

Approaches to uranium dump processing by bioleaching technology

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Abstract. This paper discusses a method of extracting uranium from uranium dumps by bacterial leaching using association of thion bacteria. Bioleaching mechanism is based on interaction of biological, chemical and electrochemical processes. The microstructure and composition of uranium-containing wastes were also investigated. The work presents the results of the study (changes in volumetric activity) before and after leaching. Results of the research proved that at microbiological processing of uranium production waste by association of thionic bacteria the degree of uranium leaching out of dumps at pachuca leaching was 59.3%. Nevertheless the residual radioactivity allows classifying the obtained material to be low-level radioactive waste one. At percolation leaching, having the aim to determine the effect of particle size of feed stock on the degree of uranium extraction, two samples were under study: breakstone in initial condition and ground breakstone. According to results of the study the degree of uranium extraction (a grinded one 0.3-0.5mm fraction) from dumps is 35.18%. Using breakstone in initial condition (10-25 mm fraction) degree of uranium leaching from dumps at percolation leaching measures up to 100%. Thus, residual radioactivity corresponds to all national sanitary standards for classifying a solid phase to a nonradioactive one. In addition, using microbial leaching there's no need in further uranium ore grinding and that reduces costs to process uranium dumps. The work proposes a scheme for processing uranium dumps in the technological process. This technology will contribute to transforming the places for uranium waste storage into anthropogenic deposits.

1. Introduction

An essential condition for safe development of nuclear power engineering is creating and adoption of new effective methods of radioactive waste disposal at all stages of a nuclear fuel cycle including uranium ore extraction. Uranium extraction results in waste rock dumps containing natural uranium isotopes (uranium-238, uranium-234, and uranium-235) and decay products (lead-206, thorium-234, thorium-230, radium-222, etc.) [1]. Meanwhile residual radioactivity of developed rocks compared with other enterprises is really low. But in case of long-term storage in dumps and tailing dump, harmful radiation doses occur [2, 3]. That's why it requires special areas of insulation. The use of standard manufacturing technology is not applicable due to extremely low economic efficiency. In this regard, the problem of uranium dumps disposal is relevant for all countries where uranium is mined. There is a need for environmentally-friendly and cost-effective technology providing extraction of residual uranium and concomitant radioactive elements out of dumps. Meanwhile, waste material from



processing must comply with sanitary regulations on the level of residual radioactivity. That will allow reusing it in nuclear power engineering and other industries. At the moment, the most promising method for this purpose is microbiological leaching which was considered to be an attractive alternative to traditional physical and chemical methods of ore dressing by reducing energy consumption, transport costs and less harmful effects on the environment [4, 5].

The role of microorganisms in bioleaching is in oxidation of Fe^{2+} to Fe^{3+} (oxidizer of metal sulfides), oxidation of elementary sulphur and reconstructed sulphur compounds, sulfuric acid obtaining, keeping medium acid reaction [6]. Today, bioleaching procedure supposes two coexisting mechanisms of bacteria–sulfide minerals interaction: direct (contact) and indirect (non–contact). Direct bacterial leaching occurs through physical contact of the bacterial cells with the mineral surface catalyzed by enzymes [7, 8]. It's supposed that bacteria do not contact the whole mineral surface but choose specific places of defects in crystal lattice [9].

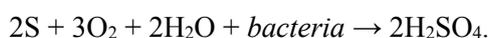
In direct bacterial mechanism, pyrite usually containing in uranium ore is an energy source for bacteria thanks to its oxidation [10].



In case of indirect bioleaching, bacteria oxidize ions of bivalent Fe up to trivalent, which oxidizes chemically sulfide mineral [11].



Sulfur released in the process is oxidized by bacteria to sulfuric acid [13]



At bioleaching of uranium minerals, insoluble tetravalent uranium transforms from insoluble uranium oxides in acid medium to soluble sulphates due to the reaction of trivalent iron and sulfuric acid resulted by microorganisms [14].



To maintain a sufficient amount of iron in the solution, chemical oxidation of metal sulfides must be done in acid environment ($\text{pH} < 5.0$) [17, 18]. At $\text{pH} 2.0 - 3.0$ bacterial oxidation of Fe^{2+} is about $10^5 - 10^6$ times faster than chemical oxidation [19].

The purpose of the work is to improve the method of microbiological processing of uranium production waste by association of thionic bacteria with high content of *Acidithiobacillus ferrooxidans* (*A. ferrooxidans*).

2. Materials and Methods

As the virgin dump samples the ones containing uranium waste (figure 1) of Pridneprovsk Chemical Plant (Ukraine) were used.



Figure1. Photo of uranium-cutting waste samples.

The experiment procedure equipment includes laboratory units for percolation (a) and pachuca (b) uranium leaching (figure2).

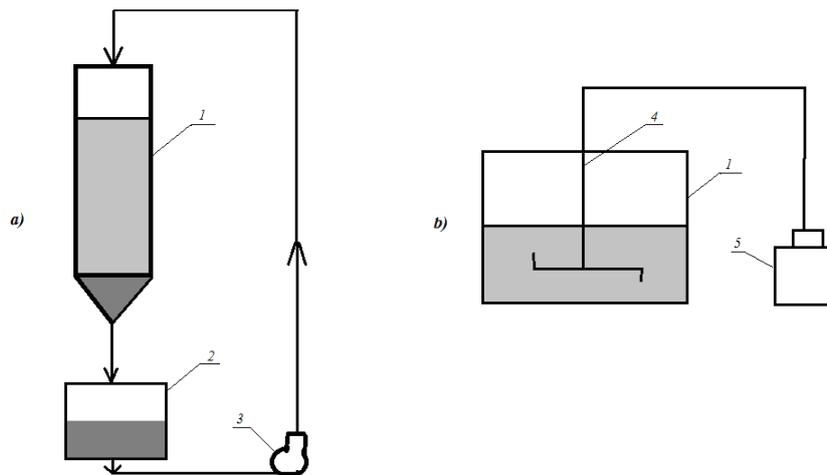


Figure 2. Functional schemes of laboratory units for percolation (a) and pachuca (b) uranium leaching out of base ore and tailing dump rock: 1–reactor; 2–collecting tank; 3– centrifugal pump; 4–mixer; 5–engine.

To study how the size of feed stock particles influences on degree of ore extraction by percolation method we've considered two sample forms – breakstone in initial condition (fraction 10–25 mm) and ground breakstone (fraction 0,3–0,5 mm).

At pachuca leaching we study only feed stock as the solution couldn't pass through ground material and there was a blockage in the result.

The temperature during the process was maintained at 25–30°C, solution pH, content of Fe^{3+} , Fe^{2+} in the solution were monitored.

Uranium wastes were put into the reactor where it was oxidized by association of thionic bacteria with high content of *Acidithiobacillus ferrooxidans* and chemical oxidation by ions of Fe^{3+} (released from microorganisms' activity) with associated reactions of electrochemical metal dissolution. At percolation leaching, bacterial solution of sulfate iron is fed self-flowing into the reactor, filtered through an ore bed and entered the filtrate collecting tank. Later this solution goes back into the sys-

tem with the help of a centrifugal pump if necessary (solution feed rate is 100 ml/day) (figure2). Pachuca leaching is realized in the reactor by continuous stirring.

The uranium content of the samples was determined by X-ray fluorescence analysis. The microstructure of uranium-containing waste was examined on a PHENOM proX SEM/EDS (Scanning Electron Microscopy with Energy Dispersive Spectroscopy) system by Phenom-World B.V. (Netherlands). Product specifications: the maximum increase of 150,000, a resolution of 10 nm, an accelerating voltage of 5, 10, 15 kV. Elemental analysis of the test samples in the set points was performed using Phenom's «Element Identification» program. The activity of the solid and liquid phase of the sample was measured on a UMF–2000 radiometer

3. Results and discussion

The scanning electron microscopy of the uranium dump samples was performed to determine the microstructure and features of uranium distribution. Figures 3 and 4 show samples before and after leaching.



Figure 3. Micrograph of uranium-containing waste sample before leaching.

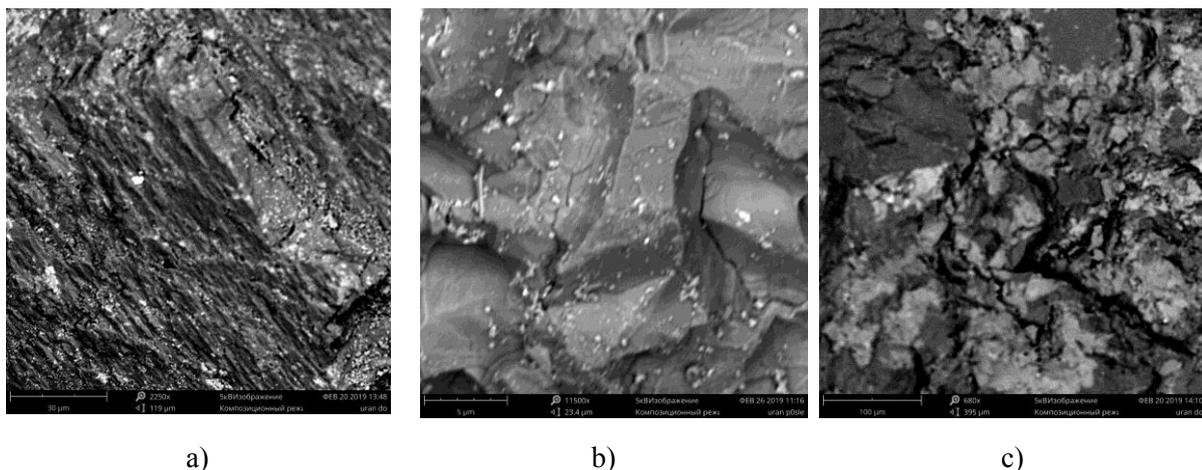


Figure 4. Micrographs of uranium-containing waste after leaching: a – pachuka method; b – percolation method (fraction 0.3 – 0.5 mm); c – percolation method (fraction 10 – 25 mm).

It can be seen that in figure 4 (after processing) there are considerably fewer areas of light color than in figure 3 (before processing).

Elemental point analysis of the uranium-containing sample prior to leaching is shown in Table 1. The element spectra are shown in figure 5 and figure 6.

Table 1. Results of elemental point analysis of the sample before leaching.

The light color phase			The dark color phase		
Element Symbol	Atomic Conc.	Weight Conc. %	Element Symbol	Atomic Conc.	Weight Conc. %
Fe	23.91	49.81	O	76.87	67.84
O	67.42	40.25	Si	8.62	13.36
Si	4.79	5.02	Na	9.40	11.92
S	3.85	4.60	Al	4.24	6.31
U	0.04	0.32	C	0.86	0.57

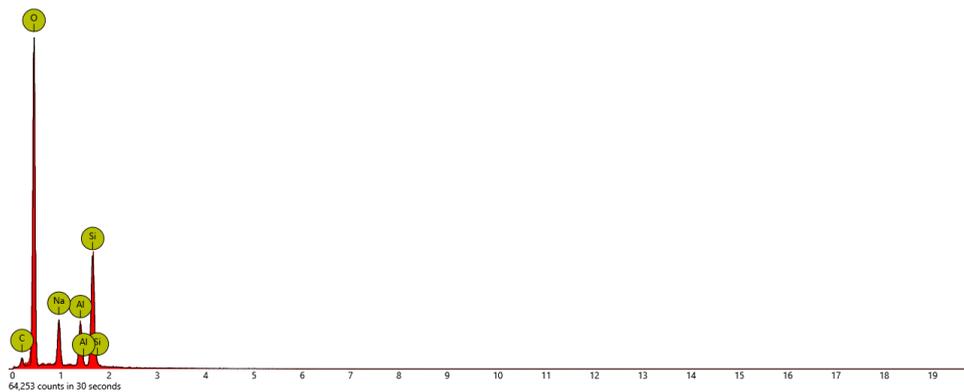


Figure 5. X-ray spectrum lines showing the presence of elements (dark phase).

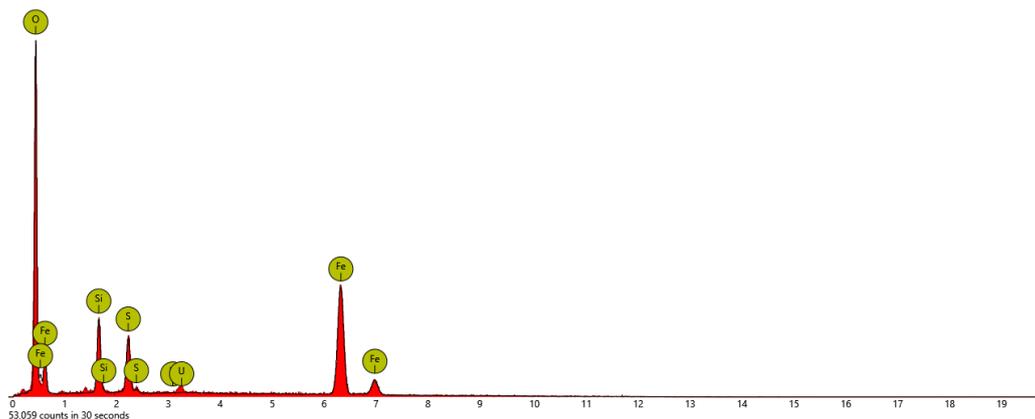


Figure 6. X-ray spectrum lines showing the presence of elements (light phase).

The results of the study of the effectiveness of uranium leaching methods are presented in table 2.

Table 2. Resulting percentage of uranium content in the samples before and after bioleaching.

Leaching method	Uranium content (%)		
	before experiment	after experiment	
		solid phase	liquid phase
Pachuca leaching	0.027	0.0110	0.0230
Percolation leaching (10–25mm fraction)	0.027	0	0.0324
Percolation leaching (0,3–0,5mm fraction)	0.027	0.0175	0.0211

As a result of bacterial leaching (table 2), we have obtained that the degree of uranium extraction from the dumps by the pachuca leaching method is 59.3%, percolatory up to 100% (fraction 10–25 mm) and 35.18% (fraction 0.3–0.5 mm).

This fact can be explained by agglomeration (adhesion) of ground particles. That’s why the leaching solution does not contact thoroughly with the whole ground ore dump.

The results showing change in activity concentration in the samples of solid and liquid phases at bacterial leaching by percolation and pachuca methods are presented in the table 3.

Table 3. Activity concentration of the samples before and after bacterial leaching by percolation and pachuca methods.

Activity concentration Bq/l	Virgin sample	Leaching methods					
		Percolation leaching (0,3–0,5mm fraction)		Percolation leaching (10–25mm fraction)		Pachuca leaching	
		Liquid phase	Solid phase	Liquid phase	Solid phase	Liquid phase	Solid phase
A_{α}	$3.85 \cdot 10^5$	18.83	3.576	13.27	0	2.1	11.9
A_{β}	$16.12 \cdot 10^5$	161.4	134.1	126.8	78	115.8	111

At percolation leaching of feed stock (breakstone 10–25mm fraction) alpha – activity decreased in $3.85 \cdot 10^5$ times and beta –activity – in $2.06 \cdot 10^4$ times. Residual radioactivity corresponds to all national sanitary standards for classifying the solid phase to a nonradioactive material that allows it to be used as breakstone in building, as a concrete filler etc. In addition, using microbial leaching (percolation method) there’s no need in further uranium ore grinding and that reduces costs to process uranium dumps.

At pachuca leaching alpha – activity decreased in $3.23 \cdot 10^4$ times and beta –activity – in $1.45 \cdot 10^4$ times. Nevertheless the residual radioactivity allows classifying the obtained material to be low-level radioactive waste material. Such low indices compared with the ones at percolation leaching, in our opinion, may result from low speed of the mixer making it impossible to mix well a pulp for complete contact of the leaching solution with the solid phase thereby creating dead zones.

The results can be shown in a scheme of operating procedures in uranium dump processing (figure 7).

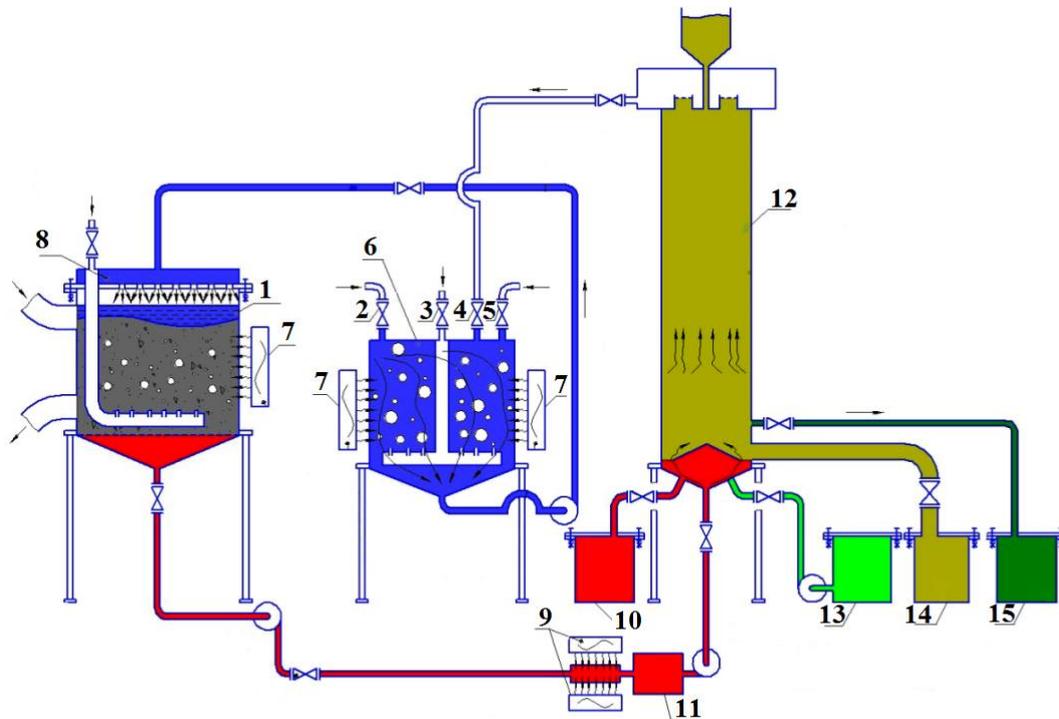


Figure 7. Scheme of microbiological processing of uranium production waste: 1 – reactor; 2 – sulfuric acid supply valve; 3 – air supply valve; 4 – return filtrate valve after the sorption column; 5 – microorganisms supply valve; 6 – unit for preparing bio-solution; 7– infrared radiator; 8 – spray line collector; 9 – ultraviolet radiators; 10 – tank for productive solution; 11 – cavitator; 12 – sorption–stripping column lock valves; 13 – tank for stripping solution; 14 – tank for sorbent; 15– tank for commercial strippant.

Using the proposed scheme of microbiological processing of uranium production waste could solve the problem of places for uranium dumps storage and transform them into anthropogenic deposits to produce uranium and rock being used for building material if needed.

4. Conclusion

There's proposed the method for microbial processing of uranium production waste by association of thionic bacteria with high content of *A.ferrooxidans*. This technology is characterized by a high rate of uranium extraction (up to 100%) and decrease in residual radioactivity for classifying the solid phase to a nonradioactive material.

The result of this work is the technological scheme of microbiological processing of uranium production waste. This technology will contribute to transforming the places for uranium waste storage into anthropogenic deposits.

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