An experimental-analytical model for the dissolution of asphaltene-resin-paraffin deposits with determination of the influence coefficients of solvent components and ultrasonic treatment

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Abstract. The purpose of this work is to predict the effectiveness of solvents, including under the influence of ultrasonic exposure, on asphalt-resin-paraffin deposits with a given group composition and to assess their economic effectiveness. The article discusses the historical aspects of the use of ultrasound. Physical and chemical studies of asphalt-resin-paraffin deposits were carried out. The process of removal of asphalt resin-refined deposits in a statistical mode with additional ultrasonic exposure and without ultrasonic exposure was investigated. Based on experimental studies, the effect of the component composition on the effectiveness of various solvents was established. An assessment of the efficiency and economic indicators of cleaning various ARPD by various methods was obtained.

Keywords: asphaltene-resin-paraffin deposits, asphaltene-resin-paraffin deposits, solvent, ultrasound, hydrocarbon, composition, deposit.

1. Introduction

In order to combat asphalt-resin-paraffin deposits (ASPO), various methods are used, such as chemical, thermal, mechanized. The most common and effective method is chemical and combined scale removal.

The essence of the chemical method is that inhibitor reagents are used. The use of solvents is an effective method, since they protect the entire production and transport chain of oil [1].

Complex solvents containing hydrocarbon compounds are used to accelerate the dissolution process and increase the decomposition of deposits. When choosing solvents, it is necessary to take into account the content and type of deposits, since their solubility depends on the specific type of deposits [2, 3].

In the experiments, sediment samples were used, which consist of resins, paraffin and aromatic hydrocarbons. The following hydrocarbon solvents were used: TSK “B” and TSK “A”.

Despite a considerable amount of research in the field of removal of asphalt-resin-paraffin deposits, it is not always possible to achieve positive results. To do this, it is necessary to pay attention to two aspects: technologies that can reduce environmental damage when removing sediments, and have low capital costs.

In world practice, methods are known to solve such problems. For example, Russian and foreign scientists have proposed methods for removing deposits using ultrasound exposure. Studies were conducted, the results of which make it possible to conclude that the use of ultrasonic cavitation will make it possible to destroy AFS at oil transport and storage facilities, restoring their design parameters, while without adversely affecting the environment and structural strength, but they do not contain requirements and recommendations for methods of using ultrasonic equipment for AFS removal. Since the analysis of the work shows that there are a large number of questions and unresolved problems and, accordingly, for a greater understanding of the mechanism of ultrasonic cleaning of tanks from deposits, as well as the development of appropriate technology, it is necessary to conduct experimental studies.

In continuation of scientific research in the field of sediment control, auto-rams, the method of cleaning from ASPO by chemical means is being investigated, additionally acting with ultrasound. But at the same time, a model will be proposed for predicting the effectiveness of solvents depending on the composition of deposits, the composition of solvents, but also under ultrasonic exposure, and assessing their economic effectiveness.

To determine the most effective cleaning technology, it is necessary to study the physicochemical properties of deposits and conduct analytical work to determine the optimal cleaning method.

Instruments used in oil transportation use ultrasound to change the physical and chemical properties of formation fluids. This prevents the formation of paraffin and asphalt-tar deposits in the oil, which allows for deeper processing and increases the recovery of light fractions.

Ultrasonic treatment due to high-frequency waves, the effect of cavitation destroys oil sludge. But besides this, on tanks with medium viscous and heavy oil, a significant decrease in oil viscosity is observed.

Physicochemical methods are more technological, but at the same time they have several advantages. But for the success of the technology, it is necessary to determine the physicochemical characteristics of the deposits.

2. Study objects, reagents, and auxiliary substances

Eight samples of asphalt-resin-paraffin deposits from existing pipelines and tanks were selected as the objects of the study. In addition to the organic fraction, asphaltene-resin-paraffin deposits contain water and mechanical impurities.

The inorganic part of the deposits can be divided into mechanical impurities and salts, which are present both in dissolved form and as part of the mechanical impurities. Table 1 presents the physicochemical characteristics of the asphaltene-resin-paraffin deposits

Table 1. Composition of organic part of the sediments

ARPD	Paraffin-naphthenic,	Aromatic, %			Resins, %		Asphalteneresins, %	Σ (Resins - Asphalteneresins), %
No. 1	23.9	9.6	7.6	14.9	4.2	39.8	8.5	52.5
No. 2	4.8	9.9	2.2	3.2	3.9	76	10.5	94.4
No. 3	28.9	45.1	2.5	4.1	2.9	16.5	4.0	23.4
No. 4	17.7	26.9	14.5	13.5	6.0	17.2	4.2	27.4
No. 5	41.5	9.0	8.5	17.4	5.9	13.6	3.8	23.3
No. 6	52.0	5.7	6.6	15.6	5.9	10.3	4.0	20.1
No. 7	52.5	8.5	6.9	15.3	5.2	8.4	3.4	17
No. 8	48.3	5.8	A2.8	19.3	5.1	14.3	4.4	23.8

The following grade reagents were used for the test: TSK “A”, TSK “B”, a mixture of technical aliphatic and aromatic hydrocarbons of grade “G”, a mixture of technical aliphatic and aromatic hydrocarbons of grade “G”, ITPS-010 (Russia). The composition of the test agents is shown in Table 2.

An ultrasonic emitter was used to further enhance the effectiveness of the solvents.

To analyze the content of aromatic hydrocarbons, asphaltenes, resins and paraffins in sediments, a method was used based on measurements of the group chemical composition of heavy petroleum products using the Gradient-M laboratory installation with silica gel with a boiling point of more than 300 °C [1, 2, 3]. The method is based on the principles of liquid adsorption chromatography with gradient displacement.

The analyzed product in a liquid chromatograph is divided into seven groups: paraffin-naphthenic hydrocarbons, light aromatic (or mono-aromatic), medium aromatic (or bi-aromatic), heavy aromatic hydrocarbons (containing more than two-benzene rings), as well as resins 1, 2 and asphaltenes.

To determine the amount of water, the Dean Stark method was used in accordance with SS 6370 and the content of mechanical impurities was determined in accordance with SS 6370. Physical and chemical characteristics of deposits are presented in Table 2, 3.

Preparation of the sample for chromatographic analysis. To determine the group composition of deposits, ASK silica gel is poured into a clean and dried chromatographic column and compacted using a vibrator. Weigh a weighed amount of the analyzed ASPO sample with a weight of 0.1 g of analytical balance, which is dissolved with a mixture of benzene and cyclohexane 1:1 at the rate of: per 7 ml of a mixture of benzene with cyclohexane per 0.1 g of the sample. Then, 1 μl of the sample solution is introduced into the upper layer of silica gel in the column using a microspray.

Eluent alone is added as solvent for desorption of deposits, and eluent is added for desorption of asphaltenes. Then 1.5-2 ml of eluent is introduced with a medical syringe. After all separated components of the maltenic part have left the column and six chromatographic peaks are recorded in the chromatogram, release the pressure.

Remove the column with a syringe and remove the remaining solvent. Then desorption of asphaltenes with eluent 2 is carried out, the column is again installed in the chromatograph and pressure is applied. After the asphaltene peak came out, the column was switched on [1]. The order of yield of the groups is as follows: 1 peak – paraffin - naphthenic hydrocarbons; 2 peak – light aromatic hydrocarbons; 3 peak – medium aromatic hydrocarbons; Peak 4 – heavy aromatic hydrocarbons; 5 peak – resins I; 6 peak – resins II; 7 peak – asphaltenes.

Chromatograms were calculated by internal normalization by measuring the peak areas recorded on the cartogram. The peak area, approximately taken as the area of the triangle (mm^2), is calculated using the formula:

$$S = ha, \tag{1}$$

where h – the height of the peak corresponding to the baseline segment to the peak top, mm; a – peak width measured at a distance equal to 1/2 of its height, mm.

Table 2. Solvent formulations tested (fractions)

Solvent grade	2-ethylhexanol	Alcohols C_8	C_5-C_6	Benzene	Aromatic hydrocarbons
TSK “A”	0,21	0,35	0	0	0
TSK “B”	0,27	0	0	0	0
STAAU “B”	0	0	0,26	0,4	0
STAAU “Zh”	0	0	0	0	0,3
ITPS-010	0	0	0,19	0	0,51

Experimental installation I100-840. A digitally controlled ultrasonic laboratory apparatus for I100-840 fluids was used as an additional source of ultrasonic waves.

The apparatus is designed to study the effect of ultrasound on liquid media in cavitation and pre-cavitation mode.

The installation allows liquids and parts to be processed in a user's container mounted on a tripod table.

Component units consist of:

– Instrument I10-840 ultrasonic generator.
– Rack.
– Replaceable waveguides.
– Magnetostrictive PMS-1-22 transducer.
– Magnetostrictive PMS-1-44 transducer.
– Studs.

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3. Experimental part

Studies to determine the effectiveness of solvents were carried out in a statistical mode without ultrasonic exposure (US) and an additional effect using an ultrasonic dispersant immersed in the volume with the processed raw materials.

In this work, in the experimental part, a methodology for studying the washing, dissolving and dispersing properties of a solvent in a static mode without ultrasonic exposure and with ultrasonic exposure to the solvent is considered.

Experiment 1. Consider a study of the effectiveness of a solvent, which includes: the washing, dissolving and dispersing abilities of the solvent in a static mode:

– Prepare 8 samples of sediments weighing within 1-3 g.
– Place in a pre-weighed basket made of metal mesh with a mesh size of 1.5×1.5 mm.
– Place the basket with the APRD sample in a sealed glass cell with a volume of 150 ml, pour the test solvent with a volume of 100 ml.
– The dissolution time of the test samples was 2 hours, and after 120 minutes the basket with the remaining undamaged part of the APRD was removed and dried to a constant weight at atmospheric pressure at a temperature of 20 °C for at least 24 hours.
– After the contact time with the sample, the deposits were filtered through a prepared paper filter placed in a glass funnel. The filter with the residue was then dried for 24 hours outdoors. A day later, the filter with the residue was dried to a constant weight in a desiccator.

After 24 hours, the baskets were removed from the wells and weighed with the remainder of the ASPO in an analytical balance with an accuracy ± 0.005 g.

The number of experiments on the effect of solvents on ASPO was 120.

Experiment 2. Consider the effectiveness of the solvent in static mode together with ultrasound action:

An ultrasonic I10-840 generator was used for the ultrasound experiments.

– Prepare 8 sediment samples weighing within 1-3 g.
– Place in a basket made of metal mesh with a mesh size of 1.5×1.5 mm.
– Place the corzine with the sample into a 150 mL cell filled with a 100 mL solvent.
– Sonicate (44 Khz) the test deposit and solvent for 5 minutes.
– After the specified time, US was turned off and the test sample was left in the cell for 115 minutes. At the end of the experiment, the metal basket with residual deposits was air dried for a day until a constant weight was achieved and weighed on an analytical balance with an accuracy of 0.005 g. The contents of the glass flask were filtered through a Blue Ribbon paper filter placed in a glass funnel.

The number of experiments on the effect of solvents on ARPD with ultrasonic exposure (HC) was 120.

4. Discussion of results

From Table 2, we set the composition of the solvents to five numbers that can be combined into a matrix:

$$S = [S_{2eg}, S_{C8}, S_{C5C6}, S_B, S_{Arom.hydrocarbon}]. \quad (2)$$

Solvent efficiency is defined as evaluation of asphalt-resin-paraffin deposits by dissolving and dispersing capacity determined by gravimetric method.

Consider the following model for determining solvent efficiency. It is believed that 1 % of each solvent component, acting on 1 % of each ARPD component, dissolves a portion thereof. The efficiency for each "solvent component" – "AFS component" pair lends itself to additivity, so the total efficiency is equal to the sum of the corresponding components. Thus, the overall efficiency is:

$$Э = \sum_{x \in X} \sum_{s \in S} k_{s,x} \cdot s \cdot x. \quad (3)$$

To improve the model, we assume that the solvent cannot dissolve any ARPD component more than it is available. Thus:

$$Э = \sum_{x \in X} \min\left(x; \sum_{s \in S} k_{s,x} \cdot s \cdot x\right). \quad (4)$$

However, within the framework of this model, it is necessary to determine the dissolution coefficients $k_{s,x}$. Their number is equal to the length S multiplied by the length X, in this case 20.

To find these coefficients, experimental studies of the effectiveness of solvents in a statistical mode were performed.

Laboratory experiments have shown that ultrasound increases the indicators of the dispersing and washing abilities of deposits.

Ultrasonic exposure increases the efficiency of solvents from 40 to 72 %. Ultrasound greatly increases the effectiveness of weak solvents, which allows the use of cheaper solvents without reducing the effectiveness of ultrasound and allows you to reduce the consumption of solvents by 1.4-13 times, since there is a synergistic effect due to the effect of ultrasound, that is, depolarization.

Also, to obtain the coefficients, there was the use of a numerical solution based on the use of the Nelder-Mead algorithm, which is intended for an effective method of solving optimization problems.

Within the framework of this algorithm, the standard error of Eq. (3) was minimized.

The bounds of the coefficients $k_{s,x}$ are taken as (0, 10).

The results of the dissolution rate algorithm $k_{s,x}$ showed the following results, which are presented in Table 3 and 4.

Table 3. Dissolution factors $k_{s,x}$ without ultrasonic exposure

Composition of solvents	Paraffins	Aromatic	Resins	Asphaltenes
2-ethylhexanol	1.11	0.28	1.29	2.84
Alcohol C_8	0.04	0.02	0.03	0.09
C_5-C_6	0.01	0.13	2.93	3.78
Benzene	3.97	4.34	0.68	0.04
Aromatic hydrocarbons	3.48	3.14	4.05	3.29

The standard error was 26.6 %.

Table 4. Dissolution factors $k_{s,x}$ using sonication

Composition of solvents	Paraffins	Aromatic	Resins	Asphaltenes
2-ethylhexanol	19.2	8.2	12.3	4.7
Alcohol C_8	0.1	2.9	0.0	0.1
C_5-C_6	4.1	1.9	3.3	4.6
Benzene	4.2	4.6	0.7	0.1
Aromatic hydrocarbons	3.5	4.0	5.8	16.3

The standard error was 28.1 %.

Comparing the results of Tables 4 and 5, it can be seen that ultrasound can intensify the destruction process.

The additional ultrasound effect values are shown in Table 5.

Table 5. Additional dissolution effect using ultrasound

Composition of solvents	Paraffins	Aromatic	Resins	Asphaltenes
2- ethylhexanol	18.14	7.90	11.02	1.88
Alcohol C_8	0.01	2.85	0.02	0.04
C_5-C_6	4.10	1.76	0.39	0.86
Benzene	0.21	0.22	0.06	0.03
Aromatic hydrocarbons	0.00	0.87	1.80	13.00

From the data in Table 5, it can be seen that an additional effect is likely to be exerted by ultrasound.

Under the influence of ultrasound, paraffin dissolves on average 4.49 % faster, aromatic by 2.72 %, resins by 2.66 %, asphaltenes by 3.16 %.

During US treatment, a time-dependent stress acts on each liquid molecule, and if the duration of exposure exceeds the time, then the intermolecular bonds break. Due to the effect of high-intensity ultrasonic vibrations, the continuous chain breaks, which leads to the destruction of the bonds between parts of the C-C bond paraffin molecules, including the physicochemical properties of liquids.

Application of cavitation changes the ability of resinous substances to bind solid particles of deposits by their destruction and transition from phase to coagulation contact. This leads to a more efficient dissolution of resinous substances and an increase in their detergency.

If the components of the solvent are analyzed through a prism, the following intensification of the effect of the solvent using ultrasound is determined:

– 2-ethylhexanol – 9.73 %.
– Alcohols C_8 – 0.73 %.
– C_5-C_6 – 1,78 %.
– Benzene - 0.13 %.
– Aromatic hydrocarbons – 3.92 %.

Let's evaluate the economic parameters of the destruction of APRD.

Let's take the following cost of solvents:

– TSK "A" – 67000 thousand rubles/ton.
– TSK "B" – 44,000, thousand rubles/ton.
– STAAU "Z" – 65,000, thousand rubles/ton.
– STAAU "B" – 48,000, thousand rubles/ton.
– ITPS-010 – 110,000. thousand rubles/tons.

We assume that the remaining components of the solvents, except for those presented in Table 1, have a negligible cost, then the cost of the components can be restored:

– 2-ethylhexanol – 125714 thousand rubles/ton.
– Alcohols C_8 – 150370 thousand rubles/ton.
– C_5-C_6 – 120000 thousand rubles/ton.
– Benzene – 64246 thousand rubles/ton.

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– Aromatic hydrocarbons – 177895 thousand rubles/ton.

Thus, the cost of the components can be represented as a matrix $C = [125714, 150370, 120000, 64246, 177895]$.

Therefore, the cost of the solvent having the predetermined composition S will be $C \times S^T$.

To determine the optimal component composition, the following expressions will be used as a loss function:

$$L = C + (1 - Э) \cdot C_S. \quad (5)$$

5. Conclusions

The idea behind this loss function on the one handreduce the cost of the solvent. On the other hand-minimize unsolved residue. We accept, that the residue of APRD can be removed with very powerful solvent ITPS-010, then as C_S parameter we accept its cost C_S = 200,000 thousand rubles/ton. This approach is used to prevent models tend to dissolve only small amounts of APRD, for which it will always be more effective.

Thus, as a result of studying the impact of ultrasound on solvents with deposition is established, that the impact of ultrasonic vibrations provides increased efficiency of dissolution of deposits, for account of change in physical and chemical properties solvents, and the process is due to the cavitation process.

Efficiency and economic cleaning parameters of different APRD of different methods. The most effective methods were selected APRD purification. For conditional average composition the results are as follows:

– If it is possible to create a solvent from the specified components (10 % 2-ethylhexanol; 3 % aromatic hydrocarbons), when ultrasound is added the cost will be 17.9 thousand rubles.

– Use of solvent mixture – 35 % TSK “B”; 10 % STAAU “B”; 3 % TSK “A” with ultrasound the cost will be 22.3 thousand rubles.

In both cases, almost 100 % removal is expected APRD.

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Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Author contributions

Anvar Valeev: investigation; software; supervision. Regina Khuramshina: validation; methodology. Tatiana Dmitrieva: writing-review and editing. Vera Muratova: writing-original draft preparation

Conflict of interest

Prof. Anvar R. Valeev is an editor in chief for Liquid and Gaseous Energy Resources and was not involved in the editorial review and/or the decision to publish this article.

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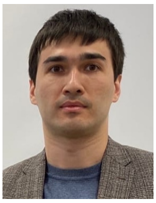

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