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OIL AND GAS GEOLOGY

THE SEARCH FOR NATURAL HYDROGEN IN RUSSIA: THE STATE OF THE PROBLEM AND POSSIBLE STARTING SOLUTIONS

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Abstract. By the time the decision was made (Rosstandart Order 07.07.2023 No. 490-st) on the inclusion of hydrogen in the all-Russian classifier of minerals in Russia, research on the geological and economic analysis of the possibilities of industrial development of hydrogen resources had not been conducted. Moreover, fossil hydrogen has been studied extremely poorly. The current situation requires the accelerated development of a hydrogen search concept based on the scientific justification of the most promising regional areas of work. In the article, the authors present their vision of this problem and propose possible solutions. In particular, the necessity of organizing scientific and technological hydrogen polygons is argued, the tasks of which will include: (i) the development of theoretical ideas about the role of hydrogen in the evolution of the Earth; (ii) detailing the mechanisms of hydrogen localization in the geological environment; (iii) the development of criteria and methods for geological and economic assessment of hydrogen prospecting, exploration and production; (iv) conducting geological and commercial research at the most promising sites for the development and testing of methods for searching for deposits of hydrogen and related minerals.

Keywords: *natural hydrogen, ancient platforms, great depths, serpentinization, radiolysis, microbiological processes of hydrogen generation, mineral and energy resources of the bowels of the Earth*

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INTRODUCTION

Hydrogen energy is a key direction of the modern economy, based on ecological imperatives. To date, significant progress has been made in the production and storage of technical hydrogen. In the last decade, there has been a steadily growing interest in natural hydrogen (H_2) as an energy resource not associated with greenhouse gas emissions.

Hydrogen produced in the Earth's interior has been extremely poorly studied. Meanwhile, the "hydrogen boom" is growing. Many countries (Australia, the USA, China, Spain, France, Mali, Oman, etc.) and dozens of companies are already working on the search for natural hydrogen, most often focusing on previously accidentally discovered outflows of this gas.

In Russia, hydrogen has been included in the national classifier of mineral resources (order of Rosstandart dated July 07, 2023, No. 490-st). This decision requires accelerated scientific justification for selecting the most promising objects within which it is advisable to prioritize geological, geophysical, geochemical, and hydrogeological studies necessary to substantiate a comprehensive methodology for conducting exploration works for hydrogen.

The purpose of this article is to substantiate proposals for forming the scientific basis of the concept of natural hydrogen exploration in Russia.

DATA AND RESEARCH METHODS

The source materials for solving the target problem have been collected from numerous literary and archival sources reflecting the results of studying the geological structure of the Earth's interior, mechanisms of hydrogen generation and accumulation.

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For generalizing the source materials, general scientific methods were used, as well as special methodological approaches focused on the analysis of geological and geophysical information. Thus, bibliometric analysis tools revealed the dynamics of publication activity on issues of natural hydrogen generation and accumulation, showing the interest of various countries, scientific centers, and resource-extracting companies in conducting exploration work to detect industrial accumulations of H_2 (Fig. 1).

It is worth noting that the number of publications devoted to natural hydrogen has been increasing in recent years with growing rapidity; many works have a high five-year citation level (over 1000).

Previous research level

To date, hundreds of hydrogen degassing manifestations are known, most of them gravitate toward lithospheric plate boundaries, fault zones, regions with increased seismicity [4, 17, 20], however only one industrially significant free hydrogen deposit is reliably known; its discovery in Mali [18] served as the "starting point" in the history of this gas as a mineral resource. Currently, global H_2 resources are estimated at 5 trillion tons. It is assumed that the cost of natural hydrogen production will be multiple times lower than that of "green hydrogen" [16].

The trend of searching for deeply submerged hydrogen accumulations is intensifying. For example, in France, H_2 of low concentration was found at a depth of 200 m, but as the sampling objects deepened, the hydrogen concentration increased to 14% at a depth of 1100 m and 20% at a depth of 1250 m [20]. Such examples are not isolated. Strong arguments have been presented in favor of the existence of favorable conditions for hydrogen generation and accumulation at great depths of ancient platforms [16].

The experience of Australia is interesting: state licenses are issued for natural hydrogen exploration there. Here, the search for H_2 is combined with the production of technical hydrogen from coal seams using the energy of solar panels, wind, and tides. Simultaneously, issues of underground hydrogen

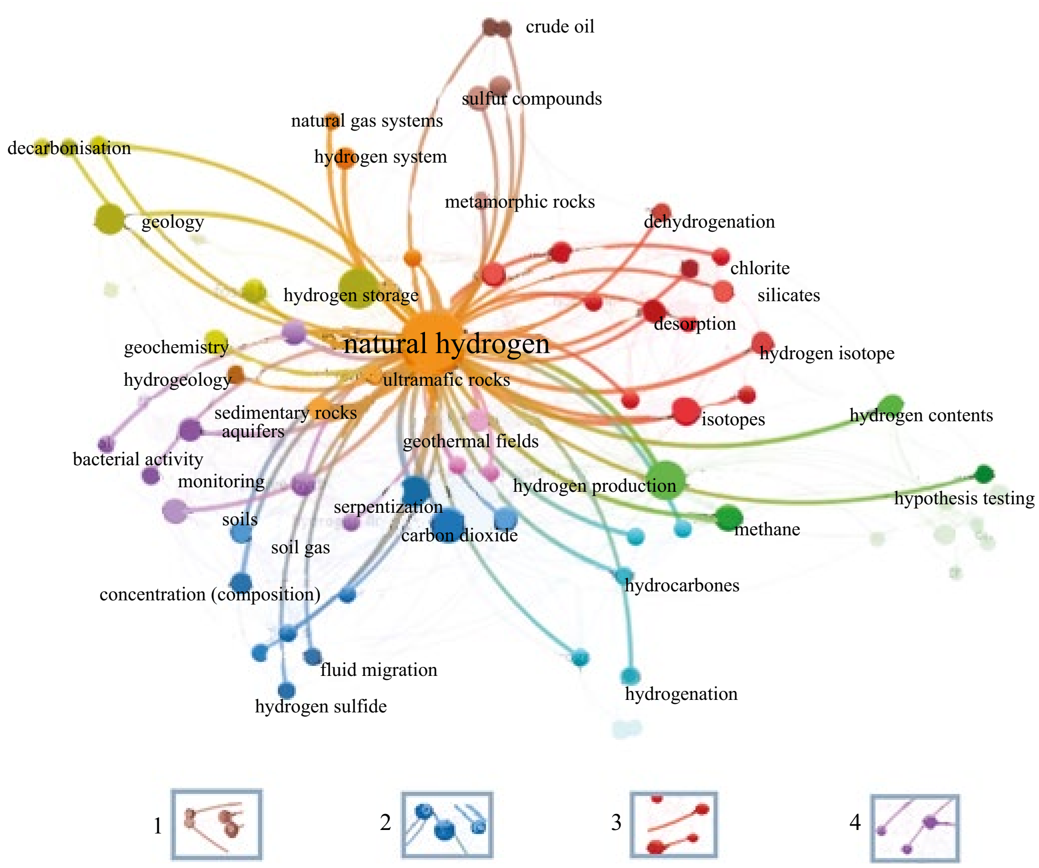

Fig. 1. Thematic landscape of the subject area "natural hydrogen" (native/natural/geological/white/gold/hydrogen) based on the Scopus database for a 20-year period (VOSviewer software). Legend: 1 – biogenic sources; 2 – abiogenic sources; 3 – interaction with rocks; 4 – microbiological processes.

storage are being addressed regardless of the method of its production [15].

In Russian scientific literature until recently, H_2 as a mineral resource was mentioned only in rare cases [5, 12, 14]; such function of H_2 is not reflected in state policy documents, including the Roadmap for the development of hydrogen energy in the Russian Federation until 2024 (Government Decree of October 12, 2020 No. 2634-r). There are no methodological developments for assessing its reserves and resources, economic criteria for profitability have not been justified, and there is no regulatory framework for conducting prospecting and exploration work for hydrogen.

There is much controversy regarding the genesis and localization conditions of natural hydrogen. However, thanks to the work of scientists from several countries (G. Etiope, I. Moretti, R. Nandi, A. Prinzhofer, L. Truche, V. Zgonnik, et al.) and Russia (E.M. Galimov, V.N. Larin, S.P. Levshunova, V.A. Nivin, V.A. Sokolov, M.A. Fedonkin, V.P. Yakutseni, et al.), certain issues regarding the generation and accumulation of hydrogen do not cause disagreements. Thus, today, magmatogenic, metamorphogenic, radiogenic, and microbiological sources of hydrogen in the geological environment are well known [6, 7, 8, 20]. In particular, there is no doubt about the global geological role of serpentinization and water radiolysis in hydrogen generation [13, 15, 17]. The absence of correlation between hydrogen and methane content for gases of metamorphic origin has been noted [8], but the presence of an inverse correlation (Fig. 2) for methane and hydrogen of biochemical genesis has been revealed [3, 14]. These issues are discussed in more detail in works [4, 14, 20].

Ideas about hydrogen accumulation processes are less definite. As a rule, they are judged by surface manifestations of hydrogen degassing, indicating the directions (and partly the scale) of unhindered hydrogen degassing. But the physical essence of degassing processes is not the concentration but the dispersion of hydrogen in the geological environment, therefore, external manifestations of hydrogen degassing (for example, bleached circles) are not always reliable diagnostic criteria for the localization of natural hydrogen.

A large volume of research has been conducted on modeling geochemical and geomechanical processes accompanying the joint storage of hydrogen and methane at natural gas storage facilities (UGS). Our results from several UGS facilities located in aquifers (Fig. 3) demonstrate the activity of methanogenic archaea and bacteria capable of using hydrogen as an energy source [9].

Studies aimed at identifying the nature of the interaction of this gas with the rocks of their host reservoirs are also of interest for the industrial development of hydrogen resources. As a particular example of such studies, we can note the identification, based on experimental models,

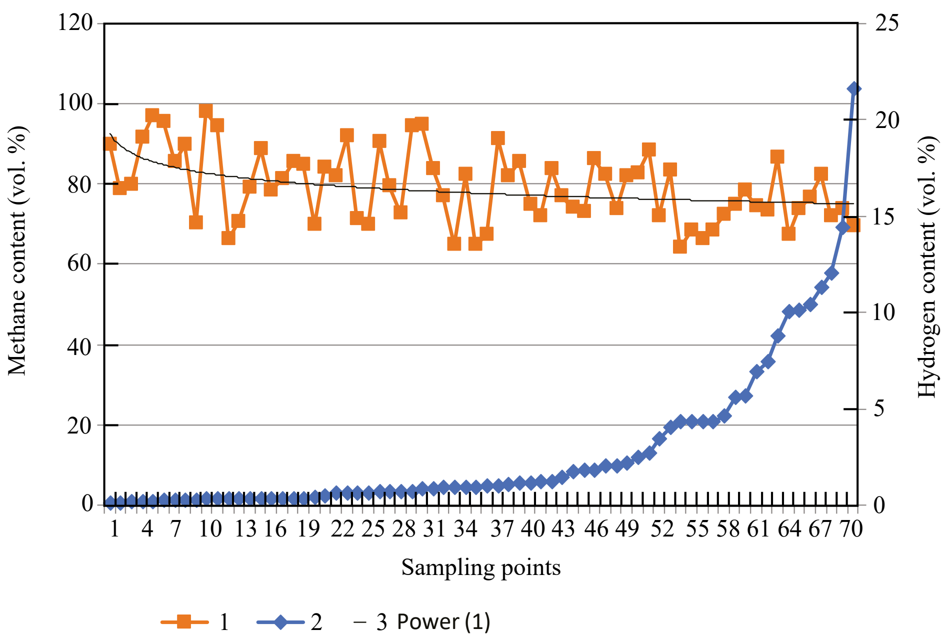

Fig. 2. Character of hydrogen and methane accumulation in the waters of the supra-salt floor of the Caspian oil and gas province. 1 – methane content (% volume); 2 – hydrogen content (% volume); 3 – power averaging of methane content.

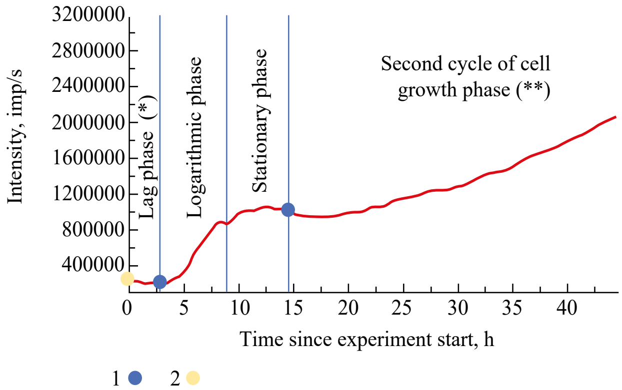

Fig. 3. Results of sequencing formation water samples from Shchelkovo UGS and growth of microbiological populations in these waters with additional hydrogen feeding according to the authors' experimental data. 1 – addition of hydrogen during the experiment; 2 – sampling for determination of microbiological composition; (*) initial microbiological composition of the studied groups of microorganisms: methanotrophs (Marinobacter) – 92% (**) final microbiological composition of the studied groups of microorganisms: methanotrophs (Marinobacter) – 9%, methanogens (Methylophaga) – 8%, sulfate-reducing microorganisms (Desulfopila) – 9%, sulfur-oxidizing (Thiohalobacter, Thioalkalispiraceae) – 30%.

of significant changes in the capacity-filtration and strength properties of terrigenous rocks induced by their geochemical interaction with hydrogen in case of their moistening and the absence of significant effects during contact of hydrogen with dry samples of terrigenous rocks, both under cyclic loading and under stationary conditions (Fig. 4) [2].

The information provided above about previously conducted research, without claiming to be a comprehensive review of accumulated knowledge in the field of natural hydrogen, demonstrates only various aspects of studying the interaction of hydrogen with the geological environment. Nevertheless, it makes it possible to express some judgments about regional directions of hydrogen exploration in Russia.

RESEARCH RESULTS: POSSIBLE DIRECTIONS OF SCIENTIFIC JUSTIFICATION FOR SEARCHING HYDROGEN ACCUMULATION ZONES IN RUSSIA

Based on the current level of our knowledge about the patterns of distribution of natural hydrogen outflows to the surface and into the near-surface layers of the Earth's crust, as well as taking into account existing hypotheses of hydrogen formation, it is possible to formulate the following basic provisions of the hydrogen exploration concept (Table 1):

Abiogenic hydrogen synthesis supplies the largest volume of hydrogen to the lithosphere. It is implemented in various ways, but the most common are serpentinization of ultrabasic rocks and water radiolysis.

1. The compositions of gas mixtures from abiogenic generation sources do not show correlation between hydrogen and methane content; methane is present in low, sometimes trace concentrations, which excludes the possibility of forming industrially significant accumulations. Hydrogen of abiogenic origin, due to its high volatility, can be detected in a wide range of depths.

2. The compositions of gas mixtures from biogenic generation sources are characterized by an inverse correlation between hydrogen and methane (Isaev's antagonism), the hydrogen content is significantly lower than the methane content. Hydrogen of biogenic origin is concentrated in the sedimentary cover, predominantly under salt-bearing and doleritic caprocks.

3. The mechanisms of hydrogen accumulation are fundamentally different for oil and gas bearing and oil and gas prospective continental territories, on the one hand, and shields and orogenic (folded and block) structures of the mobile belt, on the other (Table 1). Within oil and gas bearing territories, dominant and regional fluid seals play a key role. The role of dominant fluid seals (DFS) is especially significant, as they divide the sedimentary cover sections of oil and gas provinces in plate structures of young and ancient platforms into two stories with different types of water pressure regimes (the upper with free and the lower with (quasi)stagnant). In lithological terms, the DFS most often consist of salt-bearing and/or doleritic gigantic strata. The fluid isolation properties of the DFS can be enhanced by the presence of gas hydrate layers in the upper parts of the section, as well as by the development of hydrodynamically screened autoclave-type hydrocarbon systems below the salt (or doleritic) covers [1]. Thus, the presence of DFS, the placement of hydrogen accumulation zones in the lower geofluid dynamic story, and the spatial proximity of H_2 generation sources and hydrogen accumulation zones collectively provide the most favorable conditions for the formation of large hydrogen deposits at great depths in oil and gas provinces/basins.

Within the shields of ancient platforms and plates of young and ancient platforms, vertical migration of hydrogen predominates, which has been repeatedly recorded in the form of hydrogen (methane-hydrogen) emanations. Nevertheless, the stability of flow rates and the geochemical composition of gas mixtures are the most important objects of study.

Let us emphasize the important point: the path from orienting hydrogen searches based on individual direct and indirect indicators to creating a comprehensive methodology for forecasting industrially significant hydrogen deposits lies through conducting a large volume of fundamental research and experimental work. This, in turn, actualizes the task of organizing integrated scientific and technological test sites for detailing the scientific and methodological foundations for H_2 forecasting and developing technologies for its prospecting, exploration, and development. The question of the

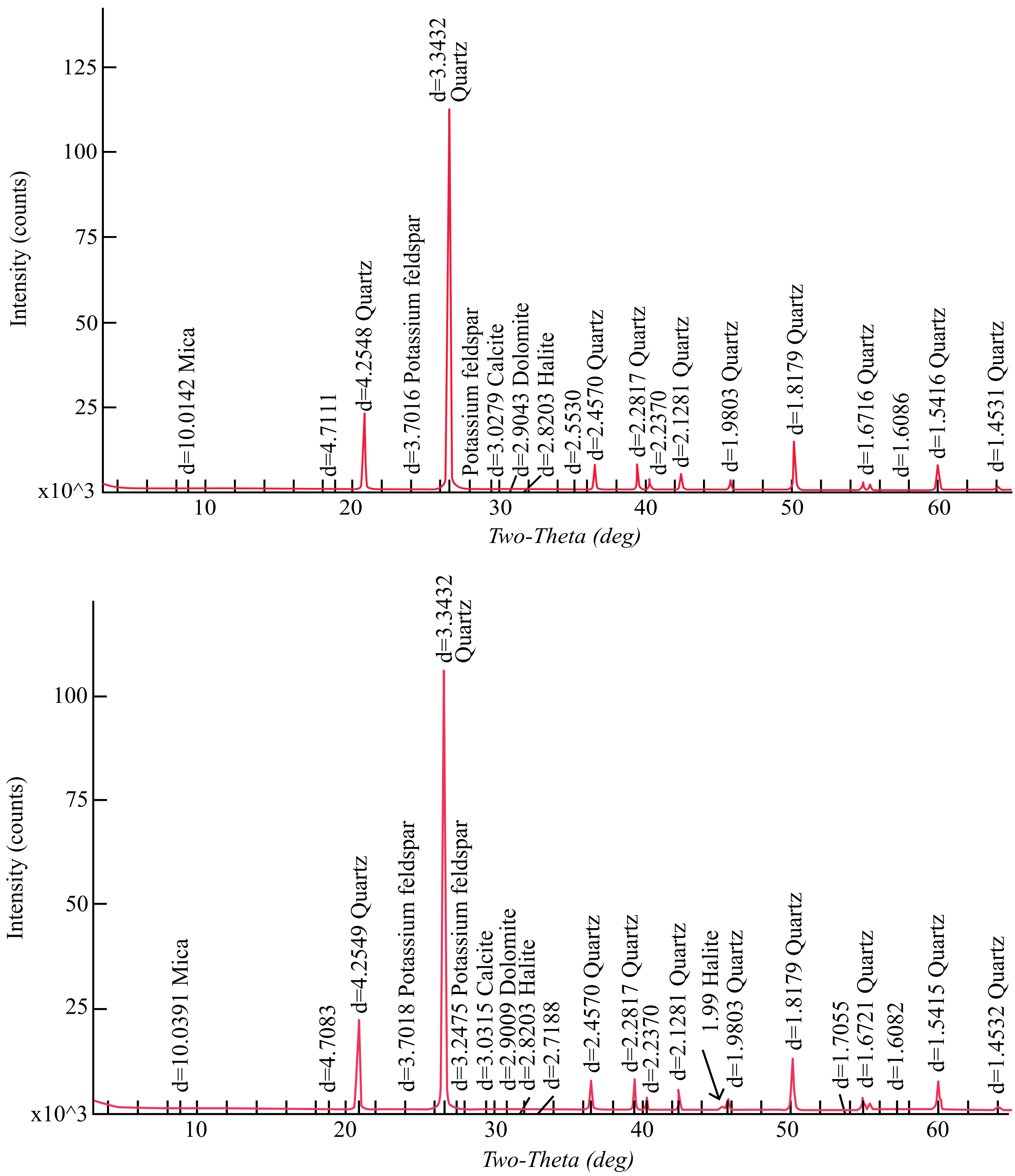

Fig. 4. Comparative evaluation of the X-ray diffraction pattern of a sandstone sample before (a) and after (b) exposure to hydrogen (mineral composition of sandstone: quartz – 94%, mica – 2%, dolomite – 1.1%, potassium feldspar – 2%, calcite – 0.6%, halite – 0.3%)

Table 1. Forecast objects and directions of scientific research on substantiation of search criteria for natural hydrogen as a mineral resource

Forecast of the nature of hydrogen generation and accumulation processes	Differences between regions by presence (absence) of regional and dominant fluid seals in the Earth's crust										
	Regions where regional and dominant fluid seals are absent			Regions where Regional and dominant fluid seals are present							
	Shields of ancient and young platforms	Block orogenic structures	Folded orogenic structures	Provinces* oil and gas bearing and prospective (tectonic-sedimentary provinces) of plate structures of ancient and young platforms					Oil and gas bearing and prospective basins* (sedimentary-rock basins)		
				located in the internal parts of plates		located at plate edges					
				of ancient platforms	of young platforms	at boundaries with folded structures	at junction of ancient and young platforms	on passive continental margins	marginal-continental seas	mediterrane-an seas	intraconti-nental rifts
Examples of territories*	Aldan, Baltic shields	Kuril-Kamchatka volcano-plutonic arc	Altai-Sayan folded structure	Lena-Tunguska, Volga-Ural OGP	West Siberian Siberian OGP	Timan-Pe-chora, Lena- Pe-chorskaya, Leno-Vily-ui OGP	Caspian, Barents-Kara OGP	East Arctic prospective OGPP	Sea of Okhotsk OGB	Black Sea OGB	Baikal OGPB
Generation sources of H_2:											
a) serpentinization	*	*	*	*	?	?	*	?	?	?	?
b) radiolysis	*	*	*	*	?	?	*	?	?	?	?

Ending

c) microbiological processes	?	?	?	?	*	?	?	?		*	
Predominant transit direction: a) vertical b) lateral	* *	* *	* ?	?	?	?	* *	?	?	?	?
Predicted accumulation features: a) H_2 focal type accumulations, small in size, frequently occurring b) H_2 reservoir type accumulations, large, rarely occurring	*	*	*	?	?	?	?	?	?	?	?
	–	–	?	?	?	?	*	?	?	?	?
Most promising regional objects	Baltic Shield						Caspian OGP				
Priority objects for placement of scientific and technological test sites	Onega trough						Astrakhan arch				

Note: provinces and basins are distinguished considering the geodynamic regime of the Earth's crust development: provinces – platform regime, basins – block-orogenic and fold-orogenic. The selection of regional search objects was performed using the following materials:
(1) structural map of the basement surface of the USSR platform territories at scale 1:5,000,000 (1982) edited by V.V. Semenovich, L.I. Rovnin, N.V. Nevolin et al.
(2) geodynamic map of Russia, scale 1:10,000,000 (1995) edited by N.V. Mezhelovsky, A.I. Burde.
(3) scheme of thicknesses of the unexplored deep part of the sedimentary cover section of oil and gas provinces of Russia and adjacent countries (Yu.A. Volozh, M.A. Fedonkin, *V.M. Tolkachev*. On new approaches to exploration in traditional production provinces //: Oil and Gas Vertical. 2020. No. 7. P. 74.
(1) the following notations are used to indicate the significance of characteristics: * – significant manifestation; – absence of manifestation; ? – nature of manifestation not identified.
(2) -OGP – oil and gas province; OGPP – oil and gas prospective province; OGB – oil and gas basin; OGPB – oil and gas prospective basin

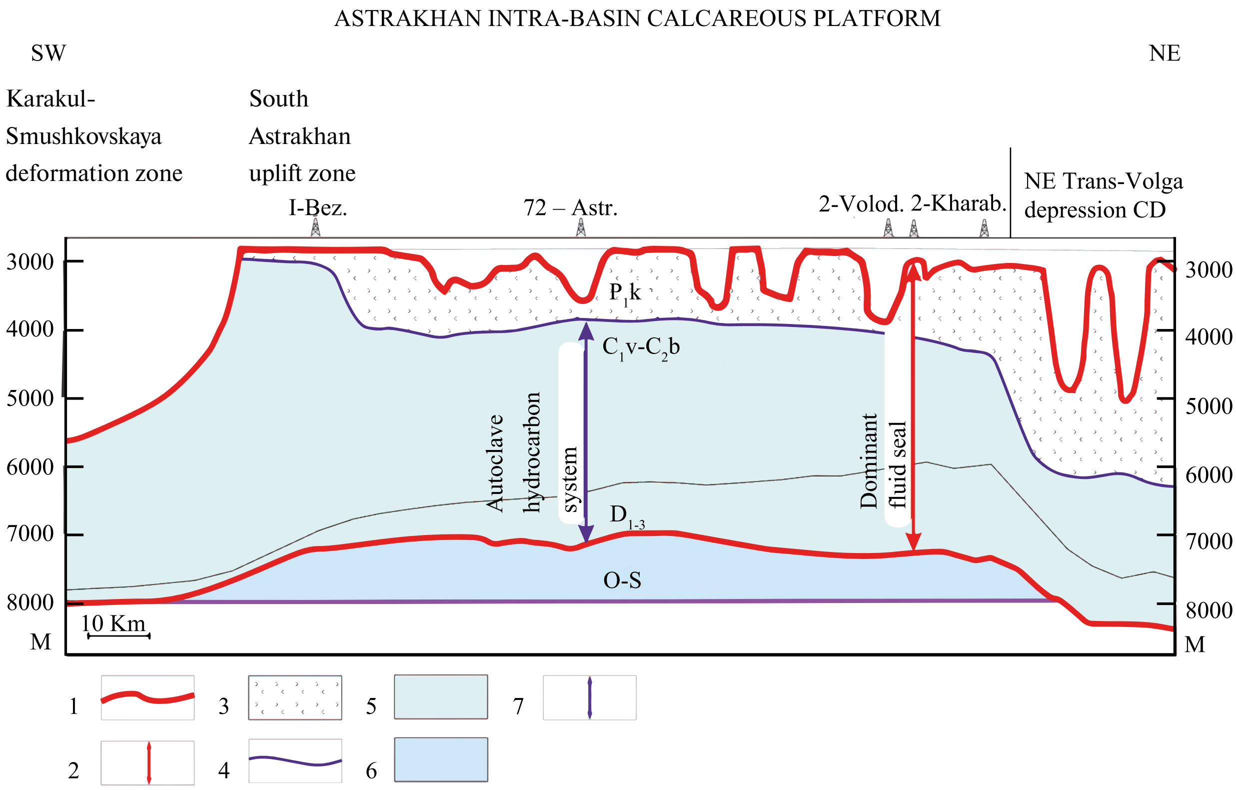

Fig. 5. Geofluid dynamic model of the Astrakhan platform, within which a scientific and technological testing site can be located. Dominant fluid seal: 1 – boundaries, 2 – distribution area (thickness 4–8 km); 3 – salt-bearing stratum with a thickness of 3–6 km; 4 – sulfate-carbonate-clay horizon at the base of the salt-bearing stratum; autoclave hydrocarbon system: 5 – distribution area, 6 – thickness 4-8 km; 7 – area of potential hydrogen accumulation.

need to create scientific and technological test sites for studying hydrogen as a mineral resource was previously raised in a work by one of the authors of this article; the latter also formulated the main tasks that are advisable to study at these test sites [14]. With some additions, they are reduced to the following: (1) development of a terminological glossary of hydrogen geology; (2) creation of a classification of planetary gases based on hydrogen content; (3) formation of a program for geophysical, geochemical, and hydrogeological studies of test wells; (4) transition from indicators (prerequisites) to criteria of hydrogen prospectivity for various geological and tectonic settings; (5) substantiation of reliable methods for delineating natural hydrogen accumulations, estimating hydrogen reserves (resources); (6) testing various criteria for ranking H_2 deposits by their reserve size and depth of placement; (7) investigating the possibilities and feasibility of accounting for volumes of sorbed and water-dissolved hydrogen when assessing resources; (8) evaluating the significance of differences in hydrogen accumulation mechanisms under conditions of closed and open hydrodynamic systems.

In terms of geology and field development, it is important to: (1) clarify the conditions for hydrogen embrittlement of reservoir rock cement and the geomechanical consequences of active hydrogen degassing; analyze the possibilities of inhibiting undesirable consequences of these processes; (2) monitor the stability of natural hydrogen emanation flow rates; (3) analyze the activity of hydrogenation of organic matter and hydrocarbon compounds under mass exposure to transit hydrogen flows; (4) account for hydrogen-induced processes of methane and carbon dioxide generation under reservoir conditions; (5) select criteria for assessing hydrogen purity, qualitative and quantitative composition of impurities.

When selecting locations for scientific and technological testing sites, preference should be given to objects located within oil and gas provinces/basins with a sedimentary cover thickness of at least 5–7 km. These may include depressions at the boundaries of ancient and young platforms, as well as depressions of intracontinental rifts. Additionally, platform shields, tectonic-sedimentary provinces of mobile belts: intermontane depressions, mediterranean and marginal-continental seas, and depressions of intercontinental rifts can be added as potential testing sites.

Given the absence of any specially conducted studies on natural hydrogen, the authors propose selecting one object each from the presumed most promising and less promising areas as priority objects. According to the authors, two testing sites within the East European platform, provisionally named “Onega” and “Astrakhan,” can be selected for this purpose.

The “Onega” testing site can be located in the area of the Onega parametric well (depth 3537 m), which penetrated the dominant fluid seal at a depth of 2944 m, represented mainly by halite [10]. The Onega structure shows an analogy with the geological structure of the industrial hydrogen development site (Bourakebougou, Mali), but has more favorable conditions for preserving this gas.

The “Astrakhan” testing site can be located on the Astrakhan arch in the area of the Astrakhan 2-D well, which penetrated a thick deposit layer (6–9 km) at a depth of more than 6 km, serving as a dominant fluid seal (Fig. 5). The predicted hydrogen accumulation zone may be located below the base of the Domanic shale sequence, which simultaneously serves as the base of the predicted autoclave hydrocarbon system [1].

It is predicted that the combination of lithological type of isolation (through salt and shale strata) with geofluid dynamic screening (due to the formation of a giant hydrocarbon system with extremely high reservoir pressures) creates unique conditions for the preservation of hydrogen (as well as helium) under the bottom of the autoclave hydrocarbon system. Such a geological situation should be considered as a very favorable environment for the formation of an industrially significant hydrogen (and helium) deposit.

CONCLUSION

1. Due to high geological risks caused by poor geological understanding of the processes of generation and accumulation of natural hydrogen, currently only preliminary studies are available at scientific and technological test sites, clarifying the conditions for the localization of hydrogen in various geological and tectonic settings. Any plans to initiate hydrogen exploration should be considered premature at present.

2. Large and unique H_2 deposits within oil and gas provinces/basins are most likely to be encountered at great depths and under autoclave hydrocarbon systems.

3. The most optimal solution (from scientific and practical perspectives) is the development of methods for extracting hydrogen of geological origin together with traditional energy carriers, helium, industrially valuable components of associated underground waters and, more broadly, hydrothermal resources and heat from the Earth’s interior. This approach will reduce geological, technical and economic risks of exploration activities, best meets the requirements of ecologization of the geological industry, and will provide a scientific and practical basis for the comprehensive development of energy and mineral resources of deep horizons of the lithosphere, which can be considered a task of the national level. In this regard, it is necessary to develop a modern concept for the search and development of resources of liquid and gaseous minerals within the deep horizons of the sedimentary cover of oil and gas provinces and megabasins, as well as the consolidated continental crust in zones where it outcrops within the shields of ancient platforms.

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