

SIMULATION OF CHANGE IN DENSITY AND VISCOSITY
OF CRUDE OIL WHEN MIXING*Konstantin Zamikula¹, Olena Tertyshna¹ ✉, Oleg Tertyshny¹, Petro Topilnytskyi²*<https://doi.org/10.23939/chcht16.03.469>

Abstract. The deviation of density and viscosity values of oil mixtures from those calculated according to the additivity rule has been examined. Mathematical models have been developed for determining the properties of mixtures with different compositions taking into account the group composition of the source components. Mixtures containing components with a high content of alkanes and a low content of arenes have been found to be characterized by extreme deviations of density towards the maximum and kinematic viscosity towards the minimum. The created models were validated and compared with existing methods of describing the oil physicochemical properties. An approach to the creation of optimal from a technological point of view formulation for compounding oil of different types is proposed.

Keywords: density, kinematic viscosity, oil mixing, mathematical model, group composition.

1. Introduction

High-quality crude oil affects the efficiency, durability, and reliability of pipelines, as well as the properties of products obtained from it. In this regard, the definition and forecasting of physicochemical parameters of prepared oil to increase the efficiency of its refining process is an important and urgent task.

With the increase in heavy and ultra-heavy oil refining volume, the values of viscosity and density became more important characteristics in determining the operation modes of compounding units.

The understanding of the initial components mutual influence on the density and viscosity of oil mixtures was

based on studies of simple ideal solutions, and as a result, errors occurred in the calculations of technological regimes.¹⁻⁵

There is a problem with predicting the viscosity of the formed oil mixtures. The linear law of mixing with respect to viscosity is not applicable. The greater the difference in the viscosity of the initial components, the greater the error of the resulting indicator.⁶ Mathematical models, which describe the viscosity of oil mixtures, take into account the degree of dilution, the corresponding viscosity, and density of oil and diluents.⁷

The formula for calculating the mixture viscosity was first proposed by Arrhenius⁶ in 1887 and became the starting point for subsequent researchers.⁸

The equation of exponent,⁹ which is based on the values of molar fractions of the mixture components, is in close agreement with the experimental results. Later developed more complex equations include parameters which are determined on the basis of experimental measurements. The corrections improved the convergence between the predicted and experimental values of viscosity.¹⁰⁻¹¹

The dependences of viscosity of heavy oils and oil fractions mixture described by several authors¹²⁻¹⁴ showed a higher correlation between the predicted and experimental values, but in a narrow temperature range. Mago¹⁵ found that Walter's logarithmic linear law does not provide sufficient flexibility to determine the viscosity of oil.

The nonlinearity of changes in properties is difficult to describe mathematically, however, it is the calculation of deviations that allows us to predict the behavior of systems and reduce the error in technological calculations.

The universal formula for determining the viscosity of the oil mixture does not exist, because it significantly depends on the physical and chemical properties of the initial components to be mixed. Mathematical dependences, which adequately describe the physical essence of the mixing process, in one or another way contain empirical coefficients which are determined experimentally. In addition, existing formulas do not take into

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account the possible manifestation of non-Newtonian nature of the components to be mixed. The rheological characteristics of the mixture vary greatly depending on the components concentration and their group composition, so the estimated viscosity for the mixtures will not be entirely correct. Existing dependencies are focused only on binary mixtures, and their direct use for multicomponent mixtures by the usual addition of new components leads to significant errors.

The aim of this work is to develop a mathematical model for determining the density and viscosity of oil during mixing, which takes into account the non-additive nature of changes in indicators depending on their group composition.

For mathematical modeling of physicochemical properties of oil fractions, we proposed and tested an approach with a preliminary division of the set of the same structure individual hydrocarbons into separate hydrocarbon groups: alkanes, cycloalkanes and arenes.¹⁶

2. Experimental

2.1. Materials and Methods

As components of the model, we used three types of oil, physicochemical parameters of which are given in Table 1. The group composition of each component was determined by the n-d-M method.¹⁶ The density and refractive index of the components were determined according to EN ISO 3675: 2012, ASTM D1218-02 (2007). The average molecular weight of the components was determined by the cryoscopic method.

The essence of the method is to determine the decrease in the temperature of crystal formation (depression) of the analyte solution in comparison with pure solvent and to determine the molecular weight of this substance by calculations.

Benzene was used as the most available solvent, which dissolves oil and petroleum products well. The crystal formation temperature of pure benzene, as well as the temperature of analyte solutions dissolved in benzene, were determined.

The calculation of molecular weight was carried out according to the formula:

$$M = \left(\frac{K \cdot g \cdot 1000}{G \cdot \Delta t} \right) \quad (1)$$

where K is the cryoscopic constant (for benzene 5.49 °C (mol/kg)⁻¹); g is the weight of dissolved analyte, g; G is the solvent weight, g; Δt is the decrease in crystal formation temperature, °C.

To describe the properties of two-component and multi-component mixtures, we used the "quasi-additivity" model.¹⁷

In accordance with the proposed model, the properties of the mixture are presented in the form of:

$$X_{\Sigma} = \sum_1^n \psi_j^* \cdot X_j \quad (2)$$

where X_{Σ} is the property of the mixture; X_j is the property of the j-th component; ψ_j^* is the "non-additive" part of the j-th component.

$$\psi_j^* = \psi_j \cdot \frac{\beta_j}{\sum_1^n \psi_j \cdot \beta_j} \quad (3)$$

where ψ_j is a share of the component in the mixture; β_j is the "quasi-additivity" coefficient calculated according to formula (4):

$$\beta_j = \beta_j^a \cdot \psi_j^a + \beta_j^n \cdot \psi_j^n + \beta_j^{ar} \cdot \psi_j^{ar} \quad (4)$$

where β_j^a , β_j^n and β_j^{ar} are the coefficients of "quasi-additivity" of alkanes, cycloalkanes and arenes of the j-th component, respectively; ψ_j^a , ψ_j^n and ψ_j^{ar} are the share of alkanes, cycloalkanes and arenes in the j-th component, respectively.

For density the coefficients of "quasi-additivity" are calculated from Eqs. (5-7):

$$\beta_j^a = 1.3476 \ln(M_j) - 4.9815 \quad (5)$$

$$\beta_j^n = 0.9854 \cdot \ln(M_j) - 3.512 \quad (6)$$

$$\beta_j^{ar} = -7.755 \cdot 10^{-5} \cdot M_j^2 + 0.0198 \cdot M_j - 0.525 \quad (7)$$

where M_j is the average molecular weight of the j-th component.

For viscosity the coefficients of "quasi-additivity" are calculated from Eqs. (8-10):

$$\beta_j^a = 12.592 \cdot M_j^{-0.578} \quad (8)$$

$$\beta_j^n = -0.375 \cdot \ln(M_j) + 1.8961 \quad (9)$$

$$\beta_j^{ar} = -0.62 \cdot \ln(M_j) + 3.1855 \quad (10)$$

Eqs. (5-10) are approximations of the experimental results of the experiments represented by Kravtsov et al.¹⁷

To realize the task, the temperatures of the mixed components were taken into account.

Table 1. Physicochemical parameters of oil

Indicator	Component 1	Component 2	Component 3
Density at 293 K, kg/m ³	828.3	861.3	911.6
Kinematic viscosity at 293 K, mm ² /s	5.273	13.287	121.364
Content of alkanes, vol %	23	25.6	40.1
Content of cycloalkanes, vol %	71.5	69.8	57.9
Content of arenes, vol %	5.5	4.6	2.0

The density of the mixture for certain mass fractions of the components was determined by the formula:

$$\frac{1}{\rho_{\Sigma}(t_{\Sigma})} = \sum_{j=1}^n \frac{\psi_j^*}{\rho_j(t_j)} \quad (11)$$

where ρ_{Σ} is the mixture density, kg/m³; t_{Σ} is the mixture temperature, °C; ρ_j is the density of the j-th component, kg/m³; t_j is the temperature of the j-th component, °C.

To determine the temperature t_{Σ} of the mixture and its density ρ_{Σ} in Eq. (11) the specific heat of each component before mixing was taken into account.

The actual heat capacity of oil at a given temperature, in the first approximation, was calculated by the formula:

$$c_j^p = 1674.7 \cdot (2.025 - \hat{\rho}_j^t) \quad (12)$$

where c_j^p is a specific isobaric heat capacity of the j-th component, J/(kg·K); $\hat{\rho}_j^t$ is a relative density of the j-th component at temperature t.

The heat capacity of petroleum products mixture is equal to the additive sum of the heat capacities of the components that form it:

$$c_{\Sigma}^p \cdot t_{\Sigma} = \sum_{j=1}^n c_j^p \cdot \psi_j^* \cdot t_j \quad (13)$$

where c_{Σ}^p is a heat capacity of the mixture at the temperature t_{Σ} .

The mixture temperature is calculated using Eq. (14):

$$t_{\Sigma} = \frac{1}{c_{\Sigma}^p} \sum_{j=1}^n (c_j^p \cdot \psi_j^* \cdot t_j) \quad (14)$$

To calculate c_{Σ}^p we used Eq. (15):

$$c_{\Sigma}^p = 1674.7 \cdot (2.025 - \rho_{\Sigma}(t_{\Sigma})) \quad (15)$$

Since in the general case the mixture temperature differs from the temperature of the components to be mixed, Eq. (11) transforms into Eq. (16):

$$\frac{1}{\rho_{\Sigma}(t_{\Sigma})} = \sum_{j=1}^n \frac{\psi_j^*}{\rho_j(t_{\Sigma})} \quad (16)$$

where $\rho_j(t_{\Sigma})$ is the density of the j-th component of the mixture at the mixture temperature t_{Σ} calculated by the formula:

$$\rho_{\Sigma}(t_{\Sigma}) = \frac{\rho_j}{1 + \alpha_j \cdot (t_{\Sigma} - t_j)} \quad (17)$$

The coefficient of thermal expansion α_j of the j-th component was determined by correlation dependence:¹⁷

$$\alpha_j = 10^{-3} \left(\frac{(\hat{\rho}_j)^2}{0.58 + \hat{\rho}_j \cdot (t_j \cdot 10^{-3} - 1.2 \cdot (\hat{\rho}_j - 0.68))} - \frac{t_j - 20}{1000} \right)^{-1} \quad (18)$$

Taking into account Eq. (14), the formula (15) looks as following:

$$c_{\Sigma}^p = 1674.7 \cdot \left(2.025 - \frac{1}{\sum_{j=1}^n \frac{\psi_j^*}{\rho_{\Sigma}(t_{\Sigma})}} \right) \quad (19)$$

To determine the density of the mixture of petroleum products $\rho_{\Sigma}(t_{\Sigma})$ and its temperature t_{Σ} , a mathematical model is proposed:

$$\rho_{\Sigma}(t_{\Sigma}) = \frac{1}{\sum_{j=1}^n \left(\frac{\psi_j^*}{\rho_j} \cdot (1 + \alpha_j \cdot (t_{\Sigma} - t_j)) \right)} \quad (20)$$

$$t_{\Sigma} = \frac{1}{1674.7} \frac{\sum_{j=1}^n c_j^p \cdot \psi_j^* \cdot t_j}{\left(2.025 - \left(\sum_{j=1}^n \frac{\psi_j^* \cdot (1 + \alpha_j \cdot (t_{\Sigma} - t_j))}{\rho_j} \right)^{-1} \right)} \quad (21)$$

The mixture temperature (Eq. 21) and its density (Eq. 20) were found by the method of successive iterations.

The kinematic viscosity of each component of the mixture at the mixture temperature was determined according to Eq. (22):

$$v_j^{t_{\Sigma}} = \frac{1000}{Z \cdot \rho_j^{t_{\Sigma}}} \cdot \left(\frac{Z \cdot v_j^n \cdot \rho_j^n}{1000} \right)^{\chi} \quad (22)$$

where $v_j^{t_{\Sigma}}$ and v_j^n is the kinematic viscosity of the j-th component at the temperature t_{Σ} and initial temperature, respectively, mm²/s; $\rho_j^{t_{\Sigma}}$ and ρ_j^n is the density of the j-th component at the temperature t_{Σ} and initial temperature, respectively, kg/m³.

The parameter χ was calculated by the formula:

$$\chi = \frac{1}{1 + \alpha(t_{\Sigma} - t_0) \cdot \lg(Z \cdot v_j^n \cdot \rho_j^n)} \quad (23)$$

where α and Z are coefficients, the values of which are taken as:

$$Z = 10, \alpha = 0.00252 \text{ at } v_j^n \cdot \rho_j^n \geq 1000 \quad (24)$$

$$Z = 100, \alpha = 0.00144 \text{ at } 10 \leq v_j^n \cdot \rho_j^n < 1000 \quad (25)$$

$$Z = 1000, \alpha = 0.00076 \text{ at } v_j^n \cdot \rho_j^n < 10 \quad (26)$$

The algorithm for calculating the kinematic viscosity at the temperature t_{Σ} is the following:

a) The viscosity value according to Eq. (22) is calculated with coefficients corresponding to the value of the known oil viscosity at the initial temperature. If the calculated value of the relative viscosity at the temperature t_{Σ} did not exceed the range of viscosity changes corresponding to the selected coefficients α and Z , the calculation ends.

b) If $v_j^n \cdot \rho_j^n \geq 1000$ and $10 \leq v_j^{t_{\Sigma}} \cdot \rho_j^{t_{\Sigma}} < 1000$, the viscosity of the component is calculated by the formula:

$$v_j^{t_{\Sigma}} = \frac{\exp\left(\frac{5}{1+7.2 \cdot \frac{t_{\Sigma} - t_{1000}}{1000}} - 2\right)}{\rho_j^{t_{\Sigma}}} \quad (27)$$

where t_{1000} is the boundary temperature, at which $v_j \cdot \rho_j = 1000$

$$t_{1000} = t_j^0 + 99.2 \cdot \left(1 - \frac{4}{1 + \lg(v_j^{t_0} \cdot \rho_j^{t_0})} \right) \quad (28)$$

c) If $v_j^n \cdot \rho_j^n \geq 1000$ and $v_j^{t_{\Sigma}} \cdot \rho_j^{t_{\Sigma}} < 10$, the component viscosity is calculated as:

$$v_j^{t_{\Sigma}} = \frac{\exp\left(\frac{5}{1+3.04 \cdot \frac{t_{\Sigma} - t_x}{1000}} - 3\right)}{\rho_j^{t_{\Sigma}}} \quad (29)$$

where t_x is an accessory parameter.

d) If $10 \leq v_j^n \cdot \rho_j^n$ and $v_j^{t_x} \cdot \rho_j^{t_x} < 10$, the component viscosity is calculated as:

$$v_j^{t_x} = \frac{\exp\left(\frac{5}{1+3.04 \cdot \frac{t-t_{10}}{1000}} - 3\right)}{\rho_j^{t_x}} \quad (30)$$

where t_{10} is the boundary temperature, at which $v_j \cdot \rho_j = 10$

After calculating viscosity of each component at the mixture temperature, the viscosity of the mixture was calculated according to Eq. (2).

3. Results and Discussion

To validate the developed mathematical model,¹⁸ the density and viscosity of mixtures consisting of components 1, 2 and 3 in different ratios were determined (Table 2) and the results of calculations were compared with experimental values.

The results of experimental determination of density and viscosity, their calculation using the developed mathematical model and their visualization are shown in Fig. 1.

Dots on triangular diagrams indicate experimentally obtained values of physicochemical properties of oil mixtures, colored stripes – the results of calculations based on developed mathematical models.

To compare the adequacy of the developed models with the traditional ones, a regression analysis of the obtained results in the STATISTICA 10.0 package was performed. The results of the analysis are given in Table 3.

The values of the Student's criterion of the proposed calculation model (0.104) and the traditional additive model (0.764) are much lower than the critical value (2.093), which indicates the adequacy of both models for technological calculations. However, the developed model, which takes into account the group composition, approximates the experimental results better than the traditional additive model.

Table 2. The composition of mixtures of oil samples for experimental verification of properties

Mixture number (N)	Amount, wt %			Mixture number (N)	Amount, wt %		
	Component 1	Component 2	Component 3		Component 1	Component 2	Component 3
1	0	0	100	11	30	50	20
2	0	50	50	12	33,3	33,3	33,4
3	0	100	0	13	50	0	50
4	10	10	80	14	50	20	30
5	10	80	10	15	50	30	20
6	20	20	60	16	50	50	0
7	20	30	50	17	60	20	20
8	20	50	30	18	80	10	10
9	20	60	20	19	100	0	0
10	30	20	50				

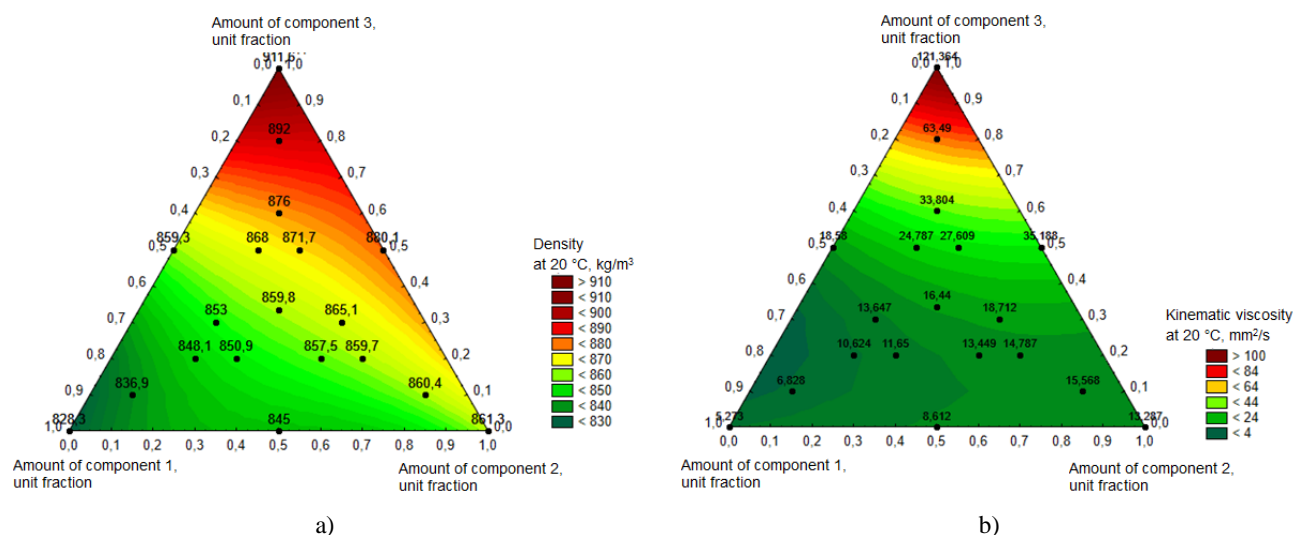


Fig. 1. Comparison of experimental and calculated results for density (a) and viscosity (b) of the mixture depending on its composition

Table 3. Comparison of adequacy criteria of different mathematical models for determining the density and viscosity of mixtures

Mathematical model	Student's criterion at $p = 0.05$	
	Calculated (t)	Critical (t_{cr}) for $N = 19$
Additive calculation of mixture density	0.104	2.093
Calculation of mixture density taking into account the components group composition	0.764	
Additive calculation of mixture kinematic viscosity	3.028	
Calculation of mixture kinematic viscosity according to Walter model	4.639	
Calculation of mixture kinematic viscosity taking into account the components group composition	1.715	

The value of the Student's criterion (4.639) for the Walter model, which is usually used in engineering calculations, is greater than the critical value and even greater than in the additive calculation (3.028). The reason is the presence of an empirical component in the Walter model making it impossible to use this model for the description of properties of high-viscous oil mixtures.

The proposed model of viscosity calculation, which takes into consideration the components group composition, approximates the experimental results with the Student's criterion of 1.715, which is significantly less than the critical value. Thus, this model is adequate for any viscosity range.

To facilitate the prediction when using the results of modeling the physicochemical properties of oil mixtures of any composition, we carried out an approximation of a series of experiments (Table 2) using the properties of simplex space.¹⁹

The composition of the mixture with any number of components is determined by a simplex, *i.e.*, the simplest geometric figure having k vertices in $(k-1)$ dimensional space. For three-component mixtures a triangle of concentrations is such a figure. Different Scheff polynomials were used to construct the composition-property model, followed by adequacy checking at every experimental point.²⁰

Equations based on the Scheff polynomial of the third order of the following type are adequate for the presented data on the density and viscosity of oil mixtures:

$$\begin{aligned} \rho = & 828.2951 \cdot x + 861.2963 \cdot y + 911.5634 \cdot z + \\ & + 1.7638 \cdot x \cdot y - 40.9864 \cdot x \cdot z - 27.0622 \cdot y \cdot z - \\ & - 0.0442 \cdot x \cdot y \cdot (x - y) + 9.7274 \cdot x \cdot z \cdot (x - z) + \\ & + 7.0381 \cdot y \cdot z \cdot (y - z) + 11.2992 \cdot x \cdot y \cdot z \quad (33) \end{aligned}$$

$$\begin{aligned} \nu = & 8.2029 \cdot x + 15.1592 \cdot y + 113.2284 \cdot z + \\ & + 3.4869 \cdot x \cdot y - 172.3395 \cdot x \cdot z - \\ & - 128,7617 \cdot y \cdot z + 130,3224 \cdot x \cdot y \cdot z \quad (34) \end{aligned}$$

where x , y and z are the concentrations of components 1, 2 and 3, respectively, unit fractions.

The simulation results were used to create formulations of oil mixtures with optimal composition, *i.e.*, the composition with the maximum amount of heavy

component 3, but with the minimum values of kinematic viscosity and density. Thus, the addition of 20 % of light components 1 and 2 as a diluent does not significantly reduce the density of the mixture, but reduces the viscosity of the mixture almost twice – from 121.36 to 63.49 mm²/s, which significantly facilitates working conditions for technological equipment and reduces its energy consumption.

4. Conclusions

Based on experimental studies on changes in density and viscosity of oil mixtures with different component proportions, the dependences of changes in density and kinematic viscosity of the mixture on the group composition of the components were obtained.

The obtained results allowed to establish the regularities of deviation of physicochemical properties of oil mixtures in the whole range of component composition.

The deviation of the mixture density from the additively calculated one toward the higher values indicates that the effect of "non-additive part" of the heaviest component 3 is greater than its actual value in the mixture. The deviation of the mixture viscosity from the additively calculated one towards the lower values, on the contrary, suggests that the effect of "non-additive part" of the most viscous component 3 is less than its actual value in the mixture, due to the presence in this component the maximum amount of alkane hydrocarbons (40.1 %) and the minimum amount of arenes (2.0 %).

The established regularities allow to reduce the number and duration of laboratory experiments in the study of rheological properties of oil and its mixtures for more efficient and rational planning of technological processes taking into account the influence of the oil environment nature.

An approach to the creation of optimal, from a technological point of view, formulation for compounding oils of different types is proposed.

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Received: November 17, 2021/ Revised: December 06, 2021 / Accepted: January 12, 2022

МОДЕЛЮВАННЯ ЗМІНИ ГУСТИНИ ТА В'ЯЗКОСТІ НАФТОВОЇ СИРОВИНИ ПРИ ЗМІШУВАННІ

Анотація. Розглянуто відхилення значень густини та в'язкості нафтових сумішей від розрахованих за правилом адитивності. Розроблено математичні моделі визначення даних властивостей сумішей різного складу з урахуванням групового складу вихідних компонентів. Встановлено, що для сумішей, які містять компоненти з великим вмістом алканів та малим вмістом аренів характерні екстремальні відхилення густини в бік максимуму та кінематичної в'язкості в бік мінімуму. Проведено перевірку створених моделей на адекватність та порівняння із існуючими методами опису фізико-хімічних властивостей нафти. Запропоновано підхід до створення оптимальних, із технологічної точки зору, рецептур компаундування нафти різних типів.

Ключові слова: густина, кінематична в'язкість, змішування нафти, математична модель, груповий склад.