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Research on the prospects for obtaining iodine from carbohydrate fields in Ukraine

The article describes that industrial waters are limited to deep parts of heavy hydraulic structures, mainly in zones of slow and very slow water exchange. It has been established that the production of iodine from reservoir waters is profitable under the condition that its concentration in the water is 18 mg/l. It is described that the concentration of iodine in formation waters of the Poltava region is 15-115 mg/l. Based on the results of water sample analysis, it was determined that the waters of the Andriyashiv gas condensate field have an iodine content of 15 mg/l or more. Water with an abnormally high iodine content of up to 115 mg/l was found among the samples of the Chizhivka field, the Rudivka gas field, the Lyman, and the Reshetnyaky oil and gas fields. It was determined that the specific gravity of the waters of the gas fields is usually low - from 1.071 to 1.085 g/cm3, and the water of the Reshetnyaky field is high - from 1.132 to 1.167 g/cm3. Formation waters of the following deposits were studied: Mashivka; Chutove; Raspashnivka; Novoukrainske; Lanna; Western Khrestyshche; Chervonoyarske; Western Starovirivske; Vedmedivka; Efremivka; Western Efremivske; Western Sosnivka; Kegichivka; Shebelinka from the intervals from 2520 to 5560 m. It was found that the mineralization of formation water was from 33.0 to 337.01 g/kg, the iodine content from 13.5 to 54.74 mg/l. It can be argued that the formation of groundwater with a high iodine content was significantly influenced by the considerable thickness of the sedimentary strata and the corresponding thermobaric conditions.

Keywords: reservoir water, iodine, deposit, raw materials

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Introduction.

Iodine is of great importance for human life and economic activity. Its global production is 15,000 to 20,000 tons per year. Iodine is one of the components traditionally obtained from hydromineral raw materials, including groundwater. It is widely used in medicine and the food industry, in the production of various chemicals. Its consumption and demand for it in the world are constantly growing. World reserves of iodine are estimated at approximately 15 million tons. According to various data, the deficit in demand for iodine is 900-1,500 tons per year. Prices for iodine on the world market, depending on its purity and characteristics, range from 16.0 to 30.0 dollars for 1 kg. Ukraine has reserves of associated and formation waters with industrial concentrations of valuable components, which the domestic industry feels an urgent need for [1].

The efficiency of toll water processing enterprises is quite high, especially taking into account the possibilities of their organization on the basis of oil, gas and gas condensate deposits. All this necessitates the development of new hydromineral resources, the development of new technologies for obtaining valuable components, in particular iodine, from hydromineral raw materials.

Ukraine's annual need for iodine for table salt iodization reaches 10 - 15 tons, the needs of the medical industry - 20 tons, for other branches of Ukraine's developed economy - 30 tons. The beginning of the history of oil production in Poltava region was the introduction of an oil well at the Radchenky field into industrial operation in September 1951 year. Commissioning was preceded by almost 20 years of intensive search and reconnaissance work in the eastern regions of Ukraine. Over the following years, dozens of oil and gas deposits were explored and put into operation in the region, including those that today constitute the raw material base of the Poltavanaftogaz oil and gas production management.

As a result of long-term exploitation, the oil deposits were significantly watered. Therefore, the main production volume is the associated formation waters. They are valuable hydro-mineral raw materials, in particular, they are characterized by industrial iodine content. Today, such valuable raw materials are irretrievably lost as a result of returning to the corresponding geological horizons to maintain reservoir pressure.

Review of the research sources and publications. Iodine in the underground waters of oil-bearing basins was discovered as early as the second half of the 19th century. Later, significant concentrations of iodine were found in groundwater in almost all oil fields of the world. There is no doubt that during the decomposition of dispersed organic matter contained in silts and sedimentary rocks, iodine is released from them and passes first into the silt and then into groundwater. [1].

According to O. Vinogradov, the accumulation of iodine in the underground waters of oil fields is connected with the history of oil formation. The formation of groundwater with a high iodine content is significantly influenced by the large thickness of sedimentary strata and elevated and high temperatures and pressures. According to the research of Y. M. Svoren (2006), the lower temperature limit for the release of iodine from the organo-mineral complex of sedimentary rocks and its accumulation in groundwater is 35–50 °C. However, the most intensive processes of destruction of iodine-containing organic substances occur at temperatures above 125–150 °C. [2].

The problem of extracting iodine from reservoir waters is considered in the works of Azerbaijani, American, and Japanese scientists [1, 3].

The results of the research highlight the main methods and directions for the modernization of the production of iodine, bromine and their compounds from reservoir waters. Iodine production in world practice is carried out on the basis of the following raw materials: formation waters, accompanying waters, seaweed (China), water from the production of saltpeter (Chile). According to regulatory documents, technical iodine of the 1st and 2nd grades contains, respectively: at least 99.0 and 97.5% of iodine; no more than 0.010 and 0.015% of chlorine and bromine; 0.1 and 0.2% of organic substances; 0.05 and 0.15% of ash [4].

Definition of unsolved aspects of the problem.

Today, there is no iodine production in Ukraine, and its extraction from formation waters of oil and gas fields of Ukraine is quite relevant.

Problem statement. The purpose of the research is to analyze the results of reservoir water samples from deposits of Ukraine since 2011.

Basic material and results. Industrial waters are natural underground waters that contain useful components in solution or their compounds in quantities that ensure their profitable extraction and processing. Iodine, bromine, Glauber salt, and soda are removed from industrial waters. Waters with high concentrations of boron, lithium, rubidium, germanium, uranium, tungsten and other substances are of industrial interest. In many countries of the world, industrial groundwater is the main source of iodine. More than 70% of bromine production is provided by industrial waters.

Industrial waters are confined most often to deep parts of heavy hydraulic structures, mainly in zones of slow and very slow water exchange. Structurally and tectonically, such systems correspond to syneclises and depressions of ancient platforms, as well as foothill depressions and intermountain depressions. Industrial waters are found in rocks of the most different geological age, composition and origin.

The depth of industrial waters varies widely from the first tens of meters to 4-5 km or more, the most common depths of their occurrence are 1000-3000 m. They are also characterized by very high pressures. In some areas, their piezometric levels in wells are set close to the surface and even higher (they spill over to the day surface).

Therefore, the existence of a genetic connection between the accumulation of iodine in underground waters and oil formation is not in doubt among researchers. Geologists use the increased content of iodine in groundwater and sedimentary layers of rocks to predict prospects for oil and gas potential [1,5].

The concentration of iodine in the waters of oil fields can vary widely. Reservoir water is under high pressure and has a high solubility. Groundwater is rich in various complexes of ionic, molecular and colloidal impurities, often saturated with gases. The degree of mineralization of groundwater usually depends on its chemical composition. In brines of medium concentration (100 - 150 g/l), sodium chloride most often prevails. In the USA and Italy, in addition to iodine and bromine, boric acid, tungsten, lithium, germanium are obtained from underground reservoir waters. A significant advantage of underground water as a raw material source of rare elements is the low cost of the product, because underground water is a complete raw material, some of its geochemical types have relatively high manufacturability, exploitation of water deposits of rare elements does not require significant capital investments. Therefore, in most countries (the USA, Italy, Israel, Japan, New Zealand, Iceland, Australia), technological research is constantly and systematically conducted to develop methods of extracting these elements from specific geochemical types of natural waters.

Within Ukraine, the Volyn-Podilsky, Dnipro-Donetsk, Black Sea artesian basins, fractured water basins of the Ukrainian shield, Donetsk, Crimean and Carpathian hydrogeological folded areas with small intermountain artesian basins are distinguished.

In the greater part of the territory, the layers of sedimentary deposits contain horizons of salty waters and brines with mineralization of 3 - 83 g/l and more and with the content of bromine and iodine ions from 20 mg/l [6].

When comparing the mineralization of surface and underground waters of the countries of the world (Fig. 1.), it was found that the highest mineralization was found in the waters of the United States, about 430 g/l.

Since mineralization depends on the chemical composition, it is possible to extract a larger amount of valuable raw materials from these waters.

The production of iodine from reservoir waters is profitable under the condition that its concentration in the water is 18 mg/l. In formation waters of the Poltava region, the concentration of iodine reaches 15-115 mg/l. Based on the results of water analyzes since 2011, a table of waters with high water content (15 mg/l and more) has been compiled.

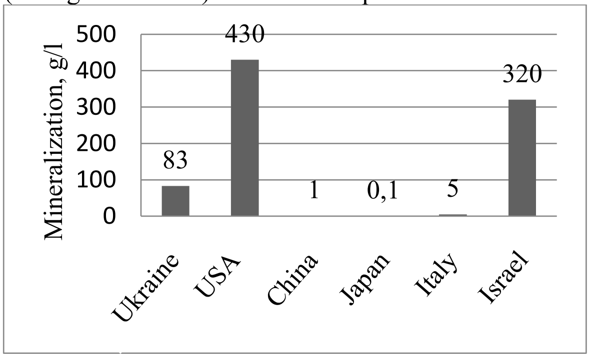

Figure 1 – Mineralization of waters of surface and underground sources [7].

Mineral iodine, bromine, and iodo-bromine waters accompany gas, oil, and gas condensate deposits, so they are most often brought to the surface by exploratory and exploitation wells for hydrocarbon minerals. But also beyond the boundaries of hydrocarbon deposits in the territories of the Dnipro-Donetsk, Black Sea, Lviv-Volyn depressions, the Pre-Kapatsky and Transcarpathian depressions, the Crimea and the folded region of the Carpathians, at depths of hundreds and thousands of meters, wells can encounter waters of increased mineralization, enriched in iodine and bromine to the level of which provides balneological conditions.

There is enough evidence of the distribution of iodine-bromine waters in the North Black Sea and Azov regions. In particular, in the area of the city of Berdyansk, Zaporizhzhia region, water with a bromine content of 38–138 mg/dm^3 and iodine content of 4–11 mg/dm^3 with a sodium chloride composition and mineralization of 11–60 g/dm^3 was found in Upper Cretaceous sands. In Koblevo (Mykolaiv Oblast), waters of the same composition were found in Miocene sediments. Water mineralization is 24.4 g/dm^3, bromine content is 48.8–56.6 mg/dm3. Bromine waters are widely distributed in the Azov region and are found in the areas of Melekino and Sedove (Donetsk region). In the Odesa region, bromine waters were found at a depth of 550 m in Upper Paleogene sands. Water mineralization is 23 g/dm^3, the bromine content is 52 mg/dm^3. Besides, sodium chloride waters with mineralization of 90–105 g/dm^3 and bromine concentration of 170 mg/dm3 and water temperature at the outlet of 35–45 ºC were discovered near Odesa by well No. 1–OT in Archaean–Proterozoic granites.

Crimea is very rich in mineral waters of this type, where unique springs are known. Well No. 905 near the city of Feodosia, which at a depth of 71 m in Lower Paleogene limestones, discovered an aquifer of sodium chloride waters with a mineralization of 7.8 g/dm^3 and an extremely high iodine content of 239 mg/dm^3. and bromine - only 57 mg/dm^3. The waters of the Sarta-Su spring, which flows from the Chokrat limestones near Lenino, also have a significant bromine content. The water of this source is poorly mineralized (3.2 g/dm^3), and the bromine content in it is 79 mg/dm^3.

The most famous resort where iodine-bromine mineral waters are used is Berdyansk (Zaporizhia region).

According to the results of the water sample analysis, it was determined that 6 water samples with an iodine content of 15 mg/l or more were taken from the wells of the Andriyashivka gas condensate field, 11 from the Chizhivka gas field, 9 from the Rudivka gas field, and 15 from the Lyman gas field. 9 samples of water with abnormally high iodine content - up to 115 mg/l were taken from the Reshetnyaky oil and gas field.

The specific gravity of gas field waters is usually low - from 1.071 to 1.085 g/cm^3, and the water of the Reshetnyaky field is high - from 1.132 to 1.167 g/cm^3.

Also, iodine and bromine are products of a single organic transformation process that takes place under conditions of high temperatures and pressures. Further, hydrocarbons and their accompanying deep chloride-sodium solutions with iodine and bromine move along zones of major tectonic disturbances to higher zones of the earth's crust. At these depths, the lithological and structural conditions are favorable for the formation of accumulations of oil, gas and accompanying iodobromine waters. The latter are localized in artesian basins associated with large tectonic structures.

The formation of groundwater with a high iodine content was significantly influenced by the considerable thickness of the sedimentary strata and the corresponding thermobaric conditions. It was established that the lower temperature limit for the release of iodine from the organo-mineral complex of sedimentary rocks and its accumulation in underground waters is 35-50°C. However, the most intensive processes of destruction of iodine-containing organic substances occur at temperatures above 125-150 °C.

The water extracted simultaneously with the oil of the NGDU "Poltavanaftogaz" is enough to extract approximately 1 ton of iodine per year [8]. Therefore, in the case of organizing production, it is necessary to use conserved water wells with their appropriate commissioning or drill new wells and develop aquifers of deposits.

An example of such a field is the Reshetnyaky mineralized water gas and oil field in the Novosanzhar district of the Poltava region, which is characterized by a high content of iodine and bromine in formation waters (Table 1).

Table 1 - Analyzes of water with high iodine content

№	Date of selection samples	Name of the field, wells	Well number	Iodine content, mg/l
1	2	3	4	6
2	28.12.2011	Andriyashivka	55	21,15
3	13.05.2014	Andriyashivka	51	29,61
4	31.01.2011	Chizhivka	37	34,90
5	22.05.2013	Chizhivka	58	25,38
6	16.05.2011	Rudivka	17	57,11
7	08.07.2014	Rudivka	1	39,13
8	24.05.2013	Lyman	17	34,90
9	03.06.2014	Lyman	52	37,01
10	18.06.2014	Lyman	16	29,61
11	04.03.2013	Holubivka	103	30,67
12	16.07.2014	Chervonozavodske	5	72,97
13	18.05.2013	Reshetnyaky	55	101,52
14	18.05.2013	Reshetnyaky	55	112,10

According to preliminary data (unpressurized water sampling, long-term storage of samples for analysis, etc.), the iodine content in the field water ranges from 15 to 50 mg/l. With an average content (30 mg/l), iodine reserves will amount to 7128 tons.

The highest concentrations of iodine and bromine are confined to chloride-sodium waters of increased mineralization. Iodine-bromine mineral waters gravitate to the zones of large ruptured landslides, which are channels for deep underground water.

Reservoir water for research was sampled at fields of the Mashiv-Shebelinka gas-bearing district [9-13]. It belongs to the Eastern oil and gas region of Ukraine. It is located on the continuation of Glynsko-Solokha, and covers the most submerged part of the axial zone of the Dnieper-Donets Trough. It is on the territory of this district that such well-known deposits as Shebelinka, Western Khrestyshche, Efremivka, etc. are located. Industrial productivity has been established by exploratory and exploratory drilling in Lower Permian and Upper Carboniferous deposits. Most of the deposits of the area, which are represented by massive stratified deposits, sometimes limited by salt diapirs and disjunctive disturbances, are confined to two long structural zones, which are simultaneously gas accumulation zones with double structural control — Raspashnivsko-Melykhivska and Sosnivsko-Belyaevska [10].

The development of a thick layer of rock salt deposits of the Lower Permian age and active salt tectonics of Devonian rock salt on the territory of the region led to the formation of unique mushroom-like salt bodies. Under their visors there are industrial accumulations of natural gas in the above-mentioned two gas accumulation zones. The main volume of explored hydrocarbon reserves is located in sandy reservoirs, but a part is concentrated in chemogenic cavernous layers (mainly anhydrites) of the Lower Permian. The data obtained during the drilling of the deepest wells indicate a higher position of the upper limit of active catagenesis than in the neighboring Glynsko-Solokha region. In addition to those listed, there are oil and gas storage zones with one and a half structural control: Shebelinska, Kopylivsko-Skhidno-Poltavska, Mashivsko-Yelizavetivska, Maryanivsko-Lanivska and Yefremivska. Formation waters of the following deposits were studied: Mashivka; Chutove; Raspashnivka; Novoukrainske; Lanna; Western Khrestyshche; Chervonoyarske; Western Starovirivske; Vedmedivka; Efremivka; Western Efremivske; Western Sosnivka; Kegichivka; Shebelinka from the intervals from 2520 to 5560 m. It was found that the mineralization of formation water was from 33.0 to 337.01 g/kg, the iodine content from 13.5 to 54.74 mg/l.

After the analysis of formation water samples from the fields, the highest iodine content was found at the Kegichivka field - 54.74 mg/l, and the mineralization of the formation water was 256.14 g/kg [9].

Conclusions. It is necessary to investigate a number of promising deposits of reservoir waters containing iodine for industrial extraction. Next, approve their reserves at the level of the state commission on reserves. The development of new deposits will provide a full-scale approach to the comprehensive development of iodine-bromide water reserves. Further, it is necessary to take measures to expand the production capacity of iodine production and obtain various chemical products from it. Groundwater is a valuable source because it contains not only iodine and bromine, but also chlorides of sodium, calcium, magnesium and other chemical elements. Their effective use is largely determined at the current stage by their availability on the market. Therefore, in parallel with the restoration of wells and the increase of production capacity, it is necessary to conduct work on the study of the domestic and foreign markets of iron bromide, ozokerite, technical carbon and other commodity products of the chemical industry.

Table 2 - Characteristics of reservoir waters of productive horizons

The name of the field	Horizon index	Mineralization, mg/l	Iodine content, mg/l
Mashivka	G-13	68,17	10,55
Chutove	A-2	307	13,7
	A-5	299	36,1
	A-6-8	333	42,0
Raspashnivka	G-9-12	337,01	45,75
Novoukrainske	A-3	263,2	48,09
Lanna;	A-8	321,46	36,50
Western Khrestyshche	G-11	315	25,1
	K-1	313	24,7
Chervonoyarske	G-13	289	15,25
Western Starovirivske	G-10н	201,359	21,0
Vedmedivka	G-10-G-13	332,2	18,61 45,68
Efremivka	A-6-8, G-3-4	220 330	40,6
Western Efremivske	G-8-13	260 270	13,5 26,2
Western Sosnivka	A-7-A-8	178,3 318,2	13,55 46,55
Kegichivka	A-6-7	256,14	54,74
Shebelinka	A-5	33,0 318,9	36,0

Therefore, the deep formation waters of the deposits of Ukraine are a promising raw material for profitable iodine extraction, which can be carried out in three directions:

1) use of available waters associated with oil production;2) use of conserved and liquidated water wells of oil deposits;
3) drilling new wells into aquifers.

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Дослідження перспектив вилучення йоду з родовищ вуглеводнів України

При значному обводнені нафтової продукції головний об'єм видобутку становлять супутні пластові води, які є цінною гідромінеральною сировиною, зокрема характеризуються промисловим умістом йоду. Значні концентрації йоду знайдено у підземних водах майже в усіх нафтових родовищах світу. Промислові води приурочені як найчастіше до глибоких частин важких гідравлічних структур, переважно у зонах сповільненого та дуже сповільненого водообміну. У стрктурно-тектонічному відношенні такі системи відповідають синеклізам та западинам стародавніх платформ, а також передгірським прогинам і міжгірським западинам. Описано існування генетичного зв'язку між накопиченням йоду в підземних водах і нафтоутворенням. На більшій частині території світу в товщі осадових відкладень містяться горизонти солоних вод і розсолів з мінералізацією 3 − 83 г/л і більше та з вмістом іонів брому, йоду від 20 мг/л. виявлено, що найбільшу мінералізацію виявлено у водах США близько 430 г/л. Мінералізація залежить від хімічного складу, то більшу кількість цінної сировини можливо вилучити саме з високомінералізованих вод. Встановлено, що виробництво йоду з пластових вод рентабельне за умови його концентрації у воді 18 мг/л. У пластових водах Полтавського регіону концентрація йоду сягає 15 – 115 мг/л. За результатами аналізів проб води встановлено, що води Андріяшівського газоконденсатного родовища мають уміст йоду 15 мг/л і більше. Серед проб Чижівського родовища, Рудівського газового родовища, Лиманського, Решетняківського нафтогазового родовища виявлено води з аномально високим умістом йоду – до 115 мг/л. Встановлено, що питома вага вод газових родовищ зазвичай низька – від 1,071 до 1,085 г/см^3, а вод Решетняківського родовища висока – від 1,132 до 1,167 г/см^3. Досліджено пластові води таких родовищ: Машівське; Чутівське; Розпашнівське; Новоукраїнське; Ланнівське; Західно-Хрестищинське; Червоноярське; Західно-Старовірівське; Ведмедівське; Єфремівське; Західно-Єфремівське; Західно-Соснівське; Кегичівське; Шебелинське з інтервалів від 2520 до 5560 м. Виявлено, що мінералізація пластової води склала від 33,0 – 337,01 г/кг, вміст йоду від 13,5 до 54,74 мг/л. На формування підземних вод з високим умістом йоду значно вплинули значна потужність осадових товщ та відповідні термобаричні умови. Підтверджено, що нижня температурна межа виділення йоду з органо-мінерального комплексу осадових порід і накопичення його в підземних водах становить 35 – 50^{o}C. При організації виробництва потрібно використовувати законсервовані обводнені свердловини з відповідним їхнім уведенням в експлуатацію або бурити нові свердловини і розробляти водоносні горизонти родовищ.

Ключові слова: пластова вода, йод, родовище, сировина

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