

Empirical correlations between viscosity, density and the cloud point of diesel oil mixtures with the straight vegetable oils (SVO): Palm, Cabbage palm and Copra

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Abstract— Although the valuation of straight vegetable oils (SVOs) into gasoil is a subject which dates from the invention of the motor current remains difficult to control due to the fluctuations of fossil fuel costs. In the search for use of SVO in substitution for gasoil, previous studies have shown that the viscosity, density and cloud point are key parameters in the use of biofuels.

Having established predictive models for these parameters in our previous work on dilution of SVO in diesel, our goal in this article is to look for correlations between these parameters. Viscosity, density and cloud point measurements were made on tropical vegetable oils mixed with gasoil. The results obtained for each blend diesel/SVO from the cloud point to 353K (mean temperature of running diesel) allow justifying the existence of correlations between these key parameters of such biofuel.

Index Terms— straight vegetable oils (SVO): *Elaeis guineensis*, *Sabal palmetto*, *Cocos nucifera*; viscosity, density, cloud point, correlations.

Abbreviations

K : the viscometer constant,
 t : the flow time of the fluid through the capillary,
 \mathcal{G} : the kinetic energy correction given by the manufacturer which is a function of the capillary type and the flow time t ,
 N : the population size,
 η : predictive,
 EXP : experimental,
 PR : calculated value,
 x : the SVO mass rate,
 $T_{cp}(x)$: the cloud point of the mixture,
 T_g : the cloud point of the pure gasoil,
 $d_{mixture}$: the mixture's density,
 T_{cp0} : the cloud point of the mixture at 273.15K,
 a_1 and a_2 : $d_{mixture}$ fitting parameters,
 ν : kinematic viscosity.
 A_1 : the slope of the line
 A_0 : the density of the gasoil at the temperature T (K).

I. INTRODUCTION

The focus of the world on petroleum and its derivatives are the origin of the explosive growth of global needs for fossil fuels even if they are the cause of global warming. Although oil and gas reserves are important, they are limited according

to forecasts by J. Tickell [1] who estimates that half of the oil reserves and a third of those of natural gas will be consumed in 2020 and that these reserves run out after 2040 and 2070 respectively.

The oil crisis of the 1970s [2] was strongly felt in the Third World because of the fragility of the trade balance of raw materials, generating a currency shortage. However, the needs of paltry amount of fossil oil greatly strike their low incomes.

Ivory Coast, third producer of palm oil after Malaysia and Nigeria (FAO, 2012), to mix a proportion of this SVO to gasoil can be an opportunity to exploit. Ramirez-Verduzco et al. [3] and Imahara H. et al. [4] consider that viscosity, density and cloud point are three essential parameters of biofuels.

The viscosity is a physical phenomenon that occurs whenever the adjacent layers of the same fluid are in relative motion, that is to say when it establishes a velocity gradient. This is the internal friction resulting from the sliding of a fluid layer over another. As it flows in a tube, the liquid exerts tangential forces that hinder its movement. According to Ramirez et al. [5], the very high viscosity of vegetable oils is the origin of the sub-supply of the combustion chamber, thus power loss when used in the existing engines.

The density is more or less weight compared to water. It allows determining the buoyancy of a material in pure water. Oils densities vary from 0.9 to 0.93 while that of the gasoil varies from 0.81 to 0.87. Grabosky et al [6] and Karasmanoglu et al [7] studied the effects of using pure SVO in diesel engines. They concluded that the density increases the inertia of jet fuel in the combustion chamber.

Cloud point is the temperature at which appear solid particles in the liquid, able to prevent the normal flow [8]. Khan et al. [9] suggested that the ability to predict the cloud point of biofuels is economically crucial for optimizing the production. This conclusion was confirmed by the findings of Marante and Coutinho [10] on SVO/gasoil mixtures.

Our previous tests conducted on six different vegetable oils mixed with diesel have enabled us to propose predictive models of viscosity [11], the density and the cloud point [12]. The correlations between these parameters object to specify for each composition of known SVO fatty acids, suitable conditions for its use. The goal in this article is to seek from the experiences, correlations between these three parameters.

II. MATERIAL AND METHODS

2.1 Material

2.1.1 Raw material

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Bio-oils

Bio-oils used are from palm, copra, cabbage, cotton and groundnut. Palm oil is from the *Ehania Palmci* society. A secondary quality palm oil (palm QS) is extracted from the seeds decomposed. Copra oil has been extracted in the traditional way by pressing. Cabbage oil is solvent extracted using cyclohexane. As for groundnut and cotton oils, they were purchased commercially. Their fatty acid compositions are shown in Table 1.

Fossil material

We got summer gasoil (cloud point = 0 ° C) at a BP station in Paris. This type of diesel is also distributed in Africa. It is directly blend in vegetable oil.

2.1.2 Technical equipment

Viscometer

An Ubbelohde capillary viscometer SCHOTT is used with capillaries reference: I ref. 53010 Ic ref. 53213 and IIc ref. 53023.

Densimeter

Specific mass measurements were performed using a densimeter Anton Paar DMA 4500, with an accuracy of 3.10^{-4} .

Device for measurement of cloud point:

It consists of a double envelope transparent thermostated bath. A circulation fluid allows regulating temperature whereas the SVO sample tubes (diameter 8mm) are plunged in the bath.

Accessories

The accessories comprise:

- A thermostat bath JULABO type F30 F10-VC (243K to 373K) with a compressor;
- A vacuum cleaner SCHOTT GERÄTE AV 310;
- A bottle of nitrogen to 200 bars with a pressure regulator from 0 to 12 bars;
- A pressure sensor.

2.2 Methods

2.2.1 Viscosity measurement at atmospheric pressure

The viscosity was measured using the Ubbelohde viscometer with the uncertainties of ± 0.02 cSt from 1 to 5 cSt; ± 0.05 cSt from 6 to 20 cSt and ± 0.1 cSt from 21 and 55 cSt. Temperature varies from the cloud point (273K to 308K depending on the mixture) to 343K. The composition of the mixture by weight percentage oil range 0% to 100%. Kinematic viscosity ν is calculated using the formula (Eq. A).

2.2.2 Density measurement

Each oil mixtures 0; 5; 10; 15; 20; 30; 40; 50 and 100% w/w of oil were performed. Density measurements are made when varying the temperature between 283K C and 353K in increments of 10 K.

2.2.3 Cloud point measurement

Each sample mixtures oil / gasoil is placed in an open test tube. These tubes are firmly fixed on a polystyrene plate and dip along with a thermometer in the thermostated bath. We begin the measurement by a temperature at which all samples are perfectly liquid (usually between 303K and

313K). Every 5 minutes, the set temperature is lowered by 0.5K. Samples are successively reviewed at the end of 5 minutes and those who have reached the cloud point are removed.

Results are compared to those obtained from the predictive equations by using Equation B to calculate the average absolute deviation (AAD) according Krisnangkura et al 2010 [13].

III. RESULTS AND DISCUSSION

Cloud points of samples from SVO are given in Table 2. The correlations were previously given according to the temperature and the oil rate in the mixture [12]. Mean quadratic differences of cloud points between the model and defined by Equation 2 and the experimental data won the maximum value of 0.6 K for SVO / gasoil (cloud point: 273K) mixture. Although all these oils are from tropical climate, some of them do not give enough point to draw a graph. This is the case of groundnut and cotton oils. The predictive model of the cloud point that had been proposed is given by Equation C.

Expressed in Kelvin, cloud points values calculated using the predictive model are shown in Table 2, and the AAD value calculated for the different oils.

Our results give 0.41 and 0.14 respectively for the maximum deviation and the maximum AAD value related to copra oil, the other oils giving lower values. J D Mejia used a model obtained by Imahara H. et al. [4]. It consists in describing the cloud point thermodynamically as a solid-liquid balance. He obtained 5.21% and 3.17 respectively for the maximum deviation and the maximum AAD value on a cloud point range between 271,15K and 289,15K.

Mejia also used empirical models of second order polynomial proposed by Tang (2008) to determine the cloud point of mixtures of palm and castor oils with gasoil. He got a maximum deviation of 8.48% and a maximum AAD value of 4.10 in the same temperature range.

For our model, the deviation and AAD values are lower. Generally, mixtures of the cloud point increase with SVO rate in the mixture. Although copra and cabbage oils have the same cloud point, they don't give exactly the same progression. At 50% SVO, mixtures cloud points are slightly higher than the weighted average of the cloud points. So SVO cloud point has more influence on the mixture.

3.1 Correlation between density and the cloud point

By plotting the density of SVO / gasoil mixture versus the cloud point, graphs are given in Figure 1. These graphs show a binomial evolution of the density versus the cloud point from 283,15K to 353,15K. Palm QS oil boils around 353,15K because of its impurities that does not allow performing the measurement at this temperature. Let fixe a reference graph (for example the one of higher temperature T_0). The other graphs can be deducted from the last one by translating on the ordinate to gasoil density at temperature T (of the new graph). A. Sarin et al. [15] worked on the effects of mixtures of biodiesel jatropha-palm-pongamia on the cloud point and Coutinho et al. [16] on fuel mixture. They proposed models versus SVO rate or versus esters saturation rate. The proposed models do not permit an evolution of clouds points according to biofuel density.

For each SVO, binomial coefficients to adjust the cloud point to the highest temperature according to Equation D are given in Table 3.

Parabolas are obtained a correlation coefficient close to 1; this well justifies the existing of correlation between the density and the cloud point.

3.2 Correlation between the density and viscosity of vegetable oil/gasoil blend

If we match for a given oil and a SVO mass rate in the mix, density and viscosity at temperature T(K), we can associate to each viscosity of the mixture, its density at the set temperature. The graphs of the densities versus the viscosities are in Figure 2.

The shape of the graphs shows a logarithmic progression of the density versus the kinematic viscosity ν of biofuels at set temperature. This ultimate notice allow drawing in each case the graphs of density versus $\ln\nu$. The linear distribution obtained is fitted by Equation E:

Straight lines are drawn with a correlation coefficient close to 1. This confirms well the existence of the correlation between the density and the viscosity that allows predicting the viscosity from the density and vice versa.

Modelling the density slope A_1

Representing the values of the slope A_1 versus oil saturation rate, graph of Figure 3 are obtained.

The graph is plotted with $R^2 = 8523$, showing that the slope A_1 evolves linearly with the mass percentage of saturated fatty acids in oils.

Schaschke Carl et al. [17] carried out tests with a high pressure cylinder viscometer fall on different types of fossil fuels from refineries in England and on a 5% mixture of methyl ester to a gasoil. The densities of these fuels were determined experimentally under pressures up to 500 MPa at various temperatures 298K, 323K, 348K and 373K. They have drawn graphs of the evolution of the dynamic viscosity and density versus the pressure. The graphs are similar for all their fuels.

Taking the pressure as a reference parameter, we draw the evolution of the density according to the viscosity for each of fuels to check if our results were transferable their conditions. The results obtained with all their fuels being similar, the one relating to fuel1 at 323K is given compared to our SVO blends to gasoil in Figure 4.

We get the same trend that confirms the logarithmic evolution obtained with our SVO/gasoil blends. We also check the consistency with the work of A. Amin et al. [17] who performed experiments mixtures of castor oil with diesel to study its effects on the kinematic viscosity, and density. Taking as reference the percentage of biodiesel, the results allow obtaining also a logarithmic pace.

3.3 Correlation between viscosity and cloud point of SVO/gasoil blends

For given oil and temperature, we match the cloud point of the mixture, the viscosity by varying SVO rate in the

mixture. The plot graphs at 303K, 353K and 343K are shown in Figure 5.

In fact, we have expressed firstly density versus the cloud point and the other density as a function of viscosity. From these two models, we can mathematically express the viscosity as a function of the cloud point.

Going from Eq (D) and Eq.(E) we equal the second members to get Eq.(F.1). From which we derive Eq. (F.2), and setting down $k_2=a_2/A_1$; $k_2=a_1/A_1$ and $k_0=\exp[(a_0-A_0)/A_1]$ we get Eq.(F.3).

Figure 5 shows comparative graphs of the evolution of the kinematic viscosity SVO/gasoil blends versus the cloud point of the mixture between the experiment and the predictive. For each SVO, we get a similarity between the curves obtained by experiment and the ones obtained by calculations at different temperatures. The evolutions show an exponential pace. The mean squared differences of the average (curve obtained by calculation) are calculated and summarized in Table 5, and deviations and AAD values of viscosities between the model Eq.(F.3) and the Experimental data are given in table 6.

Values obtained are for oil levels below 50% w / w in the blends. These lower values well confirm the existence of correlation between the kinematic viscosity and the cloud point.

Deviations give more low temperature values decreased in the case of QS palm oil, copra and cabbage. Palm oil does not follow this logic though its deviations and AAD value are generally lower compared to that of palm QS. The largest deviations and therefore the greatest value of AAD are obtained with QS palm which is highly degraded oil due to impurities contained therein. The free fatty acids rate of palm QS is more increased than the other oils, this rate could be the origin of the high viscosity value deviation predominately when the temperature increases. Taking oil rate in the mixtures as an observation criterion, we find that the deviations are not related to oil rate in the mixture.

Overall, deviations and ADD values used to justify the existence of correlations between the kinematic viscosity and cloud point.

J.D. Mejia et al. [18] studied the effect of mixtures of palm and castor oil / gasoil on the viscosity and cloud point at 313K. We represent in Figure 6 Mejia's curve and the one we have obtained with palm oil.

These graphs allow noticing a growth of viscosity versus the cloud point. All these curves show a linear evolution in temperature range of Mejia's work going from 273K to 293K. Above the 293K, our results give in reality exponential tendencies. However, over the same temperature range, the slopes we get are higher than the Mejia's one. This difference arises because Mejia's biofuels are mixtures of palm and castor oil/gasoil and have not the same characteristics as ours which are mixtures of only one vegetable oil and gasoil.

IV. CONCLUSION

Experiments attempted from clouds point to 353K relating to diesel running temperature, confirm that the density and the cloud point and the viscosity known as key parameters of biofuels are linked two by two. Knowing two of these

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parameters allows predicting the third in the case of SVO/gasoil mixtures. The results on fossil oils given by literature confirm the existence of correlation between the density and the kinematic viscosity. For a given fossil, the cloud point, being unique doesn't allow as in the case of mixing vegetable oil/gasoil to check correlations with the cloud point. Correlation coefficients are better between the density and viscosity where R² is closed to 1 for all vegetable oils. However these correlations are obtained with deviations and average absolute deviation (AAD) allow validating the results.

ACKNOWLEDGMENTS

The authors thank the CNRS and the french-ivoirien office for cooperation of the MAE for financial support.

CONFLICT OF INTEREST

The author declare no conflict of interest.

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FORMULAS AND EQUATIONS:

$$\nu = K(t - \theta) \quad \text{Eq.(A)}$$

$$AAD = \sum [|\eta_{EXP} - \eta_{PR}| \times 100 / \eta_{EXP}] / N \quad \text{Eq.(B)}$$

$$T_{cp} = 0.095[(Tg + 10.478)x^3 - 2.236(Tg + 7.625)x^2 + 1.236(Tg + 5.317)x]A - 4.371[Tg \cdot x^3 - 2.990(Tg - 0.500)x^2 - 0.229(T_{ocp} + 9.701(Tg - 0.674))x - 0.229Tg] \quad \text{Eq.(C)}$$

$$d_{mixture} = a_2 T_{cp}^2 + a_1 T_{cp} + T_{cp0} \quad \text{Eq.(D)}$$

$$d_{mixture} = A_1 \ln(\nu) + A_0 \quad \text{Eq.(E)}$$

$$\ln(\nu) = a_2 T_{cp}^2 / A_1 + a_1 T_{cp} / A_1 + (a_0 - A_0) / A_1 \quad \text{Eq. (F.1)}$$

$$\nu = \exp \left[a_2 T_{cp}^2 / A_1 + a_1 T_{cp} / A_1 + (a_0 - A_0) / A_1 \right] \quad \text{Eq. (F.2)}$$

$$\nu = k_0 \exp \left[k_1 T_{cp} + k_2 T_{cp}^2 \right] \quad \text{Eq. (F.3)}$$

TABLES

Table 1: Bio-oils composition (analysed by GC/MS) (% m/m) [13]

	Fatty acids	Copra	cabbage	Palm	Palm QS ^(a)
C8	caprylic	6.52	1.43	-	-
C10	capric	6.38	4.03	0.11	0.42
C12	lauric	27.73	28.9	1.32	0.43
C14	myristic	20.11	20.3	2.82	2.06
C16 :0	palmitic	13.37	14.04	26.38	29.60
C16 :1	pamlitoleic	0.74	-	-	-
C18 :2	linoleic	1.93	2.88	11.41	8.76
C18 :1	oleic	16.00	23.5	40.71	41.05
C18 :0	stearic	5.91	4.91	12.46	14.74
C18 :3	linolenic	-	-	2.91	2.63
C18 :3	α -linolenic	0.62	-	-	-
C20 :0	arachidic	-	-	1.88	0.31
C20 :1	gadoleic	0.42	-	-	-
C22 :0	béhenic	0.26	-	-	-
C22 :1	erucic	-	-	-	-
%					
Saturation		80.28	73.61	44.97	47.56

(a) : Secondary quality palm oil

Table 2: Cloud Point blends vegetable oil / gasoil versus the temperature in Kelvin calculated using our model Eq.(C) (cloud point gasoil : 273K)

% Oil	palm HC			palm MQ		
	EXP	PR	Dev (%)	EXP	PR	Dev (%)
0	273	273	0.00	273	273	0.00
5	275	275.8	0.29	274.9	275.2	0.12
10	278.6	278.4	0.08	278.1	277.3	0.28
15	280.9	280.8	0.05	279.4	279.3	0.04
20	282.5	283.0	0.17	280.9	281.1	0.08
30	286.9	286.9	0.00	284.5	284.5	0.01
40	290	290.3	0.11	287.4	287.6	0.06
50	293.4	293.4	0.00	290.6	290.4	0.08
100	308.5	308.5	0.00	304	304	0.00
AAD			0.11	0.09		
% Oil	Copra			Cabbage		
	EXP	PR	Dev	EXP	PR	Dev
0	273	273	0.00	273	273	0.00
5	274.2	273.1	0.41	273.5	273.5	0.00
10	274.2	273.5	0.27	273.8	274.2	0.13
15	274.9	274.1	0.29	274.9	275.0	0.04
20	274.9	275.0	0.03	275.6	276.0	0.14
30	277.2	277.3	0.04	278.7	278.3	0.14
40	280.9	280.2	0.27	280.9	280.9	0.01
50	283.5	283.2	0.09	283.5	283.7	0.06
100	293	293.0	0.00	293	293.0	0.00
AAD			0.14	0.10		

Table 3: Coefficient relating to binomial adjustment graph at 353K or 343K

SVO	a ₀	a ₁	a ₂	R ²
Palm	4301,7	-26,559	0,0502	0,9921
Palm QS	2302,1	-6,841	0,0268	0,9917
Copra	2662,3	-17,3	0,0383	0,9881
Cabbage	5707	-38,928	0,0766	0,9959

Table 4: logarithmic density adjustment parameter as a function of the viscosity

SVO	A ₁	A ₀	R ²
Groundnut	33,416	780,19	0,9999
Cotton	30,712	784,8	0,9999
Cabbage	37,220	778,18	0,9997
Copra	42,353	774,06	0,9996
Palm QS	32,124	781,07	0,9966
Palm	31,969	781,43	0,9997

Table 5: Mean quadratic differences of viscosities between the model (Eq.(F.3)) and the Experimental data

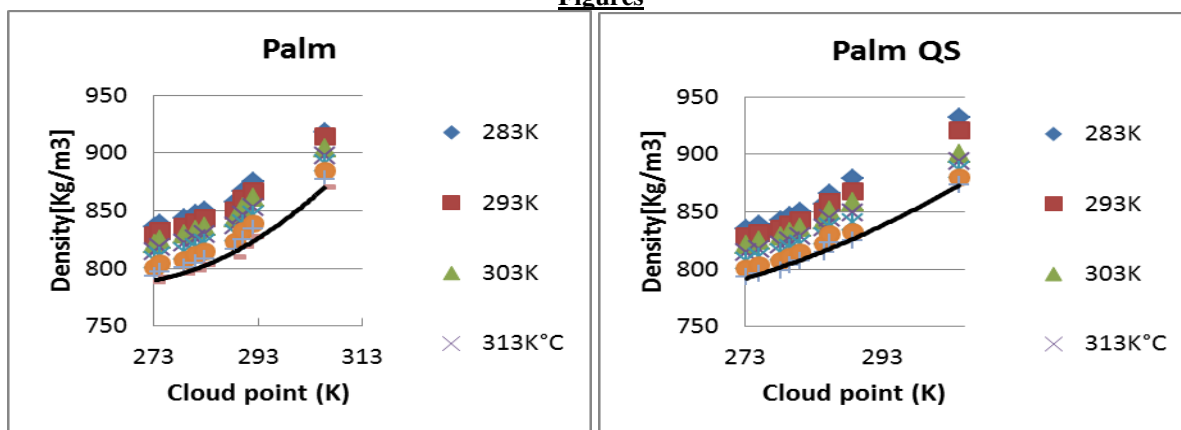
SVO	Palm	Palm QS	Copra	Cabbage
303	0.45	0.33	0.21	0.24
323	0.16	0.23	0.18	0.23
343	0.20	0.09	0.16	0.18

Table 6: Deviations and AAD values of viscosities between the model (Eq.(F.3)) and the Experimental data

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T(K)			303			323			343		
	Oil (%)	Tcp (K)	EXP	PR	Dev(%)	EXP	PR	Dev(%)	EXP	PR	Dev(%)
Palm	0	273.15	3.30	3.77	14.38	2.25	2.29	1.72	1.64	1.51	7.60
	5	274.15	3.69	3.89	5.46	2.50	2.36	5.72	1.84	1.56	14.99
	10	278.75	4.23	4.66	10.17	2.81	2.81	0.01	2.04	1.88	7.49
	15	281.05	4.83	5.22	8.17	3.16	3.14	0.48	2.32	2.12	8.51
	20	282.65	5.52	5.71	3.46	3.66	3.43	6.10	2.63	2.32	11.59
	30	288.55	7.23	8.48	17.34	4.65	5.09	9.47	3.23	3.48	7.74
	40	290.15	9.90	9.62	2.85	5.97	5.77	3.38	4.05	3.95	2.33
	50	292.15	14.10	11.38	19.31	7.68	6.82	11.16	5.04	4.69	6.97
				AAD	10.14		AAD	4.75		AAD	8.40
Palm QS	0	273.15	3.30	3.46	4.93	2.25	2.02	10.36	1.64	1.50	8.15
	5	275.05	3.66	3.91	6.89	2.47	2.25	8.70	1.82	1.68	7.73
	10	278.25	4.14	4.85	17.21	2.77	2.85	2.77	2.02	2.11	4.64
	15	279.55	4.74	5.31	12.12	3.09	3.32	7.58	2.24	2.46	10.12
	20	281.05	5.34	5.92	10.83	3.54	3.72	5.16	2.45	2.76	12.70
	30	284.65	6.99	7.75	10.84	4.47	5.82	30.26	3.01	4.34	44.25
	40	285.35	9.12	8.18	10.31	5.64	6.63	17.58	3.66	4.96	35.58
	50	288.65	12.06	10.64	11.74	7.17	7.85	9.46	4.71	5.89	25.07
				AAD	10.61		AAD	11.48		AAD	18.53
Copra	0	273.15	3.30	3.44	4.30	2.25	2.24	0.36	1.64	1.85	13.02
	5	274.35	3.63	3.90	7.65	2.44	2.54	4.20	1.80	2.10	16.90
	10	274.35	4.02	3.90	2.83	2.67	2.54	4.79	1.95	2.10	7.86
	15	275.05	4.47	4.21	5.88	2.97	2.74	7.87	2.12	2.26	6.86
	20	275.05	4.98	4.21	15.53	3.24	2.74	15.55	2.32	2.26	2.36
	30	277.35	6.27	5.37	14.27	4.05	3.49	13.81	2.80	2.89	3.41
	40	281.05	7.89	8.00	1.43	4.98	5.18	3.98	3.36	4.31	28.51
	50	283.65	11.31	10.61	6.20	6.75	6.85	1.46	4.38	5.72	30.67
				AAD	7.26		AAD	6.50		AAD	13.70
Cabbage	0	273.15	3.30	3.86	17.03	2.25	2.57	14.27	1.64	1.59	2.80
	15	275.05	4.53	4.61	1.79	2.99	3.07	2.71	2.12	1.89	10.55
	20	275.75	5.04	4.93	2.18	3.36	3.28	2.28	2.30	2.71	17.97
	30	278.85	6.30	6.69	6.21	4.05	4.45	10.03	2.67	3.43	28.35
	40	281.05	8.19	8.38	2.41	4.92	5.58	13.40	3.16	4.06	28.33
	50	283.65	10.08	11.05	9.67	6.09	7.34	20.61	4.05	4.51	11.54
				AAD	6.55		AAD	10.55		AAD	16.59

Figures



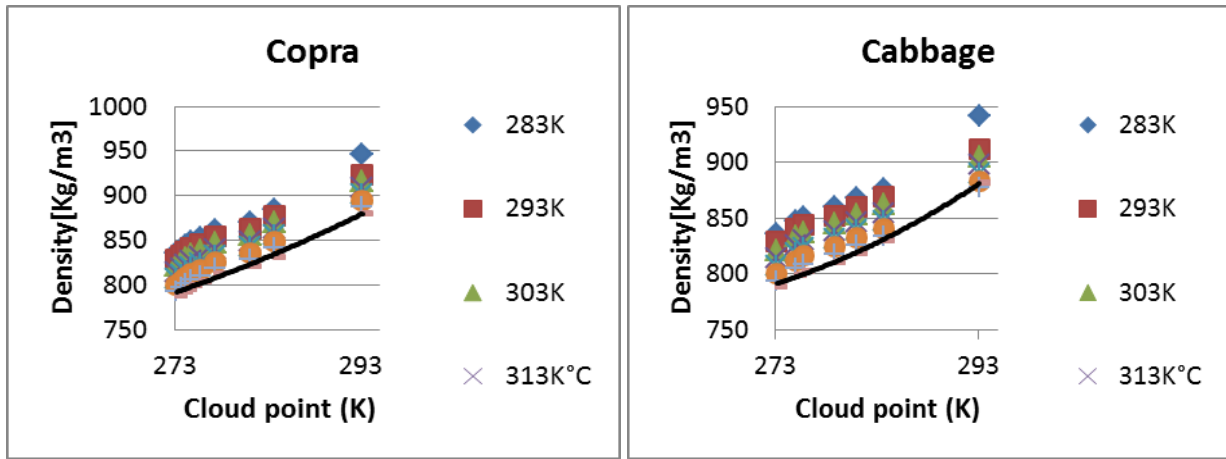


Figure 1: Evolution of the density versus cloud point

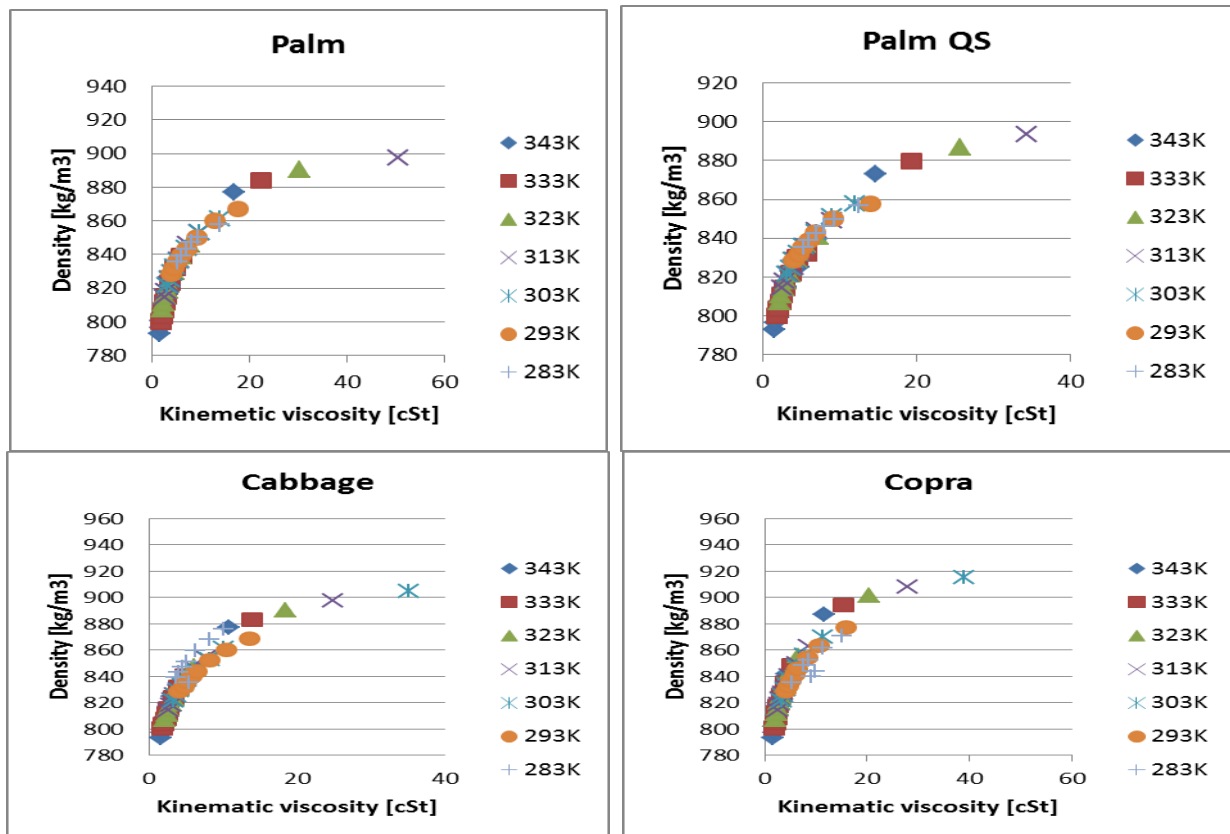


Figure 2: Graphs of densities versus viscosities of SVO/gasoil mixture at temperature T

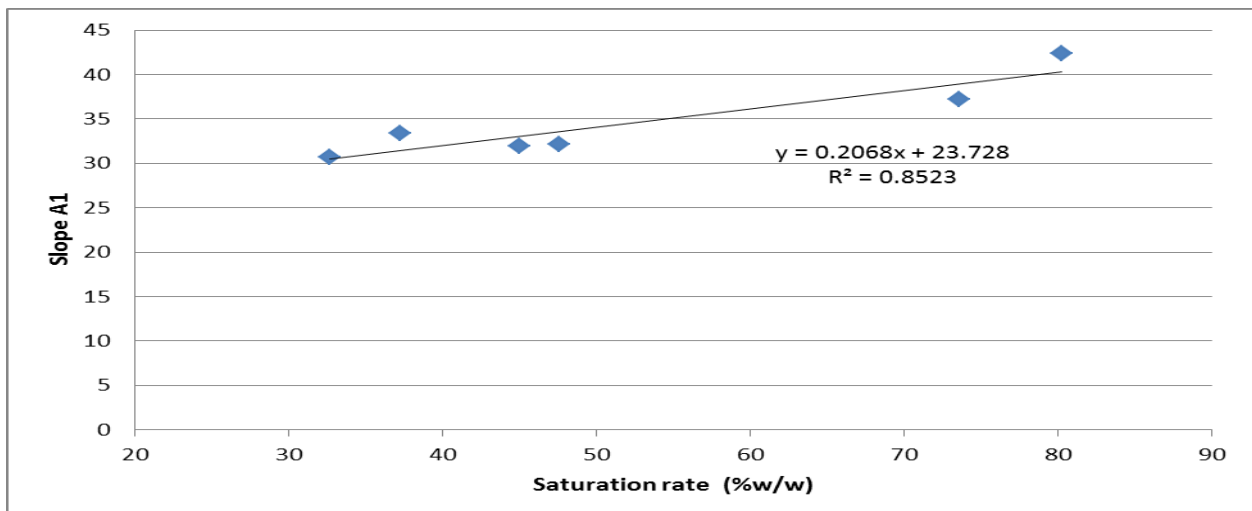


Figure 3: Evolution of the A₁ slope versus the saturation of vegetable oils

Empirical correlations between viscosity, density and the cloud point of diesel oil mixtures with the straight vegetable oils (SVO): Palm, Cabbage palm and Copra

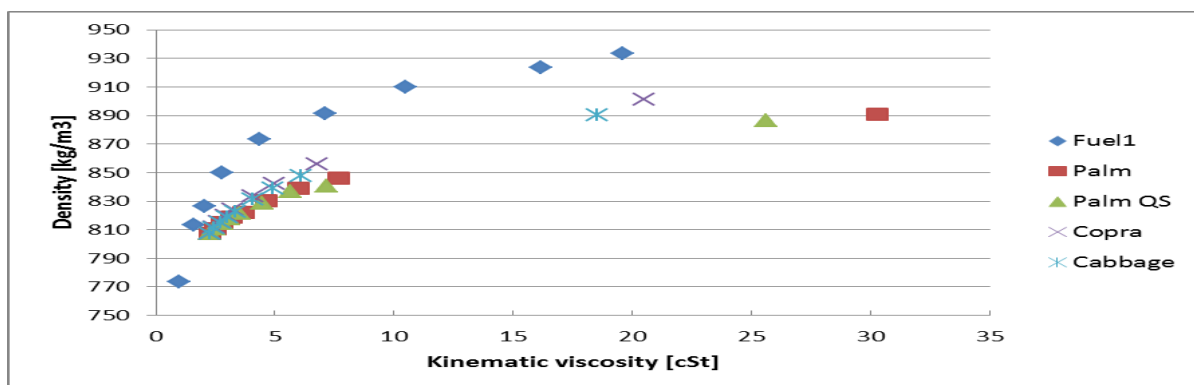


Figure 4: Comparison of evolution of the density versus the kinematic viscosity Fuel1 and the SVOs mixed with gasoil at 323K.

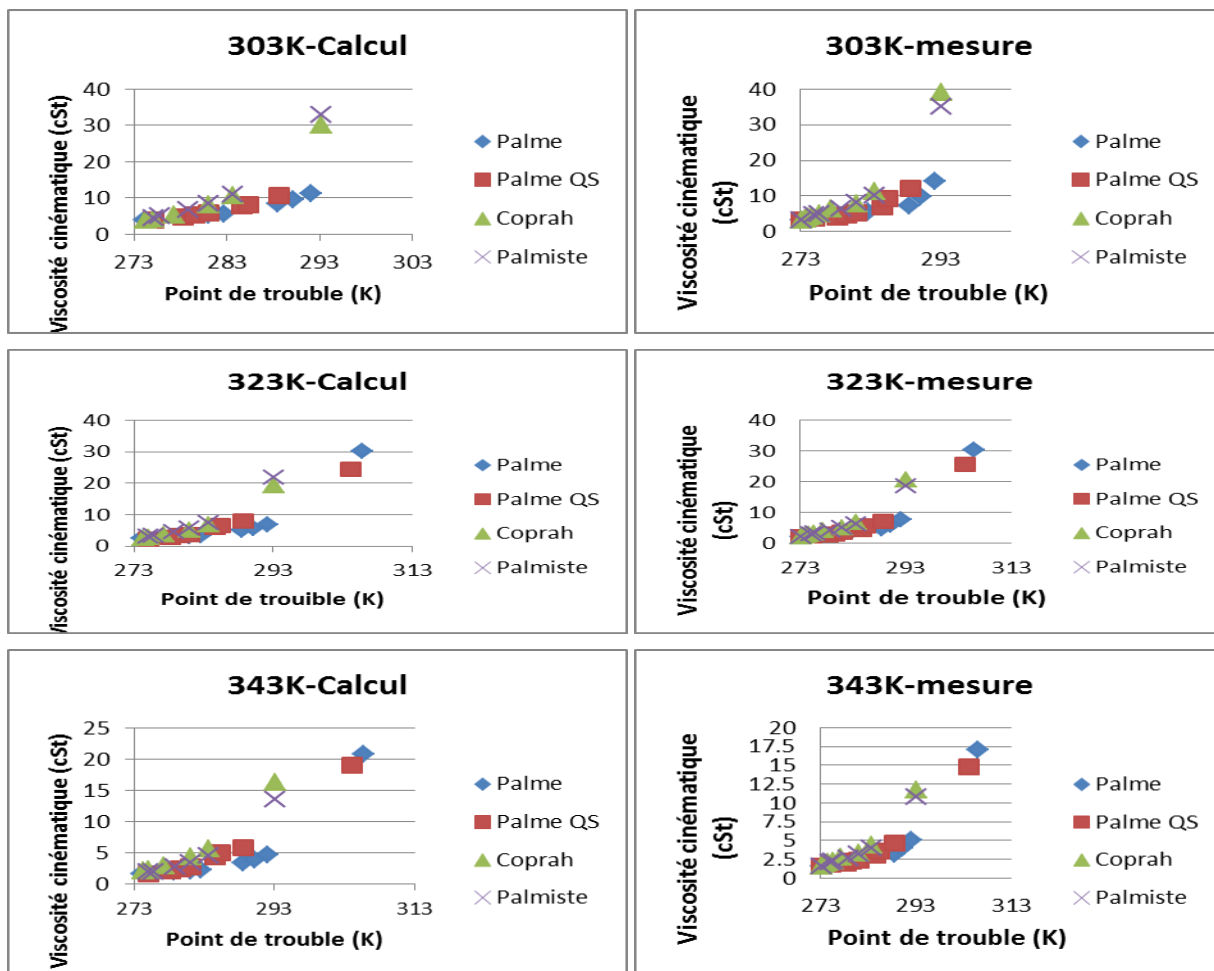


Figure 5: Comparatives graph of the evolution of kinematic viscosity of SVO / gasoil blends according to the cloud point.

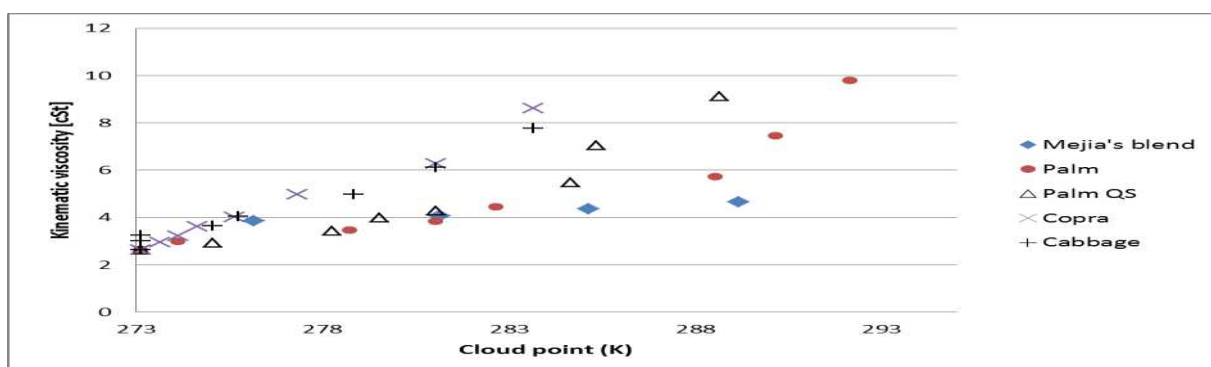


Figure 6: Evolution of kinematic viscosity of fuels depending on the cloud point