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ON SOME ASPECTS OF NAFTALAN OIL PROPERTIES

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Abstract: *Naftalan medicinal and fuel oil properties have been studied by methods of IR/UV spectroscopy, DLS, chromato-mass-spectrometry and oil used for curative purposes throughout the year as well. It revealed that two types of Naftalan oil originated not only independently from one another but also from primary organic matter with critical differences, and this can be supported by data of chromato-mass-spectrometry. Identity of Naftalan field can be explained by the fact that it is confined to faults zone and by participation of deep fluids in the formation of oil properties. Lithological composition of rocks is of significant importance in the formation of a unique deposit where rocks are mainly of sandy composition ~ 27%, and clays are ~ 73%. According to DLS, data particles with diameter ranging from 100 to 1000 nm are more intensive in medicinal Naftalan oil. As for the sample of fuel oil, particles with diameter from 50 nm and lower can be observed. Used Naftalan oil tends to aggregate particles with diameter from 100 to 8000 nm; in this case particles of more than 1000 nm are stable up to 50°C. Diffusion coefficients are higher for samples of medicinal oil than for fuel one and this probably provides for pharmaceutic effect. Comparative study of Naftalan oil samples showed DLS data can be a peculiar kind of distinctive fingerprint for used medicinal oil.*

Keywords*: Naftalan oil, IR/UV spectroscopy, DLS, chromate-mass-spectrometry.*
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Introduction

Phenomenon of medicinal effect of Naftalan oil has always been in the spotlight of scientists. Large number of studies has been made in the field of this oil. Full records of Naftalan curative properties were gathered in the monograph by G.A. Kyazimov [1].

Scientific publications pointed out that deposits of Akchagyl and Sarmatian stages were developed in the section of Naftalan pool, and these deposits form the structure of two brachy-anticline folds. Akchagyl deposits were found on rocks of Maykop suite.

Oil-bearing formations within the section of Tertiary deposits played an important role in the process of their shaping, for preservation of oil deposits in Naftalan, especially, foraminiferal layers and Maykop suite. Deposits from the upper Cretaceous to the Quaternary participate in geological structure of area. However, oil was recovered only from different horizons of Maycop suite (Oligocene-lower Miocene deposits) among drilled wells of the area. Deposits of Maycop suite are characterized by high content of C_{org} obtaining 15.1% by average grade 1.86%. Hydrogen index varies from 11 to 612 mg HC/G_{rocks}, with average value 146 mg HC/G_{rocks}.

Naftalan structure is located within Arpa-Samur zone of faults, at all times from Paleozoic up to now it is a zone of active manifestation of tectonic movements, conductor of magmatic melts, load-bearing solutions and

seismicity. Tectonic deformations with thickness up to 2000 m cut all sedimentary complex of Cenozoic deposits and penetrate Mesozoic shallow series. Therefore, it is not unlikely that individuality and unique character of Naftalan deposit can be explained by its confinedness to this area of faults and a probable participation in the formation of a specific outlook of deep fluids oil.

The presence of two types of oil different by its quality in the deposits of Maykop suite in Naftalan deposit: curative hard oil - in the upper horizons of the upper Maykop (I, loamy and II horizons) and fuel light - in lower horizons of the upper Maykop and lower Maykop were reflected in the works of Sh.F.Mekhtiyev [2].

These two types of oil generated independently from each other and from essentially different primary organic matter as well.

A great importance for formation and preservation of oil deposits in Naftalan is played by lithological composition of rocks. The works [3,4] focuses on data dealing with definition of geological structure possessing block structure and nearby disjunctive breaks. There was established reservoir properties increase in north-eastern direction in the examined interval, moreover rocks of mainly sandy composition is ~ 27% and clays ~ 73% for this interval.

According to availability of rearranged steranes, the Naftalan oil generated to clayey rocks. This oil has a high maturity ratio ~ 6; in oils of Absheron ~ 1.9 (Petrov Al.A., 1984), and it is likely connected with affection of terrigenous (clayey) rocks.

In addition to hydrocarbon components, there are also the nitrogenous compounds close to plant alkaloids (A.N. Karayev [5]) in the Naftalan oil, and it allows to suggest a domination of plant material as a source of primary organics, as well as the mixture of organics

superimposed from land given that Maykop deposits were formed due to the destruction of bedrocks of the Minor Causasus.

Transgression of Akchagyl Sea affected the process of properties change for Maykop oil by formation of noncombustible medicinal oil, all area was covered by brackish waters which penetrated into the upper horizons of Maykop suiete. As a result of this process, its physical-chemical and hydro-chemical settings changed towards a common intensification of restoration and stimulation of redox bacterial transformations of oil hydrocarbons (polymerization, cyclization, etc.).

We conducted physical-chemical research of medicinal and fuel oil of Naftalan in our earlier works [6,7]. The difference revealed can be explained by distinction in keeping with the component composition that provided for the difference in medicinal and fuel oils nature itself. However, the further study of structure and properties of Naftalan oils from different horizons by instrumental methods as well as the investigation of oil properties used for medicinal purposes throughout the year should be of some interest.

The aim of the study is to compare samples of Naftalan oil using chromatography-mass spectrometry, IR / UV spectroscopy and dynamic light scattering to establish differences in their composition and properties, which can serve as a kind of distinctive fingerprint of these oils.

Experimental part

Three samples of Naftalan oil were taken for study:

1. Medicinal Naftalan oil from oil terminal I-II horizon, filter 150-586 m.
2. Fuel Naftalan oil from oil terminal III-IV horizon, 1 well.
3. Medicinal Naftalan oil used in Wellness Centre throughout the year.

Studies were carried out by such methods as chromato-mass-spectrometry, IR/UV spectroscopy, dynamic light scattering (DLS), dynamic viscosity of oil samples was also defined.

Chromato-mass-spectrometrical study

The research into Naftalan was conducted on chromato-mass-spectrometry Perkin-Elmer on system including chromato-mass-spectrometry Clarus 680 with interface and high-efficient mass selective detector Clarus SQ8T. Chromatograms of hydrocarbons were obtained on total ion current (TIC). Chromatograph is provided by quartz capillar column with length 60 m, diameter 0.25 mm, impregnated by phase Rtx-1MS. Carrier gas-helium, flow rate - 1 ml/min. Temperature of evaporator 300°C; programming of temperature increase from 80 to 300°C by rate 2°C/min with subsequent isotherm during 70 min. Ionizing voltage of source - 70 eV, source temperature-250°C. Carbon sulphide CS_2 was used as a solvent.

UV- spectroscopic study

Spectra of visible and ultraviolet range were obtained on UV spectrometer LAMBDA, Perkin Elmer by use of quartz cells with length of optical distance 10 mm; all measuring conducted by 22°C atmospheric pressure. Samples of Naftalan oil are prepared by dissolution in hexane, qualification (ch. pure).

IR spectroscopic

Research was carried out on ^{1}, on accessory MATR. spectrometer Frontier by range $\lambda\sim$ 700-4000 sm^-

Method of Dynamic Light Scattering

Structure of oil components was studied by method of dynamic light scattering on LB-550 Horiba device which makes it possible to define size of nanoparticles by range 1-6000 nm and concentration from ppm level to 40% of mass.

Dynamic viscosity was defined on rotation viscometer LVM-D.

Results and discussions

Data on total number definition of benzene, naphthalene and phenanthrene in oil samples by method of UV spectroscopy are represented at Fig. 1-3 and on Table 1.

Table 1. Content of aromatic HC in Naftalan deposit oil on data of UV spectroscopy.

Samples of Naftalan oil	C benzene D_{200}	C naphthalene D_{230}	C phenanthrene D_{250}	Σ C arom,%
medicinal	9.666	5.512	7.284	22.463
fuel	11.246	6.163	7.387	24.796
used throughout the year	2,1011	1,0932	0,55976	11,99601

As is seen, content of aromatic HC is insignificantly more in fuel oil than in medicinal oil and mainly it is due to benzene. As for used oil sample, total content of aromatic hydrocarbons reduces in it by two times. It can equally be referred to benzene, naphthalene, phenanthrene as well (Fig 1-3).

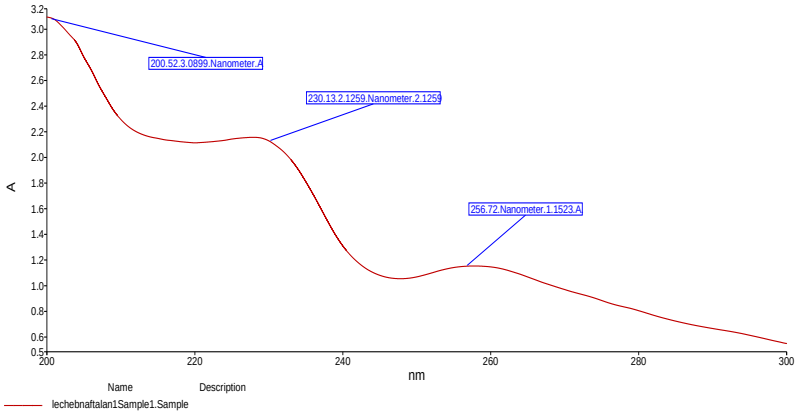

Fig. 1. UV spectrum of Naftalan medicinal.

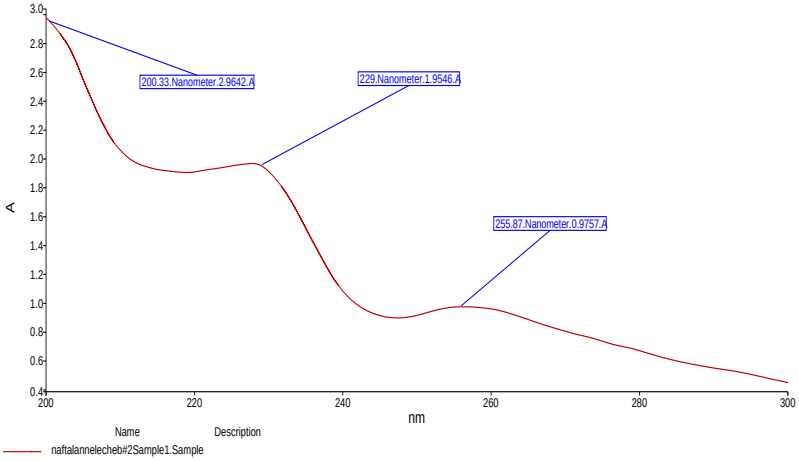

Fig. 2. UV spectrum of Naftalan fuel.

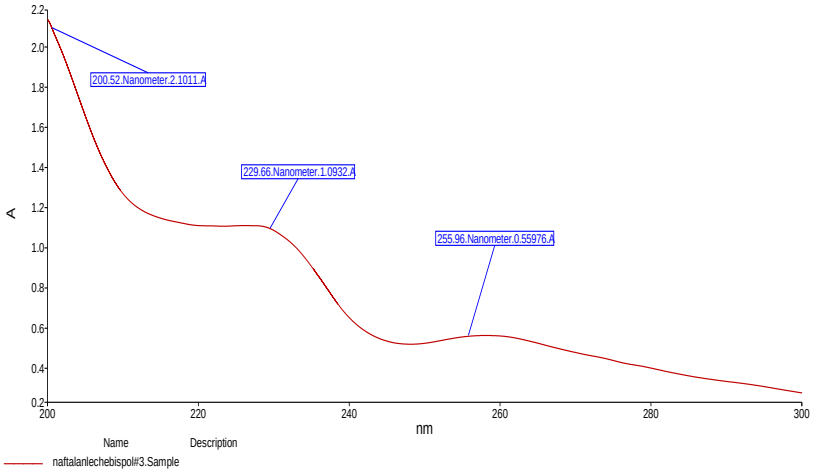

Fig. 3. UV spectrum of Naftalan used throughout the year.

Data of dynamic viscosity for three samples of Naftalan oil by temperatures T ~ 25, 35, 50°C obtained on rotation viscometer are as follows:

1. $\Sigma\eta_{25}$ = 396 mPa S ; $\Sigma\eta_{35}$ = 340 mPa S ; $\Sigma\eta_{50}$ =314 mPa S

2. $\Sigma\eta_{25}$ = 332 mPa S ; $\Sigma\eta_{35}$ = 329 mPa S ; $\Sigma\eta_{50}$ =300 mPa S

3. $\Sigma\eta_{25}$ = 391 mPa S ; $\Sigma\eta_{35}$ = 347 mPa S ; $\Sigma\eta_{50}$ =321 mPa S

Values of dynamic viscosity of fuel oil are less than for medicinal one, As test temperature increases, viscosity reduces. As for sample 3 by T=25°C, its viscosity is practically equal to sample viscosity1 (medicinal), however by growth of temperature sample viscosity 3 exceeds sample viscosity 1. Probably it occurs as a result of increase nanoassociates in the used oil which are prone to agglomeration. Therefore, study of Naftalan dispersity was of a great interest.

Dispersity of samples for medicinal and fuel Naftalan oil was studied by method of dynamic light scattering (DLS) which allows to define sizes of nanoparticles by range 1-6000 nm by concentration from ppm level to 40% of mass.

It is necessary to take into account the complex of nanocolloids features in oil and gas media to prevent the fulfillment of intensification of critical phase transformations in nanocolloid system.

Table 2. Size of particles and diffusion coefficient of Naftalan oil.

Name of sample	T=25^{0}C, g,nm / Dif.Coef., m^2/s	T=35^{0}C, g,nm / Dif.Coef., m^2/s	T=50^{0}C, g, nm/ Dif.Coef., m^2/s
Naftalan medicinal	1)340.9/ 2.305$*$ 10^{-12} 2)2.0 /3.951 $*$ 10^{-12}	1)144.2 / 6.319 $*$ 10^{-12} 2)875.0 / 1.040 $*$ 10^{-12}	1)0.4 / 3.093 $*$ 10^{-12} 2)1.4 / 7.875 $*$ 10^{-12}
Naftalan fuel	1)27.8 / 2.862 $*$ 10^{-12} 2)7.1; 556.6 / 1.071 $*$ 10^{-12}	1)35.7 / 2.638 $*$ 10^{-12} 2)42.9 / 2.130 $*$ 10^{-12}	-
Naftalan used	1)0.5 / 1.623 $*$ 10^{-9} 2)4413.4/ 1.858 $*$ 10^{-13} 3)205.3/ 3.819 $*$ 10^{-12}	1)3.1 / 2.958 $*$ 10^{-10} 2)167.5 / 5.475 $*$ 10^{-12} 3)1203.1; 6212.2 / 1.830 $*$ 10^{-13}	1)5765.6 / 1.930 $*$ 10^{-13} 2)1955.4 / 5.997 $*$ 10^{-13}

It has to be kept in mind that oil is represented by colloid system where colloid phase is represented mainly by asphalthenes [8]. Majority of self-organizing molecules of oil includes chiefly to the composition of asphalthene fractions which can be distinguished according to solubility [9]. Experimental data obtained as a result of method DLS are represented in Table 2. Measuring of dependable values of distribution density from particles diameter for solutions of Naftalan oil in toluol is carried out in the range of temperatures 25, 35, 50°C. Fig. 4 demonstrates density values (q) %, size of particles at peak (Mode) by different test temperatures, in nm; and also mean geometrical values of particles (Geo mean), in nm. Diffusion coefficient (Dif. Coef.), in m^2/sec, estimated on Stokes-Einschtein equation: $D= kT/3\pi\eta d$, its values are given for each test temperature (table 2).

There is information (N.D. Aliyev, S.K.Zeinalov a, 1983) dealing with naphtalan's fungicidal properties when its dispersity plays a certain role in the behavior of these properties. It is due to finely dispersed particles which can be formed particularly during naphtalan dilution into vaseline and then they are capable of penetrating through bacterial shell. Thus, mold fungi appear to be more sustainable to naphtalan effects than other species of fungi (N.D.Aliyev, S.K.Zeinalova, 1979).

As is seen from Fig.4, particles with diameter ranging from 100 to 1000 nm (T~25-35 °C) are more intensive in medicinal naftalan oil, and by 50°C – solution is homogeneous. Particles with diameter 50 nm and lower (T ~ 25-35 °C) can be found in the sample of fuel oil, and by 50°C the solution is homogeneous. As for used Naftalan oil, it is prone to aggregation from 100 to 8000 nm, in this case particles more than 1000 nm are sustainable up to 50°C.

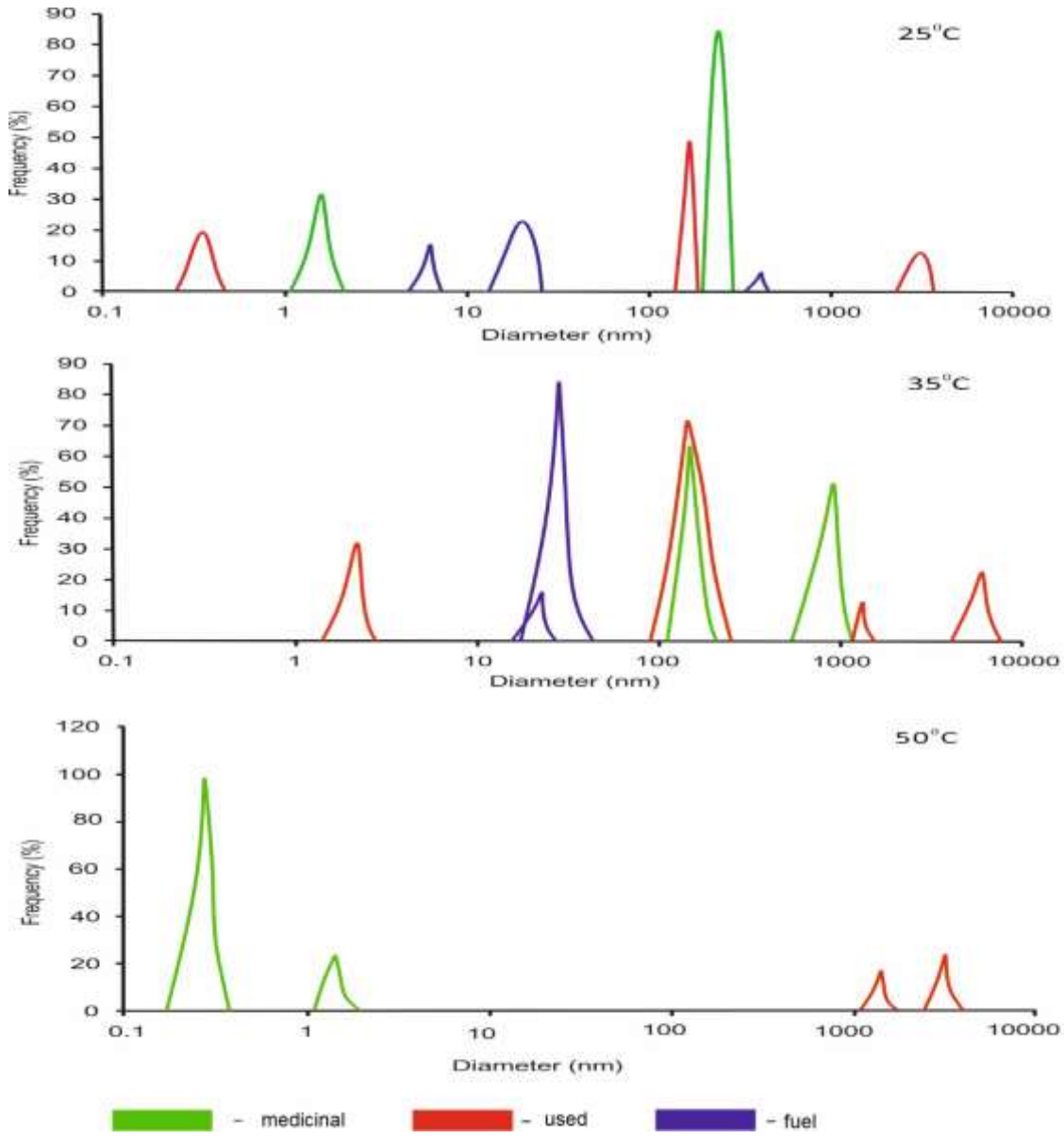

Fig. 4. Dependence diagram of distribution values on density from particles diameter for Naftalan oil solutions in toluol by different test temperatures.

As can be seen, the diffusion coefficient are more evident for medicinal oil samples than for fuel, as for density (q) value – it is ~50-80% in medicinal oil, and this probably provides for pharmaceutical effects.

According to chromato -mass spectroscopy, the HC composition of Naftalan oil was defined. Taking into account

hydrocarbon composition, the medicinal Naftalan oil differs significantly from fuel. (Table 3).

Content of naphthenic HC ~ 59.37 % in fuel oil, and in medicinal ~ 77.04 %; ∑ content of alkanes is only 4.47% in medicinal oil whereas in fuel ~ 22.93%. Content of aromatic HC is nearly the same.

Table 3. Hydrocarbon composition of Naftalan oil

Name of samples	∑ alkenes	Naphthenic					∑ naph-tenes	Arenes			∑ arenes	m/z 95
		mono	di	tri	tetra	penta		mono	di	tri (tetra)		
Naftalan medicinal	4.47	48.24	24.01	4.79	-	-	77.04	0.22	5.76	13.44	19.42	24-27
Naftalan fuel	22.93	41.7	17.14	-	0.53	-	59.37	3.7	1.76	12.02	17.48	17
Naftalan used throughout the year	9.10	48.24	35.3	4.79	-	-	77.04	3.65	5.3	5.12	14.07	

For additional information of group chemical composition of studied oil samples and nature of structural fragments, their mutual correlation IR spectroscopic analysis conducted.

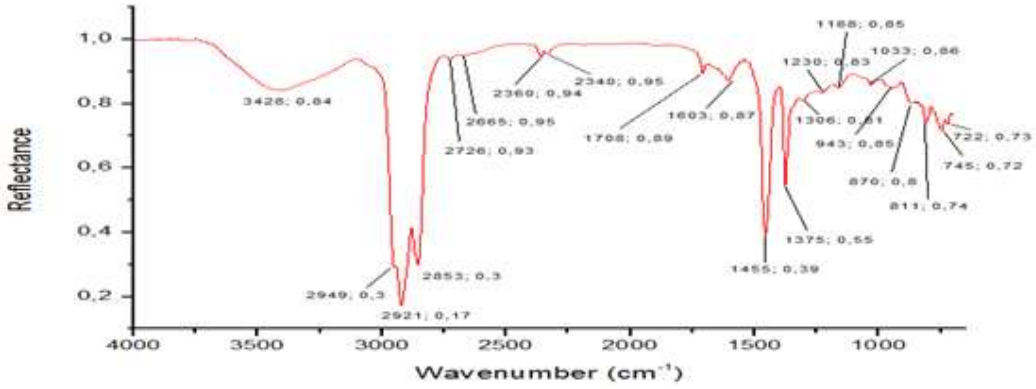

Fig. 5. IR spectrum of Naftalan medicinal oil

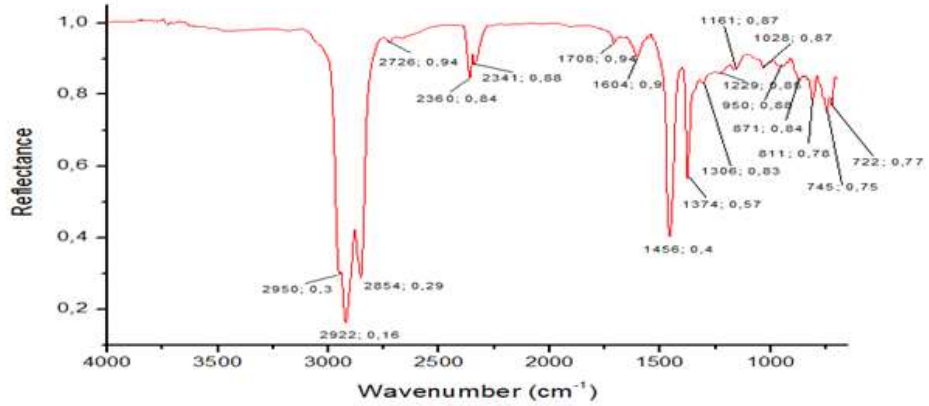

Fig. 6. IR spectrum of Naftalan fuel oil

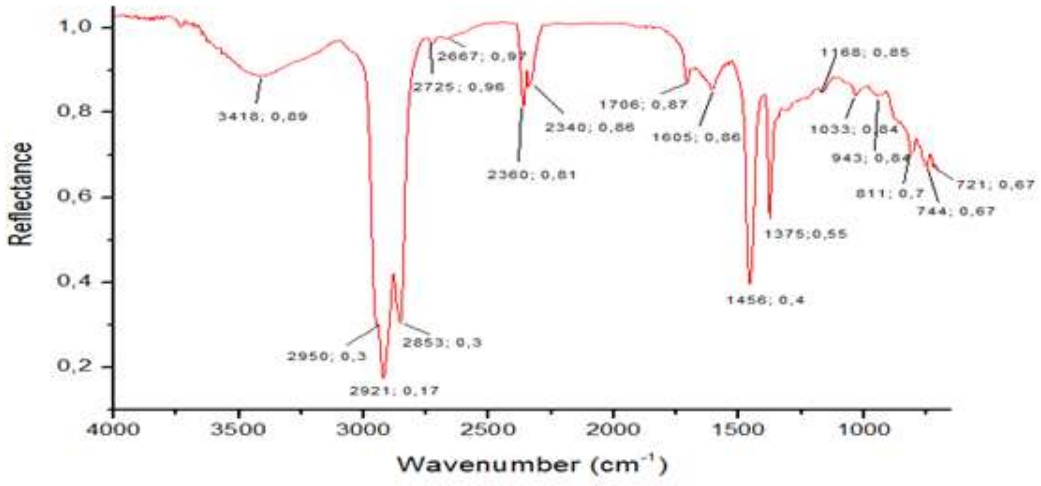

Fig. 7. IR spectrum of Naftalan used throughout the year

Spectral coefficients were estimated on IR spectra. In this respect, peak intensities of analytical absorption bands were measured - 720, 870, 1376, 1464, 1600 sm^{-1} and according to optical densities of the main absorption bands aromaticity coefficient was estimated $C_{ar}=D_{1600}/D_{720}$, aliphatic coefficient $C_{al}= D_{720} + D_{1380}/ D_{1600}$, coefficient of branching $C_{br}= D_{1380}/ D_{720}$ and of oxidation $C_{ok}=D_{1710} /D_{1465}$. There are also coefficients reflecting relationship of substituted (bi-tricyclic) aromatic structures to total content of aromatic fragments ($C_{870}/_{1600}$) and relation of aliphatic fragments sum (CH_2+CH_3) to aromatic structures ($C_{1464}/_{1600}$) [10]. There is obtained data for oil samples in Table 4.

Table 4. Spectral coefficients of Naftalan oil

Properties /naftalan samples	C_{ar} 1600/720	C_{al} 720+1380 / 1600	C_{br} 1380/720	C_{ok} 1710/1465	C_{al}/C_{ar} 1464/1600	Sub.ar ΣAr 870/1600	Polyc. str. 750/1600
1	1.191	1.471	0.753	2.282	0.448	0.919	0.827
2	1.169	1.49	0.74	2.35	0.440	0.93	0.833
3	1.283	1.418	0.820	2.175	0.465	0.900	0.744

1-Naftalan medicinal oil; 2- Naftalan fuel oil; 3- Naftalan oil used medicinally throughout one year.

Aromaticity coefficient of Naftalan fuel oil is less than of medicinal oil, moreover there is a significant amount of condensated aromatic structures among aromatic structures of this oil, and this can be supported by coefficient $C_{870}/_{1600}$(0.93) and of polycondensated asphalthene structures (C_{750}/C_{1600} = 0.833).

Naftalan oil sample (№3) has the bigger coefficient of aromaticity C_{ar}, this oil is used for medical purposes throughout the year (1.283). At the same time it possesses the less index of abundance of substituted arenas $C_{870/1600}$ (0.900), and this demonstrates the less content of polycondensated structures in this sample. There is the biggest coefficient of branching (C_{br}) and the lowest aliphatic coefficient (C_{al}) in this sample. Used medicinal oil possesses the least coefficient pointing out the relationship between polycondensated arenas and total content of aromatic structures (C_{750}/C_{1600}=0.744). It shows a high content of monocyclic arenas which are natural solvents

for polycondensated aromatic structures and eventually their aggregative stability increases.

Therefore, individuality and unique character of Naftalan deposit due to its confinedness to Arpa-Samur zone of faults, and to possible participation in formation of specific oil form of deep fluids can be confirmed by numerous comparative studies of medical and fuel oil.

Presence of two types oils is different by quality in beds of Maykop suite of Naftalan deposit: in the upper horizons of the upper Maykop - medicinal hard oil, and in the lower horizons of the upper Maykop and the lower Maykop - fuel light indicates the following: these two types of oil are generated independently from each other and generated from significantly different primary organic matter as well.

According to chromato-mass spectrometry Naftalan medicinal oil is essentially different from fuel one. Content of naphthenic HC is ~ 77.04% in medicinal oil and in fuel oil ~ 59.37%. ∑ content of alkanes is only 4.47% in medicinal oil, while in fuel oil ~ 22.93%.

On UV spectroscopy data content of aromatic HC is insignificantly more in fuel oil than in medicinal oil due to benzene. As for used oil sample, a total content of aromatic hydrocarbons is less in it than in two previous samples. It is noteworthy that there is a growth of used oil viscosity (sample 3) by increase of temperature as compared with primary medicinal oil (sample 1). Evidently this occurs as a result of nanoassociates increase in used oil prone to agglomeration.

So, according to DLS data particles with diameter ranging from 100 to 1000 nm are more intensive in Naftalan medicinal oil. Particles with diameter ranging from 50 nm and lower can be found in samples of fuel oil. Naftalan used oil is prone to aggregation of particles with diameter from 100 to 8000 nm; in this case particles with more than 1000 nm are stable up to 50°C. Diffusion coefficients are greater for samples of medicinal oil than for fuel one, evidently it provides pharmaceutic effect.

Comparative IR test of Naftalan oil samples showed less indices of abundance of substituted arenas ($C_{870/1600}$ =0.900) after medicinally used Naftalan oil throughout the year, and this points out lesser content of polycondensated structures in this sample, as well as the lowest coefficient indicating relationship between polycondensated arenas and total content of aromatic structures (C_{750}/C_{1600} =0.744). It proves a high content of monocyclic arenas which are natural solvents for polycondensated aromatic structures raising their aggregative stability to be used as an original distinctive fingerprint of used medicinal oil.

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NAFTALAN NEFTİNİN BƏZİ XÜSUSİYYƏTLƏRİNƏ DAİR

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Xülasə: *Müalicəvi və yanacaq xassəli Naftalan nefti, həmçinin bir il ərizində müalicəvi məqsədlər üçün istifadə olunmuş neft İQ/UB spektroskopiyası, işığın dinamik səəpilməsi və xromato-kütlə spektrometriyası üsullarının köməyi ilə tədqiq olunmuşdur. Xromato-kütlə spektrometrik tədqiqatlar göstərir ki, bu iki növ Naftalan nefti bir-birindən asılı olmayan və əhəmiyyətli dərəcədə fərqlənən ilkin üzvi maddələrdən əmələ gəlmişdir. Naftalan yatağının fərdliliyi onun qırılma zonası ilə məhdudlaşması və neft xassələrinin formalaşmasında dərin mayelərin iştirakı ilə izah olunur. Unikal yatağın formalaşmasında süxurların litoloji tərkibi də böyük əhəmiyyət kəsb ediir, burada əsasən qumlu tərkibli süxurlar ~ 27%, gil isə ~ 73% təşkil edir. İşığın dinamik səpilməsi məlumatlarına görə, müalicəvi Naftalan neftində diametri 100-1000 nm diapazonda olan hissəciklər daha intensivdir. Yanacaq nefti nümunəsində diametri 50 nm və ondan aşağı olan hissəciklər müşahidə olunur. İstifadə olunmuş Naftalan nefti diametri 100-dən 8000 nm-ə qədər olan hissəcikləri birləşdirməyə meyllidir, ölçüləri 1000 nm-dən yuxarı hissəciklər isə 50 ^{0}C-ə qədər dayanıqlıdır. Müalicəvi neft nümunələri üçün diffuziya əmsalları yanacaq nefti nümunələrindən daha yüksəkdir ki, bu da açıq şəkildə farmaseptik effektə təsir edir. Naftalan nefti nümunələrinin müqayisəli tədqiqi göstərmişdir ki, işığın dinamik səpilməsi məlumatları istifadə olunmuş müalicəvi neftin özünəməxsus fərqləndirici finqerprinti ola bilər.*

Açar sözlər: *Naftalan nefti, IQ/UB spektroskopiyası, işığın dinamik səpilməsi, xromato-kütlə spektrometriyası*

О НЕКОТОРЫХ АСПЕКТАХ СВОЙСТВ НЕФТИ НАФТАЛАНА

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Аннотация: *Методами ИК/УФ спектроскопии, динамического рассеяния света, хромато-масс-спектрометрии исследовались лечебная и топливная нафталанская нефть, а также нефть, использованная в течение года в лечебных целях. Показано, что два вида нафталанской нефти произошли не только независимо друг от друга, но и из существенно различного исходного органического вещества, о чем свидетельствуют данные хромато-масс-спектрометрии. Индивидуальность Нафталанского месторождения объясняется приуроченностью к зоне разломов и участием глубинных флюидов в формировании свойств нефти. Также немаловажное значение в образовании уникальной залежи имеет литологический состав пород, где породы преимущественно песчаного состава составили ~27%, а глины ~ 73 %. По данным динамического рассеяния света в лечебной нафталанской нефти наиболее интенсивны частицы с диаметром в диапазоне от 100 до 1000 нм. В пробе топливной нефти наблюдаются частицы с диаметром частиц от 50 нм и ниже. Использованная нафталанская нефть проявляет склонность к агрегированию частиц с диаметром от 100 до 8000 нм, причем частицы свыше 1000 нм устойчивы вплоть до 50 ^{0}С. Коэффициенты диффузии для проб лечебной нефти больше, чем топливной, что, очевидно, способствует фармацевтическому эффекту. Сопоставительные исследования проб нафталанской нефти показали, что данные динамического рассеяния света могут явиться своеобразным отличительным фингерпринтом использованной лечебной нефти.*

Ключевые слова: *нефть Нафталана, ИК/УФ спектроскопия, динамическое рассеяние света, хромато масс-спектрометрия*