

ISSN 2518-170X (Online)

ISSN 2224-5278 (Print)

ҚАЗАҚСТАН РЕСПУБЛИКАСЫ
ҰЛТТЫҚ ҒЫЛЫМ АКАДЕМИЯСЫ

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НАЦИОНАЛЬНОЙ АКАДЕМИИ
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N E W S

OF THE ACADEMY OF SCIENCES
OF THE REPUBLIC OF
KAZAKHSTAN
Satbayev University

SERIES

OF GEOLOGY AND TECHNICAL SCIENCES

3 (459)

MAY – JUNE 2023

THE JOURNAL WAS FOUNDED IN 1940

PUBLISHED 6 TIMES A YEAR

ALMATY, NAS RK

NAS RK is pleased to announce that News of NAS RK. Series of geology and technical sciences scientific journal has been accepted for indexing in the Emerging Sources Citation Index, a new edition of Web of Science. Content in this index is under consideration by Clarivate Analytics to be accepted in the Science Citation Index Expanded, the Social Sciences Citation Index, and the Arts & Humanities Citation Index. The quality and depth of content Web of Science offers to researchers, authors, publishers, and institutions sets it apart from other research databases. The inclusion of News of NAS RK. Series of geology and technical sciences in the Emerging Sources Citation Index demonstrates our dedication to providing the most relevant and influential content of geology and engineering sciences to our community.

Қазақстан Республикасы Ұлттық ғылым академиясы «ҚР ҰҒА Хабарлары. Геология және техникалық ғылымдар сериясы» ғылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Web of Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Геология және техникалық ғылымдар сериясы Emerging Sources Citation Index-ке енуі біздің қоғамдастық үшін ең өзекті және беделді геология және техникалық ғылымдар бойынша контентке адалдығымызды білдіреді.

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«ҚР ҰҒА Хабарлары. Геология және техникалық ғылымдар сериясы».

ISSN 2518-170X (Online),

ISSN 2224-5278 (Print)

Меншіктеуші: «Қазақстан Республикасының Ұлттық ғылым академиясы» РҚБ (Алматы қ.).

Қазақстан Республикасының Ақпарат және қоғамдық даму министрлігінің Ақпарат комитетінде 29.07.2020 ж. берілген № **KZ39VPY00025420** мерзімдік басылым тіркеуіне қойылу туралы куәлік.

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Мерзімділігі: жылына 6 рет.

Тиражы: 300 дана.

Редакцияның мекен-жайы: 050010, Алматы қ., Шевченко көш., 28, 219 бөл., тел.: 272-13-19

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«Известия НАН РК. Серия геологии и технических наук».

ISSN 2518-170X (Online),

ISSN 2224-5278 (Print)

Собственник: Республиканское общественное объединение «Национальная академия наук Республики Казахстан» (г. Алматы).

Свидетельство о постановке на учет периодического печатного издания в Комитете информации Министерства информации и общественного развития Республики Казахстан № **KZ39VPY00025420**, выданное 29.07.2020 г.

Тематическая направленность: *геология, химические технологии переработки нефти и газа, нефтехимия, технологии извлечения металлов и их соединений.*

Периодичность: 6 раз в год.

Тираж: 300 экземпляров.

Адрес редакции: 050010, г. Алматы, ул. Шевченко, 28, оф. 219, тел.: 272-13-19

<http://www.geolog-technical.kz/index.php/en/>

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News of the National Academy of Sciences of the Republic of Kazakhstan. Series of geology and technology sciences.

ISSN 2518-170X (Online),

ISSN 2224-5278 (Print)

Owner: RPA «National Academy of Sciences of the Republic of Kazakhstan» (Almaty).

The certificate of registration of a periodical printed publication in the Committee of information of the Ministry of Information and Social Development of the Republic of Kazakhstan **No. KZ39VPY00025420**, issued 29.07.2020.

Thematic scope: *geology, chemical technologies for oil and gas processing, petrochemistry, technologies for extracting metals and their connections.*

Periodicity: 6 times a year.

Circulation: 300 copies.

Editorial address: 28, Shevchenko str., of. 219, Almaty, 050010, tel. 272-13-19

<http://www.geolog-technical.kz/index.php/en/>

© National Academy of Sciences of the Republic of Kazakhstan, 2023

Address of printing house: ST «Aruna», 75, Muratbayev str, Almaty.

NEWS of the National Academy of Sciences of the Republic of Kazakhstan
SERIES OF GEOLOGY AND TECHNICAL SCIENCES
ISSN 2224-5278
Volume 3, Number 459 (2023), 96–107
<https://doi.org/10.32014/2023.2518-170X.302>

IRSTI:622.276(06)

© J.A. Ismailova^{1*}, A.R. Khussainova², Luis E. Zerpa³, D.N. Delikesheva¹,
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A NEW PREDICTIVE THERMODYNAMIC MODEL OF PARAFFIN FORMATION WITH THE CALCULATION OF THE MATHEMATICAL ORIGIN OF THE POYNTING CORRECTION FACTOR

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Abstract. One of the major and big problems of the oil industry is the deposition and crystallization of paraffin. This process can appear at any point in the design of the production process. Wax precipitation is a thermodynamically controlled process that leads to an increase in the viscosity of the oil mixture, a decrease in the internal diameter of wells and pipelines. Wax appearance temperature (WAT) is one of the important characteristics of the wax deposition process. To manage this procedure, it is required to mathematically accurately forecast the temperature at which the wax possibly could appear, to analyze the phase equilibrium of the system. This article discusses the development of a thermodynamic theoretical model for calculating the paraffin precipitation, using a mathematical approach and experimental data. To consider the issue of phase equilibrium, the fugacity parameter of the pure component is analyzed.

This article shows the Poynting correction factor, which is expressed as a function of fugacity at certain pressures and temperatures and fugacity at saturation. This aspect allows one to analyze the phase state of the system at high pressures. One of the important indicators that affects the behavior of wax deposition in the oil mixture is the melting point temperature. This paper presents the correlation of melting temperature, which is characterized by dependence on pressure and molecular weight of the components. Using this theoretical model, the calculation of the Poynting Correction factor, accurately predicts the wax appearance temperature as well as the weight of deposited wax.

Keywords: Poynting correction, phase behavior, temperature, pressure, fugacity

Acknowledgements. *This research was prepared upon the project "Development of PVT model for prediction of wax precipitation" AP09058452, within the framework of the competition is held on grant financing of fundamental and applied research young scientists on scientific and (or) scientific and technical projects for 2021-2023 by Ministry of Education and Science of the Republic of Kazakhstan.*

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ПОЙНТИНГ ТҮЗЕТУ КОЭФФИЦИЕНТІН ЕСЕПТЕУ АРҚЫЛЫ ПАРАФИН ТҮЗІЛҮДІН ЖАҢА БОЛЖАМДЫ ТЕРОМОДИНАМИКАЛЫҚ МОДЕЛІ

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Аннотация. Мұнай өнеркәсібінің негізгі және өзекті мәселелерінің бірі парафиннің тұнуы және кристалдануы болып табылады. Бұл процесс өндіріс

процесін жобалаудың кез келген нүктесінде пайда болуы мүмкін. Парафин тұнуы — мұнай қоспасының тұтқырлығының жоғарылауына, ұнғымалардың және құбырлардың диаметрінің кішіреуіне әкелетін термодинамикалық басқарылатын процесс. Парафиннің пайда болу температурасы парафин тұну процесінің маңызды сипаттамаларының бірі болып табылады. Бұл процесті басқару үшін парафиннің пайда болу температурасын математикалық дәл болжау, жүйенің фазалық тепе-теңдігін талдау қажет. Бұл мақалада математикалық тәсілді және тәжірибелік деректерді пайдалана отырып, парафин түзілудің болжамын есептеудің термодинамикалық теориялық моделін жасау қарастырылады. Фазалық тепе-теңдік мәселесін қарастыру үшін таза құрамдас бөліктің өтімділік параметрі талданады. Бұл мақалада белгілі бір қысымдар мен температуралардағы ұшымдылық және қанығу кезіндегі ұшымдылық функциясы ретінде көрсетілген Пойнтинг түзетуі қарастырылады. Бұл аспект жоғары қысымда жүйенің фазалық күйін талдауға мүмкіндік береді. Мұнай қоспасындағы парафин тұну әрекетіне әсер ететін маңызды көрсеткіштердің бірі балку температурасы болып табылады. Бұл мақалада қысым мен компоненттердің молекулалық салмағына тәуелділігімен сипатталатын балку температурасының корреляциясы берілген. Осы теориялық модельді пайдаланып, Пойнтинг түзету мерзімін есептеу, парафиннің пайда болу температурасын, сондай-ақ тұнылған парафиннің салмағын дәл болжайды.

Түйін сөздер: Пойнтинг түзету мерзімі, фазалық мінез-құлық, температурасы, қысым, құбылмалылық

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НОВАЯ ПРОГНОСТИЧЕСКАЯ ТЕРМОДИНАМИЧЕСКАЯ МОДЕЛЬ ПАРАФИНООБРАЗОВАНИЯ С РАСЧЕТОМ МАТЕМАТИЧЕСКОГО ПРОИСХОЖДЕНИЯ ПОПРАВочНОГО КОЭФФИЦИЕНТА ПОЙНТИНГА

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Аннотация. Одной из крупных и актуальных проблем нефтяной индустрии является осаждение и кристаллизация парафина. Данный процесс может появиться в любой точке проектирования процесса добычи. Осаждение парафина — это термодинамически контролируемый процесс, который приводит к увеличению вязкости нефтяной смеси, уменьшению диаметра скважин и трубопроводов. Температура появления парафина является одной из важных характеристик процесса осаждения парафина. Для контроля данного процесса необходимо математически точно прогнозировать температуру появления парафина, анализировать фазовое равновесие системы. В данной статье рассматривается разработка термодинамической теоретической модели для расчета прогноза выпадения парафина, при использовании математического подхода и экспериментальных данных. Для рассмотрения вопроса фазового равновесия анализируется параметр летучести чистого компонента. В данной статье затрагивается поправка Пойнтинга, которая выражается в зависимости летучести при определенных значениях давления и температуры и летучести при насыщении. Данный аспект позволяет анализировать фазовое состояние системы при высоких значениях давления. Одним из важных показателей, который влияет на поведение осаждения парафина в нефтяной смеси является температура плавления. В данной работе представлена корреляция температуры плавления, которая характеризуется зависимостью от давления и молекулярной массы компонентов. Использование данной теоретической модели, расчета Поправочного члена Пойнтинга позволяет точно прогнозировать температуру появления парафина, а также вес осажденного парафина.

Ключевые слова: поправка Пойнтинга, фазовое поведение, температура, давление, летучесть

Introduction

At the main stages of oil field exploration, from extraction, transportation and processing of the oil mixture, high pressure and temperature changes are observed. These facts are one of the reasons for the paraffin formation deposits. Paraffins are high molecular weight compounds that are a big problem in the oil industry. To manage this procedure, it is obligatory to analyze the behavior of wax under various thermodynamic conditions. To clear up this issue, it is needed to forecast the deposition of wax, taking into account the pressure increase and the temperature at which wax could start to precipitate.

Thermodynamic equilibrium refers to the condition of the equality within the chemical potentials or in another words fugacity.

Result and discussions

Normally, the computation of fugacity from such thermodynamic potentials is awkward, and for other substances in the condensed phase, such a potential may be absorbed (Ghanaei et al., 2007). A convenient alternative is to start with the fugacity of pure components at saturation pressure, which usually differs by only small correction from the readily available vapor pressure of the pure component, where γ_i - the fugacity coefficient at saturation (for ideal gas is equal to 1, for low and moderate pressure is typically slightly less than 1) (Reza Dalirsefat et al., 2007). Thereby the needed value are:

$$P = \frac{f_i^{pure}(T,P)}{f_i^{pure}(T,P^{sat})} \quad (1)$$

P – Poynting correction (Allan H. Harvey, 2017).

Basic idea is that we calculate the fugacity of a compressed liquid or say a solid.

For a system that is at a pressure greater than saturation at a given temperature, so if we think about compressed liquid, if we are at the same temperature, on the same isotherm but at pressure greater than saturation for that given T right compressed liquid we could do the same thing for a solid.

The mathematical origin of the Poynting correction

C — compressed phase.

Fugacity at some given temperature and pressure.

$$\ln F^C(T, p) \quad (2)$$

Add 0, so if we calculate $\log F^C$ that's equivalent to:

$$\ln F^C = \ln F^C + (\ln F^{sat} - \ln F^{sat}) \quad (3)$$

First term - state at the same temperature, but at the corresponding saturation pressure (T, P^{sat}).

$$= \ln F^{sat} + (\ln F^C - \ln F^{sat}) \quad (4)$$

At constant T

For an isothermal process the differential $\log F$ is equivalent to the differential of our dimensionless molar Gibbs free energy. Const T — DT is zero.

$$d \ln F = d \left(\frac{G}{RT} \right) = \frac{V}{RT} dp \quad (5)$$

$$P \leftarrow P^{sat}$$

$$\int_{P^{sat}}^P d \ln F = \ln F^C - \ln F^{sat} = \int_{P^{sat}}^P \frac{V}{RT} dp \quad (6)$$

or,

$$\ln F^C = \ln F^{sat} + \left[\int_{P^{sat}}^P \frac{V}{RT} dp \right] \quad (7)$$

Last term is the Poynting correction (Nichita et al., 1999). It's just the change in the log fugacity and going from P^{sat} to P at the same T .

$$\ln \frac{F^C}{F^{sat}} = \int_{P^{sat}}^P \frac{V}{RT} dp \quad (8)$$

F^C — *fugacity* of compressed phase

$$\frac{F^C}{F^{sat}} = \int_{P^{sat}}^P \frac{V}{RT} dp \quad (9)$$

$$F^C = F^{sat} \left[\exp \left[\int_{P^{sat}}^P \frac{V}{RT} dp \right] \right] \quad (10)$$

$\left[\exp \left[\int_{P^{sat}}^P \frac{V}{RT} dp \right] \right]$, again Poynting correction, is it change in log fugacity going from saturation to compressed phase.

Or you can represent this equation (9) in the form:

$$f_i = \varphi_i^{sat} P_i^{sat} \exp \frac{V_i^L(P - P_i^{sat})}{RT} \quad (11)$$

To analyze the significance of the *Poynting factor* in calculations, we can consider the case – the gamma/phi formulation of VLE (vapor-liquid equilibria).

To determine the activity coefficient in the liquid phase we use:

$$\widehat{f}_i^l = x_i \gamma_i^l f_i^l \quad (12)$$

The foundation for VLE equations represents for equilibrium:

$$f_i^v = \widehat{f}_i^l = f_i^{sat} \quad (13)$$

$$\widehat{f}_i^v = \widehat{f}_i^l \quad (14)$$

$$f_i^v = \widehat{\varphi}_i^v y_i P \quad (15)$$

$$y_i \widehat{\varphi}_i^v P = x_i \gamma_i^l f_i^l \quad (16)$$

We can transform this equation into a working formula:

$$f_i^l = \varphi_i^{sat} P_i^{sat} \exp \frac{V_i^L(P - P_i^{sat})}{RT} \quad (17)$$

As an example, in this article, the correction factor computation was carried out at different pressures. For example, the following values were taken: 0.17 MPa, 0.36 MPa and 0.57 MPa. If we consider the relationship between the correction factor and pressure, then this correlation is represented by a linear function.

The given model does not use adaptable variables; the theoretical or ideal model was also observed in the research. The principal computations are built on thermodynamic wax sedimentation modeling surveys to outline the response of the solid and liquid phases (Lira-Galeana et al., 1996).

Fusion temperature values depend on pressure. By analyzing of literary sources on this topic, a relationship is observed between the parameters of pressure and melting temperature. Based on the data, we study the correlation to describe the behavior of the melting temperature at different pressures. When analyzing this aspect, the dependence of melting properties on pressure and molecular weight of the component is observed. Applying the theoretical approach of the Wilson and the calculated previously correction term, we could obtain an algorithm for forecasting the wax sedimentation at different pressure conditions.

Phase behavior calculations using cubic equation of state to predict of reservoir fluid require components characteristics such as (1) critical pressure, (2) critical temperature, (3) acentric factor, (4) binary interaction parameters (Won, 1986). Separation techniques including gas chromatography and distillation are not 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.

For correlation computation, it is needed to take components which molecular weights are higher than heptane. Then obtained correlation was used for entire calculations.

Using the *Poynting Correction Factor*, calculations of the multisolid model were carried out, as well as modifications of the melting temperature correlations.

The correlation modification algorithm is presented below:

- Based on the results of the experiment, calculations were made to determine the results of the melting points of each component.

- A program was written in the Python programming language to determine the correlation coefficients in order to improve the accuracy of the calculations.

- A test of the calculation results was carried out using the modified correlation.

Thus, the modified correlation (2023) for kazakhstan field X is as follows:

$$T_i^f = 101,82154 + 0,02617 * M_i - 20172/M_i \quad (18)$$

The dependence of the melting point T_f on the molecular weight of the component M_i is shown in the graph below for components from C_6 hexane to C_{36+} Hexatriacontane for X and Y fields.

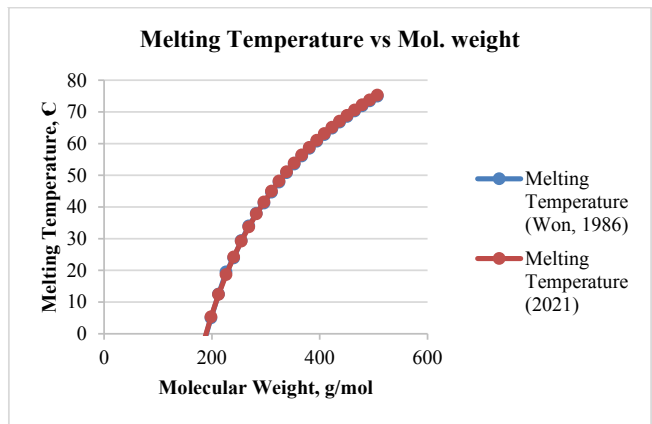


Fig. 1 - Dependence of the melting point T_f on the molecular weight of the component M_i

To obtain the necessary calculation data, samples of two different oils from the X and Y fields were taken; laboratory experiments were carried out to determine the component composition of oil necessary for the implementation of the subsequent tasks of the project from C_1 to C_{36+} , splitting of components. And also, the physical and chemical properties of the surface oil sample, necessary for calculating the multi-solid solution model, as well as the melting and transition temperatures to the solid state.

As a result, the proposed correlation gives an accuracy of 7% compared to Won's (1986) correlation for the X and Y fields.

According to the article (Pan, H, Firoozabadi et al., 1997), it was experimentally established that only heavy hydrocarbons from C_{15} to C_{20} and above are present in the precipitated paraffin.

To refine the results for Pentadecane $C_{15}H_{32}$ to Eicosane $C_{20}H_{42}$, the correlation was recalculated using the Python programming language and plotted, which showed an accuracy of 11% for the six components compared to Won's (1986) correlation.

$$T_i^f = 99,833 + 0,02617 * M_i - 20172/M_i \quad (20)$$

The calculation results are presented in tables 4 and 5.

Table 4 – Melting point modification results (Pentadecane – Eicosane)

Components	Mol. Weight (%)	Molar mass	Experimental (literature) data	Correlation Results (Won)	Correlation Results (2021)
Pentadecane	3.439	212.42	10	12.5	10.43
Hexadecane	2.878	226.41	18.1	19.8	16.66
Heptadecane	2.791	240.471	21.9	23.90775	22.24
Octadecane	2.833	254.494	28.1	29.4	27.23
Nonadecane	2.707	268.518	32	33	31.73
Eicosane	3.439	212.42	36.6	34	35.83

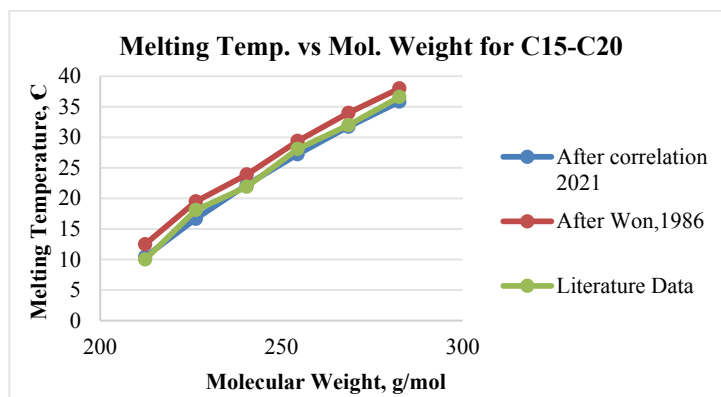


Fig. 2 – Melting point versus molecular weight for 6 components

Table 5 – Results of modification of the melting point

Component	Weight, %	Molar mass	Reference data	Correlation Results (Won, 1986)	Correlation Results (2021)
Hexane	0,984	86,18	-95	-135,286	-129,897654
Heptane	1,56	100,2	-91	-97,6	-96,9134560
Methylcyclohexane	0,632	98,18		-101,27	-101,1012375
Toluene	0,195	92,14		-115,112	-114,7199234
Octane	2,346	114,23	-70,2	-72,75	-71,6998721
Nonane	2,502	128,2	-54	-52,7	-52,09982172
Decane	2,666	142,28	-30	-36,5	-36,30156389
Undecane	2,605	156,31	-26	-23,8	-23,09983625
Dodecane	2,649	170,32	-10	-13,3698	-12,20982645
Tridecane	3,072	184,37	-5,5	-4,1564	-2,69987654
Tetradecane	2,914	198,39	5,8	6,00087	5,298376354
Pentadecane	3,439	212,42	10	12,7	12,398746531
Hexadecane	2,878	226,41	18,1	19,9	18,701123456
Heptadecane	2,791	240,471	21,9	23,8734	24,312209879
Octadecane	2,833	254,494	28,1	29,7	29,199845321
Nonadecane	2,707	268,518	32	35	33,812765864
Eicosane	2,465	282,5475	36,6	337	37,908712389
Heneicosane	2,456	296,57	40,2	42,2835	41,49912875
Docosane	2,107	310,6027	44	49,57344	45,17456448
Tricosane	2,861	324,6	47,5	49,7986	48,07896433
Tetracosane	2,574	338,66	50,6	51,85687	51,00981247
Pentacosane	2,718	352,69	53,5	55,8734	53,90012987
Hexacosane	2,882	366,71	56,3	58,22455	56,38754320
Heptacosane	3,084	380,74	58,8	59,39987	58,90012976
Octacosane	2,925	394,77	61,2	61,6554	61,12009563
Nonacosane	3,272	408,6	63,7	63,79965	63,00981754
Triacontane	3,308	422,82	65,4	69,79954	65,29844032
Hentriacontane	3,216	436,85	67,9	68,65432	67,11763943

Dotriacontane	3,099	450,86	69,7	69,98886	68,90118463
Tritriacontane	2,856	464,9	71,2	71,31009	70,61876432
Tetratriacontane	2,397	478,9	72,6	72	72,19876543
Pentatriacontane	2,655	492,9	75	73,7	73,81764493
Hexatriacontane	16,504	506,973	75,08	74,6	75,00168854

The absolute average deviation was calculated for these components using the following formula:

$$\frac{1}{n} \sum_{i=1}^n |x_i - m(X)| \quad (19)$$

Performing *material balance* for liquid-multi-solid equilibrium (Juan Carlos Martin Escobar-Remolina, 2005)

$$s_i = z_i - x_i^l \left(\frac{\varphi_i^l}{\varphi_i} \right) \quad (20)$$

Calculation of paraffin crystallization (P.c) at a given temperature

$$P. c (\%) = \frac{\sum_j^{N_s} M_j s_j}{(\sum_i^N M_i z_i)_{T_{WAP}}^{T_f}} \times 100 \quad (21)$$

T_{WAP} and T_f — is the temperature range, in which precipitation occurs from the wax appears point to the final temperature.

Using an iterative process, the *multisode* model was calculated and compared with the *PVTsim* program.

The table below indicated the values for $P_1 = 0.17$ MPa obtained by proposed model, PVTsim software and experimental data. According to the data, the results within a good agreement.

Table 6 — Wax appearance temperature results for sample 1.01 (P = 0.17MPa)

WAT model	317.5079 K
WAT PVTsim	317.57 K
WAT experimental	315.15 K

Table 7 – Experiment results for sample 1.01

Field X oil 1.01
Models
PR Peneloux (T)
CSP Visc/Thermal Cond
Wax Appearance Temperature

Pressure	Temperature
kPa	K
170.00	317.57
360.00	317.60
570.00	317.64

The line graph below showed the percentage of wax weight versus temperature for sample 1.01, $P_1 = 0.17$ MPa.

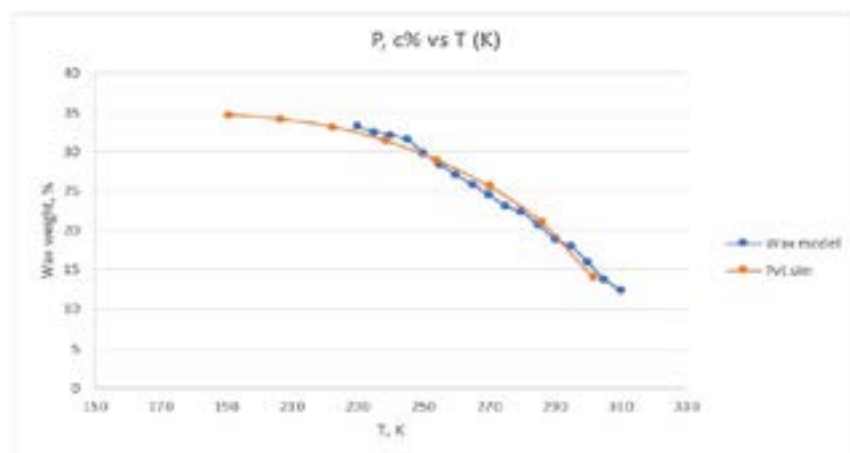


Fig. 4 – Crystallized paraffin (%) versus Temperature (K) for sample 1.01

Conclusions

Wax deposition phenomena will always be an important task in the oil industry. In this article, a mathematical theoretical model of the behavior of paraffin during precipitation was developed by analyzing the correlation of the calculation of the melting point temperature, fugacity with respect the correction factor, assessing the crystallization of paraffin at a given temperature. As results, experimental data on the dependence of melting properties on the molecular weight of the components are presented. The model presented in the article, taking into account the Poynting correction factor and modified temperature gave the best result for predicting wax deposition.

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CONTENTS

D.K. Akhmetkanov, M.Zh. Bitimbayev, V. Lozynskyi, K.B. Rysbekov, B.B. Amralinova NEW VARIANTS FOR WIDE OREBODIES HIGH-CAPACITY MINING SYSTEMS WITH CONTROLLED AND CONTINUOUS IN-LINE STOPPING.....	6
F.A. Akhundov, M. Sarbopeeve, R. Bayamirova, A. Togasheva, A. Zholbasarova ON THE ISSUE OF PREPARING THE WELLBORE FOR ITS FASTENING.....	22
A.M. Baikadamova, Y.I. Kuldeyev GEOLOGICAL STRUCTURE OF THE ZHARKENT THERMAL GROUNDWATER DEPOSIT BY THE EXAMPLE OF WELL 3-T.....	35
A.A. Yerzhan, P.V. Boikachev, B.R. Nakisbekova, Z.D. Manbetova, P.A. Dunayev METHOD OF SYNTHESIS OF MATCHING TELECOMMUNICATION DEVICES BASED ON THE METHOD OF REAL FREQUENCIES FOR 5G ANTENNAS IN A DISTRIBUTED ELEMENT BASIS.....	47
K.S. Zaurbekov, S.A. Zaurebkov, A.V. Sladkovsky, D.Y. Balgayev HYDRODYNAMIC SIMULATION OF THE STEAM-ASSISTED GRAVITY DRAINAGE METHOD FOR DIFFERENT RESERVOIR THICKNESSES USING ECLIPSE.....	60
A.T. Ibrayev, D.A. Aitimova A METHOD FOR ACCOUNTING THE IMPACT OF ERRORS ON THE QUALITY OF ANALYTICAL INSTRUMENTS AND OPTIMAL CONTROL SYSTEMS.....	70
I.G. Ikramov, G.I. Issayev, N.A. Akhmetov, SH.K. Shapalov, K.T. Abdraimova RECYCLING OF PRODUCTION WASTE AND ENVIRONMENTAL IMPACT ASSESSMENT.....	80
J.A. Ismailova, A.R. Khussainova, Luis E. Zerpa, D.N. Delikesheva, A.A. Ismailov A NEW PREDICTIVE THERMODYNAMIC MODEL OF PARAFFIN FORMATION WITH THE CALCULATION OF THE MATHEMATICAL ORIGIN OF THE POYNTING CORRECTION FACTOR.....	96
Zh.S. Kenzhetaev, K.S. Togizov, A.K. Omirgali, E.Kh. Aben, R.Zhalikyzy INTENSIFICATION OF INHIBITOR-ASSISTED URANIUM ISL PROCESS.....	108
M.A. Li, T.T. Ibrayev, N.N. Balgabayev, B.S. Kali, D.A. Toleubek SIMULATION AND OPTIMIZATION MODELING OF WATER USE MANAGEMENT IN IRRIGATION SYSTEMS.....	119
A.S. Madibekov, L.T. Ismukhanova, A.O. Zhadi, A. Mussakulkyzy, K.M. Bolatov RANKING THE TERRITORY OF THE ALMATY AGGLOMERATION ACCORDING TO THE DEGREE OF POLLUTION.....	130
E.K. Merekeyeva, K.A. Kozhakhmet, A.A. Seidaliyev CHARACTERISTICS OF THE STRUCTURAL UPLIFTS OF KURGANBAI AND BAYRAM-KYZYLADYR LOCATED WITHIN THE ZHAZGURLI DEPRESSION.....	149
R.N. Moldasheva, N.K. Shazhdekeyeva, G. Myrzagereikyzy, V.E. Makhatova, A.M. Zadagali MATHEMATICAL FOUNDATIONS OF ALGORITHMIZATION OF WATER POLLUTION MODELING PROCESSES.....	164
Y.G. Neshina, A.D. Mekhtiyev, A.D. Alkina, P.A. Dunayev, Z.D. Manbetova HARDWARE-SOFTWARE COMPLEX FOR IDENTIFICATION OF ROCK DISPLACEMENT IN PITS.....	180

M.B. Nurpeisova, Z.A. Yestemesov, V.G. Lozinsky, A.A. Ashimova, S.S. Urazova INDUSTRIAL WASTE RECYCLING – ONE OF THE KEY DIRECTIONS OF BUSINESS DEVELOPMENT.....	193
B. Orazbayev, M. Urazgaliyeva, A. Gabdulova, Zh. Moldasheva, Zh. Amanbayeva METHODS OF MULTI-CRITERIA SELECTION IN PETROLEUM GEOLOGY UNDER CONDITIONS OF FUZZY INITIAL DATA.....	206
B.R. Rakishev, A.A. Orynbay, A.B. Mussakhan AUTOMATED FORECASTING OF THE PARTICLE SIZE COMPOSITION OF BLASTED ROCKS DURING BLASTHOLE DRILLING IN HORIZONTAL UNDERGROUND WORKINGS.....	222
Y.Sh. Seithaziyev GEOCHEMICAL STUDIES OF CONDENSATE, GAS AND CORE SAMPLES DERIVED FROM GAS-CONDENSATE FIELDS IN THE MOYNKUM SAG (KAZAKHSTAN).....	242
E.Yu. Seitmuratova, R.T. Baratov, F.F. Saidasheva, V.S. Goryaeva, M.A. Mashrapova, Ya.K. Arshamov TO STUDY THE RING STRUCTURES OF CENTRAL AND SOUTHERN KAZAKHSTAN AND THEIR ORE CONTENT.....	262
J.B. Toshov, Sh.R. Malikov, O.S. Ergashev, A.K. Sherov, A. Esirkepov IMPROVING THE EFFICIENCY OF THE PROCESS OF DRILLING WELLS IN COMPLEX CONDITIONS AT GEOLOGICAL PROSPECTING SITES.....	282
V.A. Tumlert, Zh.K. Kasymbekov, R.A. Dzhaismbekova, E.V. Tumlert, B Sh. Amanbayeva INFLUENCE OF THE HYDROGEOLOGICAL MODE OF OPERATION ON THE CHARACTER OF COLLATING OF THE FILTER AND THE FILTER ZONE OF SEASONAL WELLS.....	295

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ISSN 2518-170X (Online),

ISSN 2224-5278 (Print)

Заместитель директор отдела издания научных журналов НАН РК *Р. Жәліқызы*

Редакторы: *М.С. Ахметова, Д.С. Аленов*

Верстка на компьютере *Г.Д. Жадыранова*

Подписано в печать 14.06.2023.

Формат 70х90^{1/16}. Бумага офсетная. Печать – ризограф.

20,0 п.л. Тираж 300. Заказ 3.