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Analysis of Water–Gas System Equilibria in Jurassic–Cretaceous Reservoirs (by the Example of the Yamal-Kara Depression)

D.A. Novikov ✉

Trofimuk Institute of Petroleum Geology and Geophysics, Siberian Branch of the Russian Academy of Sciences, prosp. Akademika Koptyuga 3, Novosibirsk, 630090, Russia

Novosibirsk State University, ul. Pirogova 1, Novosibirsk, 630090, Russia

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Abstract—The paper presents results of the pioneering study of equilibria in the water–gas system by the example of Jurassic–Cretaceous deposits of the Yamal–Kara depression located within northern West Siberia and its Arctic regions. Numerical modeling of physicochemical equilibria and evasion–invasion processes in the water–gas system allowed determination of the degree of groundwater saturation with gases and the nature of diffusive redistribution of gases in the media that form at the hydrocarbon deposit–groundwater contact. According to the degree of water saturation with gases (K_g), aquifers with poorly (<0.2) to ultimately (0.8–1.0) gas-saturated waters have been established. The revealed increase in the degree of groundwater saturation with gases in sinking producing reservoirs reflects its dependence on their total gas saturation. All waters with a total gas saturation of more than 1.8 L/L become ultimately saturated with gases (K_g = 1.0), thus theoretically predetermining the formation of hydrocarbon accumulations. Major gas condensate deposits are confined to the zone of gas-saturated waters with K_g from 0.8 to 1.0, while oil accumulations, to waters with lower gas saturation. Based on the established nature of water–gas, we can argue that oil and gas accumulations in Jurassic–Cretaceous reservoirs act as a conservative element of the lithosphere, i.e., its geologic and geochemical "relics". The surrounding subsurface waters, as a more active constituent of the system, have largely anticipated its geochemical development, which is manifested in the differentiation of the fugacity ratios of individual gases in groundwater and hydrocarbon accumulations. The composition of the latter is therefore subjected to slow directional changes while the equilibrium is established, to usher in the qualitatively new state of the geochemical water–gas system.

Keywords: total gas saturation, water–gas system, equilibrium, modeling, degree of water saturation with gases, fugacity, hydrocarbon deposit, Yamal–Kara depression, West Siberia, Arctic

INTRODUCTION

The fundamental problem of studying the geochemical processes and physicochemical equilibria in the water–gas system is integrated into a general geological problem of oil and gas deposits formation and degradation, and therefore comes laden with a highly topical research area. It is generally recognized that recent discoveries of new oil and gas fields within the West Siberian sedimentary basin (WSSB) are increasingly involving novel approaches in addition to geophysical imaging and geological and geochemical information analyses. In this regard, the study of physicochemical equilibria in the water–gas system encapsulating the information about region-specific conditions of oil and gas fields formation and preservation, as well as about the processes of mass exchange between reservoirs and the surrounding formation waters, opens up a new expanse of research as part of geochemical prediction of oil and gas potential and evaluation of productivity of regional geologic structures and prospecting areas (Novikov, 2018).

Due to their exceptional mobility, gases produce the greatest halos resulting from their dispersion in sedimentary basins. This phenomenon accompanies diffusion interchange of gases between hydrocarbon (HC) deposits and formation waters surrounding them, thereby providing the most reliable search criteria (Namiot, 1958; Antonov, 1963; Kartsev et al., 1992; Qin et al., 2016; Shabani and Vilcáez, 2017). Results and findings of the calculations performed by Yu.P. Gattenberger, V.M. Matusevich and colleagues (Matusevich et al., 2005) suggest that " ... at the present stage of the WSSB development, the water percolation rate in its submerged part is characterized by negligible values even from the perspective of geological time, so that water infiltration, as it is, does not take place. Under these conditions, the largest extents of mass transfer and, in particular, the appearance of aqueous dispersion halos near oil and gas accumulations are associated with the diffusion processes" (Matusevich et al., 2005, p. 160). In this context, the role of inherent processes within the water–gas system increases dramatically.

✉ Corresponding author.
E-mail address: NovikovDA@ipgg.sbras.ru (D.A. Novikov)

The study of gases dissolved in water (or water-dissolved gases, WDG) in Western Siberia was commenced more than 50 years ago by M.S. Gurevich and N.N. Rostovtsev. In the early 1950s, they introduced the concept of gas zonality of groundwater in the West Siberian artesian basin (WSAB) and stressed the importance of WDG values (determined not only by qualitative aspects of their composition, but also by the saturation pressure) for hydrocarbon prospecting. N.M. Kruglikov for the first time recognized a decrease in saturation pressure of dissolved gases with their distance increasing from the gas-water contact, which can be explained by the diffusion phenomenon. L.M. Zorkin have studied different conditions for generation of gases, along with their separation from groundwater, to form gas accumulations. A.E. Kontorovich, B.P. Stavitsky, A.A. Rozin, N.M. Kruglikov, V.V. Nelyubin, O.N. Yakovlev, V.M. Matusevich, A.A. Kartsev, S.L. Shvartsev and many other researchers have long been studying groundwater in complex with water-dissolved gases (Kartsev, 1963; Kontorovich et al., 1975; Kruglikov et al., 1985; Kartsev et al., 1986, 1992; Nazarov, 2004; Matusevich et al., 2005).

However, despite the fact that the knowledge of the conditions of WDGs' physicochemical equilibria will enable solution of many genetic aspects of oil and gas-generation (Sokolov, 1971) and enable to reveal previously unknown patterns of groundwater formation (Shvartsev, 1996), with both being of great scientific and practical interest, such conditions remain largely underexplored. The water–gas equilibria provide evidence for revealing the geochemical processes orientation in the water–gas system both at the present stage of the petroleum system evolution and in its geological past, and also help to solve the problems of oil/gas deposits preservation, as well as predicting their phase type.

The two main tasks solved within the frames of the considered problem are: 1) evaluation of the degree of groundwater saturation with gases (K_g)[1] and 2) identification of the nature of interactions in the water–gas system (formation waters – hydrocarbon deposit).

In respect to the West Siberian petroleum province (WSPP), the first problem was addressed to in the works of N.M. Kruglikov, V.N. Kortsenshtein, V.V. Nelyubin, O.N. Yakovlev, M.Y. Rudkevich, L.S. Ozeranskaya, N.F. Chistyakova and others (Kruglikov, 1964; Kortsenshtein, 1977; Bars, 1978; Kruglikov et al., 1985; Rudkevich et al., 1988). Their results provided valuable data on variations in the degree of groundwater saturation with gases in different aquifer complexes within the lower hydrogeological tier (from the perspective of a tiered classification approach). These also indicated that the values of the gas saturation coefficient for the Jurassic complex vary from 0.03 in the marginal part of the basin to 1.00 in its middle. Within the Neocomian complex, K_g values tend to increase from the marginal part towards the basin's center (up to 0.77 in the Middle Ob region), with the maximum values (up to 0.94) revealed within the Tazovskaya area. Within the Apt–Albian–Cenomanian complex, K_g values explicably increase (from 0.02 to 0.87) from south to north of the WSPP.

In research works of N.M. Kruglikov and V.N. Kortsenshtein (Kruglikov, 1964; Kortsenshtein, 1977; Kruglikov et al., 1985) some results and findings are analyzed with the focus on the second problem, involving calculations of interactions in the formation water–hydrocarbon deposit system for Cenomanian reservoir PK_1 (Medvezhie, Yamburgskoe, Ety-Purovskoe, Urengoyskoe fields) and Neocomian reservoirs BU_{10}, BU_{12} (Urengoyskoe gas field). According to these authors, the unstable position (nonequilibrium) of investigated HC deposits in respect to the waters hosting them allows suggesting that in the Upper Cretaceous and Lower Cretaceous horizons processes of degradation are progressively taking place in the existing petroleum accumulations.

Given that each geological structure, in itself, is a unique natural object with unparalleled inherent characteristics of the hydrogeological structure, hydrodynamic conditions and vertical hydrogeochemical zonality, etc., it was therefore too early to infer about the degradation processes occurring in oil/gas accumulations that formed in the Aptian-Albian-Cenomanian and Neocomian oil and gas-bearing complexes, relying on calculation results for discrete reservoirs alone.

In this regard, an in-depth study of the nature of equilibria in the water–gas system using the available unique hydrogeochemical data and oilfield hydrogeology research and physicochemical modeling methods has become highly topical for Jurassic–Cretaceous deposits in the northern West Siberia and its Arctic regions.

FACTUAL MATERIALS AND RESEARCH METODOLOGY

The water–gas system is known to be fairly complicated, which is explained by the multicomponent and multidirectional processes involved, which makes it problematic to use the previously applied simplified methods for calculating the degree of saturation of formation waters with gases, individual fugitive gas emissions (hereinafter, fugitivities), etc. Specifically, calculations for this study were performed using the HG-32 (Hydrogeo) software developed by M.B. Bukaty at Trofimuk Institute of Petroleum Geology and Geophysics, which is presently qualified as unmatched in the world, inasmuch as it allows to take into account all parameters of the studied system (density, total salinity (TDS) and chemical composition of formation waters, gas saturation, WDG composition, thermobaric conditions, etc.) in the modeling; to determine the composition and some characteristics of free gas-phase equilibria from the composition of WDG and water, or conversely, to derive the composition and other parameters of WDG from the compositions of free gas and solution; as well as to simulate gas evasion-invasion processes under changing pressure, tem-

[1] The degree of groundwater saturation with gases (K_g) is interpreted as a ratio between the gas-saturation pressure and formation pressure.

perature and composition of the solution (Bukaty, 1992, 1999).

The HG-32 (Hydrogeo) software complex is based on dependencies of the law of mass action and activity method. In this case, for each component i of n components of a gas mixture dissolved in groundwater, a reversible reaction of the transition from solution to the free-phase state is considered

$$i_{\text{д}} \Leftrightarrow i_g \,. \tag{1}$$

Mass-action law can be written for it as

$$K_i = f_i / a_i^0 \,, \tag{2}$$

where K_i is the thermodynamic equilibrium constant, f_i is the fugitivity (volatility) of free gas phase, a_i^0 is its free phase activity in the equilibrium solution (saturated with this gas) in the standard state.

The degree of aqueous solution saturation with gas i is estimated using the undersaturation index

$$L_i = \frac{a_i K_i}{f_i} \,, \tag{3}$$

where a_i is gas activity in the investigated water.

The most common approach consists in direct determination of gas fugitivity F_i for each gas i in a hypothetical free gas phase equilibrium with the solution, using semi-empirical methods proposed by A.Yu. Namiot (1991), E.S. Barkan et al. (1998). At this,

$$F_i \equiv a_i K_i \text{ and } L_i = \frac{F_i}{f_i} \,. \tag{4}$$

To calculate f_i and K_i, a number of correction coefficients and a series of regression equations resulted from the experimental data processing (obtained by M.B. Bukaty) that describe the required thermodynamic parameters, solubility and phase distributions of gases depending on the composition of each and the real thermobaric conditions.

The patterns of gases redistribution between petroleum accumulations and formation waters were estimated by the ratio of individual fugitivities of gases calculated for the two systems: HC accumulation-formation water and formation water-reservoir. In the former case, the hypothetical equilibrium composition of WDGs was calculated from the free gas phase composition, whereas in the latter case, the composition of hypothetical gas deposit was calculated from WDGs. Calculations of the degree of saturation of formation waters with gases (K_g), individual degrees of formation waters saturation with gases and individual fugitives of gases, estimated characteristics, etc. were performed for more than 400 gas deposits within the Cretaceous and Jurassic intervals and localized within 52 fields (exploration areas) in the WSSB's northern and Arctic regions (Fig. 1). This article is largely underpinned by the results of testing of a total of >2800 objects in 127 prospecting area, as well as by materials of the integrated chemical analysis (including microcomponents) conducted on samples from groundwaters (>5600), and from water-dissolved (>2500) and free gas (>1900) accumulations. The hydrogeochemical data and water-dissolved gases (WDGs) and free gases (FGs) compositions summarized in this work have been collected by the author over a long period (since 1997) from test holes at production sites of Glavtyumengeologiya (subordinate facilities: Urengoyneftegazgeologiya, Yamalneftegazgeologiya, Purneftegazgeologiya, etc). Presently, these materials are stored in the IPGG SB RAS data repository. These allowed the most exhaustive study of Cretaceous reservoirs.

RESULTS AND DISCUSSION

The degree of groundwater saturation with gases (K_g)

The region-specific hydrogeological conditions and hydrogeochemistry of oil and gas-bearing deposits of the Yamal–Kara depression have been extensively studied by O.V. Ravdonikas, A.E. Kontorovich, B.P. Stavitsky, Yu.G. Zimin, N.M. Kruglikov, V.V. Nelyubin, O.N. Yakovlev, E.G. Bro, G.A. Ivanova, G.D. Ginsburg, I.N. Ushatinsky, V.M. Matusevich, A.A. Kartsev, A.D. Duchkov, A.R. Kurchikov, S.L. Shvartsev, D.A. Novikov and many other researchers since the 1960s (Ravdonikas, 1962; Kruglikov, 1964; Zimin et al., 1967; Kontorovich and Zimin, 1968; Ushatinsky and Matusevich, 1970; Ginsburg and Ivanova, 1971, 1974; Ivanova and Melkanovitskaya, 1973; Duchkov and Sokolova, 1974; Kontorovich et al., 1975; Bro, 1977; Kruglikov et al., 1985; Kartsev et al., 1986, 1992; Balobaev et al., 1987; Kurchikov and Stavitskii, 1987; Kurchikov, 1992; Shvartsev and Novikov, 2004; Stavitsky et al., 2004; Matusevich et al., 2005; Novikov and Lepokurov, 2005; Kokh and Novikov, 2014; Novikov and Sukhorukova, 2015; Novikov, 2017a,b, 2018a,b; Kazanenkov et al., 2019).

Cretaceous and Jurassic reservoirs of the Yamal–Kara depression are characterized by wide spread high and abnormally high reservoir pressures, whose nature is associated with the drive of elision water-head systems (Novikov et al., 2018; Novikov, 2019).

Groundwaters identified within the studied geological structures are characterized by varied chemical compositions. However, all the water-bearing complexes (aquifers) are dominated by subsurface waters of sodium chloride, sodium chloride-bicarbonate and sodium bicarbonate-chloride types (after S.A. Shchukarev (Samarina, 1977)) with TDS varying from 2–5 g/dm^3 in the peripheral areas to 63.3 g/dm^3 in the basin's middle part (Shvartsev and Novikov, 2004; Stavitsky et al., 2004). Waters with the highest salinities are confined to the Upper Jurassic aquifer complex (Table 1) (Novikov, 2017a; Novikov et al., 2019). Each chemical type has its own characteristics reflected in distributions of major salt-forming macro-and micro-components, whose concentrations have a strong correlation with their TDS values. The salinity growth entails an increase in amounts of chlorine, sodium, magnesium, calcium, potassium, as well as micro-

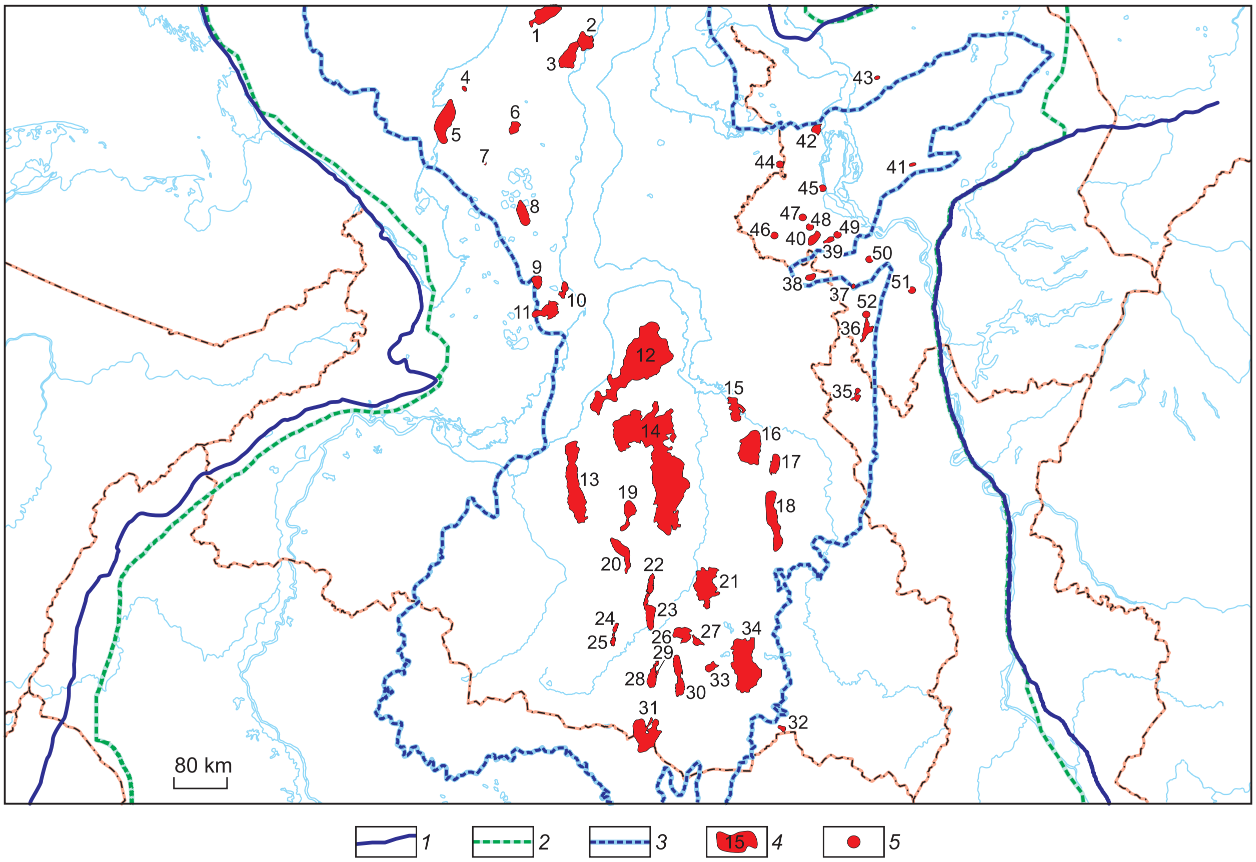

Fig. 1. Geographical location of the studied objects within the Yamal–Kara depression. Boundaries: *1*, West Siberian sedimentary basin; *2*, Jurassic sedimentary basin; *3*, Yamal–Kara depression; Oil/gas fields and exploration areas: *4*, oil/gas fields: 1, Malyginskoe; 2, Tasiyskoe, 3; West-Tarkosalinskoe; 4, Kharatskoe; 5, Kruzenshternskoe; 6, Verkhne-Tiuteyskoe; 7, Nerstinskoe; 8, Arkticheskoe; 9, Nurminskoe; 10, Khambateyskoe; 11, Rostovtcevskoe; 12, Yamburgskoe; 13, Medvezhie; 14, Urengoyskoe; 15, Tazovskoe; 16, Zapolyarnoe; 17, Russkoe; 18, Yuzhno-Russkoe; 19, Yubileynoe; 20, Yamsoveyskoe; 21, East-Tarkosalinskoe; 22, Severo-Gubkinskoe; 23, Gubkinskoe; 24, Komsomolskoe; 25, Barsukovskoe; 26, Tarasovskoe; 27, Ust-Kharampurskoe; 28, Vyngayakhskoe; 29, Vostochno-Vyngayakhskoe; 30, Yety-Purovskoe; 31, Vynga-Purovskoe; 32, Udmurskoe; 33, Yuzhno-Tarkosalinskoe; 34, Kharampurskoe; 35, Lodochnoe; 36, Suzunskoe; 37, Gorchinskoe; 38, Yuzhno-Soleninskoe; 39, Ushakovskoe; 40, Pelyatkinskoe; 41, Ozernoe; 42, Deryabinskoe; 43, Khabeyskoe; *5*, Exploration areas: 44, Tanamskaya; 45, South-Noskovskaya; 46, Yarovskaya; 47, Sredne-Yarovskaya; 48, Anomalnaya; 49, Turkovskaya; 50, Semenovskaya; 51, Bolshe-Laidinskaya; 52, Tokachinskaya.

components (bromine, iodine, boron, ammonium and strontium). At salinity levels of 15–20 g/dm^3 and more, concentration of bicarbonate ions (hydrogencarbonate ion) tends to decrease. Sulfate ion concentrations (on average, not more than 20–60 mg/dm^3) are largely dictated by the process of its reducing to hydrogen sulfide at the "oozy" stage.

The pattern of groundwater saturation with gas is highly differentiated within the lower hydrogeological tier of the Yamal–Kara depression and can show two-fold change (sometimes even greater) within one reservoir, while there is a generally persisting increasing trend with depth from 0.3–3.0 L/L in Aptian–Albian–Cenomanian to 0.9–5.7 in the Lower-Middle Jurassic complexes (Kruglikov et al., 1985; Shvartsev and Novikov, 2004). In the Jurassic–Cretaceous aquifers, groundwater methane concentrations reach 95.5 vol.% in the Aptian–Albian–Cenomanian complex to 83.3 vol.% in Lower-Middle Jurassic aquifers. Methane concentrations tend to decrease with depth in the sinking aquifers. Whereas methane homologues increase with depth: from 1.3 vol.% in Aptian–Albian–Cenomanian to 11.7 vol.% in Lower-Middle Jurassic complexes. These aquifer complexes are characterized by an increase in carbon dioxide levels accompanied by a decrease in the ratio of total amount of heavy hydrocarbons to nitrogen: from 96 vol.% (Aptian–Albian–Cenomanian) to 52 vol.% (Lower-Middle Jurassic). Accordingly, the amounts of major components are not more than 15 vol.% for nitrogen, 4 vol.% for carbon dioxide, 6 vol.% for hydrogen, 0.14 vol.% for helium and 0.19 vol.% for argon.

Results of the detailed calculations have shown a complicated and uneven pattern of groundwaters saturation with gases illustrated by the established horizons (according to the K_g value) within all the studied aquifers whose waters are saturated with gases to varying degree: from low (<0.2) to ultimately high (0.8–1.0) (Fig. 2). Its variation across major aquifers is considered below in greater detail.

Table 1. Hydrogeochemical characteristics of aquifers

Indices	Measurement unit	Aquifers				
		Aptian–Albian–Cenomanian	Neocomian	Upper Jurassic	Lower Jurassic	Pre-Jurassic
pH	–	6.1–8.7 (7.6)	6.0–9.3 (7.6)	6.2–9.3 (7.5)	6.4–9.5 (8.1)	7.6-8.9 (8.6)
HCO_3^-	mg/dm^3	31–4882 (737)	84–5490 (929)	12–3709 (833)	37–3477 (1347)	94–3184
SO_4^{2-}	mg/dm^3	1–165 (23)	1–198 (45)	1–248 (33)	1–272 (38)	4–213 (44)
Cl^-	g/dm^3	0.3–15.1 (7.8)	0.3–36.8 (6.3)	1.8–37.6 (11.9)	1.5–39.6 (7.6)	1.1–28.4 (6.2)
Br^-	mg/dm^3	0.5–70.3 (27.9)	0.5–231.8 (28.6)	6.7–207.9 (40.6)	9.5–214.9 (32.1)	2.0–106.0 (30.0)
I^-	mg/dm^3	0.4–29.9 (7.8)	0.2–114.7 (7.8)	0.4–45.0 (5.2)	0.3–135.9 (8.1)	0.5–40.6 (8.5)
F^-	mg/dm^3	0.1–10.0 (2.1)	0.1–12.0 (2.4)	0.3–8.9 (1.2)	0.3–6.0 (1.8)	0.5–3.9 (2.5)
Na^+	g/dm^3	0.3–10.0 (4.6)	0.3–20.6 (4.0)	0.4–24.5 (6.3)	0.3–23.5 (4.8)	0.3–16.2 (4.3)
Ca^{2+}	mg/dm^3	4–1672 (316)	5–3406 (393)	8–3250 (502)	2–3110 (263)	8–1640 (207)
Mg^{2+}	mg/dm^3	2–383 (78)	1–680 (29)	1–547 (62)	1–350 (34)	2–170 (32)
K^+	mg/dm^3	4–425 (43)	3–690 (60)	3–502 (114)	5–840 (110)	10–380 (108)
NH_4^+	mg/dm^3	0.1–39.2 (14.3)	0.2–90.0 (16.5)	0.2–150.0 (39.4)	3.0–112.5 (23.1)	7.5–18.0 (11.4)
SiO_2	mg/dm^3	0.9–74.0 (19.2)	1.0–115.0 (33.8)	3.0–86.0 (25.8)	5.0–130.0 (36.6)	7.0–85.0 (35.3)
B^+	mg/dm^3	0.2–39.4 (6.9)	0.2–87.3 (11.3)	0.3–200.0 (10.1)	0.5–75.0 (9.9)	2.6–107.5 (11.1)
Sr^{2+}	mg/dm^3	0.6–200.0 (31.1)	0.4–290.0 (56.6)	1.6–450.0 (154.6)	1.2–290 (77.6)	–
Napth. acids	mg/dm^3	0.1–5.8 (0.8)	0.1–8.1 (0.6)	0.1–4.0 (0.6)	0.1–9.3 (1.0)	0.2–2.6 (0.8)
TDC	g/dm^3	1.5–25.3 (13.8)	2.0–53.0 (11.6)	2.0–63.3 (19.9)	2.0–53.1 (14.5)	2.5–46.8 (12.0)
rNa/rCl	–	0.42–4.28 (1.08)	0.26–4.79 (1.16)	0.52–2.28 (1.00)	0.47–3.99 (1.23)	0.86–1.91 (1.16)
Cl/Br	–	70–1923 (247)	12–1970 (234)	52–547 (264)	27–1641 (255)	94–724 (253)
Ca/Cl	–	0.01–0.54 (0.04)	0.01–0.38 (0.06)	0.01–0.80 (0.05)	0.01–0.57 (0.04)	0.01–0.27 (0.04)
B/Br	–	0.01–4.25 (0.36)	0.01–9.79 (0.66)	0.02–6.23 (0.36)	0.01–3.44 (0.50)	0.04–3.13 (0.80)
Chemical type after S.A. Shchukarev	–	Sodium chloride, chloride sodium (with anomalously high calcium concentration), chloride–bicarbonate sodium, bicarbonate–chloride sodium				
Number of analyses	pcs.	1127	3378	644	381	73

Note: maximum values (arithmetic mean).

The degree of groundwater saturation with gases in the ***Aptian–Albian–Cenomanian aquifer complex*** was calculated on the example of the Arkticheskoe, Gubkinskoe, Polyarnoe, Medvezhie and a number of other fields. Thus, reservoir PK_1 known to be holdsing the unique Cenomanian gas deposits, is dominated by gas-saturated waters with the K_g values varying between from 0.80 and 1.00. The waters are predominantly sodium chloride, with TDS up to 20 g/dm^3 and gas-saturation 1.5–2.5 L/L. The WDGs composition is ubiquitously methane-dominated, with its concentration averaging ca. 98.5 vol.%. The amounts of other gases except nitrogen (0.5–1.5 vol.%) are ranked as minor (i.e., even lower levels). In the direction from geological structures of the Yamal–Kara depression towards the WSSB's marginal parts, the total saturation of groundwater with gas along with the K_g values have significantly reduced (0.1–0.5 and 0.05–0.2 L/L, respectively), while the composition of gases has changed to methane-nitrogen and nitrogen-methane.

In the lower horizons, the situation looks strikingly different: the central regions of the Yamal–Kara depression encompassing the Medvezhie, Urengoyskoe, Vynga-Purovskoe and other fields are dominated by groundwaters with K_g value varying between 0.8 and 1.0. At this, waters weakly saturated with gases were revealed in a number of exploration areas, e.g. chloride-sodium waters with low salinity (3.3–6.3 g/dm^3) in reservoir PK_{15} (*Udmurtskoe* field). WDGs have either methane (96.3–97.2 vol.%) or nitrogen (2.6–3.4 vol.%)-dominated composition. The total groundwater saturation with gases within the reservoir varies significantly (from 0.3 to 1.5 L/L), while the K_g value ranges from 0.08 to 0.10. The most exhaustively studied is the *Kharampurskoe* field (reservoirs PK_{13}, PK_{14}, PK_{15} and PK_{16}). The waters hosted in them are chloride sodium with TDS varying from 10.7–10.9 (PK_{13}, PK_{15}) to 15.0–17.8 (PK_{14}, PK_{16}) are widespread. WDGs of these beds are generally methane-dominated with methane concentration reaching 97.4–97.9 vol.% (PK_{13}) to 84.3–90.0 vol.% (PK_{16}), and nitrogen concentrations not exceeding 2.2 vol.%, while down the section the proportion of methane explicably decreases against the backdrop of its homologs increasing to 1.5–5.0 vol.% (PK_{16}). Noteworthy is also that gas-saturation of groundwaters increases with depth from 1.0–2.2 to 1.5–

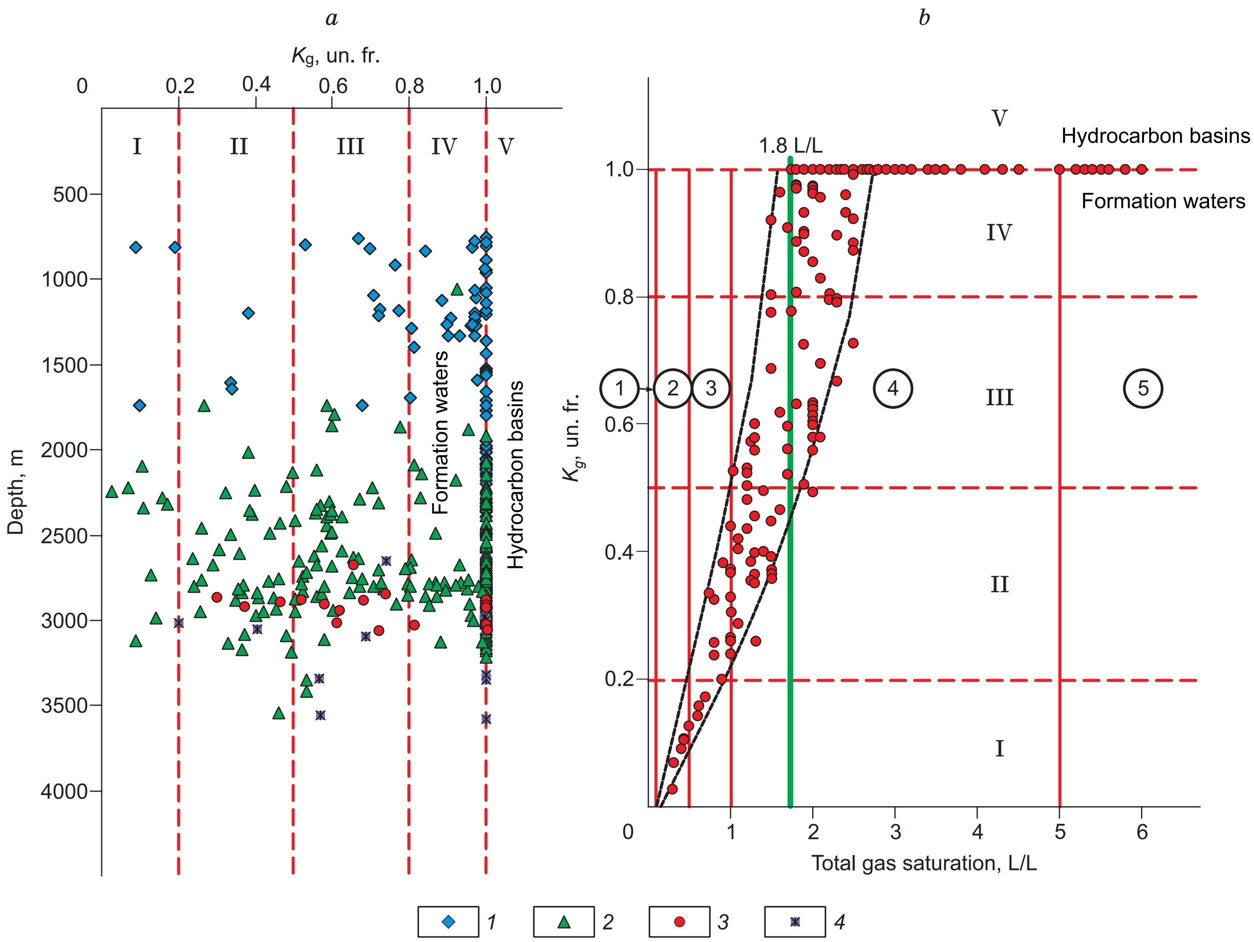

Fig. 2. Variation of coefficient of groundwaters saturation with gases (K_g) with depth (*a*) and versus total gas saturation of subsurface waters (*b*). a) Gas saturation zones determined from the coefficient of groundwaters saturation with gases as: I, low; II, moderate; III, high; IV, ultimately saturated; V, hydrocarbon accumulation; coefficient of waters saturation with gas (K_g) in major aquifers: *1*, Aptian–Albian–Cenomanian, *2*, Neocomian, *3*, Upper Jurassic, *4*, Lower–Jurassic. b) Gas saturation zones determined from the total gas saturation of subsurface waters (L/L): 1, very low (<0.1); 2, low (0.1–0.5); 3, moderate (0.5–1); 4, high (1–5); 5, very high (>5).

3.0 L/L. The value of K_g varies significantly (from 0.34 to 1.00), with its minima confined to reservoirs PK_{13} (0.34–0.98) and PK_{15} (0.33–0.34), and maxima – to PK_{16} (0.68–1.00), from which, when tested by well No. 339 in the 1798–1808 m depth interval, the obtained gas inflow flow rates (depending on the diameter of the fitting) vary in from 451.4 to 584.6 thou. m^3/d. In the studied reservoirs of the Aptian–Albian–Cenomanian complex in the adjacent areas of the Yenisei-Khatanga basin, the value of K_g at the Ozernoe and Pelyatkinskoe fields equals 0.71–0.73 and 0.53–0.70, respectively, and does not exceed 0.19 at the Nerstinskoe field (Yamal Peninsula).

Given that major oil reserves are confined to the ***Neocomian aquifer complex***, this appears to be the most extensively studied and is therefore represented by a wealth of actual material. The degree of groundwater saturation with gases was calculated for reservoirs from the Barsukovskoe, Vostochno-Tarkosalinskoe, Gubkinskoe, Deryabinskoe, Ety-urovskoe, West-Tarkosalinskoe, Komsomolskoe, Ust-Kharampurskoe and many other oil/gas fields. The evolutionary history of the aquifer complex encompassed the transgression to regression transition stage of the Late Jurassic-Cenomanian sedimentation cycle. Given the shallow-shelf environment, a sandy-clayey rhythmically layered sequence consequently formed, subdivided into sandy reservoirs persistent along the strike with multiple interbedding, and clayey impermeable beds. This formation is viewed as most favorable for the formation of multireservoir oil and gas fields. This is confirmed by the example of oil and gas fields mentioned above. The change in the degree of groundwater saturation with gases at some of them is discussed below.

Subsurface waters of the *Vostochno-Tarkosalinskoe* field are sodium-calcium chloride or sodium with salinity from 5 to 20 g/dm^3 and gas saturation varying widely from 0.6 to 3.9 L/L, averaging 2.4 L/L. WDG compositions are ubiquitously methane-dominated: CH_4 averages 84.4 vol.% , while concentrations of N_2 do not exceed 12, (on average, ca. 2 vol.%). All other gases except heavy hydrocarbons (HH) are contained in even lower amounts. The amounts of HH,

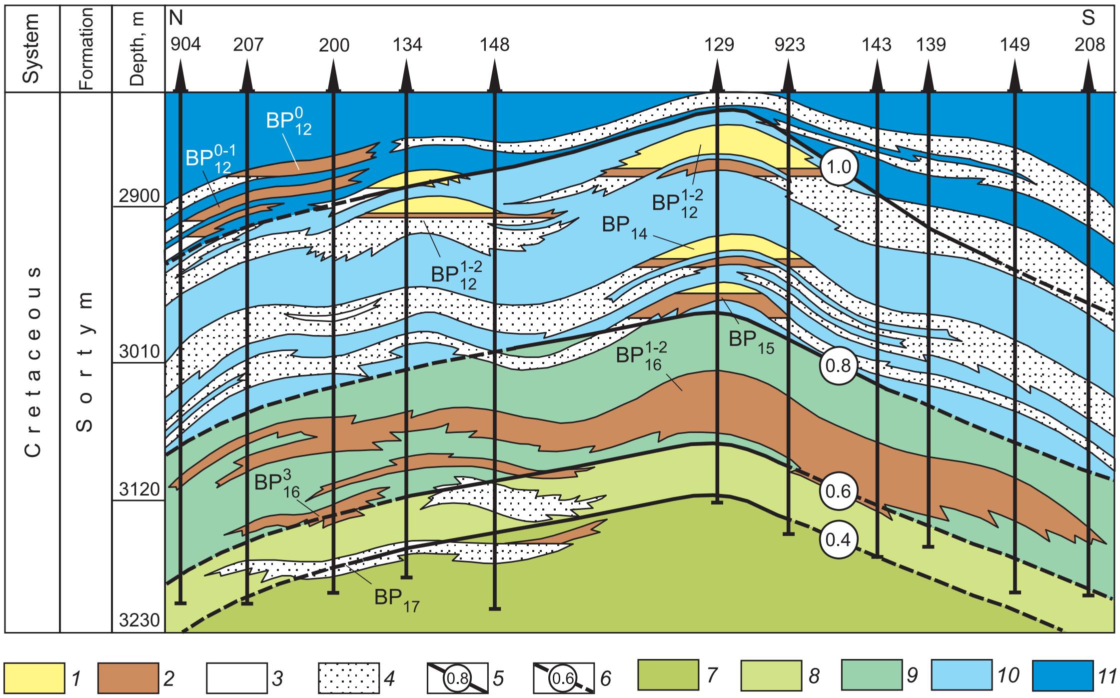

Fig. 3. The degree of groundwaters saturation with gases (derived from K_g values) in the producing interval of the Neocomian aquifer at the East-Tarkosalinskoe oil-gas-condensate field. Hydrocarbon accumulations: *1*, oil-gas-condensate; *2*, oil; *3*, predominantly mudstones and clays; *4*, sandstones; Isolines for K_g values: *5*, data-derived; *6*, putative; *7–11*, K_g value zones: *7*, less 0.4; *8*, 0.4–0.6; *9*, 0.6–0.8; *10*, 0.8–1.0; *11*, ultimaley gas-saturated.

as well as the total gas saturation, increase significantly with depth. The maximum groundwater saturation with gases was observed in reservoirs $BP_{11\text{-}12}$, where $K_g = 1.0$. Further down and up the section the total gas saturation tends to decrease. Thus, K_g equals 0.33–0.42 and 0.78 in beds labeled BP_{17} and BP_4, respectively.

The clearest picture is observed in the lower part of the producing strata of the Neocomian aquifer complex (Fig. 3). The degree of water saturation with gases gradually decreases from reservoirs with gas condensate deposits (region of K_g values: from 0.8 to 1.0) to reservoirs with entirely oil accumulations (region of K_g values: from 0.4 to 0.8). Thus, in reservoir BP_{12}, the coefficient of K_g varies from 0.95 to 1.00, i.e., saturation of water with gases has reached the maximum. The value of K_g equals 0.80–0.86 in reservoir BP_{14}, while in the underlying BP_{15} it varies from 0.74 to 0.83. Further down, the section is dominated by entirely oil reservoirs characterized by a lower degree of gas saturation. At this, a major oil reservoir BP_{16} is confined to the remarkably unsaturated zone localized in the region of K_g values from 0.56 to 0.75, whose minima (0.32–0.42) were observed in reservoir BP_{17}.

As it was noted earlier, the degree of subsurface waters saturation with gas also decreases in a region above reservoirs straddling $BP_{11\text{-}12}$. However, the gas saturation pattern is more complicated here: the locally observed reservoirs with waters fully saturated with gas ($K_g = 1.00$), against the background of waters generally under-saturated with gases (e.g. K_g averages 0.46 for reservoir BP_{10}). In the overlying BP_9, values the values of gas saturation vary from 0.23 to 1.00, averaging 0.73, while in reservoirs BP_6, BP_5 and BP_4, K_g values average 0.33, 0.99 and 0.78, respectively. A distinct relationship between the type of HC accumulation and the degree of total gas saturation of groundwater saturation with gases can be generally observed. Normally, gas condensate deposits are confined to zones with higher K_g values (from 0.8 to 1.0), while oil accumulations are associated with K_g values below 0.8 (Fig. 3).

Sodium chloride groundwaters with salinity varying from 4.3 to 28.5 g/dm^3 are widespread within Neocomian deposits of the *Zapadno-Tarkosalinskoe* field. Gas saturation of groundwater varies over a wide range from 1.0 to 5.5 L/L, averaging 3.1 L/L.WDGs are methane-dominated in composition, with CH_4 levels approaching ca. 78.1 vol.%, and N_2 not more than 16 (on average, ca. 4 vol.%). All other gases except heavy hydrocarbons are found in exceedingly minor amounts. Concentrations of heavy hydrocarbons, along with the total gas saturation, tend to significantly increase with depth. Unlike at the Vostochno-Tarkosalinskoe field, most of the studied reservoir rocks contain waters supersaturated with gases within the Zapadno-Tarkosalinskoe field (K_g values are close to 1.0).

Waters unsaturated with gases were reported from reservoirs BP_4 ($K_g = 0.5$) and BP_8 ($K_g = 0.37$). A zone of formation waters ultimately saturated with gases is established down the section of the field. Major high-yield reservoirs with gas condensate accumulations prove to have maximum values of K_g. Thus, the reported inflow rates (depending on the choke size) from gas condensate deposit confined to reservoir BP_6 were: 92.7–214.3 thou m^3/day for gas and 38.6–88.7 m^3/day for stable condensate. Given that formation waters gradually become ultimately saturated with gas here, the value K_g varies from 0.59 to 1.00 for this reservoir. Reservoir BP_7 sitting below is characterized by the presence of waters also highly saturated with gases (K_g = 1.00). It follows from the above that below reservoir BP_8 there is a zone characterized by groundwaters ultimately saturated with gases (K_g = 1.00). The K_g value therefore equals 1.00 throughout the entire BP_9 reservoir; and equals 0.58–1.00 for BP_{11} and 0.68–1.00 for BP_{12}; in BP_{16} 0.92 and in the Achimov sequence of sediments it varies from 0.29 to 1.00, averaging 0.79.

Thus, a clear relationship between the hydrocarbon phase and the degree of total saturation of groundwater with gases was revealed within the Vostochno-Tarkosalinskoye field. Gas condensate deposits are generally confined to hydrocarbon fields with elevated K_g values (in the range from 0.8 to 1.0). Given that the Zapadno-Tarkosalinskoye field is characterized by widespread waters saturated with gases, and, hence, by a greater intensity of the degassing processes in the formation waters at the present stage of the petroleum system evolution.

Sodium chloride groundwaters with salinity varying from 4.4 to 29.5 g/dm^3 were established in Neocomian reservoirs of the *Gubkinskoye* field. The degree of groundwater saturation with gas varies from 0.4 to 3.5 L/L. WDGs are methane-dominated, with its content varying from 61.0 to 92.3 vol.%, while nitrogen concentrations are in the range 1.4–18.2 vol.%. The calculations have revealed almost identical, as compared to the Zapadno-Tarkosalinskoye field, pattern of formation waters saturation with gases. As in the previous case, most of groundwaters of the Neocomian aquifer complex are found to be ultimately saturated with gases (K_g = 1). Reservoir BP_9 containing waters unsaturated with gases (K_g = 0.17) is distinguished against the backdrop of the saturated zone. Formation waters unsaturated with gases (K_g = 0.47) were revealed in BP_4 sitting above the saturated zone, likewise in the Zapadno-Tarkosalinskoe field located to the east (in the Zapadno-Tarkosalinskoe field, K_g = 0.5). The zone that includes formation waters saturated with gases measures more than 500 m in size and is confined to the interval straddling reservoirs from BP_6 through BP_{16}.

Sodium chloride groundwaters with TDS from 4.0 to 23.9 g/dm^3 were identified in Neocomian deposits of the *Yety-Purovskoye* oilfield. Their gas saturation varies widely from 0.3 to 5.2 L/L. WDGs are methane-dominated, with its amounts varying from 64.9 to 96.1 vol.% and nitrogen accounts from 0.6 to 23.6 vol.%. Methane homologs show an increasing trend with depth, along with the value of total gas saturation of groundwaters. The obtained results analysis revealed a large zone dominated by waters saturated with gases, which comprises the reservoirs labeled BP_3 to BP_{12}. Further down the section, the total gas saturation coefficient (K_g) tends to decrease, gradually reaching 0.53 in reservoir BP_{17}. Some traits of subsurface waters saturation with gases are highlighted below.

As it was noted above, within this field a large zone of waters ultimately saturated with gases has been identified. Against their backdrop, unsaturated waters were established in reservoirs BP_6 (K_g = 0.12–0.46) and BP_{10} (K_g = 0.56). In addition, a number of reservoirs marked by the presence of ultimately gas-saturated waters are characterized by a large variation in K_g values. The most representative reservoirs are: firstly, BP_7 where the degree of saturation of formation waters with gases varies from 0.32 to 1.00, averaging 0.69; BP_{11} (K_g = 0.36–1.00); BP_{12} showing the widest range of K_g values (from 0.22 to 1.00); these are succeeded by BP_{14} and BP_{17} with K_g values gradually decreasing from 0.8 to 0.53–0.54, respectively.

The *Ust-Kharampurskoe* field (the last among the targets studied in great detail) the from a hydrogeological standpoint is characterized by: chloride sodium formation waters with salinity varying from 4.6 to 22.0 g/dm^3; their gas saturation varies in the range from 0.5 to 5.0 L/L; WDGs are methane-dominated, with its concentration in the range from 69.0 to 94.4 vol.% and nitrogen from 2.5 to 11.4 vol.%. The amounts of methane homologs, as well as total gas saturation, increase significantly with depth. It was found that, likewise at the Ety-Purovskoe field, groundwaters in reservoirs BP_8, BP_9 and BP_{11} continue to be ultimately saturated with gases, with this zone confined to the interval spanning reservoirs from BP_8 to BP_{11}. Further, below reservoir BP_{11} the degree of formation waters saturation with gases decreases from 0.74–0.84 (BP_{12}) to 0.46 (BP_{17}).

In summary, it should be noted that saturation of formation waters with gases within the Neocomian aquifer complex of the Yamal–Kara depression is characterized by complicated and uneven pattern and generally follows the trend shown by the Aptian–Albian–Cenomanian hydrogeological complex: a decrease in the background K_g values in the direction from the basin's central part towards its periphery. The established direct relationship between the degree of saturation of formation waters with gases and their phase composition is corroborated by the fact that major gas-condensate deposits are confined to horizons, where the K_g values range from 0.8 to 1.0, while oil accumulations are confined to reservoirs with waters less saturated with gas.

The degree of groundwater saturation with gases in the ***Jurassic aquifer complexes*** were calculated using the data obtained from the Gubkinskoe, Deryabinskoe, Komsomolskoe, Malyginskoe, Kharampurskoe and other fields.

Results of the most detailed calculations performed for the *Kharampurskoe* field (Fig. 4) have shown that groundwaters within the producing Jurassic sediments are basically undersaturated with gases. Only waters confined to the top of the Tyumen Formation (i.e., the most deeply subsided) are interpreted as gas-saturated. The degree of saturation of groundwater with gases progressively increases with in-

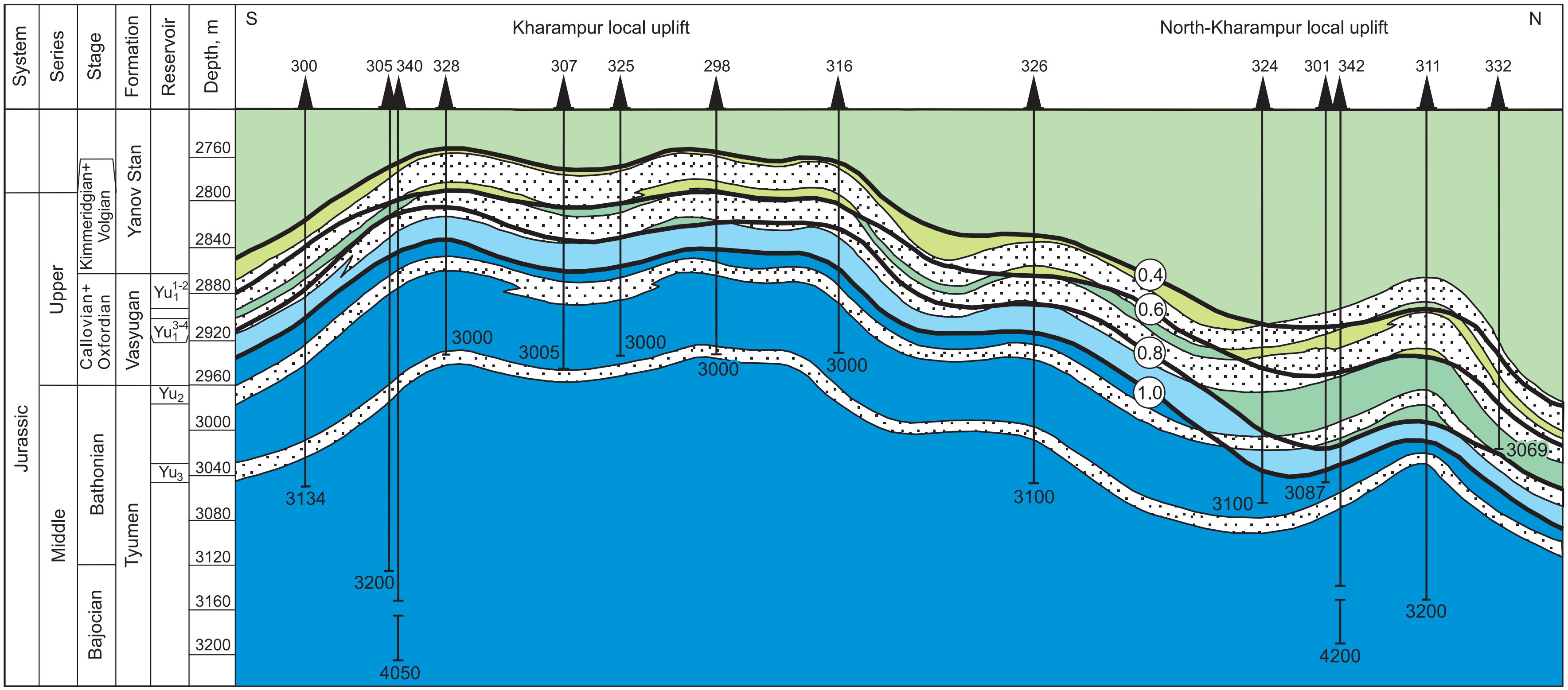

Fig. 4. The degree of groundwaters saturation with gases (derived from K_g values) in the producing interval of the Jurassic aquifer within the Kharampurskoe oil-gas-condensate field. 1, predominantly mudstones; 2, sandstones; 3, isolines for K_g values. For notation see Fig. 3.

creasing aquifers' occurrence depth. Thus, in the Upper Jurassic aquifer complex, the K_g coefficient for Yu_1^1 varies from 0.42 to 0.56 within the Kharampur local uplift area and from 0.31 to 0.55 within the North-Kharampur local uplift.

Reservoir Yu_1^2 is generally characterized by undersaturated groundwaters with gases within the Kharampur local uplift, where K_g = 0.45…0.75 (on average, 0.63). In the lower-occurring reservoir Yu_{13} (Kharampur local uplift) K_g values vary between 0.60 and 0.75, whereas their mean equals 0.55 in the North-Kharampur local uplift area. Reservoir Yu_{14} contains formation waters undersaturated with gas, which is corroborated by low K_g values (0.60–0.85). The Lower-Middle Jurassic complex (Yu_2) has a heterogeneous pattern of formation waters saturation with gases, which is illustrated by the laterally undersaturated (North Kharampur local uplift) succeeded by supersaturated waters with K_g = 1 (Kharampur local uplift).

Subsurface waters in reservoir Yu_3 are ubiquitously supersaturated with gases (K_g = 1) within the field, with K_g values generally increasing from 0.37–0.42 (2850–2910 m depth interval) to 1.00 (3000–3050 m). Isolines of the coefficient of total water saturation with gases (K_g) basically delineate the geological structure with localized permeable layers, i.e., showing a direct relationship between K_g values and structural framework of the field. This was also confirmed by the calculation results which revealed a correlation between K_g values and reservoir occurrence depth, which has shown a strong positive relationship between them (r = 0.84). There is also a strong correlation between the K_g coefficient and TDS of groundwater. Thus, if K_g is 0.37 at TDS 24 g/dm^3, then K_g = 1.00 at salinity level 42 g/dm^3. Salinities of formation waters saturated with gases were found to differ at TDS levels in excess of 38 g/dm^3, while all waters with lower salinities are undersaturated with gases. The correlation between K_g and TDS values showed a positive relationship between them (r = 0.56).

Thus, it can be stated that there are two zones – upper and lower – with varied patterns of groundwater saturation with gases within the producing interval of Jurassic deposits of the Kharampurskoe field (Fig. 4). The upper zone characterized by the presence of groundwater unsaturated with gases is dominantly extended in the deposits of the Vasyugan Formation (permeable layers of reservoir Yu_1). The only exception is the hydrogeological section confined to the North Kharampur local uplift, where the zone boundary descends lower encompassing deposits of reservoir Yu_2. It follows from the available data that groundwaters confined to the upper zone are capable of dissolving additional amounts of gases from the existing hydrocarbon accumulations, since the system is found to be nonequilibrium here. The lower zone, which includes groundwater saturated with gases, is confined to deposits of the upper Tyumen formation and embraces reservoirs Yu_2 and Yu_3, except the part of reservoir Yu_2 confining to the North Kharampur local uplift, where gases are likely to be emanated from formation waters into the free phase, if this process is viewed from the perspective of geological time.

The data obtained on the degree of groundwater saturation with gases at first glance appear to be in disagreement with the geological data. This can be accounted for the fact that oil accumulations with gas caps are confined to permeable layers of reservoir Yu_1 within the upper zone, where the water–gas system is found to be nonequilibrium. This contradiction can be explained neither by variations in temperature or general salinity, nor by pressure variations with depth, at the least because these parameters change slightly, while the values of K_g vary from 0.4 to 1.0, i.e., more than twice. Note that the formation of gas deposits occurred at the expense of contributions of water-dissolved gases, i.e., in the conditions of dissolved gases in equilibrium with free gases. Given that presently this equilibrium is non-existing, we can therefore justifiably assume that the formation of gas deposits was likely to have immediately followed by a change in geological settings, which was evidenced by the lost ability of subsurface water to emanate gases into the free phase, i.e., becoming thereby unsaturated with them, which we observe at the present time. Thus the question arises: what had caused this?

The answer to it should be sought in the established relationship between the total salinity and the degree of groundwater saturation with gas. The observed status quo can be explained by the fact that sedimentation waters in oil and gas-bearing deposits are partially diluted by ancient infiltration waters, which later penetrated into the analyzed system. This assumption is corroborated by the analysis of hydrogeochemical and hydrodynamic conditions in the region. Firstly, the analysis of relationships between total salinity of groundwater and Cl/Br ratio revealed its absence (which is possible, if saltier water is diluted with fresh water that have low Cl and Br concentrations affecting the Cl/Br ratio only slightly). Secondly, the analysis of region-specific hydrodynamic conditions found reservoir pressures to be lower in the southern part of the Kharampurskoe field, against its northern end, suggesting therefore groundwater migration through Jurassic sediments in a north-south direction. The northern part is also distinguished by lower salinities and gas saturation of groundwaters.

Thus, at the onset of the sedimentation processes in the permeable layers of reservoir Yu_1 groundwaters were supersaturated with gases, which prompted the formation of the free-gas zones. The change in the hydrogeological conditions caused by later ingression of infiltration waters unsaturated with gases, has led to the violation of gas equilibria. At this, the restrained contact between groundwater and gas accumulation precludes their rapid (from the perspective of geologic time) dissolution and thus contributes to sustainable nonequilibrium in the free gas-WDG system. Deeper-sitting horizons, where infiltration waters have not penetrated, remain saturated with gases at the present time. A new mechanism is thereby provided for assessing the possible extent of petroleum loss due to effects of groundwaters.

The calculation results of the degree of groundwater saturation with gases in the adjacent regions of the Yenisei-Khatanga basin (Deryabinskaya, Semenovskaya, Sredne-Yarovskaya, Turkovskaya and Ushakovskaya exploration areas) showed that the highest gas saturation have waters of reservoir Yu_2, where K_g values vary from 0.57 to 1.00. In the underlying layers of reservoir Yu_4 (Ushakovskaya area) and Yu_{17} (Semenovskaya area), the degree of groundwater saturation with gases is 0.57 and 0.74, respectively. On the Yamal Peninsula, within the group of layers bracketing reservoirs $Yu_{2\text{-}3}$ waters are also seen as ultimately saturated with gases (K_g = 1.00).

THE NATURE OF PHYSICOCHEMICAL EQUILIBRIA IN THE WATER–GAS SYSTEM

The study of the nature of equilibria in the water–gas system is largely based on the analysis of groundwater chemistry and gas composition, along with the data on the composition of free gas phase deposits and the well test results. Physical and chemical calculations performed in accordance with the above-described technique allowed to estimate the mechanism of their interaction with the host groundwater (Figs. 5–8).

Calculations for the ***Aptian–Albian–Cenomanian aquifer complex*** were performed on an example of hydrocarbon accumulations at the Arkticheskoe, Polyarnoe, Messoyakhskoe, Kruzenshternskoe, Malyginskoe, Tasiyskoe, Kharampurskoe and other fields. The calculation results have demonstrated significant distinctions in the process of gases redistribution between the studied reservoirs.

In northern West Siberia, reservoir PK_1 containing unique gas deposits of the Urengoyskoe, Medvezhie, Yamburgskoe and other gas fields has been studied most exhaustively, revealing thereby patterns of gases redistribution between gas deposits and invaded waters. Inasmuch as gas in reservoir PK_1 is mostly dry and contain no methane homologs, it was impossible to determine the nature of diffusive redistribution from heavy hydrocarbons. There was observed methane and carbon dioxide dispersion from almost all hydrocarbon accumulations, which was compensated by simultaneous contributions of helium, argon and nitrogen from groundwater. The equilibria were observed in gas-bearing reservoirs of the Komsomolskoe and Kruzenshternskoe fields (methane) and Nerchinskoe field (ethane). Analysis of relationships between individual fugitives of gases in HC accumulations and formation waters allowed an inference that giant gas deposits in reservoir PK_1 formed by ascending migration of hydrocarbons.

The process of methane, argon and carbon dioxide dispersion, i.e., dissolution into the surrounding groundwater has been established for hydrocarbon accumulations of the

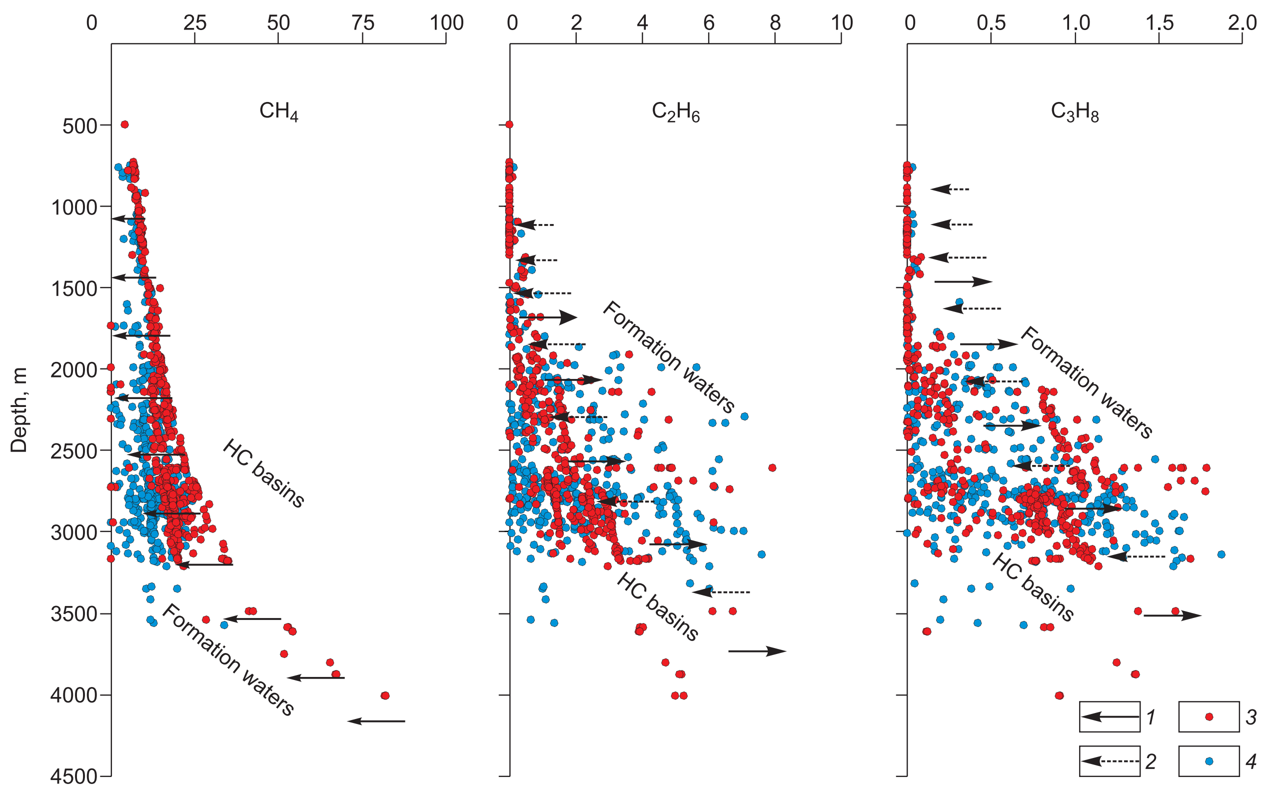

Fig. 5. Depth-dependent fugitivities of CH_4, C_2H_6 and C_3H_8 in formation waters and hydrocarbon accumulations. Direction of diffusion transfer: *1*, from HC accumulation into formation waters; *2*, from formation waters into HC accumulation; gas fugitivity: *3*, in the HC accumulation; *4*, in formation waters.

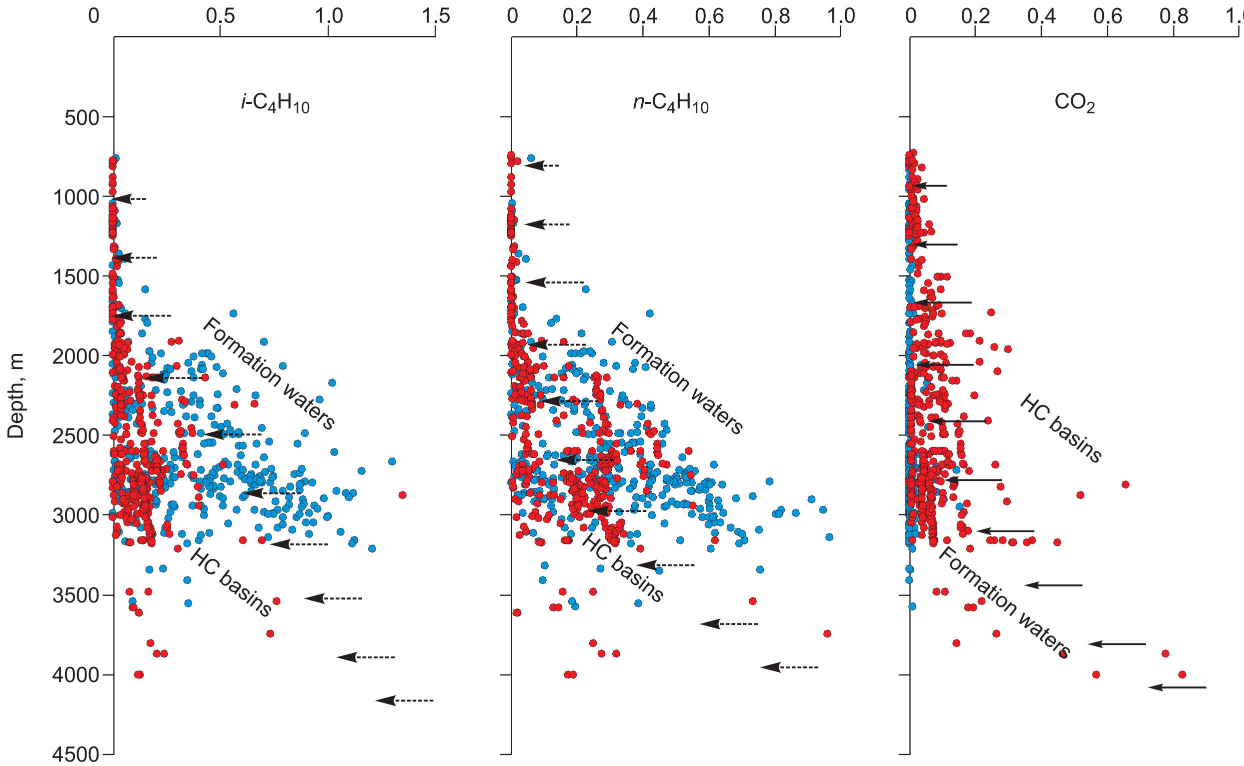

Fig. 6. Depth-dependent fugitivities of iC_4H_{10}, nC_4H_{10} and CO_2 in formation waters and hydrocarbon accumulations. For notations see Fig. 5.

Yamal Peninsula: Malyginskoe (HM_1, TP_1, TP_3, TP_6, TP_8), Kruzenshternskoe (TP_9, TP_{10}, TP_{13}), Nurminskoe (TP_3) and Tasiyskoe (TP_4^2, TP_5, TP_{11}, TP_{13}). Their dissolution is compensated by the simultaneous inflow of heavy hydrocarbons, helium and nitrogen from the peripheral formation waters. Methane and carbon dioxide dispersion, which is offset by simultaneous arrival of helium, argon and nitrogen from groundwater, is characteristic of almost all the studies fields. An equilibrium with respect to methane is observed in HC accumulations of the Komsomolskoe and Krusensternskoe fields, and with respect to ethane at the Nerchinskoe field. Relationships between individual volatile gases in HC accumulations and formation waters allow an inference that the formation of giant gas deposits in reservoir PK_1 owes to ascending migration of hydrocarbons. It is evident that the gaseous part of deposits is presently being transformed towards weighting of its composition, and changes in concentrations of non-hydrocarbon gases. This suggests that the processes of oil generation, migration and accumulation have continued until the modern stage of geochemical evolution of the water head system, against the backdrop of cessation of gas generation and accumulation.

Reservoirs PK_{13}, PK_{14}^1, PK_{15} and PK_{16} were studied within the Kharampurskoe field in the middle Nadym-Taz interfluve. The calculation results have revealed the mechanism of exchange between oil/gas accumulations and formation waters to be almost identical to the Yamal fields. The distinctions established in PK_{13} and PK_{15} which are also characterized by dispersion from ethane, propane and butane accumulations. The process of dispersion of all components of the gas cap into the surrounding formation waters (hydrocarbon and non-hydrocarbon components) was established in reservoir PK_{15} of the Udmurtskoe field.

A number of multireservoir fields of varied phase composition selected within the ***Neocomian aquifer complex*** of the study area were investigated to clarify the nature of equilibria between deposits and formation waters. These are Gubkinskoe, Zapadno- and Vostochno-Tarkosalinskoe, Komsomolskoe oil and gas condensate fields (Nadym-Taz interfluve); Yety-Purovskoe oil and gas field; Ust-Kharampurskoe and Yuzhno-Tarkosalinskoe oil fields; Malyginskoe and Khambateiskoye gas condensate field from Yamal, and Ozernoe gas field in the adjacent areas of the Yenisei-Khatanga regional trough, etc.

The most intriguing pattern of gases redistribution between HC deposits and formation waters was revealed within oil and gas condensate fields. Thus, in the Neocomian section of the Gubkinskoe field, a number of reservoirs ($AP_{9\text{-}10}$, $BP_{4\text{-}5}$, BP_9^1 and BP_9^2, BP_{15} and $BP_{16\text{-}21}$) can be distinguished, in which the gas component of the deposits was subjected to weighting (due to influx of methane homologs – ethane, propane, butane – from the surrounding formation waters), along with pentane and hexane in reservoirs $AP_{9\text{-}10}$, $BP_9{}^2$ and $BP_{16\text{-}21}$. Enrichment of HC accumulations with helium and nitrogen is observed almost in all reservoirs of this field, except BP_7 and BP_9. As such, the redistribution

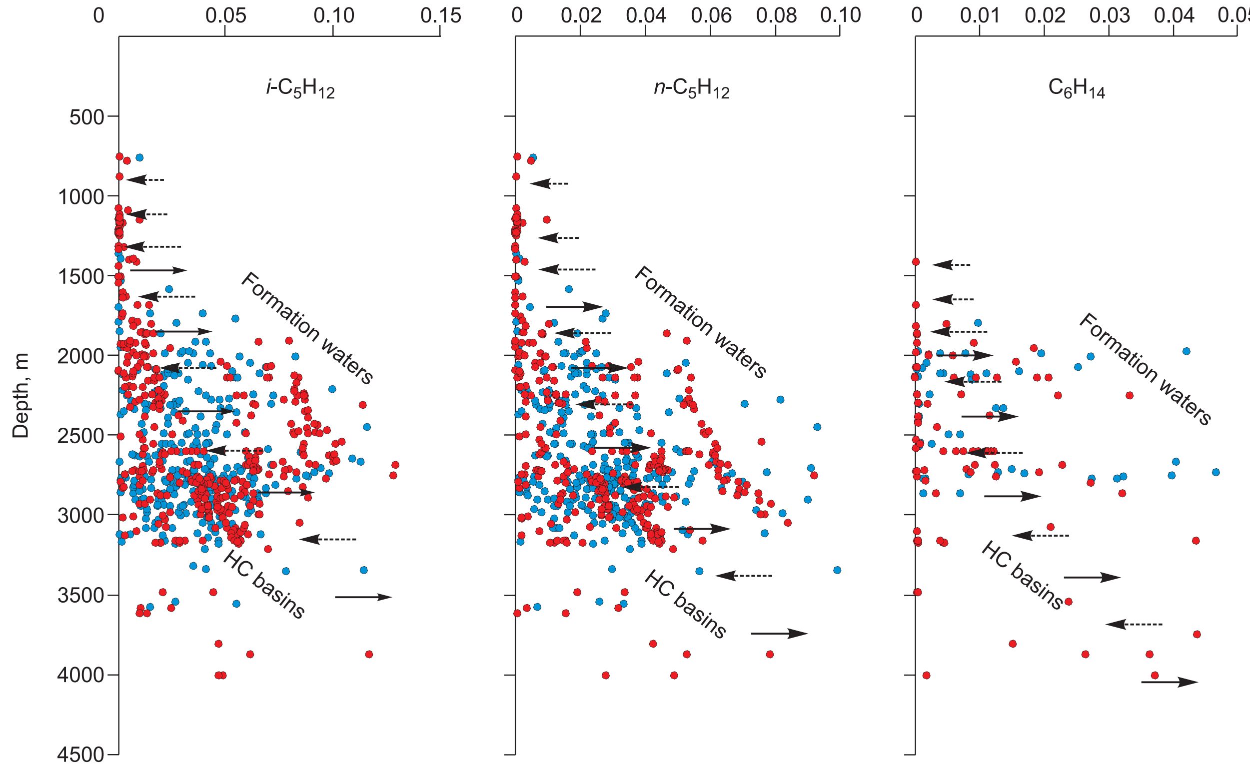

Fig. 7. Depth-dependent fugitivities of iC_5H_{12}, nC_5H_{12}, and C_6H_{14} in formation waters and hydrocarbon accumulations. For notations see Fig. 5.

is compensated by the simultaneous dispersion of hydrogen, methane, carbon dioxide and argon into the surrounding formation waters. At this, almost complete dispersion of the gas cap into peripheral waters takes place in reservoirs BP_3, BP_7, BP_8 and BP_9.

Within the *Vostochno-* and *Zapadno-Tarkosalinskoe* oil and gas condensate fields, as noted above, most of the formation waters are ultimately saturated with gases, with K_g values approaching 1.00 (Fig. 3), which has entailed activation of gas formation, as was inferred from the obtained data. The ongoing process of diffusion-driven transport of ethane, propane, butane, helium and nitrogen into HC accumulation is observed in all reservoirs, except BP_{10} and BP_{14} (Zapadno-Tarkosalinskoe field) and $BP_{2\text{-}3}$, $BP_{3\text{-}4}$, BP_6 and BP_7 (Vostochno-Tarkosalinskoe field). All the reservoirs continuously emanate methane, carbon dioxide; and most of them – hydrogen, argon, pentane and hexane.

Hydrocarbon deposits of the *Yety-Purovskoe* oil and gas field are characterized by the dissolution into the surrounding groundwater of mainly hydrogen, methane, ethane, propane, pentane, hexane, carbon dioxide and argon, which in turn is compensated by influx of isomeric and normal form of butane (BP_5, BP_6, BP_7, BP_8, etc.), helium and nitrogen (except BP_5). The process of dispersion of hydrogen, methane, hexane, carbon dioxide and to a lesser extent pentane from almost all gas caps of oil deposits was revealed within the *Ust-Kharampurskoe* oil field. Against this background, there is observed influx of ethane, propane, isomeric and normal forms of butane, helium, argon and nitrogen into the HC accumulations, while isomeric and normal forms of pentane are augmented in reservoirs BP^0_{10}, BP_{11}, BP^2_{14} and BP_{15}. The equilibrium between pentane and formation waters is reported from reservoirs BP_{12} and BP^1_{14}.

The fields located on the Yamal Peninsula (Malyginskoe, Kharatskoe, Kruzenshternskoe and Verkhne-Tiuteyskoe) are characterized by the dispersion of hydrogen, methane and carbon dioxide from almost all the studied HC deposits, which is compensated by the influx of helium, nitrogen, and in varied degree by methane and argon homologs (Novikov, 2018a). Gas fugitivities show an increasing trend with depth. Most of HC accumulations experience transportations towards the weighting of their composition. The calculation results analysis have shown that the nature of the interaction between formation waters and HC deposits at the Rostovtsevskoe field, located in southern Yamal Peninsula (Fig. 1), differs considerably. Thus, in reservoirs TP_{14} and NP_8, hydrogen, methane, carbon dioxide, nitrogen and most of heavy hydrocarbons are dissolved into the surrounding groundwater, despite the fact that it is classified as oil and gas condensate field.

In the studied reservoirs of the Neocomian aquifer complex of the Deryabinskoe (SD_{IV}), Ozernoe (SD_{VI}), Pelyatkinskoe (SD_{III}) and Suzunskoe (SD_{XIII}) fields in the adjacent areas of the Yenisei-Khatanga basin, the process of gases redistribution between hydrocarbon accumulations and peripheral waters is largely similar to most of the fields located

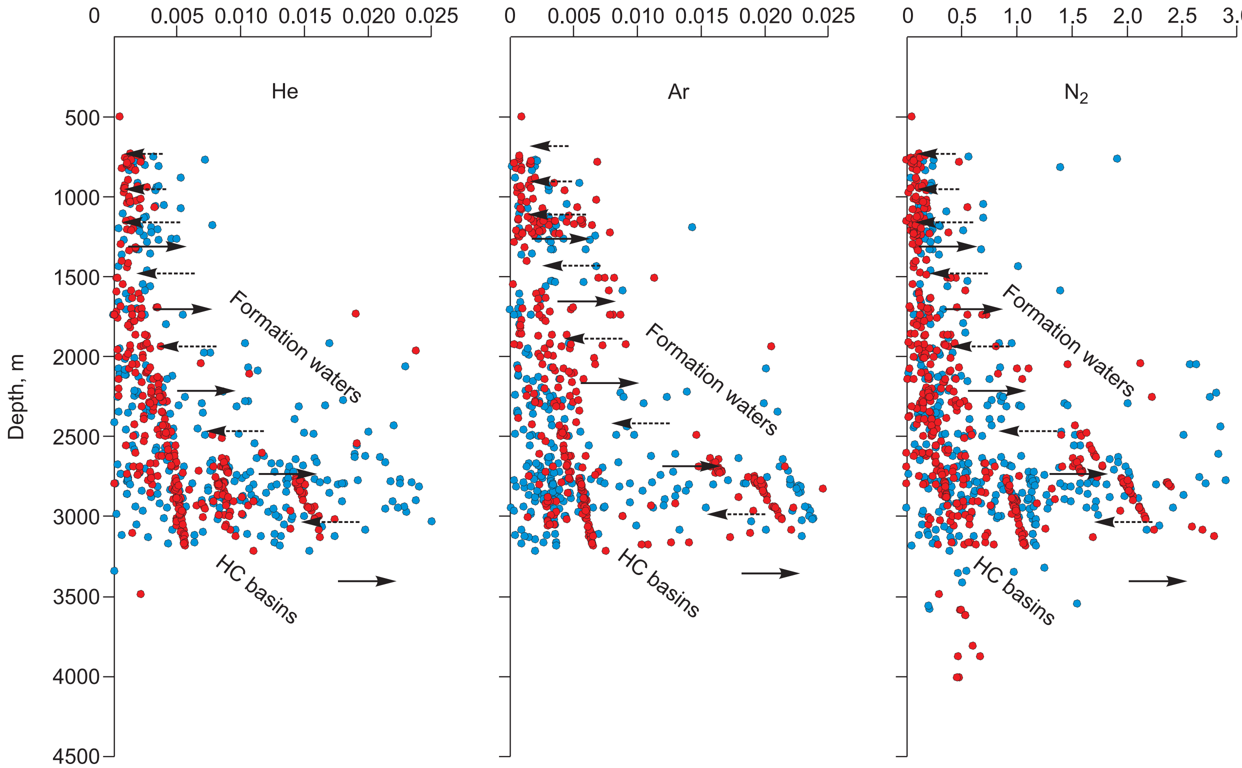

Fig. 8. Depth-dependent fugitivities of He, Ar and N_2 in formation waters and hydrocarbon accumulations. For notations see Fig. 5.

in the Nadym-Taz region and in the Yamal Peninsula. Methane and carbon dioxide are continuously dispersed, which is compensated by the arriving to varying degrees helium, nitrogen, and methane homologues in their stead.

The nature of physicochemical equilibria between deposits and the surrounding formation waters of the ***Jurassic aquifer complexes*** was estimated for major reservoirs of the Kharampurskoe, Gubkinskoe, Komsomolskoe oil and gas condensate fields within the Nadym-Taz interfluve, Malyginskoe gas condensate field in the northern Yamal Peninsula and Yuzhno-Soleninskoe gas condensate field in the adjacent areas of the Yenisei-Khatanga basin. Most thoroughly studied was the Kharampurskoe field, which can perfectly exemplify the nature of equilibria in the Upper Jurassic and Lower-Middle Jurassic aquifers.

In the producing intervals confined to the Upper Vasyugan subformation (reservoir Yu_1) and in reservoirs Yu_2 and Yu_3 subsumed into the upper sunformation of the Tyumen Formation, the mechanism of interactions between them was markedly different, which is expressly evident from by the calculation results analysis.

Thus, in reservoir Yu_1^1 the ratios of individual fugitivity coefficients of gases in HC deposits and groundwaters reflected the hydrogen, methane, helium and nitrogen transport from the surrounding subsurface waters into the reservoir, while other gases, specifically, heavy hydrocarbons, undergo dissolution into formation waters. This allows to infer that HC accumulation in reservoir Yu_1^1 which has been intensely diluted with methane contained in groundwaters is therefore experiencing directional transformation from oil to gas-oil type.

A hydrocarbon accumulation in reservoir Yu_1^2 is characterized by dispersion of mainly methane, ethane, propane, pentane and carbon dioxide from the gas component of HC deposits into the surrounding groundwater, achieving a relative equilibrium for argon, which in turn is compensated by the influx of hydrogen, butane, helium and nitrogen into the accumulation. The distinction between this HC accumulation from the overlying one accounts for the fact that methane dispersion and weighting of its composition was prompted by contributions of butane. The pattern of redistribution of gases between deposits and groundwater in reservoir Yu_1^3 is almost identical to reservoir Yu_1^2, but with the difference observed only for hydrogen, helium and argon.

A different process takes place in the deposits of reservoir Yu_1^4: methane, light hydrocarbons and carbon dioxide are dissolved from the deposits into the surrounding groundwater, which is compensated by the simultaneous inflow of heavy hydrocarbons, noble gases and nitrogen from the surrounding formation waters. It stands to reason that the gaseous part of the HC accumulation is currently being transformed towards weighting of its composition and changing concentrations of non-hydrocarbon gases. With this in mind, we can assume the existence of continued processes of oil generation, migration and accumulation until the present stage of the geochemical evolution, against the backdrop of the cessation of gas generation and accumulation. The latter has been seeing the initial period of dispersion-dissolution into the surrounding groundwaters.

Reservoirs Yu_2 and Yu_3, confined to the upper subformation of the Tyumen Formation, are characterized by the pattern of redistribution of gases, identical to the one described above for reservoir Yu_1^4.

HC accumulations of the Gubkinskoe, Komsomolskoe, Yuzhno-Soleninskoe fields are largely the same nature of equilibria in the water – gas system, with the Kharampurskoe field studied in detail. A deposit in reservoir $Yu_{2\text{-}3}$ of the Malyginskoe gas condensate field is interpreted as outstanding, being the deepest of the studied ones (intervals: 3612–3620, 3636–3644 m, well No. 35). Its main distinction is that besides the normal form of pentane, methane, carbon dioxide, it emanates nitrogen as well, which altogether is compensated by the weighting of gas composition of the HC accumulation by the heavy hydrocarbons (ethane, propane, butane and the isomeric form of pentane) transport from the surrounding subsurface waters.

Thus, the analysis of redistribution of gases between hydrocarbon accumulations and near-contour formation waters showed that the deposits are predominantly in an unstable position with respect to the waters hosting them (Figs. 5–8). Almost all the studied HC deposits emanate methane, carbon dioxide and argon, which is compensated by helium, nitrogen (and to varied degree methane homologs) influx into the deposits. A number of major HC accumulations undergo the process of their phase reformation towards weighting of their composition. The revealed specificity of the processes of gases redistribution between HC deposits and formation waters surrounding them assumingly depends on the phase composition of HC accumulations. The most intensive processes of exchange and transformation are inherent in oil-gas-condensate and gas-condensate accumulations, whereas oil accumulations experience their least intensive action. The analysis of changes in the relations of individual fugitives of gases in formation waters and hydrocarbon deposits revealed some regularities. Thus, a depthwise increase shown by the methane, hydrogen, ethane, propane and carbon dioxide ratios confirms the earlier research results obtained by of B.N. Ryzhenko and V.P. Volkov on increase in gas volatility for a wide range of temperatures and pressures (Ryzhenko and Volkov, 1971). The behavior of butane, pentane and hexane is known to be much more complicated.

The modeling results for interactions in the water–gas system showed that the zoning of the predicted-hypothetical composition of the free gas phase (except hydrocarbon and non-hydrocarbon gases) is also manifested in distributions of the helium-argon ratio with depth, which is associated with the absolute age of hydrocarbon accumulations. Unlike the widespread age determination methods for gases which are largely based on the helium-argon ratio in groundwaters and yielding rather questionable results, the algorithm used in the HG-32 (Hydrogeo) software is based on V.P. Savchenko's empirical equation for free gas derived from the data on a large number of hydrocarbon fields across the world.

Thus, the predicted age of potential HC accumulations determined from water-dissolved gases, increases from 20–23 Ma (Upper Oligocene) in the upper parts of the Aptian-Albian-Cenomanian complex to 40–87 Ma (Eocene-Upper Cretaceous) in the lower parts of the Neocomian complex. The results are in good agreement with the data obtained by N.N. Nemchenko, A.S. Rovenskaya and M. Shoell on the isotopic composition of natural gases in the discovered giant deposits in the northern West Siberia (Nemchenko et al., 1999).

CONCLUSION

To summarize the above, the following inferences were made:

1. Using the pioneering approach, the processes involved in saturation of groundwaters with gases in Jurassic and Cretaceous oil and gas-bearing deposits of the Yamal–Kara depression were studied on a unified methodological basis. Formation waters are interpreted to vary from unsaturated to ultimately saturated with gases (according to the K_g values analysis). The growth of formation waters saturation with gases with increased the subsidence depth of producing reservoirs was established, along with the dependence of the degree of formation waters saturation with gases on the value of their total gas saturation.

2. All subsurface waters that have total gas saturation greater than 1.8 L/L become saturated with gases ($K_g = 1.0$), providing thus the theoretical prerequisites for the formation of hydrocarbon deposits. Whereas unsaturated formation waters are capable of dissolving previously formed oil and gas accumulations.

3. A direct relationship between the degree of formation waters saturation with gases (K_g) and the phase composition of deposits is established. The main gas condensate deposits are confined to the zone of development of K_g values in the range from 0.8 to 1.0, while oil accumulations are associated with less waters saturated with gas. An explicit complexity of the revealed dependencies attests to the diversity of chemical and gas composition of groundwater, as well as to the presence of different genetic types of subsurface water in oil and gas-bearing horizons in the section.

4. The established equilibria pattern allows to confidently suggest the presence of oil and gas deposits in Jurassic–Cretaceous deposits of the Yamal–Kara depression, which act as a conservative element of the lithosphere representing geological and geochemical "relics" that survived from the past stages. The surrounding subsurface waters are interpreted as a more active component of this system, significantly outpacing its geochemical evolution. This translates to differentiation in the fugitivity ratios of individual gases in groundwaters and HC accumulations. As a result, the chemical composition of the latter experiences a slow direc-

tional change in parallel with the equilibrium establishing itself, to measure up to the qualitatively new geochemical state of the water–gas system.

5. Results of the regional hydrogeochemical and gas-hydrogeochemical studies have prompted an inference that favorable conditions for the formation and preservation of hydrocarbon accumulations in most of the study area, except the peripheral areas of the West Siberian sedimentary basin.

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