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Surface tension prediction of vegetable oils using artificial neural networks and multiple linear regression

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Abstract

The surface tension is one of the main properties for characterization of the quality of the fuel atomization process for its use in a diesel engine. There is a lack of published information about the values of surface tension of vegetable oils. The aim of this research is to obtain a mathematical model based on physical properties that establishes a relationship between the surface tensions of different vegetable oils and their fatty acid composition. For this reason, from literature reports, experimental data of oils related to the surface tensions was collected. Knowing that surface tension as a function of temperature, a total amount of 15 oils from different feedstocks at 20 °*C* was selected. The obtained models were developed based in the use of artificial neural networks and multiple linear regressions fits, based on the experimental data available in the literature. Also, the obtained models present a good correlation between surface tension and the fatty acid composition, with a 95 % of confidence interval and coefficient of correlation higher than 0,95. The coefficient of correlation obtained shown a high correlation between the analyzed variables. According to the obtained results, the proposed models are a useful tool for the surface tension estimation from the oils fatty acid composition.

Keywords: Surface tension, fatty acid, biofuels, spray, performance, engine

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1. Introduction

The world is presently confronted with the twin crises of fossil fuel depletion and environmental degradation [1].The majority of mankind's current energy demand is satisfied from petrochemical, coal and natural gas sources, which are all non-renewable resources and are estimated to approach depletion within next 50–100 years [2]. During the last century, the worldwide energy consumption showed more than 20-fold increase and is expected to continue its growth in the future [2]. Thus, the search for alternative sources of renewable and sustainable energy has gained importance with the potential to solve many current social issues such as the rising price of petroleum crude and environmental concerns like air pollution and global warming caused by combustion of fossil fuels [3].

According to the limited resources of fossil fuels, rising crude oil prices and the increasing concerns for environment, there has been renewed focus on vegetable oils and animal fats as an alternative to petroleum fuels [4]. Small scale use of vegetable oils is also considered an interesting option because they can be obtained from agricultural or industrial sources with simple processing [5].

The major disadvantages of the use of vegetable oils as alternative fuel are the different in order of magnitude between the physicochemical properties of these compared to diesel fuel. Kinematic viscosity as well a high surface tension could significantly influence the injection process as well as the process of mixture formation [6]. The influence of these fuel properties in the injection process, combustion y emissions of the engine are studied and reported [7] [4] [8] [9]. Other authors [10] [11] [12] emphasizes the importance and the strong influence of the surface tension in the drops formations, as well as on the atomization properties related to the quality of ignition.

The vegetable oils are obtained from different feedstock available around the world. Therefore, different oils properties are expected. These differences can be attributed to the differences in the fatty acid composition. The fatty acids vary in their carbon chain length and in the number of double bonds or unsaturations [13]. Surface tension (*ST*) is a physical property that is closely related to the molecular structure [10]. Both the length of the fatty acid hydrocarbon chain and the number of unsaturations affect the surface tension [10]. Also, the surface tension is increased with an increase in the chain length [14].

Several models and a computer program for estimation of some physical properties of biofuels from their fatty acid compositions can be found in the literature. E.g., the viscosity model by Allen et al. [15], the cetane number model development by Sánchez et al. [16] and Piloto et al. [17], and a computer program development by Yuan et al. [18] to predict a normal boiling point, critical points, vapor pressure, enthalpy, heat of vaporization, viscosity and surface tension. Also, there exist models to predict the surface tension based on their fatty acid composition, but they only had been development for biodiesel fuels. In 1999, Allen et al. [19] published a model to predict the surface tension of biodiesel from 15 feedstocks; Shu et al. [10] in 2008 published a paper to predict the surface tension of biodiesel fuels by a mixture topological index method.

The mentioned models for estimation of oils and derived biofuel's physical properties had been developed using different methods as Multiple Linear Regression (MLR) techniques, Artificial Neural Networks (ANNs) and others. The MLR procedure requires the user to specify a priori a mathematical model to fit the data in order to obtain the empirical correlation, but an alternative to avoid that problem is the use of artificial neural networks [17]. Unlike the correlation techniques, the neural network can identify and learn the correlative patterns between the input and output data once a training set is provided [17].

Different methods to measure the surface tension of liquids exists, such as the capillary rise method, the drop weight method and its variant, the drop volume method, the pendant drop method and the ring method among others [20]. However, in spite of the simplicity and suitability of several methods, always are necessary laboratory instrumentations and special laboratory conditions. For this reason, in the case of

oils and its derived biofuels, the obtaining of mathematical models to predict the surface tension through its fatty acid compositions is a good solution.

The aim of this research is to obtain a mathematical model to predict the surface tension of vegetable oils from their fatty acid composition, using artificial neural networks and multiple linear regressions.

2. Methods

In this investigation, 15 vegetable oils from different feedstocks and a pure fatty acid (oleic) were selected for obtaining a mathematical model based on the relationship between the surface tensions of the vegetable oils and their fatty acid composition. The surface tension values of these vegetable oils and pure fatty acids were found in experimental data reports (20°*C*), knowing that the surface tension is a function of temperature. The collected values are in Table 1 shown.

Table 1. Values of surface tension (*ST*) reported in the literature at 20°*C*

Items	ST values (*mN/m)*	Reference
Oleic acid	32,50 - 33,00	[21-23]
Olive	33,00 - 33,06	[24-26]
Coconut seed	33,40	[21, 24, 25, 27, 28]
Peanut	35,50	[29, 30]
Cottonseed	35,00 - 35,40	[21, 23-25, 27, 28, 30]
Neem	39,00	[31]
Jatropha	31,00	[31]
Rapeseed	32,90 - 33,83	[20, 32, 33]
Sunflower	33,50 - 34,00	[20, 25, 32]
Soybean	33,85 - 33,90	[20, 32]
Corn	33,40 - 33,80	[20, 29, 32]
Grapeseed	33,89 - 33,90	[20, 32]
Castor	39,00	[21, 23, 27, 28]
Mahua	37,00	[31]
Palm Olein	33,20	[28]
Camelina sativa	32,90	[6]

The fatty acid compositions corresponding to the 15 vegetable oils previously presented in Table 1, are presented in Table 2 covering eight fatty acids found in the oils composition. Only eight fatty acids were taken into account because their weight percent (wt. %) are the most representatives in the full composition. The rest of non-representatives fatty acids were gathered in another classification named Other (*Ot*), also used in modeling process. As is shown in Table 2, the fatty acids compositions percent varies among reports for the same oil source.

Table 2. Values of the fatty acid composition [1, 3, 5, 6, 15, 20, 28, 34-46]

Items	Lauric	Myristic	Palmitic	Stearic	Oleic	Ricinoleic	Linoleic	Linolenic
Oleic acid	0	0	0	0	100	0	0	0
Olive	0	0-1,3	7-18,3	1,4-3,6	55,5-84,5	NR	4-19	0,6
Coconut	44-51	13-20,57	7,5-10,5	1-3	5-8,2	NR	1-2,6	0
Peanut	0	0-0,5	6-12,5	2-8,9	37-61	NR	13-41	0,5-1
Cottonseed	0	0-1,5	22-28,7	0,9-5	13-19	NR	50-58	0-0,5
Neem	NR	0,03-0,26	13,6-17,8	14,4–24,1	49,1–61,9	NR	2,3–15,8	NR
Jatropha	0,31	0,1	13,38-14,2	5,44-7	43,1-45,79	NR	32,27-34,4	0,2
Rapeseed	NR	<0,1	3-4,7	1-2	62,2-65,3	NR	19,2-22	8-9
Sunflower	0-0,5	0-0,2	6,7-3,5	1,3-5,9	14-43	NR	44-74	0-0,8
Soybean	NR	<0,1	11,2	2,9	25,2	NR	55,4	5,0
Corn	0	<0,1	9,9-11,4	1,7-3,1	29,1-32,8	NR	53,3-56,8	0,5-1,1
Grapeseed	NR	<0,1	7,2	3,9	20,2	NR	68,4	0,2
Castor	NR	NR	0,7-1,3	0,9-1,2	2,8-5,5	84,2-94	4,2-7,3	0,2-0,5
Mahua	NR	NR	16,0–28,2	20,0–25,1	41,0–51,0	NR	8,9–13,7	NR
Palm Olein	0,13–0,23	0,85-0,91	36,75-40	2,49	43-49,48	NR	11-12,26	0,1-0,54
Camelina sativa	NR	NR	NR	NR	14,1–19,5	NR	18,8 –24	27–34,7

NR: no reported fatty acid compositions values.

The chemical formula and the basic structure of the fatty acids presented in this research are in Table 3 shown. The nine fatty acid listed represent the inputs for the *ST* modeling. The basic structural description for the input fatty acid used in this work (XX:X) covers the information about the number of carbon atoms (XX) and the number on the right (X) represents the number of unsaturations in the molecule [17].

Table 3. Basic chemical structure of the fatty acids

Fatty acid	Nomenclatures	Structure	Type*	Formula
Lauric	La	12:0	S	$C_{12}H_{24}O_2$
Myristic	M	14:0	S	$C_{14}H_{28}O_2$
Palmitic	P	16:0	S	$C_{16}H_{32}O_2$
Stearic	S	18:0	S	$C_{18}H_{36}O_2$
Oleic	O	18:1	US	$C_{18}H_{34}O_2$
Ricinoleic	Ri	18:1	US	$C_{18}H_{34}O_3$
Linoleic	Li	18:2	US	$C_{18}H_{32}O_2$
Linolenic	Ln	18:3	US	$C_{18}H_{30}O_2$

*S, saturated fatty acids; US, unsaturated fatty acids.

In order to obtain models for surface tension estimation from the fatty acid composition, two methods based in the use of ANNs and MLR fits were used. The first step was the implementation of ANNs. The

networks were development using a topology 9:2:1. The input data consists of nine elements, each of the fatty acid presented in Table 1 and Table 2 plus the additional element named *Ot*. The output data covers the experimentally evaluated surface tension. The ANNs used were the multilayer Perceptrons, with one hidden layer.

In the training step two phases were implemented, keeping constant the phase 1 (back propagation) for all the ANNs evaluated. The training was development for 10000 epochs with a learning rate of 0,01. Linear and logistic functions in the range of 0,9 were used as the output functions in different network variants. The phase one was a back propagation (BP) and the second phase was varied among different possibilities: back propagation, conjugate gradient descend (CGD), Levenberg-Marquardt (LM), quick propagation (QP), quasi-Newton (QN) and Delta-bar-Delta (DBD) [47]. Twelve ANNs (9:2:1) were evaluated keeping constant the function for phase 1 (back propagation) and changing phase 2 and the output function [17] . The better ANNs were selected through the lower Absolute Mean Error (AME) and the higher coefficient of correlation (*R*).

On the other hand, to predict the surface tension from their fatty acid composition, a multiple linear regression model was developed. The same surface tension values and fatty acids composition used to ANNs modeling were given as inputs to develop the regression model. In multiple linear regression, the surface tension was taken as the dependent variable, while the fatty acids composition were taken as the independent variables. The best MLR model was selected through the lower Absolute Error Mean and the higher coefficient of correlation (*R*).

Finally, when the ANNs and MLR models were obtained, a comparison of both methods was assessed in order to select the best way to predict the surface tension.

3. Results and discussion

3.1 Artificial neural networks

The results from the ANNs in order to correlate the surface tension with the fatty acid compositions from the better combinations between phase 1 and phase 2 are shown in Table 4. The best ANNs (9:2:1) was obtained for back propagation in the phase 1 and quick propagation in the phase 2, as the training algorithm. In this training step (BP-QP) the coefficient of correlation (R = 0,960) is the highest and its absolute mean error (AME = 0,44) is the lowest. The the best learning algorithm and network architecture found for prediction surface tension is shown in Fig 1.

Table 4. Results using the betters ANNs (9:2:1) obtained

Function	Phase1	Phase 2	*R*	AEM
Linear	BP	CGD	0,901	0,68
	BP	LM	0,901	0,57
	BP	QP	0,901	0,95
	BP	DBD	0,912	0,67
Logistical	BP	BP	0,925	0,59
	BP	CGD	0,908	0,68
	BP	LM	0,919	0,51
	BP	QP	0,960	0,44
	BP	QN	0,903	0,79

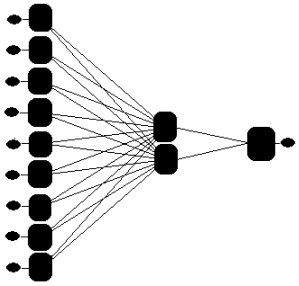

Fig. 1. Basic topology (9:2:1) used in the modeling of the surface tension

The residuals values obtained from the surface tension estimation using ANNs are shown in Table 5. Except for Coconut seed and Mahua, the residuals values are smaller than 1 % for the testing data. According to these result, the ANNs model analyzed shows a good prediction capacity.

Table 5. Comparison of surface tension expected and predicted values from the selected ANNs

	Surface tension (*mN/m*)		
Items	Expected (Literature Values)	Predicted	Residuals
Oleic acid	32,76	32,25	0,51
Olive	33,03	32,89	0,14
Coconut seed	35,40	34,14	1,26
Peanut	35,50	34,83	0,67
Cottonseed	35,20	35,05	0,15
Neem	39,00	38,38	0,62
Jatropha	31,00	31,88	0,88
Rapeseed	33,51	32,75	0,76
Sunflower	33,82	33,78	0,04
Soybean	33,88	34,09	0,21
Corn	33,60	33,58	0,02
Grapeseed	33,90	34,05	0,15
Castor	39,00	39,15	0,15
Mahua	37,00	38,41	1,41
Palm Oleim	33,20	33,15	0,05
Camelina sativa	32,90	32,85	0,05

3.2 Multiple linear regression

To estimate the surface tension from their fatty acid composition a multiple linear regression model was developed using the values showed in Table 1 and Table 2. The MLR model obtained is shown in the following equation.

$$ST = -0{,}201777 \cdot La + 0{,}276565 \cdot P + 0{,}300558 \cdot Ot + 0{,}321542 \cdot O + 0{,}333744 \cdot Li + 0{,}363275 \cdot Ln + 0{,}394795 \cdot Ri + 0{,}6142218 \cdot S + 2{,}00272 \cdot M \quad (1)$$

where *La, P, Ot, O, Li, Ln, Ri, S, M* are the independent variables. A high coefficient of correlation (R = 0,999) indicates the excellent correlation between *ST* and the fatty acid composition. Then, the absolute error mean (AME = 0,69) shows a low mean error for the residuals.

The predicted values using multiple linear regression with the reported values from literature. The results are in Table 6 shown. The residuals values are under 2 %, except for Jatropha oil.

Table 6. Comparison of surface tension of expected and predicted values calculated from Eq. (1)

	Surface tension (*mN/m*)		
Items	Expected (Literatures Values)	Predicted	Residuals
Oleic acid	32,76	32,15	0,61
Olive	33,03	34,64	1,61
Coconut seed	35,40	35,38	0,02
Peanut	35,50	34,37	1,13
Cottonseed	35,20	34,57	0,63
Neem	39,00	37,44	1,56
Jatropha	31,00	33,75	2,75
Rapeseed	33,51	33,14	0,37
Sunflower	33,82	33,83	0,01
Soybean	33,88	33,55	0,33
Corn	33,60	33,25	0,35
Grapeseed	33,90	33,98	0,08
Castor	39,00	39,00	0,00
Mahua	37,00	37,90	0,90
Palm Oleim	33,20	32,70	0,50
Camelina sativa	32,90	33,05	0,15

3.3 Comparison among ANN and MLR prediction capability

A comparison between the *ST* values reported in the literature and the values obtained using ANNs and MLR models are in Fig. 3 shown. The results indicate that both methods are suitable for estimating the *ST* with low residual values. However, coefficient of correlation of MLR model is more higher than the ANNs. On the other hand, the lowest absolute mean error values were obtained with the ANNs.

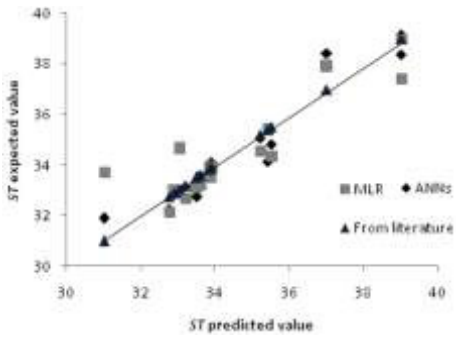

Fig. 3. Comparison between ST predicted using ANNs and MLR

According to the residuals analysis, both models show a good prediction capacity. However, the worst residual values were obtained using MLR model.

It is necessary to point out that the models developed in this research can be used to predict the surface tension of vegetable oils from the fatty acid composition but limited to a number of carbon atoms settled in the range 12-18.

3.4 Further work

The work developed here is an approach in the aim of for the obtaining of a tool that brings good prediction capability for the surface tension. It is demonstrated in this paper that the MLR and the ANNs are both good ways to reach this goal according to the results shown. The most important step in the modeling that is the experimental validation is still pending due to the necessity of performing experiments for comparison between actual values and the predicted values by both methods and therefore, to evaluate the real residual values and the accuracy of the models. An increasing in the number of data used for the input and output elements is also necessary in order to increase the precision, accuracy and the range of fatty acid composition and carbon number to be applied for.

4. Conclusions

A model to predict the surface tension of vegetable oils using an artificial neural network was obtained. The best neural network for predicting the ST was a back propagation network (9:2:1) using a quick propagation algorithm for the second training step and showing a coefficient of correlation (R = 0.960). A multiple linear regression model was obtained showing a coefficient of correlation (R = 0,999) . Good correlations between the surface tension and the fatty acid composition were found. The models based on artificial neural networks and multiple linear regressions are suitable for estimating the surface tension with good precision based on the low absolute mean error found.

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