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Statistical Analysis of Surface Tension and Viscosity of Heavy Oil Crude by Mixing Rule

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Abstract Crude oil data analysis is necessary to know the possibilities in energy reduction in transport and some uses in many areas, such as materials construction or emissions reduction. Currently, extra-heavy crude oil is produced in Mexico and other countries like Canada, Venezuela, and the United States. For Mexico, in many cases, it is found as a part of "chapopote" in soils; solids and oils can be mixed to produce valuable materials on building construction. Models and representations of oils and many liquids are essential in the industry, allowing to predict a priori the characterization in many application cases. This first paper presents the experimental results related to the behavior of the surface tension of crude oil and a mixture of crude oil and a flow improver and the behavior of the viscosity concerning the temperature and the improver fraction in volume by statistical analysis and application of a mixing-rule for two liquids. From these results, statistical models were adjusted to be taken as a basis for interpreting the physical-chemical mechanism. It is possible to improve the transport and extraction of heavy crudes through chemical additives that act as flow improvers opening the possibility to combine with solids for some proposes as permeability change.

Keywords Mixing Rule Crude Oil, Viscosity Correlation, Statistical Analysis

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1. Introduction

For several centuries, Mexico has been an oil producer where heavy crude oil has been presented at “chapopoteras” [1]. Models have been made to predict viscosity changes from a theoretical mathematical analysis [2] and others that work to predict the apparent viscosity of death crude oil or emulsion [3].

Several sustainable earth-based building systems [4] are proposed as ecological solutions; some of the soil materials can be stabilized with various natural oils that increase the hydrophobic character [5]. Stabilizers generally promote the increase in durability of ground-based materials [6]. Formerly oil and chapopote was used to stabilize buildings presenting some examples in archaeological centers in Mexico [7].

At present various materials are presented as a sustainable source alternative [8]; indeed, some waste can be used as a partial substitute for traditional materials such as cement [9], heat stabilization treatments have been proposed, and the and Utilization of Ash as a Construction

Material [10]. Although certain plants are another alternative for the remediation of soils with crude oil in their composition [11]

To propose the use of heavy and extra-heavy crude oil, know its transport, and know the possibility of mixing, it is necessary to understand the rheological properties [12] and the change with the use of viscosity improvers.

Various effects have been found in the development of rocks, i.e., Ahmadi, M. et al. [13] presents that in some soils, such as Sand-kaolinite proportions with oil, it changes the shear stresses on the element. These shear stresses depend on the composition of the crude oil, but mainly on its rheological characteristics accompanied by various models such as soil and concrete since they can be used to positively modify other properties such as the passage or storage of water in this medium [14].

2. Materials and Methods

In all cases, we used extra-heavy crude oil from the Northwest of Mexico.

2.1. Surface Tension

In this study, we used the ring method for surface tension with an Automatic surface tensiometer TEN 202 equipment using the ring method. In this method, the force to separate the surface of a metal ring from a liquid surface is measured, ensuring the ring is completely immersed in the liquid before measuring.

From the laws of thermodynamics and assuming that crude behaves roughly like a pure liquid (the effects related to chemical potential are disregarded), it was estimated the behavior of surface entropy and surface internal energy.

Figure 1. Tension determination with a platinum ring

2.2. Viscosity

Viscosity analyses were carried out on a rheolabQC rheometer model Anton Paar (Figure 2), with a geometry of concentric cylinders, the samples were measured at a constant temperature of 25°C, 30°C, 40°C, and 60°C and with cutting speeds between 0 and 100 s^{-1}.

Figure 2. RheolabQC rheometer model Anton Paar brand.

In this model, the temperature is expressed in degrees Kelvin and the composition of the improver in % by volume. The adjustment was carried out using the Marquardt method. The results of the estimate were as follows.

Based on these results, a nonlinear regression statistical adjustment of the model was made:

$$\mu = Aexp\left(\frac{B \times 1000}{T}\right)\exp(-kx)$$

In this model, the temperature is expressed in degrees Kelvin and the composition of the improver in % by volume. The adjustment was carried out using the Marquardt method. The results of the estimate were as follows:

Nine iterations and 39 function calls were carried out before reaching convergence in the results. For the multiple regression statistical study, we used the STATGRAPHICS program.

3. Results and Discussion

3.1. Analysis of the Influence of the Flow Improver on Surface Tension

The interfacial tension behavior for a crude oil concerning temperature was carried out with and without a flow improver. Table 1 shows the surface tension values of crude oil and improver at a temperature of 25°C.

Table 1. Surface tension values of crude oil and fluidity improver at 25°C

component	Surface tension mN/m
Crude oil	61,1316
Fluency enhancer	29,22

Surface tension values were taken for different temperature values. The observed experimental results are shown in Figure 3, presenting the adjusted linear trend lines.

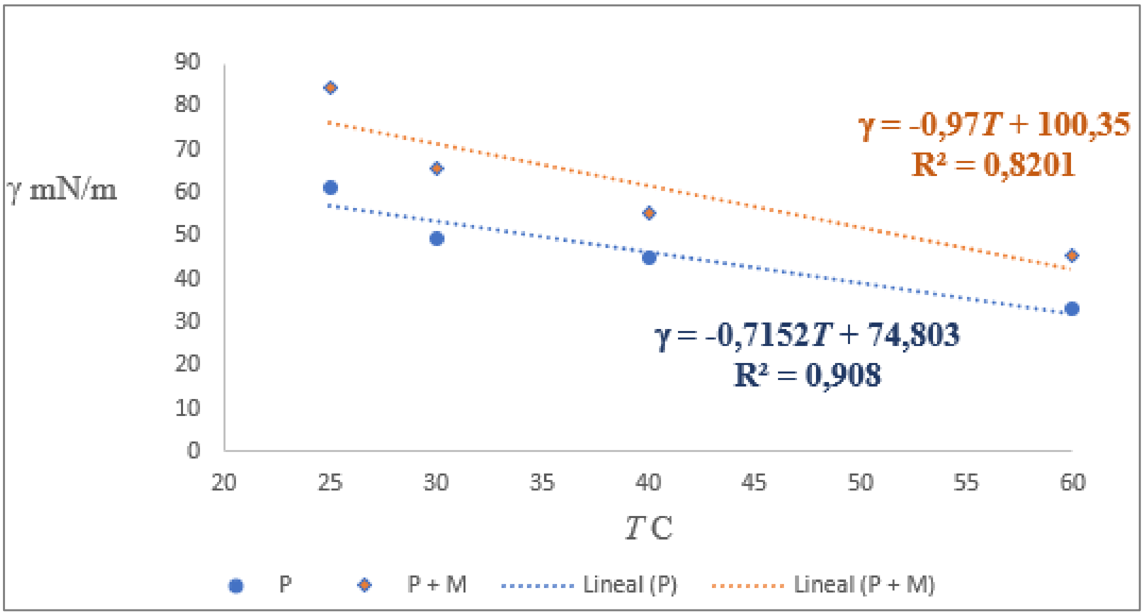

Figure 3. The behavior of the surface tension of crude oil (P) and a mixture of crude oil and fluidity improver (P + M)

From the laws of thermodynamics and if crude behaves roughly like a pure liquid (the effects related to chemical potential are disregarded), one can estimate the behavior of surface entropy and surface internal energy.

In this case, considering the fitted models, we obtain for the surface entropy:

$$S_s = -\frac{\partial \gamma}{\partial T} = 0{,}7152 \times 10^{-3}\,\frac{\mathrm{mJ}}{\mathrm{m}^2}$$

Raw + fluidity improver

$$S_s = -\frac{\partial \gamma}{\partial T} = 0{,}97 \times 10^{-3}\,\frac{\mathrm{mJ}}{\mathrm{m}^2}$$

And for internal surface energy:
raw:

$$E_s = \gamma - T\frac{\partial \gamma}{\partial T} = 74{,}803 \times 10^{-3}\,\frac{\mathrm{mJ}}{\mathrm{m}^2}$$

Raw + fluidity improver

$$E_s = \gamma - T\frac{\partial \gamma}{\partial T} = 100{,}35 \times 10^{-3}\,\frac{\mathrm{mJ}}{\mathrm{m}^2}$$

From these estimations, the following facts are obtained:

The fluidity improver caused an increase in the surface tension in crude oil for all the temperature values considered, where this increase cannot be estimated considering the standard equations. Traditional equations estimate the behavior of the surface tension of a mixture of liquids from the composition of the system and the surface tensions of the components (Priogine's equation). As can be seen, the surface tension of the improver is lower than the surface tension of the crude oil; however, the surface tension of the mixture is significantly higher than both fluids separately.

It is known that substances that act as surfactants for a given liquid cause a decrease in surface tension. This effect is not observed in the system, where you have to have that the surface tension far from decreasing is increased; therefore, it can be raised without doubts that the improver has no surface activity in the raw system – air.

For highly viscous asphaltic crudes the presence of asphaltenes (a polar substance present in crude oil) causes the appearance of a dispersed phase that influences the laminar flow patterns. It causes an appreciable increase in apparent viscosity and a non-Newtonian behavior at low temperatures typical of dispersed systems. The improver causes a decrease in the size of the asphaltenes aggregation, avoiding deposition on tubes and reducing viscosity.

If the hypothesis is considered valid, the flow improver decreases the aggregation of asphaltenes. When grouped in such a way that they present polar and non-polar groups that confer surface activity in the raw interface – air, then the partial destruction of these aggregates would cause a decrease in the concentration of the same on the surface with the consequent increase in surface tension. Note that the estimated surface entropy is more significant than the improver's presence in the absence of an improver, which indicates a higher order on the surface. The more excellent self-organization of these aggregates can explain it.

The internal surface energy, associated with the kinetic energy of the molecules, is increased in the presence of the enhancer. Destruction of these aggregates can also explain this, being structures of higher average molecular weight experience lower kinetic energy (speed) at the same temperature.

It is important to note that these analyses are related to the raw–air system. For a raw system – water, the effects of flow enhancers on the interfacial tension between both liquids may be different.

3.2. Analysis of the Effect of the Flow Improver on Viscosity

The effect of the flow improver on the viscosity of the crude oil was studied experimentally for different values of temperature and volume fraction of the improver. The results obtained are shown in Figures 4 to 6. Surface tension values were taken for different temperature values. The observed experimental results are shown in Figure 1, presenting the adjusted linear trend lines.

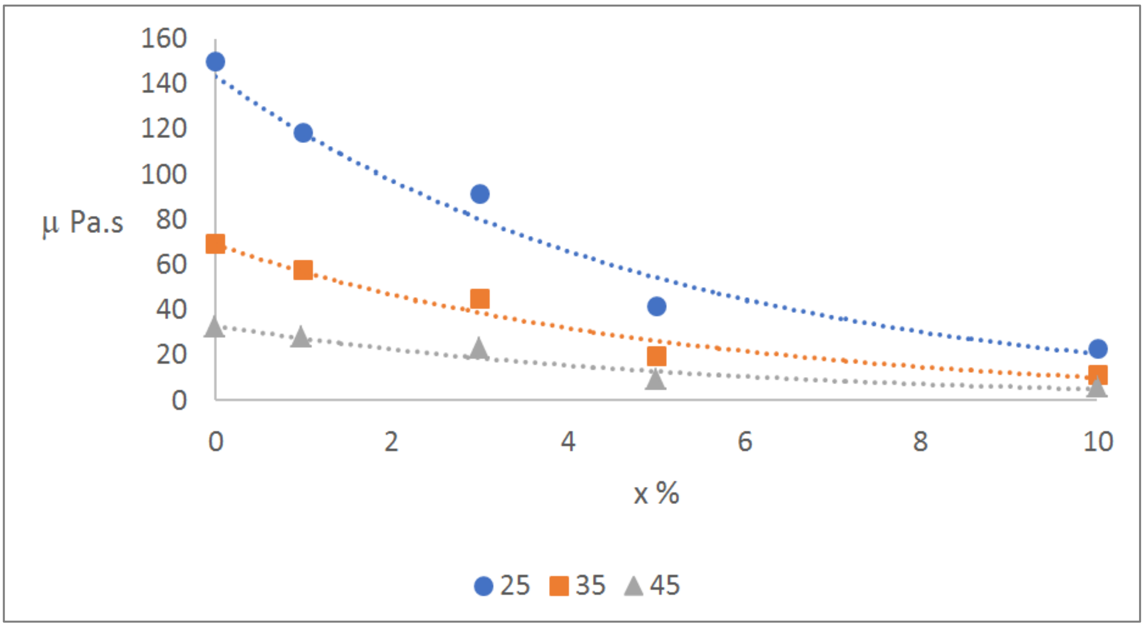

Figure 4. Viscosity behavior for the volume fraction of improver for temperatures of 25, 35, and 45°C

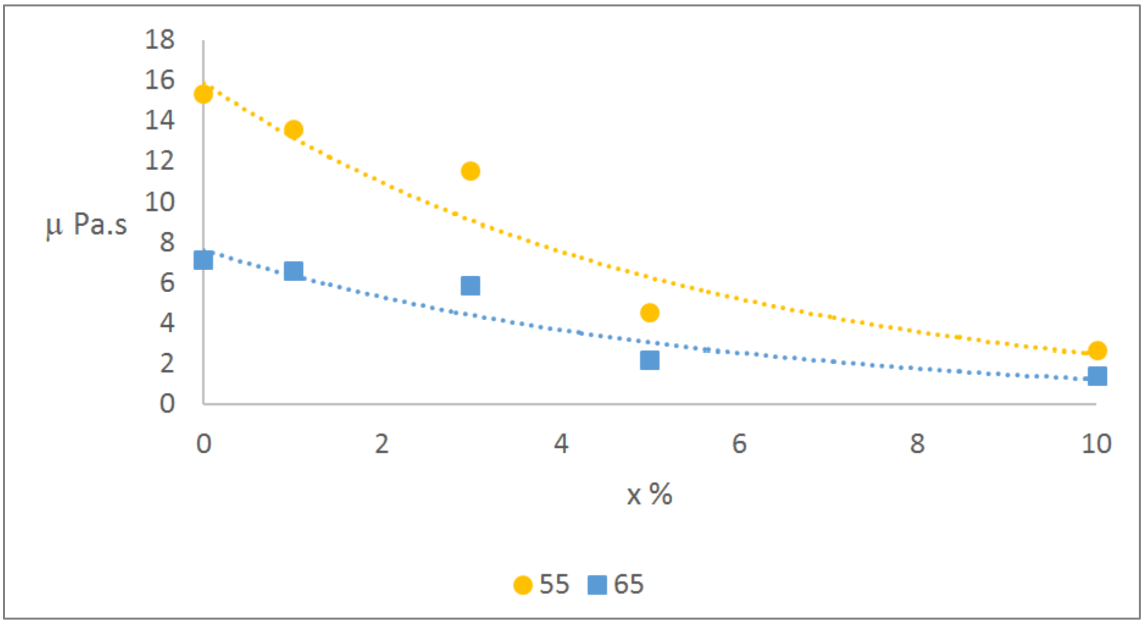

Figure 5. Viscosity behavior concerning the volume fraction of improver for temperatures of 55 and 65°C

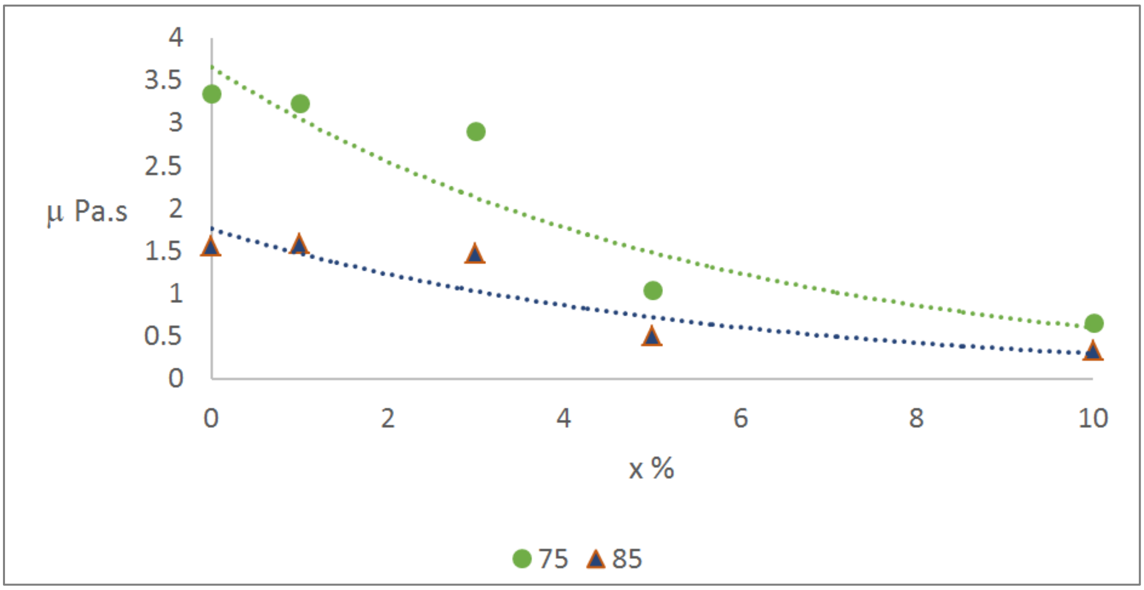

Figure 6. Viscosity behavior for the volume fraction of improver for 75 and 85°C

The first step was to take these experimental data to perform a multiple regression statistical study using the STATGRAPHICS program. The results were obtained with a Multiple Regression Analysis with Dependent variable: Pa.s viscosityγ in Table 2 with R^2 =63.3206% and R^2 (adjusted to degrees of freedom) =62.2257, Standard estimate error = 18.5831rr. Mean absolute error = 13.2758, Durbin-Watson statistic = 0.44002 (P=0.0000), Self-correlation of residues = 0.614013.

Table 2. Results obtained with model.

Parameter		Estimated error	P value
C	-323.177	36.4057	0.0001
T	116.45	11.9392	0.0001
x	-2.83968	0.62672	0.0001

The adjusted regression model showing the dependence between viscosity (Pa.s), temperature T (0C) and γpercent in volume x of the flow improver is:

$$\mu = -323{,}177 + 116{,}45T - 2{,}83968x$$

Because the P-value<0.01, there is a statistically significant dependency between the independent and dependent variables with 99% reliability. A self-correlation between the residues is obtained, shown in Figure 7, showing the behavior between the observed and predicted values. These results indicate a nonlinear dependence between the variables, manifested precisely in a non-random distribution of the points plotted in Figure 7 for the straight line of slope equal to 45 degrees, which indicates equality between the value predicted by the model and that observed experimentally.

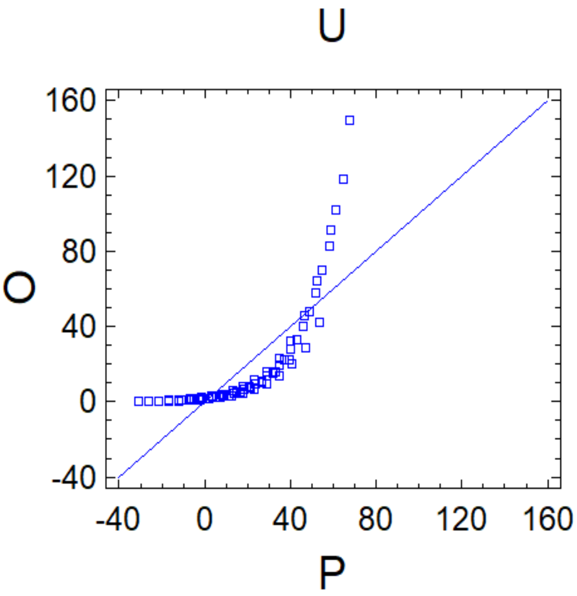

Figure 7. Values predicted by the regression model (P) vs. experimentally observed values (O). In this case, although the regression analysis showed a dependence between viscosity, temperature, and the volume fraction of the improver, it can be pointed out that this dependence is strongly nonlinear.

The results of the estimate are shown in table 3. R-squared = 98.9572 %, R-squared (adjusted to degrees of freedom) = 98.9261 %, Standard estimate error = 3.13332

Average absolute error = 1.84037, Durbin-Watson stratigraphy = 0.514016. Autocorrelation of residuals = 0.742799.

Table 3. Results for model 2

Parameter		Standard Error
A	8.24795E-9	1.23061E-9
B	7.04007	0.113125
k	0.202327	0.00641978

The equation of the fitted model is:

$$\mu = 8{,}24795 \times 10^{-9} exp\left(\frac{7{,}04007 \times 1000}{T}\right)\exp(-0{,}202327x)$$

Figure 8 shows the relationship between the values predicted by the model (P) and the values observed experimentally (O), being able to appreciate that the plotted points are randomly distributed to the line of 45 degrees, representing the equality between the values of the prediction and those observed.

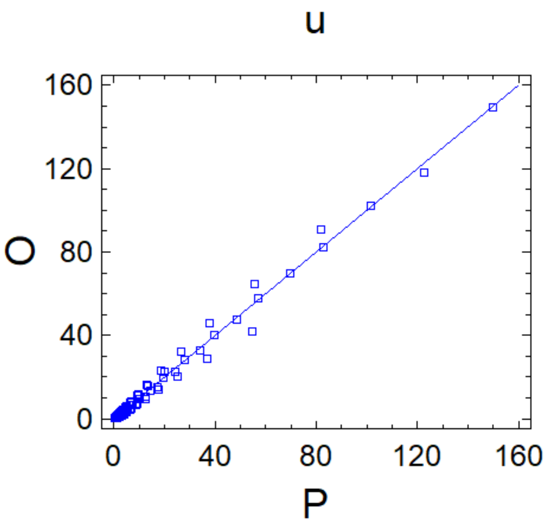

Figure 8. Relationship between the values predicted by the fitted model (P) and the experimentally observed values (O)

The results obtained indicate that the proposed model can analyze the studied system within the specified ranges of values

4. Conclusions

We found a mathematical correlation between oil crude viscosity reduction and an exponential model allowing viscosity prediction when a fluid flow improver is dosed.

Surface tension changes with the compounds and reproduces with a statistical equation with a high correlation.

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