

DEVELOPMENT AND RESEARCH OF DEMULSIFIER COMPOSITIONS FOR DEWATERING OF HEAVY CRUDE OILS FROM UKRAINIAN FIELDS

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Abstract. Heavy crude oils from the fields of three Ukrainian regions (Lviv, Sumy, and Poltava regions) have been studied and characterized. For dewatering these oils, a number of compositions using Dissolvan demulsifiers have been developed and investigated. The synergistic effect of oxyethylated and oxypropylated block polymers was confirmed. The crude oils were diluted with heavy gas condensate to reduce viscosity and increase dewatering efficiency. For each oil under study, the optimal compositions of demulsifiers were determined. Four compositions that showed the highest demulsifying ability (90–94.5 %) when working with the studied oils were selected for further research.

Keywords: heavy oil, demulsifying ability, degree of dewatering, diluent.

1. Introduction

A characteristic feature of modern oil production is the growing share of hard-to-recover reserves in the global structure of raw materials. These resources include mainly heavy and high-viscosity oils. The reserves of these oils are estimated to be at least 1 trillion tons^{1,2}. In industrialized countries, they are viewed not as a reserve for oil production, but as the main basis for its development.

A number of heavy oil deposits have been found in Ukraine's oil and gas condensate fields. Among them are the Orkhovytske field located in the Lviv region (reserves are estimated at about 3 mln t)³, the Buhryvatske field (Sumy region, estimated reserves of about 53 mln t)⁴, and the Yablunivske field (Poltava region, 50 mln t)⁵. Since the oil reserves of these fields are quite significant, they

can be associated with an increase in oil production, which is extremely important for the current energy sector of Ukraine. Despite the long development period of the deposits, the data on the physical and chemical properties and composition of crude oil from these fields are very incomplete, contradictory, and unsystematic. The lack of information makes it difficult to attract new investors to the processing of these types of raw materials.

The main problem that arises during the production, transportation and refining of heavy, high-viscosity oils is the formation of oil-water emulsions, which in turn lead to a number of problems, including increased production costs and the use of chemicals that affect the environment⁶. To solve the problem, various demulsification methods are used: physical, biological, chemical and combined ones⁷. Chemical demulsification is considered the most widely used method of emulsion destruction due to its ease of implementation and economic feasibility. When using this method, the emulsion separation is stimulated by the addition of chemicals known as demulsifiers (DE). Despite the large number and variety of compounds used as DEs, the development of effective demulsifiers is still the subject of intensive research⁸. Several factors drive the research. First, the growing oil production of most fields is accompanied by an increase in their watering, which means an increase in the DE amount, which is economically unprofitable. To save money, it is necessary to increase the efficiency of dewatering by developing and using new DEs. Secondly, even when oil is produced from one field, its physical and chemical properties and the properties of oil-field water change over time, which also affects the efficiency of using dewatering equipment.

Due to the lack of colloidal-chemical bases for obtaining composite demulsifiers, there has been no significant increase in their efficiency for a long time⁹. For this reason, intensive work has been carried out in recent years to improve the efficiency of demulsifiers. A method of increasing the efficiency of demulsifiers by obtaining their modifications has been proposed. The essence of this

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method is to achieve such an optimization of intermolecular interactions that they acquire the ability to be in their solutions in the form of a critical emulsion. Gurbanov and Gasimzade¹⁰ note that the development of a demulsifier composition is currently considered an important aspect in the preparation for the processing of heavy oils. The use of composite compounds is, in most cases, more effective than the use of any compound in its pure form. The advantages of using a demulsifier composition are also confirmed by Kumar *et al.*¹¹.

The Dissolvan brand of demulsifiers, manufactured by Clariant (Switzerland), is designed to separate oil-water emulsions and is widely used in the oil industry. These commercially available demulsifiers have been used for study and/or comparison in several researches^{9, 10}, but their effectiveness for Ukrainian oils has not been studied. Moreover, the authors have not found any studies on the effectiveness of compositions consisting of Dissolvan demulsifiers.

This work is aimed at developing DE compositions, studying their demulsifying ability, and selecting the most effective compositions for dewatering heavy oils from the fields of three regions of Ukraine.

2. Experimental

2.1. Materials

Oil samples were taken from three fields in different regions of Ukraine: Orkhovytske (Lviv region), Bugruvatske (Sumy region), and a mixture of oils from Yablunivske field (Poltava region). The physical and chemical characteristics of the oils under study were determined according to well-known standardized methods.

To develop the compositions, we used demulsifiers of the Dissolvan series (Clariant, Switzerland), the chemical nature and purpose of which are presented below.

Dissolvan 3245 is a cross-linked oxypropylated block polymer containing diepoxy groups with RSN 9 (relative solubility number characterizing the hydrophilic-lipophilic balance or affinity of DE for a particular phase) and an active base content of 100 %. This DE can be the basis for any composition, especially when working with heavy oils. It can be used with all components of the Dissolvan series. It is recognized as an environmentally safe product.

Dissolvan 3431 is a crosslinked oxypropylated block polymer containing diepoxy groups with RSN 12 and an active base content of 100 %. The component is often used when working with heavy oils. It can be both the main component and an additional component. The best effect is achieved with Dissolvan 3245, Dissolvan 5022 and Dissolvan 5252. It is recognized as an environmentally safe product.

Dissolvan 3567 is an oxyethylated resin with RSN 23.8 and an active base content of 88 %. This product works particularly well with hard-to-break emulsions of light or medium crudes as a main component; when working with easily broken emulsions it is used as a final additive to obtain high-quality commercial oil. It can be combined with Dissolvan 3431 and Dissolvan 3879.

Dissolvan 4006 is an oxyethylated resin with RSN 22.1 and an active base content of 80 %. An overdose effect is possible when using Dissolvan 4006.

Dissolvan 4064 is an oxyethylated resin with RSN 14 and an active base content of 60 %. It is usually used as a main or additional component in combination with Dissolvan 3879 or Dissolvan 5022. It is recognized as an environmentally safe product.

Dissolvan 4411 is a crosslinked oxyethylated block polymer with RSN of about 20 and an active base content of about 100 %. It is used as the second component in the composition, usually with Dissolvan 3245, with synergism being particularly strong when De is used in medium and heavy oils.

Dissolvan 4472 is an oxyethylated / oxypropylated resin with RSN 15 and active base content of 90 %.

Dissolvan 5022 is an oxypropylated polyamine with RSN 6.5 and an active base content of about 100 %. It is used as an additional component with Dissolvan 3245 and Dissolvan 3431 for heavy oils. It is recognized as an environmentally safe product.

Dissolvan 5252 is an oxyethylated block polymer with RSN 19 and an active base content of about 100 %. It is used as an additional component in combination with Dissolvan 3245, Dissolvan 3879 and Dissolvan 3431 for heavy oils.

Dissolvan 2830 (commercial form) is a mixture of non-ionic surfactants dissolved in a mixture of methanol and xylene. It is used in industrial oil treatment processes for dewatering and desalination, as well as for the destruction of emulsions formed by different types of oils.

The physical and chemical properties of demulsifiers were determined by well-known standard methods.

To improve the efficiency of dewatering and reduce the consumption of DE, heavy oil is diluted with a diluent, for example, light low-viscosity oil (30–70 wt. % relative to the feedstock), thermal cracking gasoline, or stable gas condensate^{12, 13}. This synergetic effect contributes to the fact that lowering the viscosity reduces the obstacle to DE diffusion and increases the demulsification rate¹⁴. In the previous studies⁵ light and heavy gas condensates were examined as the diluents and the heavy gas condensate (HGC) produced by Yarivsky Gas Processing Department (Poltava region, Ukraine) was found to be the most effective for heavy oils of Ukrainian fields. HGC characteristics are presented in Table 1. The amount of HGC, 30 wt. % per crude oil, was chosen as the optimal value, taking into account the earlier results⁵.

Table 1. Characteristics of heavy gas condensate⁵

Index	Value
Initial b.p., °C	56
10 % distilled at, °C	80
20 % distilled at, °C	92
30 % distilled at, °C	102
40 % distilled at, °C	111
50 % distilled at, °C	122
60 % distilled at, °C	139
70 % distilled at, °C	160
80 % distilled at, °C	210
90 % distilled at, °C	320
End b.p., °C	350 (92 %)
Density, kg/m ³	758
Refractive index n _D ²⁰	1.4353
The mixture of oil and condensate is homogeneous	

2.2. Methods

A thermostatically controlled separating funnel equipped with a stirrer was loaded with 100 ml of the investigated emulsified oil, 0.2 ml of a 5 % solution of DE composition (100 ppm), and 30 wt. % of heavy gas condensate. The resulting mixture was heated to 60 °C and stirred for 15 s. The mixture was settled for 3–5 h. The temperature regime, DE dosage, and settling time were selected based on literature data^{9,11,15} and previous studies.⁵

The efficiency of oil emulsion dewatering was determined by the ratio of the amount of water removed from the oil after adding the demulsifier to the initial amount of water in the oil emulsion. The amount of residual water in the original emulsion and dewatered oil was determined by the Dean-Stark method and calculated by the following formula:

$$W = \frac{V \cdot \rho}{C} \cdot 100 \%,$$

where W is the water content in oil, wt. %; V is the water volume in the trap, mL; ρ is the density of the dispersed phase, kg/mL; C is the weight of the oil sample, kg.

At least three parallel determinations were made for each oil sample under study. In cases of discrepancies in the results by more than 5 %, parallel studies were repeated. The arithmetic mean of the three experiments was taken as the true value.

3. Results and discussion

Characteristics of the studied oils are presented in Table 2.

The obtained values confirm that the investigated samples belong to heavy, viscous oils with a high water content (especially the oil of the Bugruvatske field, the water content is 16 wt. %), and therefore with high emulsification.

To develop demulsifying compositions, we used DEs, the chemical nature and purpose of which are given in Section 2, and their characteristics are presented in Table 3.

Based on the manufacturer's recommendations, and taking into account the results presented in the literature,^{9,15-20} the eleven compositions of demulsifiers with different variations in both the composition components and their amount were developed (Table 4). Composition 12 is actually not a composition but a commercial form of Dissolvan 2830. Since this demulsifier is widely used in oilfields for dewatering and desalination of various types of oils, it was advisable to determine its effectiveness for dewatering the studied Ukrainian heavy oils.

Xylene mixture and isopropyl alcohol were used as epy solvent for the preparation of the compositions. This ensures the solubility of the demulsifier in both water and oil media and improves its demulsifying ability. There are different views on whether DE should be soluble in oil or water. The mechanism of emulsion destabilization during treatment with water- and oil-soluble demulsifiers is different. In the first case, the armoring shells on the water globules are destroyed first, and then the adsorption activity of natural stabilizers is inhibited. In the case of using oil-soluble DE, the adsorption ability of natural stabilizers is first inhibited, and the armor shells are destroyed later or in parallel with the first process²¹. Water and isopropanol were the solvents for composition 5.

Table 2. Characteristics of the initial crude oils

Index	Oil from the Orkhovyske field	Oil from the Bugruvatske field	Mixture of oils from Yablunivske field
Density, kg/m ³	973.8	935	965
Water content, wt. %	3.8	16	4.2
Sulfur content, wt. %	5.17	1.48	2.4
Mechanical impurities content, wt. %	0.120	0.16	0.14
Pour point, °C	+3	+5	+18
Relative viscosity at 50 °C, °RV	4.59	2.95	2.54
Fractional composition			
Initial b.p., °C	162	92	90
Distilled up to 200 °C	3	20	14.6
Distilled up to 350 °C	18	46	42.3

Table 3. Characteristics of DE

DE	Index						
	Color	Pour point, °C	Initial boiling point, °C	Flash point, °C	Density at 20 °C, kg/m ³	Dynamic viscosity, mPa·s (20 °C)	Solubility in water or crude oil
D-3245	Yellow-brown	−9	300	200	1030	≈10000	w/s (suspension)
D-3431	Yellow-brown	−10	400	200	1050	7500	w/s
D-3567	Brown	+10	190	62	1030	34000	o/s
D-4006	Yellow-brown	+5	185	63	1050	8000	mixture, phase separation
D-4064	Yellow-brown	−20	185	65	1020	500	o/s
D-4411	Yellowish	+15	—	100	1050	1500	w/s
D-4472	Brown	−6	185	63	1010	2500	o/s
D-5022	Yellow	−24	320	100	1000	4000	o/s
D-5252	Yellow	−12	320	100	1040	1300	w/s
D-2830	Yellow-brown	−50	64	11	947	120	o/s (in water suspension is formed)

Note: w/s – water-soluble; o/w – oil-soluble

Table 4. Components of the developed compositions

Components, wt. %	Composition number											
	1	2	3	4	5	6	7	8	9	10	11	12
D-3245	25	30	20	30	10	35	35	25	25	—	6	—
D-3431	—	—	20	—	—	—	—	—	—	—	—	—
D-3567	—	—	—	—	—	—	—	10	—	—	—	—
D-4006	—	—	—	—	—	—	15	—	—	—	—	—
D-4064	—	—	—	—	—	15	—	—	—	—	—	—
D-4411	15	10	—	20	30	—	—	15	15	15	40	—
D-4472	—	—	—	—	—	—	—	—	10	10	—	—
D-5022	10	—	10	—	—	—	—	—	—	25	—	—
D-5252	—	10	—	—	10	—	—	—	—	—	5	—
D-2830	—	—	—	—	—	—	—	—	—	—	—	100
Xylene mixture	40	40	40	40	—	40	40	40	40	40	39	—
Isopropyl alcohol	10	10	10	10	30	10	10	10	10	10	10	—
Water	—	—	—	—	20	—	—	—	—	—	—	—

The determination of the demulsifying efficiency of the compositions was carried out according to the method described in Section 2, the results are shown in Figs. 1–3. It should be noted that, despite the difference in the physicochemical characteristics of oils, the tendency to change the dewatering degree under the influence of all the studied DE compositions is the same for the three oils, differing only in quantitative values.

As can be seen from the data obtained, the highest dewatering degree for Orkhovytska oil is 93.1 % (Fig. 1). This value is obtained by using composition 11, which contains D-3245, D-4411, D-5252, and D-4064. The hyd-

rophobic and hydrophilic properties are attributable to the use of oxyethylated and oxypropylated block polymers with diepoxy groups. The presence of oxyethylene groups can significantly increase the hydrophilicity of DE without significantly affecting its hydrophobic properties, which increases the demulsifying ability and allows to increase the dewatering degree. The demulsification efficiency of non-ionic demulsifiers is less dependent on oil quality than the demulsifying effect of ionic demulsifiers²². Non-ionic demulsifiers provide rapid stratification of the water layer, low residual water content, minimal or no intermediate layer, and a pure aqueous phase.

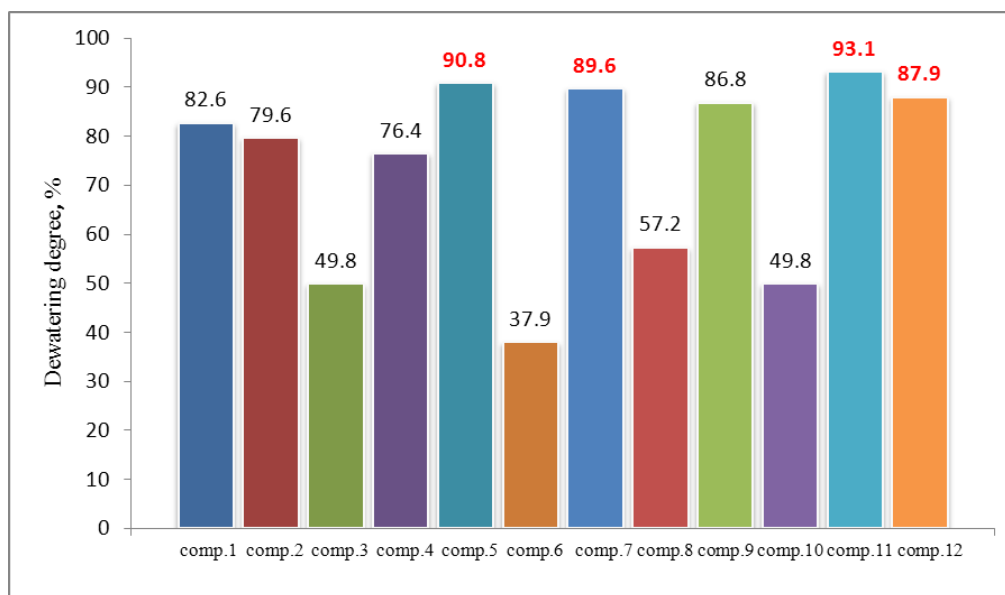


Fig. 1. Demulsifying efficiency of the developed compositions for oil from the Orkhovytske field

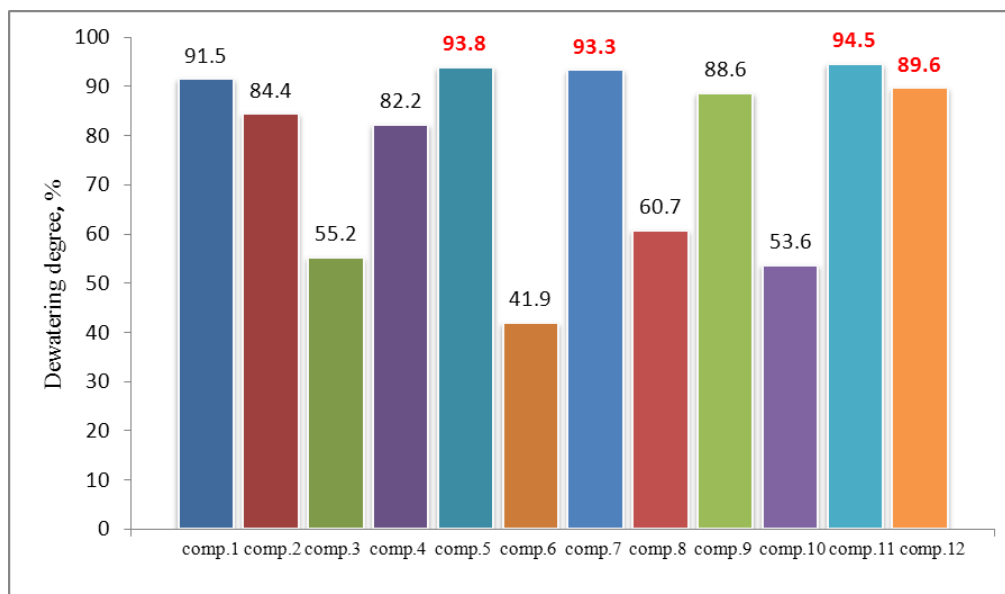


Fig. 2. Demulsifying efficiency of the developed compositions for oil of the Bugrurvatske field

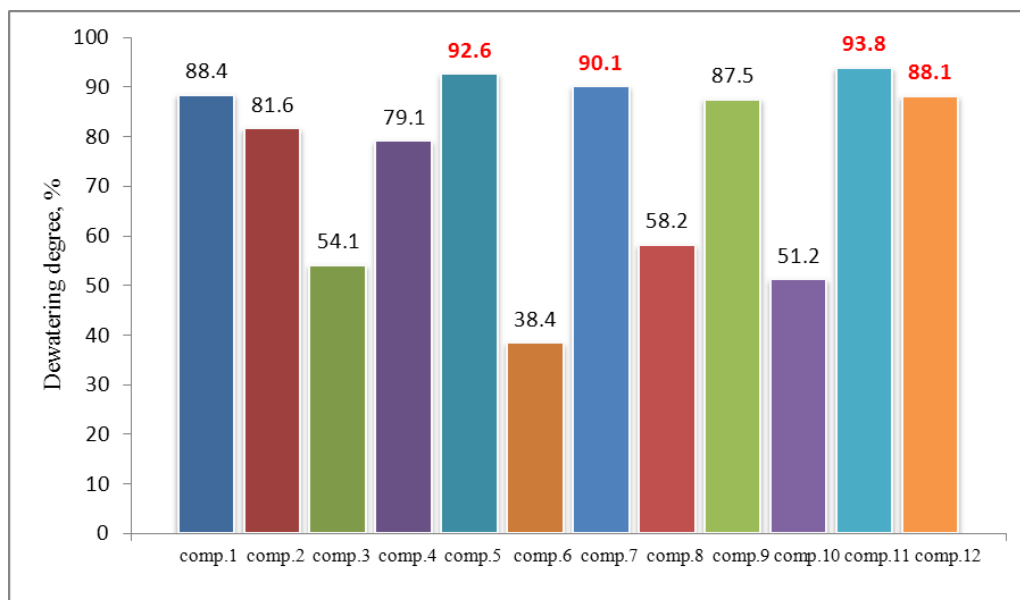


Fig. 3. Demulsifying efficiency of the developed compositions for a mixture of oils from the Yablunivske field

Composition 11 also proved to be the most effective for both Bugrurvatska oil (dewatering degree is 94.5 %, Fig. 2) and for a mixture of oils from the Yablunivske field (dewatering degree is 93.8 %, Fig. 3). Compositions 5, 7, and Dissolvan D-2830 (comp. 12) also showed quite high results. When using the above compositions, the dewatering degree of all studied oils ranges from 87.9 % to 93.8 %. The use of these compositions changes the ability of compounds to be adsorbed at the oil-water interface. At the same time, they act differently on the boundary film, *i. e.*, the placement of molecules in the adsorption layer is not the same²³.

It should be noted that for all three kinds of investigated oils, the same trend of the developed compositions is observed.

Obviously, the demulsifying activity of the synthesized compositions is related to the components ratio and structure of the components themselves. Given the nature of those DEs that showed the best results, their effectiveness was expected. When D-3245 and D-4411 are used simultaneously, their synergetic effect is revealed, which is confirmed by a rather significant dewatering degree (from 76.4 % to 82.2 %) of composition 4, which consists only of the two mentioned demulsifiers.

Compositions 1, 2, 8-10 show moderate demulsifying ability. The dewatering degree ranges from 57.2 % to 86 %. The worst results were obtained for composition 6 (the dewatering degree is 37.9 % for Orkhovytska oil, 41.9 % for Bugrurvatska oil, and 38.4 % for a mixture of oils from the Yablunivske field). The composition includes only two demulsifiers, based on an oxypropylated copolymer (D-3245), which is responsible for hydro-

phobic properties, and an oxyethylated resin (D-4064), which is a hydrophilic agent. Low values of the dewatering degree can be the result of an unsuccessfully selected composition or components ratio. Another reason may be poor compatibility of the compositions with other additives present in the oil, such as corrosion inhibitors, water clarifiers (reverse emulsifiers), anti-foam additives, *etc.*

Since the best results were obtained when using compositions 5, 7, 11, 12, it is these compositions will be used for further research.

4. Conclusions

Taking into account the country's growing demand for raw oil materials and the tendency of a further increase in oil prices, three representatives of heavy Ukrainian oils – Orkhovytska, Bugrurvatska, and a mixture of oils from the Yablunivske field – were studied in this paper. To solve the problem of demulsification of such oils, twelve compositions consisting of different representatives of the Dissolvan series were developed and studied for the first time. It was found that the highest dewatering degree (93.1 %, 94.5 %, and 93.8 % for Orkhovytska, Bugrurvatska, and a mixture of oils from the Yablunivske field, respectively) was obtained using a composition consisting of a mixture of oxyethylated and oxypropylated block polymers. Their synergistic effect was confirmed by a rather significant dewatering degree. Heavy gas condensate was used as a solvent for the studied oils to reduce their viscosity and increase the dewatering efficiency. For further research, four compositions that showed the highest

demulsifying ability when working with the studied oils were selected. The results obtained may be important for the development of a set of measures to increase production, rational transportation, and methods of oil processing.

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РОЗРОБЛЕННЯ І ДОСЛІДЖЕННЯ КОМПОЗИЦІЙ ДЕЕМУЛЬГАТОРІВ ДЛЯ ЗНЕВОДНЕННЯ ВАЖКИХ НАФТ УКРАЇНСЬКИХ РОДОВИЩ

Анотація. Досліджено й охарактеризовано важкі нафти з родовищ трьох областей України (Львівської, Сумської та Полтавської). Для зневоднення цих нафт розроблено та досліджено низку композицій із використанням деемульгаторів Dissolvan. Підтверджено синергійний ефект оксидильованих та оксипропілованих блок-кополімерів. Для зменшення в'язкості і підвищення ефективності зневоднення нафти розріджували важким газовим конденсатом. Для кожної досліджуваної нафти визначено оптимальні складі деемульгаторів. Чотири композиції, які продемонстрували найвищу деемульгувальну здатність (90–94,5 %) під час роботи з досліджуваними нафтами, відібрано для подальших досліджень.

Ключові слова: важка нафта, деемульгувальна здатність, ступінь зневоднення, розріджувач.