

The effect of oil-water interfacial tension on the stability of oil emulsions

Wpływ napięcia międzyfazowego między ropą a wodą na stabilność emulsji ropnych

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ABSTRACT: The stability of crude oil emulsions sourced from wells in two distinct fields distinguished by varying physical-chemical properties was analysed. The oil taken from the wells of Muradkhanli and Balakhani fields of State Oil Company of the Azerbaijan Republic (SOCAR) has been used as the research object. Demulsification of both oils, whose composition differs in the amount of high molecular compounds, has been investigated through the static settling method for 2, 4, 6, 8, 10 and 12 hours, and the amount of residual water in the oil has been recorded. In the same time interval, the oils have been tested by adding n-hexane and phenol to them, respectively. During the experiments, it was discovered that after the addition of n-hexane, which is a saturated hydrocarbon, the surface tension force increased in both oil samples and caused the formation of water in the form of a dispersed phase in the oil, which resulted in an increase in stability. In particular, as the concentration of n-hexane in oil increased, oil emulsions became aggregative and kinetically stable. The experiments conducted involving the addition of phenol to oils revealed that an increase in the volume of phenol in the oil led to a decrease in the surface tension force at the oil-water boundary, thereby accelerating the demulsification process. Subsequently, the research extended to investigating the emulsion stability of the oil samples by adding phenol and n-hexane in varying proportions. It was observed that an increase in the phenol ratio expedited the demulsification process, whereas a higher hexane ratio weakened it. Throughout the experimental tests, fluctuations in demulsification rates were noted upon the addition of organic substances to the oil samples. Furthermore, an increase in the concentration of high molecular compounds in the oil composition resulted in elevated surface tension, posing challenges in separating the water phase from the oil. The demulsification ability of organic substances added to crude oil samples, compositions A, B and C (25% n-hexane and 75% phenol (conditional name A), 50% n-hexane and 50% phenol (conditional name B), 75% n-hexane and 25% phenol (conditional name C)) prepared on the basis of these organic substances was examined depending on the surface tension.

Key words: emulsion, demulsification, demulsifier, surface tension, interfacial effect, degree of effectiveness, crude oil.

STRESZCZENIE: Przeanalizowano stabilność emulsji ropy naftowej pobranej z odwiertów w dwóch odrębnych złożach, charakteryzujących się zróżnicowanymi właściwościami fizykochemicznymi. Ropa pobrana z odwiertów na złożach Muradkhanli i Balakhani należących do Państwowej Kompanii Naftowej Republiki Azerbejdżanu (SOCAR) została wykorzystana jako obiekt badawczy. Deemulgacja obu rodzajów ropy, których skład różni się ilością związków o dużej masie cząsteczkowej, została zbadana za pomocą metody statycznej sedymentacji przez 2, 4, 6, 8, 10 i 12 godzin, po czym zarejestrowano ilość wody pozostającej w próbce. W tych samych przedziałach czasowych przetestowano analizowane próbki ropy poprzez dodanie do nich odpowiednio n-heksanu i fenolu. Podczas eksperymentów odkryto, że po dodaniu n-heksanu, który jest węglowodorem nasyconym, siła napięcia powierzchniowego wzrosła w obu próbkach ropy, co spowodowało powstanie wody w postaci fazy rozproszonej w ropie, co zaowocowało zwiększeniem stabilności. W szczególności, w miarę wzrostu stężenia n-heksanu w ropie, emulsje ropne stały się agregacyjne i kinetycznie stabilne. Przeprowadzone eksperymenty dotyczące dodania fenolu do ropy wykazały, że wzrost objętości fenolu w ropie spowodował zmniejszenie siły napięcia powierzchniowego na granicy ropa-woda, przyspieszając tym samym proces deemulgacji. Następnie badania zostały rozszerzone na badanie stabilności emulsji próbek ropy poprzez dodanie fenolu i n-heksanu w zróżnicowanych proporcjach. Zaobserwowano, że wzrost stosunku fenolu przyspieszył proces demulsyfikacji, podczas gdy wyższy stosunek heksanu osłabił go. W trakcie testów eksperymentalnych odnotowano wahania w tempie deemulgacji po dodaniu substancji organicznych do próbek ropy. Ponadto wzrost stężenia związków o dużej masie cząsteczkowej w składzie ropy spowodował wzrost napięcia powierzchniowego, co utrudniło oddzielanie fazy wodnej od ropy. Zbadano zdolność deemulgacyjną substancji organicznych dodanych do próbek ropy naftowej, kompozycji A, B i C (25% n-heksanu i 75% fenolu (nazwa warunkowa A), 50% n-heksanu i 50% fenolu (nazwa warunkowa B), 75% n-heksanu i 25% fenolu (nazwa warunkowa C)), przygotowanych na podstawie tych substancji organicznych, w zależności od napięcia powierzchniowego.

Słowa kluczowe: emulsja, deemulgacja, deemulgator, napięcie powierzchniowe, efekt międzyfazowy, stopień skuteczności, ropa naftowa.

Introduction

The world's major oil fields are currently in their final stages of production, leading to a notable increase in hydration levels of up to 80–90%. This surge in hydration presents challenges in production, resulting in changes in the rheological properties of oil, particularly an elevation in viscosity. Consequently, the inevitable formation of water-oil emulsions during oil production becomes more pronounced, especially with the depletion of active resources of light oils, prompting global trends towards increased production of high-viscosity heavy oils. In contrast to oils containing low and medium concentrations of dispersed systems, high-viscosity heavy oils are characterized by highly concentrated associated dispersed systems, significantly impacting the efficiency of their production, transportation, and treatment processes.

Water-oil emulsions formed through vigorous mixing of formation waters and oils are stabilized by the presence of asphaltene, resin, paraffin, naphthenic acids, clay, and clay-like solids (Lutfullin et al., 2005; Nebogina et al., 2008). A notable characteristic of stable crude oil emulsions is their interfacial viscoelasticity, reinforced by asphaltene action, which impedes droplet coalescence. However, these stable emulsions present challenges throughout production and treatment processes, leading to issues such as pipeline corrosion, corrosion of oil storage tanks, pump malfunction, and operational complications in production and treatment facilities. Additionally, transportation via pipelines is hindered due to increased oil viscosity and the presence of dispersed water in the water-oil emulsion. Emulsions containing high concentrations of resin, asphaltene, and naphthenic acids, termed natural emulsifiers, yield more stable emulsions with formation waters. As a result of these emulsifiers remaining suspended in the oil, the colloidal property of the oil increases, and during the intensive mixing of oil and water, they create a framework structure around the water phase.

The surfactants added to the oil, performing the function of demulsifier, should first free the water phase from this structure, i.e. reduce the colloidal properties by dissolving the mentioned high molecular compounds (Bakhtizin et al., 2016; Gurbanov and Gasimzade, 2022).

Diverse methodologies are employed for the demulsification of stable water-oil emulsions, encompassing ultrasonication, electro sedimentation, centrifugation, and chemical demulsification. Chemical demulsification entails the addition of demulsifiers possessing surfactant properties at low concentrations to stable water-oil emulsions. Surfactants are categorized based on their nano and ionic structures, characterized by amphoteric properties (Mukhamadiyev and Notov, 2008; Gurbanov and Gasimzade, 2023).

The surfactant chain consists of two parts, one of which is called a polar group and the other is called a non-polar group.

Numerous studies have demonstrated the profound impact of emulsion demulsification on oil rheology, including viscosity reduction and alteration (Schubert and Armbruster, 1992; Verruto and Kilpatrick, 2008). Demulsifiers play a pivotal role in destabilizing stable emulsions by modulating oil rheology, diminishing interfacial surface tension, and attenuating the electric double layer. Upon reducing interfacial strength and surface tension to a critical threshold, demulsifiers facilitate the separation of water from oil (Matiyev et al., 2016; Ali and Alqam, 2000).

The aim of the study was to investigate the impact of both saturated and unsaturated organic compounds on the stability of water-oil emulsions.

Research methodology

One widely used method for assessing the demulsification ability of reagents and their compositions is the "Cold finger test" (static settling) method. In this approach, reagents or compositions are initially dissolved in a solvent. Subsequently, they are added at various concentrations to packaged oil samples for investigation. After the addition of reagents, thorough mixing is conducted for 1 minute, followed by placement in a water bath. The separation of water is recorded at different time intervals, depending on the demulsification ability of the tested demulsifier. The demulsification index can be calculated using the following equation:

$$\text{Water separation\%} = V/V_0 \times 100$$

Where V represents the volume of water separated after the addition of the demulsifier, and V_0 represents the volume of initial water in the crude oil emulsion

All studies were conducted at a minimum temperature of 20°C to examine the alterations in surface tension at the oil-water interface induced by the added organic substances. Oil samples were extracted from the Muradkhanli and Balakhani fields, presenting varying degrees of dilution, serving as the subjects of research (Table 1).

To facilitate a comparative analysis, both oil samples were initially demulsified without reagents at a temperature of 20°C. The results obtained are provided in Table 2.

Due to the compositional differences between Muradkhanli and Balakhani oils, the amounts of water separated during the same time period varied.

Laboratory experiments continued to investigate the effects of saturated and unsaturated organic substances on the stability of water-oil emulsions. n-hexane served as the saturated

Table 1. Physical and chemical properties of crude oils**Tabela 1.** Właściwości fizyczne i chemiczne ropy naftowej

Parameters	Muradkhanli	Balakhani
Density 20°C [kg/m ³]	976.1	924.2
Viscosity 20°C [mP·san.]	2260.3	2128.5
Water content [wt.%]	45	30
Amount of chlorine salts [mg/l]	546.6	374.4
Mechanical mixtures content [wt.%]	6.51	3.79
Resin content [wt.%]	15.5–18.9	6.2–8.10
Asphaltene content [wt.%]	4.7–5.9	0.1–0.16
Paraffin content [wt.%]	4.1–6.2	0.3–0.45
Freezing temperature [°C]	+12	+5

Table 2. Demulsification of oil samples at a temperature of 20°C (without reagent)**Tabela 2.** Deemulgacja próbek oleju w temperaturze 20°C (bez odczynnika)

Time [hour]	The residual water content in the oil after demulsification [%]	
	Muradkhanli	Balakhani
2	35	22
4	31	19
6	27	15
8	24	12
10	20	8
12	15	6

organic compound, while phenol served as the unsaturated organic compound. Reagent concentrations of 0.1%, 0.2%, 0.3%, 0.4%, and 0.5% were utilized in the research process.

Tables 3–7 show the results of the effect of different percentages of hexane on the stability of water-oil emulsions of Muradkhanli and Balakhani oil samples.

Table 3. Effect of 0.1% n-hexane on the demulsification process**Tabela 3.** Wpływ 0,1% n-heksanu na proces deemulgacji

Time [hour]	The residual water content in the oil after demulsification [%]		
	percentage of n-hexane	Muradkhanli + n-hexane	Balakhani + n-hexane
2	0.1	35	22
4		31	19
6		27	15
8		25	12
10		22	9
12		16	7

Table 8 and Figure 1 summarizes the results of experiments conducted with n-hexane.

Table 4. Effect of 0.2% n-hexane on the demulsification process**Tabela 4.** Wpływ 0,2% n-heksanu na proces deemulgacji

Time [hour]	The residual water content in the oil after demulsification [%]		
	percentage of n-hexane	Muradkhanli + n-hexane	Balakhani + n-hexane
2	0.2	35	22
4		32	19
6		28	15
8		23	13
10		22	9
12		18	8

Table 5. Effect of 0.3% n-hexane on the demulsification process**Tabela 5.** Wpływ 0,3% n-heksanu na proces deemulgacji

Time [hour]	The residual water content in the oil after demulsification [%]		
	percentage of n-hexane	Muradkhanli + n-hexane	Balakhani + n-hexane
2	0.3	36	22
4		32	19
6		28	16
8		26	14
10		22	10
12		19	9

Table 6. Effect of 0.4% n-hexane on the demulsification process**Tabela 6.** Wpływ 0,4% n-heksanu na proces deemulgacji

Time [hour]	The residual water content in the oil after demulsification [%]		
	percentage of n-hexane	Muradkhanli + n-hexane	Balakhani + n-hexane
2	0.4	36	23
4		33	20
6		30	17
8		26	15
10		23	11
12		21	10

As can be seen from the tables, as the percentage of n-hexane increases in both oil samples, the amount of released water decreases. This observation can be explained by the fact that n-hexane, a saturated organic substance, increases the surface tension of the system and causes an increase in the kinetic and aggregative stability of water-oil emulsions. Thus, one of the most important theories explaining the different effects of added chemical reagents on different oil samples is surface tension, which was reflected in the research work. Although having similar properties to resin, asphaltenes, which are high molecular components of oil, differ in structure. They

Table 7. Effect of 0.5% n-hexane on the demulsification process

Tabela 7. Wpływ 0,5% n-heksanu na proces deemulgacji

Time [hour]	The residual water content in the oil after demulsification [%]		
	percentage of n-hexane	Muradkhanli + n-hexane	Balakhani + n-hexane
2	0.5	39	24
4		36	20
6		35	18
8		33	17
10		32	14
12		31	12

Table 8. Demulsification of oils with added n-hexane for 12 hours

Tabela 8. Deemulgowanie olejów dodatkiem n-heksanu przez 12 godzin

Percentage of n-hexane	Time [hour]	The residual water content in the oil after demulsification [%]	
		Muradkhanli + n-hexane	Balakhani + n-hexane
0	12	15	6
0.1		16	7
0.2		18	8
0.3		19	9
0.4		21	10
0.5		31	12

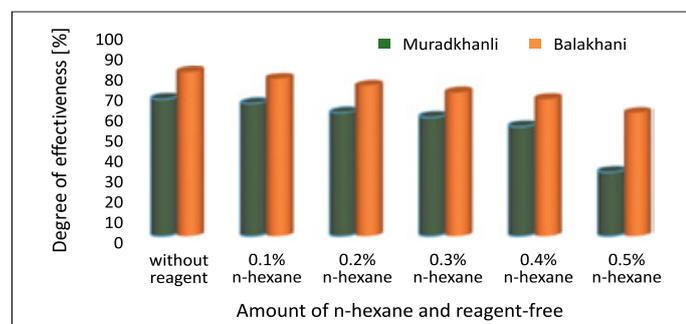


Figure 1. The effect of n-hexane on the demulsification process

Rysunek 1. Wpływ n-heksanu na proces deemulgacji

also constitute the most important component of the structures formed in the oil, as they combine with the resins making the structure more complex and stable. The accumulation of such continuous structures around the water-dispersed phases inside the oil increases the surface tension between water and oil, leading to an increased emulsion stability. After adding n-hexane to both oil samples, an increase in the surface tension at the oil-water interface was observed. However, since the amount of asphaltene high-molecular compounds predominated in Muradkhanli oil, the surface tension was higher in this oil.

These observations were also confirmed by the results of laboratory experiments conducted on the effect of n-hexane on the surface tension of both oil samples given in Table 9.

Table 9. Effect of n-hexane on the surface tension of crude oils

Tabela 9. Wpływ n-heksanu na napięcie powierzchniowe ropy naftowej

Percentage of n-hexane	Time [hour]	Surface tension [N/m ²]	
		Muradkhanli + n-hexane	Balakhani + n-hexane
0	12	31.7	22.5
0.1		32.5	22.9
0.2		34.1	23.6
0.3		36.8	23.9
0.4		39.9	26.3
0.5		41.9	27.9

Research efforts were extended to investigate the effect of varying percentages of phenol, an unsaturated compound, on the stability of water-oil emulsions in Muradkhanli and Balakhani oils. The outcomes of the laboratory experiments are presented in Tables 10–14.

Through investigations conducted at various time intervals and concentration levels, it was observed that the addition of

Table 10. Effect of 0.1% phenol on the demulsification process

Tabela 10. Wpływ 0,1% fenolu na proces deemulgacji

Time [hour]	Percentage of phenol	The residual water content in the oil after demulsification [%]	
		Muradkhanli + phenol	Balakhani + phenol
2	0.1	35	20
4		30	16
6		26	13
8		23	10
10		19	7
12		14	5

Table 11. Effect of 0.2% phenol on the demulsification process

Tabela 11. Wpływ 0,2% fenolu na proces deemulgacji

Time [hour]	Percentage of phenol	The residual water content in the oil after demulsification [%]	
		Muradkhanli + phenol	Balakhani + phenol
2	0.2	34	19
4		28	15
6		25	10
8		22	8
10		18	6
12		12	4

Table 12. Effect of 0.3% phenol on the demulsification process**Tabela 12.** Wpływ 0,3% fenolu na proces deemulgacji

Time [hour]	Percentage of phenol	The residual water content in the oil after demulsification [%]	
		Muradkhanli + phenol	Balakhani + phenol
2	0.3	33	16
4		26	14
6		23	9
8		20	6
10		15	5
12		11	3

Table 13. Effect of 0.4% phenol on the demulsification process**Tabela 13.** Wpływ 0,4% fenolu na proces deemulgacji

Time [hour]	Percentage of phenol	The residual water content in the oil after demulsification [%]	
		Muradkhanli + phenol	Balakhani + phenol
2	0.4	32	14
4		24	11
6		20	7
8		18	5
10		11	3
12		8	2

Table 14. Effect of 0.5% phenol on the demulsification process**Tabela 14.** Wpływ 0,5% fenolu na proces deemulgacji

Time [hour]	Percentage of phenol	The residual water content in the oil after demulsification [%]	
		Muradkhanli + phenol	Balakhani + phenol
2	0.5	31	12.0
4		22	9.0
6		19	5.0
8		15	3.0
10		10	2.0
12		6	0.8

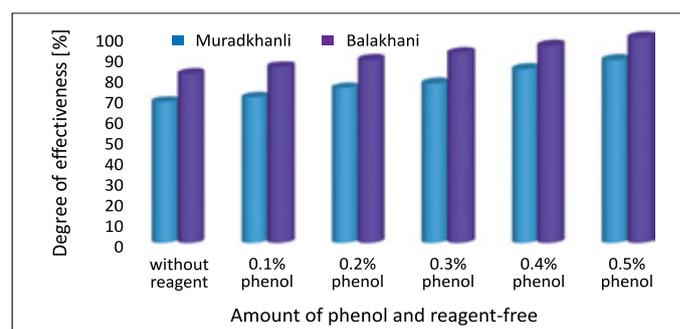
phenol to oil samples accelerates the demulsification process, unlike n-hexane. Consequently, there is an increase in the quantity of water-dispersed phases separated from the oil medium. Experimental data suggests that higher concentrations of phenol, along with increased time, lead to a swifter demulsification process. The rise in released water in the presence of phenol can be attributed to the destabilizing effect of unsaturated organic compounds on water-oil emulsions. The differences in physical-chemical properties between Muradkhanli and Balakhani oils, particularly in the amount of high-molecular compounds, were evident. This diversity was observed not

only when adding n-hexane but also when introducing phenol.

The most effective results were observed at the highest concentration of phenol and the longest duration. Table 15 and Figure 2 present the results obtained from both crude oil samples used.

Table 15. Demulsification of oils with added phenol for 12 hours**Tabela 15.** Deemulgowanie rop dodatkiem fenolu przez 12 godzin

Time [hour]	Percentage of phenol	The residual water content in the oil after demulsification [%]	
		Muradkhanli + phenol	Balakhani + phenol
0	0.3	16	6.0
0.1		14	5.0
0.2		12	4.0
0.3		11	3.0
0.4		8	2.0
0.5		6	0.8

**Figure 2.** Effect of phenol on the demulsification process**Rysunek 2.** Wpływ fenolu na proces deemulgacji

The addition of n-hexane to oils leads to changes in the structural configuration of high molecular components, resulting in alterations in surface tension at the oil-water phase boundary. Similarly, upon the addition of phenol, there are observable changes. However, it is noteworthy that based on the study results, after the addition of phenol, the separation of high-molecular components from the water phase boundary reduces surface tension at the phase boundary, enhances the coalescence of water droplets to form larger droplets, and accelerates the demulsification process (Table 16). Acceleration of the coalescence process was observed when phenol was added to oil, especially at a high concentration ratio.

After conducting a comparative analysis of the results obtained from experimental tests, wherein both organic substances were added to the oil in different proportions (25% n-hexane and 75% phenol (conditional name A), 50% n-hexane and 50% phenol (conditional name B), 75% n-hexane and 25% phenol (conditional name C)), the findings were examined. Based

Table 16. The effect of phenol on the surface tension of crude oils
Tabela 16. Wpływ fenolu na napięcie powierzchniowe ropy naftowej

Percentage of phenol	Time [hour]	Surface tension [N/m ²]	
		Muradkhanli + phenol	Balakhani + phenol
0	12	31.7	22.5
0.1		29.8	20.4
0.2		28.1	18.8
0.3		23.9	15.3
0.4		20.8	10.2
0.5		16.2	3.50

on the results obtained, it can be concluded that the effects of composition B were additive, while compositions A and C exhibited antagonistic properties (Tables 17–19).

The changes in surface tension during the application of the prepared compositions to both oil samples were also examined. The study results indicated that composition A, containing a significant quantity of phenol, exhibited a notable reduction in surface tension, demonstrating higher efficacy compared to the other compositions. Additionally, in compositions B

Table 17. The effect of composition A on the demulsification of crude oils

Tabela 17. Wpływ kompozycji A na deemulgację ropy naftowej

A composition	Time [hour]	The residual water content in the oil after demulsification [%]	
		Muradkhanli	Balakhani
0	0	45	30
0	12	15	6
0.1		15	6
0.2		13	5
0.3		12	4
0.4		10	3
0.5		8	1

Table 18. The effect of composition B on the demulsification of crude oils

Tabela 18. Wpływ kompozycji B na deemulgację ropy naftowej

B composition	Time [hour]	The residual water content in the oil after demulsification [%]	
		Muradkhanli	Balakhani
0	0	45	30
0	12	15	6
0.1		16	6
0.2		15	5
0.3		14	5
0.4		12	4
0.5		11	3

Table 19. The effect of composition C on the demulsification of crude oils

Tabela 19. Wpływ kompozycji C na deemulgację ropy naftowej

C composition	Time [hour]	The residual water content in the oil after demulsification [%]	
		Muradkhanli	Balakhani
0	0	45	30
0	12	15	6
0.1		16	6
0.2		17	7
0.3		18	8
0.4		21	9
0.5		24	10

and C, the reduction in surface tension of Balakhani oil was more pronounced than that of Muradkhanli oil (Tables 20–22).

Based on the conducted studies, summarizing the aforementioned findings, it can be concluded that composition A demonstrated superior results compared to the other two compositions in terms of reducing surface tension, and it exhibited higher demulsification ability. However, when compared to the direct effect of phenol, composition A yielded lower results. The investigation of two different oil samples revealed that during the addition of n-hexane at the highest concentration and for the

Table 20. The effect of composition A on the surface tension of crude oils

Tabela 20. Wpływ kompozycji A na napięcie powierzchniowe ropy naftowej

A composition	Surface tension [N/m ²]	
	Muradkhanli	Balakhani
0	31.7	22.5
0.1	30.0	24.4
0.2	28.7	25.6
0.3	27.5	26.8
0.4	25.3	27.5
0.5	23.2	10.9

Table 21. The effect of composition B on the surface tension of crude oils

Tabela 21. Wpływ kompozycji B na napięcie powierzchniowe ropy naftowej

B composition	Surface tension [N/m ²]	
	Muradkhanli	Balakhani
0	31.7	22.5
0.1	30.4	24.4
0.2	29.7	25.6
0.3	28.3	26.8
0.4	27.4	27.5
0.5	26.6	12.2

Table 22. The effect of composition C on the surface tension of crude oils

Tabela 22. Wpływ kompozycji C na napięcie powierzchniowe ropy naftowej

C composition	Surface tension [N/m ²]	
	Muradkhanli	Balakhani
0	31.7	22.5
0.1	30.8	24.4
0.2	29.9	25.6
0.3	29.5	26.8
0.4	29.1	27.5
0.5	28.3	14.2

longest duration, no water separation occurred in Muradkhanli oil, whereas demulsification was observed in Balakhani oil. Demulsification occurred simultaneously in both Muradkhanli and Balakhani oils at the same concentration of phenol. Due to the effects of the compositions, varying amounts of water were separated from both oil samples (Figure 3).

Thus, the results of numerous laboratory experiments conducted provide a basis for the expediency of using unsaturated organic compounds as an effective reagent or composition for the demulsification of stable water-oil emulsions.

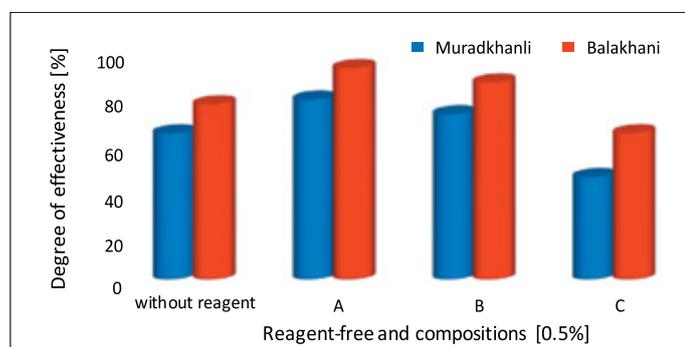


Figure 3. Effects of compositions on the demulsification process

Rysunek 3. Wpływ kompozycji na proces deemulgacji

Conclusion

1. The effects of n-hexane as a saturated compound and phenol as an unsaturated compound on the durability of the oil-water emulsion of Muradkhanli and Balakhani field oils were studied for the first time under laboratory conditions.

The process was carried out for twelve hours, using 0.1%, 0.2%, 0.3%, 0.4%, and 0.5% amounts of reagents.

2. It was determined that as the percentage of n-hexane increases in both oil samples, the amount of separated water decreases. These observations can be attributed to the fact that n-hexane enhances the surface tension of the system, thereby increasing the kinetic and aggregative stability of water-oil emulsions.
3. Unlike n-hexane, phenol reduces the surface tension of the system, reduces the stability of the water-oil emulsion and causes the separation of the water and oil phases.
4. It was determined that the synergistic effect does not occur in compositions with different molar ratios of n-hexane and phenol; instead, both components retain their individual effects.

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