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AN APPLICATION OF STABILITY ANALYSIS OF HYDROCARBON MIXTURES FROM X AND Y FIELDS TO PREDICT WAX PRECIPITATION

BAZARBAYEVA M.B.[1], ISMAILOVA D.A.[1], ABDUKARIMOV A.К[1], DELIKESHEVA D.N[1], MOMBEKOV B.I.[1], ZERPA L.[2]

[1]Satbayev University, 050000, Almaty, Kazakhstan
[2]Colorado School of Mines, 80401, Colorado, USA

Abstract. *The thermodynamic description of wax deposition is a relatively new approach to solving the problem of wax precipitation, which the oil and gas industry has been struggling with for a long time. Many models exist in the literature to predict the thermodynamic conditions under which the first paraffin crystal is formed. The first task in all models is to determine the stability of the hydrocarbon mixture in order to define the possibility of wax precipitation. The stability of a mixture is determined by the thermodynamic behavior of the phases of a multicomponent mixture, namely, the presence of all existing phases in equilibrium. To this end, a new stability algorithm with Gibbs energy minimization to determine the wax precipitation in hydrocarbon mixtures has been developed. The algorithm is based on multi-solid thermodynamic model with EOS concepts. The main criterion for stability is the existing of the mixture at its global minimum. Proposed stability analysis predicts whether a given mixture will be split into multiple phases or will exist as the single phase at a given temperature and pressure conditions. The model was proven with 6 samples from fields X and Y showing the instability of original petroleum mixtures that agree with the real behavior of the oil in in-situ conditions. The results of the new stability algorithm are comparable with the results of the models presented previously. The main advantage of the method is its simplicity and reliability.*

Key words: *Stability analysis, Gibbs energy, global minimum, fugacity, heptane plus characterization, wax deposition, chemical potential, splitting, phase equilibrium, multi-solid model.*

ПАРАФИН ТҮСУІН БОЛЖАУ ҮШІН X ЖӘНЕ Y КЕНДЕРІНІҢ КӨМІРСУТЕГІ ҚОСПАСЫНЫҢ ТҰРАҚТЫЛЫҚ АНАЛИЗІН ҚОЛДАНУ

БАЗАРБАЕВА М.Б.[1], ИСМАИЛОВА Д.А.[1], АБДУКАРИМОВ А.К.[1], ДЕЛИКЕШЕВА Д.Н.[1], МОМБЕКОВ Б.И.[1], ЗЕРПА Л.[2]

[1]Қ.И. Сәтбаев атындағы Қазақ Ұлттық техникалық зерттеу университеті,
050000, Алматы қ., Қазақстан
[2]Колорадо тау-кен мектебі, 80401, Колорадо, АҚШ

Аңдатпа. *Парафин шөгуінің термодинамикалық сипаттамасы мұнай-газ өнеркәсібі біраз уақыттан бері күресіп келе жатқан парафин шөгуінің мәселесін шешудің салыстырмалы* *түрде жаңа тәсілі болып табылады. Әдебиетте бірінші парафин кристалы түзілетін термодинамикалық жағдайларды болжауға арналған көптеген модельдер бар. Барлық модельдердегі бірінші міндет парафин тұнудың мүмкіндігін тексеру үшін көмірсутекті қоспаның тұрақтылығын анықтау болып табылады. Қоспаның тұрақтылығы көпкомпонентті қоспаның фазаларының термодинамикалық күйімен, атап айтқанда, барлық фазалардың тепе-теңдікте болуымен анықталады. Осы мақсатта көмірсутек қоспаларындағы парафинның шөгуін анықтау үшін Гиббс энергиясын минимизациялау арқылы жаңа тұрақтылық алгоритмі әзірленді. Алгоритм күй теңдеулері концепциялары бар multi-solid термодинамикалық модельге негізделген. Тұрақтылықтың негізгі критерийі қоспаның оның жаһандық минимумында болуы. Ұсынылған тұрақтылық алгоритмі берілген қоспаның белгілі температура мен қысым жағдайында бірнеше фазаға бөлінетінін немесе бір фаза ретінде болатынын болжайды. Модель X және Y кен орындарынан алынған 6 үлгіден сыналды, бұл кендердегі мұнайдың нақты әрекетіне сәйкес келетін бастапқы мұнай қоспаларының тұрақсыздығын көрсетті. Жаңа тұрақтылық алгоритмінің нәтижелерін бұрын ұсынылған үлгілермен салыстыруға болады. Әдістің басты артықшылығы оның қарапайымдылығы мен сенімділігі болып табылады.*

Түйінді сөздер: *тұрақтылық анализі, Гиббс энергиясы, жаһандық минимум, ұшпа, гептан-плюс фракциясының сипаттамасы, парафин жиналуы, химиялық потенциал, бөліну, фазалық тепе-теңдік, multi-solid моделі.*

ПРИМЕНЕНИЕ АНАЛИЗА СТАБИЛЬНОСТИ СМЕСИ УГЛЕВОДОРОДОВ МЕСТОРОЖДЕНИЯ X И Y ДЛЯ ПРОГНОЗИРОВАНИЯ ВЫПАДЕНИЯ ПАРАФИНА

БАЗАРБАЕВА М.Б.[1], ИСМАИЛОВА Д.А.[1], АБДУКАРИМОВ А.К.[1],
ДЕЛИКЕШЕВА Д.Н.[1], МОМБЕКОВ Б.И.[1], ЗЕРПА Л.[2]

[1]Казахский национальный исследовательский технический университет
имени К.И. Сатпаева, 050000, г. Алматы, Казахстан
[2]Колорадская горная школа, 80401, Колорадо, США

Аннотация. *Термодинамическое описание осаждения парафина – относительно новый подход в решении проблемы выпадения парафина, с которой нефтегазовая индустрия борется довольно продолжительное время. В литературе существует множество моделей с целью прогнозирования термодинамических условий, при которых образуется первый кристалл парафина. Первой задачей во всех моделях является определение стабильности смеси углеводородов для того, чтобы определить перспективу выпадения парафина. Стабильность смеси определяется термодинамическим поведением фаз многокомпонентной смеси, а именно нахождением всех существующих фаз в равновесии. С этой целью разработан новый алгоритм стабильности с минимизацией энергии Гиббса для определения осаждения парафина в углеводородных смесях. Алгоритм основан на термодинамической модели multi-solid с концепциями УС. Главный критерий устойчивости – нахождение смеси в ее глобальном минимуме. Предлагаемый анализ стабильности предсказывает, будет ли данная смесь разделена на несколько фаз или будет существовать как одна фаза при заданных условиях температуры и давления. Модель была проверена на 6 образцах из месторождений X и Y, показывающих нестабильность исходных нефтяных смесей, что согласуется с реальным поведением нефти в условиях месторождения. Результаты нового* *алгоритма устойчивости сопоставимы с результатами моделей, представленных ранее. Главное достоинство метода – простота и надежность.*

Ключевые слова: *анализ стабильности, энергия Гиббса, глобальный минимум, летучесть, описание фракции гептан-плюс, отложение парафина, химический потенциал, расщепление, фазовое равновесие, модель multi-solid.*

Introduction

Crude oil is a complex mixture of hydrocarbons, consisting of alkanes, naphthenics, asphaltenes, resins and aromatics. Paraffins (normal alkanes) are the first precipitating components and their amount in wax are greater than other components. Hence, wax deposition is often associated with paraffin deposition. Wax precipitation substantially affects oil production and transportation. Wax deposition increases pressure drop and therefore power requirements, and decreases effective flow area of the pipeline reducing oil production. To prevent these problems, it is required to predict the thermodynamic conditions under which the waxes will precipitate in functions of pressure, temperature and composition. The thermodynamic- predictive models are useful to complete this task.

Thermodynamic behavior of petroleum mixtures can lead to a number of multiphase equilibrium states, including vapor–liquid, liquid–multi-solid (wax) and vapor–liquid–multi-solid (wax) equilibria. Multiphase equilibrium calculations are solved by two widely used stability analysis algorithms. The first is the solution of a system of equations that describe the phase equilibrium [1], [2] and the second approach is consist of Gibbs free energy minimization. [3], [4]. Stability analysis predicts whether a given mixture will be split into multiple phases, the number of phases at equilibrium and the distribution of components within these phases [5]. Both approaches are based on multi-solid thermodynamic model. Multi-solid model assumes that precipitated wax predominantly consists of mutually immiscible pure hydrocarbon components. The number of precipitating species can be determined at fixed temperature and pressure using a stability analysis.

[2] showed that at constant temperature, T, and pressure, P, a component i may precipitate as a pure solid if it fulfills this expression:

$$\mathrm{f_i^L(P,T,Z_i) - f_{i,pure}^S(P,T) \geq 0}$$

where $\mathrm{f_i(P,T,Z_i)}$ is the fugacity of component i in an original petroleum mixture with composition Z and $\mathrm{f_{i,pure}^S(P,T)}$ is the fugacity of component i as a pure solid. This equation is easily derived from Eq. (5) of [4]. The vapor–liquid phase behavior is described using Peng–Robinson EOS [6]. The criterion for thermodynamic equilibrium is that the fugacities of every component in each phase must be equal implying zero net mass transfer between the phases.

The second assumption is numerical and geometrical methods of stability analysis based on Gibbs' tangent plane and global minimum criterions. The tangent plane criterion theory suggests that if the tangent hyperplane to the Molar Gibbs energy surface at original composition z to the energy surface at composition y, F(y) is the at no point lies above the energy surface, the system is stable. If any of the parallel tangent planes lie below the energy surface the mixture is unstable and will split into at least two phases. Numerically, the necessary and sufficient condition for global stability is: [3]

$$F(y) = g(y) - L(y;z) = \sum_{i}^{N} y_i(\mu_i(y) - \mu_i(z)) \geq 0$$

At this work based on the multi-solid phase model for petroleum mixtures, a new and effective stability analysis using Gibbs energy minimization is proposed.

Main part

This section considers the fundamental theoretical concepts regulating the stability of hydrocarbon mixture that are the foundation of this paper. In addition, the heptane plus characterization methodology is presented, and equations for calculating the critical properties of alkanes heavier than C_{7+} and their chemical potentials are developed. These values are required to obtain the final inequality equation.

Problem statement

The Gibbs energy of an M-component mixture with mole fractions (z_1, z_2, …, z_M) at given temperature and pressure (T_o, P_o) is

$$G_o = \sum_i^M n_i \mu_i^0$$

where μ_i^0 is the chemical potential of i-component of the mixture.

We have an assumption that this mixture is divided into two phases with mole numbers $N - \epsilon$ and ϵ, respectively. The number of moles ϵ of the second phase is infinitesimal and the mole fractions in phase II are (y_1, y_2, …, y_M).

The change in Gibbs energy after splitting is:

$$\Delta G = G_I + G_{II} - G_o$$

or using a Taylor series expansion of G_I, discarding second order terms in ϵ, gives

$$\Delta G = G(\epsilon) - \epsilon \sum_i^N y_i \, \mu_i^0 = \epsilon \sum_i^N y_i(\mu_i(y) - \mu_i^0)$$

Original mixture is stable when its Gibbs energy is at the global minimum. Consequently, a necessary criterion for stability is:

$$F(y) = \sum_i^N y_i(\mu_i(y) - \mu_i^0) \geq 0$$

for all trial compositions y.

For binary mixtures stability criterion is that the tangent hyperplane at no point lies above the energy surface. Multiphase system is in an equilibrium when the individual species have identical chemical potentials in all phases and global minimum criterion is also satisfied [4].

Materials

Three oil sample from field X and as many from field Y were obtained to produce the laboratorial analysis. Composition of the crudes was investigated using gas chromatography. Table 1 presents the composition of six oils. Study of the group composition of oil by SARA analysis was performed. More than half of the mixtures consist of saturated hydrocarbons, the portion of aromatic hydrocarbons is about one fifth, the rest belongs to resins and asphaltenes. The molecular weight of the mixture varies from 270-275 g / mol.

Table 1 – Composition of six oil samples obtained from gas chromatography

Component	Mole fraction					
	1	2	3	4	5	6
i-butane	0,00047	0,00047	0,00054	0,00072	0,00065	0,00068
i-pentane	0,00102	0,00105	0,00113	0,00251	0,00239	0,00254
n-pentane	0,00115	0,0012	0,00128	0,00419	0,00403	0,00428
hexane	0,00337	0,00343	0,00379	0,00984	0,00981	0,01029
heptane	0,00602	0,00614	0,0064	0,0156	0,01584	0,0169
octane	0,0119	0,01235	0,01274	0,02346	0,02399	0,02581
nonane	0,01556	0,01592	0,01631	0,02502	0,02552	0,02748
decane	0,02192	0,02234	0,02226	0,02666	0,02712	0,02961
C_{11}	0,02438	0,02473	0,0243	0,02605	0,02642	0,029
C_{12}	0,02679	0,02706	0,02633	0,02649	0,02679	0,02936
C_{13}	0,03264	0,0329	0,03166	0,03072	0,03105	0,03395
C_{14}	0,03227	0,0325	0,03101	0,02914	0,02946	0,032
C_{15}	0,03837	0,03862	0,03681	0,03439	0,03476	0,03731
C_{16}	0,03275	0,03301	0,03121	0,02878	0,0291	0,03081
C_{17}	0,03238	0,03262	0,03055	0,02791	0,02825	0,02956
C_{18}	0,03302	0,03329	0,03107	0,02833	0,02864	0,02964
C_{19}	0,03185	0,03198	0,02979	0,02707	0,02734	0,0278
C_{20+}	0,64539	0,63826	0,64047	0,61379	0,60087	0,57849
i-hexane	0,00049	0,00051	0,00054	0,00137	0,00136	0,00144

methylcyclopentane	0,00067	0,00069	0,00072	0,00211	0,0021	0,00222
benzene	0,00064	0,0008	0,00084	0,00206	0,00207	0,0022
cyclohexane	0	0	0,00003	0,00006	0,00006	0,00006
methylcyclohexane	0,0032	0,00332	0,00347	0,00632	0,0064	0,00683
Toluol	0,00012	0,00315	0,0132	0,00195	0,01043	0,00566
Ethylbenzene	0,00083	0,00086	0,00087	0,00146	0,0015	0,00162
m-xylene	0,0012	0,00123	0,00123	0,00156	0,00159	0,00178
o-xylene	0,00036	0,00037	0,00018	0,00054	0,00055	0,0006
Trimethylbenzene	0,00121	0,00123	0,00125	0,00186	0,00188	0,00204
Total	1,0	1,0	1,0	1,0	1,0	1,0

Methods

Phase behavior calculations using cubic equation of state to predict the reservoir fluid properties require components characteristics such as critical pressure and temperature, acentric factor and binary interaction parameters (BIP). Separation techniques including gas chromatography and distillation are not able to identify all components in mixture, particularly, heavier than C_7. These components are grouped in heptane-plus fraction and next calculations are performed to simulate C_{7+} characterization. [10]

Molecular weight and density of C_{7+}. Density is calculated at standard conditions (P = 101325 Pa, T = 293 K)

Mole fraction of heptane-plus:

$$z_{C7+} = \sum_{i=7}^{n} z_i$$

Molecular weight of C_{7+} fraction:

$$M_{wC7+} = \frac{\sum_{i=7}^{n} z_i * M_i}{z_{C7+}}$$

Density:

$$\rho_{C7+} = \frac{M_{wC7+}}{V_{C7+}}$$

Total volume calculation: since the pressure is low, the mixture might be considered as ideal solution and the total volume can be calculated summarizing component's partial volume.

Sum of partial volumes based on the known densities at standard conditions.

$$V_{C7+} = \sum_{i=7}^{n} z_i * V_i$$

Specific gravity:

$$SG = \rho_{C7+}/\rho_w$$

Split the C_{7+} fraction:

Universal Oil Products (UOP) characterization factor [7]:

$$K_{wC7+} = 4.5579 M^{0.12178} \gamma^{-0.84573}$$

Molar weight [8]:

$$M_i = 14n - 4$$

$$\gamma_i = \left(\frac{K_{wC7+}}{4.5579 M_i^{0.15178}} \right)^{\frac{1}{-0.84573}}$$

Mole fraction [8]:

$$z_{cn} = 1.38205 z_{C7+} \exp(-0.25903n)$$

Boiling point in R:

$$T_{bi} = (K_{wC7+} \gamma_i)^3$$

Knowing boiling temperature and specific gravity of all components, other characteristics can be calculated using Lee-Kesler correlation.

Lee-Kesler correlations, $T_c\ in\ °R$, P_c in psi [7]:

Critical temperature:

$$T_{ci} = 341.1 + 811\gamma_i + (0.4244 + 0.1174\gamma_i) T_{bi} + (0.4669 * -0.2623\gamma_i) * 10^{-5} T_{bi}^{-1}$$

Critical pressure:

$$lnP_{ci} = 8.3634 - \frac{0.0566}{\gamma_i} - \left[\left(0.24244 + \frac{2.2898}{\gamma_i} + \frac{0.11857}{\gamma_i^2} \right) * 10^{-3} \right] T_{bi} + \left[\left(1.4685 + \frac{3.648}{\gamma_i} + \frac{0.47227}{\gamma_i^2} \right) * 10^{-7} \right] T_{bi}^2 - \left[\left(0.42019 + \frac{1.6977}{\gamma_i^2} \right) * 10^{-10} \right] T_{bi}^3$$

Acentric factor:

$$\omega_i = \frac{-\ln\left(\frac{P_c}{14.7}\right) + A_1 + A_2 T_{bri}^{-1} + A_3 lnT_{bri} + A_4 T_{bri}^6}{A_5 + A_6 T_{bri}^{-1} + A_7 lnT_{bri} + A_8 T_{bri}^6}$$

where A1= −5.92714; A2=6.09648; A3=1.28862; A4=−0.169347; A5=15.2518; A6=−15.6875; A7=−13.4721; A8=0.43577

Binary interaction parameters:

$$k_{ij}^{EOS} = 1 - \left[\frac{2V_{c_i}^{1/6}V_{c_j}^{1/6}}{V_{c_i}^{1/3} + V_{c_j}^{1/3}}\right]$$

Gibbs energy minimization

Global minimum:

$$F(y) = \sum_i z_{Cn}(\mu_i(y) - \mu_i^0) \geq 0$$

Chemical potential of pure component:

$$\mu_0 = \frac{G_0}{n_i}$$

Gibbs energy of pure component G_0 is obtained from [9].

Chemical potential of component i in the mixture:

Results and discussions

Heptane-plus fraction is characterized in Table 2. The portion of intermediate and heavy hydrocarbons is extremely high, because the petroleum mixtures produced from X and Y fields are characterized by a great amount of paraffins and resins present. The pour point temperature is also relatively high.

Table 2 – Properties of heptane-plus fraction

Sample	zC_{7+}	MwC_{7+}, g/mol	VC_{7+}, m3/mol	ρC_{7+}, g/m3	SG
1	0,98524	341,476	0,000429004	795974,038	0,75597404
2	0,98172	339,901	0,000427143	795755,809	0,7957558
3	0,97091	344,09	0,000432035	796440,235	0,79644024
4	0,96341	336,747	0,000423413	795316,126	0,795316
5	0,95515	333,924	0,000420134	794802,822	0,7948028
6	0,95772	327,314	0,000412325	793826,029	0,793826

In Fig. 1 splitted molar composition of the first sample from field X is presented. Hydrocarbons with carbon number higher than 20 are grouped and their 4% of heptane-plus fraction. A great part of the fraction consists from liquid hydrocarbons and the portion of heavy paraffins are significant.

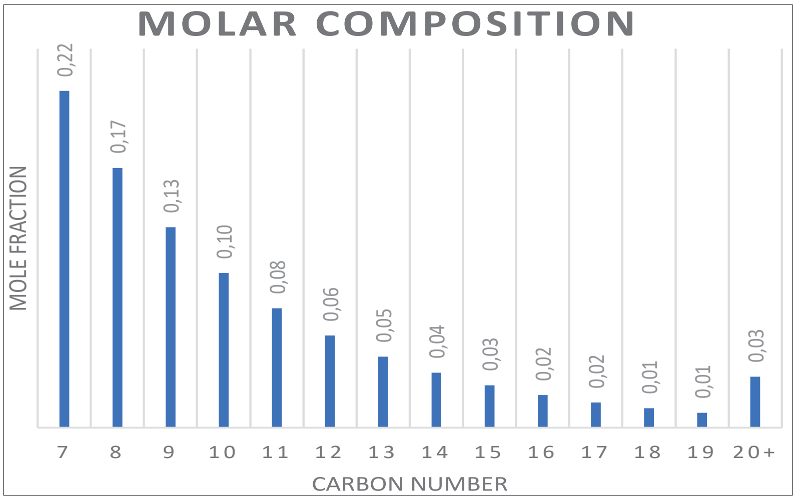

Figure 1– Molar composition of the oil sample #1 from field X after splitting

Heptane-plus characterization results with critical properties are tabulated in the Table 3.

Table 3 – Properties of heptane-plus fraction components from field X, sample #1

Carbon number	Mole fraction	MW, g/mol	Pc, MPa	Tc, K	Acentric factor
7	0,222125641	94	2,660247011	642,1377145	-0,388615554
8	0,17143654	108	2,395102473	663,0170572	-0,388615554
9	0,132314699	122	2,174872927	682,429072	-0,388615554
10	0,102120468	136	1,988558059	700,6390911	-0,388615554
11	0,078816563	150	1,828582633	717,8421369	-0,388615554
12	0,060830613	164	1,689535127	734,1863416	-0,388615554
13	0,046949059	178	1,567432645	749,7872435	-0,388615554
14	0,036235278	192	1,459270229	764,7369485	-0,388615554
15	0,027966383	206	1,362732803	779,1102394	-0,388615554
16	0,02158445	220	1,276004529	792,9687869	-0,388615554

17	0,016658876	234	1,197638844	806,3641357	-0,388615554
18	0,012857318	248	1,126467572	819,3398726	-0,388615554
19	0,009923277	262	1,061535934	831,9332351	-0,388615554
20+	0,033561697				

This work is based on multi-solid thermodynamic model and performing splitting, we assume that our mixture is divided into pure hydrocarbon components. After splitting we know the exact number of components present in heptane-plus fraction. In our samples the highest carbon number is 149.

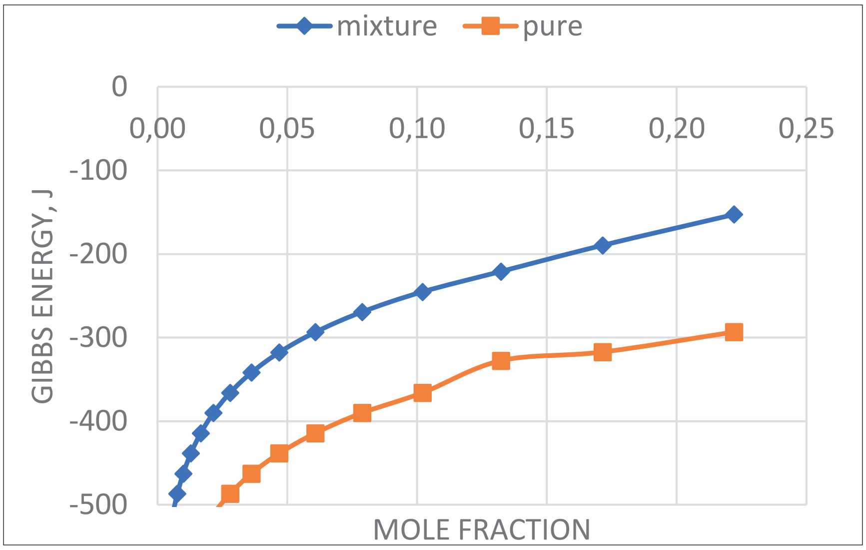

Figure 2 – Gibbs energy as a function of molar composition

In fig 2 the Gibbs energy of pure hydrocarbon components and their Gibbs energy as the part of the mixture are compared. Pure components have Gibbs energy less than when they are in the mixture. Consequently, their chemical potential as in the negligible pure phase are smaller than in the original petroleum mixture. In the global minimum criterion, our potential differences are negative and the instability of the mixture is proved.

Conclusion

Problems related to the wax precipitation during production and transportation of crude oil cause major difficulties in these processes. To prevent the wax segregation from the original mixture it is recommended to predict the thermodynamic conditions at which the components are not at equilibrium. There are thermodynamic models based on solid-solution and multi-solid theories to predict the wax appearance at certain pressure temperature conditions. The main part of these models is stability analysis, which predicts whether a given mixture at equilibrium or not. At this work a new stability algorithm based on Gibbs tangent criterion is proposed.

The stability of the mixture requires that its Gibbs energy be at a global minimum. If the mixture of hydrocarbons is stable under certain thermodynamic conditions of the formation it will exists as a single phase, but if the stability criterion is not satisfied, the mixture will split into two or more different phases. In our case the petroleum mixtures are not at their global minimum. It means that the mixtures are unstable and segregated into at least liquid and solid phases. The solid phase is our wax that consists of pure hydrocarbons, since our work is based on the multi-solid model. A new algorithm is simple and does not require a lot input data.

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Information about authors

Bazarbayeva Makpal Bulatovna (corresponding author)
Bachelor student, Satbayev university, Satpayev st. 22, 050000, Almaty, Kazakhstan
ORCID ID: 0000-0002-7217-6238
E-mail: makpal.bazarbayeva.pe@gmail.com

Ismailova Jamilyam Abdulakhatovna
PhD, Satbayev University, Satpayev st. 22, 050000, Almaty, Kazakhstan
ORCID ID: 0000-0002-7680-7084
E-mail: j.Ismailova@satbayev.university

Abdukarimov Aibek Kenzhebekovich
MSc in Petroleum Engineering, PhD student, Satbayev University, Satpayev st. 22, 050000, Almaty, Kazakhstan
ORCID ID: 0000-0001-6570-2810
E-mail: abdukarimov1303@gmail.com

Delikesheva Dinara Nasipulovna
MSc, Satbayev University, Satpayev st. 22, 050000, Almaty, Kazakhstan
ORCID ID: 0000-0001-5442-4763
E-mail: d.delikesheva@satbayev.university

Mombekov Bagdat Isakhanovich
MSc, Satbayev University, Satpaev st. 22, 050000, Almaty, Kazakhstan

ORCID ID: 0000-0002-7112-8920
E-mail: b.mombekov@satbayev.university

Dr. Luis Zerpa
Associate Professor of Petroleum Engineering, Colorado School of Mines, 1500 Illinois St., Golden, CO, 80401, USA
ORCID ID: 0000-0003-3211-2704
E-mail: lzerpa@mines.edu

Авторлар туралы ақпарат

Базарбаева Мақпал Булатовна (корреспонденция авторы)
Бакалавриат студенті, Қ.И.Сәтбаев атындағы ҚазҰТЗУ, Сәтпаев көш., 22, Алматы қ., Қазақстан
ORCID ID: 0000-0002-7217-6238
E-mail: makpal.bazarbayeva.pe@gmail.com

Исмаилова Джамилям Абдулахатовна
Техника ғылымдарының докторы, Қ.И.Сәтбаев атындағы ҚазҰТЗУ, Сәтпаев көш., 22, Алматы қ., Қазақстан
ORCID ID: 0000-0002-7680-7084
E-mail: j.Ismailova@satbayev.university

Абдукаримов Айбек Кенжебекович
Техника ғылымдарының магистрі, PhD докторанты, Қ.И.Сәтбаев атындағы ҚазҰТЗУ, Сәтпаев көш., 22, Алматы қ., Қазақстан
ORCID ID: 0000-0001-6570-2810
E-mail: abdukarimov1303@gmail.com

Деликешева Динара Насипуловна
Техника ғылымдарының магистрі, Қ.И.Сәтбаев атындағы ҚазҰТЗУ, Сәтпаев көш., 22, Алматы қ., Қазақстан
ORCID ID: 0000-0001-5442-4763
E-mail: d.delikesheva@satbayev.university

Момбеков Бағдат Исаханұлы
Техника ғылымдарының магистрі, Қ.И.Сәтбаев атындағы ҚазҰТЗУ, Сәтпаев көш., 22, Алматы қ., Қазақстан
ORCID ID: 0000-0002-7112-8920
E-mail: b.mombekov@satbayev.university

Доктор Луис Зерпа
Мұнай инженериясы кафедрасының ассоциацияланған профессоры, Колорадо тау-кен мектебі, Иллинойс көшесі, 1500, Голден, CO 80401, АҚШ
ORCID ID: 0000-0003-3211-2704
E-mail: lzerpa@mines.edu

Информация об авторах

Базарбаева Макпал Булатовна (автор для корреспонденции)
Студент бакалавриата, КазНИТУ им. К.И. Сатпаева, ул. Сатпаева, 22, 050000, г. Алматы, Казахстан
ORCID ID: 0000-0002-7217-6238
E-mail makpal.bazarbayeva.pe@gmail.com

Исмаилова Джамилям Абдулахатовна
Доктор технических наук, КазНИТУ им. К.И. Сатпаева, ул. Сатпаева, 22, 050000, г. Алматы, Казахстан
ORCID ID: 0000-0002-7680-7084
E-mail: j.Ismailova@satbayev.university

Абдукаримов Айбек Кенжебекович
Магистр технических наук, PhD докторант, КазНИТУ им. К.И. Сатпаева, ул. Сатпаева, 22, 050000, г. Алматы, Казахстан
ORCID ID: 0000-0001-6570-2810
E-mail: abdukarimov1303@gmail.com

Деликешева Динара Насипуловна
Магистр технических наук, КазНИТУ им. К.И. Сатпаева, ул. Сатпаева, 22, 050000, г. Алматы, Казахстан
ORCID ID: 0000-0001-5442-4763
E-mail: d.delikesheva@satbayev.university

Момбеков Багдат Исаханович
Магистр технических наук, КазНИТУ им. К.И. Сатпаева, ул. Сатпаева, 22, 050000, г. Алматы, Казахстан
ORCID ID: 0000-0002-7112-8920
E-mail: b.mombekov@satbayev.university

Доктор Луис Зерпа
Ассоциированный профессор нефтяной инженерии, Колорадская горная школа, 1500 Иллинойс-Стрит, Голден, Колорадо, 80401, США
ORCID ID: 0000-0003-3211-2704
E-mail: lzerpa@mines.edu