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INNOVATIVE METHODS FOR RESTORING FILTRATION CHARACTERISTICS OF BOREHOLE URANIUM ORES IN KAZAKHSTAN'S FIELDS

Abstract. The practice of operating technological wells in deposits with low filtration characteristics of ores shows that over time there is a decrease in their productivity. The aim of the study is to increase the efficiency of downhole uranium production by selecting special decolmating solutions and selecting rational parameters of the technology of influencing the near-filter zone of the formation of geotechnical wells, improving the filtration characteristics of the formation depending on the mineralogical composition and structure of sedimentary materials. The main method of research is the sampling of sedimentation from the productive horizon at the uranium deposits of the Shu-Sarysu and Syrdarya depressions. Quantitative and qualitative characteristics and features of mineral compositions were determined by the X-ray phase method. A technique was developed and laboratory experiments were performed on the processing of sedimentation samples by the drip method using selected decolmating solutions.

Key words: *downhole production, sedimentation, decolmating solution, X-ray phase analysis, microscopic studies, colmatation, uranium production, process solutions, sedimentary materials, mineralogical composition.*

Қазақстан кен орындарында уранды ұңғымалық өндіру кендерінің сүзу сипаттамаларын қалпына келтірудің инновациялық әдістері

Аңдатпа. Кендердің сүзілу сипаттамалары төмен кен орындарында технологиялық ұңғымаларды пайдалану тәжірибесі уақыт өткен сайын олардың өнімділігінің төмендейтінін көрсетеді. Зерттеу мақсаты арнайы декольматирлеуші ерітінділерді іріктеу және геотехнологиялық ұңғымалар қыртысының сүзгіш аймағына әсер ету технологиясының тиімді параметрлерін таңдау есебінен уранды ұңғымалық өндірудің тиімділігін арттыру, минералогиялық құрамы мен тұнба түзуші материалдардың құрылымына байланысты қыртыстың сүзгіш сипаттамаларын арттыру болып табылады. Зерттеудің негізгі әдістері Шу-Сарысу және Сырдария депрессиясының уран кен орындарында өнімді горизонттан тұнба түзілу сынамаларын іріктеу болып табылады. Рентгендік фазалық әдіспен минералдардың сандық және сапалық сипаттамалары мен құрамының ерекшеліктері анықталды. Таңдалған деколматизация ерітінділерін қолданып, тұндыру сынамаларын тамшылату әдісімен өңдеу әдістемесі әзірленді және зертханалық тәжірибелер жүргізілді.

Түйінді сөздер: *ұңғымаларды өндіру, тұндыру, деколматизациялау ерітіндісі, рентгендік фазалық талдау, микроскопиялық зерттеулер, уран өндіру.*

Инновационные методы восстановления фильтрационных характеристик руд скважинной добычи урана на месторождениях Казахстана

Аннотация. Практика эксплуатации технологических скважин на месторождениях с низкими фильтрационными характеристиками руд показывает, что с течением времени наблюдается снижение их производительности. Целью исследования является повышение эффективности скважинной добычи урана за счет подбора специальных декольматирующих растворов и выбора рациональных параметров технологии воздействия на прифильтровую зону пласта геотехнологических скважин, повышение фильтрационных характеристик пласта в зависимости от минералогического состава и структуры осадкообразующих материалов. Основными методами исследований является отбор проб осадкообразования из продуктивного горизонта на урановых месторождениях Шу-Сарысуйской и Сырдариинскй депрессии. Рентгенофазовым методом установлены количественно-качественные характеристики и особенности составов минералов. Разработана методика и произведены лабораторные опыты по обработке проб осадкообразования капельным методом с применением подобранных декольматирующих растворов.

Ключевые слова: *скважинная добыча, осадкообразование, декольматирующий раствор, рентгенофазовый анализ, микроскопические исследования, добыча урана.*

Introduction

Climate change due to disproportionate human production activity on the planet is becoming more and more tangible [1]. Concrete measures are being taken to protect and preserve biodiversity and reduce the negative impact of climate change in the World. In this regard, the recognition of nuclear generation projects for green energy types will double electricity generation by 2050, which will make an important contribution to the fight against global warming [2]. According to the IAEA research, nuclear power has a significant potential to reduce greenhouse gas emissions to mitigate the effects of climate change in certain regions of Europe, Asia and Africa (IAEA) [3, 4]. The growth of nuclear energy will lead to an increase in the demand for natural uranium and its products. The uranium industry of Kazakhstan, based on progressive, highly efficient borehole extraction of uranium ores, can make a worthy contribution to solving the issues of natural uranium supplies.

Kazakhstan has 14% of the world's proven uranium reserves and ranks second after Australia, with 70% of them suitable for downhole development. Borehole development of uranium ores in the Republic of Kazakhstan is carried out at 26 sites, united in 13 uranium mining companies. The total volume of natural uranium production is more than 40% of the global level [5].

Uranium deposits in Kazakhstan are located in six provinces: Shu-Sarysui, Syrdarya, North Kazakhstan, and the Caspian region, Balkash, Ili regions. The first two

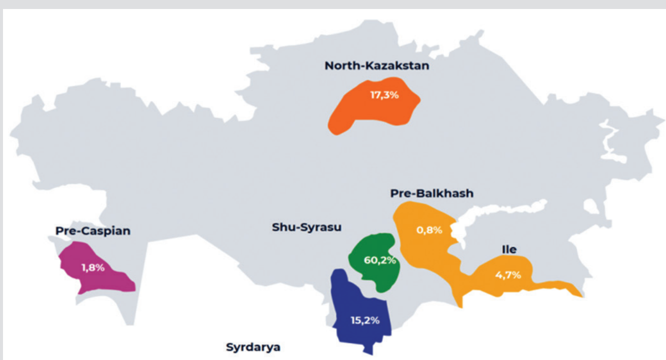

Figure 1. Scheme of location of regions of explored uranium deposits.
Сурет 1. Барланған уран кен орындары өңірлерінің орналасу схемасы.
Рис. 1. Схема расположения регионов разведанных месторождений урана.

Table 1

Mineralogical composition of sediments from the uranium deposit

Кесте 1

Уран кен орнының шөгінділерінің минералогиялық құрамы

Таблица 1

Минералогический состав осадков из месторождения урана

Minerals	Formula	Syrdarya depression (concentration, %)	Chu-Sarysui depression (concentration, %)
Gypsum	$CaSO_4 \cdot 2H_2O$	52,0	81,8
Quartz	SiO_2	42,3	3,2
Potassium feldspar	$KAlSi_3O_8$	5,7	2,2
Sillimanite	$Al_2O_3 \cdot SiO_2$		12,8

provinces, located in the northern part of the country, are currently producing oil. Kyzylorda and Turkestan regions. They are shown schematically in Figure 1.

Downhole mining of minerals, in particular uranium, involves the dissolution of the useful component by a moving flow of solvent at the location of the ore body, followed by the removal and lifting of the formed compounds to the surface [6]. The positive aspects of using sulfuric acid solutions at enterprises in Kazakhstan are its low cost, widespread use in the national economy, and the possibility of complete dissolution of uranium mineralization [7]. However, there are negative aspects, such as the high reactivity of the interaction of sulfuric acid with carbonate and clay minerals of ore-bearing rocks. When sulfuric acid interacts with carbonate minerals, gypsum is formed, and clay minerals swell and increase in size, these factors prevent the leaching process [8].

Difficult-to-dissolve sediments and swollen clay particles in the productive horizon increase hydraulic resistance and form impenetrable sections of geochemical barriers that overlap the solution flow lines. As a rule, decrease in the filtration characteristics of the productive horizon leads to a decrease in the uranium content in the productive solution, and a decrease in the flow rate and the period of uninterrupted operation of wells. This increases the development period of technological units, as a result of which the consumption of sulfuric acid, electricity, and other operational components increases [9, 10]. In some cases, it is necessary to carry out costly, heavy complex treatments using drilling rigs, including washing, chemical treatment, swabbing and compressor pumping.

Laboratory research methods

Detailed determination of the physical and chemical characteristics of sediments will make it possible to develop more effective reagents and methods for restoring the filtration characteristics of ores in the borehole zone of the formation, ensuring an increase in productivity and uninterrupted operation of geotechnical wells. To determine the quantitative and qualitative characteristics of sedimentation, sediment samples were taken from the Shu-Sarysu and Syrdarya depressions.

X-ray diffractometric analysis was performed on an automated DRON-3 diffractometer with $Cu_{K\alpha}$-radiation, β-filter. Conditions for shooting diffractograms: $U = 35$ kV; $I = 20$ mA; shooting θ-2θ; detector 2 deg/min. X-ray phase analysis on a semi-quantitative basis was performed based on diffractograms of powder samples using the method of equal weightings and artificial mixtures. Quantitative ratios of crystal phases were determined. Interpretation of diffraction patterns was carried out using data from the ICDD card file: Powder diffraction data base PDF2 (Powder Diffraction File) and diffractograms of minerals free of impurities. For the main phases, the content was calculated. Figures 2 and 3 show diffractograms of samples in which crystal phases were identified by the radiation intensity.

Research results

According to X-ray phase analysis, the sediments have high crystallization in several phases. B Table 1 shows the results of X-ray phase analysis of sedimentation from the Shu-Sarysu uranium deposit and The Syrdarya depression.

Table 1 shows that the basis of the sample from the Syrdarya depression deposit was made up of chemical compounds $Ca(SO_4)(H_2O)_2$ (52%) and SiO_2 (42%), minerals – gypsum and quartz. About the steel part of the sample is potassium feldspar (5,7%). The characteristics of samples from the Syrdarya depression indicate a combined origin of sedimentation, chemical-gypsum, and mechanical-quartz and potassium feldspar. The bulk of the sample from the Shu-Sarysu depression consists of the chemical compound $Ca(SO_4)(H_2O)_2$ (81,8%), the mineral gypsum. The rest of the sample consists of quartz (3,2%), potassium feldspar (2.2%), sillimanite (12,8%). The data of the samples of the Shu-Sarysu depression show the predominance of the chemical origin of gypsum sedimentation and the presence of mechanical impurities of quartz and potassium feldspar.

Analysis of X-ray images of samples from different deposits shows that the main sedimentary component of colmatant is calcium sulfate $CaSO_4 \cdot 2H_2O$. In this regard, it can be said that the main cause of colmatation is the interaction of a leaching solution of uranium leaching with calcium carbonate, which proceed according to the formula:

$$CaCO_3 + H_2SO_4 = CaSO_4\downarrow + H_2CO_3.$$

For effective destruction and prevention of such sedimentation, it is necessary to develop a decolmating solution using hydrofluoric acid with the addition of surfactants with complexing properties. To improve the solubility, sulfamic acid was used as a surfactant, which has the properties of lowering pH and binding metal ions.

Laboratory experiments on the selection of chemical reagents

Experiments on the treatment of sediments were carried out on samples from the same sample with different compositions of chemical reagents of decolmating solutions. To determine the effective composition of the solution, the most solvent properties were selected. The experiment included treatment with a solution of ammonium bifluoride (5,0%) and sulfuric acid (10,0%), Surfactant (1,0%) and industrial water (84%). The choice of ammonium bifluoride as the main component is due to its high ability to exchange reactions with mineral acids (sulfuric, hydrochloric, nitric acids) and the formation of hydrofluoric acid according to the formula:

$$NH_4HF_2 + H_2SO_4 = NH_4SO_4 + HF.$$

$$CaSO_4{\cdot}2H_2O + 2HF = CaF_2 + H_2SO_4 + 2H_2O.$$

$$CaAl_2SiO_8 + 16HF = 2AlF_3 + 2SiF_4 + 8H_2O + CaF_2.$$

$$6HF + SiO_2 = SiF_4 + 2HF + 2H_2O.$$

As a result of the interaction of hydrofluoric acid with sedimentation, both the colmatant and part of the terrigenous component of the sands are dissolved, which increases the effective porosity of the ore block massif. The addition of surfactants increases the interaction of hydrofluoric acid with sedimentary minerals. At the same time, hydrofluoric acid is completely utilized due to the large amount of quartz contained in the sands.

Discussion of the results of laboratory tests

After carrying out laboratory experiments on the processing of samples by the drip method with a different composition of decomposing solutions, the sedimentation was dried at room temperature. A scanning electron microscope was used for a detailed examination of the sample surface. A comparative analysis of the images after processing with a particular solution and comparing it with the original image allowed us to visually establish the effectiveness of the composition of the decollating solution.

Images of the precipitation surface before and after treatment with various solutions were recorded using a high-resolution analytical scanning electron microscope. It is manufactured for a wide range of research tasks and quality control at the submicron level of Tescan MIRA 3 FEG-SEM. SEM TESCAN MIRA electron column, electron source: Schottky auto-emission cathode. The energy range of the electron beam incident on the sample is from 200 eV to 30 keV (from 50 eV with the option of braking the BDT beam). To change the beam current, an electromagnetic lens is used as an aperture changer. Beam current: from 2 nA to 400 nA with continuous adjustment. Maximum field of view: more than 8 mm at WD = 10 mm, more than 50 mm at maximum WD. Electron column resolution, high vacuum mode 1.2 nm at 30 keV, detector SE. 3.5 nm at 1 keV, In-Beam detector SE. 1.8 nm at 1 keV, beam braking option BDT. Figure 4 shows images of samples from the Syrdarya and Shu-Sarysu depressions before and after treatment with special solutions.

It can be seen from Figures 4a and 4b that the surface of the initial sample is formed of dense lamellar crystals with sizes from 5 µm to 30 µm with a characteristic frame structure without breaks and cracks in the body. The crystal shapes are elongated with a chaotic arrangement and uniform surface relief. It can be seen from Figures 4c and 4d that after treatment with decollating solutions, there was a noticeable destruction of the structure and a change in the shape of crystals with a decrease in their size and density with the formation of small loosened flakes. It can be seen the rounded edges of the crystals and the formation of cracks in the bodies. The arrangement of crystals has become less dense with the formation of voids and gullies in the pore space. Partial dissolution of the sample is noticeable, the

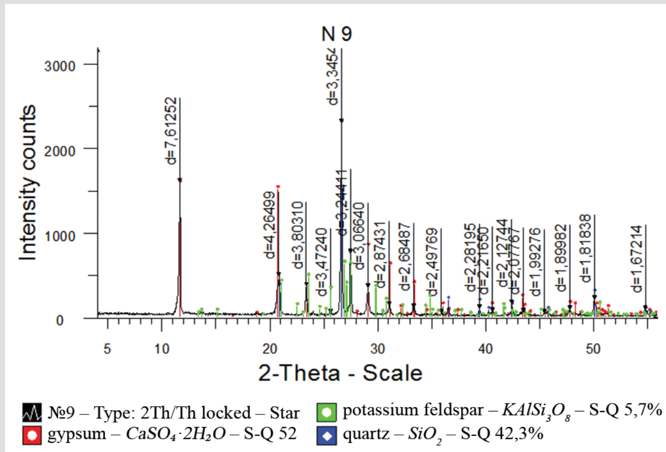

Figure 2. Diffraction pattern of the sample of the Syrdarya depression.

Сурет 2. Сырдария депрессиясы үлгісінің дифрактограммасы.

Рис. 2. Дифрактограмма образца Сырдарьинской депрессии.

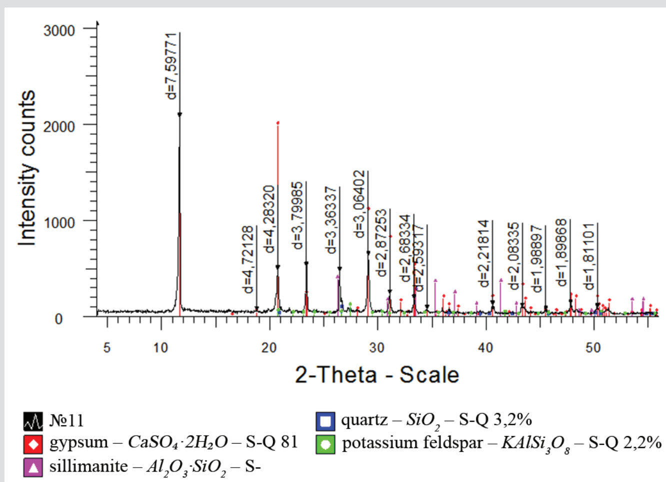

Figure 3. Diffraction pattern of a sample of the Chu-Sarysu depression.

Сурет 3. Шу-Сарысу депрессиясы үлгісінің дифрактограммасы.

Рис. 3. Дифрактограмма образца Чу-Сарысуйской депрессии.

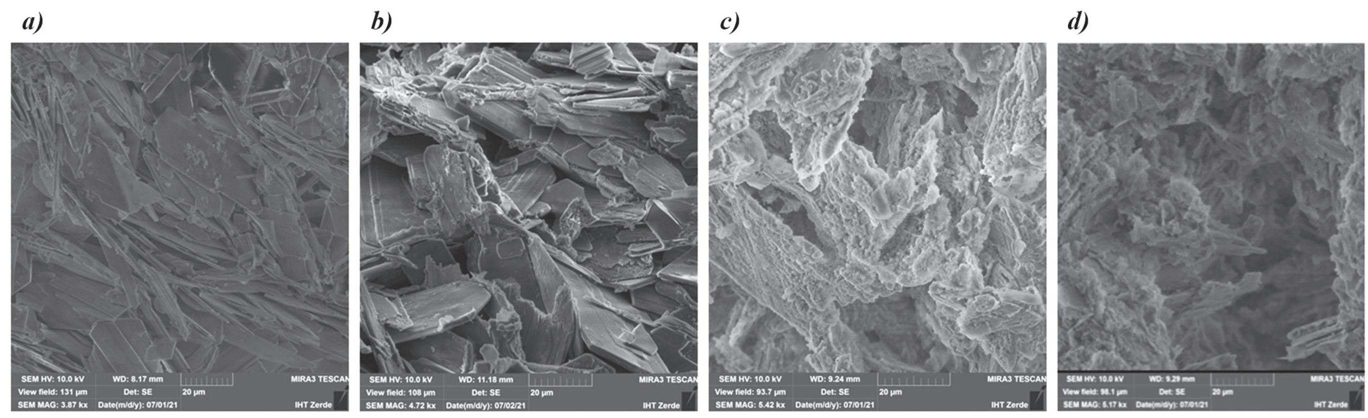

Figure 4. Image of surface samples of Syrdarya and Shu-Sarysu depression:
***a* and *b* – initial samples; *c* and *d* – after the experiment.**
Сурет 4. Сырдария және Шу-Сарысу депрессиясы сынамаларының үстіңгі бетінің бейнесі:
***a* және *b* – бастапқы сынамалар; *c* және *d* – тәжірибеден кейін.**
Рис. 4. Изображение поверхности проб Сырдарьинской и Шу-Сарысуйской депрессии:
***a* и *b* – исходных проб; *c* и *d* – после опыта.**

crystal sizes decreased from 30 μm to 15 μm. This is due to the dissolution of part of the sample in hydrofluoric acid.

The application of the decollating solution must be carried out according to a special method on special technological equipment. The innovative method provides for the treatment of the filter area of the well with a decomposing solution directly and the maximum destruction and prevention of sedimentation in the formation. The method provides an increase in the productivity of operational units and the completeness of metal extraction from them by removing and preventing sedimentation in a porous medium. In addition, a reduction in the specific costs of sulfuric acid, electricity, labor costs and other production costs is achieved in the process of borehole extraction of uranium from various mining and geological blocks.

Conclusions

The quantitative and qualitative studies of the sedimentation composition of the deposits of the Shu-Sarysu depression indicate that the main part of the sample is gypsum (81,8%). The rest of the sample consists of quartz (3,2%), potassium feldspar (2,2%), sillimanite $Al_2O_3{\cdot}SiO_2$ (12,8%). The basis of the sample of the Syrdarya depression deposit consists of gypsum (52%) and quartz (42,3%), the rest of the sample consists of potassium feldspar (5,7%).

Due to the interaction of sulfuric acid solutions with carbonate minerals, sedimentation is deposited mainly in the discharge zone. They cause low efficiency of hydro-dynamic methods and difficulties in processing technological blocks, lead to a decrease in the productivity of production and intake of injection wells due to the deterioration of the filtration characteristics of the formation. They excite additional costs for restoring the permeability of the productive horizon and increasing the productivity of production and intake of injection wells, increase the operating costs of developing blocks.

Preparation of a decomposing solution based on ammonium bifluoride (5%), sulfuric acid (10%) and surfactants in small quantities can increase the dissolving ability of the decomposing solution and prevent sedimentation in the formation for a longer time.

The developed technique for restoring the filtration characteristics of the productive horizon based on the treatment of the filter part of wells allows to reduce the specific consumption of chemical reagents and increase the efficiency of the decomposing solution.

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