DISCUSSION ARTICLE

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Factors of gas accumulations formation in oil-bearing sediments and in casing annulus of wells

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Abstract. Gas component study is one of the important tasks of petroleum geology. Gas component can exist in various forms in sedimentary rocks. Of great interest is nitrogen, the gaseous accumulations of which are formed in oil-bearing strata, causing complications during the oilfield development. The problem of abnormal nitrogen accumulations had great relevance in the fields of the Volga-Ural petroleum province, which is one of the long-term developed with a large stock of wells for various purposes.

This article discusses possible sources of gaseous nitrogen and the reasons for its accumulations in oil-bearing reservoirs. The main purpose of the article is to clarify the reasons for the gaseous nitrogen and its deposits formation. The main patterns of the areal distribution of nitrogen gas accumulations in oil-bearing strata are revealed on the basis of field, hydrogeological, geological and geophysical researches data analysis. It has been established that during the gas caps formation, the source of gaseous nitrogen is its dissolved compounds in groundwater and oil, biochemical decomposition of which leads to the dissolved molecular nitrogen accumulation in a liquid medium.

The release of free gaseous nitrogen and the formation of its accumulations is associated with the decompression of formation waters for natural (geological) or man-made reasons (hydrocarbons extraction). Disturbance of the natural hydrodynamic regime in oil-bearing formations leads to the release of gaseous nitrogen and the formation of its accumulations under favorable conditions (the presence of reservoirs, structures and impermeable rocks in the top of the formation).

Key words: Gases, nitrogen, oil-bearing strata, dissolved forms, organic matter, microbial activity

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Introduction

Gases are a natural component of oil and gas basins, where they occur in dissolved form in groundwater and oil. The main gases are hydrocarbons (methane, propane, isobutane, etc.), hydrogen sulfide, hydrogen, carbon dioxide, as well as nitrogen, argon, and helium (Zorkin, 1973, 2008). They have a different origin and haracterized by different concentrations, ratios, distribution patterns. Study of the gas component speciation and composition in oil-bearing formation is an important geological task due to the production of associated petroleum gas (Subbota et al., 1980). The gaseous (free) nitrogen accumulations is stand more interesting to study as the result of the hydrocarbon development intensification. Gaseous (free) nitrogen accumulations are specific to many Paleozoic oil and gas basins with evaporite formations, chloride-calcium brines, and high-sulfur oils (Trunova, 2005; Tikhomirov, 2014). Free gases are contained in voids under high pressures and release during formation drilling (Tikhomirov, 2014).

In a large number of gaseous nitrogen accumulations (deposits) are found within the Volga-Ural petroleum province (Republic of Tatarstan). The problem of gaseous nitrogen deposits drilling is relevant. The study area is an old, long-term developed oil-producing region with large stock of wells of different types.

Gaseous nitrogen accumulations presence can create significant difficulties during oil exploration. Nitrogen production during well drilling often results into complications and emergencies, has a negative impact on technological processes, deteriorating the

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clogging technology of gas producing intervals, creates problems in the utilization of poor associated petroleum gas (APG).

In this work, we consider gas caps placement patterns, nitrogen-bearing formations' zonal distribution in productive sediments of Middle and Lower Carboniferous at the territory of South Tatar arch. Possible reasons for their formation in order to develop methodology of a nitrogen deposits identifying and localizing are considered.

Object and research methods

Intensive nitrogen occurrence, which are gaseous nitrogen accumulations (deposits), were identified during drilling of the Nalimovskoye anticline (Novo-Elkhovskoye oilfield). The study area is located in the southeast parts of the Republic of Tatarstan (the east of the European part of Russia) (Figure 1). The Novo-Elkhovskoye oilfield is located on the western slope of the South Tatar arch and is controlled by a same named positive structure associating with the Novo-Elkhovskoye block of the crystalline basement. The Novo-Elkhovsky basement block is bounded by the Kuzaikinsky (from the west) and Altunino-Shunaksky (from the east) deep faults of submeridional strike (Geology of Tatarstan ..., 2003; Voitovich, 1998). These faults expressed as Kuzaikinsky and Altunino-Shunksky depressions in the structure of the sedimentary cover. These depressions outline the Aktashsko-Novo-Elkhovsky swell, which is complicated by positive oil-bearing structures of the third order – uplifts. Novo-Elkhovskoye field's geological structure is presented by Precambrian, Devonian, Carboniferous, Permian, Neogene and Quaternary formations. The oil-bearing capacity of the Novo-Elkhovskoye field includes terrigenous and carbonate deposits of the Upper Devonian, carbonate and terrigenous strata of rocks of the Lower Carboniferous, mainly carbonate deposits of the Middle Carboniferous. The oil-bearing capacity of the Novo-Elkhovskoye field includes terrigenous and carbonate formations of the Upper Devonian, Lower Carboniferous and predominantly carbonate formations of the Middle Carboniferous. The main productive layers are the Kynovian and Pashian horizons of the Lower Frasnian substage of Devonian. Middle and Lower Carboniferous formations, despite there are oil-bearing, are poorly studied so far. In the same time, structural and tectonic, mining-geological and hydrogeological features of the Lower and Middle Carboniferous formations have great importance for the oilfield development. The structural plan of the Lower Carboniferous formations is more dislocated in comparison with the Devonian formations. Devonian top includes a large number of oil-bearing uplifts with an amplitude of 60–70 m, separated by third-order depression network. These depressions are zones of increased permeability. Various gases occurrence, the most important of which is nitrogen, are noted in this formations. According to drilling data, a lot of gas accumulations of nitrogen are detected in oil-bearing formations of the Devonian and Carboniferous.

This article discusses nitrogen gas accumulations and deposits in the oil-bearing formations of the

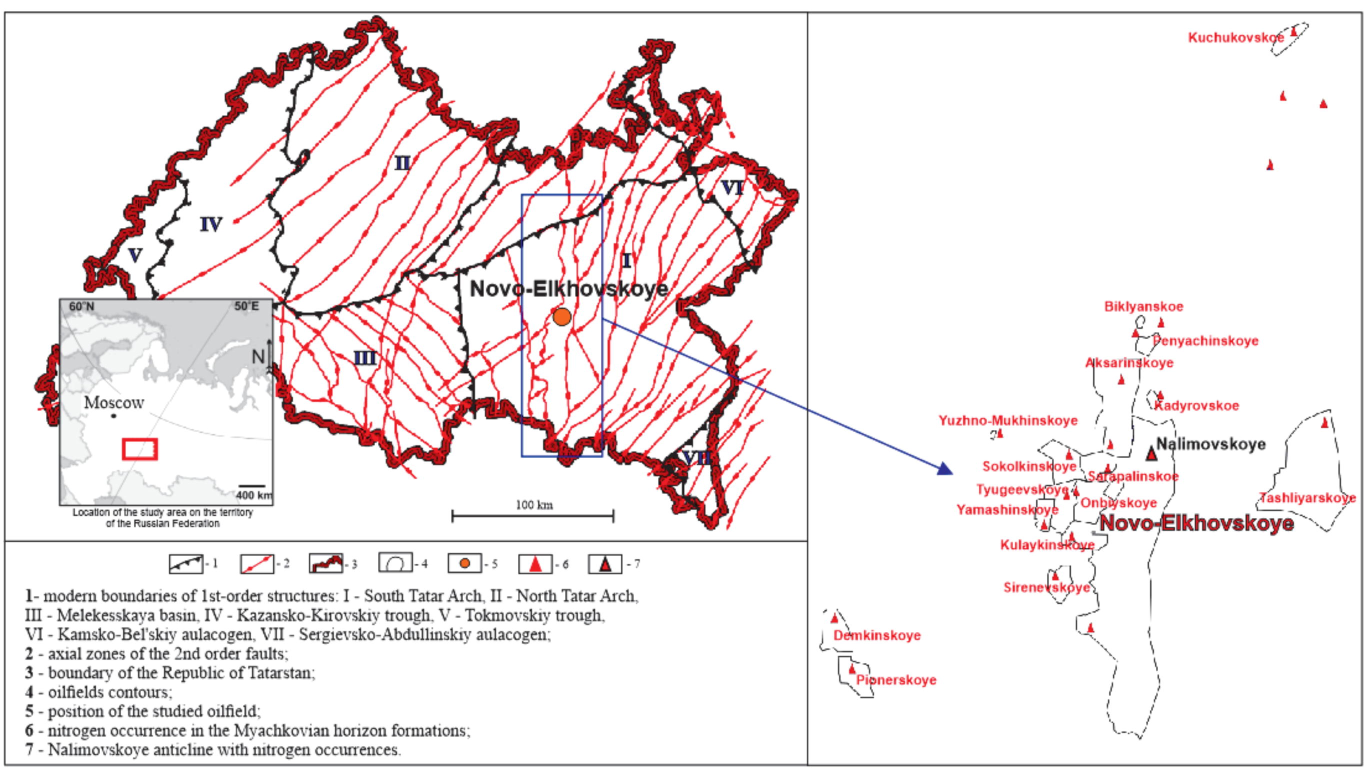

Fig. 1. Overview scheme of nitrogen occurrences on the western slope of the South Tatar arch

Nalimovskoyey anticline (Figure 1), where they represented by the Moscovian stage formations. Moscovian stage at the study area includes Vereyian, Kashirian, Podolskian, Myachkovian horizons. Gas accumulations are represented by almost pure nitrogen with a small admixture of hydrocarbon gases (the nitrogen content in the gas mixture exceeds 90%).

Vereyian horizon's formations are oil-bearing and have in this oilfield thickness up to 50 meters and are composed of alternating terrigenous (siltstones, mudstones) and carbonate (limestones, dolomites, marls) rocks (Geology of Tatarstan ...,2003). Gray bioclastic limestones with interlayers of clayey limestones, clays, siltstones are located in the lower part of the horizon. Rocks have enhanced reservoir properties and are oil-bearing. Seven strata are distinguished (from bottom to top: C_2vr-1 – C_2vr-7) in Vereyian horizon, but commercial oil-bearing capacity is associated only with C_2vr-2, C_2vr-3 strata. Other strata in the area are composed of low-capacity varieties (clays and carbonate rocks) and cannot be considered as reservoir strata. The Kashirian horizon is composed of bioclastic limestones and dolomites. The Podolskian horizon is represented by microgranular and fine-grained, dense dolomites and limestones with interlayers of mudstones. Uneven-grained limestones and dolomites with inclusions of gypsum and anhydrite nests and with layers of mudstone take part in the composition of the Myachkovian horizon.

The main purpose of the work is to clarify the reasons for the nitrogen-gas accumulations formation on the basis of their lateral distribution analysis in productive reservoirs of Carboniferous (Vereyian horizon), connection with the tectonic structure of the territory and the content of dissolved nitrogen in oil and formation water.

Some regional patterns of the geological and structural-tectonic features of the study area and their possible impact on the formation of strata containing nitrogen in the form of gas accumulations and deposits are analyzed by authors in order to identify the features of the nitrogen areal distribution and abnormal accumulation.

Research and discussion result

Based on the drilling results, it was found that gaseous nitrogen accumulations are connected with positive structures of the Nalimovskoye anticline. Nitrogen occurrence zones of the Moscovian stage deposits are localized in the depth range of 450–650 m and are unevenly distributed over the section. Their maximum occurrence is associated with the Vereyian and Myachkovian formations. In the Vereyian horizon gaseous nitrogen are found in the C_2vr-2, C_2vr-3, C_2vr-4, and C_2vr-5 formations, where their intensity increases up the section. Gas accumulations are localized in the domes of positive oil-bearing structures. Gas accumulations contain mainly mixture of hydrocarbon gases with a predominance of nitrogen. According to the Elkhovneft Petroleum Production Department (PJSC «Tatneft») data, the nitrogen content in the gas mixture varies from 84.7 to 93.7% and increases up the section. The gases accumulation in the dome of the structure leads to the gas-oil contact formation and the oil rim displacement. Development of the deposit's central part is complicated due to this reasons and only the marginal zone development is possible.

At the same time, it should be noted that the largest number of nitrogen occurrences is associated with the Myachkovian horizon formations, where they are in large quantities. The geological conditions and nitrogen gas accumulations areal distribution are considered to clarify their nature.

Comprehensive analysis of factors that could have significant influence on the nitrogen gas accumulations formation, showed that the structural and tectonic factor has the greatest impact. South Tatar arch sedimentary cover's structural plan inherits block structure of the crystalline basement (Voytovich, 1998; Larochkina, 1993), due to the deep faults series superposition. Faults have two orthogonal directions: 1) northwest – northeast and 2) sublatitudinal – submeridional.

The gas accumulations' spatial distribution depends on the tectonic structure of the study area. The relationship between the location of nitrogen occurrences (red triangles) and the fractured zones of the crystalline basement is quite well according to the map (Figure 2).

Nitrogen occurrence are observed almost along the entire Kuzaikinsky depression (fault) and partly along the northern part of the Altunino-Shunaksky depression (fault) within the Novo-Elkhovsky structure. This is reasonable if we take into account that the faults are high fracturing zones with high permeability. This is confirmed by signs of hydrothermal activity in crystalline basement faults (Khasanov et al., 2017). We can assume the possibility of vertical migration of nitrogen and its compounds in the composition of formation fluids. According to (Hunt, 1982), the source of nitrogen is basement rocks degasification, which may be evidenced by the presence of helium in natural gas. In this case, nitrogen occurrences are located either outside zones of active vertical fluid exchange, or on their periphery. It should be mentioned that the Altunino-Shunaksky depression zone is geodynamically more active, but does not contain nitrogen occurrences. This can be explained by the absence of favorable conditions for the gas accumulations formation (absence of appropriate structures and seals).

This assumption is partly confirmed by the lineaments map (Figure 3) analysis results. Lineaments map was constructed using digital topographic map analysis and

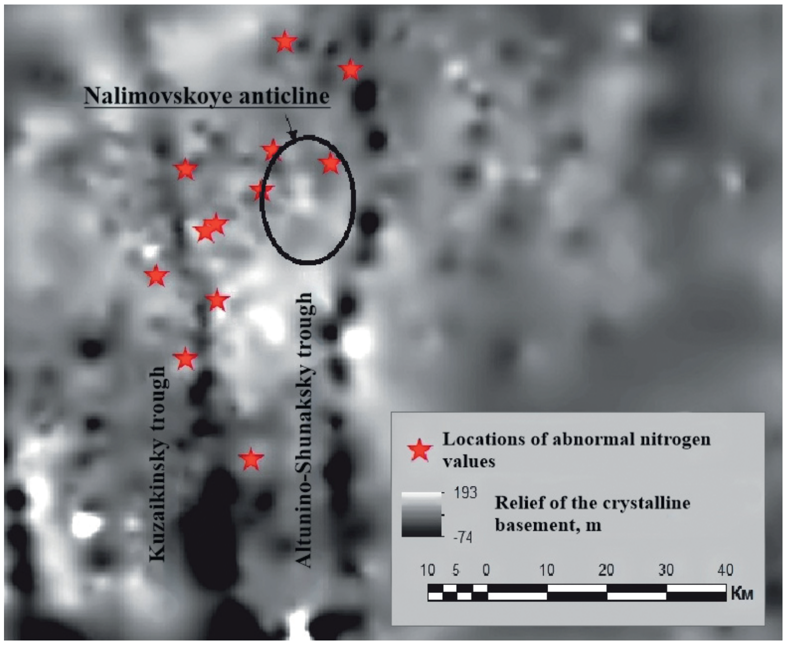

Fig. 2. Relief map of the crystalline basement on the western slope of the South Tatar arch

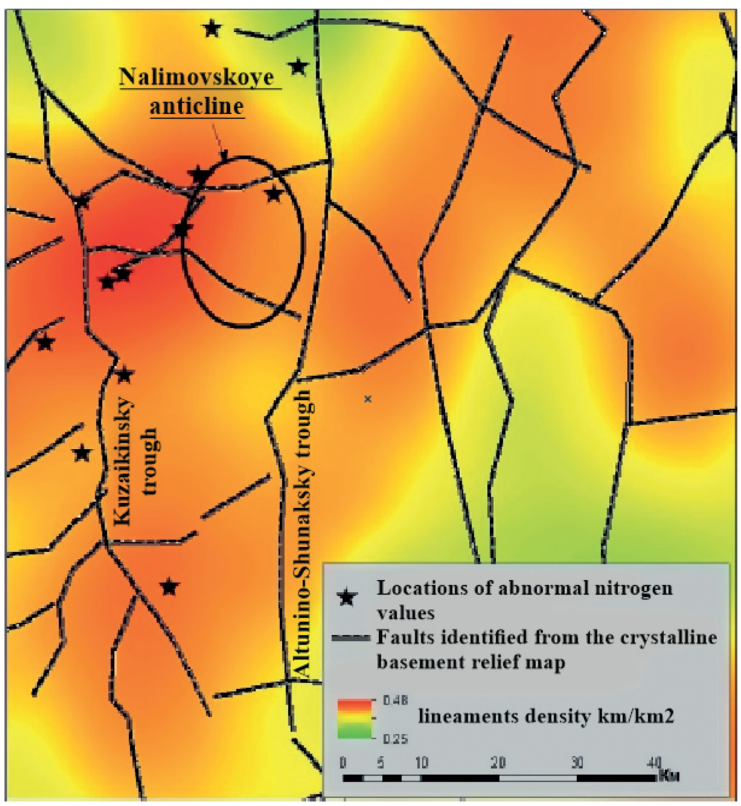

Fig. 3. Map of lineaments density on the western slope of the South Tatar arch

the geological interpretation of satellite data. This map reflects the macropermeability of the sedimentary cover. Map analysis allows sedimentary cover's increased or decreased permeability areas forecasting.

The map shows that almost all cases of nitrogen occurrence are associated with a high lineaments density areas. Therefore, formation of high nitrogen content zones requires the presence of subvertical conductive channels (cracks) and the presence of lateral aquifers in the upper structural layers of the sedimentary cover.

Relation of free nitrogen occurrences to fault zones indicates hydrodynamic regime violations and permeable sections formation. This sections makes possible gas migration and accumulation, if any seal rock present.

However, question about nitrogen sources remains open. In this regard, we will consider its content in fluid (oil, water) component of the oil-bearing complexes of the Novo-Elkhovskoye oilfield. Nitrogen is a common component of groundwater and oils in the Volga-Ural petroleum province (Khamidullin et al., 2000), where it presents as a dissolved one (N2). Its contents vary within 0.13–0.73 (wt.%) and within 10.46–26.42 (% mol) in the composition of one-time degassed gas, produced from Carboniferous oil-bearing formations within the Novo-Elkhovskoye oilfield. The gas content in the oils of the Vereyian horizon is 3–7 m^3/t at a reservoir temperature equal to 23°C.

Carboniferous groundwater also contains dissolved gases. In terms of chemical composition, the formation waters of the Vereyian horizon at the Novo-Elkhovskoye oilfield are classified as calcium chloride brines (Sulin, 1948) with a total dissolved salts content of 77–201 g/dm^3. However, waters with a total dissolved salts content of 19–124 g/dm^3 are widespread in the sediments of the same horizon, in some areas of the neighboring Romashkino oilfield. The groundwater gas saturation in the Vereyian terrigenous-carbonate formations within the South Tatar arch is 90–170 m^3/m^3 (water) (Khisamov et al., 2009). Dissolved gas has a predominantly nitrogen composition. The content of dissolved nitrogen in the studied formations groundwater varies from 50 to 150 m^3 per 1 m^3 of water (Khisamov et al., 2009). Nitrogen is characterized by low solubility, and such high levels of nitrogen in groundwater are due to high reservoir pressure. Reservoir pressure in groundwater of the Vereyian horizon at the Novo-Elkhovskoye field reaches 6.0–8.0 MPa.

Nitrogen is one of the most common chemical element on Earth. It occurs naturally in the composition of chemical compounds with other elements and in the gaseous state in the form of a diatomic N_2 molecule. The gaseous nitrogen content in atmosphere is more than 78%. In a dissolved state, nitrogen is present in all types of natural waters, where it is also presented in the form of ions of soluble chemical compounds or dissolved gas. Nitrogen exists in all types of natural waters, in the form of ions of soluble chemical compounds or dissolved gas. The main components are ammonium (NH_4), nitrate (NO_3), nitrite (NO_2) and molecular nitrogen (N_2) ions. Nitrogen in the form of N_2 is presented in gaseous dissolved form. The solubility of nitrogen in water is low and equals to 23.6 ml/l (29.5 mg/l) at 0°C and a partial pressure of 0.1 MPa. The nitrogen solubility decreases with an increase in temperature and increases with an increase in pressure. According to the Henry-Dalton law, at a constant temperature, the gas mixture's each component solubility in a liquid located under this mixture is proportional to their partial pressure (Glinka, 2003). Therefore, the gas dissolved in water will tend to come to equilibrium with the partial pressure of this gas in the atmosphere. Hence, the gas dissolved in groundwater tends to come to equilibrium with the pressure of this gas in the gas caps. The gas is absorbed by the water, when the gas content in the water is less than it is necessary to achieve equilibrium with the gas in the gaseous medium. Liquid degassing are observed at a high gas content.

Nitrogen in nature has a mixed biogenic and deep abiogenic origin (Lukin, Dontsov, 2009). In the groundwater of the earth's crust's upper horizons, nitrogen is mainly of atmospheric origin, and its composition depends on the partial pressure of nitrogen in the atmospheric air. Abiogenic (mantle) origin nitrogen comes to the surface in areas of recent volcanism, from where it can get into the groundwater. Nitrogen has a heterogeneous nature in groundwater of oil-bearing regions (nitrogen released from sediments during their compaction, atmospheric nitrogen, biogenic nitrogen formed during the decomposition of organic matter). However, due to the groundwater composition features, a lot amount of nitrogen is biogenic, the formation of which is associated with the decomposition of both organic matter of sedimentary rocks and dissolved petroleum fractions.

Let's consider the nitrogen occurrence forms in groundwater of oil-bearing deposits in order to understand the origin and the concentration reasons of dissolved nitrogen in groundwater. As mentioned earlier, nitrogen in natural waters is present in the form of ammonium ions (NH_4), nitrates (NO_3), nitrites (NO_2) and molecular nitrogen (N_2). They both can get into composition of underground waters as part of the earth's crust's upper horizons water, and can form in situ. If nitrogen forms in situ, the nitrogen source in petroleum basins groundwater is mainly dispersed organic matter (OM) of sedimentary rocks, coal seams (Littke et al., 1995) and the sedimentary rocks themselves. Its content in sedimentary rocks reaches 170–1200 ppm (Zorkin, 1973, 2008).

Significant quantities of coal resources in Tatarstan associated with the Lower Carboniferous formations (Visean stage). Furthermore, dissolved petroleum matter can be a source of nitrogen in oil-bearing formations. Oil-bearing formation's groundwater is strong calcium-chlorine brines that lead to the organic molecules destruction, including protein ones, which mainly include nitrogen atoms. Bacterium are involved in organic molecules decomposition processes. Bacterium live at oil-water contact. Such bacteria can use various types of hydrocarbons, as well as nitrogen-containing organic substances for their vital activity. As a result of complex chemical and biochemical decomposition, the content of normal alkanes decreases (an increase in the isobutane/butane ratio), and the composition of nitrogen-containing organic substances is decomposed to ammonia (NH_3) and ammonium (NH_4), which are oxidized to nitrates and nitrites (Freedman, 1995). This process is carried out by nitrifying bacteria in two stages. One group of bacteria decomposes ammonia to nitrite, while another oxidizes nitrite to nitrate. The products of nitrification (NO_3^-) and (NO_2^-) are further subjected to denitrification. The denitrification process also occurs as a result of the denitrifying bacteria activity, which live in an oxygen-free environment. These bacteria have the ability to take oxygen from nitrates, reducing nitrate via nitrite to nitrous oxide gas (N_2O) and nitrogen gas (N_2) (Skiba, 2008).

In addition, the gaseous nitrogen formation can be associated with the anammox process (anaerobic oxidation of ammonium), which is also carried out by bacterial communities (Reimann et al., 2005). During anammox, nitrite ion and ammonium ion are converted directly to molecular nitrogen (Strous et al, 1999): $NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O$. Anammox has been described in hot springs at temperatures of 36–52°C (Andrea Jaeschke et al., 2009) and in hydrothermal vent along the Mid-Atlantic Ridge at temperatures of 60–85°C (Byrne et al., 2009). These conditions are close to the Vereyian horizon formation waters conditions, the salinity of which is about 77–201 g/dm^3 at temperatures of 23–25°C.

It should be noted that the Novo-Elkhovskoye oilfield has been developed over a long period of time. Surface fresh water and also prepared waste water were injected into reservoirs to stimulate oil production. These waters contain foreign microbiota and dissolved oxygen, which significantly changes both the composition of bacterial strains and the physicochemical and thermodynamic properties of groundwater. Therefore, their gas composition changes too (Khisamov et al., 2009). In particular, waters total dissolved salts content decrease to the level of saline (within the Romashkino field, values up to 19 g/dm^3 are noted). It can be assumed that locally changed conditions are favorable for the various microbial processes development, including nitrification and anammox. These processes lead to the formation of water-dissolved gaseous nitrogen (N_2) in the oil-bearing formations groundwater. A high concentration of nitrogen in groundwater and in the composition of associated petroleum gases within the Volga-Ural petroleum basin may be connected mainly with a high content of protein in organic matter (Zorkin, 1973, 2008). The content of dissolved ammonium NH_4^+ in groundwater of the Vereyian horizon within the Novo-Elkhovskoye field is 72.9–90.6 mg/dm^3 (Khisamov et al., 2009).

Thus, a nitrogen source for the formation nitrogen gas caps (nitrogen content in the gas mixture is 84–93%) are formation waters, where the molecular nitrogen formation is associated with organic matter molecules decomposition. High concentrations of dissolved nitrogen in groundwater cannot be explained by its input from the underlying horizons of the earth's crust (Hunt, 1982) along the fault zones due to the fact that the magnitude of reservoir pressures in undisturbed layers is higher than in fault zones.

The release of gaseous nitrogen from groundwater and its accumulation in the form of separate deposits and caps is associated with the decompression of formation waters. In zones of reduced pressure, dissolved nitrogen tends to equilibrium with environmental pressure.

The reasons for the low pressure local areas occurrence can be *geological* and *man-made*.

The main geological reasons are formation of faults in the earth's crust, subsidence of more dense mineralized solutions (in saline basins), etc. The tectonic factor controls the natural gas deposits location. Fault zones are permeable areas of the earth's crust where a decrease of hydrostatic pressure can occur. Groundwater degasification occurs in the low pressure zones with the release of excess gases quantities. Most of the gases are removed to the atmosphere, however some part may be retained in natural traps (presence of reservoir rocks, positive structures, seals) and form gas caps and accumulations. Most of the nitrogen gas accumulations are associated with the positive structures of the Myachkovian horizon, in the top of which there are low-permeable carbonate complexes of the Upper Carboniferous, represented by dense dolomitized limestones and dolomites with interlayers of anhydrite. Low-capacity rocks of the C_2vr-6 and C_2vr-7 strata can be seals during the formation of gas accumulations in the Vereyian horizon.

Man-made causes are associated with the disruption of the natural hydrodynamic regime in oil-bearing reservoirs during the oilfields development. It has been empirically established that gaseous nitrogen release is observed in zones of intense drilling mud loss, where the fluid level in the wellbore does not provide sufficient backpressure on the gas-bearing formation. Thus, the development of the reservoir leads to the formation of a reduced pressure zone in the radius of well influence, where formation waters degasification occurs. Gas component floating in a liquid medium leads to the gas accumulations and deposits formation. At the same time, the gas pressure in gas caps and accumulations will tend to the equilibrium state with the liquid and will be proportional to the groundwater pressure of corresponding layers. The released nitrogen gas, if appropriate conditions are present, can form accumulations and caps in the formation. Nitrogen is accumulated in the behind-the-casing space of wells and with associated petroleum gas is discharged into the oil gathering system.

Conclusions

As a result of the research it was found that the gas accumulations of nitrogen identified by drilling are connected with the fault zones, where its migration and localization in the void space of rocks conditions exist. The main source of nitrogen for the formation of gas accumulations are its dissolved forms in the liquid medium of oil-bearing formations (groundwater, oil). Isolation of nitrogen gas from groundwater and its accumulation in the form of separate deposits and caps occurs as a result of formation waters decompression due to the natural (geological) or man-made reasons (development of oil fields).

The nitrogen gas deposits formation, in which the nitrogen content reaches more than 90%, occurs under favorable conditions (the presence of reservoirs, structures and seals). Within the Nalimovskoye anticline, they are confined to the positive structures of the Myachkovian horizon, overlain by seals in the form of low-permeable carbonate complexes of the Upper Carboniferous. Low-capacity rocks of the C_2vr-6 and C_2vr-7 strata can be seals during the formation of gas accumulations in the Vereyian horizon. The nitrogen gas accumulations localization in the domed areas of positive oil-bearing structures above the oil reservoir leads to the deposits formation.

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