DETERMINATION OF RHEOLOGICAL PROPERTIES OF CRUDE OIL BLENDS

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The article presents an overview of widely applicable methods of calculation of viscosity of liquid mixtures, as well as rheology characterisation of crude oil. It is focused on the evaluation of rheological properties, particularly in order to assess the flow conditions in pipelines at different temperatures and flow rates, especially in the case of waxy crude oils. A specific calculation procedure was proposed and described, suitable for practical purposes, i.e. field implementation in the form of a spreadsheet calculator. Using this method, it is possible to calculate the rheological properties of a mixture of known components at a given temperature over a range of shear gradients corresponding to a specified range of flow rates in a circular pipeline of specified diameter. The proposed calculation method was verified on the properties of specific types of oils and their mixtures.

Keywords: crude oil; mixtures; mixing rules; rheological properties, waxy crude oil

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1. Introduction

1.1. Calculating the viscosity of liquid mixtures

Viscosity is an input parameter included in the calculation of many criteria important in processes used in chemical engineering such as mixing, pipeline transport, and many others. Since liquid mixtures are often handled in real operations, methods have been developed to calculate the viscosity of liquid mixtures with known properties of individual components [1,2]. Examples are mixtures of different types of crude oil or crude oil and fraction added as a diluent, mixtures of heavy residual fraction with crude oil [2] or distillates to mix fuel oil, or dilution of heavy oils to improve transportability [2-10]. In all these cases, the viscosity of the key is to determine the blend or blending ratio at which the viscosity of the blend reaches a predetermined limiting value.

Currently, a large number of equations can be used to determine the viscosity of crude oil mixtures [11,12]. These can be divided into theoretical, semi-empirical, or empirical, depending on whether they have been designed based on knowledge of the physical principles determining the mechanical properties of substances or on regression analysis. The basis for capturing non-Newtonian behaviour is the determination of the viscosity of the dispersion of solid particles, which is also the case for oil below the wax appearance temperature [13]. However, the problem lies in describing the size, content, and properties or morphology of the particles. Some modern approaches use models of the behaviour of substances that involve the description of the internal interactions of the components [14]. These procedures are typically characterised by small requirements on experimental data sets for their validation, but require extensive analytical data for the characterisation of the considered mixtures. Another approach used in recent years is based on the use of neural networks, whose parameters are determined in a "learning" process on large sets of measurements with known samples [15], genetic algorithms [16,17], or using AI methods [18].

Table 1 gives an overview of the reported equations or procedures to calculate the viscosity of mixtures used for the calculations of mixtures of crude oils, oils with light fractions or hydrocarbon fractions. They replace the additivity of the viscosity by the additivity of its function (logarithm, third root, or inverse):

$$f(\eta_{mix}) = \sum_{i=1}^{n} x_i \, f \qquad (1)$$

These simple equations were compared for mixtures of Newtonian and non-Newtonian liquids by Zhang [19], finding that the reliability of the result is significantly lower for non-Newtonian liquids. These equations can be further corrected by additional terms involving binary interaction coefficients (Table 2). However, the coefficients must be determined on knowledge of the viscosities of the binary mixtures; in other words, they assume knowledge of the experimental data for all combinations of the components of the mixtures under evaluation. The procedure involving the use of binary interaction coefficients can be replaced by other types of equations involving smaller numbers of constants [11], but even in this case the values of these constants must be determined on the basis of sufficient experimental data for real mixtures. Exceptionally, when light fractions are used to dilute oils, the resulting effect is expressed as a relative reduction in the viscosity of the diluted component [20]:

$$DVR\% = \frac{(\eta_r - \eta_c)}{\eta_r} * 100 \qquad (2)$$

where index r denotes the reference (original) viscosity and index c the reduced viscosity.

Tab. 1 Simple mixing equations and viscosity blending index (VBI) equations

Additivity	$\eta_{mix} = \sum_{i=1}^{n} x_i \eta_i$	(3)
Power function	$\eta_{mix} = (w_A\eta_A^n + w_B\eta_B^n)^{\frac{1}{n}}$	(4)
Arrhenius equation	$\ln \eta_{mix} = \sum_{i=1}^{n} x_i \ln \eta_i$	(5)
Bingham	$\eta_{mix}^{-1} = \sum_{i=1}^{n} x_i \ln \eta_i^{-1}$	(6)
Koval	$\nu_{mix}^{-0.25} = \sum_{i=1}^{n} \nu_i \eta_i^{-0.25}$	(7)
Kendall-Monroe	$\eta_{mix}^{\frac{1}{3}} = \sum_{i=1}^{n} x_i \ln \eta_i^{\frac{1}{3}}$	(8)
Cragoe*	$\eta_{mix} = 5.10^{-4} \exp\left[1000 \ln\left(\frac{20}{L_{mix}}\right)\right],$ $L_{mix} = \sum_{i=1}^{n}\left[\frac{(1000 \ln 20)}{(\ln \eta - \ln 5.10^{-4})}\right]$	(9)
Wallace and Henry	$\nu_{mix} = \exp\exp\left(\frac{1}{I_{WH}}\right), I_{WH} = w_A I_{WH_A} + w_B I_{WH_B}, I_{WH_i} = \frac{1}{\ln\frac{\nu_i}{0,01}}$	(10)
Reid**	$\upsilon_{mix} = \frac{(x_A + x_B)\nu_A\nu_B}{x_a\nu_A + x_B\nu_B}$	(11)
Chirinos**	$\log\log(\nu_{mix} + 0,7) = w_A \log\log(\nu_A + 0,7) + w_B \log\log(\nu_B + 0,7)$	(12)
Refutas	$VBI_i = 23,097 + 33,469 \log\log(\nu_i + 0,8)$ $VBI_{mix} = w_A VBI_A + w_B VBI_B$ $VBI_{mix} = 10^{10^{\frac{VBI_{mix}-23,09}{33,469}}} - 0,8$	(13)
Chevron	$VBI_i = \frac{\log \nu_i}{3+\log \nu_i},\ VBI_{mix} = \sum_{i=1}^{n} x_i VBI_i,\ \nu_{mix} = 10^{\frac{3VBI_{mix}}{1-VBI_{mix}}}$	(14)
Han (1)	$\log \eta_{mix} = \sum_{i=1}^{n} x_i \log \eta_i + \sum_{j=1}^{n-1}\sum_{k=j+1}^{n} C_{jk}\, x_j\, x_k$ $C_{jk} = -0,0613(\log \eta_i + \log \eta_k) + 0,134$	(15)
Han (2)	$\log\log \eta_{mix} = \sum_{i=1}^{n} x_i \log\log \eta_i + \sum_{j=1}^{n-1}\sum_{k=j+1}^{n} C_{jk}\, x_j\, x_k$ $C_{jk} = -0,064(\log\log \eta_i + \log\log \eta_k) + 0,1706$	(16)

* first the "mixed liquidity" is calculated and in the second step the viscosity of the mixture is calculated

** kinematic viscosity, the relationship is applicable for a two-component mixture

The reliability of the procedures for calculating the viscosity of mixtures decreases with increasing density or decreasing API value, but at the same time the results improve with increasing measurement temperature [21]. This is most likely due to a change in the nature of the oil towards non-Newtonian behaviour. This is more pronounced for heavier oils and at lower temperatures, when a higher proportion of a heterogeneous solid phase in the form of paraffin particles or asphaltenes is present in the oils.

The Arrhenius equation is suitable for high viscosity ratios of the input components, e.g. for the calculation of mixtures of heavy oils or fractions with light oils. The Walther equation is not suitable for mixtures exhibiting the behaviour of non-Newtonian liquids. The Kendall-Monroe model is recommended by the API for calculating the viscosity of mixtures of pure hydrocarbons at low pressures, assuming that the input components do not differ much in the values of their molecular weights, so that the possibilities of its use are limited [22].

Tab. 2 Mixing equations with binary interaction coefficients, additional functions, and other parameters

Modified Arrhenius equation	$\ln \eta_{mix} = \sum_{i=1}^{n} x_i \ln \eta_i + \sum_{j=1}^{n-1} \sum_{k=j+1}^{n} C_{jk} B_{jk} x_j x_k$	(17)
Modified Bingham	$\eta_{mix}^{-1} = \sum_{i=1}^{n} x_i \ln \eta_i^{-1} + \sum_{j=1}^{n-1} \sum_{k=j+1}^{n} C_{jk} B_{jk} x_j x_k$	(18)
Modified Cragoe*	$\eta_{mix} = 5.10^{-4} \exp\left[1000 \ln\left(\frac{20}{L_{mix}}\right)\right],$ $L_{mix} = \sum_{i=1}^{n} \left[\frac{(1000 \ln 20)}{(\ln \eta - \ln 5.10^{-4})}\right] + \sum_{j=1}^{n-1} \sum_{k=j+1}^{n} C_{jk} B_{jk} x_j x_k$	(19)
Walther	$\log\log(\nu_{mix} + C) = w_A \log\log(\nu_{mix} + C) + w_B \log\log(\nu_B + C)$	(20)
Lederer	$\ln \eta_{mix} = x'_A \ln \eta_A + x'_B \ln \eta_B$ $x'_A = \frac{\alpha x_A}{\alpha x_A + x_B}$ $x'_B = 1 - x'_A$	(21)
Van der Wyk	$ln\, \eta_{mix} = x_1^2\, ln\left(\frac{\eta_1 \eta_2}{\eta_{12}^2}\right) + 2x_1\, ln\left(\frac{\eta_{12}}{\eta_2}\right) + ln\, \eta_2$	(22)
Grunberg and Nissan	$\ln \eta_{mix} = m_A \ln \eta_A + m_B \ln \eta_B + m_A\, m_B G_{AB}$	(23)
Tamura a Kurata	$\log \nu_{mix} = m_A \nu_A x_A + m_B \nu_B x_B + 2\nu_{AB}(m_A m_B x_A x_B)^{0,5}$	(24)

* first the "mixed liquidity" is calculated and in the second step the viscosity of the mixture is calculated

The Bingham equation was proposed considering the properties of ideal liquids and shows a large error in the calculated viscosity values of mixtures of oils [23].

1.2. Rheological behaviour of crude oil

Commonly published mixing rules assume that viscosity is an invariant parameter independent (except for temperature) of other conditions. In contrast, for most oils, the viscosity changes with shear gradient (shear rate) due to the fact that they are not homogeneous liquids but mixtures with emulsified water and, in particular, particles and crystals of high molecular weight substances such as linear alkanes (paraffins) and asphaltenes. Thus, especially at low temperatures, these are non-Newtonian liquids, exhibiting pseudoplastic behaviour - the viscosity decreases with increasing shear rate and at zero shear rate these materials exhibit non-zero shear stress, the so-called yield stress [24,25].

At the same time, the viscosity exhibits a time dependence, i.e. thixotropy, which means that the viscosity decreases with time at an imposed shear gradient. This is due to the gradual destruction of the structure formed by the aggregation of paraffin particles.

Since the non-newtonian behaviour of waxy crude oil is caused by the presence of paraffinic particles of different size which can even be interconnected in a three-dimensional network, conditions of paraffin crystallisation can affect the rheology of such crudes. In particular, heating or cooling rates can change the size and morphology, resulting in a change of viscosities, measured under identical conditions [26].

Although some authors still express the flow characteristics of oil mixtures in terms of simple viscosity parameters [27], the above facts suggest the need for a more comprehensive approach taking into account the shear properties of oil.

1.3. Rheological models

The change in viscosity with shear gradient is usually expressed by so-called rheological models, i.e. functions approximating the interdependence of shear stress or viscosity on the shear gradient. Commonly reported models are summarized in Table 3.

Individual models differ mainly in the number of parameters used and their interpretation. Models that can be related to real behaviour when measuring the dependence of shear stress or viscosity on the shear gradient are mainly the theoretical flow limit τ_0, viscosity at zero (η_0) or infinite (η_∞) shear rate.

1.4. The influence of temperature

Theoretical considerations suggest that the viscosity of homogeneous liquids should follow the Arrhenius relation:

$$\eta = A e^{\frac{E_a}{RT}} \tag{25}$$

where A is the pre-exponential factor, E_a the activation energy of the viscous flow, R the gas constant and T the absolute temperature. However, in the case of real liquids, this relationship does not give satisfactory results, so other functions have been developed for engineering purposes.

Tab. 3 Rheological models for non-newtonian fluids

Model	Shear stress	Viscosity		Suitable for materials
Power-law (Ostwald - de Waele)	$\tau = k\dot{\gamma}^{n}$	$\eta = k\dot{\gamma}^{n-1}$	(26)	Dispersions, chocolate
Bingham	$\tau = \tau_0 + k\dot{\gamma}$	$\eta = \frac{\tau_0}{\dot{\gamma}} + k$	(27)	Coatings
Herschel a Bulkey	$\tau = \tau_0 + k\dot{\gamma}^{n}$	$\eta = \frac{\tau_0}{\dot{\gamma}} + k\dot{\gamma}^{n-1}$	(28)	Coatings
Casson	$\sqrt{\tau} = \sqrt{\tau_0} + \sqrt{k\dot{\gamma}}$	$\sqrt{\eta} = \sqrt{\frac{\tau_0}{\dot{\gamma}}} + \sqrt{k}$	(29)	Blood, blood plasma
Luo-Kuang	$\tau = \tau_0 + k\sqrt{\dot{\gamma}} + \eta_\infty\dot{\gamma}$	$\eta = \frac{\tau_0}{\dot{\gamma}} + \frac{k}{\sqrt{\dot{\gamma}}} + \eta_\infty$	(30)	Blood, blood plasma
Sisko	$\tau = \eta_0\dot{\gamma} + k\dot{\gamma}^{n+1}$	$\eta = \eta_0 + k\dot{\gamma}^{n}$	(31)	Plastics, lubricants
Cross	$\tau = \eta\dot{\gamma}$	$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + \alpha\dot{\gamma}^{n}}$	(32)	Dispersions,suspensions
Carreau	$\tau = \eta\dot{\gamma}$	$\eta = \eta_\infty + (\eta_0 - \eta_\infty)(1 + \alpha\dot{\gamma}^{a})^{\frac{n-1}{a}}$	(33)	Polymer melts

They have an empirical basis given by the behaviour of media used mainly in the chemical industry.

To express the effect of temperature on the viscosity of lubricating oils, Walther [28] proposed the following equation, which was further adapted by the ASTM technical standard [29]. It was published along with other equations in SAE [30]:

$$\log\log(\nu + 0{,}7) = a + b\log T \qquad (34)$$

where ν is the kinematic viscosity in mm^2s^{-1} or cSt, a and b are constants and T is the absolute temperature. The strongly nonlinear viscosity versus temperature is also expressed by other proposed relations:

Reynolds equation $\eta = be^{-aT}$ (35)

Slott equation $\eta = \frac{a}{(b + T)^c}$ (36)

Slott-Fahrenheit equation $\eta = k.t^{a}$ (37)

Vogel equation $\eta = ae^{\frac{b}{T-c}}$ (38)

where η is the dynamic viscosity (mPa.s, cP).

1.5. Practical implications: calculation of viscosity for determination of pressure drop in pipes

Practically the only source of pressure drop in a horizontal pipe is the internal friction in the flowing media. The pressure drop calculation depends on the flow regime, which is given by the Reynolds number. In the laminar flow regime, characterized by sub-critical Re, the friction factor equals 64/Re, which results in well-known Hagen-Poiseuille equation:

$$\Delta p = \frac{32\eta L v}{D^2} \qquad (39)$$

where L and D are the pipe length and diameter, respectively, and the pressure drop is directly proportional to the dynamic viscosity.

In the case of a fully developed turbulent regime, the influence of viscosity can be almost eliminated at high flow velocities and high internal surface roughness. However, crude oil pipeline transport is often characterised by relatively low flow velocities and transient or even laminar flow regimes can occur, where the viscosity effect is significant, directly influencing the pump energy consumption necessary for flow assurance.

Since viscosity depends on the shear gradient for non-Newtonian fluids, it is necessary to determine a representative shear rate in the pipe at a given flow rate. Obviously, the shear rate is given as follows:

$$\dot{\gamma}_{rep} = \frac{\tau_w}{\eta_{rep}} \qquad (40)$$

where τ_w is the shear stress on the pipe wall:

$$\tau_w = \frac{\pi D}{4L} \qquad (41)$$

However, the representative viscosity, which is unknown, cannot be used for the calculation of the representative shear rate. Therefore, it must be derived only from the flow rate (or velocity) in this phase of the calculation. For Newtonian fluids, the representative shear gradient is the shear gradient at the pipe wall, resulting from the calculation of the (parabolic) velocity profile in the pipe [31]:

$$\dot{\gamma}_{rep} = \frac{8v}{D} \qquad (42)$$

where v is the flow velocity and D is the inner diameter of the pipe. A slight modification is also given in the literature:

$$\dot{\gamma}_{rep} = \frac{2\pi v}{D} \qquad (43)$$

In the case of non-Newtonian fluids, this expression is not entirely accurate because the variable viscosity value with the shear gradient results in a different shape of the velocity profile in the pipe, and, therefore, mean (representative) value of shear rate. At the same time, these relationships are only valid for laminar flow. However, since the following procedure for estimating the viscosity and Reynolds number aims to describe the behaviour in the transition to laminar flow, this method of calculating a representative shear gradient will be further used.

2. Proposed method for calculating the rheological properties of crude oil mixtures under conditions of pipeline transport

2.1. Calculation approach

It is assumed that the input for calculating the viscosity of oil mixtures will be data collected for individual components, i.e., viscosities determined at several shear gradient values at given temperatures. The problem can occur when the input data for the mixture components can be provided at different range of shear rates or different temperatures. The proposed procedure for the calculation of the viscosity of mixtures includes the following steps:

1) Approximation of the rheogram (viscosity or shear stress vs. shear rate) of the mixture components using a suitable rheological model. The measured rheological data of the components are converted into an approximation function (rheological model) in a suitable form. This shape should preferably include constants in the form of a component representing the viscosity at (infinitely) high shear gradient and a component corresponding to the interflow.

2) Correction of component viscosities to the target temperature. Because the measurement temperature of the component viscosities may not agree with the final temperature for which the mixture viscosities are determined, they must be corrected by an appropriate relationship.

3) Calculation of the rheological model parameters of the mixture based on the parameters of the components corrected to the target temperature. For this purpose, a selected type of mixing equation suitable for describing the viscosity of oil mixtures should be used.

4) Calculation of viscosities of the mixture for given range of shear rate (according to the engineering needs, particularly at representative shear rates for a pipe of known diameter at given flow rates). For this purpose, the rheological model of the mixture obtained in the previous step will be used.

The input data for the calculation are the viscosities of the oils determined at five values of the shear gradient. The available rheological models differ in the number of constants of the approximation function, and it is clear that a larger number of parameters gives better assumptions for a successful approximation of the real data. However, in such a case the approximation function (successfully) interleaves deviations due to the mere error of one or more measurements, and in addition, worse behaviour can be expected in case of an extrapolation attempt. Among the rheological models (Chapter 2.3), those that do not require more than 3 parameters were selected and compared on data for oils measured in the laboratory. An example of the results is shown in Figure 1, while in other cases the three-parameter model also gave very good results, according to Sisko. Slightly better results were obtained only with the four-parameter models, but these were excluded from further processing for the reasons given above.

2.2. Selection of a rheological model based on data knowledge for real oil samples

Example input data for the calculation are the viscosities of the oils determined at five shear rate values. The available rheological models (Table 3) differ in the number of constants of the approximation function, and it is clear that a larger number of parameters gives better assumptions for a successful approximation (or even extrapolation) of the real data. On the other hand, there is a risk that in such a case the model (successfully) approximates errors of one or more measurements., and even worse behaviour can be expected in case of an extrapolation attempt. Among the rheological models (Chapter 2.3), those that do not require more than 3 parameters were selected and compared on the data for the sample oils used for the study. An example of the results is shown in Figure 1, while in other cases the three-parameter model also gave very good results, according to Sisko. Slightly better results were obtained only with the four-parameter models, but these were excluded from further processing for the reasons given above.

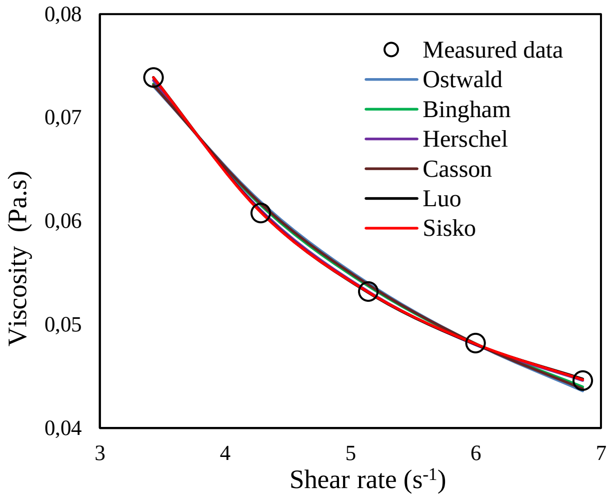

Figure 1 Sample viscosity data (waxy crude A) at 4 ° C approximated using different rheological models

2.3. Viscosity correction to temperature

The correction of viscosity (if a target temperature for a mixture differs from the measurement temperature of components) was carried out using the equation successfully used in the viscosity versus temperature measurements of a wide set of waxy crudes:

$$\eta = \eta_{ref} \cdot 10^{a \Delta t} \qquad (44)$$

where η is the corrected viscosity, η_{ref} is the viscosity measured at the original temperature, a is a constant and Δt is the difference between the temperature for which the corrected viscosity is calculated and the measurement temperature η_{ref}.

Correction was applied to the constant η_0 (viscosity at infinite shear gradient) of the Sisko equation, used as a rheological model.

2.4. Calculation of the mixture rheological model

The Han relationship (15), was applied to the η_0 constant (viscosity at infinite shear gradient) of the Sisco equation, used as a rheological model. The Han equation was chosen because it was verified on a relatively large set of oil blend samples [11]; the choice was also dictated by the absence of unknown binary interaction coefficients, additional functions, or other additional parameters whose values would have to be obtained from measurements for a sufficient number of real mixtures.

The mixture parameter k, representing the fluid yield stress, was calculated from those for the mixture components using the Arrhenius equation (5), since the data from the literature [19] for the yield stress were found to be consistent with the additivity of the logarithm of the flow limit.

2.5. Calculation of the flow curve of a mixture

Next, the viscosities at selected representative shear gradient (see Section 1.5) corresponding to the diameter of the pipe in the specified flow range were calculated from the parameters of the rheological model of the mixture.

2.6. Example calculation results

As an example, rheological data of two components A and B measured at different temperature and representative shear rates corresponding to certain flow conditions (Tab. 4) were used to calculate viscosities at the specified temperature and flow in a smaller diameter pipe.

Calculated and experimental results for the above-mentioned pair of crude oils and flow conditions is shown in Figure 2. There, the input data for mixture components were provided at different temperatures, and the resulting output viscosity was needed for different range of shear rates given by the need to predict pressure drop in a pipe of smaller diameter, thus at higher flow rates.

3. Conclusions

The proposed calculation procedure allows the viscosity of oil mixtures at given temperatures and shear rates to be determined based on the input data on the components of the mixture. It has been taken into account that consistent data are not available in all cases, so the method can use viscosities of mixture components measured in different shear rate ranges at different temperatures.

Tab. 4 Measured properties of components and conditions for the mixture results

Crude oil (blend)	A	B	Mix
Measurement			
Density @ 15 °C (kg.m^{-3})	826.5	808.9	-
Pour point (°C)	-3	-12	-
Viscosity measurement temperature (°C)	4	10	-
Shear rate at			-
Pipe diameter (mm)	700	700	
Flow rate min (m^3.h^{-1})	400	400	
Flow rate max (m^3.h^{-1})	800	800	
Calculation			
Viscosity calculation temperature (°C)	-	-	10
Shear rate at			
Pipe diameter (mm)	-	-	500
Flow rate min (m^3.h^{-1})	-	-	400
Flow rate max (m^3.h^{-1})	-	-	800

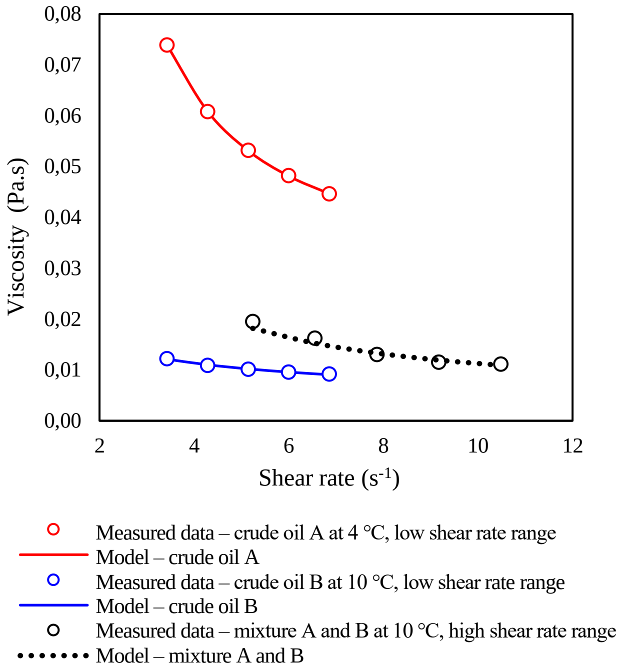

Figure 2 Example of the calculation inputs and results for different temperature and shear rate range
Sisko equation was used to approximate input data

It does not include any interaction coefficients, so it does not need to be tuned on data sets for known pairs of raw materials, which makes the procedure easy to use. For the same reason, it is not as accurate as more sophisticated equations, especially for highly non-Newtonian liquids. The procedure also does not account for the thermal and mechanical history of the samples, which could be of importance, especially in the case of viscous waxy samples.

On the other hand, the procedure provides a very good estimate for practical purposes - the calculation of pressure drop in a pipe - particularly when combined with a correct determination of the representative shear rate in the pipe at given flow rates.

Nomenclature

C	Walther equation constant
C_{jk}, B_{jk}	viscosity mixing parameter
DVR%	relative viscosity reduction
G_{AB}	empirical parameter (Grunberg Nissan eq.)
k	rheology model constant
L_{mix}	viscosity/weight fraction function
n	rheology model constant
VBI	viscosity blending index
w	mass fraction
x	molar fraction
α	empirical parameter
$\dot{\gamma}$	shear rate
ρ	density
η	dynamic viscosity
η_{12}	coefficient of internal friction (Van der Wijk equation)
η_0	rheology model parameter, zero shear viscosity
η_∞	rheology model parameter, infinite shear viscosity
ν	kinematic viscosity
τ	shear stress
τ_0	rheology model parameter, yield stress

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