

Correlations for Predicting Viscosity of Vegetable Oils and Its Derivatives for Compression Ignition Engines

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Abstract

Vegetable oil and its derivatives as an alternative diesel fuel have become more attractive recently because of its environmental benefits and the fact that they are made from renewable resources. Viscosity is the most significant property to affect the utilization of vegetable oil and its derivatives in the compression ignition engines. This paper presents the existing correlations for predicting the viscosity of vegetable oil and its derivatives for compression ignition engines. According to the parameter considered in the correlations, the empirical correlations can be divided into six groups: correlations as a function of temperature, of proportion, of composition, of temperature and composition, of temperature and proportion, and of fuel properties. Out of physical properties of fuel, there exist in the literature several parameters for giving the influence on kinematic viscosity such as density, specific gravity, the ratio of iodine value over the saponification value, higher heating value, flash point and pressure. The study for the verification of applicability of existing correlations to non-edible vegetable oil and its derivatives is required.

1. Introduction

Due to the shortage of petroleum diesel fuel and its increasing cost, an alternative source of fuel for diesel is very much required. Several vegetable oils and its derivatives are known to have the potential for use as the alternative diesel fuel. Vegetable oils and its derivatives in this study mean four different kinds of fuel such as neat vegetable oil, vegetable oil and diesel blends, neat biodiesel, and biodiesel and diesel blends.

Biodiesel is an alternative diesel fuel derived from vegetable oils or animal fats. The transesterification of a vegetable oil or animal fat with a monohydric alcohol, in most cases methanol, yields the corresponding mono-alkyl esters, which are defined as

biodiesel⁽¹⁾.

Vegetable oils can be grouped as edible and non-edible oils. The use of edible vegetable oils or biodiesels from them as a substitute for diesel fuel may lead to a problem of self-sufficiency in vegetable production. Therefore, non-edible oils instead of edible vegetable oils as a substitute for diesel fuel are getting a renewed attention because of global reduction of green house gases and concerns for long-term food and energy security⁽²⁾.

In addition, the European Union has issued Directive 2003/30EC, which mandates the use of biofuels in a percentage ranging from 2% in 2005 to 5.75% in 2010, and 20% in 2020 for all transportation fuels marketed within the member country, and it is expected that a significant portion of this amount will be biodiesel^(3,4). The cost of producing biodiesel is, however, greater than that of normal diesel fuel and biodiesel is often only competitive because it is tax-favoured.

One of the main concerns in direct injection com-

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pression ignition engine research is the fuel-air mixture process which is strongly influenced by the spray characteristics of fuel. It is well known that viscosity, density and surface tension are the main physical properties of a liquid fuel that affect its atomization in a compression ignition engines. Out of three physical properties of vegetable oils and its derivatives, viscosity is one of the most important properties to affect the spray characteristics.

Because of relatively low cetane number and high viscosity of the vegetable oil, particularly non-edible vegetable oil compared to diesel, several difficulties in diesel engines such as engine chocking, cease of fuel injector, gum formation and piston sticking under long term use are noted⁽⁵⁾.

The most widely known and accepted models for viscosity prediction of hydrocarbon and their mixtures were reviewed by Mehrotra et al.⁽⁶⁾. They found that there is no widely accepted simple theoretical method for liquid viscosity so that the methods were categorized as semi-theoretical or empirical one. Allen et al.⁽⁷⁾ introduced the several models for predicting the viscosity of fatty acids. They pointed out that those methods were applicable to individual fatty acid components but not to acyl esters or mixtures. Krisnangkura et al.⁽⁸⁾ explained the various methods for estimation of viscosity of liquid. However, they discussed the methods applicable for fatty acids and for fatty acyl esters as well as the methods for pure hydrocarbon systems. Recently, Ejim et al.⁽⁹⁾ introduced the prediction methods of viscosity in performing an analytical comparison of atomization characteristics of 7 biodiesels and 17 binary and ternary blends with diesel fuel. However, they tested the limited number of methods to evaluate the viscosity of mixtures.

If the temperature which the viscosity of vegetable oil or vegetable oil and diesel blends becomes close to the viscosity of diesel fuel can be predicted, the preheating temperature of vegetable oil before injecting it into the engine can be selected in advance. In addition, when the blended fuel are applied to the diesel engine, blending ratio based on the prediction of viscosity can be selected

The purpose of this work is to classify the existing empirical correlations for the prediction of viscosity for vegetable oils and its derivatives and to suggest the future studies on the prediction of viscosities of non-edible vegetable oils and its derivatives.

2. Correlations for viscosity prediction

There exist in the literature several empirical correlations for predicting the viscosity of vegetable oil and its derivatives. Based on the parameters considered in the correlations, the existing correlations can be classified into six groups as a function: of temperature, of proportion, of composition, of temperature and composition, of temperature and proportion, and of fuel properties.

2.1 Correlations with temperature

The simplest empirical correlation for the variation of liquid dynamic viscosity with temperature is the following correlation known as the Andrade equation.

$$\ln \mu = A + B/T \quad (1)$$

where A and B are constants, μ is the dynamic viscosity in mPa s, T is absolute temperature in K. This correlation was developed for pure hydrocarbons and further utilized for their mixtures.

Recently, the application of empirical correlation Eq. (1) for the prediction of temperature dependent viscosities of biodiesel in the range of 258-303 K was reported by Kerschbaum and Rinke⁽¹⁰⁾. For four different biodiesels selected in their study, $A = -6.1$ and $B = 2359$ were obtained. Initially, exponential equations for the dynamic viscosity derived from experiment were derived for four biodiesel fuels separately. They obtained the exponential equation by computing the mean values for slope and offset of the exponents. It should be pointed out that this correlation describes as a first approximation the dynamic viscosities of the four different biodiesel fuels for temperatures above 273 K. Even though they also suggest the dynamic viscosities of biodiesel

fuels for temperature below 273 K, they failed to generate an empirical correlation like the above one valid for all four biodiesel fuels.

Even though Eq. (1) has been successful from the freezing to boiling points for many liquids, it does not, however, include the effect of pressure, which has resulted in several modifications, mostly in the form of density dependence. A third parameter C was, therefore added to Eq. (1) so that the following correlation known as the Vogel equation was obtained.

$$\ln \mu = A + B/(T + C) \quad (2)$$

Values of constants A, B and C in Eq. (2) have published for seven vegetable oils and eight fatty acids by Nouredini et al.⁽¹¹⁾, for four biodiesel fuels and twelve its blends with diesel fuel by Yuan et al.⁽¹²⁾, and four triglycerides by Goodrum and Eiteman⁽¹³⁾ selected in their study, respectively. It should be noted that in Eq. (2), Yuan et al.⁽¹²⁾ had used the kinematic viscosity in mm²/s instead of dynamic viscosity.

Based on Eq. (2), Dutt and Prasad⁽¹⁴⁾ suggested the generalized correlation for the prediction of dynamic viscosity of fatty oil with the variation of temperature. The generalized values of A, B and C were -0.6298, 273.66 and 88.81, respectively. In Eq. (2), μ was the dynamic viscosity in centipoises and T was the temperature in °C. They state that the result of the above correlation predict viscosity of vegetable oils with an average absolute deviation of 14.5%.

The correlation for the predicting the viscosities of both soybean oil biodiesel and diesel fuel, and for their blends were proposed by Tat and Van Gerpen⁽¹⁵⁾ as follows.

$$\ln \nu = A + B/T + C/T^2 \quad (3)$$

where A, B and C are constants for the fluid, T is the temperature in K, and ν is the kinematic viscosity in cSt (mm²/s). The coefficients for neat soybean oil biodiesel, 75, 50 and 20% biodiesels with No. 2 diesel fuel, and 75, 50 and 20% biodiesel with No. 1 diesel fuel were given as well as standard deviation. It is noted that coefficient table includes the lowest

temperature at which repeatable viscosity data could be collected before the onset of crystallization. The correlations are valid from this temperature to 100°C.

Tate et al.⁽¹⁶⁾ verified the prediction models proposed by Tat and Van Gerpen⁽¹⁵⁾, Eq. (3) and Yuan et al.⁽¹²⁾, Eq. (2) over the temperature range of 20 to 300°C for three biodiesels selected in their study which is a temperature just below the boiling point of the lightest oil fraction. They found that the predicted results by Eq. (3) show good agreement with the experimental results. However, Eq. (2) overestimated the kinematic viscosity when T < 80°C and underestimated it when T > 80°C.

2.2 Correlations with proportion

The empirical correlation for the prediction of kinematic viscosities of vegetable oil-diesel blends, i.e. five soybean oil-diesel blends, five sunflower oil-diesel blends and neat No. 2 diesel fuel based on the data measured at 85°C, the typical temperature of fuel in a diesel injection nozzle, was suggested by Msipa et al.⁽¹⁷⁾

$$\ln \nu = 0.2488 + 0.019477\chi \quad (4)$$

where χ is percentage of vegetable oil and ν is the kinematic viscosity in mm²/s.

They found that the viscosities for the sunflower oil/diesel blends were almost identical to those for soybean oil/diesel blends. In addition, the neat vegetable oils were nearly 900% more viscous than the diesel fuel at 85°C.

In addition, Tat and Van Gerpen⁽¹⁵⁾ applied the mixing equation similar in form to one originally proposed by Arrhenius and described by Grunberg and Nissan⁽¹⁸⁾ to the measured biodiesel and the No. 2 and No. 1 diesel fuel data to correlate the kinematic viscosities of the 75, 50, and 20% biodiesel blends with No. 2 and No.1 diesel fuels. As a result, a blending equation that allows the kinematic viscosity to be calculated as a function of the biodiesel fraction was suggested as

$$\log \nu_B = m_1 \log \nu_1 + m_2 \log \nu_2 \quad (5)$$

where ν_B is the kinematic viscosity of the blend, m_1

and ν_1 are the mass fraction and the viscosity of component 1, and m_2 and ν_2 are the mass fraction and viscosity of component 2. Mass fraction instead of mole fraction in the original Arrhenius equation was used here because there are usually more readily available. Allen et al.⁽⁷⁾ and Allen and Watts⁽¹⁹⁾ introduced the mass fraction for the comparative analysis of the atomization characteristics of fifteen biodiesel fuel types selected in their study and for the prediction of viscosity of biodiesel fuels from their fatty acid ester composition. Recently, even though the kinematic viscosities predicted from Eq. (5) using the mass fraction were always underpredicted, Yuan et al.⁽¹²⁾ also used the mass fraction to calculate the biodiesel mixture viscosity.

Recently, the empirical correlation as a function of biodiesel fraction was suggested by Alptekin and Canakci⁽²⁰⁾ as follows.

$$\nu = A \chi^2 + B\chi + C \quad (6)$$

where ν is the kinematic viscosity in mm^2/s , A, B and C are coefficients and χ is biodiesel fraction. The calculated viscosity values from Eq. (6) was validated by using the measured viscosity values for all the blends. The maximum absolute error between the measured and calculated values comes from Eq. (6) for biodiesel-diesel fuel blends was 1.58%.

In addition, they suggested the general form of the equation as a function of biodiesel and diesel fuel concentration as follows.

$$\log \nu_B = \nu_1 \log \nu_1 + \nu_2 \log \nu_2 \quad (7)$$

where ν_B is the kinematic viscosity of the blend in mm^2/s , ν_1 and ν_1 are the kinematic viscosity of component 1 and its volume fraction, and ν_2 and ν_2 are the kinematic viscosity of component 2 and its volume fraction. It should be noted that volume fraction instead of mass fraction used by Allen et al.⁽⁷⁾ and Tat and Van Gerpen⁽¹⁵⁾ was used because the estimated values were closer to the measured values by using volume fraction than using the mass fraction. This is related to the mole fraction used in the original equation.

However, Yuan et al.⁽¹²⁾ had compared the pre-

dicted kinematic viscosities from Eq. (5) and Eq. (7) for the calculation of biodiesel mixture viscosity. They found that the kinematic viscosities predicted from Eq. (5) using the mass fraction were always underpredicted. If the volume fraction or mole fraction is used, the viscosities is even more underpredicted since pure biodiesel fuels have higher viscosities with higher densities and larger molecular weight. Therefore, they also used the mass fraction to calculate the kinematic viscosity of biodiesel mixture.

2.3 Correlations with composition

Various studies have shown that the viscosity of vegetable oils or biodiesels depends on the fatty acid composition⁽²¹⁾. The kinematic viscosity of biodiesel is intrinsically related to its ester content⁽⁴⁾. Since the transesterification reaction of an oil or fat leads to a biodiesel fuel corresponding in its fatty acid profiles with that of the parent oil or fat, biodiesel is a mixture of fatty esters with each ester component contributing to the properties of the fuel⁽¹⁾.

The empirical correlation for predicting the dynamic viscosities of biodiesel fuels from the knowledge of their fatty acid composition was developed by Allen et al.⁽⁷⁾

For methyl ester

$$\mu_{me} = 1.05 \times 10^{-4} M^2 - 0.0242M + 2.15 \quad (8)$$

For ethyl ester

$$\mu_{ee} = 1.16 \times 10^{-4} M^2 - 0.0264 M + 2.15 \quad (9)$$

A logarithmic mixture equation is

$$\ln \mu_m = \sum_{i=1}^n y_i \ln \mu_i \quad (10)$$

where μ_{me} and μ_{ee} are the dynamic viscosities for methyl ester and ethyl ester, respectively ($\text{mPa} \cdot \text{s}$), M the molecular weight (g/mol), m viscosity of pure ith component ($\text{mPa} \cdot \text{s}$), y_i mass fraction of the ith component and μ_{me} mean viscosity of mixture ($\text{mPa} \cdot \text{s}$). The viscosity of individual ester components can be calculated by Eqs. (8) and (9) and then viscosity of biodiesel and its blends can be obtained from Eq.

(10) by introducing the results from Eqs. (8) and (9).

This mixture equation was based on the Grunberg-Nissan method, one method among three methods presented by Poling et al.⁽¹⁸⁾ They introduced two assumptions that the interaction parameter can be neglected and the mass fraction was used instead of mol fraction.

Vegetable oils have a different chemical structure and the fatty acids commonly found in vegetable oils vary in their carbon chain length and in number of double bonds. Based on this background, Goering et al.⁽²²⁾ proposed the correlation with two independent variables able to account for most of the differences in viscosity among the eleven vegetable oils selected in their study.

$$\nu = -73.14 + 6.409 \text{ WACL} - 3.028 \text{ WADB}^2 \quad (11)$$

where ν is the kinematic viscosity in mm^2/s at 38°C , WACL the weighted average number of carbon atoms in the fatty acid chains, WADB² the square of the weighted average number of double bonds in the fatty acid.

2.4 Correlations with temperature and composition

A thermodynamic parameter model is proposed by Krisnangkura et al.⁽⁸⁾ for the determination of kinematic viscosities of saturated fatty acid methyl esters (FAMES) of various chain lengths at different temperatures.

For short chain FAME,

$$\ln \nu = -0.158 Z + (492.12/T) + (108.35Z/T) - 2.915 \quad (12)$$

For long chain FAME,

$$\ln \nu = -0.202 Z + (403.66/T) + (109.77Z/T) - 2.177 \quad (13)$$

For free fatty acid

$$\ln \nu = -0.326 Z + (657.12/T) + (173.31Z/T) - 2.496 \quad (14)$$

$$\ln \nu_m = \sum_{i=1}^n y_i \ln \nu_i \quad (15)$$

where Z is the number of carbon atom of the homologous series, T the absolute temperature in K , ν the kinematic viscosity of each component in cSt , ν_m the mean kinematic viscosity of biodiesel in cSt .

In the study on the mixture properties of low molecular weight triglycerides, Goodrum and Eiteman⁽¹³⁾ introduced the following empirical model for the prediction of dynamic viscosities of triglyceride mixture.

$$\mu_m^{0.5} = \sum_{i=1}^n x_i \ln \mu_i^{0.5} \quad (16)$$

where μ is the dynamic viscosity in cP , T is the temperature in $^\circ\text{C}$, A , B and C are fitted parameters, μ_m is the dynamic viscosity of mixture in cP , x is the mole fraction of i th component. In this study, viscosity of individual triglycerides was calculated by Eq. (2) in the above section. It should be pointed out that the powers of dynamic viscosities for pure triglyceride and its mixture are different with Eq. (10).

2.5 Correlations with temperature and proportion

Recently, an empirical correlation was proposed in order to interpolate viscosity to any kind of diesel oil/ neat vegetable oil blend by Abolle et al.⁽²³⁾ The dependence of kinematic viscosity on composition of neat vegetable oil/ diesel fuel blend at each temperature can be expressed as

$$\ln \nu = \alpha \text{ PVO} + \ln \nu_d \quad (17)$$

where ν is the kinematic viscosity of vegetable oil and diesel mixtures in m^2/s , ν_d the kinematic viscosity of the petroleum diesel fuel at temperature T in K , PVO the percentage of vegetable oil in the mixtures in %, α depends on temperature and the nature of the oil to be involved in the fuel.

Parameter α can be modeled as

$$\alpha(T) = aT + \kappa \quad (18)$$

where the mean value of $a = (-1.18 \pm 0.07) \times 10^{-4} (\text{K}^{-1})$, T the absolute temperature in K . The mean value of α can be obtained from

$$\kappa(S) = 1.78 \times 10^{-4} S + 2.53 \times 10^{-2} \quad (19)$$

where S is the saturates sum: palmitic + stearic + arachidic + behenic.

The viscosity of petroleum diesel fuel ν_d (10^{-6} m²/s) in Eq. (17) was modeled versus temperature from

$$\ln \nu_d = -1.84 \times 10^{-2} T + 6.764 \quad (20)$$

They predicted the kinematic viscosity for the six vegetable oils-diesel fuel blend by Eqs. (17)-(20) and compared with the experimental results. It should be noted that this predictive method is valid for any neat vegetable oil-diesel fuel blend for temperatures above the cloud point.

2.6 Correlations with fuel properties

Fuel properties for the spray and combustion analysis of vegetable oils and biodiesels can be grouped conveniently into physical, chemical and thermal properties. Physical properties include viscosity, density, specific gravity, cloud point, pour point, flash point, boiling point, freezing point and refractive index etc. It is known that there are the strong relationships between physical properties of fuel.

There exist in the literature several parameters for giving the influence on kinematic viscosity such as density, specific gravity, the ratio of iodine value over the saponification value, higher heating value, flash point and pressure.

Rodenbush et al.⁽²⁴⁾ developed the correlation related viscosity of vegetable oils to density as follows.

$$\mu^{0.5} = 2405/(\rho-513) \quad (21)$$

where μ is the dynamic viscosity in centipoises and ρ the density in kg/m³.

An advantage of this correlation is that viscosity can be calculated from density data, which are somewhat easier to obtain experimentally than viscosity data, particularly for highly viscous oils. They state that in the absence of this viscosity data, the above correlation can predict the viscosity of a vegetable oil from its density with reasonable accuracy.

The kinematic viscosities of vegetable oils and biodiesels decrease with increase in density. The correlations between viscosity and density was, therefore, suggested by Demirbas⁽²⁵⁾.

For vegetable oils:

$$\nu = -0.7328 \rho + 938.57 \quad (22)$$

For biodiesels:

$$\nu = -16.155 \rho + 930.78 \quad (23)$$

where ν is the kinematic viscosity in cSt and ρ the density in g/l.

These correlations showed the coefficient of regression values of 0.9398 and 0.9902, respectively. The comparative study on viscosities of vegetable oils obtained from Eqs. (21) and (22) are required.

For the application of Eq. (21) to biodiesel and its blends, Yuan et al.⁽¹²⁾ suggested the following empirical correlation.

$$\nu^{0.5} = [B/(SG-A)] \quad (24)$$

where SG and ν represent the specific gravity and kinematic viscosity in mm²/s, respectively of pure biodiesel or diesel fuel. A and B are correlation parameters that are dependent on different biodiesel fuels.

In addition, they suggested the empirical correlations for the evaluation of mixture of biodiesel and diesel as follows.

$$\nu_{mix}^{0.5} = [B_{mix}/(SG_{mix}-A_{mix})] \quad (25)$$

$$A_{mix} = A_{pb} \times F_{pb} + A_d \times F_d \quad (26)$$

$$B_{mix} = B_{pb} \times F_{pb} + B_d \times F_d \quad (27)$$

where A and B are correlation parameters for mixtures with subscripts mix, pb and d representing the mixture, pure biodiesel, and diesel fuel, respectively. The fraction F in this method was mass based fraction for simplicity.

Dutt and Prasad⁽¹⁴⁾ presented the correlation for the prediction of the dynamic viscosity in relation to the ratio of iodine value over the saponification value (IV/SV) as follows.

$$\log \mu = (-1.4 + 1.25IS) + (500 - 375IS) / [(t+140) - 85IS] \quad (28)$$

where μ is the dynamic viscosity in centipoises, t is the temperature in °C and IS is the IV/SV. They state that the result of the above correlation predict viscos-

ity of vegetable oils with an average absolute deviation of 13.0%.

The correlation have been developed to calculate the viscosity of various vegetable oils from their higher heating value by Demirbas⁽²⁶⁾ as follows.

$$\nu = \text{HHV}/0.0491 - 772.81 \quad (29)$$

where ν is the kinematic viscosity of vegetable oil in mm^2/s , HHV the higher heating value of vegetable oil in kJ/g . This correlation was originally developed for the prediction of higher heating value from the viscosity data. The correlation coefficient of the above equation was 0.998 for the vegetable oils.

Recently, correlations for predicting the viscosity from the higher heating values for vegetable oils and biodiesel, respectively were reported by Demirbas⁽²⁵⁾.

For vegetable oils:

$$\nu = \text{HHV}/0.0317 - 1200.41 \quad (30)$$

For biodiesels:

$$\nu = \text{HHV}/0.4625 - 85.297 \quad (31)$$

where ν is the kinematic viscosity of vegetable oil or biodiesel in cSt , and HHV the higher heating value of vegetable oil or biodiesel in MJ/kg . It should be noted that this correlation is also developed for the calculation of higher heating value from the viscosity data of vegetable oil or biodiesel.

It should be pointed out that Eqs. (29) and (30) are proposed by same author in 2000 based on the data for ten vegetable oils and in 2008 based on twenty two vegetable oils selected in their study, respectively. In addition, the slightly different data for density and higher heating values for same vegetable oil were introduced for the calculation.

Viscosity of biodiesel is closely correlated with one of physical properties, flash point. The correlation between viscosity and flash point for biodiesels was proposed as⁽²⁵⁾

$$\nu = 22.981 \text{ FP} + 346.79 \quad (32)$$

where ν is the kinematic viscosity in cSt , and FP the

flash point in K.

The flash point of a liquid is the lowest temperature at which enough vapors are given off to form a mixture that will ignite when exposed to an ignition source. The flash point is an important property for indication of volatility and for storage requirements⁽²⁷⁾.

It is well known that the viscosity of liquids increases considerably with increase in pressure. A rather general rule is that the more complex the molecular structure of the liquid, the larger is the effect of pressure. Two empirical correlations have been proposed relating viscosity of olive oil to pressure by Schaschke et al.⁽²⁸⁾

The exponential curve fit was given by

$$\mu = 89.648 e^{0.013P} \quad (34)$$

while the best-fit quadratic curve is

$$\mu = 0.0162 P^2 + 0.8658P + 89.474 \quad (35)$$

where μ is the dynamic viscosity of liquid in mPa s and P the pressure in MPa

Both correlations provide a good fit with experimental data with the quadratic relationship providing the best fit. The exponential relationship has a standard deviation of 3.32% and R^2 of 0.9972, while the quadratic relationship has a standard deviation of 1.93% and R^2 of 0.9994. However, the applicability of both correlations should be verified for the other vegetable oils or biodiesels than olive oil.

3. Discussion

The existing correlations for estimating viscosity of vegetable oil and its derivatives are reviewed and classified. The predictive methods discussed in the previous section are by no means a complete list of available methods. They are all predictive, but limited in the types of compounds to which it can be applied and also limited to temperatures of the components comprising the mixture. The study for the verification of applicability of predictive correlations discussed here to non-edible vegetable oil and its derivatives is required.

4. Nomenclature

A, B, and C	constants
FP	flash point (K)
HHV	higher heating value (MJ/kg, kJ/g)
M	molecular weight(g/mol)
m	mass fraction
P	pressure (MPa)
PVO	percentage of vegetable oil(%)
T	temperature (°C, K)
Tr	reduced temperature (T/Tc)
Tc	critical temperature (K)
S	saturates sum
v	volume fraction
x	mole fraction
y	mass fraction
Z	number of carbon atom

Greek

α	parameter
κ	parameter
μ	dynamic viscosity (mPa · s, cP)
ν	kinematic viscosity (mm ² /s)
χ	biodiesel fraction
ρ	density (kg/m ³)

Subscripts

B	blend
d	diesel
<i>i</i>	component <i>i</i>
m	mixture

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