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Mathematical Description of Fractioning Curves as a Basis for Balance Modeling of Oil Feedstock Refining Processes

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Abstract

The article gives the analysis of the existing models for mathematical description of oil feedstock fractional composition, comparison of theoretical and experimental data for oils and oil residues, including those contained in oil sludges of reservoir type. It has been shown that modeling of oil feedstock fractional composition by using empirical dependencies allows rather accurately defining the actual values of oil feedstock of various heaviness.

Keywords: oil, oil wastes, fractional composition, mathematical description, boiling point.

INTRODUCTION

The oils of various origin and from various fields represent unique blends of natural compounds mainly represented by hydrocarbons of various structure with admixture of heteroorganic components boiling out within wide temperature ranges. The distribution of constituent compounds by their boiling points, built according to the results of distillation of samples and supplemented by the results of determining physical and chemical characteristics of fraction (density, molecular weight, elemental and microelemental composition, etc.), is a basis for solving a wide range of tasks.

One of the primary tasks is preliminary sorting (with respect to possible blending of oils and their fractions) and selection of optimal types and processes to refine oil feedstock of various origin [1], in particular, the organic part of oil wastes (oil sludges) [2], to assess the yield of overall product range.

The information concerning the fractional composition of oils and oil products allows calculating their most relevant performance characteristics, defining the type of refining and establishing the required depth for taking distillate fractions in refining flow diagrams.

To formalize these tasks, it is required to develop a mathematic description of curves for dividing fractions by TBPs – true boiling points, allowing for the use of any oil and oil sludge fractions when modeling refining processes within the limits of oil initial boiling t_{IBP} and end t_{EBP} boiling points. TBP curves are used to define the fraction composition of oils, calculating physical, chemical and performance characteristics of oil products and process parameters of distillation and rectification of oil blends.

The article gives the applicability analysis of the existing approaches to mathematical description of data concerning boiling points of oil feedstock components, comparison of theoretical and experimental data for fractional composition of oils, oil residues, including those contained in oil sludges of reservoir type that are annually generated in Russia in the amount of more than 3 million tons.

METHODOLOGY

TBP curves are obtained in the conditions of laboratory and industrial distillation by means of sampling distillates described by initial and end boiling point limits with constantly increasing boiling point. While low-boiling components are distilled, the residue is enriched with high-boiling components. Fractions below 300 °C are sampled at atmospheric pressure, followed by irreversible thermal decomposition (cracking) of high-molecular hydrocarbons, so the distillation of high-boiling hydrocarbon compositions is done in the conditions of technical vacuum, and laboratory data obtained is recalculated into the values corresponding to normal pressure. As a rule, distillation is performed by using standard rectifiers equipped with distillation columns. The standards regulate the distillation conditions: rate of distillation, residual pressure, quantity of reflux, etc. Currently, the oil fractional composition is determined under GOST 11011-85.

The article considers some empirical models described in literature [3-5, 6] referring to mathematical description and interpretation of curves for oils and oil products fractional distribution. In particular, empirical description of integral distillation curves was applied with the distillation cuts share (v) equal to 1 when reaching the end boiling point (t_{EBP})), according to the analysis of a typical curve for oil TBP that is represented as follows (as given in [3-5]):

$$v = \frac{a\tau^k}{1+a\tau^k}, \quad (1)$$

where v = distillation cuts share; a and k = coefficients, the values of which are determined by processing TBP curves; t = normalized temperature parameter determined upon the formula:

$$\tau = \frac{t-t_{IBP}}{t_{EBP}-t_{IBP}}, \quad (2)$$

where t = current temperature value.

If we take the logarithm of the model (1), we obtain the following linearized dependency:

$$\ln\left(\frac{v}{1-v}\right) = \ln a + k \ln \tau, \quad (3)$$

On the one hand, fulfilling this dependency proves the adequacy of describing the oil distillation curve under the model (1) as described above, and on the other hand, it makes it possible to determine the parameters *a* and *k* presented in the mathematical description of the model.

The paper also adopts a general model obtained for plant-tank oil and oil product streams in arbitrary parts of oil refining flow diagrams as proposed by TOTAL (Compagnie Française Pétroles) personnel and described by the following formula according to [6]:

$$G(t_b) = 100\left\{1 - \exp(-(t_r/\alpha)^\beta)\right\}, \quad (4)$$

where $G(t_b)$ = share of product boiling out before t_b (expressed in mass, volumetric or molar %); t_r = reduced temperature.

The value of t_r can be calculated under the following formula:

$$t_r = \frac{t_b-t_{IBP}}{t_{EBP}-t_{IBP}}, \quad (5)$$

In formula (4), the variables of α and β represent the parameters defining the nature of oil components distribution by boiling point. The values of α and β parameters can be found in the calculation methodology given in [6] and implementing the non-linear least-square method.

The differential form proposed in literature [6] for the model of distribution oil compounds under consideration by boiling point in the range between t_{IBP} and t_{EBP} when normalized to 1 can be recorded as follows:

$$P(t_b) = a t_r^{\beta-1}\left\{\exp(-(t_r/\alpha)^\beta)\right\}, \quad (6)$$

a parameter can be calculated under the following formula:

$$a = \beta \frac{\alpha^{-\beta}}{t_{EBP} - t_{IBP}}, \quad (6.1)$$

The fraction % share boiling out from t_1 to t_2 will be:

$$\Delta G_{1,2} = 100 \int_{t1}^{t2} P(t_b) d\, t_b = G(t_2) - G(t_1), \quad (7)$$

in particular, the share of the oil heavy residue can be defined under the formula:

$$\Delta G_R = 100 - G(t_R), \quad (8)$$

where t_R = true point of atmospheric or vacuum distillation end, usually 350 or 520 °C in case of heating oil and tar as fuel oil residue, respectively (for fuel oil, the limit values of 400 and 430 °C are also used, with 500 °C for tar).

By using the distribution density $P(t_b)$ defined under formula (6), physical and chemical parameters of heaviness for modeled oils were also defined in the form of average values

$$\overline{\varepsilon_i} = \int_{t_{IBP}}^{t_{EBP}} \varepsilon_i(t_b) P(t_b) d\, t_b, \quad (9)$$

where $\varepsilon_i(t_b)$ = dependency of the i-th property of an oil fraction boiling at t_b.

As the properties of oils and oils fractions under consideration, molecular weight M and a dimension-less parameter S_g ("*specific gravity"*), used in international practice and designated in the same way as d_{15}^{15} – ratio of product density at 60 °F (15.6 °C) to the density of water at the same temperature, were considered. The oil molecular weight is usually related to the boiling point of its components. As density, it allows making a conclusion concerning oil composition. For low-density oils, high content of naphtha and kerosene fractions is typical, as well as low content of resins and asphaltene components. High-density oils show increased concentration of resins-and asphaltene components.

Converting d_{15}^{15} to conventional density values d_4^{20} was done under the following formula (according to [7]):

$$d_4^{20} = 1.0049 d_{15}^{15} - 0.0082, \quad (10)$$

By knowing average values of S_g, it is also possible to determine the parameter of feedstock heaviness using the American Petroleum Institute method [8]:

$$^0API = \frac{141.5}{S_g} - 131.5, \quad (11)$$

Changes in API degrees allow determining the relative oil density in relation to water density at the same temperature. If the density in API degrees exceeds 10, the oil is lighter and floats on the water surface, and if it is lower than 10, it sinks. This oil parameter is frequently used in classifications and to solve calculation tasks.

To assess the applicability of mathematical model, theoretical and experimental data for various oils and oil wastes were compared. The experimental results of distillation of light and heavy oil [9-11] were considered, as well as the organic part of reservoir oil sludges, which have been obtained by using the method described in [12]. The oil sludge organic part represents a dehydrated product containing hydrocarbon and mineral components.

RESULTS

Determination of oils and oil products fractional composition is based on distillation in specific conditions and in a specific reference system (temperature-distillation part). The distillation amount is measured in volume fractions, more rarely in weight or molar fractions. Distillation results are represented as tables or graphic dependencies used to determine the yield of a specific fraction.

The analysis of how the most general model (4) can be applied to building oil distillation curves requires prior specification of the distillation methodology effect upon the experiment results. In particular, many literature data concerning oil fractional composition is given in various measurement units of distillation cuts. To analyze the effect of this factor, the article considers exemplary data for oil distillation where the oil density is 870.3 kg/m^3 and the average molecular weight is 230, as given in Table 1. It can be seen that the TBP curve may significantly vary depending on the measurement units used to measure the share of the distilled fraction. The calculation results for parameters α and β defining the nature of oil components distribution by boiling points obtained when processing the data of Table 1 vary largely depending on the measurement units of distillation cuts share (Table 2).

Table 1. Fractional composition and characteristics of oil fraction [9]

Temperature, °C	Density. g/cm^3	Molecular weight	distillation % MU		
			wt.	vol.	molar
< 100	0.674	84	8.6	12.76	24.23
100-150	0.737	118	14.9	21.31	36.45
150-200	0.780	136	21.9	30.28	48.24
200-250	0.815	165	30.3	40.59	59.90
250-300	0.851	197	39.4	51.28	70.47
300-350	0.875	247	48.9	62.14	79.28
350-400	0.903	313	58.2	72.44	86.09
400-450	0.926	402	67.2	82.16	91.21
> 450	~0.980*	~700*	100	100	100

*according to extrapolation results

Table 2. Rated values of α and β parameters for oil

Parameter	When distillation % is expressed in		
	wt.	vol.	molar
α	0.6692	0.4515	0.3241
β	2.0013	1.6098	1.3370

The comparison of the theoretical TBP curve obtained for this oil under the model (4) by applying weight units for distillation fraction with experimental data shows high repeatability (Fig. 1). A wider comparison of calculation and experimental values made with respect to the share of distillation cuts expressed in weight, volumetric and molar measurement units is given in Fig. 2. It can be seen that the highest discrepancy between the calculation and experimental data occurs in case of volumetric units and the least in case of weight units.

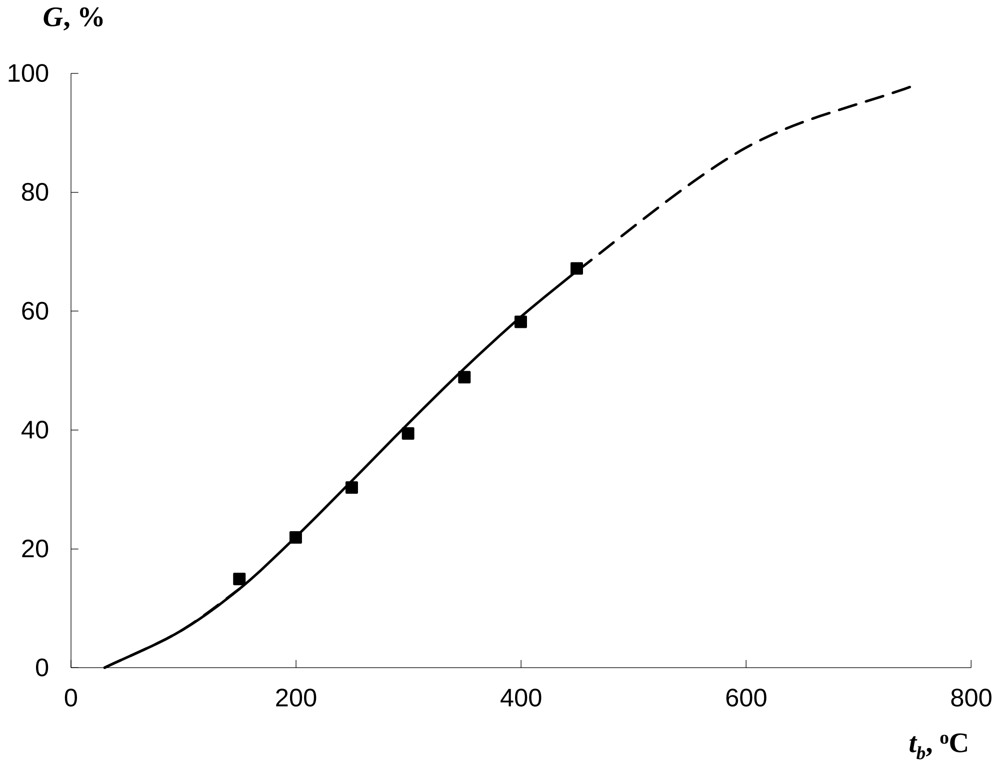


Figure 1. Model curve of oil distillation for the density of 870.3 kg/m^3 and average molecular weight of 230 obtained under the model (4). The points indicate experimental data according to [9].

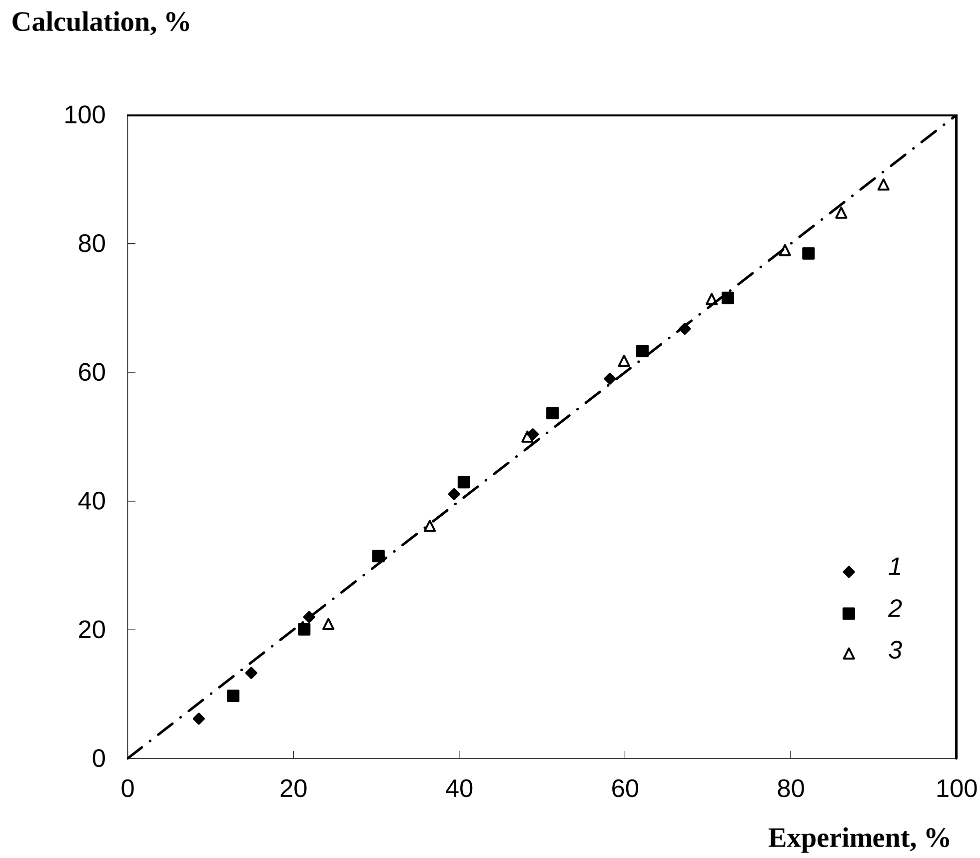


Figure 2. Comparison of experimental and calculation data for oil distillation in various measurement units of distillation cuts share (%): 1 – weight; 2 – volume; 3 – molar.

To model the fractional composition of oils having different degree of heaviness when varying α and β parameters in the model (4), it should have been checked that the value of parameters changes with oil density. This analysis was conducted upon the example of [11] according to the results of distillation of samples of more than 50 various oils, the heaviness of which can be described by density d_4^{20} and shares of distillation cuts boiling out below 200 °C, and a heavy residue boiling out above 360 °C. The results of analyzing their interrelations are given in Fig. 3 (a, b). The dependencies obtained illustrate that when the distillation share of a lighter fraction boiling out below 200 °C is increased, the density is decreased (Fig. 3 (a)), and in case of a heavier fraction boiling out above 360 °C, to the contrary, the density is increased.

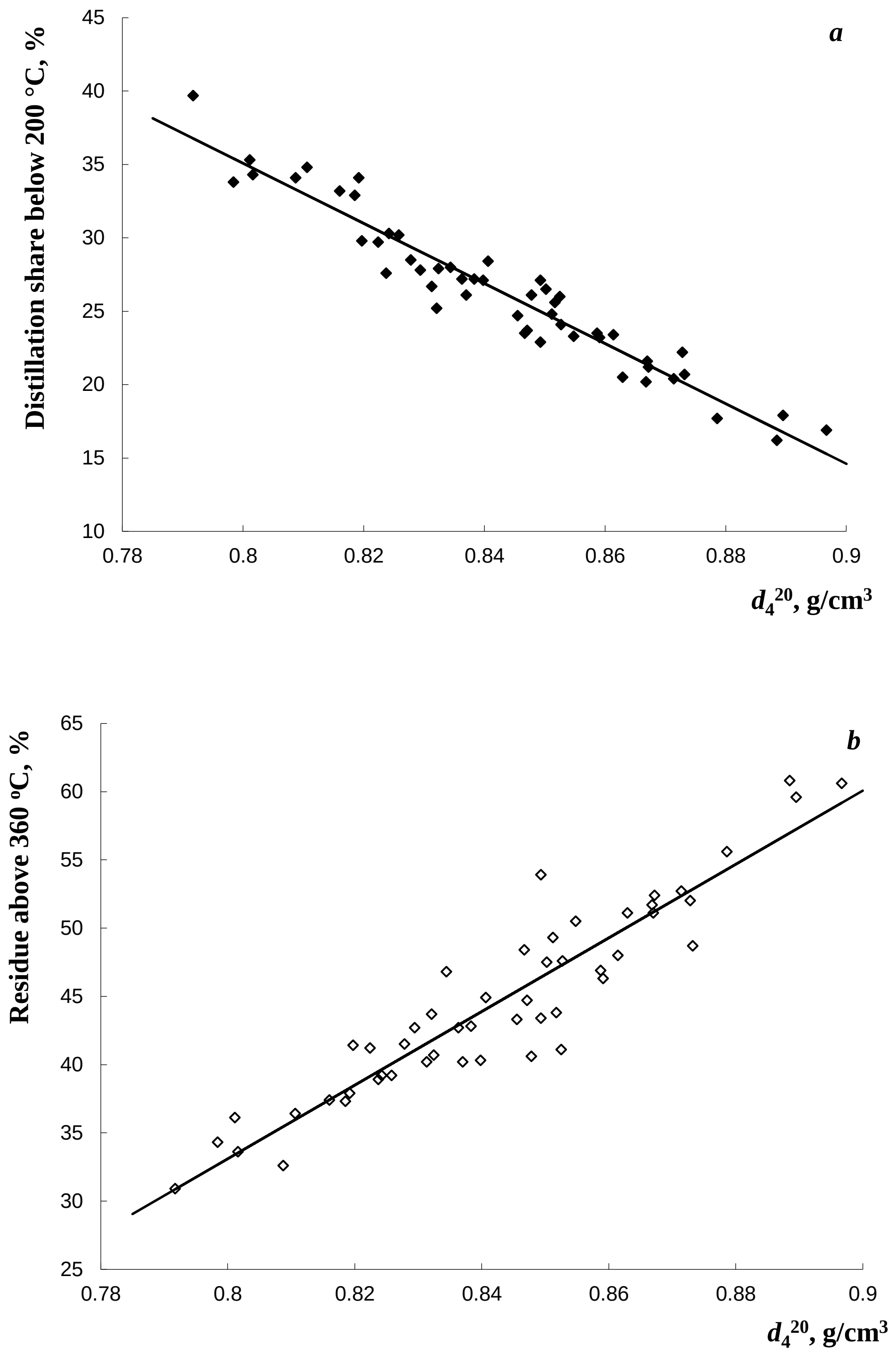


Figure 3. Dependency between the oil density d_4^{20} and fractional composition data (in %): a = shares of distillates boiling out below 200 °C; b = shares of heavy residue boiling out above 360 °C.

The graphical representation of oil distillations (TBP) corresponding to extreme points of dependencies given in Fig. 3 is represented in Fig. 4. By processing the data of curves under the model (4), the distribution parameters of components of oils under consideration by boiling points was determined. The calculation results are given in Table 3.

Table 3. Rated values of parameters α and β for oils corresponding to extreme points in Fig. 3.

Oil	α	β
Nevyanskaya (light)	0.4435	1.5229
Mikelonskaya (heavy)	0.5082	1.7502

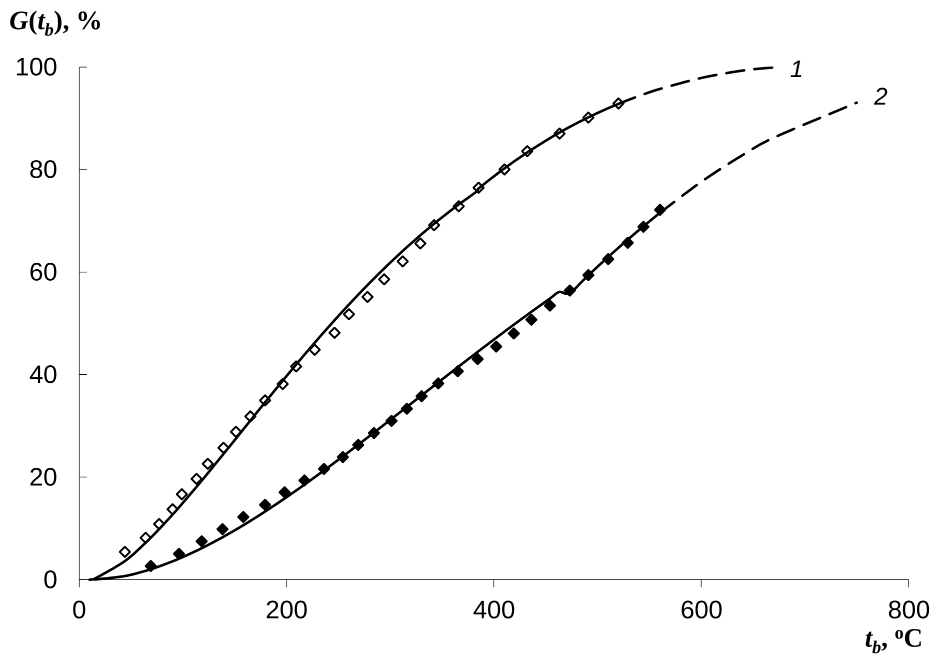


Figure 4. Curves of fractional distillation (TBP) for Nevyanskaya (1) and Mikelonskaya (2) oils. Points indicate experimental data according to [11]; solid lines indicate calculation data obtained under the model (4); dashed lines indicate calculation data obtained upon extrapolation results.

Then we built distillation curves for modeled oils of various heaviness in accordance with equations (4) and (6). We varied parameter α within 0.3 to 0.7. The second parameter was defined as $\beta = 4.51\alpha - 0.08$ (according to [12]). Distillation curves corresponding to some of the considered values of distribution parameters α and β are shown in Fig. 5.

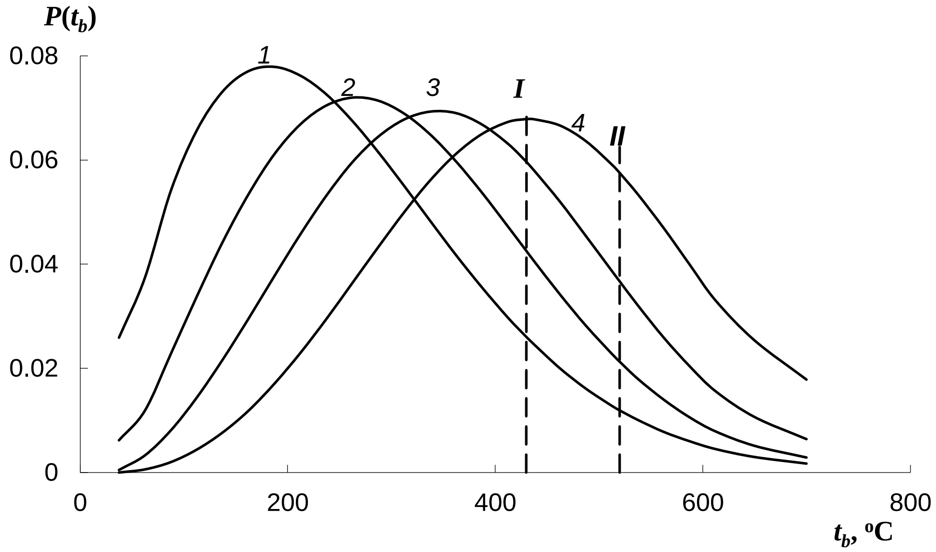


Figure 5. Differential curves of distribution by boiling points t_b of modeled oils at the values of α being 0.4 (curve 1), 0.5 (curve 2), 0.6 (curve 3) and 0.7 (curve 4). Dashed lines correspond to marginal boiling points of heave oil residues, °C: *I* – 430; *II* – 520.

The nature of yield change of individual oil fractions depending on the model parameter values as adopted in calculations is reflected by the data from Table 4. In the results, the following fractions were distinguished (with boiling point range, °C, given in brackets): gasoline fraction (< 105); naphtha (105-160); kerosene fraction (160-230); gas oil fraction (230-240); heavy residue fraction (> 430). According to the data of Table 4, we can say that when the rated model parameters change, the content of individual oil fraction also changes as obtained in the result of calculations.

Table 4. Characteristics of fractional composition of modeled oils of various heaviness

Parameters		d_4^{20},	Yield (%) of fractions with boiling point, °C				
α	β	g/cm³	< 105	105-160	160-230	230-430	> 430
0.40	1.724	0.8195	11.72	15.65	21.49	40.21	10.93
0.45	1.950	0.8395	7.30	12.42	20.01	46.08	14.19
0.50	2.175	0.8590	4.40	9.28	17.43	50.23	18.66
0.55	2.400	0.8774	2.58	6.60	14.33	52.07	24.42
0.60	2.626	0.8951	1.47	4.51	11.21	51.42	31.39
0.65	2.852	0.9108	0.83	2.98	8.41	48.51	39.27
0.70	3.077	0.9252	0.45	1.91	6.09	43.85	47.70

To calculate average values of physical and chemical parameters of modeled oil heaviness under the model (9), model dependencies were used to calculate $M(t_b)$ and $Sg(t_b)$, as described in literature [7]. Average molecular weights of modeled oil samples calculated under (9) were increased from 217 to 367 when the α parameter increased from 0.4 to 0.7, respectively.

When increasing the α parameter for modeled oil from 0.4 to 0.7, the average API index calculated under the model (11) decreased from 40.3 to 20.8°, respectively. The dependency of the distillation heavy residue yield and API index from α parameter is given in Fig. 6. The interrelation of average heaviness values and molecular weight of initial oil and the same parameters for heavy residues of distillation of the same oil feedstock is given in Fig. 6 and 7.

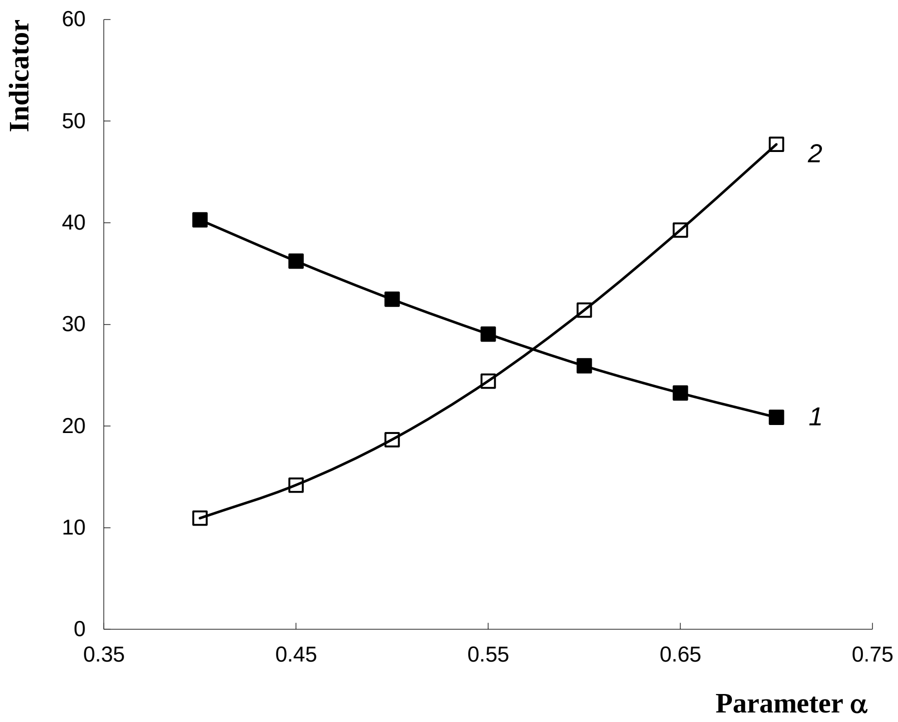
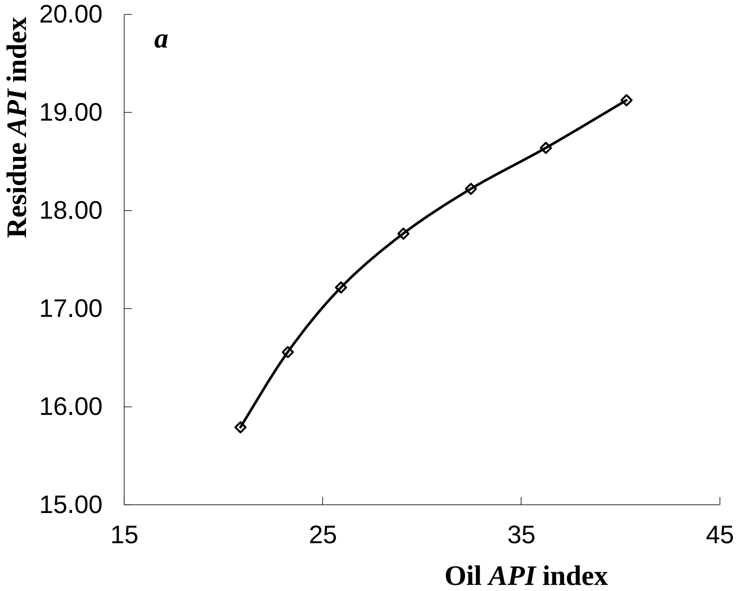


Figure 6. Effect of distribution parameter α of modeled oil: *1* – *API* value; *2* – residue yield (%) with boiling point > 350 °C.



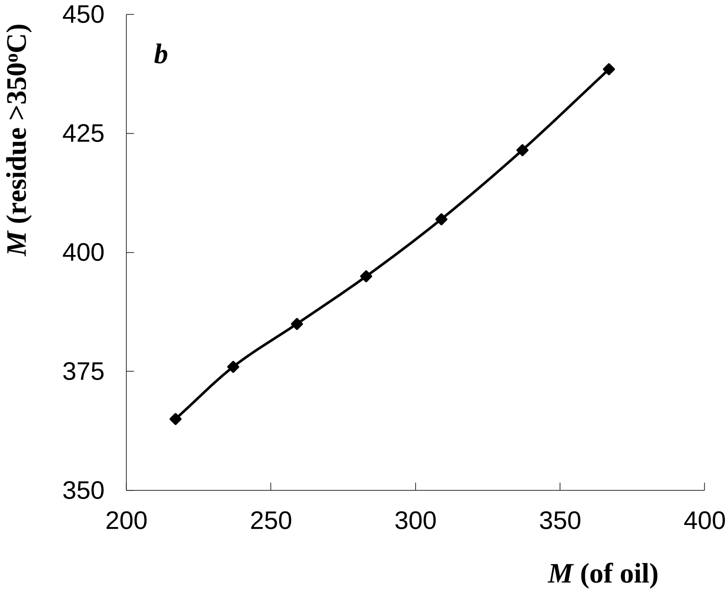


Figure 7. Interrelation of average values of oil and heavy residue (t_b > 350 °C): *a* – *API index*; b – molecular weight M.

DISCUSSION

With theoretical approach to describing the distribution of crude oil compounds in a formation by their boiling points, the data of distillation curves for tank-stock oil samples correlate to partial degassing of oil during extraction. In literature [14], it has been shown that the differential form of adjusted TBP curves is symmetrical to the bell shaped form satisfying the Gauss formula (so called normal distribution). The model (6), adopted for mathematical description of distillation curves, very well describes experimental data for oil distillation. Differential curves of boiling point distribution given in Fig. 5 have a bell shape. The model (4, 6) is quite acceptable for the purpose of determining the oil fractional composition. Additional proofs of this model reliability were obtained in [15] by analyzing more than 100 distillation curves of various oil products.

By density, oil is usually divided into light (0.800-0.839), average (0.840-0.879) and heavy (> 0.880) [16-17]. In 1987, at the XII World Petroleum Congress in Huston, the following classification for oils and natural bitumens was adopted, depending on density (kg/m^3): light oils (< 870.3), average oils (870.3-920), heavy oils (920-1000), extra-heavy oils (> 1000 kg/m^3 for viscosity $\mu < 10^4$ MPa·s); natural bitumens (the same at $\mu > 10^4$ MPa·s). According to reference data [18], the lightest oil in the Russia's European part was found at the island of Kolguyev, whereas the heaviest in the Volga region (Ulyanovsk oblast). For these oils, the yield of heavy residues with boiling point > 350 °C are 14 and 71%, respectively, and their density is 878 and 1006 kg/m^3.

An additional source of oil stock of various heaviness is oil wastes, in particular, reservoir oil sludges, the organic part of which contains more than 46 wt% of the fraction boiling out above 520 °C, according to [12]. Depending on the origin, oil sludges may differ in fractional composition, but in general, as described in [19], the products correspond to the requirements to oil feedstock for oil refineries. The papers [19-20] recommended using oil sludges for compounding when producing furnace fuel oil.

Siberian oils vary in heaviness from very light Markovskaya to rather heavy Yagerskaya [10]. The Markovskaya oil boils out below 300 °C almost completely, and heavy resinous Yagerskaya oil only starts evaporating at this temperature.

The article shows the applicability of the model (4) to the oils of various heaviness, the refining of which has become extremely relevant recently, since the reserves of oils stock of various heaviness are rather significant.

According to the results of studying a similar issue for mineral coals [13], it could have been expected that an increase in average oil density should result in both increased α and β. To model the fractional composition of oils of various heaviness under the model (4) in the conditions of parameters α and β, the effect of oil density was preliminary studied. More than 50 various oils [11] were taken for study. The analysis of the obtained data revealed that density and yield value of various fractions correlate to each other, and when the density is increased, the yield of light fractions is decreased, and the yield of heavy residues is increased (Fig. 3). The extreme points in correlations of Fig. 3a and Fig. 3b correspond to Nevyanskaya (d_4^{20} 0.7917 g/cm^3;

distillation of 39.7%; residue of 30.9%) and Mikelonskaya oil (d_4^{20} 0.8967 g/cm^3; distillation of 16.9%; residue of 60.6%). Thus, Nevyanskaya and Mikelonskaya oils can be characterized as the lightest and heaviest oils, respectively. The organic part of the reservoir oil sludge (d_4^{20} 0.9589 g/cm^3; distillation of 3%; residue of 73%) [*12*] can be characterized as heavy oil.

Indeed, as Tables 3-4 show, the distribution in case of heavy oil is characterized by higher values of parameters α and β in the model (4) as compared to light oil. This aspect was considered in modeling the fractional composition and properties of several oils with consistently changing heaviness. It can be seen that when the value of α (and respectively β) is increased, distillation differential curves are shifted to increased temperatures with increased share of heavy residues (areas under curve beyond the marginal temperatures) (Fig. 5).

Calculations of average molecular weights and API indexes of modeled oils under the model (9, 11) have shown that average molecular weights lie within 217-367, with α parameter respectively increased from 0.4 to 0.7. In this case, the average API index decrease from 40.3 to 20.8°, respectively. The dependency of the distillation heavy residue yield and API index from α parameter (Fig. 6) shows that when α is increased, the yield of the residue is also increased and the API index is decreased. The interrelation of average heaviness values and molecular weight of initial oil and the same parameters for heavy residues of distillation of the same oil feedstock is given in Fig. 7. It can be seen that high API indexes and molecular weights of feedstock correspond to high value of respective parameters of the residue boiling out above 350 °C. It should be noted that *API* = 40.3° obtained for α = 0.4 is typical of Altamont paraffin oil, and *API* = 20.8° (for α = 0.7) is close to the index value for heavy Maya oil with intermediate group composition [21]. Arabian Heavy oil (*API* = 27.7° [21]) corresponds to the modeled oil at α = 0.57. Thus, the higher is the heaviness of crude oil, the closer these parameters of heavy distillation residues approach the critical level corresponding to heavy oil feedstock (Fig. 7).

CONCLUSIONS

By modeling the factional composition of oil feedstock using empirical dependencies (4) and (6), it is possible to accurately determine the actual parameters of oils of various heaviness and oil wastes, in particular, the organic part of reservoir oil sludges. The models applied reproduce the increased yield of oil distillation heavy residue at the qualitative level when its heaviness is increased (Fig. 6). This refers both to the API index (Fig. 7) and molecular weight M (Fig. 7). To elaborate the mathematical description of fractional composition and oil properties in more details, it is reasonable to supplement the adopted model with data concerning the change in the elemental and group hydrocarbon composition of the fraction with temperature, and with a more complete set of physical and chemical properties, including viscosity, surface stress, molecular refraction and cocking behavior. It is also interesting to reveal differences in the composition and properties in relation to narrow fractions with similar average boiling points in oils of various nature (various heaviness).

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