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Predicting Temperature Dependent Viscosity for Unaltered Waste Soybean Oil Blended with Petroleum Fuels

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Abstract Using vegetable oil based alternative fuels for diesel engines has grown in interest over recent years due to the rising cost of petroleum products and instability in the energy marketplace. One of the major hurdles to overcome in using vegetable oil as a diesel fuel is high viscosity. Here, we experimentally determine the viscosity of unaltered waste soybean oil (WSO) blended with petroleum fuels. Three blend viscosity models Arrhenius, Wright, and the ASTM D7152-05 Standard were evaluated for viscosity prediction accuracy over a temperature range of −10 to 40 °C. Results indicated that the Arrhenius method using volume fractions was the most accurate predictor of viscosity for binary blends made of WSO and diesel (2.31% absolute average deviation) as well as multi-component blends made from WSO, diesel, kerosene, and gasoline (8.72% absolute average deviation). An intermolecular interaction correction factor was empirically determined for each model in an effort to improve prediction accuracy for the multi-component blends. Using the correction constants improved the absolute average deviation for the Arrhenius method to 6.85%, 5.87% for the Wright method based on mass fractions, and 9.67% for the ASTM method based on mass fractions. The use of this correlation constant for the Arrhenius method was only helpful for blends containing more that 30% WSO, indicating that molecular interaction behavior only deviates significantly from ideality at these higher WSO fractions.

Keywords Kinematic viscosity · Diesel fuel · Alternative fuels · Viscosity modeling · Vegetable oil · Waste vegetable oil

Introduction

The possibility of using vegetable oil as a fuel in diesel engines has always existed. In fact, the original diesel engine by Rudolph Diesel was designed to run on peanut oil in an effort to solve the problem of a limited petroleum fuel supply for farmers [1]. As petroleum based fuels became easier to obtain and more economical, diesel engine designs focused on using these fuels instead of vegetable oil. In recent years, the use of vegetable oil based fuels has once again gained interest [2] due to sharp increases in crude oil prices and the detrimental effect that petroleum fuels have on the environment. However, current diesel automotive technology does not match well to widespread use of vegetable oil as fuel due to its high viscosity and high gel-point temperature.

One solution to the high viscosity and gel-point is to chemically alter the vegetable oil through a synthesis process where the lipid triglyceride (oil) molecule is broken apart at the carbonyl functional group on each fatty acid chain connected to the triglyceride backbone through nucleophilic attack with methoxide [3]. The end result of this base catalyzed transesterification reaction is the creation of four lower molecular weight molecules, glycerol and three methyl esters. This allows easy removal of the highly viscous glycerol compound so that less viscous methyl esters can be used as fuel. The resulting biodiesel

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fuel can be used directly in a vehicle or as a percent component in petroleum diesel fuel. Most commercially available biodiesel is between 5 and 20% biodiesel blended with petroleum diesel [4]. The main drawback to creating 100% biodiesel fuel is the limited supply of vegetable oil due to insufficient agricultural production to supply the world's need for both food and raw material for biodiesel production. As a result the price for 100% biodiesel is higher than petroleum diesel fuel. For example, in June of 2009, the price for 100% biodiesel was $3.50/gallon compared to diesel at $2.53/gallon [5, 6]. While operating a diesel engine on 100% biodiesel is possible, the commercialization likelihood is unlikely due to the economics and the limited supply of vegetable oil [7, 8].

An alternative approach to creating a diesel biofuel is to blend chemically unaltered vegetable oil with petroleum fuels. Blending reduces the viscosity and gel-point allowing more appropriate flow through the fuel system, fuel injector spray patterns and atomization of the fuel. The advantage of such an approach is that fuel processing only requires mixing of the vegetable oil and diesel. If waste vegetable oil is used from a deep fryer, the oil must be de-watered and filtered before mixing. There is no significant waste created, such as glycerol, when blending and no hazardous reagents, such as methanol and sodium hydroxide, are required. Cost of creating this type of fuel is generally based on the price of the oil used. For example, if refined deodorized bleached (RDB) vegetable oil is used in the blend, commonly referred to as straight vegetable oil (SVO), no processing other than mixing is required. Therefore, the blend fuel price is based simply on the raw material costs and will typically be comparable to petroleum diesel fuel. If waste vegetable oil (WVO) is used, the raw material costs are significantly lower. However, the WVO must be heated to drive off water and filtered before use. In our work, we have been able to obtain large quantities of waste soybean oil (WSO) for free and our processing costs are approximately $0.30/gallon. There have been a number of reports and evidence indicating good engine power and acceptable emissions with diesel engines operating on SVO blended with petroleum diesel [9–14]. In our own research, we have operated six different diesel vehicles on WVO-petroleum fuel blends for a combined total of over 30,000 miles [15]. In that time, we have not experienced any engine problems or difficulty in moving the blended fuel through the vehicle fuel supply system.

Since viscosity plays a critical role in determining the percent loading of unaltered vegetable oil that can be blended with petroleum fuels, it is useful to model the viscosity of a potential blend composition over a wide range of ambient temperatures before creating and using the fuel in diesel engines. The purpose of this research is to investigate and evaluate three mathematical models for predicting viscosities of WSO blended with diesel, kerosene and gasoline fuels. The ability to accurately predict viscosity based on the fraction of each component in the fuel allows one to model and design a fuel blend with appropriate viscosity characteristics for a specific application.

There have been many theoretical models created for predicting the viscosity of blended liquids. The earliest and most fundamental of these dates back to the equation developed by Arrhenius, as discussed by Grunberg and Nissan, where the sum of the logarithmic viscosity of each blend multiplied by its mole fraction equals the log of the overall blend viscosity [16].

$$\log v_b = N_1 \log v_1 + N_2 \log v_2 \qquad (1)$$

where v_b is the viscosity of the blend, N_1 and v_1 are the mole fraction and viscosity of component 1, N_2 and v_2 are the mole fraction and viscosity of component 2 at the same temperature. This equation often referred to as the semi-log blending model, can be expanded for multi-component blends. Depending on the nature of the blend components, mole, mass, or volume fractions can be used in viscosity blend calculations. Recent reports have investigated the ability of this equation to predict viscosities of biodiesel blended with petroleum diesel [17–20]. However, no studies investigating viscosity modeling accuracy for binary vegetable oil–diesel blends or for multiple component blends with vegetable oil have been reported.

Other well known and utilized viscosity blend models also use mass or volume fractions of each component. For example, the ASTM D7152-05 Standard uses mathematically transformed viscosity values for each component at a given temperature [21]. The transformed viscosities are multiplied by the loading fraction of the respective component and then summed together. Inverse transformation of this value produces the blend viscosity. When mass fractions are used, it is referred to as the Modified ASTM method. The Wright method, also referenced in the ASTM D7152-05 Standard, uses transformed viscosity data as well as transformed temperature values to create an empirical viscosity–temperature relationship (slope) over a finite temperature range for each component [21]. These values are then used, along with the fraction base for each component, to calculate the transformed blend value. Finally, the blend viscosity value is obtained by mathematical untransformation. As in the ASTM Standard method, if mass fractions are used, it is referred to as the Modified Wright method.

The use of the Arrhenius, ASTM and Wright blend viscosity models are well known and accepted models that have generally been employed on low viscosity blend components all with similar viscosities. However, the extreme difference in viscosity between vegetable oil and petroleum fuels makes it more difficult to predict blend viscosity and it is not clear which method would produce the most accurate results. Here we investigate the predictive ability of the Arrhenius semi-log blending model, the ASTM 7152 Standard, and the Wright method on chemically unaltered waste soybean oil blended with petroleum fuels. The results are compared, and a modification of each method to improve predictive accuracy is investigated and discussed.

Materials and Methods

The work described here consisted of two sequential sets of experiments. In the first set of experiments, the predictive capability of the viscosity blend models was investigated for two component WSO–diesel blends. Viscosity values were obtained for each component and then used in the mathematical models to predict viscosity at the same temperatures for the WSO–diesel blends. Results from these experiments were evaluated to determine which predictive models should be subsequently employed to predict viscosity for more complicated, multi-component blends of WSO, diesel, kerosene and gasoline. While binary blends of WSO and diesel are practically possible, the resulting viscosity is high. Therefore, the percent of WSO that can be used in such a blend is limited. By adding in other low viscosity components, such as kerosene, the WSO percent can be maximized. Thus, the second set of experiments evaluated performance of viscosity prediction models on blends created from WSO, diesel, kerosene, and gasoline. Again, viscosity data was obtained for each component at a range of temperatures. The temperature–viscosity correlation for each component was then fit for slope (*B*) and intercept (*A*) values as shown in Eq. 2.

$$\log v = A - B(T) \qquad (2)$$

The viscosity (v) is in centistokes (cSt), and A and B are the intercept and slope of the straight line formed when $\log v$ is plotted against temperature, T, in Kelvin. Interpolated slope and intercept data (constants B and A) for each of the individual components were used to calculate the viscosity of each component at a given temperature. The interpolated viscosity values were then used in the mathematical models to predict blend viscosity correlation to temperature from −10 to 40 °C.

All viscosity measurement were conducted with calibrated Cannon–Fenske kinematic viscometers in accordance with the ASTM D445-88 Standard [22]. Viscosity measurements for each sample were taken at 10 °C intervals and repeated five times at each temperature. The temperature range of interest was from 40 °C down to the lowest temperature obtainable without clouding or gelling of the sample. The viscometer water bath was insulated, except for a viewing window to observe viscometer levels, and temperature was regulated by a water chiller/heater. Ethylene glycol was added to the bath as needed to avoid freezing. For diesel, kerosene, and gasoline, the lowest temperature for viscosity measurement was −10 °C, and for pure WSO it was 10 °C. It should be noted that the highest temperature measurement for pure gasoline was 30 °C, due to its volatility. The two component blends created from diesel and WSO were measured down to 0 °C. For multi-component blends, the lowest temperature for viscosity measurements was −10 °C. Viscosity measurements above 40 °C were deemed unnecessary since ASTM diesel fuel viscosity standards are measured at 40 °C. Further, viscosity is a non-issue at higher temperatures in diesel engines. Diesel, kerosene, and regular unleaded gasoline were obtained from commercial fueling stations. Waste soybean oil, provided by a local restaurant, was stored undisturbed for 1–2 weeks to settle out large particulate material. The oil was decanted and heated to approximately 65 °C for a time period long enough to drive off water and lower the content to no more than 0.25%. Water content was determined by calculating mass loss of a representative 300-g sample after heating it to 250 °F for 15 min. The oil was then passed through a 1-µm polymicro microfiber filter bag a minimum of two times before use [23]. The WSO free fatty acid content, determined through base titration, ranged from 3 to 9%. Since the mathematical models for predicting blend viscosity use both volume or mass fractions, the density of each component was experimentally obtained (Table 1). Microsoft Excel was used to record all data and analyze the results.

Results and Discussion

In part one of these experiments, four WSO–diesel blends were created for analysis. The fraction ratios selected were based on providing a wide range of WSO–diesel blends to comprehensively evaluate the viscosity models. The maximum amount of WSO was limited to 50%. Any blend that contains a WSO loading higher than 50% will not produce a viscosity that is acceptable for fuel use at typical atmospheric temperatures. The experimentally determined viscosities for theses binary blends and the predicted values are shown in Table 2. Without exception, both the Wright and ASTM methods consistently under predicted the experimental viscosity suggesting that the actual molecular interactions between diesel and WSO are stronger than ideal behavior. The Arrhenius volume fraction base method

Table 1 Molecular weight and density of blend components

	WSO	Diesel	Kerosene	Gasoline
Molar mass (g/mol)	900	230	170	103
Density (g/mL)	0.9117 ± 0.0024	0.8376 ± 0.0025	0.7995 ± 0.0008	0.7407 ± 0.0024

Table 2 Comparison of experimental and predicted viscosities (cSt) of WSO–diesel blends

Percent WSO	Temperature (°C)	Experiment (±STD)	Arrhenius (mass)	Arrhenius (volume)	Wright	Wright (modified)	ASTM	ASTM (modified)
50	0	40.87 ± 1.1	45.16	41.81	33.10	35.75	29.73	29.73
	10	25.73 ± 2.49	28.04	26.28	22.29	23.77	19.12	20.36
	20	18.49 ± 0.55	19.55	18.38	15.56	16.55	13.44	14.27
	30	13.36 ± 1.63	13.73	12.96	11.12	11.78	9.71	10.26
	40	10.05 ± 1.33	10.39	10.19	8.79	8.96	7.72	7.86
35	0	24.65 ± 3.62	26.00	24.21	19.65	20.98	16.94	17.98
	10	16.92 ± 2.42	17.65	16.62	14.34	15.16	12.63	13.30
	20	11.93 ± 0.54	12.55	11.86	10.22	10.78	9.06	9.51
	30	9.07 ± 1.61	9.10	8.63	7.53	7.91	6.73	7.04
	40	6.96 ± 0.65	7.02	6.90	6.05	6.15	5.44	5.52
25	0	18.16 ± 0.45	17.85	16.82	14.20	14.95	12.67	13.25
	10	12.02 ± 1.57	12.87	12.25	10.86	11.35	9.85	10.24
	20	9.14 ± 0.8	9.28	8.85	7.84	8.18	7.15	7.42
	30	6.74 ± 1.31	6.87	6.58	5.89	6.12	5.40	5.59
	40	5.35 ± 0.72	5.40	5.32	4.79	4.85	4.41	4.46
15	0	12.68 ± 0.9	12.17	11.69	10.43	10.79	9.70	9.98
	10	8.9 ± 0.63	9.34	9.03	7.32	8.57	7.82	8.02
	20	6.68 ± 1.26	6.83	6.61	5.39	6.26	5.74	5.88
	30	5.03 ± 3.93	5.17	5.01	4.16	4.78	4.40	4.50
	40	4.08 ± 1.73	4.15	4.11	3.43	3.86	3.63	3.65

produced the best prediction values and the mass fraction, being nearly as accurate, typically over predicted the viscosity. The percent absolute average deviations (AAD) for each model from the corresponding experimental values are shown in Table 3. Since the Modified Wright model using mass fractions was significantly better at prediction than with volume, use of volume fractions was not further investigated. This finding was also true for the ASTM method. However, since the Arrhenius method using either mass or volume yielded similar results, both fraction bases were kept for further analysis in part two of these experiments.

In part two of these experiments, four different multicomponent blends ratios were created for analysis (Table 4). As before, a wide range of WSO, diesel, kerosene and gasoline loading fractions were used for the blends to comprehensively evaluate the viscosity models and the maximum amount WSO was limited to 50%. In addition, the amount of gasoline was limited to no more than 10%, due to its low cetane value and high auto ignition temperature. Interpolated slope and intercept data (constants *B* and *A* in Table 5) for each of the individual components were used to determine the viscosity of each component at a given temperature. Theses values were then used in the prediction models. Calculated blend viscosity values are shown along with the experimental results in Table 6. Finally, the percent AAD for each model is shown in Table 7.

A review of the results presented in Tables 4 and 7 indicates that the Arrhenius method based on volume fractions was the best overall predictor of viscosity for all WSO blends created in this study. However, the percent AAD was greater for the multi-component blend predictions than for the simple oil–diesel blends. The increase in error may be due to the fact that interpolated component viscosity data was used for the multi-component blend prediction rather than the exact experimental values, as in the WSO–diesel blend predictions. However, the

Table 3 WSO–diesel blend average absolute deviation for each prediction method

Percent WSO	AAD%					
	Arrhenius (mass)	Arrhenius (volume)	Wright	Wright (modified)	ASTM	ASTM (modified)
50	6.26	1.88	15.51	10.67	26.15	23.17
35	3.23	1.98	15.99	11.88	25.68	22.38
25	2.62	3.10	13.80	10.48	21.57	18.92
15	3.05	2.28	17.65	7.13	14.69	12.89
Average	3.79	2.31	15.74	10.04	22.02	19.34

Table 4 Multi-component blend compositions

Blend[a,b]	Waste oil	Diesel	Kerosene	Gasoline
O50DKG	50.0	20.0	20.0	10.0
O37DKG	37.0	37.0	17.0	9.0
O25DKG	25.0	55.0	15.0	5.0
O10DKG	10.0	80.0	8.0	2.0

[a] All values are percent composition by volume

[b] Blend notation: *O*-WSO followed by the percent WSO, *D*-diesel, *K*-kerosene, *G*-gasoline

Table 5 Equation 2 slope (*B*) and intercept (*A*) values for pure fuel components used for predicting the multi-component fuel blends

Fuel component	Coefficient[a]		R^2
	A	*B*	
Diesel	9.12	−0.0264	0.992
WSO	17.5	−0.0449	0.990
Kerosene	6.45	−0.0191	0.983
Gasoline	2.54	−0.0101	0.998

[a] The coefficients *A* and *B* represent the curve intercept and slope, respectively when viscosity, in cSt, is plotted on the *y*-axis and temperature, in Kelvin, is plotted on the *x*-axis

correlation coefficients for line fitting of the individual component temperature–viscosity curves were excellent (Table 5). Further, when the true experimental values were used in place of the interpolated data for multi-component blend viscosity predictions, a minimal improvement in prediction error was realized.

Another possible explanation is that the molecular interactions between oil, diesel, kerosene, and gasoline all combined together are further away from ideal behavior compared to only oil and diesel molecular interactions. In past literature many cases were cited where a molecular interaction correction factor was experimentally determined and employed in attempt to minimize the prediction error for blend viscosity [18, 19, 23–25]. One of the first uses of such corrections was reported by Grunberg and Nissan [16]. More recently, Yuan et al., using the Arrhenius model, incorporated a correction factor directly into the base fraction of the component deemed most non-ideal in behavior [16]. Their approach was to determine empirically the best molecular interaction correction factor by minimizing the prediction error through reiterative methods. This approach was determined to be the most applicable for our work presented here. Consequently, it was used on the prediction models and fraction bases shown in Table 7.

In the Arrhenius method, the correction constant was incorporated as a multiplier with the WSO fraction base (Eq. 1). In the Modified Wright method, the correction factor was incorporated as a multiplier to the calculated inverse slope of the WSO as described in the ASTM 7152-05 Standard [21]. Finally, the correction factor for the Modified ASTM method was incorporated as a multiplier with the transformed viscosity of the WSO and WSO fraction [19]. The Microsoft Excel solver function was used to determine these constants by minimizing the total AAD for each model.

Improvement of the prediction models with the molecular interaction correction constants are shown in Table 8. There was improvement in percent AAD for the Modified Wright and Modified ASTM methods on all fuel blends. However, mixed results were found with the Arrhenius model, with the lower WSO fraction blends actually increasing in percent AAD. It is possible that the non-ideal molecular interactions of WSO with petroleum fuels are relatively insignificant at lower WSO fractions and become significant only at WSO fractions higher than 30%. Interestingly, the Arrhenius method generally over-predicted the viscosity of the higher WSO fraction blends. Thus, it appears that molecular interactions forces are weaker than ideality at WSO fractions above 30%. While further investigations are necessary to determine the exact fundamental nature of these molecular interactions, the results presented indicate that a correction factor for the Arrhenius model is only necessary when blends have greater than 30% WSO.

Any viscosity blend model can be improved with the addition of correction constants. However, their use should

Table 6 Comparison of experimental and predicted viscosities (cSt) of multi-component blends

Fuel blend	Temperature (°C)	Experiment (±STD)	Arrhenius (mass)	Arrhenius (vol)	Wright (modified)	ASTM (modified)
O50DKG	−10	32.18 ± 0.49	40.17	34.51	24.58	19.66
	0	22.71 ± 0.98	28.68	24.86	18.22	14.76
	10	14.34 ± 0.08	20.47	17.91	13.53	11.10
	20	10.49 ± 0.11	14.61	12.90	10.06	8.35
	30	8.11 ± 0.08	10.43	9.30	7.49	6.30
	40	6.81 ± 0.04	7.45	6.70	5.58	4.75
O37DKG	−10	19.86 ± 1.22	26.01	22.93	17.67	14.58
	0	14.46 ± 1.49	18.99	16.86	13.29	11.08
	10	10.61 ± 1.86	13.86	12.40	10.01	8.43
	20	8.26 ± 0.69	10.12	9.12	7.55	6.42
	30	6.37 ± 1.71	7.39	6.71	5.70	4.89
	40	5.10 ± 2.64	5.39	4.94	4.30	3.73
O25DKG	−10	17.37 ± 0.42	18.28	16.73	14.30	12.28
	0	12.37 ± 0.20	13.58	12.49	10.82	9.36
	10	9.17 ± 0.43	10.08	9.32	8.19	7.14
	20	7.03 ± 0.14	7.48	6.96	6.21	5.46
	30	5.00 ± 0.12	5.56	5.19	4.71	4.17
	40	4.31 ± 0.36	4.13	3.88	3.57	3.18
O10DKG	−10	12.76 ± 0.48	11.60	11.16	10.52	9.81
	0	9.02 ± 0.04	8.80	8.49	8.04	7.52
	10	6.55 ± 0.07	6.67	6.45	6.15	5.77
	20	4.96 ± 0.02	5.06	4.91	4.70	4.43
	30	3.99 ± 0.17	3.84	3.73	3.60	3.40
	40	3.21 ± 0.06	2.91	2.83	2.75	2.61

Table 7 Average absolute deviation for multi-component blend viscosity predictions

Fuel Blend	%AAD			
	Arrhenius (mass)	Arrhenius (vol)	Wright (modified)	ASTM (modified)
O50DKG	28.53	13.48	13.16	28.26
O37DKG	22.86	11.33	9.93	23.83
O25DKG	7.80	3.54	12.58	23.48
O10DKG	4.76	6.54	10.68	16.00
Average	15.99	8.72	11.59	22.89

Table 8 Average absolute deviation for multi-component blend viscosity predictions using molecular interaction correction constants

	Arrhenius (mass)	Arrhenius (volume)	Wright (modified)	ASTM (modified)
Correction constant	0.9031	0.9642	1.191	1.321
Blend (AAD%)				
O50DKG	7.87	7.74	8.04	6.92
O37DKG	5.47	6.19	5.63	7.45
O25DKG	5.70	5.42	3.80	12.52
O10DKG	7.96	8.04	6.03	11.79
Average	6.75	6.85	5.87	9.67

be carefully considered and employed only when necessary. The value of the correction factor is empirically derived and will likely change based on the type of vegetable oil used. In our case, viscosity predictions were excellent using the Arrhenius method with volume fraction on the WSO–diesel blends, suggesting that the fundamental interaction of WSO and diesel does indeed behave in an ideal manner with WSO fractions as high as 50%. Only when kerosene and gasoline are added to the blend at WSO fractions greater than 30% does this ideal interaction start to break down, requiring correction. While the corrected Wright method produced the most accurate results, we believe it is best to use the fundamental Arrhenius equation using volume fractions and incorporate the molecular interaction correction parameter only when necessary. In doing so, viscosity predictions are most accurate and based on fundamental molecular interactions rather than on empirical correction factors.

The ability to model viscosity of blended petroleum fuels has certainly been an area of interest and importance for many years. As the use of alternative fuels becomes more prevalent, the need to predict viscosity of biofuel blends will become just as important as for traditional petroleum fuel blends. The research and results presented here help in the understanding, prediction and potential commercialization of biofuel blends created from unaltered vegetable oil and petroleum fuels. While there are many critically important parameters that must be considered when creating a fuel blend, such as gel-point, energy content and cetane rating, viscosity is equally important. The ability to accurately model biofuel blend viscosity allows one to tailor the fuel to specific diesel engine designs and operating temperatures. In doing so, vegetable oil content can be maximized while maintaining acceptable engine and fuel flow performance.

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