ҚАЗАҚСТАН РЕСПУБЛИКАСЫ ҰЛТТЫҚ ҒЫЛЫМ АКАДЕМИЯСЫНЫҢ

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ХАБАРЛАРЫ

ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ НАУК РЕСПУБЛИКИ КАЗАХСТАН Казахский национальный исследовательский технический университет им. К. И. Сатпаева

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Қазақстан Республикасы Ұлттық ғылым академиясы "ҚР ҰҒА Хабарлары. Геология және техникалық ғылымдар сериясы" ғылыми журналының Web of Science-тің жаңаланған нұсқасы Етегдіпд Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Ехрапдед, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Webof Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Геология және техникалық ғылымдар сериясы Етегдіпд Sources Citation Index-ке енуі біздің қоғамдастық үшін ең өзекті және беделді геология және техникалық ғылымдар бойынша контентке адалдығымызды білдіреді.

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TO THE QUESTION OF RECOVERY OF URANIUM FROM RAW MATERIALS

Abstract. The problem of extracting uranium from difficult-to-hide ores can be attributed to the most important scientific and technical problems facing the modern uranium mining industry.

One of the areas of modern scientific and technological progress in the development of radioactive ore deposits is using of a practically non-waste method of mining uranium – underground leaching (UL). A great interest to the method of underground leaching is determined by the positive experience of its application and high technical and economic indicators.

In Kazakhstanin underground uranium leachinga sulfuric acidis used as a leaching reagent. In this case, the degree of transition of uranium to the productive solution can be due to a variety of reasons, primarily related to the physico-chemical properties of uranium-containing minerals.

Thus, the use of sulfuric acid as a leaching reagent allows the extraction of only uranium (VI) compounds, since uranium compounds (IV) in sulfuric acid do not dissolve. To convert uranium (VI) to uranium (IV), oxidizers are used in industry, most often-ferric compounds. In this regard, the main problem of increasing the extraction of uranium in underground leaching is the transfer of uranium (IV) to a soluble state.

Analysis of scientific and technical literature has shown that recently one of the main directions for increasing the extraction of uranium from poorly soluble ores is the use of intensifiers that allow the most complete transfer of uranium from one valence state to another. In the article, the catalyst "M-1", which is a compound of transition metals, is considered as an intensifier. Comparative data are given for the study of agitation leaching of uranium from ore in the presence of a number of oxidants. Priorityuseofthe "M-1" catalystwasnoted.

Sorption methods are used to extract uranium from productive solutions. Ion exchange has certain advantages over others, since it allows to extract uranium from solutions of various concentrations.

Comparison of the kinetic dependencies of the sorption extraction of uranium by the ionites of Purolite A-500 and Ambersep-920 from the productive solutions of leaching of uranium-bearing ore in the presence of the «M-1»catalyst showed that they differ insignificantly. The values of activation energy for sorption of uranium on these ion exchangers and diffusion coefficients are calculated. Their magnitude is characteristic of a mixed external and internal diffusion process type.

In the literature, there are data on the use of natural sorbents for the extraction of uranium from productive solutions with low content of uranium. The advantages of such sorbents are their low cost. Within the framework of the present work, the possibility of sorption of uranium from productive solutions by natural zeolite and schungite in comparison with synthetic sorbents is investigated.

Key words: uranium, leaching, sorption, intensifier, catalyst, sorbent.

Introduction. Kazakhstan ranks second in the world in terms of natural uranium reserves and the leading positions in its production. Uranium is present in ores as part of a large number of mineral formations that differ in their physicochemical properties. According to the literature, the main uranium ores are the minerals of the tetravalent uranium: oxides (nasturan, uranium black) silicates (coffinite). The most common in the ores of almost all genetic types are simple oxides of U (IV). Minerals of hexavalent uranium are of lesser industrial importance [1-3].

Specificity of technological schemes for processing uranium ores is determined by the composition and properties of raw materials. Usually, the content of uranium in ores varies from tenths to hundredths of a percent. Because of the fine impregnation of uranium minerals in ores, the latter are not amenable to mechanical enrichment and the extraction of uranium from ores is in most cases carried out chemically [4, 5].

Depending on the composition and properties of the initial ore, one or another method of chemical processing is used.

Meanwhile, the method of underground well leaching finds an increasingly wide application in the development of uranium deposits, which, due to complex bedding conditions, as well as high specific investments and operating costs, cannot be worked out by traditional methods. In Kazakhstan, underground well leaching is the only effective method of uranium mining. Sulfuric acid is used as a leaching reagent [6, 7].

Minerals of tetravalent uranium do not dissolve in sulfuric acid, unlike to hexavalent. To dissolve the minerals of tetravalent uranium in dilute solutions of sulfuric acid, leaching is conducted with the addition of oxidizing agents. It is believed that the main problem of increasing the extraction of uranium from ore during underground leaching using existing technologies is the conversion of uranium (IV) to a soluble state. Therefore, at the heart of the overwhelming majority of research works is the question of the most complete transfer of uranium compounds into a soluble state, i.e. uranium (IV) in uranium (VI) [8-10].

In this connection, the purpose of this work was to study the effect of the "M-1" catalyst on uranium leaching and to assess the extent of its subsequent extraction from the resulting productive solutions.

In accordance with the complex chemical composition of uranium-containing solutions of underground well leaching and low uranium content, the most acceptable method of their processing is sorption methods, which are based on the use of ion exchange. Ion exchange has certain advantages over other methods (precipitation and extraction) since it allows to extract uranium from solutions of various concentrations [11-15].

In the literature, there are data on the use of natural sorbents for the extraction of uranium from productive solutions with low content [16-18]. The advantages of such sorbents are their low cost. Within the framework of the present work, the possibility of sorption of uranium from productive solutions by natural zeolite and schungite in comparison with synthetic sorbents is investigated.

Experimental part. As a raw material for experimental purposes, uranium-containing ore was used, in which the main uranium-containing mineral is coffinite. The composition of the ore is given below.

Sulfuric acid was used as the reagents which has the qualification "technical", GOST 2184-77, ammonium nitrate of the qualifications "Reagent", GOST 20478-75, sodium peroxoborate of the qualification "reagent", GOST 22567.10-93, ferric chloride (III) six-grade of the qualification "pure", GOST 4147-74, as well as the synthesized "Muhamedzhan-1" (M-1) catalyst [19, 20].

Experimental technique. Experiments on the leaching of uranium ore were conducted in a thermostated reactor according to a generally accepted procedure. Stirring of the pulp was carried out with a Stirrer-BS stirrer with a variable speed. The stability of the temperature was stabilized with a thermostat of the brand "TERMEX M01M". The maximum duration of the process was 48 hours. The ratio of S:L = 1:4.

Sorption of uranium was carried out on the anionites Purolite-A500 and Ambersep-920 in a static mode according to the procedure described in [21] from the productive solution from leaching of uranium-bearing ore. The composition of the solution is given below. There were used the anionitesPurolite-A500 andAmbersep-920 and natural materials – zeolite and schungite.

Methods of analysis. The quantitative content of uranium was determined by chemical methods of analysis.

Results and its discussion. As mentioned above, the main task of the vast majority of research works is the question of the most complete transfer of uranium compounds into a soluble state, i.e. uranium (IV) into uranium (VI).In this connection, we chose the initial ore, in which the main mineral is U [SiO₄], which makes up about 95 % of the total uranium mineralization balance.Uranium-containing minerals are also represented by leucoxene, accompanying - native selenium, cobalt-nickel pyrite (bravoite) and sphalerite. The composition of different rocks: quartz - 70–80 %, feldspars - 10–20 %, siliceous rocks - 5–10 %. The average content of uranium is 0.031 %.

At the first stage of the research, the effectiveness of the "M-1" catalyst's influence on the uranium extraction degree by agitation leaching with sulfuric acid was evaluated.

Agitation leaching is a preliminary stage in the study of technological properties of the ore, it makes it possible to estimate its excavation with an optimal concentration of leaching reagents, as well as assess the effectiveness of the effect of some additives on the process. The investigations were carried out in the presence of the "M-1" catalyst. Further, for comparison, in the presence of traditional oxidizing agents: ferric iron, ammonium nosulphateand sodium peroxoborate.

To date, great interest is, along with the study of the effect of traditional oxidants on the process of leaching of difficult-to-hide uranium-bearing ores, as well as the search for new intensifiers for increasing the extraction of uranium from persistent raw materials. In this regard, the leaching of uranium-bearing ore in the presence of the "M-1" catalyst was studied in comparison with the traditional oxidants: ferric iron, ammonium nosulphateand sodium peroxoborate.

Preliminary studies of the catalyst "M-1" are presented in [20]. The catalyst "M-1" is a mixture of nitrates, sulfates, chlorides, bromides and iodides of transition metals (Fe, V, Cu, Mn). The total concentration of metals in the catalyst was 0.3 g/dm³. Data characterizing the extraction of uranium by sulfuric acid in the presence of a catalyst are presented in table 1.

Catalyst content, g/dm ³	рН	ORP, mV	Extraction U, mg/dm ³	The acid consumption ($C_{\rm H2SO4} = 5.0 \text{ g/dm}^3$), g/g of uranium
0	2,00	378	35	39.1
0,3	2,01	388	80	14.2
0,6	1,99	396	82	12.8
0,9	1,98	402	84	11.1
1,2	2,00	408	86	9.7

Table 1 – Results of sulfuric acid leaching of uranium in the presence of "M-1" catalyst

From the data presented, it can be seen that the use of the "M-1" catalyst in the leaching process leads to an increase in the extraction rate of uranium from the ore. The maximum increase in the concentration of uranium in the solution is observed at the site of the change in the catalyst content in the range 0-0.3 g/dm³.

Figure 1 shows the leaching of uranium-bearing ore by sulfuric acid in the presence of a catalyst in comparison with oxidants.

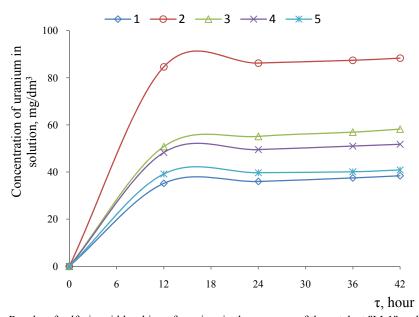


Figure 1 – Results of sulfuric acid leaching of uranium in the presence of the catalyst "M-1" and oxidants: 1 – sulfuric acid without oxidants and catalyst; 2 – "M-1" catalyst; 3 – ferric ion; 4 – ammonium nosulphate; 5 – peroxoborate

It can be seen from the table that the uranium content in the leaching solution without catalyst and oxidizers was 35 mg/dm³ after 12 hours of the experiment and increased to 38 mg/dm³ after 48 hours after the beginning of the leaching. In the case of using traditional oxidants, an increase in uranium extraction was also observed with increasing leaching time. The use of ferric chloride during the leaching process allowed to increase the concentration of uranium in the solution from 50.8 mg/dm³ for 12 hours of leaching to 58.2 mg/dm³ for 48 hours, and for ammonium nitrate and sodium peroxoborate from 48.3 and 39.2 to 51.8 and 40.9 respectively. When the "M-1" catalyst was used after 12 hours of the experiment, 84 mg/dm³ of uranium extraction was achieved, with increasing leaching time this value varies 88 mg/dm³.

Thus, the highest concentration of uranium in the solution is achieved when the ore is leached with sulfuric acid in the presence of a catalyst and decreases in the series: the catalyst "M-1" - ferric chloride III - ammonium perchlorate - sodium peroxoborate - sulfuric acid in the absence of oxidants and catalyst.

It should be noted that the catalyst "M-1" was successfully tested under production conditions and recommended for use.

The technological cycle of uranium mining by the method of underground borehole leaching includes the following interrelated technological processes: the actual underground uranium leaching, sorption-desorption of uranium and the production of concentrate. In accordance with the complex chemical composition of productive solutions and the low content of useful components, the most suitable variant of uranium concentrating is the sorption methods.

Unlike other methods, uranium sorption is characterized by the possibility of achieving high purification factors and a degree of concentration, high selectivity, the possibility of flexible regulation of selectivity by changing pH, oxidation-reduction conditions, complexation, simplicity and compactness of instrumentation, and a wide variety of artificial and natural sorbents.

Sorption of uranium together with the process of leaching is a kind of indicator of its extraction from this type of raw material.

Sorption of uranium was carried out in a static mode on Purolite-A500 and Ambersep-920 anion exchangers. The selected anion exchangers - strongly basic macroporous Purolite-A500 anioniteon the basis of styrenedivinylbenzene matrix and macroporous strongly basic anionite Ambersep-920 based on cross - linked polystyrene are effectively used in uranium mining enterprises of Kazakhstan.

At the first stage, comparative studies were conducted uranium sorption studies were carried out according to the above procedure. In the course of the experiment, a productive solution was used from the leaching of uranium-bearing ore with sulfuric acid in the presence of the "M-1" catalyst. The characteristics of the productive solutions are given in table 2, and the kinetic curves of sorption are shown in figure 2.

From the data given, it can be seen that the degree of uranium sorption most rapidly increases during the first hour, then the extraction rate decreases. The maximum degree of uranium recovery - 89.13% was achieved from the productive solution from ore leaching using the catalyst "M-1". From the leaching solution of uranium ore in the presence of an oxidizer-ferric chloride, uranium extraction amounted to 76.35%, and in directleaching with sulfuric acid (without additives) it was 63.31%.

Further, we determined the kinetic dependencies of uranium extraction from solution №1 by Purolite A-500 and Ambersep-920 ion exchangers in comparison. The results of the experiment are shown in figure 3, from which it can be seen that the ion exchangers practically equally sorb uranium from the solution. The degree of extraction of uranium on ion exchanger Ambersep-920 reaches 87.6%, Purolite A-500 - 89.0% for 8 hours of contact.

#	Productive solutions		Concentration H ₂ SO ₄ , g/dm ³	рН	Oxidation-reduction potential
1	With the use of the catalyst "M-1"	86.0	5.0	2.0	408
2	With the use of an oxidizing agent (trivalent iron chloride)	65.0	5.0	1.6	550
3	Productive solutions(without oxidizers and catalysts)	53.9	5.6	2.1	480

Table2 – The characteristics of the productive solutions

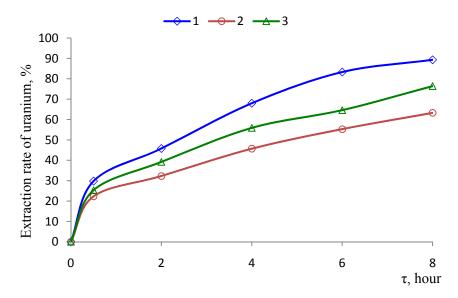


Figure 2 – The degree of extraction of uranium from solutions 1–3. The characteristics of solutions are given in table 2

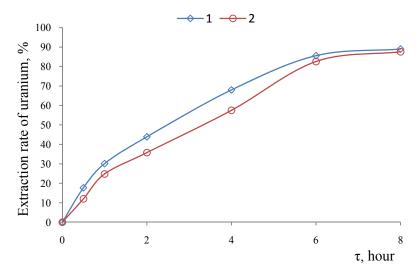


Figure 3 – Kinetic curves of sorption of uranium from the productive solution on anion exchangers: 1 – Purolite-A500; 2 – Ambersep-920

In addition, studies have shown that increasing the duration of the process beyond 8 hours is ineffective.

It is known from literature sources [16-18] that at low concentrations of uranium it is possible to use natural sorbents - shungite, zeolite - to extract it.

During the research, sorption extraction of uranium was tested with natural sorbents. The main constituents of the natural zeolite that we used are chabasite - (Ca, Na)[Al $_2$ Si $_4$ O $_{12}$]·6H $_2$ O, desmin (stilbite) - Ca[Al $_2$ Si $_7$ O $_{18}$]·7H $_2$ O, analcite - Na $_2$ [AlSi $_2$ O $_6$]·2H $_2$ O and the harmonic - Ba[Al $_2$ Si $_6$ O $_{16}$]. The composition of schungite for the main components is given in table 4. It should be noted that in the microquantities the composition of schungite includes such elements as:Sr, Co, Zn, Y, Cu, Sn, Mo, Ba, Ni, Mn, V, etc.

Content, % wt. Sorbent C SiO₂ Al_2O_3 Fe₂O₃ MgO CaO Na₂O K₂O Shungite 10 26.01 6.55 3.81 2.5 13.5 0.6 1.46

Table 3 – Chemical composition of schungite

The results of uranium sorption from productive solutions are presented in table 4.

#	Sorbent	Sorbent weight, g	with a in the mother productive liquor of sorpt	Uranium content in the mother liquor of sorption,	The extraction of uranium into the sorbent mg %	the sorbent	Calculated content of U in the sorbent without taking into account sorbed impurities,
			solution, mg	mg/dm³		/0	% wt.
1	Shungite	10	21.25	0.85	19.3	90.9	0.193
2	Zeolite	12	22.0	0.97	19.8	90.1	0.165

Table 4 – Results of sorption of uranium from productive solutions

It states in the table that the degree of extraction of uranium by natural sorbents is quite comparable with the synthetic ones (T: F for synthetic sorbents 1: 500, and for natural sorbents 1-1.2: 25), but their sorption capacity is low. The main disadvantage of synthetic sorbents is their high cost, and therefore, the use for uranium sorption from solutions with low concentrations (usually for pre-extraction) is unreasonable. To extract uranium from productive solutions, or from liquid waste of uranium chemical-metallurgical industries, it is more profitable to use natural sorbents. However, due to the low sorption capacity, their use is limited. At present, we are working to increase the sorption capacity of natural ion exchangers for uranium.

Conclusions. At the heart of the hydrometallurgical method of extracting uranium from ores in the industry of Kazakhstan is the method of underground leaching with sulfuric acid.

The tendency in the development of the world uranium mining industry in the development of the world uranium mining industry has also affected the uranium mining industry of the republic, which necessitated the involvement of persistent uranium-bearing ores. The minerals of tetravalent uranium insoluble in sulfuric acid represent such ores, as a rule. Therefore, at the heart of the overwhelming majority of research works is the question of the most complete transfer of uranium compounds into a soluble state, i.e. uranium (IV) into uranium (VI).

When poorly soluble uranium ores containing mainly tetravalent uranium bysulfuric acid is leached, the main task is the most complete oxidation of uranium (IV) to uranium (VI). Uranium (IV) in sulfuric acid is insoluble. In this connection, the study of the effect of new intensifiers on the process of uranium leaching is of great interest.

The influence of "M-1" catalyst on uranium leaching from persistent uranium-containing ore is studied and its advantage over traditional oxidants is shown.

Sorption methods are used to extract uranium from productive solutions. The concentration of uranium on Purolite-A500 and Ambersep-920 ion exchangers and natural sorbents - zeolite and shungitewas studied. In view of the low cost of natural sorbents, their priority use is considered for the extraction of uranium from productive solutions and liquid waste of uranium chemical and metallurgical industries.

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МИНЕРАЛДЫ ШИКІЗАТТАРДАН УРАНДЫБӨЛІП АЛУ МӘСЕЛЕСІ

Аннотация. Радиоактивті кен орындарынан уран өндірудезаманауи ғылыми-технологиялық прогрестің бағыттарының бірі іс жүзінде қалдықсыз игеру әдісі жерасты шаймалау (ЖШ) болып табылады.

Күкірт қышқылын шаймалау реагент ретінде қолдану тек уранның (VI) қосылыстарын алуға мүмкіндік береді. Уранның (IV) қосылыстары күкірт қышқылында ерімейді. Индустрияда, уранды (VI) уранға (IV)

айналдыру үшін тотықтырғыштар көбінесе, құрамында темір бар қосылыстар қолданылады. Осыған байланысты жер асты шаймалауда уранды өндіруді арттырудың негізгі мәселесі - уран (IV) ерігіш күйге ауысу.

Мақалада өтпелі металдардың қосындысы болып табылатын «М-1» катализаторы күшейткіш ретінде қарастырылады. Салыстырмалы деректер рудадан уранды үдетпелі шаймалауды бірнеше оксиданттың қатысуымен зерттеулержүргізілген. «М-1» катализаторының басымдықты пайдаланылуы байқалды.

Уранды ерітінділерден алу үшін сорбциялық әдістер қолданылады.«М-1» катализаторының қатысуымен ураны бар кендерді шаймалау ерітінділерінен уранды Purolite A-500 және Ambersep-920 иониттерімен сорбциялаудың кинетикалық тәуелділіктері олардың айырмашылығының шамалы екенін көрсетті. Әдебиет көздерінде уранның аз мөлшерімен өнімді ерітінділерден уран алу үшін табиғи сорбенттерді пайдалану туралы мәліметтер кездеседі. Мұндай сорбенттердің артықшылықтары олардың құнының арзандығы болып табылады. Табиғи цеолит пен шунгитпен уранның сорбциялануы туралы мәліметтер берілген.

Түйін сөздер: уран, шаймалау, сорбция, күшейткіш, катализатор, сорбент.

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К ВОПРОСУ ОБ ИЗВЛЕЧЕНИИ УРАНА ИЗ МИНЕРАЛЬНОГО СЫРЬЯ

Аннотация. Одним из направлений современного научно-технического прогресса в разработке месторождений радиоактивных руд является применение практически безотходного способа добычи урана – подземного выщелачивания (ПВ).

Применение серной кислоты в качестве выщелачивающего реагента позволяет извлечь только соединения урана (VI), так как соединения урана (IV) в серной кислоте не растворяются. Для перевода урана (VI) в уран (IV) в промышленности используют окислители, чаще всего – соединения трехвалентного железа. В этой связи основная проблема повышения извлечения урана при подземном скважном выщелачивании - перевод урана (IV) в растворимое состояние.

В статье в качестве интенсификатора рассматривается катализатор «М-1», который представляет собой соединения переходных металлов. Приведены сопоставительные данные исследования агитационного выщелачивания урана из руды в присутствии ряда окислителей. Отмечено приоритетное использование катализатора «М-1».

Для извлечения урана из продуктивных растворов применяют сорбционные методы. Сравнение кинетических зависимостей сорбционного извлечения урана ионитамиPurolite A-500 и Ambersep-920 из продуктивных растворов от выщелачивания урансодержащей руды в присутствии катализатора «М-1» показало, что они отличаются незначительно.

В литературе имеются сведения о применении природных сорбентов для извлечения урана из продуктивных растворов с низким его содержанием. Преимущества таких сорбентов – их невысокая стоимость. Приведены данные о сорбции урана природными цеолитом и шунгитом.

Ключевые слова: уран, выщелачивание, сорбция, интенсификатор, катализатор, сорбент.

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