9642	6.14	0.8638
logsig-logsig-purelin	4.24	0.9770	3.02	0.9099
tansig-logsig-purelin	5.12	0.9661	23.71	0.0903
tansig-tansig-purelin	3.32	0.9860	4.53	0.8382
logsig-purelin-logsig	**5.14**	**0.9656**	**2.41**	**0.9355**
logsig-purelin-tansig	6.16	0.9499	3.20	0.9290
tansig-purelin-logsig	5.51	0.9597	4.11	0.8855
tansig-purelin-tansig	6.42	0.9444	3.30	0.8993
purelin-logsig-logsig	6.15	0.9502	2.73	0.9342
purelin-logsig-tansig	**6.28**	**0.9479**	**2.44**	**0.9391**
purelin-tansig-logsig	6.90	0.9364	3.51	0.9074
purelin-tansig-tansig	6.12	0.9512	3.29	0.8535
purelin-purelin-purelin	12.51	0.7589	5.98	-1.2950
purelin-purelin-tansig	10.99	0.8307	8.50	-34.7200
purelin-purelin-logsig	11.51	0.8104	8.14	-15.0800
purelin-tansig-purelin	6.40	0.9460	5.89	0.7532
purelin-logsig-purelin	6.47	0.9448	2.68	0.9042
logsig-purelin-purelin	6.89	0.9369	2.91	0.9002
tansig-purelin-purelin	6.71	0.9403	7.10	0.4931

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Among the excellent models appeared during validation session, ANN models with activation function architecture of logsig-purelin-logsig and purelin-logsig-tansig are the best two with respectively *RRMSE* of 2.41% and 2.44%, and R^2 of 0.9355 and 0.9391. During the training session these models (logsig-purelin-logsig and purelin-logsig-tansig) are also among the excelent models with *RRMSE* less than 10% and R^2 greater than 0.90. Figure 4 and 5 show the scater between the predicted values of bio-diesel yield observation values, both in training sessions and validation sessions.

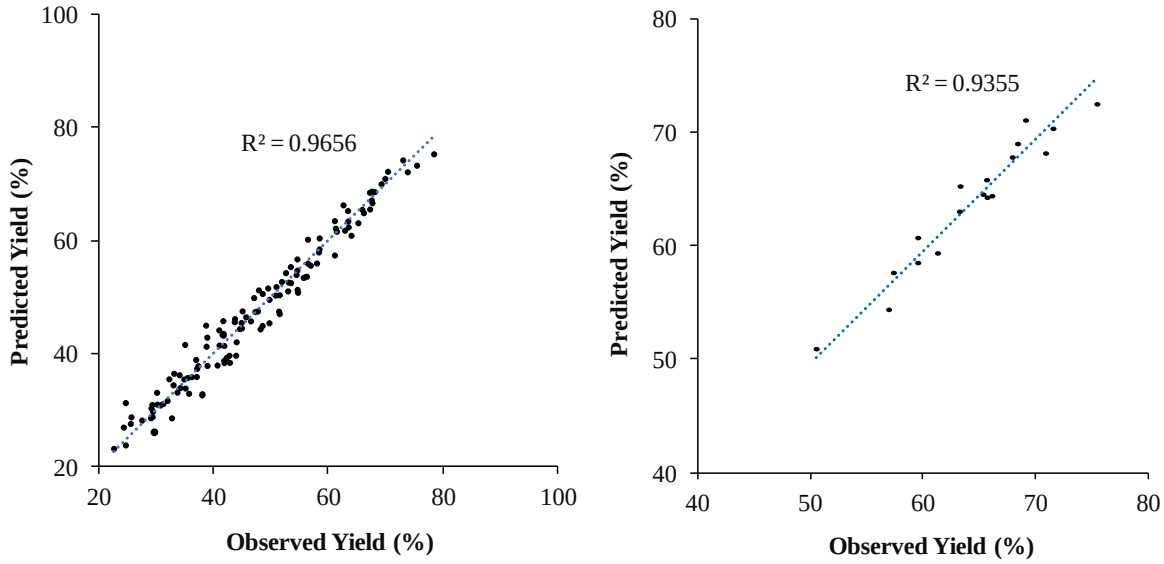

Figure 4. Scatter diagram of observation vs. prediction yield from ANN model with architecture of logsig-purelin-logsig: training result (left) and validation result (right)

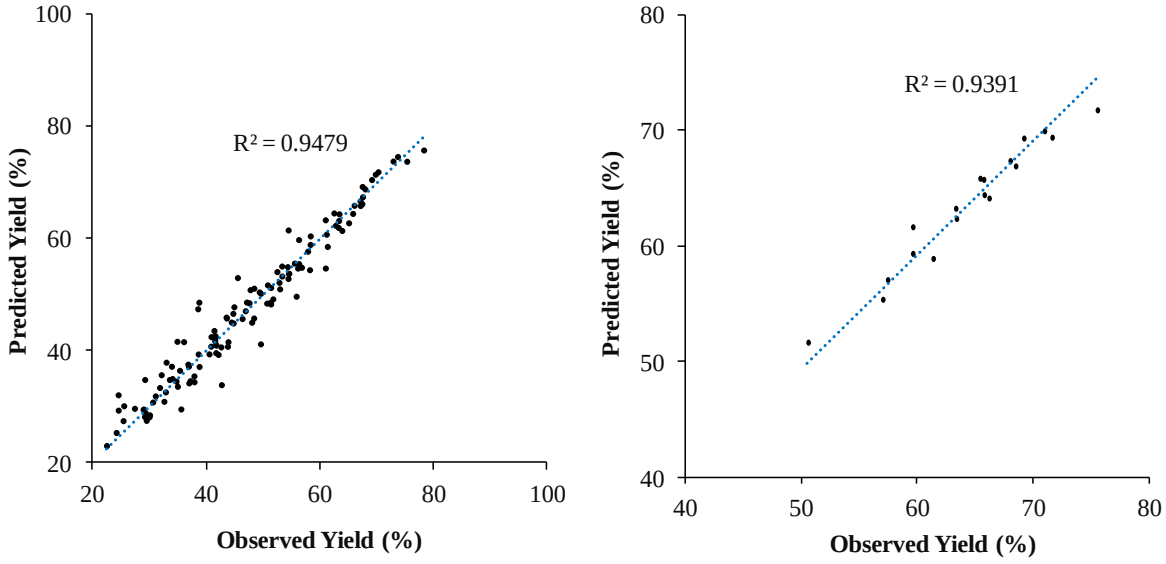

Figure 5. Scatter diagram of observation vs. prediction yield from ANN model with architecture of purelin-logsig-tansig: training result (left) and validation result (right)

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Previously we have reported the prediction of biodiesel yield at the same conditions using first order kinetic approach (Authors, 2019). The result showed excellent predictions with *RRMSE* of 3.39%, but R^2 value of 0.8454. Compared to the results from this study, it is clear that application of ANN model provide better prediction than those of first order kinetic.

4. CONCLUSION

The network architecture of the ANN model consists of three layers with three nodes in the input layer, five nodes in the first and the second hidden layer, and one node in the output layer. The type of training used is the Levenberg-Marquardt (*trainlm*) with a learning rate of 0.001. Twenty seven architectural combinations of three activation function (logsig, tansig, purelin) had been trained using 126 data set of biodiesel yield observed at three different molar ratios, six different temperatures and seven points reaction time, and had been validated using 18 data set observed at reaction time of eight minutes. Results showed that models with activation function of logsig-purelin-logsig and purelin-logsig-tansig be the best with *RRMSE* of 2.41% and 2.44%, respectively, and R^2 of 0.9355 and 0.9391%, respectively.

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6. AUTHORS' NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. Authors confirmed that the data and the paper are free of plagiarism.

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Authors wrote well. Minor correction: Discussion must be compared with references. Other is OK.

Reviewer 2:
1. Please complete the author names.
2. The authors should briefly explain the content of data training used in this research.
3. The authors should present the parameters assigned in the experiments.

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Application of Artificial Neural Network to Predict Bio-diesel Yield from Waste Frying Oil Transesterification

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ABSTRACTS

Used frying oil (UFO) has great potential as feedstock for biodiesel production. This study aims to develop an artificial neural network (ANN) model to predict biodiesel yield produced from base-catalyzed transesterification of UFO. The experiment was performed with 100 ml of UFO at three different molar ratios (oil:methanol), namely 1:4, 1:5, 1:6, reaction temperatures of 30 to 55 °C (raised by 5 °C) and reaction time of 0.25, 0.5, 1, 2, 3, 6, 8, and 10 min. Prediction model was based on ANN model consisting of three layers with 27 combinations of three activation functions (tansig, logsig, purelin). All the 27 activation function architectures were trained using Levenberg-Marquardt train type with 126 data set (87.5%) and learning rate of 0.001. Model validation used 18 data set (12.5%) measured at reaction time of 8 min. Results show that two ANN models with activation function of logsig-purelin-logsig and purelin-logsig-tansig be the best with *RRMSE* of 2.41% and 2.44%, respectively, and R^2 of 0.9355 and 0.9391, respectively. Predictions of biodiesel yield using ANN models are significantly better than those of first-order kinetics.

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1. INTRODUCTION

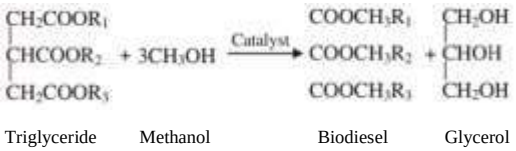
Biodiesel is an alternative energy source for diesel fuel offering several advantages. First, biodiesel is made from vegetable oils or animal fats so it is classified as a renewable, biodegradable, and nontoxic energy source (Khan *et al*., 2013). Biodiesel, therefore, is ecologically friendly because the production and application of biodiesel results in lower greenhouse gas (GHG) emissions than petroleum fuels do. Studies in the United States (Sheehan *et al*., 1998) revealed that based on life cycle analysis biodiesel is able to decrease GHG emission 78% as compared to petroleum diesel fuel. Other works using various feedstock in different countries also reported that

biodiesel application results in lower GHG such as in India with jatropha curcas (Kumar *et al*., 2012; Achten *et al*., 2010), China with various oils (Hou *et al*., 2011; Guo *et al*., 2010), Southeast Asia (Indonesia, Malaysia, Thailand) with palm oil (Siregar *et al*., 2015; Harsono *et al*., 2012; Hassan *et al*., 2011; Silalertruksa & Gheewala, 2012), Brazilia with soybean oil (Oliveira *et al*., 2017), and Europe with rapeseed oil (Malça & Freire, 2011). Second, unlike fossil fuels which are bestowed to a few countries, oil-plants producing biodiesel feedstock are spread throughout the world, so that geopolitically biodiesel can be one that increase energy security (Paltsev, 2016).

Utilization of vegetable oils as feedstock for biodiesel production, however, is more expensive because the cost of raw materials can reach 80 to 85% of operational costs (Canakci & Sanli, 2008; Hindryawati *et al*., 2014). One potential cheaper raw material is used frying oil (UFO), which is not alowed to be dumped directly because it has a high COD value. In addition, UFO contains toxic compound formed during high temperature heating like hydroperoxides and aldehydes. When these compounds ingested through consumed food they may be responsible for increasing blood pressure (hypertension) and attributable to cardiovascular diseases and diabetes (Leong *et al*., 2015; Jaarin *et al*., 2018). Repeatedly heated cooking oils even produce carcinogenic compounds such as polycyclic aromatic hydrocarbons that relate to the incidence of tumor and cancer diseases (Ganesan *et al*., 2017). Therefore, using UFO repeatedly to fry food or to make food-related ingredients, such as chili sauce, may endanger human health.

The potential of used cooking oil in Indonesia is quite large because the consumption of cooking oil tends to increase from 0.198 L/capita per week in 2007 to 0.205 in 2012 and 0.221 in 2017 (BPS, 2018). This figure is very close with the study of Fujita *et al*. (2013) in Bogor, which found cooking oil consumption of one family is 3L/month or 36 L/year. With a proper management, UFO has great potential to be used as energy sources in the form of biodiesel to replace diesel oil (Chhetri *et al*., 2008). The development of biodiesel from UFO will provide a healthier choice in the utilization of UFO.

The reaction usually chosen to produce biodiesel from oil is transesterification with methanol and a base catalyst (Fajardo *et al*., 2011). For every mol of triglyceride or vegetable oil, the process stoichiometrically required three mols of methanol to produce three mols biodiesel or FAME (fatty acid methyl ester) and one mol glycerol, as presented in Equation (1).



Triglyceride Methanol Biodiesel Glycerol (1)

Important factors in biodiesel synthesis, among other, include molar ratio (MR) of oil to methanol, reaction temperature (*T*), and reaction duration (*t*). Transesterification is a reversible and equilibrium reaction so that to obtain maximum yield should be carried out with excess of methanol (Ejikeme *et al*., 2010), rather than 1:3 as in Equation 1. The disadvantage of this reaction is that there may be a side reaction in the form of safonification which will consume the catalyst so that reducing biodiesel yield, especially if the FFA content in UFO is quite high (Narasimharao *et al*., 2007). The relation of biodiesel yield and dependent variables is unlinear and so complex that require a robust model to accurately predict the yield. Conventional approaches such as kinetics analysis can be applied successfully in certain constrained environments, but it is not flexible in different conditions. The complexity of these relationships can be

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analyzed by using ANN (artificial neural network) models so as to produce high accuracy prediction of biodiesel yield (Lavalle *et al*., 2012; Seo, 2013).

ANN model has been an important tool to solve many problems in a wide range of areas such as pattern recognition, function approximation, categorization, prediction, optimization, associative memory, and control (Jain *et al*., 1996). Recently, ANN model is explored to predict biodiesel yield (Thoai *et al*., 2018). The ANN model has also been used to predict biodiesel properties such as viscosity, cloud point, fash point, pour point, and cetane number (Giwa *et al*., 2015; Al-Shanableh *et al*., 2016).

The general objective of this study is to predict biodiesel yield resulted from UFO transesterification reaction which is broke down into three specific objectives. First, examine the relationship between molar ratio, reaction temperature and reaction time as independent variables to biodiesel yield as the dependent variable. Second, develop ANN models to predict biodiesel yield from UFO transesterification reaction. Third, validate ANN models to obtain the most accurate models in predicting biodiesel yield.

2. MATERIALS AND METHODS

2.1. Biodiesel synthesis

The UFO was collected from fried food pedlars in the vicinity of the University of Lampung. Biodiesel was sinthesized by transesterification reaction using 100 ml UFO with methanol and NaOH. Combination of three different molar ratios (*MR*) of oil to methanol (1:4; 1:5; 1:6), six temperature (*T*) levels (30, 35, 40, 45, 50, 55^{o}C), and eight points of reaction time, *t* (0.25, 0.5, 1, 2, 3, 6, 8, 10 minutes) was run to evaluate their effect on biodiesel yield. Detil of materials and processing method have been recently reported elsewhere (Haryanto *et al*., 2019).

2.2. ANN model development

Figure 1 shows a step-by-step ANN model development to validation. The ANN model to be developed is back-propagation type with supervised learning method.

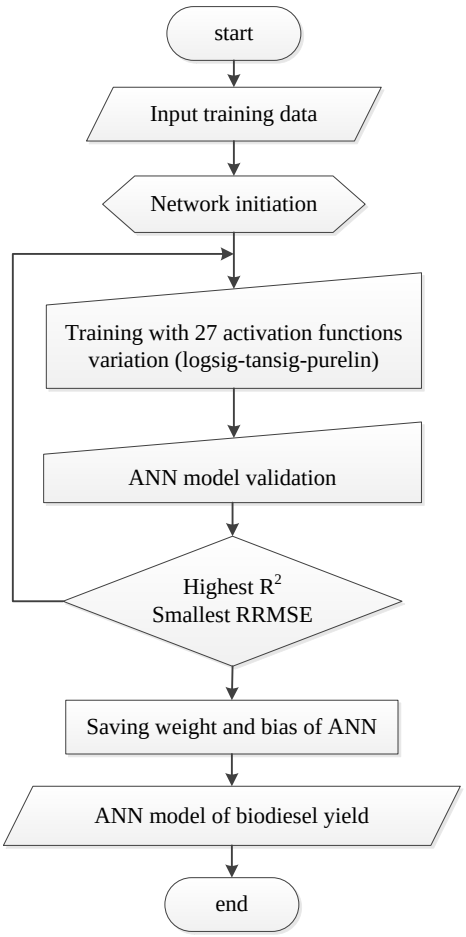

Figure 1. Flowchart fot ANN model development and validation

Figure 2 presents a form of ANN model for biodiesel yield prediction based on molar ratio, reaction temperature, and reaction time. The model consisted of three layers, namely input layer, two hidden layers, and output layer. Due to complexity of involved variables, two hidden layers are used, each with five neurons or nodes. Karsoliya (2012) guided that neuron number of hidden layer is less than twice of the number of neurons in input layer.

The ANN model works in three steps, namely feed forward, back-propagation, and

weight adjustments which are calculated based on the established equations (Widodo *et al*., 2013). At the first step, each input node receives an input value, x_i (i = 1, 2, 3, ..., n) and forwards the signal to all nodes in the hidden layer. Each hidden layer node will add all the weighted input signals (z_in_j), which is x_i multiplied by the weight (v_{ij}) and added by received bias, b_j (j = 1,2,3, ..., p) as in Equation 2. By using activation function as in Equation 3, the signal coming out from the hidden layer node is then calculated.

$$z\_in_j = b_j + \sum_{i=1}^{n} x_i v_{ij} \quad (2)$$

$$z_j = f(z\_in_j) \quad (3)$$

In this work, three activation functions (logsig, tansig, and purelin) were selected that making 27 combinations need to be validated. These three functions are defined and presented graphically in Figure 3.

Each output layer node, y_k (k = 1,2,3, ..., m), will add up all signals from the hidden layer nodes (multiplied by weight, w_{jk} and added bias, b_k) as in Equation 4. The signal coming out of the output node is calculated by using activation function (Equation 5).

$$y\_in_k = b_k + \sum_{j=1}^{p} z_i w_{jk} \quad (4)$$

$$y_k = f(y\_in_k) \quad (5)$$

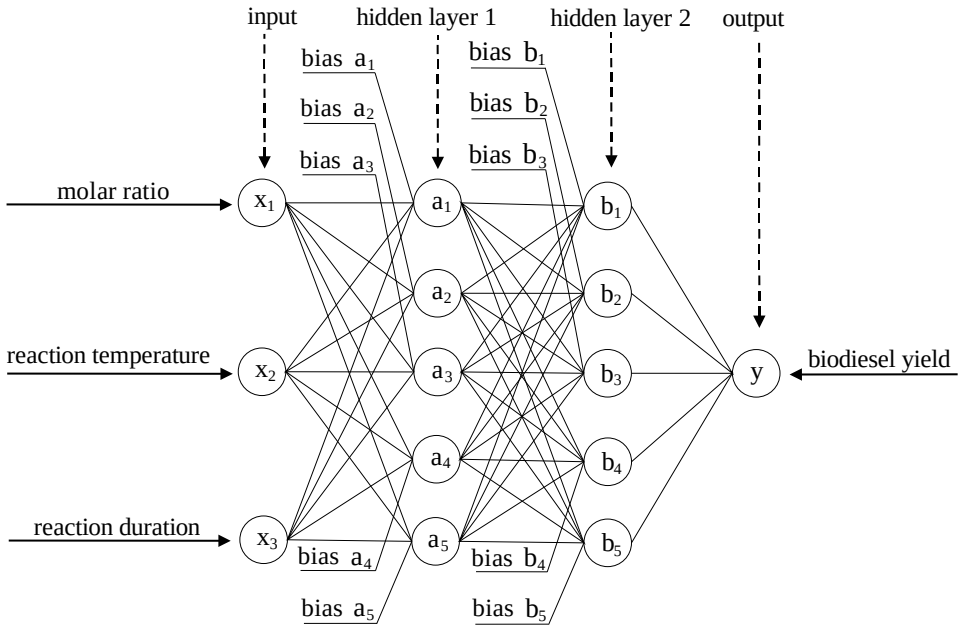

Figure 2. Form of ANN model consisting of three layers for biodiesel yield prediction

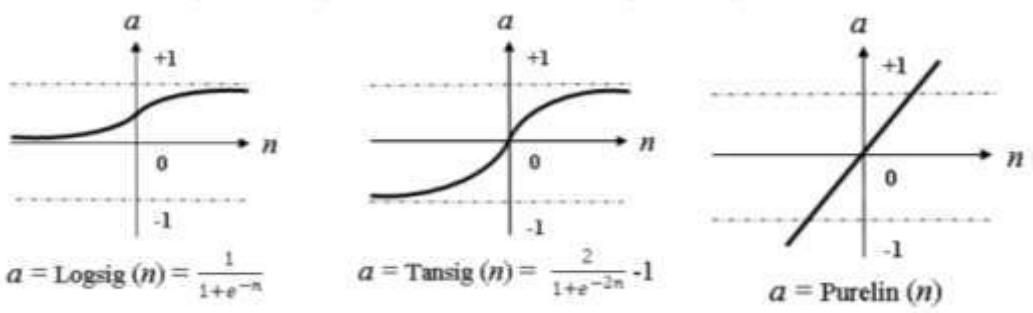

Figure 3. Definition and graphical representation of logsig, tansig, and purelin

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The second step (back-propagation) starts by calculating the error information (δ_k) between each output node (y_k) with target value (t_k) associated with data for learning as in Equation 6. In order to correct the weight and bias values, correction for weight (Δw_{jk}) and bias (Δb_k) are calculated using the predetermined learning rate (α) as given in Equation 7 and Equation 8.

$$\delta_k = (t_k - y_k)f'(y\_in_k) \quad (6)$$

$$\Delta w_{jk} = \alpha\delta_k z_j \quad (7)$$

$$\Delta b_k = \alpha\delta_k \quad (8)$$

The third step (weight adjustment) starts by calculating the error information (δ_j) between each hidden layer node (z_j, j = 1,2,3, ..., p) with the nodes in the input layer as in Eq. 9. Then calculate the correction weight (Δv_{ij}) and correction of bias (Δb_j) to correct the weight value (v_{ij}) and bias value (b_j) as in Equation 10 and Equation 11 by using learning rate (α).

$$\delta_j = \left(\sum_{k=1}^{m} \delta_k w_{jk}\right) f'(z\_in_j) \quad (9)$$

$$\Delta v_{ij} = \alpha\delta_j x_j \quad (10)$$

$$\Delta b_j = \alpha\delta_j \quad (11)$$

Each output unit is fixed by new value of its weight and bias as in Equation 12 and 13, and imilarly, for each hidden unit as in Equation 14 and 15 (Widodo *et al*., 2013).

$$w_{jk}\,(\text{new}) = w_{jk}(\text{old}) + \Delta w_{jk} \quad (12)$$

$$b_k\,(\text{new}) = b_k\,(\text{old}) + \Delta b_k \quad (13)$$

$$v_{ij}\,(\text{new}) = v_{ij}\,(\text{old}) + \Delta v_{ij} \quad (14)$$

$$b_j\,(\text{new}) = b_j\,(\text{old}) + \Delta b_j \quad (15)$$

The ANN model construction was build using toolbox of MATLAB.

2.3. Model training

The ANN training process is preceded by network initialization to determine initial network architecture so that the network training process can be carried out. Twenty seven variations in activation function are combination of logsig, tansig, and purelin (Dorofki *et al*., 2012). Input data involved molar ratio, reaction time, and reaction temperature; whereas biodiesel yield was target parameter. Out of 144 data set (Table 1), 126 data pairs (87.5%) were used for the training by using Levenberg-Marquardt training type (Anandhi *et al*., 2012) with learning rate (α) of 0.001 (Amini, 2008). The maximum number of iterations was set at 1000 and the smallest mean square error (MSE) is 0.00001 (Kusuma & Abadi, 2011).

2.4. Model validation

Model validation was performed to assess the accuracy between predicted and observed values. All data values (Table 1) collected at reaction time of eight minutes (12.5%) were used for validation session. In this case, validation was evaluated by using relative root mean square error (*RRMSE*) and coefficient of determination (R^2) with a target to obtain the smallest *RRMSE* and the highest R^2 values.

$$RRMSE = \frac{\sqrt{\frac{1}{n}\sum_{i=1}^{n}(O_i - P_i)^2}}{\overline{O}} \times 100 \quad (16)$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(O_i - P_i)^2}{\sum_{i=1}^{n}(O_i - \overline{O})^2} \quad (17)$$

where n is number of data, O_i is observed value of i^{th}, P_i is predicted value of i^{th}, and $\overline{o}$ is average observed value. The predicted yield is classified as excellent with *RRMSE* < 10%, good (10-20%), fair (20-30%), and poor (> 30%) (Li *et al*., 2013).

The R^2 is used to assess the closeness of calculated values to the measurement data. The predictions are considered excellent if R^2 close to one (Despotovic *et al*., 2016).

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Tabel 1. Biodiesel yield at different *MR*, *T*, and *t* used as training set data

Time (min)	Biodiesel yield (%)					
	30°C	35°C	40°C	45°C	50°C	55°C
MR 1:4						
0.25	24.27	25.49	29.38	35.67	24.63	29.26
0.5	29.10	31.16	35.06	37.28	29.30	37.07
1	34.83	37.01	42.23	40.93	36.15	41.64
2	38.72	40.98	48.14	47.02	38.84	55.98
3	41.69	46.46	51.37	53.08	45.63	61.12
6	47.69	52.96	56.43	61.47	54.57	65.93
8	50.52	56.97	61.32	63.33	65.37	69.15
10	53.43	58.44	65.22	67.23	69.27	73.04
MR 1:5						
0.25	30.14	25.60	32.68	30.70	27.49	29.57
0.5	32.95	37.99	37.97	32.21	34.14	31.92
1	36.90	41.81	41.85	34.98	43.97	43.88
2	41.69	44.89	44.83	45.01	50.79	47.23
3	43.64	51.49	49.71	50.92	54.55	54.61
6	53.46	55.67	58.00	56.41	63.41	63.53
8	57.39	59.56	59.56	66.15	68.44	70.93
10	61.29	63.46	67.27	68.09	70.40	73.84
MR 1:6						
0.25	38.88	35.44	33.64	24.62	30.08	22.57
0.5	41.82	42.72	40.63	34.01	42.80	29.02
1	43.69	48.50	44.54	41.50	49.69	33.07
2	47.86	51.49	48.52	49.47	51.87	38.64
3	52.59	56.24	54.43	56.91	58.34	54.69
6	58.46	64.02	62.90	61.11	62.62	67.56
8	63.27	65.73	65.68	67.97	71.59	75.50
10	66.19	67.67	67.61	69.92	75.46	78.44

3. RESULTS AND DISCUSSION

3.1. ANN model for yield prediction

Table 2 shows a summary of the results from training and validation of 27 models of architectural combination of logsig-tansig-purelin activation functions. Model accuracy was evaluated from the *RRMSE* and R^2 values of the models. We observed that during training session 24 out of 27 combination models demonstrate excellent accuracy with *RRMSE* values less than 10% (between 3.32 and 6.90%) and R^2 more than 0.90 (between 0.9364 and 0.9860). The remaining three models (purelin-purelin-purelin, purelin-purelin-tansig, and purelin-purelin-logsig) can still be grouped as good models with *RRMSE* values between 11 and 12.5% and R^2 values between 0.7589 and 0.8307. During the training session, the best architecture is tansig-tansig-purelin with *RRMSE* value of 3.32% and R^2 of 0.9860. However, this model failed as the best model during validation session. The *RRMSE* value of this model rised to 4.53% and its R^2 decreased significantly to 0.8382.

During the validation session, 25 out of the 27 models met the excelent criteria with *RRMSE* values of less than 10%. Two models fallen into fair group with *RRMSE* values of 23.71% and 27.95%. Even the three lowest

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models previously mentioned which have *RRMSE* values greater than 10% in the training session, exhibited excelent *RRMSE* (< 10%) during validation. However, if the models are examined from its determination coefficients, some of the best models have very low R^2 values, and five models have even negative R^2. This means that the accuracy of the results during the training session does not always reflect that the validation results will also be accurate. The mentioned three lowest models in the training session even displayed the worst performance with a negative R^2 values during validation session. Nine architectural models defended themselves as excelent models with R^2 values greater than 0.90 and *RRMSE* values lest than 10%. During validation session, however, the *RRMSE* of these models are lower as compared to those values during training session.

Based on the discussion above, the selection of the best activation function network architecture that in turn will be used as a prediction model must be based on both the smallest *RRMSE* value and the highest R^2 value in the validation session. A low *RRMSE* value indicates a small deviation from all data so that the prediction model successfully achieves a high level of accuracy. On the other side, high R^2 value indicates a very close relationship between calculated and observed biodiesel yields.

Table 2. Result from training and validation of ANN models

Activation function	Training		Validation	
	***RRMSE* (%)**	**R^2**	***RRMSE* (%)**	**R^2**
logsig-logsig-logsig	4.22	0.9772	5.15	0.8049
logsig-logsig-tansig	3.79	0.9817	3.83	0.7978
logsig-tansig-logsig	5.20	0.9648	6.34	-0.8360
logsig-tansig-tansig	3.87	0.9809	4.86	0.7865
tansig-logsig-logsig	3.99	0.9793	27.95	-0.2730
tansig-tansig-logsig	3.66	0.9829	4.04	0.8332
tansig-tansig-tansig	3.49	0.9843	2.77	0.9110
tansig-logsig-tansig	5.14	0.9658	4.98	0.3850
logsig-tansig-purelin	5.25	0.9642	6.14	0.8638
logsig-logsig-purelin	4.24	0.9770	3.02	0.9099
tansig-logsig-purelin	5.12	0.9661	23.71	0.0903
tansig-tansig-purelin	3.32	0.9860	4.53	0.8382
logsig-purelin-logsig	**5.14**	**0.9656**	**2.41**	**0.9355**
logsig-purelin-tansig	6.16	0.9499	3.20	0.9290
tansig-purelin-logsig	5.51	0.9597	4.11	0.8855
tansig-purelin-tansig	6.42	0.9444	3.30	0.8993
purelin-logsig-logsig	6.15	0.9502	2.73	0.9342
purelin-logsig-tansig	**6.28**	**0.9479**	**2.44**	**0.9391**
purelin-tansig-logsig	6.90	0.9364	3.51	0.9074
purelin-tansig-tansig	6.12	0.9512	3.29	0.8535
purelin-purelin-purelin	12.51	0.7589	5.98	-1.2950
purelin-purelin-tansig	10.99	0.8307	8.50	-34.7200
purelin-purelin-logsig	11.51	0.8104	8.14	-15.0800
purelin-tansig-purelin	6.40	0.9460	5.89	0.7532
purelin-logsig-purelin	6.47	0.9448	2.68	0.9042
logsig-purelin-purelin	6.89	0.9369	2.91	0.9002
tansig-purelin-purelin	6.71	0.9403	7.10	0.4931

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Among the excellent models appeared during validation session, ANN models with activation function architecture of logsig-purelin-logsig and purelin-logsig-tansig be the best two with *RRMSE* of 2.41% and 2.44%, respectively, and R^2 of 0.9355 and 0.9391, respectively. During the training session these models (logsig-purelin-logsig and purelin-logsig-tansig) are also among the excelent models with *RRMSE* less than 10% and R^2 greater than 0.90. Figure 4 and 5 show the scater between predicted and observed values of biodiesel yield, both in training session and validation session.

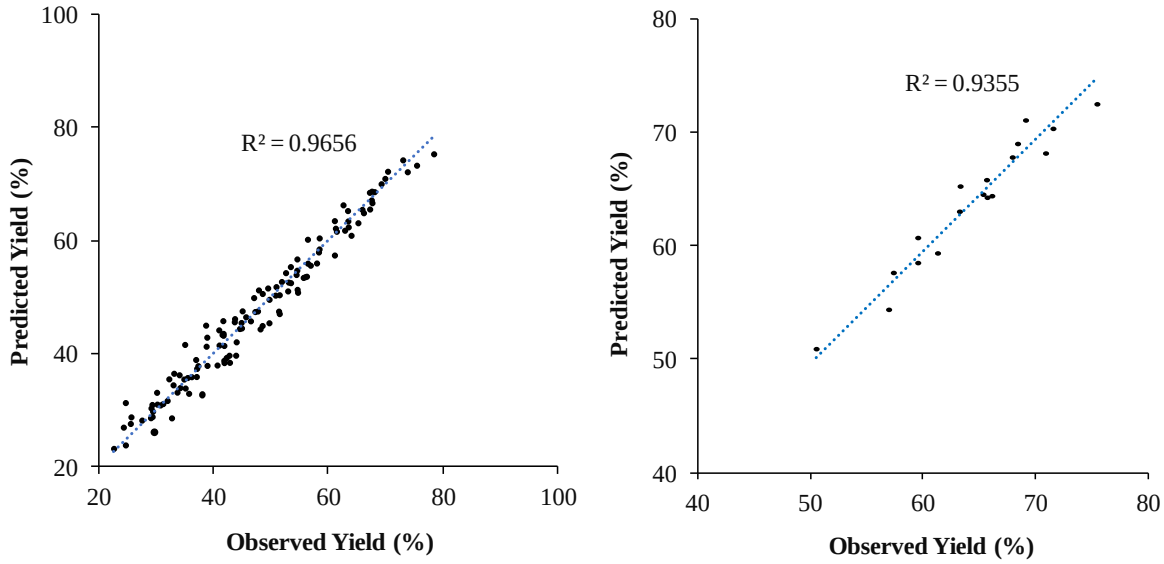

Figure 4. Scatter diagram of observation vs. prediction yield from ANN model with architecture of logsig-purelin-logsig: training result (left) and validation result (right)

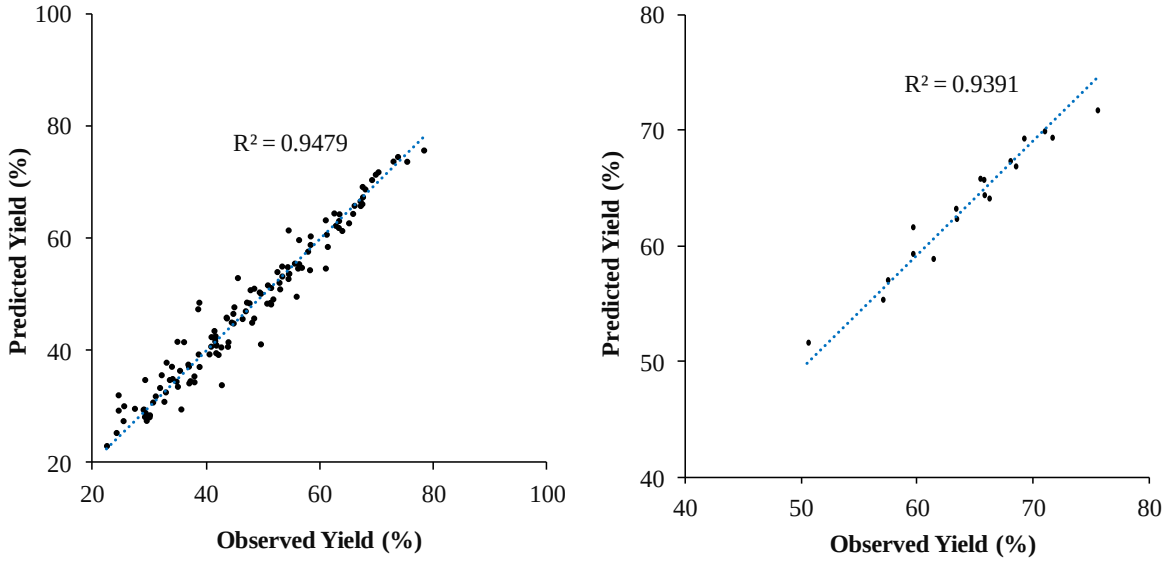

Figure 5. Scatter diagram of observation vs. prediction yield from ANN model with architecture of purelin-logsig-tansig: training result (left) and validation result (right)

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Previously we have reported the prediction of biodiesel yield at the same conditions using first order kinetic approach and the result showed excellent predictions with *RRMSE* of 3.39%, but R^2 value of 0.8454 (Haryanto *et al*., 2019). Compared to the results from this study, it is clear that application of ANN model provide better prediction than those of first order kinetic. Other works also reported the superiority of the ANN model for biodiesel prediction compared to other common tools such as linear regression and partial least squares regression (Ozgur & Tosun, 2017; Balabin *et al*., 2011). For a comparison, Table 3 summarized other studies on the application of ANN in predicting biodiesel content and biodiesel properties. It can be surmised that our result is comparable with other works and further emphasize that ANN model is a powerfull tool to predict biodiesel yield from different reaction conditions.

Table 3. Comparison of ANN application for biodiesel-related research

Feedstock	Input variables	Output target	ANN model architecture*	Model performance**	Reference
WFO	*MR, T, t*	Biodiesel yield	(3:5:5:1)	R^2 = 0.94; RRMSA = 2.41%	This works
Oil (unspesific)	*MR, T, t, P*	Biodiesel yield with Super-critical Methanol	(4:17:1)	R^2 = 0.9980; MSE = 4.49×10^{-4}	Farobie et al., 2015
Refined palm oil	*MR, T, t,* catalyst	FAME content	(4:3:3:1)	R^2 = 0.9958; RMSE = 0.0313	Thoi et al., 2018
Cotton oil	*T*, blend ratio	Viscosity	(2:3:1)	MAPE = 0.19%	Ozgur & Tosun, 2017
Cotton oil	*T*, blend ratio	Density	(2:4:1)	MAPE = 0.02%	Ozgur & Tosun, 2017
Refined canola oil	Fatty acid composition	Cloud point	(9:6:3)	R^2 = 0.98; SE = 1.7	Al-Shanableh et al., 2016
Refined canola oil	Fatty acid composition	Pour point	(9:6:3)	R^2 = 0.94; SE = 2.1	Al-Shanableh et al., 2016
Different oils	Chemical composition	Cetane number	(5:2:4)	R^2 = 0.9349; MAE = 0.955	Giwa et al., 2015
Some oil types	Fatty acid composition	Flash point	(5:2:4)	R^2 = 0.981; MAE = 1.705°C	Giwa et al., 2015

*) First figure is the neuron number of input layer, last figure is the number of output, and figure(s) in the middle is the neuron number of hidden layer (one or more hidden layers).

**) MAE = Mean Absolute Error; MAPE = Mean Absolute Percentage Error; RMSE = Root Mean Squared Error; SE = standard Error.

4. CONCLUSION

The network architecture of the ANN model consists of three layers with three nodes in the input layer, five nodes in the first and the second hidden layers, and one node in the output layer. The type of training used is the Levenberg-Marquardt with a learning rate of 0.001. Twenty seven (27) architectural combinations of three activation function (logsig, tansig, purelin) have been trained using 126 data set (87.5%) of biodiesel yield observed at three different molar ratios, six different temperatures and seven points reaction time, and have been validated using 18 data set (12.5%) observed at reaction time of eight minutes. Results confirm that models

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with activation function of logsig-purelin-logsig and purelin-logsig-tansig be the best with *RRMSE* of 2.41% and 2.44%, respectively, and R^2 of 0.9355 and 0.9391%, respectively.

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6. AUTHORS' NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. Authors confirmed that the data and the paper are free of plagiarism.

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#15334 SUMMARY

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Authors	Agus Haryanto, Tri Wahyu Saputra, Mareli Telaumbanua, Amiera Citra Gita
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Title and Abstract

Title	Application of Artificial Neural Network to Predict Bio-diesel Yield from Waste Frying Oil Transesterification
Abstract	Used frying oil (UFO) has great potential as feedstock for biodiesel production. This study aims to develop an artificial neural network (ANN) model to predict biodiesel yield produced from base-catalyzed transesterification reaction of UFO. The experiment was performed with 100 ml of UFO at three different molar ratios (oil:methanol), namely 1:4, 1:5, 1:6, reaction temperatures of 30 to 55 ^{o}C (raised by 5 ^{o}C) and reaction time of 0.25, 0.5, 1, 2, 3, 6, and 10 min. Prediction model was based on ANN back propagation type with supervised learning method. Validation model was carried out using data set measured at reaction duration of 8 min. The results showed that network architecture of the ANN model consists of four layers, namely input layer, first hidden layer, second hidden layer, and output layer. The training used in this work is the Levenberg-Marquardt train type with

learning rate of 0.001 and activation function of purelin-logsig-purelin. The model validation revealed that predicted yield is excellently accurate with %RMSE value of 1.51% and the coefficient of determination (R^2) of 0.946. Prediction values resulted from the ANN model were better than those of first-order kinetics.

Indexing

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