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RADIO-GEOCHEMICAL METHODS AT SURFACE EXPIOTATION OF OIL AND GAS FIELDS

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A review of the situation with radio-geochemical methods of searching oil and gas fields is presented. Potential reasons of radio-geochemical anomalies formation are considered, some approaches to interpretation of radio-geochemical datum are listed.

During the last 15 years in the majority of world regions, where oil and gas prospecting work are conducted, the positive trend of using various geochemical and geophysical methods of surface survey is observed. Such a tendency is typical for Russia, where due to stabilization of economical situation, and the beginning of important international projects realization of oil and gas transportation to Asian parts of the Pacific region, more attention is paid to growth of resource and hydrocarbon materials. At the same time, the territories of the East Siberia and the Far East, which are studied quite poorly, are regarded as potentially main oil and gas base. To solve a complicated task of new pool discovery at the earliest possible date, along with traditional prospecting seismology one need to attract various number of additional efficient and high-performance methods of exploration activity in hydrocarbons.

Today in the world's practice, with different frequency and in different combinations, a great number of direct and indirect methods of oil and gas deposit search are used.

Direct methods are hydrocarbon survey, where depending on modification, free, odsorbed and solute hydrocarbon gases are analyzed, and bituminological researches are quite rare. Soil, snow, water, sludge from drilling-and-blasting well, artificial sorbents are used as sample objects. Among methods directed to define oil-and-gas fields assist nonhydrocarbon components and exposure of geochemical and geophysical fields epigenetic transformations, the following types of survey are established: microbiological, helium, neon-argon, ozone and thermal survey. The following methods are also used: magnetometry (in surface and aerial variants), electrometry (by methods of induced polarization, field formation, etc.) electrochemical research (method of partial metal extraction, redox potential, etc.), electromagnetic research (maghetotelluric survey, etc.). A special complex of lithogeochemical methods is developed; it allows evolving oil-bearing areas, basing on the analysis of rock's petrophysical characteristic, concentration, form of presence and distributional character of some chemical elements isotope correlation, peculiarities of mineral compound of a rock, etc.

In the 20th of the past century, when direct methods of oil and gas field search were just conceived, in the USSR information about studying the influence of hydrocarbon accumulation on ground radioactivity was published [1]. Properly investigated Maykopskoe oil field was the research subject, where on two profiles views radiation intensity were measured with the help of collision chamber. As a result of work over oil field, a radioactivity rise, exceeding the bounds of experimental error, was revealed.

Practically at the same time a great number of information on the studying of radioactivity of stratal water in oil field appeared, which clearly showed that contact water of hydrocarbon deposits is characterized by anomalously high radium concentration.

However, later on radioactive survey during oil field search were practically stopped, because of development of methods aimed at direct definition of hydrocarbons, and because of absence of appliances, permissive to register comparatively quickly and with fast response parameters of radio geochemical areas.

An impulse to start the research into evaluation of radio geochemistry methods of surface search was development and application of gamma-ray spectrum apparatus into exploration work, basing on the scintillation counter. H. Lunberg cites data about airborne- radiometric profile, crossed oil field Redwater in Canada [2]. The survey route shows distinctly the low-level radioactivity sector, coinciding with the projection fragment of oil pool. At the same time, relative increasing of field radioactivity is typical for field boundaries. Appearance of radioactive anomalies over oil and gas accumulation is explained by diffusive transportation of water-soluble radium from deep levels to surface. At that, the very hydrocarbon accumulations, by the authors opinion, performed a shield function on the way of the substance that explains fixed areas of low-level radioactivity [3].

Radioactive survey, carried out next 10 years on over than 30 Canadian and US oil-and-gas fields, displayed that overwhelming majority of them prove themselves in radio-geochemical fields.

Since 1956, researches on effectiveness evaluation of radiometrical methods during oil and gas field search were started in USSR. In a brief space of time the researches were carried out on well-known oil and gas fields of Nizhniy Volga region, Ciscaucasia, West Turkmenia. Along with measuring of radioactivity and radioactive elements concentration, examining of some geochemical characteristics of surface sediments were realized, peculiarities of geomorphologic and lithologic structure of the territories were analyzed. An important conclusion was made at that time, that radioactive anomalies over hydrocarbon pools are particular case of general epigene transformation of geochemical fields [4].

The results of numerous test-systematical and laboratory researches, which were carried out that time at the laboratories of nuclear geology and geophysics of Oil Institute AN USSR, allowed to call in question the depth migration of anomaly-formative radioactive elements. Assuming an epigenetic character of anomalous radio geochemistry effects, radioactive elements redistribution in surface sediments of oil and gas bearing territory, according to researchers opinion, connected with components, changing the geochemical situation in the hyperkinesias zone, coming from hydrocarbon pools. On the one hand, this leads to changes in radionuclides migration capacity, on the other hand, to decreasing of sorptive capacity of sedimentary over productive of structures [5].

Practically at the same time, the article of A.F. Gregory was published; it casts doubt on connection between radioactive anomalies and hydrocarbon pools presence at a depth [6]. As examples, he introduced oil fields Redwater and Coalinga, where earlier H. Lunberg basing on airborne radiometric data, evolved radioactive anomalies coinciding with oil-drainage boundary. On the basis of comparison of radioactivity maps and geological and landscape territories structure, soil structure, he concluded that the character of radioactivity changes is determined not by oil accumulation influence, but by changes in lithologic and mineral structure of surface sediments and by salinity degree.

Similar point of view was expressed by W.G. Kellog, who analyzed the connection of radioactive field with subsurface-tectonic pattern of the oil field Tejon Grapevine (USA) and adjacent territories. He concluded that low radioactive anomalies control anticline, elevated at the relief, and that formation of such a radioactive field disturbance is not connected with hydrocarbon pools, but determined by more intensive leachability and radioactive elements transport from elevated structures [7].

During the process of development of radioactive methods of oil and gas field search, carried out in laboratories of radio-geochemical and isotope analysis of the All-Russia Scientific Research Institute of Nuclear Geophysics and Geochemistry, the following facts appears: when conducting surface and aerial survey, productive and nonproductive anticline prove themselves identically in distribution of radioactivity field and natural radioactive elements. Relying on materials of gamma-ray logging of deep well and data on lithologic structure, dense correlation was shown between radioactivity meanings and rock granulometric composition. At the same time, the tendency is observed for all stratigraphical horizons to increase crest of fold grittiness of inherited folds with simultaneous growth of flank shaliness, and this determines the character of radionuclides distribution. There were not founded any essential distinctions during gamma survey in the Caspian Sea and the Sea of Azov's area of water, where physicochemical conditions differs greatly from that of the hyperkinesias zone. As the result, radio-geochemical methods played a supporting role in research of structure-tectonic pattern of oil and gas bearing territories [8].

It should be mentioned, that in the 50^{th}–60^{th} of XX century it was considered, that gas and water soluble components from oil and gas accumulation can not migrate through overlying pervious bulk of basic sediments to surface. Exploration possibilities of direct methods suffered serious criticism that time [9].

Discovery of paragenesis of the subvertical zonoringshaped geochemical and geophysical fields in sedimentary cover of the earth's crust, successes in geochemical and geophysical peculiarities of oil and gas fields investigation, progress in high-precision, recording equipment development, all these created the favorable prerequisites for more active application of direct and subdirect geochemical and geophysical researches in oil and gas exploration work.

Increasing interest in geochemical research of hydrocarbon pools affected radio- geochemistry methods. In the USA the results of reinterpretation of gamma-roy spectrometry data of oil and gas bearing territories, which were received as a result of NURE program realization and other aerographical researches, are cited [10–12], ground total counting [13] and radon survey [14] are carried out. The question of connection between radioactive anomalies and epigenetic rocks' carbonate formation is studied [15], the character of relation between natural radioactive elements is analyzed [16, 17]. Thermoluminescent, radiometrical charting is conducted extensively during the oil-and-gas content evaluation of some Chinese regions [18, 19]. Experimental, systematic and prospecting work using radioactive methods are executed in Russia [20–22], Israel [23, 24] and India [25]. These researches results prove the presence of radioactive anomalies over oil and gas fields, and authors support epigenetic nature of destruction in the structure radioactive fields. However, this is not enough in the process of realization of the radioactive anomalies formation. In most articles one can find just reference to well-known models of formation of anomalous effects, or some marginal changes are made.

The suggestion about possible relative unification of surface basic sediments by natural radioactive elements as the result of more intensive corrosion of rock forming minerals in the oil-and-gas drainage boundaries is of interest [24]. Oxidation products of migratory hydrocarbons, carbonic acid is one of them, destroy crystalline matrix minerals, releasing their chemical elements, which are carried out by hypergene water to periphery of pools.

Learning the potential contribution of biogenic magnetofossilium in magnetizability increasing in the influence zone of hydrocarbon flow, some data was received about inclusion of uranium bioaccumulation to the process [26]. Some species of aerobic, hydrocarbon-absorbing, magnetotropic microorganisms decreased three times U concentration in solution, and during magnetic separation, biomass contains U 1000 times more than in the original solution. Sulfate-reducing bacteria are also characterized by good absorptance. It was determined, that magnetizability increases sharply in terrigenous formation with traces of old oil spills and ventilated bitumen, though source oil is diamagnetic.

This allows pointing out potential contribution into anomalous radioactive effects formation of microorganisms, feeding on hydrocarbon components.

Redox potential meaning plays a great role in uranium migrating capacity. According to S.J. Pirson, hydrocarbon pools are in a way of natural galvanic element [27, 28]. On the rock pole boundaries intrusion of gas and liquid hydrocarbon components due to appearing of Eh boundary, electrochemical reactions take place, they activate movement of electro telluric current. Current vertical circulation occurs between oil and gas accumulation and low boundary of hypergenesis zone. As a consequence meaning gradient of redox potential appears above hydrocarbon-water contact on the redox boundary level. Moreover, as the result of electrophoresis, uranium redistribution in buffer area is possible, between rocks, affected and not falling under epigenetic influence of hydrocarbon pools.

Number of researches on commercially productive hydrocarbons of East Siberia, Kazakhstan and Ukraine show that soil and ground samples, selected in the field contour, on average are characterized by Eh meaning less than 150 mV, out of fields influence zone more than 250 mV. Medium acidity for terrigenous deposits of upper part of a section above oil and gas accumulation is 1–2 unit more on average, than beyond the productivity boundary. Great variation of values of such physical-chemical indicators is stated in the intermediate region [29].

Thus, hydrocarbon pools create contrast geochemical barriers, uranium, attenuated in oxygen waters, can precipitate on them, this process is more active on the boundary of oxidative and restoration environment of a cut, at the places of intense substance inflow from hydrocarbon polls.

Indeed, analyzing role of hydrochemical factor in the radioactive anomalies formation, one points out a considerable deviation of the radioactive balance value in the active water cycle with surfase of oil and gas fields. At the same time, waters of hypergenesis zone, suffered hydrocarbon component influence, are characterized by radioactive balance change towards Ra increasing; it is connected with U concretion on restoration barrier [30].

Moreover, combined research of concentration of natural radioactive elements (NRE), basing on γ-ray spectrometry and radioactivity, which are measured by highly sensitive thermo-luminescent detector (TLD), allows to suppose that short-living isotopes participate actively in the process of loaded radionuclides redeposit. This supposition is based on the fixed effects in the structure of anomalous radio-geochemical fields, when peculiar zoning in the character of NRE distribution and integrated radioactivity is observed. In such cases, we observe invert correlation between U (Ra) anomalies signs (positive – negative) and thermoluminescense detector. It should be mentioned, that basing on full-scale experiment, the main contribution to accumulated light-sum by TLD is made by β-active ^{210}Po and γ-active ^{214}Bi, switch to them in radioactive decay series ^{238}U is conducted through ^{226}Ra and radioactive gas ^{222}Rn.

Today, researches of radioactive elements behavior in lower geochemical zone of oil and gas fields lacks of information, so it is impossible to evaluate correctly the character and scale of NRE redistribution over the whole sediment, crossing oil and gas accumulation. However, there exists information about radio-geochemical characteristics of terrigenous deposits of productive and semiproductive horizons, where researchers distinguished zones of epigenetic uranium redeposit, they based on the results of quantitative analysis of radionuclides accumulation level [31, 32].

Contemporary fluid-dynamic oil and gas evolution models are based on sediment characteristic to laminate, during lithogenesis, into compacting and decompacting zones, saturated with fluids, at high, internal pressure, this leads to creation of a massive fluid-dynamic system, expressed by ascending thermal current, activating oil and gas formation processes [33, 34].

The fluid-dynamic systems are characterized by connection with tectonomagnetic structures, formed in the deep fault crossing, and «through-formation» fluid absorb systems are distinguished among them. These systems are the products of abyssal physico-tectonic factors and degassing processes, they perform fluid-constringent function and control spatio-temporal associations of various oil and gas pools.

Thus, the unity of various geophysical and geochemical anomalies above oil and gas accumulation is determined by oil-and-gas bearing region history and mechanisms of development, which are heterogeneous, long-lived and self-developing systems, related to deep tectonic structures, these systems are hyperpermeability zones, where intensive substance migration is taking place, it defines complex mechanisms of geophysical and geochemical fields transformation. We consider it would be wrong to deny the possibility of radioactive elements involving into the process of formation and further destruction of hydrocarbon accumulation.

Basing on researches of elemental composition of fluids relic, closed in microcracks of rocks and mineral sediments of oil and gas bearing regions, R.P. Gottih and coauthors, suggest that uranium, lithophilous and chalcophilios metals migrate in bituminous phase as metalloorganic complex [35]. Moreover, the presence of considerable concentration of radioactive elements in restoration fluids of oil and gas bearing systems can be one of the factors in the high-molecular hydrocarbon compound synthesis.

Group of researches from St. Petersburg proved experimentally and theoretically that transportation of some heavy metals from hydrocarbon pools in gas-bubble flow is possible [36].

To sum up, it is possible to say, that today there exist separate, in most cases indirect facts, proving the epigenetic character of radio geochemical anomalies above oil and gas fields.

Considerable progress exists in the development of methodic of radio-geochemical information receiving and processing. There is a definite number of radio-ge-

ochemical searching methods, allowing to delineate oil and gas accumulation in the territories of various geological and landscape structure.

Radio-geochemical researches in landscapes with low watering and restricted development of allochthonic sediment are most informative. Though, in most cases it is possible to fixate oil and gas fields presence even in adverse for radioactive survey Siberian landscapes, when special particularized ways of isolation of epigenetic part of radioactive fields are used [17].

Analyses of numerous literary data actually shows that majority of anticline deposits prove themselves with anomalies of circular type («halo» effect in foreign literature) with relatively low radioactivity values and NRE

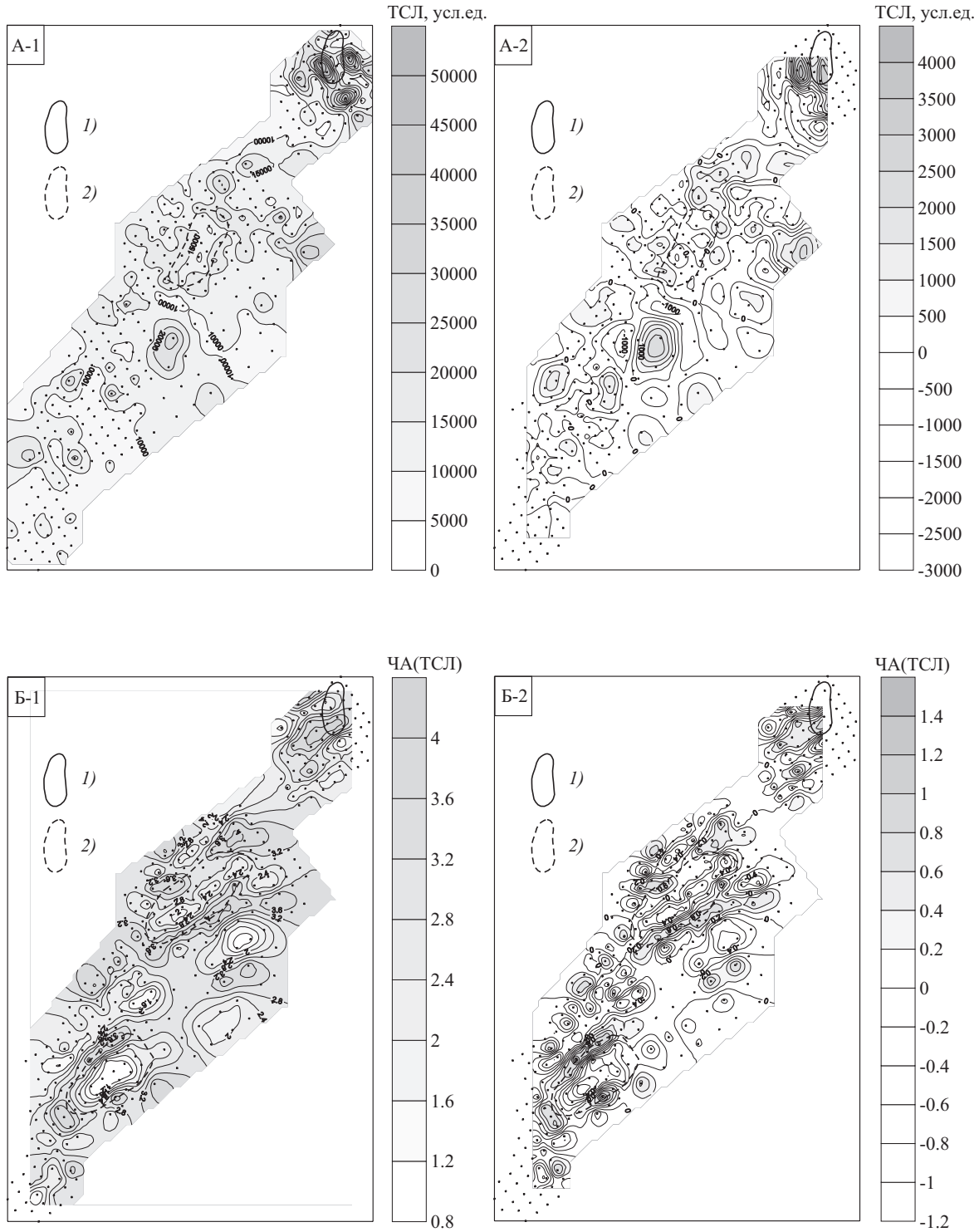

Fig. 1. *Fields of distribution of thermostimulated fluorescence (А-1), frequency and amplitude parameters (В-1) and their residual components (А-2, В-2 correspondingly) in the ground sediments of the Gulf of Ob: 1) Ob field; 2) boundary of perspective area*

accumulation level at oil-drainage boundary and splashes of anomalously high indices in the influence zones of water-hydrocarbon contact. Morphology of radio-geochemical fields above pools, localized in stratigraphic traps is more drawn towards linear and semiring anomalies [37].

Though, as practice shows, not all oil and gas fields are controlled by anomalies of radio-geochemical fields distractions, which are relatively easy to distinguish. Disclosure of epigenetic radio geochemical anomalies is practically impossible when analyzing only construction of NRE accumulation fields, in oil and gas bearing fields with variegated lithofacies solution of sediment.

To reduce interfering factors, various procedures of mathematical treatment of radio-geochemical data are used. Most frequently and effectively, to distinguish «naphthagenious» radio-geochemical anomalies, one uses examining of local constituent of radio geochemical fields, received by subtraction measured radio-geochemical parameters from the regional background, which was counted as averaging mathematical window or regression analysis. Then amendments are introduced into lithologic solution of horizon testing, soil horizon structure, landscape condition [5, 12, 25, 38].

Indices, evaluating the character of interconnection between NRE are highly informative.

DRAD index was used on group of USA oil bearing objects, during oil-drainage boundary definition, it was directed at the revelation of areas with anomalous divergence of meanings of selective uranium and potassium canals from «ideal» meanings of these elements, normalized to thorium equivalent [16].

To define the areas of imposed redeposit of U, the most labile from radionuclides, one can normalize its concentration according to sediments, characterizing its lithologic structure comparatively to inert thorium or alumina [21, 22, 39].

One of the most stable oil-and-gas content's characteristic is a structural failure of correlation between NRE group [40]. There is a tendency to divergence from background data of pair correlation for oil fields, mainly, because of U redistribution, U and K behave more chaotic in gas fields.

During radioactive data interpretation, as it seems to us, not sufficient attention is paid to heterogeneity of radioactive fields. A number of works about gas-geochemical, litho-geochemical, magnetometer, aerogramme-spectrometric researches of oil-and-gas bearing region, bring out clearly, that heightened «tension» of geochemical and geophysical fields is one of the peculiar features of the productive regions [20, 40–42].

As an example of a good informativeness of the analysis of the heterogeneity level of radioactive fields, we can cite the results of interpretation of thermoluminescence measuring of ground soil in the Gulf of Ob's south part. During hydrocarbon survey («Pangeya») the ground sediments samples selection was carried out, hydrocarbon component content was defined right up to C_{20}. Moreover, the sediment samples were handed to TPU to study the thermoluminescense. In the Fig. 1 two variants of mathematical processing of thermoluminescense study results are presented.

In the first case, the source field of thermoluminescence distribution (TFD) was smoothed through averaging in mathematical window, the size is 1,5×1,5 km with further residual component allocation.

At the second case, the same algorithm was used during mathematical processing of field of frequency and amplitude parameters (FAP) of change. These frequency and amplitude parameter brings information about extremum quantity per unit area, taking into account changes amplitude of analyzed parameter.

As a result, «reference» field and group of halo anomalies, which are classified as perspective for hydrocarbon pools' detection, prove more clearly in the character of field irregularity of ground sediments' thermostimulated fluorescence.

Thus, today there are perfect prerequisites for wider usage of radio-geochemical researches during oil and gas field ground search. As any other method, radio-geochemical survey has some restrictions and there are under masking factors' influence. Nevertheless, progress in the sphere of radio-geochemical fields measuring means' development, systematic data about approaches to interpretation of radio-geochemical data, constantly accumulative information about radio-geochemical peculiarities of sediments on the section from hydrocarbon pools to day, all these allow us to be optimistic concerning the future of radio-geochemical methods in the complex of oil and gas exploration work.

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