Theoretical substantiation of application of the hydrocarbon accumulation prospecting technique in Western Siberia based on the study of water-gas equilibria

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Abstract. Computer simulation of physicochemical equilibria and evasion-invasion processes in the water-gas system enables modeling gas saturation degree variability and the physical nature of gaseous diffusion redistribution at the hydrocarbon accumulation – formation waters interface and in the transition zone from hydrocarbon accumulation to the peripheral waters. Aquifers with waters characterized by low (< 0.2) to maximum saturation (0.8-1.0) with gases have been established within Cretaceous, Jurassic and Paleozoic deposits of the West Siberian sedimentary basin (WSSB), along with an increase in the saturation degree of formation waters with gases as the occurrence depth of reservoir intervals increases, and a relationship between gas saturation of formation waters and their total gas saturation value. All waters with total gas saturation exceeding 1.8 L/L become ultimately saturated with gases (Kg = 1.0), providing thereby theoretical prerequisites for the formation of hydrocarbon accumulations. The zone of Kg values spanning from 0.8 to 1.0 is associated with major gas condensate accumulations, while less saturated waters – with oil accumulations. The simulation results allow to assess the current state of the petroleum system of the West Siberian sedimentary basin, its part or a separate horizon within its limits, and to substantiate forecasts of its oil and gas potential at regional, zonal or local levels. The application of modeling techniques to water-gas equilibrium enables significant improvement of the reliability of oil and gas reserves assessment and largely contributes to the solution of problems with respect to searches of skipped accumulations in multi-reservoir fields during their further exploration.

1. Introduction
The fundamental task of studying geochemical processes and physicochemical equilibria in the water – gas system, being closely related to the backbone geological problem of the formation of oil and gas accumulations and degradation, is presently a topical research direction [1-4]. It is generally recognized that the present-day discoveries of new oil and gas fields in the West Siberian sedimentary basin (WSSB) can no longer rely on structural imaging and analysis of general geological and geochemical indicators derived predominantly from geophysical data. In this context, the study of physical and chemical equilibria in the water-gas system, providing information about the region-specific conditions for the oil and gas fields formation and conservation, as well as mass exchange processes with the surrounding groundwater, opens a new promising area of extensive research conjugated to geochemical prediction of oil and gas potential and evaluation of reservoirs in local structures [5, 6].

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Because of their extraordinary mobility, gases generate the largest scattering halos within sedimentary basins, during diffusion processes between hydrocarbon accumulations and surrounding reservoir waters, representing therefore one of the most reliable criteria for hydrocarbons prospecting [4, 7-12]. The factual knowledge resulted from the calculations performed by Yu. P. Gattenberger, V. M. Matusevich and others have provided a compelling evidence that "...at the present stage of the WSSB development, the water percolation rate in its submerged part is marked by negligible values even from the perspective of geological time, so that water percolation, as it is, does not take place. Under these conditions, the largest extents of mass transfer and, in particular, the appearance of water dispersion halos near oil and gas accumulations are associated with the diffusion processes» [13]. In this context, the role of inherent processes within the water-gas system increases far and away.

The study of water dissolved gases (WDG) in Western Siberia given a was start to more than 50 years ago by M. S. Gurevich and N. N. Rostovtseva. In the early 1950ies of the 20th century they were the first to define the nature of gas zonation of groundwater within the WSSB and revealed the prospecting value of WDG, which is largely determined by extent of their tension rather than qualitative composition alone. N. M. Kruglikov made a discovery about the diffusion and dispersion of gas being responsible for a decrease in dissolved gas tension as the distance from the gas-water contact increases.

L. M. Zorkin investigated different conditions potential for gas generation, their extraction from groundwaters and formation of gas accumulations. The study of groundwater and water-dissolved gases in WSSB attracted interest of many researchers who have greatly contributed to its advancement: A. E. Kontorovich, B. P. Stavitskii, A. A. Rozin, N. M. Kruglikov, V. V. Nelyubin, O. N. Yakovlev, V. M. Matusevich, A. A. Kartsev, A. D. Nazarov, S. L. Shvartsev and D. A. Novikov and many others [3, 4, 9, 11].

At this, conditions of physicochemical equilibria of gases remain largely understudied despite their representing a problem of great scientific and practical importance, which allows to solve more purposefully many genetic issues inherent in crude petroleum generation [14], and to reveal hitherto unknown patterns of the formation of groundwater [5, 6].

The above discussed equilibria allow revealing the direction of geochemical processes in the "water-gas" system both at the present stage of the petroleum system development, and in its geological past. These also address the problems of the deposits' preservation and enable prediction of their phase type. Their solution will result in providing a substantiation of a new oil and gas prospecting technology applicable to the conditions of WSSB.

2. Materials and Methods

In itself, the water-gas system is very complex, which is accounted for by the multi-component and multidirectional processes involved therein, which makes problematic the use of previously employed simplified calculus methods for gas saturation levels in formation waters, individual gases fugacities etc. The calculations in this research were made with the use of HG-32 (Hydrogeo) software package, currently having no analogs in the world, which was developed by M. B. Bukaty at the Trofimuk Institute of Petroleum Geology and Geophysics SB RAS. The simulations based on it allow considering all the parameters of the investigated system (density, TDS and the composition of groundwater, gas saturation, WDG composition, temperature and pressure conditions and other indicators) and determining the composition and a number of characteristics of the equilibrium free gas phase from the composition of dissolved gas and water, or conversely, the composition and other parameters of dissolved gas from the composition of free gas and solution, as well as modeling evasion-invasion of gases under variable pressure, temperature and composition of the solution [1, 2, 5].

The theory is underlaid by dependences of the mass-action law and the activity method. For each i component from n components of the gas mixture dissolved in groundwater, in this case, a reversible reaction of the solution-to-free phase transition is considered:

\[ i_p \Leftrightarrow i_r. \]
The mass-action law can be written for it as

$$ K_i = f_i / a_i^0, $$

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

The degree of water solution saturation with gas i is estimated using undersaturated index

$$ L_i = a_i K_i / f_i, $$

where $a_i$ - is activity of gas in the investigated water.

The simplest approach consists in direct identification of fugacity $F_i$ of each gas i in free gas phase in hypothetical equilibrium with the solution, employing semi-empirical methods developed by A.Yu. Namiot, E.S. Barkan and others, while

$$ F_i \equiv a_i K_i \quad \text{and} \quad L_i = F_i / f_i. $$

The $f_i$ and $K_i$ were calculated using a number of correction factors and a series of regression equations obtained M. B. Bukaty as a result of processing of experimental data [1, 2], which describe required thermodynamic parameters, solubility and distribution of gas between the phases depending on the composition of each, and real temperature and pressure conditions.

This paper summarizes the results of numerical physicochemical modeling of water-gas equilibria for more than 700 hydrocarbon accumulations localized within Cretaceous, Jurassic and Paleozoic sedimentary strata and delineated 132 oil and gas fields of WSSB.

3. Results and Discussion

Within the scope of the considered problem of interaction processes in the water-gas system, there are two main issues to be solved:

1. assessment of the groundwater saturation with gases $(K_g)$,
2. interaction in the water – gas system (formation water – hydrocarbon accumulation).

In the context of WSSB, the former aspect of the problem has been thoroughly discussed in papers authored by N.M. Kruglikov, V.N. Kortenshtein, V.V. Nelyubin, O.N. Yakovlev, M.Ya. Rudkevich, L.S. Ozeranskaya, N.F. Chistyakova and others [15, 16, 17]. Their works provide data on the variability of groundwater saturation with gases at the regional level within different aquifers of the lower hydrogeological stage. Specifically, it is indicated that the gas saturation coefficient values for Jurassic complexes vary from 0.03 in the marginal part of the basin to 1.00 in its center.

Within the Neocomian complex, $K_g$ values tend to increase from the margins to the center of the basin (to 0.77 in the Middle Ob district), with the maximum values up to 0.94 identified in the Taz area (within the Nadym-Taz interfluve). While the Aptian-Albian-Cenomanian complex is characterized by a regular increase in $K_g$ values from 0.02 to 0.87 in the direction from south to north of the WSSB. Calculations performed by D. A. Novikov showed that the saturation of groundwaters with gases appears of more complex nature at zonal and local levels (within a specific field) [4-6, 10, 18]. Within Paleozoic, Jurassic and Cretaceous water-bearing complexes, gas saturation of waters in the aquifers established from $K_g$ values varies from low (< 0.2) to maximum (0.8-1.0) (figure 1). Gas saturation levels of the detected formation waters vary from the unsaturated to extremely saturated (from $K_g$ values).

An increase in the gas saturation degree of formation waters with deeper occurrence of reservoirs and its dependence on the value of their total gas saturation is established. All waters whose value of total gas saturation is greater than 1.8 L/L tend to be extremely saturated with gases $(K_g = 1.0)$, providing thereby theoretical prerequisites for the formation of hydrocarbon accumulations. Whereas formation waters not saturated with gases are capable of dissolving the previously formed oil and gas.
accumulations. A direct relationship between the degree of saturation of formation waters with gases (Kg) and the phase composition of deposits is established. The zone with Kg values varying from 0.8 to 1.0 is associated with major gas condensate accumulations, and less saturated waters – with crude oil accumulations.

**Figure 1.** Variation of gas saturation coefficient of waters (Kg) with depth (a) and versus total gas saturation value of groundwaters (b).

a) Zones by gas saturation coefficient of water: I - low, II - medium, III - high, IV – very rich, V – a hydrocarbon accumulation; gas saturation coefficient (Kg) in the objects of major hydrogeological complexes: 1 – Aptian-Albian-Cenomanian, 2 – Neocomian, 3 – upper Jurassic, 4 – Lower-Middle Jurassic.

b) Zones by total gas saturation of ground waters (L/L): 1 – fairly low (< 0.1), 2 – low (0.1-0.5), 3 – medium (0.5-1), 4 – high (1-5), 5 – fairly high (> 5).

Gas saturation degree of water there shows a gradual decreasing trend from beds the with gas condensate accumulations localized within the Kg values spanning from 0.8 to 1.0, to beds holding mostly oil accumulations confined to the Kg values in the range from 0.4 to 0.8. The total gas saturation Kg therefore varies from 0.95 to 1.00 in bed BP12, indicating a maximum saturation of waters with gases.

Kg values constitute 0.80-0.86 in bed BP14 while in the lower lying bed BP15 they range from 0.74 to 0.83. The lower part the section is dominated by beds holding purely oil accumulations characterized by a lower degree of gas saturation. At this, the major oil-bearing bed BP16 is confined to the remarkably unsaturated zone, located within the Kg values range from 0.56 to 0.75. Bed BP17 is marked by the minimum Kg values, within 0.32-0.42. As such, a rather complicated pattern of the revealed dependences indicates diverse chemical and gas composition of groundwaters, as well as the
presence of different genetic types of water in the horizons within the oil-and-gas-bearing portion of the section.

In their works [15, 16, 17], N. M. Kruglikov and V. N. Kortenshtein have analyzed data on interaction in the water–gas system (formation water–hydrocarbon accumulation) for PK1 bed from Medvezhiye, Yamburgskoe, Yety-Purovskoe, Urengoyskoe fields and beds BU10, BU12 of the Urengyoyskoe field. In their opinion, the investigated hydrocarbon accumulations are in unstable (non-equilibrium) position with respect to the surrounding waters, which allowed them to deduce that the processes of degradation of the existing HC accumulations are currently taking place in Upper and Lower Cretaceous horizons.

In this paper, the redistribution of gases between HC accumulations and formation waters was estimated from the ratios of individual fugacities of gases calculated in the HC accumulation-groundwater and groundwater-HC accumulation systems. In the first case, the composition of the free gas phase was calculated from hypothetical equilibrium composition of WDG, while in the second – by the WDG calculated from the composition of hypothetical gas accumulation. The physicochemical calculations for more than 700 oil and gas accumulations performed using with this method allowed to estimate the direction of their interaction with the surrounding groundwaters. It has been established that the HC accumulations are mainly in an unstable position in relation to the surrounding waters. The analysis of methane, ethane and propane behaviors shown in figure 2 indicates that methane and carbon dioxide and argon dissipate almost in all of the investigated accumulations, which is compensated by the intake of helium and nitrogen, and to varying degrees, methane homologues. Quite numerous accumulations are found to be experiencing their phase transformation towards weighting of structure.

This suggests some specific features of the processes of redistribution of gases between HC accumulations and the surrounding formation waters, depending on the phase composition of HC accumulation. The exchange and retransformation processes occur most intensely within oil-gas-condensate and gas-condensate accumulations, and the least intensely – in oil accumulations. Analysis of changes in the ratios between individual fugitives of gases in formation waters and hydrocarbon accumulation has revealed some regularities. Thus the observed increase is in the methane, hydrogen, ethane, propane and carbon dioxide ratios versus their depth confirms inferences made earlier by B. N. Ryzhenko and V. P. Volkov about an increase in gas volatility within a wide range of PT conditions [19]. In this respect, butane, pentane and hexane appear much more complex.

Results of interactions in the water–gas system modeling showed that the zonality of the predicted-hypothetical composition of free gas phase, except hydrocarbon and non-hydrocarbon gases, is also manifested in the depth-dependent distribution of the helium-argon ratio which is directly related to the absolute age of hydrocarbon accumulations.

Unlike the widely applicable methods for determining the age of gases from the helium-argon ratio in groundwater and giving fairly problematic results, the algorithm used in the HG-32 (Hydregeo) software package is based on the empirical equation by V. P. Savchenko for free gases, obtained by him from the data generalization for a multitude of hydrocarbon fields around the globe [20]. Thus, projected age of probable deposits, estimated from water-soluble gases, increases from 20-23 MA (Upper Oligocene) at the top of the Aptian-Albian-Cenomanian complex to 40-87 MA (Eocene-Upper Cretaceous) in the lower Neocomian [4]. The results obtained are in good agreement with the data on the isotopic composition of natural gases in giant deposits of the northern West Siberia obtained by N. N. Nemchenko, A. S. Rovenskaya and M. Shoell [21].
Figure 2. A relationship between CH$_4$, C$_2$H$_6$ and C$_3$H$_8$ fugacities in formation waters and hydrocarbon accumulations and their occurrence depths. Direction of diffusion flow: 1 – from HC accumulation to formation waters, 2 – from formation waters to HC accumulation; gas fugacity: 3 – in HC accumulation, 4 – in formation waters.

4. Conclusions

The application of numerical modeling of water-gas balances thus allows to most reliably solve the problems on prediction of oil and gas content in sedimentary basins. However, the final stage of the calculations within the water-gas system additionally requires conducting extensive complex hydrogeological studies. In a nutshell, the proposed technology can be presented as the algorithm laid down below:

1. Collection and generalization of all available materials on the investigated object in the database within one of the common GIS packages.
2. GIS package-based generation of a set of electronic maps of the chemical and gas composition of groundwater, as well as hydrodynamic, geothermal, paleogeographic and lithological-facies maps of the main complexes with the obligatory allocation of the region-specific characteristics of gas-hydrogeochemical background.
3. Detailed paleohydrogeological reconstructions aiming to clarify the spatial boundaries of the main zones of oil and gas accumulations.
4. Construction of hydrogeological and hydrodynamic models of potential oil and gas accumulation zones, along with specifying the zonal gas-hydrogeochemical background.
5. Creation of hydrogeological models of reference fields, “empty” and promising structures.
6. Development of gas-hydrogeochemical field models within the reference structures in order to identify actual concentrations of oil and gas accumulations.
7. Computer simulation of physicochemical equilibria and processes in the water-gas system.
8. Modeling variability of the degree of gas saturation and the nature of diffusion redistribution of gases at the of hydrocarbon accumulation – formation water interface and in the transition zone from the HC accumulation to the peripheral waters.
9. Substantiation of recommendations on the direction of geological prospecting works and drilling the first and subsequent exploration wells.

In the context of the West Siberian sedimentary basin, the established nature of the water-gas equilibria allows to ascertain that the oil and gas deposits held in Mesozoic and Paleozoic sedimentary strata act as a conservation element of the lithosphere, representing "relics" of previous stages of its geological and geochemical evolution; while the surrounding groundwaters represent a more active component of the system, and appear to be significantly more advanced in their geochemical development. This is evidenced in the difference in the fugacity ratio of individual gases both in groundwater and HC accumulations. As a result, the composition of the latter undergoes a slow directional change towards establishing balance, which corresponds to the qualitatively new state of the water – gas geochemical system. The results of regional, hydrogeochemical and gas-hydrogeochemical studies suggest favorable conditions for the formation and conservation of hydrocarbon accumulations in most of the study area, with the exception of near edge zones of the sedimentary basin.

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