UDC 669.822:544.77.052.22 DOI: https://doi.org/10.15407/geotm2021.158.084

INDUSTRIAL TECHNOLOGY OF PROCESSING CARBONACEOUS URANIUM ORE IN UKRAINE

[1]Mukhachev A.P., [2]Yelatontsev D.O., [3]Nefedov V.G.

[1]Institute of Geotechnical Mechanics named by N. Poljakov of NAS of Ukraine, [2]Dniprovsk State Technical University MES of Ukraine, [3]Ukrainian State University of Chemical Technology

ПРОМИСЛОВА ТЕХНОЛОГІЯ ПЕРЕРОБКИ КАРБОНАТНИХ УРАНОВИХ РУД В УКРАЇНІ

[1]Мухачов А.П., [2]Єлатонцев Д.О., [3]Нефьодов В.Г.

[1]Інститут геотехнічної механіки ім. Н.С. Полякова НАН України, [2]Дніпровский державний технічний університет МОН України, [3]ДВНЗ «Український державний хіміко-технологічний університет»

ПРОМЫШЛЕННАЯ ТЕХНОЛОГИЯ ПЕРЕРАБОТКИ КАРБОНАТНОЙ УРАНОВОЙ РУДЫ В УКРАИНЕ

[1]Мухачёв А.П., [2]Елатонцев Д.А., [3]Нефёдов В.Г.

[1]Институт геотехнической механики им. Н.С. Полякова НАН Украины, [2]Днепровский государственный технический университет МОН Украины, [3]ГВУЗ «Украинский государственный химико-технологический университет»

Abstract. The purpose of the study was to represent the results of the first industrial tests of the technology for processing uranium carbonate ore with a target component content of 0.3%, by selective carbonate leaching with the separation of the uranium-containing solution from the rock, in the processes of thickening and decantation with using flocculants. This study was carried out to find out more effective flocculating reagents. Experimental data were collected from industrial slurries with the addition of polyacrylamide-based flocculants. The study showed that leaching in sodium carbonate media provided a high degree of purity of the product because metals impurities during leaching did not pass into solution. In line with earlier research, the process studies have confirmed the hypothesis that replacing of low-performance disc filters with automatic settling tanks significantly reduces labor and energy consumption. The results of the study convincingly show that the scheme for processing carbonate uranium ore made it possible to reduce the time of personnel contact with toxic and radioactive products. The main technical and economic indicators of the decantation scheme are given, methods for improving the decantation washing of uranium from pulps are described. Methods for more efficient use of flocculants and reduction of their consumption are proposed. Optimal modes of the process allowed to increase productivity and reduce production costs by 10% at the expense to reduce the consumption of electricity and steam, decommission energy-intensive vacuum pumps and filters. At the same time, the average extraction of uranium in the commercial solution – 90%. The results obtained can be applied in the uranium mining industry. To increase the productivity of the decantation scheme, further tests of new flocculating reagents, methods of their introduction are necessary; it is also of practical interest to study the conditions of flocculation depending on the salt composition of the washing solutions.

Keywords: uranium ore, flocculant, polyacrylamide, leaching, filtration, sedimentation rate.

INTRODUCTION. In uranium hydrometallurgy, the separation of solid and liquid phases and leaching of precipitation from valuable solutes are based on time-consuming processes of filtration and repulpation from acidic uranium-containing pulp [1, 2]. Previous studies have shown the effectiveness of flocculants based on polyacrylamide (PAA) for the separation of various sludge pulps that do not contain uranium [3].

Involvement in the production of various types of uranium ores remains a very important issue.

The content of uranium in ores mined in the 1950s reached 1%, but the low technical level of processing technology did not allow to achieve a degree of uranium extraction of more than 90% [4].

At that time, uranium shortages for the production of nuclear weapons forced the processing of poor uranium ores, and more than 90% of them were processed by a low-efficiency filtration scheme [5]. Studies [6] shown the possibility of leaching carbonate ores with soda with an efficiency of up to 90%. The high content of carbonates in the ore increased the consumption of acid during leaching, which significantly increased the cost of production [7].Until 1959, the uranium industry of Ukraine worked on the scheme of acid leaching with filtration of ore pulps on press filters and disk vacuum filters. From the resulting acidic solution, uranium precipitated in the form of ammonium uranyl tricarbonate [8]. The first carbonate uranium ores of industrial importance began to be processed in the United States in the late 1950s by acid and soda leaching by filtration-repulpation schemes [9–11]. Given the high content of carbonate compounds (> 10%), the most economically feasible scheme was soda autoclave leaching, which reduced reagent consumption and obtain fairly pure uranium concentrates [12–14]. A feature of the autoclave technology for processing carbonate ores in the United States was the low content of uranium (0.2%), high content of SiO_2 (up to 85%), and moisture (up to 18%) [15]. For crushing, the ore was dried in drum furnaces, then leaching was carried out in autoclaves with heat recovery in water heat exchangers. The cooled pulp was thickened with low productivity (up to 1 t/m^2·day) and filtered on disk vacuum filters. This technology required significant capital and operating costs [16].

Uranium is a very harmful chemical to human health, while the processing of acidic pulps by filtration requires a long time of contact with aggressive environments [17]. Carbonate leaching of uranium abroad was first reported in [18]. The first carbonate ore supplied to Ukraine from Romania (the Chudanovitsy deposit) contained up to 0.35% uranium and up to 30% carbonate compounds. For its processing, it was necessary to determine the reagents for leaching and to develop an experimental scheme for the separation of liquid and solid phases, because the previously used scheme was inefficient. This required solving of many technical tasks: determining the optimal conditions for ore leaching, providing conditions for a high degree of uranium extraction, the minimum consumption of flocculant and reagents, as well as studying the conditions of their utilization.

As a leaching agent, soda, sulfuric or nitric acid were used, and as an oxidant U(IV) to U(VI) - air, nitric acid, sodium hypochlorite, potassium permanganate were used. The use of soda required the utilization of Na^+ ions, for example, in the production of sodium nitrate. In addition, it was necessary to create and implement industrial technology for the production of effective flocculant, because this product was not produced by industry.

The purpose of this work is to present the results of the first in Ukraine industrial tests of carbonate ore leaching technology according to the settling-decantation scheme, determination of optimal technological parameters of pulp separation process after leaching with using flocculants, selection of the most effective one.

EXPERIMENTAL PART. The chemical and mineralogical composition of the ore is given in Tables 1 and 2.

Table 1 – Chemical composition of ore deposit "Chudanovitsy"

Component	Contents, %
U	0,33
SiO_2	51,5–52,85
Fe_2O_3	4,81–5,1
Al_2O_3	11,5–14,05
CaO	6,18–7,4
MgO	3,51–3,82
MnO	0,09–0,12
S	0,37–0,5
P_2O_5	0,033–0,094
V_2O_5	0,016–0,02
TiO_2	0,6–0,62
Cu	0,008–0,019
CO_2	9,77–10,44
Residue	13,22–13,53

Table 2 – Mineralogical composition of carbonate ore "Chudanovitsy"

Component	Content, %
Quartz	35
Carbonates	25
Sericite	25
Talc	10
Chlorite	
Hydroxides	
Kaolinite	
Pyrite	1,5
Hydroxel	1,0
Iron oxides	0,39
Anthraxolite	1,64
Accessory minerals	0,01

Based on the results of ore analysis, a basic technological scheme of carbonate ore processing was proposed for testing shown in Figure 1.

During the tests, the optimal technological parameters of all uranium extraction processes, its losses, reagent consumption, the chemical composition of raw materials, intermediates, finished products, tailings were determined. The calculations below were used to substantiate the required number of samples to obtain a representative sample of different products.

To justify the required number of samples to obtain a representative sample of products, the following formula is used

$$n = \left(\frac{t \cdot V}{P}\right)^2,$$

where n – the number of partial samples that make up the representative sample; t – probability coefficient equal to 2; P – permissible relative error when testing chemical concentrates; V – coefficient of variation of uranium content in the rock, determined by the formula

$$V = \frac{\sigma \cdot 100}{a},$$

where σ - the standard deviation of the arithmetic mean uranium content in the considered series is multiplied by 1.25, if the number of members of the series is more than 50; a - sample weight.

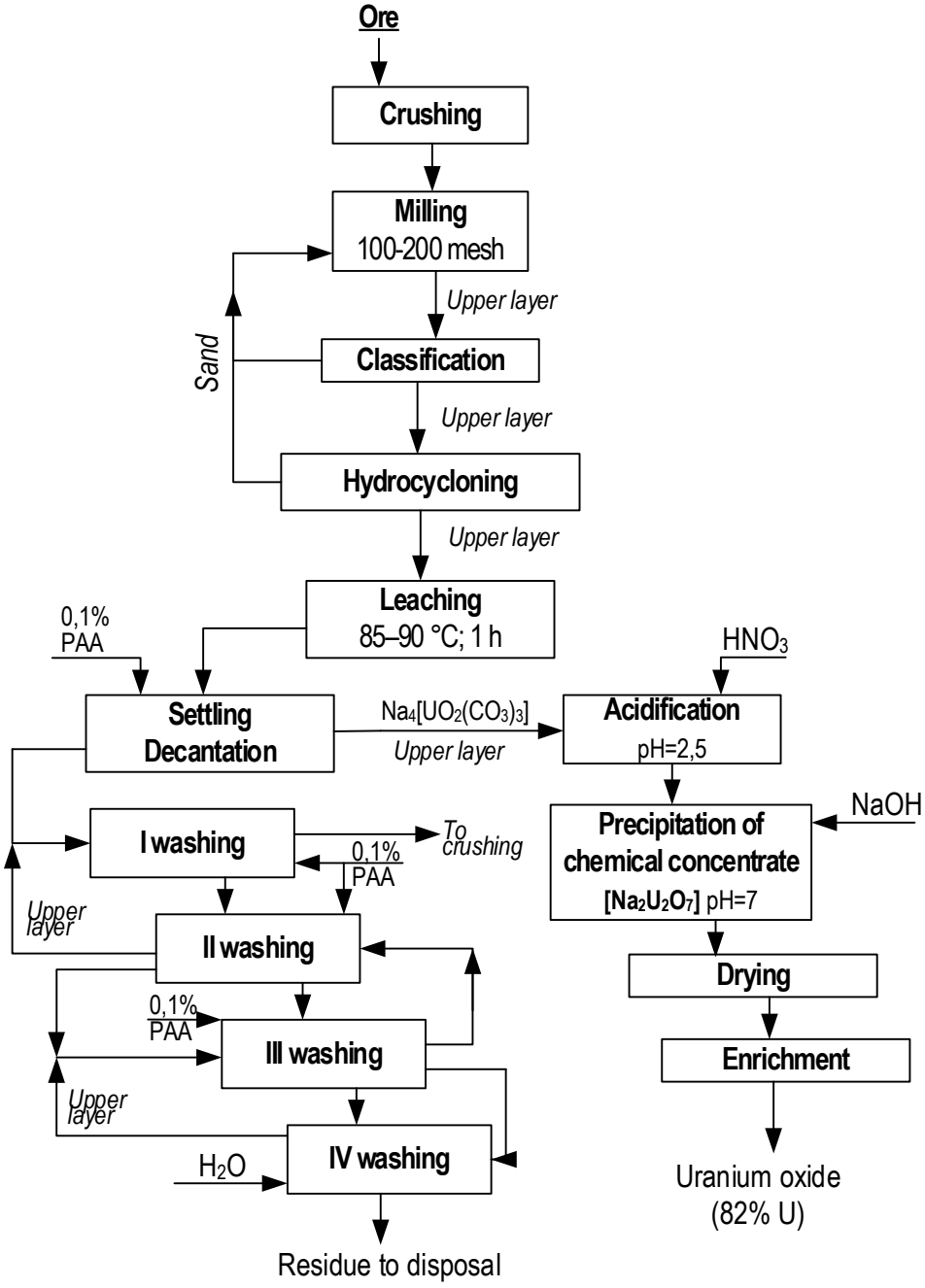


Figure 1 – Schematic technological scheme of processing of carbonate ore "Chudanovitsy"

As the members of the series decrease, the coefficient σ decreases accordingly. When determining the number of samples, the relative error of the analysis was taken into account. The content of uranium in raw materials and products is given in Table 3.

Table 3 – Uranium content in raw materials and products

Name	Uranium content	Determination limit,%	Relative allowable error of analysis,%
Sewage	0.001 g/dm^3	0–0.010 0.011–0.020	30 20
Pulp of dump cakes	0.032 %	0.021–0.050 0.051–0.070 0.071–0.120	15 8 8
Source ore	0.365 %	0.021–0.50 0.51–1.0	7 6
Solution for precipitation	1.25 g/dm^3	1.10–10.0 10.05–20.00 20.10–40.00	6 6 2
Pulp of chemical concentrates	68.5 %	< 40	1

Determination of the coefficients of variation of uranium content and the number of partial samples to obtain representative samples of different products was performed from the analysis of daily and variable samples. The costs of ore, the pulp of dump cakes, waste solutions, the pulp of chemical concentrates, uranium solution for precipitation, solution after the 4th filtration were analyzed.

The original ore was crushed to a size of 2–5 mm with further grinding in the ball mill. Classification of pulp after grinding was performed on spiral classifiers and hydro cyclones. Sands bigger than 0.15 mm were returned for grinding.

The quality of ore grinding for completeness of uranium leaching was studied. The optimal process parameters were determined: temperature, time, type and consumption of oxidant, excess carbonate. To ensure the completeness and quality of the processes of separation of solid and liquid phases, the quality of solidification of the solid phase in thickeners, the flow rate of wash water, the composition of decanters and cakes on uranium at each stage were studied. To study the quality and degree of extraction of uranium in the chemical concentrate, uranium samples were regularly taken. The number of stages of thickening and washing was determined by the analysis of waste cakes.

$KMnO_4$, NaClO, and air oxygen having standard oxidizing potentials +0.59 V, +0.89 V, +0.40 V, respectively, were used as oxidants in the alkaline medium. To determine the yield of uranium chemical concentrates, the processes of its deposition were studied and its quality was determined.

To select the most effective flocculant, studies were conducted with the addition of the following brands of flocculants:

- PAA – polyacrylamide obtained by polymerization of crystalline acrylamide in aqueous solution;

- AA + AMAC - a copolymer of acrylamide and methacrylic acid amide;
- styromal - saponified NaOH copolymer of styrene and maleic anhydride;
- ponimel - a copolymer of vinyl acetate and maleic anhydride;
- catapine - cation-active drug (para-alkylbenzylpyridine chloride);
- AMP-2.23 and AMP-1.7 - experimental polyacrylamide flocculant with a viscosity of 2.23 and 1.7 mm^2/s, respectively.

Synthesis and properties of polyacrylamide flocculant AMP

The AMF flocculant was obtained on an industrial scale by polymerization of acrylic acid amide in an aqueous solution in the presence of polymerization initiators – ammonium persulfate or sodium. Neutralization was carried out with ammonia, the acrylamide solution immediately after neutralization was fed for polymerization. The polymerization gave 8% jelly-like AMP, which was then dissolved in water to obtain a solution of the desired concentration. The average molecular weight of AMP is 10^6. In polymers, as a rule, molecules of different lengths and shapes are formed, so depending on the structure, the dissolution time of AMP was from 3 hours to 6 hours.

Semi-industrial tests of the settling-decantation scheme

Washing and thickening of carbonate pulp in the process of semi-industrial tests of the decantation scheme were first performed in thickeners with a diameter of 1800 mm and in spiral classifiers with a spiral diameter of 600 mm, with a flocculant consumption in the first stage of thickening 0.4 kg/t. Washing of the pulp was carried out in 5 thickeners with a diameter of 9000 mm and a height of 3600 mm. Pachuks with a diameter of 3100 mm and a height of 16000 mm were used for uranium leaching. The airflow rate for mixing and oxidation of uranium was varied from 100 m^3/t to 500 m^3/t.

Study of the effectiveness of batch administration of flocculant

A solution of AMF flocculant at a flow rate of 100 g/t , 200 g/t , 300 g/t and 450 g/t was introduced into the pulp in one go and when crushing it into separate portions. These amounts of flocculant were divided into 1, 2, 3, 4, etc. portions, up to 12 portions, successively introduced into the pulp. After the introduction of a certain number of portions, the suspension was stirred and the average thickening rate was determined.

Influence of mixing duration

To determine the optimal rate of pulp thickening, the effect of the duration of mixing with the addition of flocculant on the formation of flocs and their resistance to destruction was studied. Mixing was carried out by uniform rotation of the cylinders with the pulp. The rate of pulp thickening with different amounts of flocculant at different mixing times was determined.

Influence of pulp temperature

Thickening was performed at different temperatures (from 20 °C to 80 °C) both with flocculant and without it. The consumption of flocculant in all experiments was constant and amounted to 300 g/t.

The influence of the salt composition of the liquid phase of the pulp

The study of the rate of thickening of the carbonate pulp in the process of washing the solid phase was performed as follows. Different amounts of AMP flocculant were

introduced into the pulp and the thickening rate was determined, then the clarified solution was decanted and the same volume of soda solution with a concentration of 20 g/dm^3 after the first decantation and 10 g/dm^3 after the second, 5 g/dm^3 after the third and 1 g/dm^3 after the fourth decantation. The flocculant solution was introduced only during the first thickening. In each experiment, the average rate of pulp thickening was determined, and the concentration of uranium and carbonate ions in cakes and decantates was analyzed.

RESULTS AND DISCUSSION. To develop the technology of carbonate ore processing, laboratory and semi-industrial tests of individual processes were carried out, as a result of which it became possible to develop technological and hardware schemes for ore processing. During the test period, ~ 7000 tons of ore were processed.

In the course of tests, the corrosion of materials and the equipment was tested, including steel grade steel 3. The best results in terms of corrosion resistance were shown by steel C18N10T, due to which the main material for leaching was chosen bimetal, consisting of an inner layer, steel C18N10T with a thickness of 2 mm, and an outer layer of steel with a thickness of 6 mm.

To increase the extraction of uranium in the solution to 98.5%, it was necessary to provide grinding of ore to a size of 20–30% +100 mesh.

High extraction of uranium from carbonate ore was ensured by raising the leaching temperature to 130 °C and treatment in in autoclaves. In the absence of autoclaves, the the process of treatment was limited by the size of the grinding +10% 0.15 mm and a temperature of 90 °C.

The process of uranium leaching with soda is described by the equation

$$UO_2 + 0.5O_2 + 3Na_2CO_3 + H_2O \rightarrow Na_4[UO_2(CO_3)_3] + 2NaOH \qquad (1)$$

The uranium carbonate complex is stable and is in solution with an excess carbonate of 25–30 g/dm^3.

The process of precipitation of uranium in the chemical concentrate was carried out by acidification of the solution with nitric acid to pH = 2.5, followed by the introduction of NaOH solution to pH = 7.0–7.5.

The precipitation reaction of chemical concentrates is described by the equation

$$2Na_4[UO_2(CO_3)_3] + 6HNO_3 \rightarrow Na_2U_2O_7 + 6NaNO_3 + 3H_2O + 6CO_2\uparrow \qquad (2)$$

Blowing the solution with air was used to remove CO_2. The optimal airflow rate, as the cheapest oxidizer, was 100 m^3/t of ore. This ensured the complete oxidation of U(IV) to U(VI). Uranium extraction without oxidant did not exceed 88%.

The presence of sodium bicarbonate in the solution prevents the precipitation of uranium by the alkali released by reaction (1).

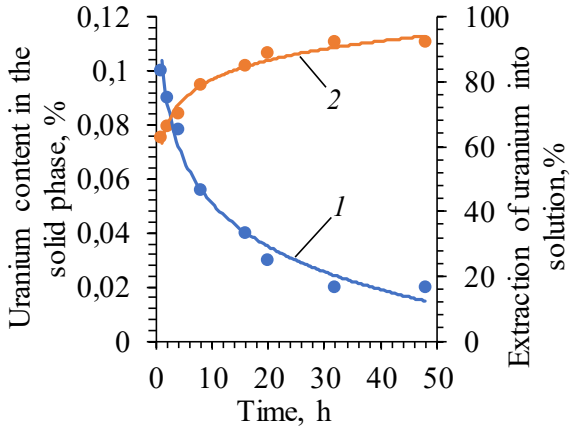
The consumption of soda was 5% by weight of ore. In the case of the introduction of reversible mother liquor (II decantate) soda consumption is reduced to 1.5%.

To purify $Na_2U_2O_7$ concentrate, Na_2CO_3 was used, which converted uranium into a soluble reaction complex

$$Na_2U_2O_7 + 6Na_2CO_3 + 3H_2O \rightarrow 2Na_4[UO_2(CO_3)_3] + 6NaOH \quad (3)$$

Excess NaOH must be neutralized with acid to pH = 2.5 so that the reaction to obtain ammonium uranyl tricarbonate is complete. Recrystallization of ammonium uranyl tricarbonate allowed, after drying and calcination, to obtain uranium oxide of nuclear purity, with a uranium content of 82%.

The influence of leaching time on the extraction of uranium at a hydraulic modulus of 1:1.6, a temperature of 85 °C, and a uranium content in the ore of 0.266% is shown in Figure 2.



1 – uranium content in the solid phase; 2 – extraction of uranium into solution

Figure 2 – The influence of leaching time on the extraction of uranium

As it can be seen from the fig. 2, the required leaching time of uranium is at least 32 hours. This ensures the extraction of uranium in the solution of at least 92%.

Uranium extraction is directly affected by oxidant consumption. Of all the studied types of oxidants, the most economical and efficient was air. The extraction of uranium depending on the consumption of the oxidant is shown in Table 4.

Table 4 - The results of precipitation of uranium from solution after leaching for 1.5 hours

Component	Concentration, g/dm^3
basic solution	
Uranium	1.63–2.16
carbonate	40–50
bicarbonate	7.6–16.8
mother liquor	
Uranium	0.024–0.064
carbonate	56.2–74.0
bicarbonate	0–16
alkalinity	0.8–9.2
chemical concentrate	
Uranium	58.5–66.2

The consumption of NaOH was 1% by weight of the ore, the extraction of uranium in the concentrate – 97.0–98.7%. When diluting the stock solution with the first repulpate (1 g/dm^3), 150 mg/dm^3 of uranium remains in the mother liquor. Therefore, it is advisable to use for re-leaching of ore.

In the course of industrial tests of carbonate ore processing, the following technical indicators were obtained (Table 5).

Table 5 – Test results of carbonate leaching of uranium

Indicator	Unit	Value
Uranium content in the ore	%	0.347
Average daily processing volume	t	320
Extraction of uranium into the solution	%	84.7–89.2
Soluble uranium losses	%	
with cake		8.3–9.8
with mother liquor		0.03–0.05

The average uranium content in the products by stages of technology is shown in Table 6.

Table 6 – Average uranium content in the products

Naming	Unit	Value
Source ore	%	0.347
I cake		0.12
II cake		0.08
III cake		0.067
IV cake		0.04
Dump cake		0.035
Chemical concentrate		63.5
The main filtrate	g/dm^3	1.36
I repulpate		0.27
II repulpate		0.13
III repulpate		0.1
Mother liquor		0.001

Consumption of reagents and materials is shown in Table 7.

Table 7 – Consumption of reagents and materials

Naming	Unit	Value
Na_2CO_3	t	0.1
HNO_3	t	0.09
NaOH	t	0.03
Electricity	kW·h	108
Vapour	MW	0.79
Water	m^3	35
Air	m^3	200

Technological indicators of the decantation scheme are given in Tables 8–10.

Table 8 – Technological indicators of uranium extraction according to the settling-decantation scheme

Characteristic	Value
Specific productivity of thickening, t/m^2·day	5.0
Consumption of AMP, kg/t	0.49
Uranium content,%	
in the ore	0.305
in washed cakes	0.030
in the chemical concentrate	57.6
Uranium extraction,%	
when leaching	90.15
after washing	89.8
Loss of uranium with cake, %	0.35

Table 9 – Chemical composition of I decantate (commodity solution)

Component	Content, g/dm^3
U	0.67
SO_4^{2-}	4.04
N	5.85
V_2O_5	<0.01
P	0.02
Al_2O_3	0.15
Fe	<0.01

Table 10 – Chemical composition of waste tails according to the settling-decantation scheme

Component	Content, g/dm^3
SiO_2	49.97–50.4
Fe_2O_3	4.68–5.09
Al_2O_3	12.72–13.51
CaO	6.29–7.41
MgO	3.80–4.53
MnO	0.13–0.22
S	0.19–0.42
P_2O_5	0.084–0.090
V_2O_5	0.019–0.060
TiO_2	0.56–0.59
Cu	0.01–0.012
Residue	14.57–15.1

Chemical analysis of the tailings confirmed the high purity of the chemical concentrates because the impurities of transition metals and silicon remained in the solid phase.

In the study of the decantation scheme, the optimal technological parameters of each operation were determined:

- ore grinding: grinding size 10% fraction +100 mesh; hydraulic module of the pulp: at crushing – 0.4–0.55:1; in the drain of the classifier – 0.8–1.2:1; in the hydrocyclone – 1.8–3.0:1.

- leaching: hydraulic module of the source pulp 2.0–2.5:1; leaching temperature 80–90 °C; excess carbonate content of 30 g/dm^3; leaching duration 30 hours; airflow for mixing 200 m^3/t.

- decantation washing. I stage of washing: the hydraulic module of the source pulp 3.0-3.5:1; the temperature of the pulp at thickening – 70–80 °C; excess carbonate content of 25 g/dm^3; hydro module of condensed pulp 1.0–1.2:1; the content of the solid phase in the drain is not more than 0.4 g/dm^3; consumption of flocculant 0.1–0.3 kg/t II, III, IV and V stages of washing: the hydro module of the original pulp 3.0–3.5:1; pulp temperature 45–50 °C; hydro module of condensed pulp 1.0–1.2:1; solids content <0.4 g/dm^3; consumption of flocculant 0.3 kg/t

- deposition of chemical concentrate: uranium content in the initial solution 0.6–0.8 g/dm^3, in the filtrate – 0.002 g/dm^3, in the chemical concentrate – 55%; solid phase content <0.4 g/dm^3; pH = 2.5; blowing air to the absence of CO_2.

In the process of leaching, the airflow was reduced from 500 m^3/t to 200 m^3/t by crushing the airflow with ceramic dispersants. Pachuks were reconstructed, each had an additional airlift for pumping sand (fraction +0.074 mm). During the development of the settling and decantation scheme, shortcomings in the design of pumps and automation schemes were eliminated, after which it began to work stably.

Technical and economic indicators of soda leaching technology are given in comparison with the standard filtration scheme (Table 11).

Table 11 – The main technical and economic indicators of the schemes

Indicator	Filtration scheme	Decantation scheme
Uranium extraction into the product solution,%	86.9	86.9
Quantity vacuum pumps disk filters repulpators thickeners	 6 48 68 -	 - - - 5
Consumption of filter cloth, m/t	0.8	-
Electricity, kW· h	100	65
Uranium content in the liquid phase of the marketable product, g/dm^3	1.0	0.731
The solid phase content in the solution before the deposition of the chemical concentrate, g/dm^3	≤1.0	0.4
The cost of 1 ton of uranium, USD	100	87.4

The simplicity of operations of thickening and washing, reduction of complexity of service of devices, and elimination of contact of personnel with uranium solution allowed to replace the process of filtration on disk vacuum filters by thickening. This has significantly improved industrial sanitation and facilitated working conditions for

staff. The cost of ore processing decreased by 12.6%, compared to the filtration scheme.

Research of different types of flocculants

The graph (Figure 3) shows the dependence of the rate of thickening on the consumption of different types of flocculants.

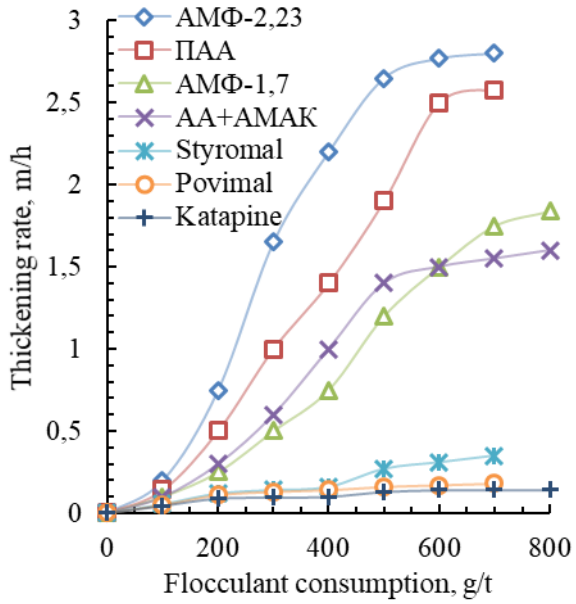


Figure 3 – The rate of thickening of the carbonate pulp depending on the consumption of flocculants

As it can be seen from Figure 3, the most effective flocculant for thickening the carbonate pulp is AMP. The molecular weight of the polymer depends on the viscosity of the solution, so the graph (Figure 4) shows the dependence of the rate of thickening on the consumption of flocculant with different viscosity.

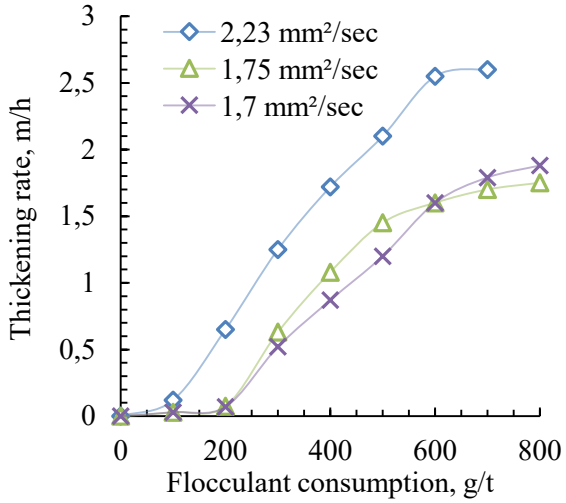


Figure 4 – The rate of thickening of the carbonate pulp with the addition of flocculant AMP with different viscosities

As expected, polyacrylamide flocculant with higher viscosity has a more pronounced effect. Thus, when obtaining a higher molecular weight polyacrylamide, it is possible to reduce its consumption while maintaining high process productivity.

Fig. 5 shows the dependence of the rate of thickening of the carbonate pulp with the addition of flocculant AMP at different numbers of portions.

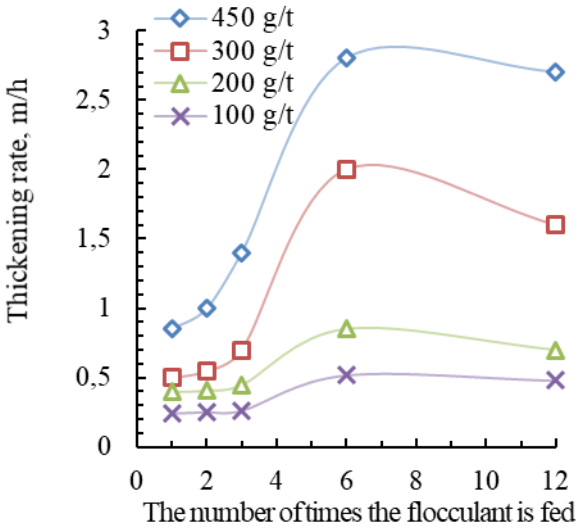


Figure 5 – The rate of thickening of the pulp with the introduction of portions of the AMP flocculant

As it is seen from the fig. 5, the best effect is achieved by introducing a solution of flocculant, crushed into 6–8 portions. Further crushing leads to deterioration of the flocculation process. Batch administration is usually carried out by dosing a solution of flocculant at several points in the pulp. For each suspension, the number of flocculant injection points, as well as the mixing time of the flocculant solution with the pulp, was determined experimentally.

The results of studies of the rate of thickening of the pulp are shown in Fig. 6.

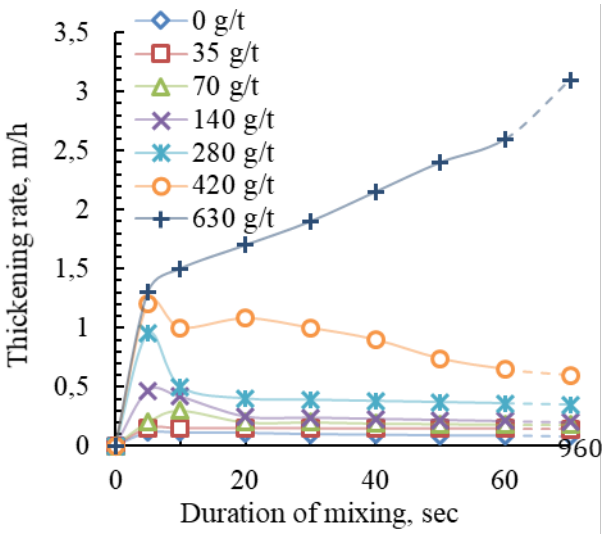


Figure 6 – The rate of thickening of the pulp with the addition of flocculant with different duration of mixing

As it is seen from the fig. 5, with a single introduction of a small amount of flocculant, the duration of mixing significantly affects the process of formation and destruction of flocs. Thus, at a consumption of up to 420 g/t after stirring for 5–6 sec there is a decrease in the rate of thickening. This is especially noticeable with small additions of flocculant. Destruction of the flocs is achieved after 8–10 sec and with subsequent mixing, the rate of thickening does not change or changes very little. With an excessive amount of flocculant, increasing the duration of mixing, on the contrary, leads to the enlargement of the flocs and increases the average rate of thickening. Most likely, this phenomenon is associated with the continued adsorption of excess flocculant molecules on solid particles and their aggregation.

The results of experiments to establish the effect of temperature on the thickening process are shown in Fig. 7.

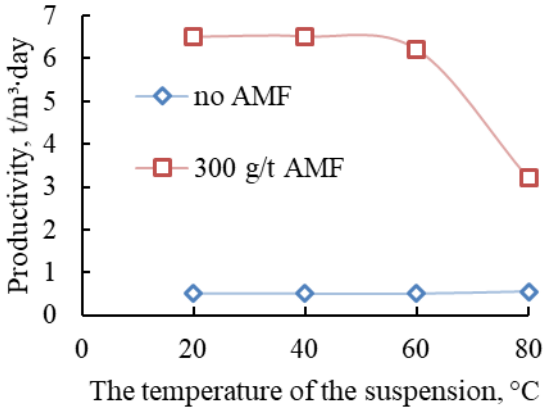


Figure 7 – Influence of temperature on the process of flocculation of carbonate pulp

With the increasing temperature of the carbonate pulp, there is a decrease in the rate of thickening. Especially sharp decrease is shown at the thickening of pulp with the temperature exceeding 60 °C. When thickening the pulp without the addition of flocculant with increasing temperature, there is a slight increase in the speed of the process. The decrease in the efficiency of the flocculant with increasing pulp temperature is probably due to the change in the properties of AMP in hot saline solutions.

The results of the study of the productivity of pulp thickening are shown in Fig. 8.

In the process of washing, there is some increase in the rate of thickening of the pulp, as a result of reducing the salt content of the liquid phase. However, at the final stage of washing, the thickening of the pulp is slowed down, probably as a result of partial hydration of the surface of the solid particles.

Ways of further development of the decantation method

Studies of the process of decantation washing of uranium-containing carbonate pulps obtained after ore leaching have shown the prospects of the soda method of processing carbonate uranium ores. Large-scale use of decantation processes in the uranium and rare earth industries can be considered promising if the following tasks are solved:

1. If necessary, processing of complex uranium-molybdenum, uranium-vanadium, uranium-iron-containing, and other ores to obtain solutions for sorption.

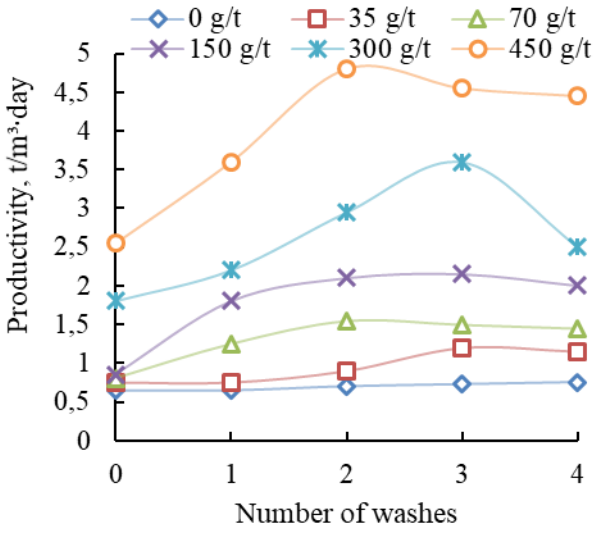


Figure 8 - The productivity of pulp thickening during the washing the solid phase

2. After the development of cheaper highly effective flocculants to reduce their consumption.

3. Creating of more efficient devices for pulp separation.

4. After improving the design of thickeners-settlers and the organization of spontaneous movement of the liquid phase through the system of thickeners.

As a result of research, a technological scheme for processing uranium iron carbonate ore was developed, shown in Fig. 9.

Improvement of the developed scheme of uranium extraction from pulp consists in full automation of the process of decantation washing of condensed pulp with use of sensors and improvement of pumps at a stage of unloading. For stable operation of the process, it is necessary to continuously measure the density of the condensed product and adjust the speed of its pumping from one thickener to another, depending on the density.

CONCLUSIONS

1. Technological indicators of the scheme of uranium extraction from carbonate ore of the Chudanovytsia deposit using soda leaching and countercurrent decanting leaching of uranium using AMF flocculant on an industrial scale confirmed the high efficiency of the proposed technical solutions. The consumption of acid to the weight of the ore (~ 21%) was significantly reduced compared to the technology of sulfuric acid leaching (~ 35%).

2. With an average uranium content in the source ore of 0.3%, the following indicators were achieved:

- number of stages of washing – 4;
- hydro module of the source pulp – 3,5–3,2:1, dump pulp – 1,0–0,9:1;
- uranium content in washed cakes after leaching – 0.03%;
- uranium content in the liquid phase of the dump pulp – 0.036 g/dm^3;

- the average uranium content in the first decantate is 0.73 g/dm^3;
- flushing water consumption – 2.5 m^3/t;
- specific productivity of thickener – 5–6 t/m^2·days;
- total consumption of flocculant – 0.4–0.5 kg/t;
- air consumption – 100 m^3/t;
- average extraction of uranium in the commercial solution – 90%;

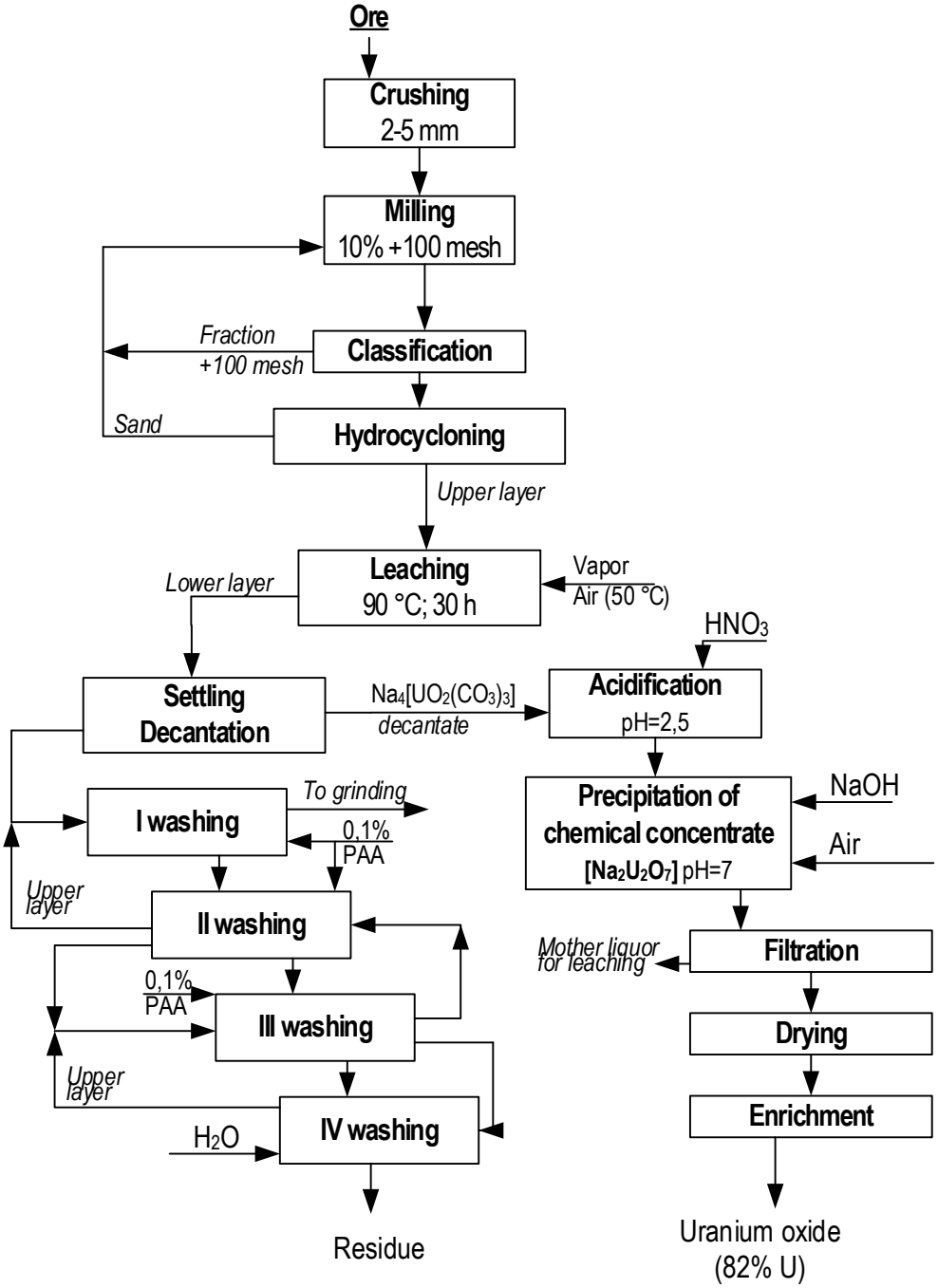


Figure 9 – Improved technological scheme of iron carbonate uranium ore processing

3. The introduction of the decantation scheme allowed to reduce the consumption of electricity and steam, decommission energy-intensive vacuum pumps and filters, and reduce the total cost of ore processing by 12.6%.

REFERENCES

1. Namazov, Sh.S., Beglov, B.M., Seitnazarov, A.R., Dekhkanov, Z.K. and Abdukodirov, E.A. (2017), “Enrichment of natural phosphate raw materials”, *Chemical Journal of Kazakhstan*, no. 2, pp. 204-226.

2. Svetlov, A.V. (2018), “Development of beneficiation methods for complex objects of non-ferrous metallurgy in the Murmansk region”, *Mineralogy of Technogenesis*, no. 19, pp. 205-216.

3. Yakubovich, I.A. (1961), “The flocculation of pulp and polyacrylamide-type flocculants”, *The Soviet Journal of Atomic Energy*, vol. 8, no. 6, pp. 449–454. https://doi.org/10.1007/BF01483664

4. Petukhov, V.I. (ed.) (2016), *Kompleksnoe ustoychivoe upravlenie otkhodami. Gornodobyvayushchaya promyshlennost* [Integrated sustainable waste management. Mining industry], Academy of Natural Sciences, Moscow, Russia.

5. Allabergenov, R.D. (2012), *Fundamentalnaya i prikladnaya gidrometallurgiya* [Fundamental and applied hydrometallurgy]. State Enterprise "Scientific Research Institute of Mineral Resources", Tashkent, Uzbekistan.

6. Serajuddin, M. and Sreenivas, T. (2015), "Application of response surface method of optimization in flocculant assisted filtration of uranium bearing alkaline leach slurry of limestone origin", Mineral Processing and Extractive Metallurgy, vol. 124, pp. 199-207. https://doi.org/10.1179/1743285515Y.0000000003

7. Serajuddin, M., Rao, K.A. and Sreenivas, T. (2016), "Studies on optimization of parameters for dewatering of ground calcite-rich ore", Russian Journal of Non-Ferrous Metals, vol. 57, no. 4, pp. 273-286. https://doi.org/10.3103/S106782121604012X

8. Kozhura, O.V., Pasenko, A.A., Ishchenko, L.E., Pismenny, B.V. and Chuiko, T.V. (2013), “Application of the product of hydrolysis of polyacrylonitrile waste in uranium hydrometallurgy”, *Voprosy khimii i khimicheskoy tekhnologii*, no. 2, pp. 89-92.

9. Vitam, K.A., Hansen, M.J., Hitt, J.E., Moler, K.E. and Poindexter, M.K. (2019), *Polimernye flokulyanty dlya obrabotki suspenziy mineralov* [Polymeric flocculants for the treatment of suspensions of minerals], Patent Office, Moscow, Pat. № RU2685217C2.

10. Trampus, B.C. and Franca, S.C. (2020), "Performances of two flocculants and their mixtures for red mud dewatering and disposal based on mineral paste production", Journal of Cleaner Production, vol. 257, 120534. https://doi.org/10.1016/j.jclepro.2020.120534

11. Wolkersdorfer, C., Nordstrom, D.K., Beckie, R.D., Cicerone, D.S., Elliot, T., Edraki, M. and Gil, A. S. (2020), "Guidance for the Integrated Use of hydrological, geochemical, and isotopic tools in mining operations", Mine Water and the Environment, vol. 39, no. 2, pp. 204-228. https://doi.org/10.1007/s10230-020-00666-x

12. Golberg, G.Yu. (2019), "Development of the theory of formation and destruction of flocculation structures in the processes of separation of suspensions of finely dispersed products of coal concentration", D. Sc. Thesis, Institute for the Problems of Complex Development of Subsoil Academician N.V. Melnikov, Karaganda, Kazakhstan.

13. Dhawan, N., Safarzadeh, M.S., Miller, J.D., Moats, M.S. and Rajamani, R.K. (2013), "Crushed ore agglomeration and its control for heap leach operations", Minerals Engineering, vol. 41, pp. 53-70. https://doi.org/10.1016/j.mineng.2012.08.013

14. Shakhalov, A.A., Ospanov, E.A., Naboychenko, S.S. and Fomenko, I.V. (2019), "Features of pressure oxidative leaching of substandard copper-zinc sulfide concentrates", Non-ferrous metals, vol. 1, pp. 13-19. https://doi.org/10.17580/tsm.2019.01.02

15. Zhang, P., Liang, H., Jin, Z. and DePaoli, D. (2017), "The ultimate mineral processing challenge: Recovery of rare earths, phosphorus and uranium from Florida phosphatic clay", Minerals and Metallurgical Processing, vol. 34, no. 4, pp. 183-188. https://doi.org/10.19150/mmp.7858

16. Hall, S.M., Van Gosen, B.S., Paces, J.B., Zielinski, R.A. and Breit, G.N. (2019), "Calcrete uranium deposits in the Southern High Plains, USA", Ore Geology Reviews, vol. 109, pp. 50-78. https://doi.org/10.1016/j.oregeorev.2019.03.036

17. Mattus Jr, A.J. and Torma, A.E. (1980), "A comparison of carbonate leaching of a low-grade uranium ore at atmospheric and increased pressures", Hydrometallurgy, vol. 5, no. 2-3, pp. 179-190. https://doi.org/10.1016/0304-386X(80)90037-7

18. Zefirov, A.P., Nevsky, B.V. and Ivanov, G.F. (1962), *Zavody po pererabotke uranovykh rud v kapitalisticheskikh stranakh* [Uranium ore processing plants in capitalistic countries], Nedra, Moscow, SU.

СПИСОК ЛІТЕРАТУРИ

1. Обогащение природного фосфатного сырья / Намазов Ш. С., Сейтназаров А. Р., Беглов Б. М. и др. / Химический журнал Казахстана. 2017. № 2. С. 204–226.

2. Светлов А. В. Разработка методов обогащения для сложных объектов цветной металлургии Мурманской области / Минералогия техногенеза. 2018. Т. 19. С. 205–216.

3. Якубович И. А. Флокуляция пульп и синтетические флокулянты полиакриламидного типа / Атомная энергия. 1960. Т. 8, вып. 6. С. 489–591. https://doi.org/10.1007/BF01483664

4. Комплексное устойчивое управление отходами. Горнодобывающая промышленность: учебное пособие / под ред. В.И. Петухова. М.: Изд. дом Академии Естествознания, 2016. 638 с.

5. Аллабергенов Р. Д. Фундаментальная и прикладная гидрометаллургия. Ташкент: ГП «Научно-исследовательский институт минеральных ресурсов», 2012. 276 с.

6. Serajuddin, M. and Sreenivas, T. (2015), "Application of response surface method of optimization in flocculant assisted filtration of uranium bearing alkaline leach slurry of limestone origin", Mineral Processing and Extractive Metallurgy, vol. 124, pp. 199-207. https://doi.org/10.1179/1743285515Y.0000000003

7. Serajuddin, M., Rao, K.A. and Sreenivas, T. (2016), "Studies on optimization of parameters for dewatering of ground calcite-rich ore", Russian Journal of Non-Ferrous Metals, vol. 57, no. 4, pp. 273-286. https://doi.org/10.3103/S106782121604012X

8. Применение продукта гидролиза отходов полиакрилонитрила в гидрометаллургии урана / Кожура О. В., Пасенко А. А., Ищенко Л. Е. и др. / Вопросы химии и химической технологии. 2013. № 2. С. 89–92.

9. Полимерные флокулянты для обработки суспензий минералов: пат. 2685217C2 RU / Витам К.А. и др. № 2016 146717; заявл 30.05.2018; опубл. 16.04.2019, Бюл. № 11. 8 с.

10. Trampus, B.C. and Franca, S.C. (2020), "Performances of two flocculants and their mixtures for red mud dewatering and disposal based on mineral paste production", Journal of Cleaner Production, vol. 257, 120534. https://doi.org/10.1016/j.jclepro.2020.120534

11. Wolkersdorfer, C., Nordstrom, D.K., Beckie, R.D., Cicerone, D.S., Elliot, T., Edraki, M. and Gil, A. S. (2020), "Guidance for the Integrated Use of hydrological, geochemical, and isotopic tools in mining operations", Mine Water and the Environment, vol. 39, no. 2, pp. 204-228. https://doi.org/10.1007/s10230-020-00666-x

12. Гольберг Г.Ю. Развитие теории образования и разрушения флокуляционных структур в процессах разделения суспензий тонкодисперсных продуктов обогащения углей: дис. ... докт. техн. наук. / Караганда: Институт проблем комплексного освоения недр им. академика Н.В. Мельникова, 2019. 148 с.

13. Dhawan, N., Safarzadeh, M.S., Miller, J.D., Moats, M.S. and Rajamani, R.K. (2013), "Crushed ore agglomeration and its control for heap leach operations", Minerals Engineering, vol. 41, pp. 53-70. https://doi.org/10.1016/j.mineng.2012.08.013

14. Особенности автоклавного выщелачивания некондиционных сульфидных медно-цинковых концентратов / Шахалов А. А., Оспанов Е. А., Набойченко С. С. И др. / Цветные металлы. 2019. № 1. С. 13–19. https://doi.org/10.17580/tsm.2019.01.02

15. Zhang, P., Liang, H., Jin, Z. and DePaoli, D. (2017), "The ultimate mineral processing challenge: Recovery of rare earths, phosphorus and uranium from Florida phosphatic clay", Minerals and Metallurgical Processing, vol. 34, no. 4, pp. 183-188. https://doi.org/10.19150/mmp.7858

16. Hall, S.M., Van Gosen, B.S., Paces, J.B., Zielinski, R.A. and Breit, G.N. (2019), "Calcrete uranium deposits in the Southern High Plains, USA", Ore Geology Reviews, vol. 109, pp. 50-78. https://doi.org/10.1016/j.oregeorev.2019.03.036

17. Mattus Jr, A.J. and Torma, A.E. (1980), "A comparison of carbonate leaching of a low-grade uranium ore at atmospheric and increased pressures", Hydrometallurgy, vol. 5, no. 2-3, pp. 179-190. https://doi.org/10.1016/0304-386X(80)90037-7

18. Зефиров А. П., Невский Б. В., Иванов Г. Ф. Заводы по переработке урановых руд в капиталистических странах. М.: Недра, 1962. 371 с.

About the authors

Mukhachev Anatolii Petrovych, Candidate of Physics and Mathematics (Ph.D), Senior Researcher in Department of Vibratory Transporting Systems and Complexes, Institute of Geotechnical Mechanics named by N. Poljakov of the National Academy of Sciences of Ukraine (IGTM of NAS of Ukraine), Dnipro, Ukraine, map45@ukr.net

Yelatontsev Dmytro Oleksandrovych, Candidate of Technical Sciences (Ph.D), Associate Professor of the Department of Chemical Technology of Inorganic Substances, Dnipro State Technical University under the Ministry of Education and Science of Ukraine (DSTU of MES of Ukraine), Kamianske, Ukraine, ya.nah2015@yandex.com

Nefedov Volodymyr Heorhiiovych, Doctor of Technical Sciences (D. Sc), Professor, Professor of the Department of Technology of Inorganic Substances and Ecology, Ukrainian State University of Chemical Technology (USUCT), Dnipro, Ukraine, vnefedov@i.ua

Про авторів

Мухачов Анатолій Петрович, кандидат фізико-математичних наук, старший науковий співробітник у відділі вібротранспортних систем и комплексів, Інститут геотехнічної механіки ім.М.С. Полякова Національної академії наук України (ІГТМ НАН України), Дніпро, Україна, map45@ukr.net

Єлатонцев Дмитро Олександрович, кандидат технічних наук, доцент кафедри хімічної технології неорганічних речовин, Дніпровський державний технічний університет Міністерства освіти і науки України (ДДТУ МОН України), Кам'янське, Україна, ya.nah2015@yandex.com

Нефьодов Володимир Георгійович, доктор технічних наук, професор, професор кафедри технології неорганічних речовин і екології, ДВНЗ «Український державний хіміко-технологічний університет» (ДВНЗ «УДХТУ»), Дніпро, Україна, vnefedov@i.ua

---

Анотація. Метою дослідження був розгляд результатів перших промислових випробувань технології переробки уранової карбонатної руди із вмістом цільового компоненту 0,3 %, методом селективного карбонатного вилуговування з відділенням розчину урану від породи, в процесах згущення і декантації із застосуванням флокулянтів. Це дослідження було здійснено для з'ясування ефективніших флокулюючих реагентів. Дослідні дані були зібрані на цехових пульпах із добавкою флокулянтів на основі поліакриламіду. Дослідження показало, що вилуговування у середовищі карбонату натрію забезпечило високий рівень чистоти готової продукції, тому що домішки перехідних металів при вилуговуванні не переходили в розчин. Відповідно до більш раннього дослідження, вивчення процесів підтвердило гіпотезу про те, що заміна низькопродуктивних дискових фільтрів на відстійники, що працюють в автоматичному режимі, дозволило значно знизити трудомісткість та енергоспоживання. Результати дослідження переконливо показують, що схема переробки карбонатної уранової руди дозволила скоротити час контактування персоналу з токсичними та радіоактивними

продуктами. Наводяться основні техніко-економічні показники декантаційної схеми, описуються методи удосконалення декантаційного відмивання урану з пульпи. Запропоновано способи більш ефективного використання флокулянтів та скорочення їх витрати. Оптимальні режими ведення процесу дозволили збільшити продуктивність та знизити собівартість продукції на 10 % за рахунок зниження витрат електроенергії та пари, виведення з експлуатації енергоємних вакуумних насосів та фільтрів. При цьому середнє вилучення урану в товарний розчин становило 90%. Таким чином, отримані результати можуть бути застосовані в урано-видобувній промисловості. З метою збільшення продуктивності декантаційної схеми, необхідні подальші випробування нових флокулюючих реагентів, методів їх введення, також представляє практичний інтерес вивчення умов флокулоутворення залежно від сольового складу промивних розчинів.

Ключові слова: уранова руда, флокулянт, поліакриламід, вилуговування, фільтрація, швидкість осадження.

Аннотация. Целью исследования было рассмотрение результатов первых промышленных испытаний технологии переработки урановой карбонатной руды с содержанием целевого компонента 0,3 %, методом избирательного карбонатного выщелачивания с отделением урансодержащего раствора от породы, в процессах сгущения и декантации с применением флокулянтов. Это исследование было осуществлено для выяснения более эффективных флоккулирующих реагентов. Опытные данные были собраны на цеховых пульпах с добавкой флокулянтов на основе полиакриламида. Исследование показало, что выщелачивание в среде карбоната натрия обеспечило высокую степень чистоты готовой продукции, т.к. примеси переходных металлов при выщелачивании не переходили в раствор. В соответствии с более ранним исследованием, изучение процессов подтвердило гипотезу о том, что замена низкопродуктивных дисковых фильтров на отстойники, работающие в автоматическом режиме, позволило значительно снизить трудоемкость и энергопотребление. Результаты исследования убедительно показывают, что схема переработки карбонатной урановой руды позволила сократить время контактирования персонала с токсичными и радиоактивными продуктами. Приводятся основные технико-экономические показатели декантационной схемы, описываются методы усовершенствования декантационной отмывки урана из пульп. Предложены способы более эффективного использования флокулянтов и сокращения их расхода. Оптимальные режимы ведения процесса позволили увеличить производительность и снизить себестоимость продукции на 10 % за счет снижения расхода электроэнергии и пара, выведения из эксплуатации энергоемких вакуумных насосов и фильтров. При этом среднее извлечение урана в товарный раствор составило 90%.. Таким образом, полученные результаты могут быть применены в уранодобывающей промышленности. С целью увеличения производительности декантационной схемы необходимы дальнейшие испытания новых флоккулирующих реагентов, методов их введения, также представляет практический интерес изучение условий флоккулообразования в зависимости от солевого состава промывных растворов.

Ключевые слова*:* урановая руда, флокулянт, полиакриламид, выщелачивание, фильтрация, скорость осаждения.

The manuscript was submitted 24.03.2021