

Uranium Leaching by Sulfuric Acid in the Presence of a Catalyst under Laboratory Conditions

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ABSTRACT

The article presents data on the extraction of uranium from ores by leaching with sulphuric acid in the presence of a catalyst.

Keywords: Uranium, Agitation Leaching, The Redox Potential, Catalyst.

I. INTRODUCTION

Intensification of the uranium extraction practise from ores is achieved by using a variety of physical and chemical methods. In the international scientific and technical literature, there is a huge amount of work about a wide variety of efforts which intensify the uranium leaching from ores with the help of various chemical (type reagent, a method of supply of a reagent, combination of reactants in solution and others) and physical factors (temperature, electric current, mechanical vibrations, pulsation of the system under the influence of mechanical work, etc.) [1 - 4]. For the underground and downhole uranium extraction from ores, the problem of increasing the degree of uranium extraction from refractory ores, reduction of well colmatation and life of mine can be classified among the most important scientific and technical problems, precede of a modern uranium mining industry of the Republic of Kazakhstan. Despite the large number of innovative technical solutions, major problems in the uranium hydrometallurgy remain: the transfer of metal from ore to a solution, transfer of uranium compounds into a soluble form and uranium extraction from solutions. Applied on a commercial scale in-situ leaching methods do not provide the required performance in terms of business competitiveness. This is due to insufficient study of a number of key issues related to the concepts of the mechanism of reactions in the hydrometallurgical systems. Recently, the major among of research question is the transfer of uranium

compounds in the soluble state through transfer of uranium (IV) to uranium (VI). In this case, it does not consider the achievements of natural science disciplines in the study of the microstructure and submicrostructure condensed systems, and the achievements of theoretical metallurgy in the research areas of structural changes and the formation of structure of substances on the micro level. It does not take into account that the uranium oxide is a continuous series of solid solutions of oxygen in uranium. The presence of singular entities in this row can significantly affect the solubility of the uranium compounds. Scientific forecasting of possible directions of chemical reactions involving uranium compounds practically does not exist. A forecast of the reaction path consists of experimentally not verified schemes based on the electron theory of behaviour in chemical systems. Therefore, the great interest, in particular, researches on the effect of oxidants on the transfer of uranium into a soluble form and the leaching process of refractory uranium ores. In this regard, our study on the effect of catalysts on the process of sulfuric acid leaching of uranium ore were taken.

II. METHODS AND MATERIAL

Analysis of the composition of the core material were conducted by X-ray and electron microscopy. The X-ray diffraction (XRD) analysis was performed using the device of D8 Advance (Bruker), radiation of α -Cu, tube voltage - 40 kV, current - 40 mA. Processing of the

diffraction patterns was performed using EVA software security. Table 1 shows the phase composition of the uranium-containing core.

Table 1. The phase composition of uranium-containing core

Name	Formula	SQ
Quartz	SiO ₂	28.4
Okermanit	Ca ₂ SiO ₄ · MgSiO ₃	18.2
Hematite	Fe ₂ O ₃	10.6
Aluminium oxide (corundum)	Al ₂ O ₃	10.5
Forsterite	Mg ₂ SiO ₄	8.8
Calcium silicate	CaSiO ₃	7.5
Orthoclase	K (Al, Fe) Si ₂ O ₈	5.3
Muscovite	(H, K) AlSiO ₄	3.8
Hematite	Fe ₂ O ₃	2.7
Wustite	FeO	2.4
Clinochlore	Mg-Fe-Fe-Al-Si-O-OH	1.7

X-ray diffraction (XRD) analysis showed that, silicates of various compositions dominated in the core phase.

Figure 1 shows the diffraction pattern of the core material.

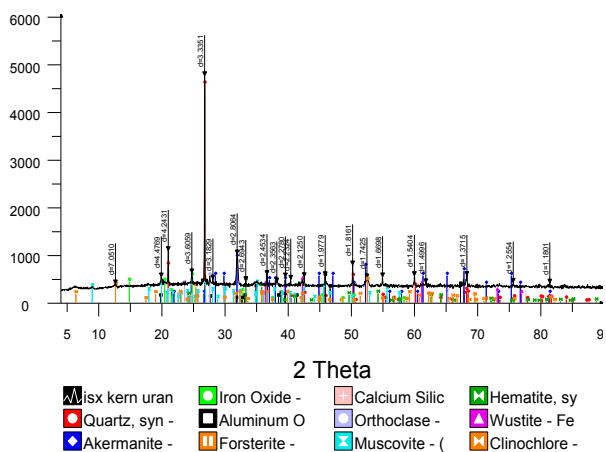
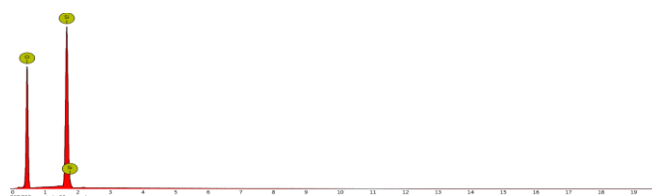
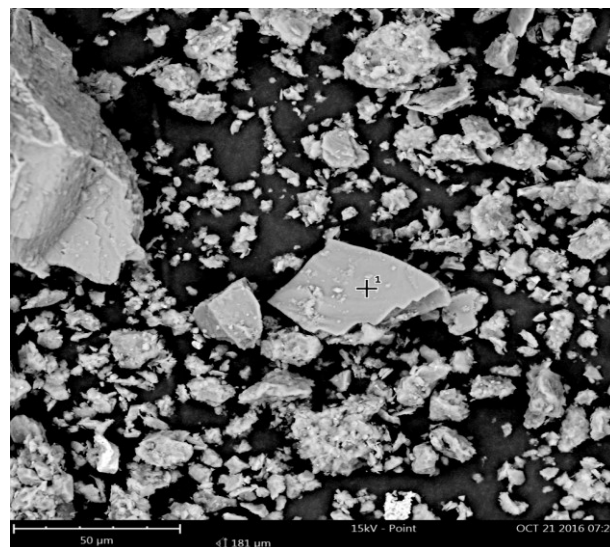


Figure 1. The XRD pattern of the original uranium-containing core

Analysis of cakes after the leaching process confirmed the presence of silicates of various compositions, not only calcium silicate, which according to the literature is considered to be the main reason of well colmatation at ISL.

The pattern of cake after the leaching process was analyzed by a scanning electron microscope PHENOM XL "PHENOM WORLD" (Netherlands), (Cake after leaching by sulfuric acid - concentration of 10 g / l in the presence of catalyst, room temperature, leaching time - 96 hours, the mixer speed rpm - 480 revolutions / min). Figure 2 shows the elemental analysis of cake particle in point 1. The particle size of 35 microns.

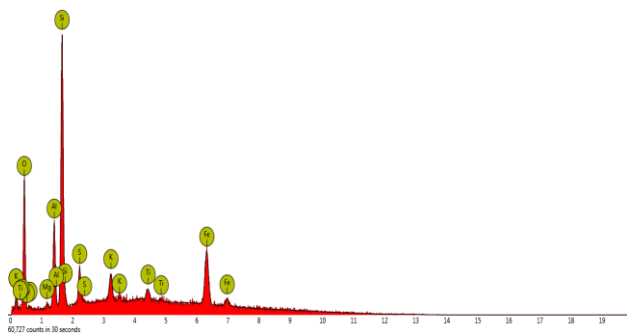
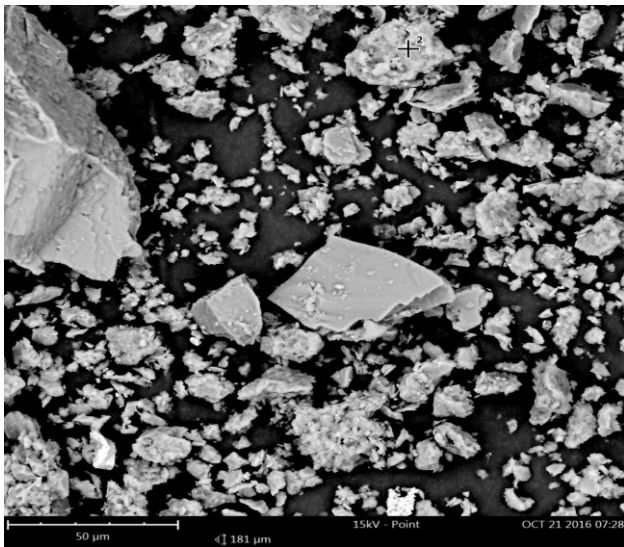


Element Number	Element Symbol	Element Name	Atomic Concentration	Error
14	Si	Silicon	22.8	0.0
8	O	Oxygen	77.2	0.0

Figure 2. The elemental analysis of a particle at point 1

The data of the analysis shows that the point 1 is a particle of free quartz.

Figure 3 shows the elemental analysis of particles at point 2. The cake particle at point 2 has a very complicated element structure which is characterized by the presence of silica in combination with oxides of various elements up to the sulfur-containing compounds (Fig. 3).



Element Number	Element Symbol	Element Name	Atomic Concentration	Error
14	Si	Silicon	22.0	0.1
8	O	Oxygen	50.0	0.0
26	Fe	Iron	13.1	0.1
13	Al	Aluminium	7.7	0.0
16	S	Sulfur	3.2	0.1
19	K	Potassium	2.4	0.7
22	Ti	Titanium	1.2	0.9
12	Mg	Magnesium	0.5	0.7

Figure 3. Elemental analysis of particle at point 2

These and other data suggest the complexity of the chemical reactions occurring in situ leaching of uranium ore by sulfuric acid, which causes the need to study the leaching process with regard to specific uranium containing materials.

Uranium leaching was conducted by sulfuric acid in the laboratory conditions and in the presence of a catalyst "PM" based on iron oxide. Samples of uranium ore core contained pitchblende - 58% and coffinite - 42%.

Leaching was conducted in a glass equipped with a powered movement pulp at different ratios of S: L and concentrations of sulfuric acid at room temperature (25⁰C). Hinge of core material is 50 grams (dry weight).

III. RESULTS AND DISCUSSION

The dependency of uranium extraction on concentration of the sulfuric acid by using the PM catalyst was determined. Table 2 shows the redox potential, pH of the solutions, the uranium concentration in the solution after leaching, the share of the reduced iron.

Table 2. Uranium leaching depending on the concentration of sulfuric acid

	5 g / dm ³	10 g / dm ³	25 g / dm ³	50 g / dm ³
The uranium content in solution, mg / dm ³	17	32	37	39
pH	1.96	1.89	1.86	1.78
AFP	318.0	371.0	378.1	389.0
Fe + 2	1.21	1.23	1.20	1.03
Fe + 3	0.11	0.17	0.16	0.18

For this type of ore, an increase in the concentration of sulfuric acid substantially affect the extraction of uranium, the content of which is increased in a solution from 17 mg / dm³ to 39 mg / dm³. As seen from Table 2, the sulfuric acid solution with a concentration of 50 g / dm³ has a high oxidation reduction potential (389 mV), nevertheless, during the uranium leaching, the acceptable concentration of sulfuric acid can be considered and 10 g / dm³, since it reduces the overall consumption of sulfuric acid (Fig. 4).

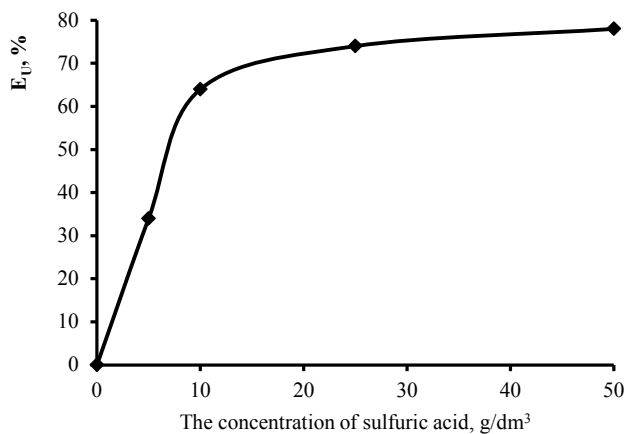


Figure 4. Dependence of uranium extraction on the concentration of sulfuric acid

The greatest effect during the uranium leaching process obtained by using solutions containing ferric ions. Uranium concentration in the solution increases from 3 to 19 mg / dm³. The proportion of the reduced iron is provided in Table 3.

Table 3. Uranium leaching depending on S: L

	1: 2	1: 4	1: 6	1: 8
The uranium content in solution, mg / dm ³	3	9	17	19
pH	1.98	2.0	1.89	1.97
AFP	298.3	299.0	298.1	298.0
Fe + 2	0.98	1.07	1.19	1.03
Fe + 3	0.25	0.17	0.20	0.23

The oxidation potential, according to published data, is regulated by the ratio of ferrous / ferric iron and requires ferric ions to produce soluble compounds of uranium (IV). As can be seen from Table 3, with the increase in the ratio L: S values of the oxidation reduction potential hardly changes. The highest uranium extraction value observed at a ratio of L: S = 8: 1.

Figure 5 shows the dependency of uranium extraction on the ratio of solid to liquid conditions.

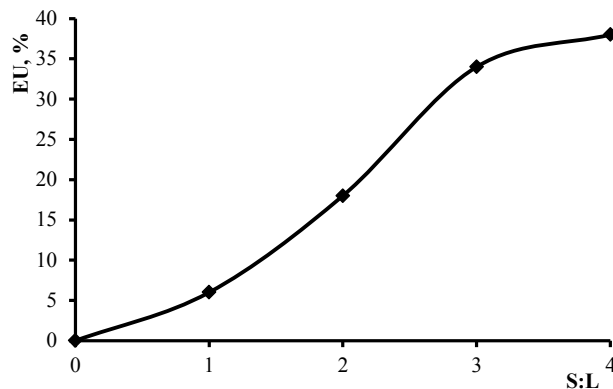


Figure 5. Dependency of uranium extraction on the ratio of S: L (1 - 1:2; 2 - 1:4, 3 - 1:6, 4 - 1:8)

The ratio of ferric to ferrous ions in the solution also affects the rate and extent of dissolution of the uranium; extraction increases with the amount of divalent iron ions, even if the concentration of trivalent iron ions remains constant. The greater ratio of divalent ions / trivalent iron is especially important at leaching of refractory uranium ores.

In the next stage, the research of dependency of uranium extraction on the leach duration was carried out. The results are shown in Table 4 and Figure 6.

Table 4. Uranium leaching depending on leaching duration

	12 h	24 h	36 h	48 h
The uranium content in solution, mg / dm ³	31	29	20	17.4
pH	1.78	1.78	1.84	2.24
AFP	314.0	298.2	275.0	249.0
Fe + 2	1.11	1.07	1.18	1.03
Fe + 3	0	0.1	0	0

As it is clear from table 4, with the increasing leaching time of uranium extraction into the solution in this case is reduced, which is probably due to changes in the structure of the leach solutions and inverse output of uranium from leach solution (Fig. 6).

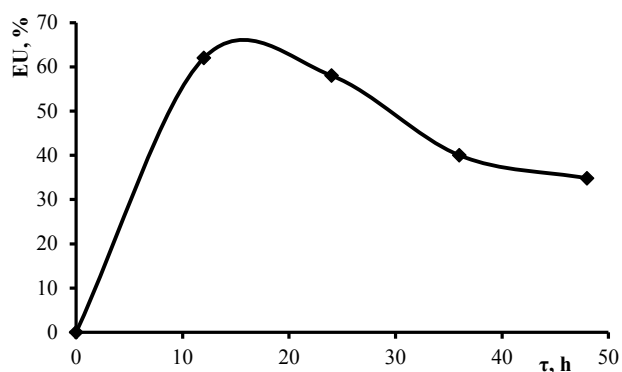


Figure 6. Dependency of uranium extraction on the leaching duration

What is indirectly confirmed by the increase in the oxidation - reduction potential of the solution with increasing leaching time.

IV. CONCLUSIONS

A comprehensive analysis of the solid leaching reaction products showed the complexity of the cakes formed during the agitation leaching under laboratory conditions. The data of the analysis suggests the formation of silicates not only of calcium, but also sodium and potassium. These silicates can lead to faster well colmatation in situ leaching than gypsum.

It has been shown that the increase in concentration of leaching agent in the aqueous solution leads to a rapid increase in uranium extraction only within the sulfuric acid concentration of 10 - 20%, which suggests a possible inappropriate use of solutions with a high concentration of sulfuric acid to activate wells in the processes by applying the in-situ leaching.

A similar assumption can be made on the basis of the influence of the ratio S: L on the extent of uranium extraction during agitation leaching.

Indicators of uranium extraction into solution, depending on the leaching duration, demonstrate the complexity of the behaviour of uranium compounds in an inorganic aqueous solution. The reverse transition of uranium from solution to the solid phase predetermines the need to study the behaviour of uranium in an aqueous solution of an inorganic complex microstructure.

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