Available online at www.worldscientificnews.com

World Scientific News

An International Scientific Journal

WSN 162 (2021) 148-160 EISSN 2392-2192

Investigation of effect of additives on rheological properties and asphalt-rezin-paraffin compounds of crude oil

V. H. Nurullayev[1,a], B. T. Usubaliyev[2,b], G. R. Gurbanov[3,c], Z. A. Abdullayeva[3,d]

[1]State Oil Company of the Azerbaijan Republic,
Khodzhala Ave., Baku, 1025, Republic of Azerbaijan

[2]Scientific Research Institute, Technical Sciences, Azerbaijan State Oil and Industry University,
Baku, 1010, Republic of Azerbaijan

[3]Azerbaijan State Oil and Industry University, Baku, 1010, Republic of Azerbaijan

[a-d]E-mail address: veliehet1973@mail.ru , ubeybala@gmail.com , ebikib@mail.ru , abddali@yandex.ru

ABSTRACT

Rheological properties of asphaltene-resin-paraffin sediments (ARPS) are shown depending on content of resinous components in crude oil. The results of experimental studies on low-paraffinic, high-paraffinic oil show that the presence of asphaltenes and resins in the oil dispersion system can lead to depressant effects. According to the obtained results, it was found that asphalteno-resinous components are natural depressants that reduce the temperature of wax crystallization in oil, and their depressant properties depend on the type of oil. The following types of additives were studied as additives in the work - Difron 4201 and BAF-1. It also follows that the greatest decrease in oil viscosity is observed in the presence of BAF-1 additive.

***Keywords*:** Bulla, Surakhani, Balakhany, Difron 4201, Baf-1, pour point, paraffin, resin

(Received 25 September 2021; Accepted 15 October 2021; Date of Publication 16 October 2021)

1. INTRODUCTION

Oil is a natural multicomponent organic liquid. It is based on a mixture of naphthenic, aromatic and paraffinic hydrocarbons. Oil is a mixture of about 1000 individual substances, of which most are liquid hydrocarbons and heteroatomic organic compounds, mainly sulfurous, nitrous and oxygen, organometallic compounds, the remaining components are dissolved hydrocarbon gases, water, mineral salts, solutions of organic acid salts, etc., mechanical impurities.

The content of all these components can vary widely and depends on the oil field. The oil consists of about 425 hydrocarbon compounds. Any oil under natural conditions consists of a mixture of methane, naphthenic and aromatic hydrocarbons.

By hydrocarbon composition, all natural oils are divided into: methane-naphthenic, naphthenic-methane, aromatic-naphthenic, naphthenic-aromatic, aromatic-methane, methane-aromatic, as well as methane-aromatic-naphthenic [1]. In other words, the oil can consist of a plurality of hydrocarbon and non-hydrocarbon compounds with different boiling points, so one of the most commonly used properties of the oil is its fractional composition, characterized by the content of individual fractions of oil boiling out at different temperature ranges. With a huge variety of components up to 300 °C, it usually boils no more than 50% of the weight of oil. The residue consists of high molecular weight hydrocarbons, resins, asphaltenes, minerals, the more, the heavier the oil. At the moment, there is a stable growth in the production of hard-to-recover oil reserves, which are characterized by an increased content of asphaltenes [2]. The occurrence of asphaltenes deposits on various surfaces and at different stages of the technological process complicates its flow.

Formation of asphaltene deposits can be formed both in formation and in tubing, pipelines and in reservoirs. Asphaltenes are very negative on the technological and economic side. In the end, there is a need to carry out work to prevent and remove deposits, thereby increasing the costs of the production process, as well as the transportation of oil. Knowledge about the influence of heavy organic compounds in the operating conditions of oil production and transportation processes will enable to model asphaltenes formation processes with high accuracy and prevent deposition of high molecular weight organic compounds. In order to study asphaltenes, their property of separation from the oil system is used when low molecular weight n-alkanes are added [3-7].

Knowledge about the influence of heavy organic compounds in the operating conditions of oil production and transportation processes will enable to model asphaltenes formation processes with high accuracy and prevent deposition of high molecular weight organic compounds. In order to study asphaltenes, their separation property from the oil system is used when low molecular weight n-alkanes are added. Asphaltenes are isolated by adding solvents of n-alkane, for example n-heptane or n-propane. The remaining oil components, called maltenes, are separated by passing them through a chromatographic column with an adsorbent. Each of the components is isolated by washing it with various solvents. Saturated hydrocarbons and solid paraffins with n-alkanes. Resins form a special class that differs in solubility characteristics, and in this they are similar to asphaltenes: resins are a non-volatile polar component of oil, soluble in n-alkanes, but insoluble in liquid propane. From the point of view of colloidal chemistry, it is important that the temperature interval of the liquid state of the oil components is significantly different. In other words, the melting point on the one hand and the boiling point on the other may vary greatly.

Currently, the issue of improving the efficiency of viscous and highly viscous oils is being raised in the work of many scientists. For efficient pumping of viscous oil, several main methods are used: oil heating; using hydrocarbon diluents; hydraulic transport; construction of lupings, inserts; increase in the number of pump stations (PS) on the pipeline; increase of PS productivity; adding additives [8-11].

The most common method of pipeline transport of viscous oil is pumping of preheated oil. In this method, preheated oil enters the main oil pipeline, then, after a certain distance, thermal stations where oil is heated are installed on the route. The initial data is a pipeline, the route of which runs in conditions of heavily watered, marshy terrain and lens permafrost throughout the territory. In addition to the Arctic climate, the construction area is characterized by unique geological and hydrological conditions. Increase capacity by heating only is not rational to avoid environmental impact [12]. In this regard, the authors proposed the use of additives. An analysis was made of the use of various additives both in domestic and foreign practice. Applied additives by type of commercial form are divided into two groups: dispersion and gel [13].

The first kind of additives is a suspension of the polymer in various liquids that do not react with it. Alcohols, glycols and esters thereof are usually used as such liquids. The active polymer content of these additives is up to 25%. Such anti-turbulent additives include Necadd 447, Liquid Power, FLO XL, FLO MXA, M-FLOWTREAT, Alfakauchuk-C, Koltek PTN 3170, Turbulent Master 8010. Gel additives (CDR 102, FLO, Necadd 547, Viol, X-PAND, HIPR) are made in the form of a polymer solution in any hydrocarbon liquid (gasoline, kerosene, etc.).

The active polymer in such additives contains about 10%. The use of gel additives is especially advantageous for pipelines where the processes of their dissolution speed are important. These can be short pipelines of offshore fields, loading and shipping terminals. Recently, work has been underway on the development and implementation of suspension-emulsion additives designed for use in oil and gas production conditions on field pipelines. Due to the variety of physical and chemical properties of pumped oil and oil products through pipeline systems in each case, the use of chemical reagents is an individual feature of the pipeline. Rheological properties of oil are the main initial data for solution of design and operational tasks during development of the field and further pipeline transport [14].

2. MATERIALS AND METHODS

Taking into account the parameters of the tested pipeline, an anti-turbulent additive was chosen, which reduces the coefficient of hydraulic resistance and increases the throughput of the pipeline.

One of the main tasks of rheological research is to determine the relationship between the forces acting on the studied environment and the deformations caused by these forces.

Liquids called Newtonian are described by the following equation:

$$\tau = \eta \cdot \gamma$$

where: τ - damping voltage damping voltage, η - dynamic viscosity factor - proportionality factor, Pa s; γ - velocity gradient.

The hypothesis of the linear relationship between tangent stresses and the velocity gradient proposed by Newton is not true for all liquids. Fluids whose rheological behavior differs from the equation are called non-Newtonian. Non-Newtonian liquids are usually divided into three groups [15]:

1) nonlinear viscous liquids (tangent stress is a nonlinear function of shear rate);
2) fluids with transient rheological characteristics (functional relationship between tangent stress and shear rate depends on the time or history of the process);
3) viscoelastic fluids (exhibit elastic shape recovery after stress relief).

The test oil sample can be referred to as nonlinear viscous liquids. An overview classification of rheological models of nonlinear-viscous media proposed by various researchers was presented in the work [16].

It is known that the bulk of the oil produced is heavy in composition, containing mainly paraffinic hydrocarbons and/or resinous asphaltenes (RA). Some oil is characterized by a relatively high paraffin content, they are characterized by the highest paraffin content (25%). Studies have shown that the more paraffin in oil, the less resins and asphaltenes in its composition. The following pattern is also observed that the greater the geological age of oil, the greater the paraffin in its composition. Highly araffinic oils are also characterized by the lowest content of sulfur, vanadium and nickel [17]. The development and use of high-paraffin deposits requires the application of unconventional methods of oil extraction from the formation, its collection, preparation and pipeline transport. Pipeline transport of such oil causes serious problems, for which it becomes necessary to regulate their rheological properties by various methods of influence.

The production of highly viscous paraffin oils leads to the colmatation of the bottomhole zone of the formation, the formation of asphalt-resin-paraffin deposits (AFS) and plugs in the process chain of the formation - well - pipeline, and, therefore, to a decrease in well yields, wear and breakdown of downhole equipment and, as a result, to its unscheduled replacement with the need for underground well repair. Paraffin deposition is complex and very expensive, and as a result, a pressing problem of Kazakhstan's oil industry, which has been widely studied and covered in recent decades [18]. One of the current current control methods is the use of depressor additives and inhibitors on APRS [19].

3. RESULTS AND DISCUSSIONS

The purpose of this article is to study the effect of additives on the rheological properties of the oil field of Azerbaijan Bulla, Balakhany heavy and Surakhany. Additives of Difron 4201 and BAF-1 grades are used as additives [4]. Separation of paraffins, asphaltenes and resins from oil was carried out according to the procedures GOST 11851-85 and GOST 11858-66. The pour point, or the pour point, was determined in the S.D.M. - 530 (Germany), RK 1530-2006 supplied with three cameras for maintenance of temperatures of 0,-17 and 34 ^{o}C wasps according to ST. Effective viscosity and shear stress were measured on a rotary rheometer REOTEST-2 using a measuring system standard GOST 26581-85 [20]. The main physicochemical characteristics of oil, such as viscosity, solidification temperature, paraffin content, etc. from Bulla, Balakhany heavy and Surakhani deposits are studied in the work, which are given in table 1.

Table 1. The main physical and chemical characteristics of oil from the Bulla, Balakhany heavy and Surakhany fields.

Indicators	Bulla	Balakhany heavy	Surakhani
Density, at 20 °C kq/m^3	842,1	921,7	859,3
Amount of water, mass %	0,76	0,21	0,79
Chloride Salt Content, mq/l	138,6	38,4	139,5
Amount of mechanical impurities, mass %	0,007	0,009	0,008
Resins content, mass, %	9,6	16,2	14,8
Asphaltenes content, mass, %	0,22	2,8	2,94
Paraffins content, mass, %	13,1	0,31	2,73
Puor point temperature, °C	+12	-33	-21
Viscosity at 20°C mm^2/sec	23,2	160,3	20,2
Fractional composition:			
Overpoint,temperature, °C	71,5	97,5	78
To 200 °C, by volume %	24,5	16	21
To 300 °C, by volume %	44	32,5	40,5
To 350 °C, by volume %	63	54	61,5
Final boiling point, °C	341,5	302	328

Table 1 shows that oil from Bulla field belongs to high-paraffinic oil, compared to oil from Balakhana fields heavy and Surakhany. It should also be noted that oil from Surakhany fields has a positive freezing point, which also indicates the presence of a large number of paraffins. It is known that when pumping highly paraffinic oils, paraffin deposition is observed on the inner walls of the pipeline. In order to prevent this phenomenon, a hot pumping method is used when transporting high-paraffin oils. For this purpose, oil is further heated every 25-150 km of pipeline length. Heating oil solves the problem of pumping high-paraffinic oil, but this complicates and increases the cost of its production, transportation and processing. In terms of the dissolution efficiency of ARPS, most often these compositions differ slightly from natural solvents, and in some cases are even less effective. Natural ARPS removers associated with oil production, such as gas condensate, gas gasoline, a mixture of liquefied petroleum gases, light oil, have become the most widespread. The undeniable advantage of such solvents is their availability. They are usually produced or obtained in oil areas, have a low cost, do not affect further oil processing processes. To increase the effectiveness of ARPS removers, compositions comprising a hydrocarbon solvent and various surfactants are often provided, the addition of which increases the surface activity of the solvents and the dispersion effect of ARPS to the compositions up to 3 wt%. Such surfactants include composite additives, polyalkylbenzene resin (PABR), heavy pyrolysis resin (PR), catalytic cracking thermogasoil (TG), nitrogen-containing block copolymer (BC) of ethylene oxide and propylene oxide with molecular weight of 5000 and phenol-formaldehyde resins [21]. As solvents prepared on an aqueous basis, reagents manufactured by Neftepromchem LLC are used: SNPX-7p-1 - a mixture of paraffin hydrocarbons of normal and isostroy, aromatic hydrocarbons; SNPX-7p-2 is a hydrocarbon composition consisting of light pyrolysis resin and hexane fraction thereof; SNPX-7200 and SNPX-7400 are complex mixtures of oxyalkylated surfactants and aromatic hydrocarbons, ash soda solution, alkali solution, trisodium phosphate, a composite mixture consisting of surfactants, phosphates, carbonates, sulfates and silicates. Surfactants based on phenol formaldehyde resins are also used. Solvents and solutions of surfactant compositions are more effective at elevated temperatures. In practice, chemical methods for removing paraffin deposits are often used in combination with thermal and mechanical methods.

Paraffin in crude oil   Resin in crude oil

Asphaltenes in crude oil

Figure 1. Appearance of resinous asphaltene paraffin compounds extracted from oil.

At the same time, the greatest technological and economic effect is achieved as a result of significant acceleration of the process and completeness of ARPS removal [22]. There are no particular restrictions on the use of ARPS removal methods. However, care must be taken when using chemical methods in combination with thermal and mechanical methods. Intensive removal of such deposits from highly paraffined objects can cause the formation of paraffin plugs in the pipeline. It is advisable to process such objects in two to three stages: first with a stripper at ordinary temperature, and then, to more fully remove the resin-paraffin deposits, at an elevated temperature (60-70 °C). Light hydrocarbon solvents are generally used without heating. It was found that the addition of the reagent increases the solubility and dispersibility of ARPS to 10% of the total weight of the precipitate due to the development of the "proppant" effect by the surfactant. The optimal weight content of solvent at which equilibrium solubility and dispersibility increase and stabilize is 20 % of the total weight of ARPS. Currently, one of the most effective ways to improve the physicochemical properties of high-viscosity and high-paraffin oils is the use of depressant additives that improve their low-temperature properties.

To date, there is no universal depressor additive for all types of oil. Studies have shown that the most optimal are synthetic and natural depressants. As depressant additives, the following types of additives were studied in the work - Difron 4201 and BAF-1. The results of the study of the effect of the above additives on the properties of the most highly paraffinic oil from the Bulla field are shown in Table 2.

Among the ARPS inhibitors studied, Difron 420:BAF-1 (1:1 ratio) was the most effective composition, which, within the concentration range of 200-800 kg/tone, reduces the solidification temperature. The optimum concentration of the Difron 420:BAF-1 composition is 600 kg/tone. It can be seen that a further increase in concentration practically does not affect the temperature of solidification and decrease in oil viscosity. Thus, a BAF-1 additive was chosen for further research. It also appears from Table 2 that the greatest decrease in oil viscosity occurs in the presence of the additive BAF-1.

The process of extracting ARPS from crude oil was investigated by a unit simulating the process of depositing ARPS on a main pipeline.

Table 2. Change of dynamic viscosity of oil produced from oil fields of Azerbaijan at different temperatures

Speed stages (corresponds to velocity gradient c^{-1})	Dynamic viscosity of oils after demulsification, cPz.					
	20 °C		10 °C		5 °C	
	Bulla	Balakhany heavy	Bulla	Balakhany heavy	Bulla	Balakhany heavy
1a (0,3333)	594,6	681,3	887,3	996,7	1049,2	1214,3
2a (0,6)	378,4	421,9	592,7	653,2	671,8	792,7
3a (1,0)	231,8	363,5	486,9	534,6	518,5	613,9
4a (1,8)	119,3	214,8	227,4	297,8	291,7	417,2
5a (3,0)	72,9	112,3	138,6	189,3	178,4	209,6
6a (5,4)	43,5	69,7	81,3	111,8	109,6	127,4
7a (9,0)	26,3	43,9	49,5	84,5	58,3	96,3

8a (16,2)	12,9	23,8	22,7	41,2	32,1	58,9
9a (27,0)	6,3	10,3	11,5	19,9	18,3	27,4
10a (48,6)	4,1	6,1	7,8	12,4	11,7	18,2
11a (81,0)	2,4	4,3	4,9	7,5	7,3	11,4
12a (145,8)	1,8	2,8	3,7	4,9	5,8	8,1

Table 3. Change of dynamic viscosity of oil produced from oil fields of Azerbaijan at different temperatures

Speed stages (corresponds to velocity gradient c^{-1})	Dynamic viscosity after addition of Baf-1 reagent to oil (0.8 kg/t) cPz.					
	20 °C		10 °C		5 °C	
	Bulla	Balakhany heavy	Bulla	Balakhany heavy	Bulla	Balakhany heavy
1a (0,3333)	581,3	596,8	972,6	748,6	1328,4	1182,36
2a (0,6)	369,7	387,6	710,3	507,4	843,5	765,9
3a (1,0)	223,1	314,9	574,7	421,9	683,2	588,2
4a (1,8)	110,8	195,3	304,8	204,1	401,7	387,9
5a (3,0)	78,3	94,8	191,3	112,6	238,6	187,4
6a (5,4)	39,1	57,4	134,7	85,9	185,2	102,5
7a (9,0)	22,6	38,4	71,3	72,7	94,1	81,7
8a (16,2)	11,2	17,4	40,2	33,7	48,2	50,2
9a (27,0)	5,9	7,9	23,1	13,5	25,4	21,3
10a (48,6)	3,6	4,6	10,2	12,4	14,9	12,8
11a (81,0)	2,3	3,5	6,8	5,1	9,1	9,7
12a (145,8)	1,6	2,3	5,1	3,7	6,9	7,4

At the same time, the consistency of the released ARPS becomes denser, which makes their mechanical removal difficult. Moreover, in the composition of such AFS there is an increase in asphalt-resin substances. By their physical state, ARPS samples become looser, fluid at room temperature, and easily removed mechanically (Figure 1). As a result of experimental studies, the main rheological dependencies were obtained, which show the presence of non-Newtonian properties in oil transported without an additive (Tables 2-5) and in oil samples transported with an additive of 800 g/t (in the temperature range of 5-20 °C.

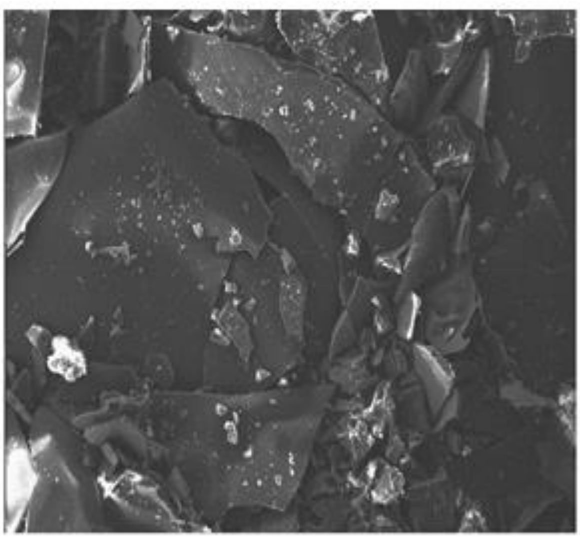
Table 4. Change of dynamic viscosity of oil produced from oil fields of Azerbaijan at different temperatures

Speed stages (corresponds to velocity gradient c^{-1})	Dynamic viscosity after addition of Diphron 4201 reagent to oil (0.8 kg/t) cPz.					
	20 °C		10 °C		5 °C	
	Bulla	Balakhany heavy	Bulla	Balakhany heavy	Bulla	Balakhany heavy
1a (0,3333)	518,3	672,1	695,8	1174,6	896,7	1396,2
2a (0,6)	302,7	413,2	413,6	782,8	568,3	891,3
3a (1,0)	216,1	354,8	325,3	598,4	465,2	748,6
4a (1,8)	97,6	209,3	143,2	347,1	236,3	503,9
5a (3,0)	69,6	102,7	97,4	234,8	125,1	311,4
6a (5,4)	38,6	61,2	68,7	176,2	91,8	183,1
7a (9,0)	21,9	40,2	36,2	125,8	43,2	143,6
8a (16,2)	9,4	21,6	18,4	65,3	20,4	95,2
9a (27,0)	5,8	9,4	9,1	28,6	11,5	43,0
10a (48,6)	3,5	5,8	6,3	19,5	8,1	26,9
11a (81,0)	2,1	4,1	3,6	11,2	4,9	19,5
12a (145,8)	1,4	2,6	2,5	7,1	3,1	12,7

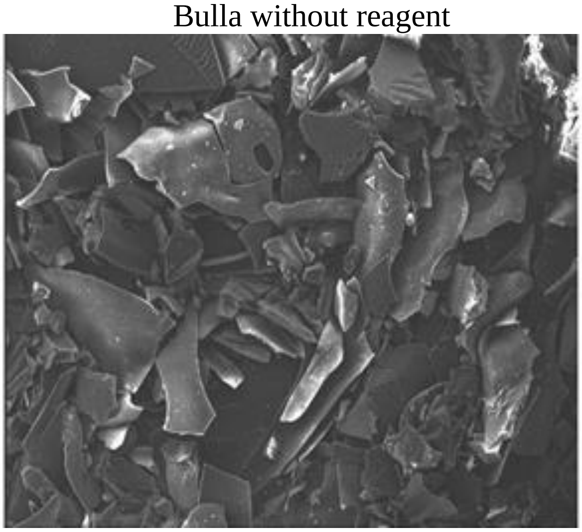
Table 5. Change of dynamic viscosity of oil produced from oil fields of Azerbaijan at different temperatures

Speed stages (corresponds to velocity gradient c^{-1})	Dynamic viscosity, after addition of 1:1 Baf-1: Diphron 4201 cps reagent to the oil (0.8 kg/t) cPz.					
	20 °C		10 °C		5 °C	
	Bulla	Balakhany heavy	Bulla	Balakhany heavy	Bulla	Balakhany heavy
1a (0,3333)	584,8	661,8	792,6	902,8	1027,4	1203,8
2a (0,6)	365,6	393,1	489,3	598,9	663,6	783,9

3a (1,0)	220,3	326,7	403,6	493,8	511,3	602,5
4a (1,8)	111,6	194,3	182,9	256,4	279,5	405,7
5a (3,0)	67,4	106,5	104,8	157,5	169,3	198,5
6a (5,4)	39,1	58,6	65,1	96,3	98,4	119,1
7a (9,0)	22,7	39,2	38,7	73,8	50,9	90,2
8a (16,2)	10,7	20,6	17,9	37,2	29,3	52,7
9a (27,0)	5,8	8,5	8,7	16,7	14,7	24,1
10a (48,6)	3,4	5,2	6,7	11,3	9,5	15,0
11a (81,0)	2,1	3,8	4,1	6,2	6,2	9,2
12a (145,8)	1,3	2,4	2,8	3,4	5,1	6,8


Bulla without reagent


Bulla Baf-1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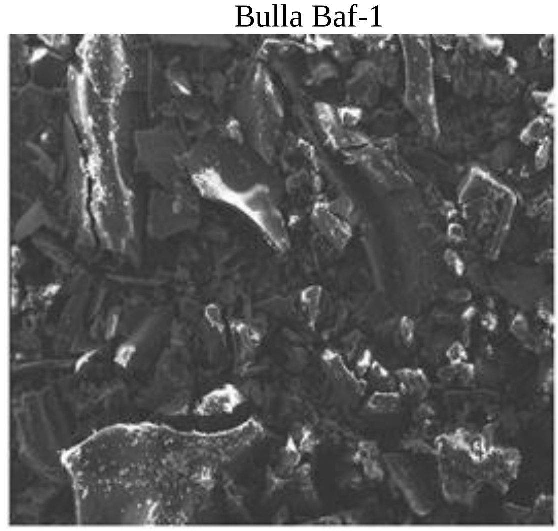
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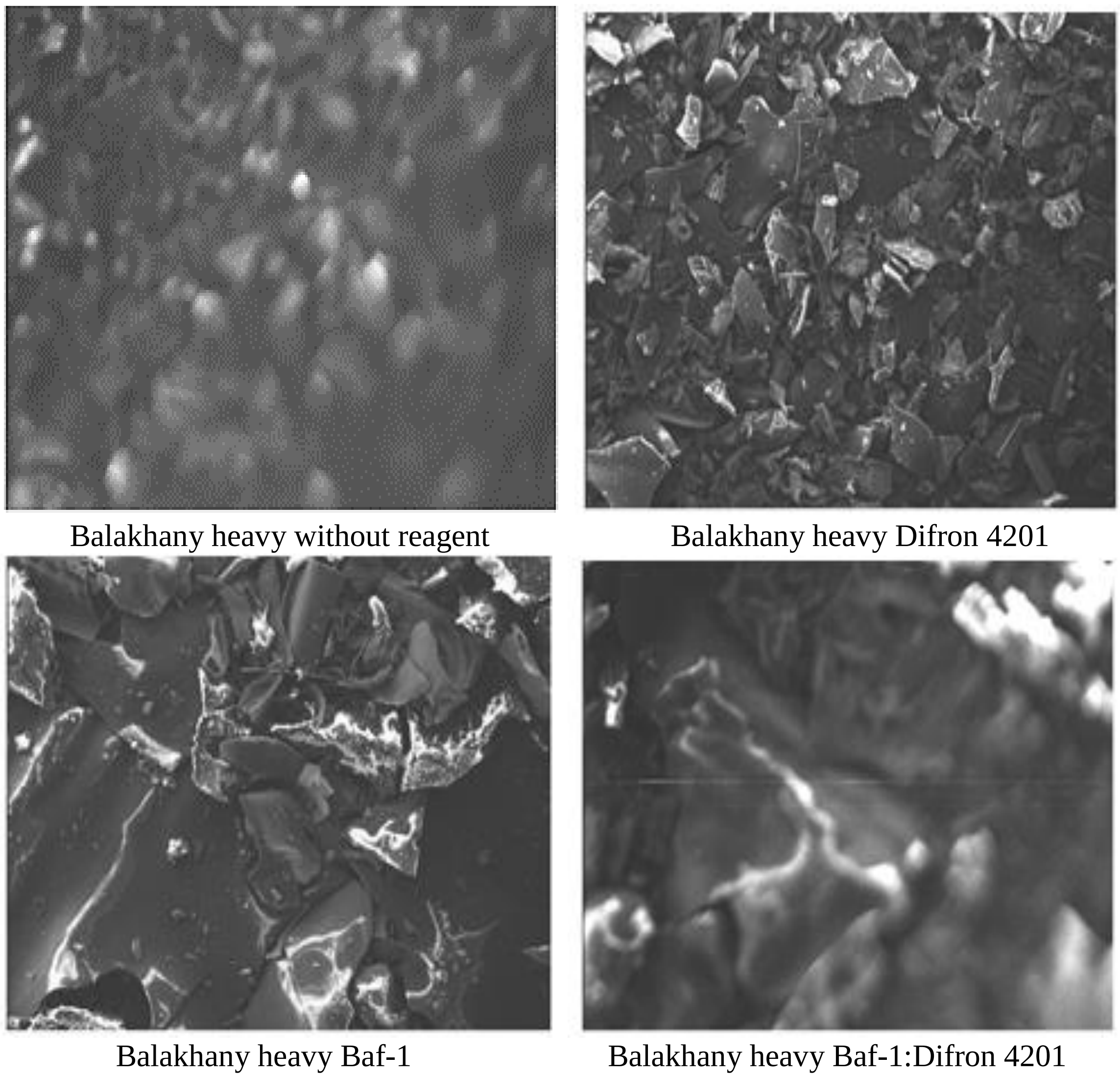

Figure 2. The result of electron microscopy of resin-asphaltene-paraffin compounds.

After the reagent was added to the asphaltene-paraffin resin compounds separated from the oil, the results of their electron microscopy were obtained (Figure 2). The results of electron microscopy are of great importance in the analysis of the structural viscosity of oils.

Our studies show that the rheological properties of oils can be improved by selecting a suitable chemical reagent for the group composition.

4. CONCLUSION

Given that, in reality, most solid bodies and liquids, especially oil and petroleum products, behave like galoid systems, rheological physico-chemical science helps explain the causes of changes and changes in mechanical effects in dispersed systems and high molecular compounds.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the research council of State Oil Company of the Azerbaijan Republic and Geotechnological Problems of Oil, Gas and Chemistry Scientific Research Institute, within Azerbaijan State Oil and Industry University.

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