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Prediction of cold flow properties of biodiesel fuel using artificial neural network

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Abstract

Artificial neural network (ANN) can be utilized as a tool for modeling the properties of biodiesel fuel those are related to the fatty acid (FA) composition of a feedstock. The cold flow properties (CFP) define the operability for diesel fuel which are strongly influenced by the FA composition of feedstock. Cloud point (CP), pour point (PP) and cold filter plugging point (CFPP) are used commonly to characterize CFP. Prediction of CFP based on the FA composition of feedstock can reduce the experimental effort to produce a biodiesel suitable for a regional climate. In an attempt for this, 9-6-3 back-propagation ANN architecture was implemented to estimate CP, PP and CFPP of biodiesel samples using nine FA components as input data of 103 biodiesel study collected from literature. To check the accuracy of the model developed, refined canola oil (RCO) and waste frying oil (WFO) were converted to biodiesel then, their CP, PP and CFPP temperatures were determined following the EN and ASTM standards. The CFP estimated by the ANN model were in close agreement with the experimental values. When compared with the experimental data, ANN model predicted the CP, PP and CFPP temperatures within 98%, 94% and 96% accuracy, respectively. The model developed has revealed that CFP of biodiesel were influenced primarily by saturation or unsaturation of FA components with a few exceptions. Since the ANN model can be trained from iterations, it predicted CFP with high accuracy inspite of the presence of nonlinearities, i.e. the average of the mean R^2 value for the three CFP was found as 0.96.

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Keywords: Artificial neural network; biodiesel; cold flow properties; fatty acid composition

1. Introduction

Biodiesel can be considered as an alternative fuel derived from renewable sources such as vegetable oils or animal fats and can substitute the petroleum diesel fuel. The usage of biodiesel have effects beneficial on

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environmental pollution, dependency to petroleum exporting countries, as well as improvement of domestic industry. Biodiesel produced should meet the specifications of ASTM D6751 or EN 14214 standards before being commercialized in the market[1,2]. The current EN 590:2009 standard that defines the automotive diesel fuel to be sold in the European Union allows the blending diesel fuel with biodiesel up to 7%. The high viscosity of vegetable oil is not suitable for the current diesel vehicles. The most common procedure to lower viscosity is via a base catalyzed transesterification[3,4] in which a feedstock is reacted with a short chain alcohol such as methanol in the presence of a base catalyst to yield FAME (Fatty Acid Methyl Esters), i.e., biodiesel. Biodiesel is a mixture of FA (fatty acid) esters with each ester component contributing to the properties of the fuel and those are identical to that of its parent oil or fat. Some of the physical and chemical specifications of biodiesel such as CFP (cold flow properties), cetane number, ignition quality, heat of combustion, oxidative stability, viscosity and lubricity are directly related to FA profile of the feedstock[5]. Present work investigates the influence of the FA composition of feedstock on the CFP of biodiesel fuel produced. The CFP include three important parameters for low temperature characteristics of biodiesel fuel, namely, CP (cloud point), PP (pour point) and CFPP (cold filter plugging point). At the CP, the crystals first become visible as cloud when fuel is cooled under the conditions described by ASTM D2500-09 or ISO 3015:1992. The PP is the lowest temperature at which fuel can flow and the amount of wax that crystallizes is sufficient to gel the fuel. The sample must be cooled following the procedure outlined either in ASTM D97-05 or ISO 3016:1994. The CFPP specified in ASTM D6371-05 and EN 116:1998 directly affects the diesel engine performance in winter. It determines the lowest temperature where 20 ml of fuel can be drawn through a 45 micron screen in 60 seconds with 200 mm of water (1.96 kPa) vacuum. The CFPP is usually between the CP and PP temperatures. The CP temperature is a must in the ASTM D6751 standard while PP and CFPP are listed in EN 14213 and EN 14214 standards. Prediction of CFP of biodiesel based on the FA composition of the feedstock prior to production can lead to a way for a suitable biodiesel for the regional climate conditions. The best feedstock composition that would enhance the biodiesel quality in cold weather is an important subject to be searched. ANN prediction and modeling techniques identify and learn complex nonlinear or linear relations between the input and output data. The ANN prediction models have been adopted to evaluate density, cetane number, CFP and kinematic viscosity of diesel fuels[6-12] but none of them is on CFP, yet. In a recent work, Piloto et al.[13] successfully predicted cetane number of biodiesel using an ANN model and showed that ANN is superior to the other methods. Meng et al.[14] modeled the prediction of kinematic viscosity at 313 K with the highest correlation coefficient of 0.9772 as compared to other viscosity prediction methods.

The research reported here was a part of a more general investigation on the production and characterization of a biodiesel project at Near East University (NEU)[15-19]. The two types of feedstock, namely waste frying oil (WFO) with high saturated (having only single C-C bonds) FA content and refined canola oil (RCO) with high unsaturated (having double C-C bonds) FA content were converted into biodiesel fuel via a one-step base-catalyzed transesterification reaction. Next, the CFP (CP, PP and CFPP) of the biodiesel were measured following the ASTM and EN standards. For the prediction of CFP, experimental data of 103 biodiesel fuel samples were traced from the literature with their corresponding FA compositions of parent feedstock. The values collected were utilized as input and output data for ANN prediction models. Finally, predicted CFP values were compared with the measured counterparts of the two biodiesel samples of the current work.

2. Material and Methods

2.1. Biodiesel Production

RCO that was purchased from a local supermarket and WFO which was collected from the university cafeteria were converted into biodiesel via a base catalyzed transesterification as prescribed by Freedman et al[20]. FA compositions of feedstock used were determined following the EN ISO 5508 and are tabulated in Table 1. Each feedstock was taken as 1.0 kg then converted into FAME using 1:3 oil to alcohol molar ratio of methanol and 0.5 wt % of NaOH as catalyst at 60°C under 1 atm for 1 hour by transesterification reaction.

Table 1. FA compositions and % FFA contents of biodiesel feedstock used in the current work.

	Fatty Acid Composition (wt %)								
	C12:0	C14:0	C16:0	C18:0	C18:1	C18:2	C18:3	C20:0	C20:1
RCO	0.08	0.0	5.63	1.57	62.97	21.34	6.99	0.46	1.04
WFO	1.18	0.10	37.29	4.04	40.42	17.84	0.18	0.0	0.0

The reaction was end up with the conversion of RCO to RCOME (Refined Canola Oil Methyl Ester) and WFO to WFOME (Waste Frying Oil Methyl Ester). Chemical composition and fatty acid profiles of biodiesel samples were analyzed using the Gas Chromatography– Mass Spectrometry (GC-MS) in accordance with the EN 14103 standard.

2.2. Measurement of CFP of Biodiesel

ASTM D2500-09, ASTM D97-05 and ASTM D6371-05 were used to determine the CP, PP and CFPP of the biodiesel samples. The apparatus for CP and PP measurements were manufactured following the ASTM standards[15]. The same set-up was also used for CFPP measurements with the addition of a vacuum system and a pipette with a filter unit.

2.3. Artificial Neural Network Prediction

For the prediction of CFP, experimental data of 103 biodiesel fuel samples were collected from the literature[21-30] with their corresponding FA compositions of parent feedstock and CFP. Literature data was utilized as input and output data for the ANN prediction model developed. The most common FA components of biodiesel feedstock were chosen as input variables, namely, lauric acid (C12:0), myristic acid (C14:0), palmitic acid (C16:0), stearic acid (C18:0), oleic acid (C18:1), linoleic acid (C18:2), linolenic acid (C18:3), arachidic acid (C20:0) and gondeic acid (C20:1). The maximum and the minimum values for the nine fatty acids present in the 103 feedstock, and CFP of biodiesel fuels produced from the feedstock are listed in Table 2. When a feedstock's FA composition falls within these ranges then, models developed here could be employed to predict the biodiesel CFP.

Table 2. Minimum and maximum values of FA and CFP of literature data[21-30].

	Min.	**Max.**	**Ave.**	**Count out of 103**
C12:0	0.0	49.2	8.95	36
C14:0	0.0	25.9	3.25	53
C16:0	0.9	44.1	13.0	103
C18:0	0.3	23.5	5.48	103
C18:1	1.8	92.5	38.2	103
C18:2	0.0	77.3	27.2	103
C18:3	0.0	72.3	7.77	94
C20:0	0.0	7.5	0.605	74
C20:1	0.0	66.5	2.34	48
CP (˚C)	-13.4	17.0	2.67	86
PP (˚C)	-23.0	15.0	-1.11	57
CFPP (˚C)	-13.0	17	-0.870	88

Artificial Neural Network (ANN) is an information processing methodology that replicates the biological nervous systems. To solve specific problems, a large number of interconnected processing elements (neurons) work together.

The multilayer feed forward neural network consists of an input layer, one or more hidden layers and an output layer. The number of the nodes in the input and output layers are related to the nature of the problem[31]. In the model developed here, the input layer is composed of 9 nodes, which are FA composition in weight fractions of feedstock in reference data. The output layer has 3 nodes, i.e., CP, PP and CFPP values of current biodiesel samples. One hidden layer was chosen initially and a suitable number of nodes in the hidden layer were determined by trial and error. The nodes between each layer were connected with adaptable weights. Equation 1 below describes how an artificial neuron or a node functions:

$$y_i = f(\sum^{n}_{j=1} x_j w_{ij} + b_i) \quad (1)$$

Where; x_j is the input from the previous node j and it is multiplied with the adapted weight w_{ij} that connects node i and node j. The total number of previous nodes connecting with node i is n. The products of all the inputs and weights are summed up and a bias b_i of node i is often added to the summation. The final summation is transferred by an activation function f to get the output of node i, y_i . The hyperbolic tangent sigmoid (tansig) function and the

linear (purelin) function are commonly utilized as activation functions[14]. The tansig function is used for a non-linear relationship approximation while the purelin function is used for a linear relationship approximation. The hidden layer activation function was chosen as sigmoid whilst the output layer activation function was chosen as linear (purelin) function. This combination was found to be valid and accurate, as mentioned in the litarature[13,14].

The critical step when developing an ANN architecture is the training procedure, where the weights and biases are adjusted to minimize the difference between the output of the ANN model and the actual value. The mean squared error (MSE) is usually applied as the performance function to interpret the difference between the output of the ANN and the actual value. The training procedure is achieved through the training algorithms and the back-propagation algorithm is one of the most popular training algorithm[31]. The problem in the training procedure is over-fitting, in which the ANN obtained memorizes the training examples and does not learn the ability to generalize on unseen data. To avoid over-fitting, the actual data can be divided into 3 data sets; the training set, the validation set and the test set. Both of the MSE obtained for the training set and the validation set should decrease during a training process. When the ANN starts to over fit, the error on the validation set will increase though the error on the training set continues decreasing. The training should be stopped at that stage. The test set is treated as an unseen data and will show accuracy of the trained ANN model developed[31,32].

The literature data from 103 biodiesel samples were divided into 2 data sets in the present work, 88 samples were used as training set while last 15 were used as validation set and the measured values for the two biodiesel samples produced were utilized as the test data. Each data set consisted of 9 FA compositions as inputs to the model and the three CFP; namely, CP, PP and CFPP as outputs. The ANN prediction system was implemented using the NN-pred, a Microsoft Excel™ software package. The ANN was trained to choose a suitable number of hidden layers starting from 1 to 12 nodes and the output should achieve the desired error goal of 0.01. In all variations the learning rate, momentum and initial weights were kept constant as 0.1, 0.1 and 0.3, respectively. ANN with 6 neurons in the hidden layer appears to give the best performance, and additional nodes over 6 did not make a significant improvement in performance. Finally, an ANN model was executed with the 9-6-3 architecture for prediction of CP, PP and CFPP temperatures as illustrated in Fig. 1. .

3. Results and Discussion

3.1. Characterization of Biodiesel Produced

To ensure the conversion of RCO and WFO to biodiesel, i.e. to RCOME and WFOME, ester contents determined according to EN 14103. Ester contents of RCOME and WFOME were found as 97 % and 96.4 % respectively, which were within the limits of the standards. CFP of the biodiesel samples were also tested and the results are shown collectively in Table 3.

Table 3. CFP of WFOME and RCOME

	Method	WFOME	RCOME
Cloud point (˚C)	ASTM D 2500	15	-3.5
Pour point (˚C)	ASTM D 97	12	-10
Cold filter plugging point (˚C)	ASTM D 6371	14	-7.5

It is evident that the CFP of WFOME and RCOME are rather poor, i.e. they become jeely at higher temperatures, compared with the commercial diesel no:2 fuel that has CP -16 ˚C, PP -27 ˚C and CFPP -18 ˚C. The WFOME showed inferior CFP as compared to RCOME. This behavior is attributed primarily on the large amounts of saturated fatty acid compounds present in WFO samples and a high content of palmitic acid portion. Udomsup et al.[33] noted that high content of palmitic acid (63 wt %) in palm stearin methyl ester caused an extreme increase in CP and PP values and reported as 18 ˚C and 19.4 ˚C, respectively.

3.2. ANN Model to Predict CFP of Biodiesel

While implementing the ANN model, the best architecture without over fitting was chosen as 6 neurons in one hidden layer. Models with up to 6 neurons in a hidden layer could not achieve the desired MSE of 1% neither in training nor in validation tests. All models with different number of neurons could achieve an acceptable error goal in training phase. However, they failed to achieve good results in the validation phase since they were trying to obtain a very small error goal in the training phase except the model that consists of six neurons. As a result, 9-6-3

back-propagation ANN architecture was implemented for prediction of CP, PP and CFPP of present biodiesel samples as shown in Fig. 1.

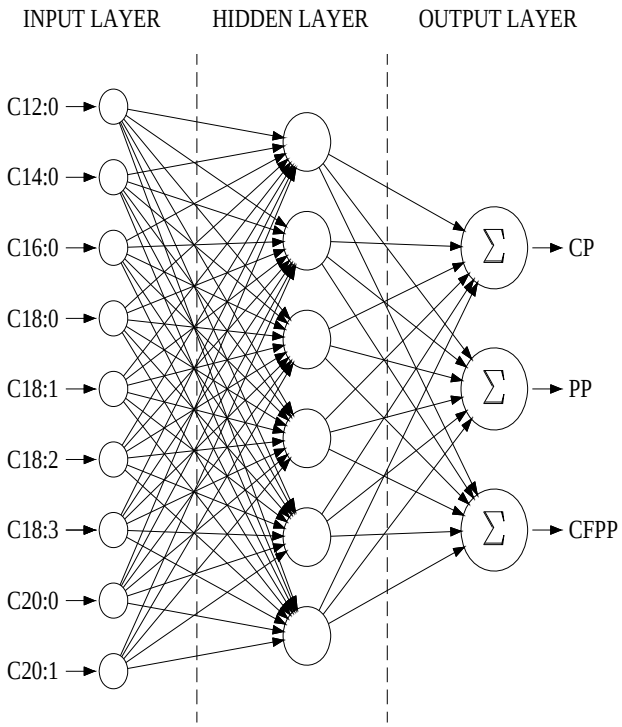

Fig. 1. The ANN architecture implemented for prediction of three CFP of biodiesel.

The R^2 from the results of ANN model between FA composition and CP, PP and CFPP were found as 0.98, 0.94 and 0.96, respectively while the standard error of estimation for CP, PP and CPPP were noted as 1.7, 2.1 and 1.8, respectively. A comparison of R^2 and absolute standard error (SE) values for ANN prediction models can be seen in Table 4.

Table 4. R^2 and SE values for ANN models for CFP of biodiesel fuel.

	CP	PP	CFPP
R^2	0.98	0.94	0.96
SE(˚C)	1.7	2.1	1.8

The saturated FA components showed a negative influence on CFP except C12:0 and C14:0 which had a positive effect for all CFP. The unsaturated components provided positive influences in the ANN modelling on CFP except C20:1 which had a negative influence on PP and CFPP temperatures.

The CP predicted seems to be the most accurate one, i.e., the closest to the measured values, among the CFP values. The CP value for biodiesel is more critical than the CFPP and PP both in ASTM D6751 and EN 14214 specifications, because of its practical significance. The PP and CFPP are not included in those standards directly. Nevertheless, CFPP is a useful parameter for diesel engines and PP is required for storage and pipelines.

CP, PP and CFPP temperatures for the WFOME were predicted as 13.5 ˚C, 11.0 ˚C and 10.9 ˚C, respectively using the ANN model. The same temperatures for RCOME were estimated as -3.6 ˚C, -9.5 ˚C and -6.8 ˚C, respectively and are listed in Table 5.

Table 5. A comparison of measured and predicted temperatures (˚C) for CFP of biodiesel samples.

	WFOME			RCOME		
	CP	PP	CFPP	CP	PP	CFPP

Measured CFP	15	11	12	-3.5	-10	-7.5
ANN - CFP	13.5	11.0	10.9	-3.6	-9.5	-6.8

The CP, PP and CFPP temperatures evaluated from the ANN models and measured CFP counterparts are compared in Fig. 2. to Fig. 4.

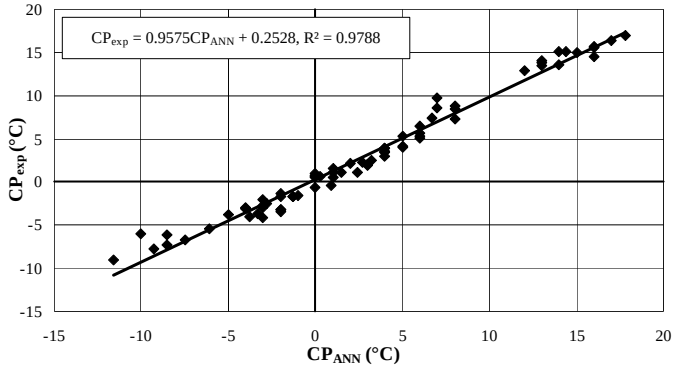

Fig2. A comparison of CP temperatures predicted by the ANN model with the experimental counterparts.

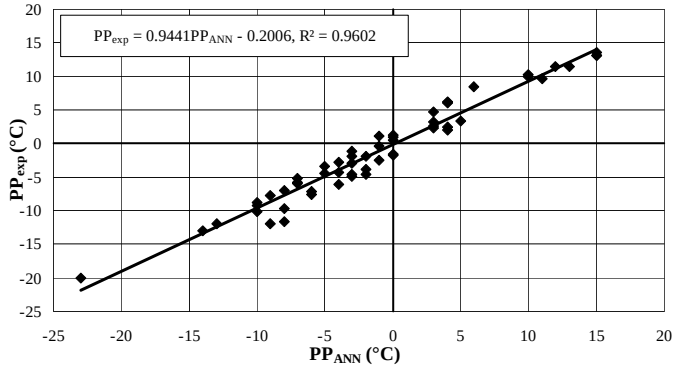

Fig. 3. A comparison of PP temperatures predicted by ANN model with the experimental counterparts.

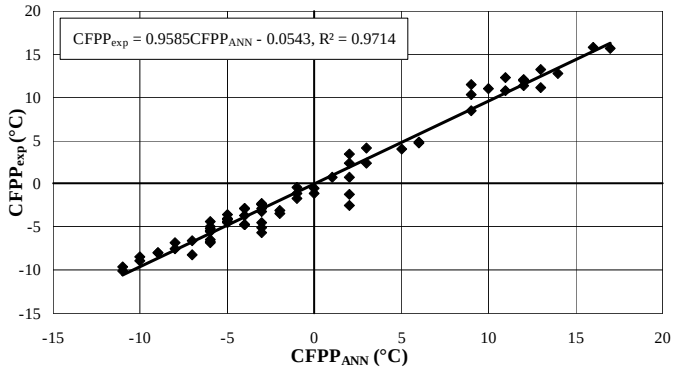

Fig. 4. A comparison of CFPP temperatures predicted ANN model with the experimental counterparts.

The ANN model developed here has not shown any clear evidence for an outlier. Consequently, one can estimate adequately the CFP of biodiesel depending on FA composition of a feedstock by implementing the ANN model. Saturated FA components were expected to have negative influences on CFP, nevertheless both C12:0 and C14:0 had positive influences. Since ANN is capable to recognize patterns whether there is a linear or nonlinear relation in the data available, prediction of CFP by ANN model yields a better estimation than the traditional statistical models such as multiple linear regression (MLR) in which R^2 for CP, PP and CFPP were evaluated as 0.91, 0.84 and 0.88, respectively, while the standard error of estimation for CP, PP and CPPP were 3.4, 2.2 and 2.6, respectively. Hence, it confirms the advantage of ANN modelling which can recognize these nonlinearities and predicts CFP temperatures more accurately.

4. Conclusions

WFOME and RCOME were waxed at higher temperatures than the commercial diesel fuel. CFP of WFOME occurred at higher temperatures than that of RCOME because of its higher saturated FA compositions. The three CFP temperatures, i.e., CP, PP and CFPP were measured as 15 ˚C, 11 ˚C and 12 ˚C, respectively for WFOME whereas -3.5 ˚C, -10 ˚C and -7.5 ˚C, respectively for RCOME. To this end, an estimation of the temperatures of CFP of a potential biodiesel fuel from a new feedstock in prior to the experimental work would be valuable and also would shorten the practical effort.

When the ANN model with 9-6-3 back propagation algorithm was implemented, the CP, PP and CFPP temperatures for WFOME were predicted as 13.5˚C, 11.0˚C and 10.9 ˚C, respectively and for RCOME they were estimated as -3.6 ˚C, -9.5 ˚C and -6.8 ˚C, respectively. It was noted that CFP of the biodiesel was positively influenced by unsaturated FA; however, negatively influenced by saturated FA. Nevertheless, palmitic (C12:0), stearic (C14:0), and gondoeic (C20:1) acids added an opposite influence on CFP. Since the ANN could be trained from iterations, a high accuracy was achieved even some nonlinearities were exist. When compared with the experimental data, the ANN model predicted the CP, PP and CFPP temperatures within 98%, 94% and 96% accuracy, respectively.

As a faster alternative to the time consuming experimental procedures, the model developed here may be used to estimate the CFP temperatures of a biodiesel produced from different feedstock with known FA compositions. It would be a challenging study to employ the models to arrange the FA compositions of a batch of feedstock that would yield exactly the CFP temperatures needed for a regional climate.

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