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ХАБАРЛАРЫ

ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ
НАУК РЕСПУБЛИКИ
КАЗАХСТАН
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NEWS

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NAS RK is pleased to announce that News of NAS RK. Series of geology and technical sciences scientific journal has been accepted for indexing in the Emerging Sources Citation Index, a new edition of Web of Science. Content in this index is under consideration by Clarivate Analytics to be accepted in the Science Citation Index Expanded, the Social Sciences Citation Index, and the Arts & Humanities Citation Index. The quality and depth of content Web of Science offers to researchers, authors, publishers, and institutions sets it apart from other research databases. The inclusion of News of NAS RK. Series of geology and technical sciences in the Emerging Sources Citation Index demonstrates our dedication to providing the most relevant and influential content of geology and engineering sciences to our community.

Қазақстан Республикасы Үлттық гылым ақадемиясы «ҚР ҰҒА Хабарлары. Геология және техникалық гылымдар сериясы» гылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрi the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруды. Web of Science зерттеушілер, авторлар, баспашилар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Геология және техникалық гылымдар сериясы Emerging Sources Citation Index-ке енүі біздің қоғамдастық үшін ең өзекті және беделді геология және техникалық гылымдар бойынша контентке адалдығымызды білдіреді.

НАН РК сообщает, что научный журнал «Известия НАН РК. Серия геологии и технических наук» был принят для индексирования в Emerging Sources Citation Index, обновленной версии Web of Science. Содержание в этом индексировании находится в стадии рассмотрения компанией Clarivate Analytics для дальнейшего принятия журнала в the Science Citation Index Expanded, the Social Sciences Citation Index и the Arts & Humanities Citation Index. Web of Science предлагает качество и глубину контента для исследователей, авторов, издавателей и учреждений. Включение Известия НАН РК. Серия геологии и технических наук в Emerging Sources Citation Index демонстрирует нашу приверженность к наиболее актуальному и влиятельному контенту по геологии и техническим наукам для нашего сообщества.

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INTENSIFICATION OF INHIBITOR-ASSISTED URANIUM ISL PROCESS

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Abstract. Decrease in filtration characteristics of ores under complex mining and geological conditions affects the dynamics of uranium extraction from the subsurface and increases the repayment period of technological units and operating costs. In this regard, prevention of gypsum sludging during the in-situ leaching (ISL) of uranium ores for its intensification at enterprises in the Republic of Kazakhstan is a crucial task. Development of productive horizon treatment techniques with specific reagents accounting the mineralogical composition of ores and the structure of sludge-forming materials shall increase the efficiency of the mining. The X-ray phase studies of core mineralogical composition and sludging of productive horizons in Santonian, Maastrichtian and Campanian ore intervals allowed determining the factors impacting the ore filtration characteristics. Laboratory tests on agitated leaching of uranium from core samples using inhibiting additives enabled to select an effective composition and concentration of chemical reagents to prevent the gypsum sludging. The process borehole experiments resulted in development of a technique for treatment of productive horizons with chemical inhibiting agents. Laboratory studies on effective inhibiting additives for uranium ISL processes led to formulation of a productive horizon treatment procedure to provide the intensification of uranium recovery under complex mining and geological conditions. The developed method of uranium ISL intensified production allowed increasing its efficiency and reducing production costs by 5-7%. Besides, it reinforced the environmental and industrial safety of the uranium ISL process.

Key words: uranium leaching, inhibitor, sludging, well operation cycle, uranium content, leaching solution, productive solution

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ИНГИБИТОРЛАРДЫ ПАЙДАЛАНА ОТЫРЫП УРАННЫҢ ҰҢҒЫМАЛЫҚ ЖЕРАСТЫ ШАЙМАЛАНУЫН ҚАРҚЫНДАТУ

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Аннотация. Құрделі тау-кен-геологиялық жағдайларда кендердің сүзу сипаттамаларының төмендеуі жер қойнауынан уран алу динамикасына теріс әсер етеді және технологиялық блоктарды өндіреу кезеңін және өндіруге арналған пайдалану шығындарын арттырады. Осыған байланысты Қазақстан Республикасының қасіпорындарында уранды ұңғымалық өндіруді қарқыннату мақсатында уран кендерін жерасты шаймалау кезінде гипс тұнбасының пайда болуын болдырмау аса маңызды және өзекті міндет болып табылады. Кендердің минералогиялық құрамына және тұнба түзетін материалдардың құрылымына байланысты арналы реагент — ингибиторлармен өнімді горизонтты өңдеу технологиясын әзірлеу уранды жерасты ұңғымалық шаймалау тиімділігін арттыруға мүмкіндік береді. Керн үлгілерінің минералогиялық құрамын және Сантон, Маастрихт және Кампан кен аралықтарының өнімді горизонттарының шөгінділерін зерттеу үшін рентгендік фазалық әдісті қолдану кендердің сүзілүү сипаттамаларының төмендеуіне әсер ететін негізгі факторларды анықтауға мүмкіндік береді. Ингибиторлық қоспаларды қолдана отырып, керн сынамаларынан уранды агитациялық шаймалау бойынша зертханалық тәжірибелер гипс

тұнбаларының пайда болуын болдырмау үшін химиялық реагенттердің тиімді құрамы мен концентрациясын таңдауға мүмкіндік берді. Технологиялық ұнғымаларда эксперименттік тәжірибелер жүргізу нәтижесінде өнімді горизонты химиялық реагенттер-ингибиторлармен өндеудің арнайы әдістемесі жасалды. Уранды ұнғымалы жерасты шаймалау процестеріне ингибиторлық қоспалардың әсер ету тиімділігін таңдау және анықтау бойынша зертханалық және практикадағы тәжірибелік зерттеулер нәтижесінде күрделі тау-кен-геологиялық жағдайларда уран алу қарқындылығын арттыруға мүмкіндік беретін өнімді горизонты өндеудің арнайы әдістемесі әзірленді. Уранды ұнғымалық өндіруді қарқыннатудың әзірленген әдістемесін қолдану уранды ұнғымалық өндірудің тиімділігін арттыруға және өндіруге арналған пайдалану шығындарын 5-7% -ға қысқартуға мүмкіндік береді. Бұл ретте уран кендерін шаймалауды қарқыннату жұмыстарының экологиялық және өндірістік қауіпсіздігі артады.

Түйінді сөздер: уранды шаймалау, ингибитор, ұнғымалардың тұнба түзілу циклі, уран мөлшері, өнімді ерітінді

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ИНТЕНСИФИКАЦИЯ ПОДЗЕМНОГО СКВАЖИННОГО ВЫЩЕЛАЧИВАНИЯ УРАНА С ПРИМЕНЕНИЕМ ИНГИБИТОРОВ

Аннотация. Снижение фильтрационных характеристик руд в сложных горно-геологических условиях негативно отражается на динамике извлечения урана из недр и увеличивает период отработки технологических блоков и эксплуатационные расходы на добычу. В этой связи предотвращение гипсовых осадкообразований при подземном выщелачивании урановых руд с целью интенсификации скважинной добычи урана на предприятиях Республики Казахстан является весьма важной и актуальной задачей. Разработка технологии обработки продуктивного горизонта специальными реагентами — ингибиторами в зависимости от минералогического состава руд и структуры осадкообразующих материалов, позволит повысить эффективность подземного скважинного выщелачивания урана. Применение рентгенофазового метода исследований минералогического состава керновых проб и осадкообразований продуктивных горизонтов Сантона, Маастрихтского и Кампанского рудных интервалов позволило определить основные факторы влияющие на снижение фильтрационных характеристик руд. Лабораторные опыты по агитационному выщелачиванию урана из керновых проб с применением ингибирующих добавок позволили подобрать эффективный состав и концентрацию

химических реагентов для предотвращения гипсовых осадкообразований. В результате проведения экспериментальных опытов на технологических скважинах была разработана специальная методика обработки продуктивного горизонта химическими реагентами-ингибиторами. В результате лабораторных и опытно-практических исследований по подбору и определению эффективности воздействия ингибирующих добавок на процессы подземного скважинного выщелачивания урана была разработана специальная методика обработки продуктивного горизонта, позволяющая повысить интенсивность извлечения урана в сложных горно-геологических условиях. Применение разработанной методики интенсификации скважинной добычи урана, позволяет повысить эффективность скважинной добычи урана и сократить эксплуатационные расходы на добычу на 5 – 7 %. При этом повышается экологическая и производственная безопасность работ интенсификации выщелачивания урановых руд.

Ключевые слова: выщелачивание урана, ингибитор, осадкообразование, цикл работы скважин, содержание урана, продуктивный раствор

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Introduction

Uranium is the most significant element fundamental to nuclear power. According to the International Energy Agency (IEA), the renewable and nuclear power will dominate the growth of global electricity supply over the next three years, meeting on average over 90 % of additional demand (Khawassek et al., 2016). China, India and South East Asia are expected to account for more than 70 % of growth in global electricity demand over the next three years. The China's share of global electricity consumption is now projected to reach a new record: one-third up by 2025 compared to one-quarter in 2015 (Rashad, 2020; Atia et al., 2018). At the same time, advanced economies are looking to expand electricity use to displace fossil fuels in sectors such as transport, heating and industry (Chen et al., 2018).

The increasing demand for uranium can be met by exploitation of hydrogenous deposits, represented by poor or low-grade ores (chats), as well as ores under complex mining, geological and hydrogeological conditions (Nikitina et al., 2019: 1). Deposits of the second group of poor ores containing uranium reserves until recently have not been commercially exploited by conventional mining methods due to technical and economic reasons (Panfilov et al., 2016:160). Introduction of the economically viable ISL technology has expanded the mineral resource base of natural uranium by mining the poor ores (Kuandykov et al., 2020: 6). However, under difficult mining and geological conditions, deposits with low filtration characteristics of ores are subject to decline in production rates (Yusupov et al., 2018: 2). This is caused by use of sulphuric acid solutions as a solvent in uranium mineralization, which interacts with carbonate and clay minerals of the productive horizon, resulting in sludging of quartz, gypsum, and clay minerals (Rakishev et al., 2019: 7) (Rakishev et al., 2019: 6). This reduces

productivity of production wells and injectivity of injection wells, create impermeable areas and reduce the uranium content in the productive solution (Kenzhetaev et al., 2022: 3).

Improvement of filtration characteristics in low permeability ores requires new approaches, scientific and technical solutions to effectively disrupt and prevent sludging in productive horizons (Yusupov et al., 2017: 4). One method is the addition of chemicals to the leaching solution, inhibiting the gypsum sludging process during the uranium leaching (Zammit et al., 2014). In order to select an effective inhibitor, laboratory studies were carried out to determine the mineralogical characteristics of ore core samples and sludge-forming components from productive horizons of an uranium deposit in the Syrdarya depression.

Materials and basic methods

Establishing the composition, quantitative and qualitative characteristics of ores and host rocks, and sludging should enable determining the minerals reacting with sulphuric acid solutions in order to select optimal parameters and concentrations of chemical reagents. Use of inhibitors during uranium leaching under complex mining and geological conditions should prevent sludge formation, increase productivity of geotechnological wells, reduce production costs by reduction of well downtime and repayment time of technological units. Table 1 shows results of the X-ray phase studies of core samples from productive horizons of the Syrdarya depression uranium deposit.

The X-ray diffractometric analysis was carried out with an automated diffractometer DRON-3 of Cu_{Kα} radiation, β-filter, under the following conditions: U=35 kV; I=20 mA; 0-20; detector 2 deg/min. The X-ray phase analysis on a semi-quantitative basis was carried out using diffractograms of powder samples using the method of equal weights and artificial mixtures. Quantitative ratios of crystalline phases were determined. Interpretation of diffractograms was carried out using ICDD: Powder Diffraction File (PDF2) and diffractograms of impurity-free minerals (Rakishev et al., 2020).

Table 1. The X-ray diffraction analysis of core samples

Mineral	Formula	Santonian tier, %	Maastrichtian tier, %	Campanian tier, %
Quartz	SiO ₂	90.8	54.7	66.3
Smectite	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂	-	27.0	-
K-Feldspar	KAlSi ₃ O ₈	9.2	10.1	5.7
Kaolinite	Al ₂ (Si ₂ O ₅)(OH) ₄	-	6.7	11.6
Gypsum	-	-	-	16.4

Results of the X-ray phase analysis of core samples from productive horizons show partial similarity in the mineralogical composition of the Santonian, Maastrichtian and Campanian tiers. However, the presence of kaolinite 6.7 % and smectite 27 % in the Maastrichtian sample indicates the formation of ion-exchange colmatation caused by swelling of clay minerals due to interaction with sulphuric acid solutions. The presence of kaolin 11.6 % and gypsum 16.4 % in the core sample of the Campanian tier indicates

the formation of complex soluble gypsum sludge and the swelling of kaolinite. All this leads to multiple increase in volume, reduction of pore space and formation of impermeable areas in the productive horizon. The latter causes the flow of leaching solutions through the non-ore areas, so that the uranium content in the production solution falls, resulting in slower recovery of uranium from the subsurface. Table 2 shows results of the semi-quantitative analysis of sludge samples taken from wells penetrating various productive horizons.

Results of the X-ray phase studies of samples from the Santonian tier well indicate that the sediments consist of gypsum (100 %). Sediments from boreholes in the Maastrichtian tier predominantly consist of gypsum (88.9 %), and quartz (5.1 %), albite 6.0 %. Sediment samples from wells of the Campanian horizon are multicomponent and consist of quartz (35.6 %), gypsum (16.7 %), calcite (8.9 %), albite (33.9 %), microcline (4.9 %). The Campanian tier sediments, as compared to the Santonian and Maastrichtian tiers, consist of many components and have a complex structure. The Campanian ore horizon is dominated by mechanical sludging, whereas in the Santonian and Maastrichtian tiers the sediments are entirely of chemical origin.

Table 2. The X-ray phase analysis of sludge samples

Mineral	Formula	Santonian tier, %	Maastrichtian tier, %	Campanian tier, %
Quartz	SiO ₂	-	5.1	35.6
Gypsum	Ca(SO ₄)(H ₂ O) ₂	100	88.9	16.7
Calcite	Ca(CO ₃)	-	-	8.9
Albite	(Na _{0.75} Ca _{0.25}) (Al _{1.26} Si _{2.74} O ₈)	-	6.0	33.9
Microcline	(K _{0.95} Na _{0.05})AlSi ₃ O ₈	-		4.9

The studies established quantitative and qualitative characteristics of core and sludge samples taken from process wells penetrating the different tiers of a productive horizon. The wells in the Santonian and Maastrichtian tiers were found to have predominately gypsum sediments of chemical origin. Prevention of gypsum sludging, increase of filtration characteristics of productive horizons in order to intensify the ISL processes are complex procedures requiring the specific chemical inhibitors. In order to determine an effective dissolution of uranium minerals, the laboratory studies of agitated uranium leaching from core samples were carried out using various inhibitors.

The laboratory utilised the most well-known and available inhibitors: the nitrilotrimethylene phosphonic acid (NTP) and the oxyethylidiphosphonic acid (OEDP) and the “BP 8000 EKROS” mechanical stirrer. The procedure envisages pouring of 150 grams of the leaching solution into a thermostatic beaker, then adding of 50 grams of the core material into the beaker. The leaching time varied from 10 to 30 minutes; concentration of sulphuric acid in the leaching solution was 10 g/l. Table 3 shows results of the analysis, the uranium content in the leaching solution of different composition.

Table 3. Effect of IOMS on uranium content in the productive solution

Parameters	Exposure time, min					Type of leaching solution
	10	15	20	25	30	
C, mg/l	4,09	4,54	4,73	4,81	4,91	Initial
C, mg/l	4,16	4,48	4,56	4,63	4,69	Input of 10 mg/l of OEDP
Effect, %	+1,71	-1,32	-3,59	-3,74	-4,48	
C, mg/l	4,31	4,52	4,83	4,74	4,65	Input of 10 mg/l of NTP
Effect, %	+5,4	-0,44	-2,11	-1,46	-5,29	

Table 3 shows the uranium content in the productive solution in the initial leaching solution ranged from 4.09 to 4.91 mg/l. When 10 mg/l of the OEDP inhibitor was added to the leaching solution, the uranium content in the productive solution at the initial stage showed 4.16 mg/l; when 10 mg/l of the NTP was added, the uranium content in the productive solution reached 4.31 mg/l. The change of uranium content in the productive solution with time depending on leaching solutions of different composition is shown below (Fig. 1).

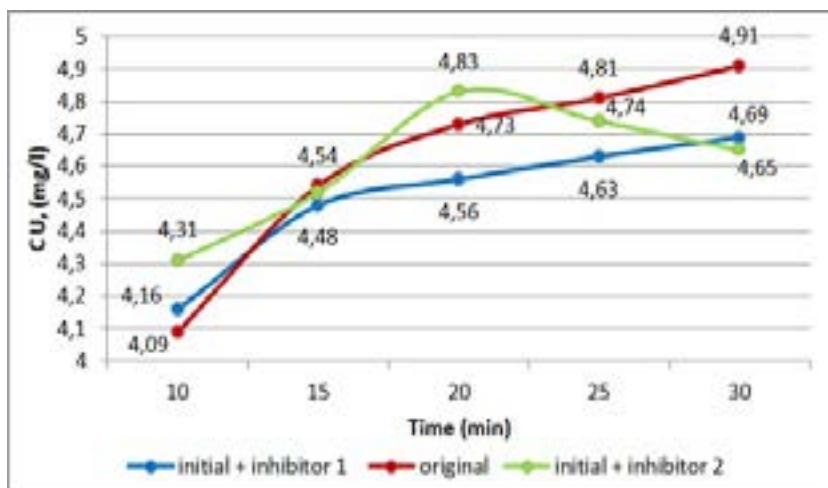


Fig. 1. The uranium content in the productive solution as a function of the composition of leaching solutions

The figure 1 shows that the uranium content in the productive solution gets higher after adding inhibitors 1 and 2 (4.16 and 4.31 mg/l respectively) compared to that in the initial leaching solution (4.09 mg/l) with 10 g/l of sulphuric acid. That is, the addition of inhibitors into the leaching solution prevents the neutralizing of sulphuric acid by host minerals, the sludging of gypsum and diverts the dissolving energy on uranium minerals. This can increase the productivity of geotechnical wells and uranium content in the productive solution, reduce the consumption of sulphuric acid and the mining operating costs. An effective use of inhibitors intensifying the in-situ uranium

production (ISL) under complex mining and geological conditions requires a special treatment technique of productive horizons (Rakishev et al., 2022: 4). Figure 2 shows the feeding of inhibitors into the leaching solution during the uranium ISL process.

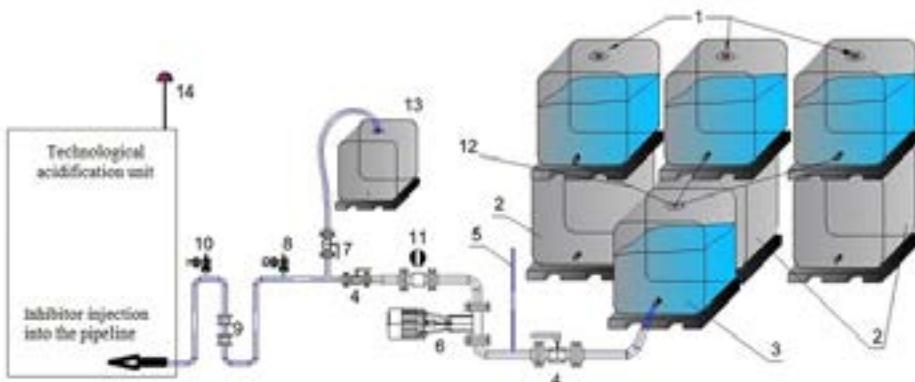


Fig. 2. Outline of supply of inhibitors into the leaching solution through the acidification process unit (APU):

1 - replaced tanks with inhibitors; 2 - fixed empty containers; 3 - fixed container with an inhibitor; 4 - shut-off valve; 5 - pressure release line; 6 - dosing pump; 7 - discharge valve; 8 - manometer; 9 - check valve; 10 - electric contact manometer; 11 - flow meter; 12 - reagent supply line to a fixed container; 13 - emergency drain container; 14 - emergency light signalization.

Inhibiting agents are supplied from transported tanks into a pipeline with the leaching solution through the acidification process unit (APU), under continuous injection of the leaching solution. Inhibitors are added to the leaching solution until reaching the assumed radius of spreading from the injection well filter of 25–30 m and the evacuation well. Reagents reaching the borehole, the feeding of inhibitors to the leaching solution stops. Injection of the chemicals and the leaching solution into injection wells lasts depending on uranium content in the productive solution and the capacity of evacuation wells.

Discussion

Further, studies were carried out to test effectiveness of chemical treatment using inhibitor reagents of the productive horizon in the Syrdarya depression deposit. In order to determine the impact on geotechnological properties of chemical inhibitors, it was decided to feed the chemical reagents, according to the developed procedure, through the acidification process unit (APU). Receptivity of wells has a direct proportional influence on spreading of chemical reagents over the productive horizon out of the borehole filter, where chemical reagents interact with host rocks. Estimates of spreading and removal of chemical agents, implemented via effective thickness of productive horizons, are provided for an assumed radius of 18–25 m from the filter of a treated well. While feeding chemical inhibitors to the productive horizon, the following parameters have been fixed: volume of the injected leaching solution (LS), consumption of sulphuric

acid, borehole receptivity, volume of rock-ore mass, the LS spreading area, the assumed radius of the LS spreading in the productive horizon, and consumption of sulphuric acid. Upon injecting the chemicals into the productive horizon, the well was put into production, and measurements of uranium content in the productive solution, borehole productivity and estimated uranium recovery from the productive solution were fixed. Figure 3 shows monitoring results of the borehole geotechnological parameters after injecting the inhibitors into the productive horizon.

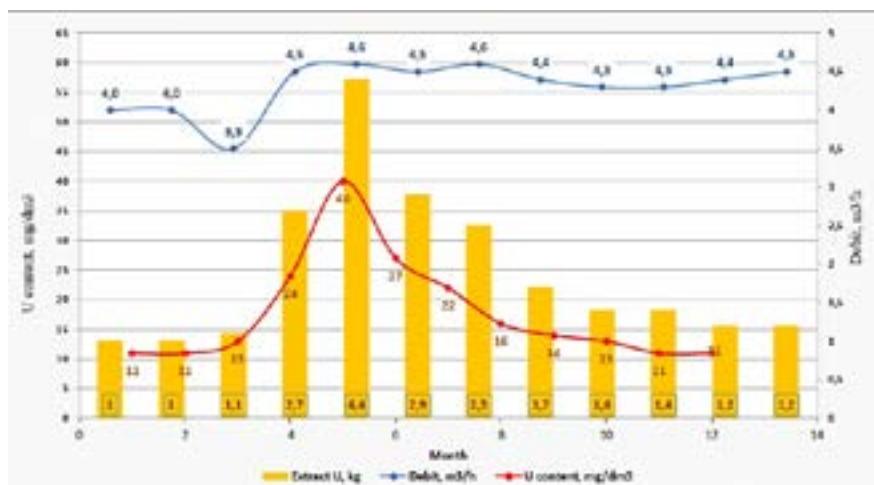


Fig. 3. The monitored results of geotechnical well parameters before and after inhibiting treatment.

As shown in the Figure 3, the uranium content in the productive solution before treatment was 11 mg/l and the well flow rate did not exceed 4.0 m³/hr, with an average recovery of 1.0 kgU/24hrs. After the treatment of the well with the IOMS inhibitors, the uranium content in the productive solution increased from 11 to 40 mg/l, and the well flow rate increased from 4.0 to 4.5 m³/hr, while uranium recovery increased from 1.0 to 4.4 kgU/24hrs. However, the uranium content in the productive solution fell to the initial values of 11 mg/l, while the well production rate increased and stabilised at 4.3 m³/hr. This confirms the assumption of sludge prevention resulting from the inhibitor's sulphuric acid acting on carbonate minerals and maintenance of sulphuric acid active in the productive solution. This way, higher uranium content in the productive solution has been ensured, as well as the increase of ore filtration characteristics and uranium recovery from the subsurface.

Conclusion

Core samples from the Syrdarya depression uranium deposit with producing horizons in Santonian, Maastrichtian and Campanian ore intervals were collected to implement quantitative and qualitative studies. Results of the X-ray phase studies indicate predominance of quartz (90.8 %) and presence of K-feldspar (9.2 %) at the Santonian ore interval. The quartz content (54.7 %), presence of smectite (27.0 %), K-feldspar (10.1 %), kaolinite (6.7 %) have been established in the Maastrichtian horizon, and the

quartz content (66.3 %), presence of K-feldspar (5.7 %), kaolinite (11.6 %), presence of gypsum (16.4 %) in the Campanian ore horizon.

Results of the X-ray phase analyses of the sludge show that its main mass has chemical origin, deposition of calcium salts, sand and clay. Predominant sludging of gypsum during uranium leaching points to the process of interaction of sulphuric acid with carbonate rocks as dominating one in the productive horizon. This decreases filtration characteristics of the ores and forms the impermeable sections, which further cause the fall in uranium content in the productive solution and growth of repayment time of technological units.

The laboratory carried out experiments on agitation leaching of uranium from the core samples using the OEDF and NTP (10 mg/l) as inhibiting additives to the leaching solution with sulphuric acid concentration (10 g/l). It was determined that, at initial stage of leaching, upon action of the above two inhibitors in the productive solution, the uranium content grew by 1.71 % and 5.4 % accordingly compared to the values of the process without the additives. This demonstrates the high efficiency of the OEDP and NTP inhibitors in preventing the neutralisation of sulphuric acid and formation of gypsum deposits.

The experiments on in-situ leaching intensification in geotechnological boreholes with use of the NTP chemical inhibitor, according to a developed procedure, showed a significant advantage over conventional methods of intensification. The post-treatment monitoring of the productive horizon in the Syrdarya depression showed the growth of uranium content in the productive solution from 11 to 40 mg/l and of the borehole productivity from 4.0 to 4.5 m³/hr. As a result, the uranium recovery increased from 1.0 to 4.4 kgU/24hrs while the operating costs and unit development time fell by 5-7%.

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