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INTENSIFICATION OF LEACHING OF URANIUM CONCENTRATES

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In work results of experiments on a lixiviation of a concentrate of uranium are explained. It is shown that transfer of process of a lixiviation to the pulsation device allows to intensify it, to reduce losses of uranium and hydrogen nitrate and to reduce process temperature. The pulsation device allows to automate process and to provide positive control to them.

INTRODUCTION

Chemical concentrates of rare metals are important reprocessing products in hydrometallurgical processes; their processing should be effective because of the high cost. This is especially important for the production of uranium, nuclear fuel for Ukrainian NPPs. The share of the uranium extracted by a lixiviation method continuously grows [1]. The concentrate of uranium is a commercial product.

Processing of a concentrate is transfer of uranium to a liquid phase. Uniform mixing is not ensured.

The theoretical foundations of the leaching process and the problems of its intensification are discussed in detail in [2], but the combination of physical effects with leaching is not fully described.

The pulsation columns with a nozzle which are more efficient devices for a lixiviation [3] allow to eliminate the specified defects. They well fit into any operating instrument scheme of production, are devices of ideal replacement and allow to maintain the given time of a lixiviation precisely.

Air and nozzles in plates increase a response time of components in the pulsation column. The device does not contain moving parts, is characterized by high removal of a product from unit volume. In the course of a lixiviation decrease in specific energy consumption is reached [4, 5].

During pulsation, the liquid phase flows from the plate to the plate, without lingering or creating a dense layer. The presence of distribution properties of the nozzle leads to the fact that the liquid phase and the reagent are evenly distributed over the section and height, which excludes the possibility of formation of stagnant zones or “breakthroughs”.

The solids are leached in the surrounding liquid medium and their entire surface is accessible for contact.

The rate of the process is determined mainly by the kinetics of internal diffusion and by the time necessary for the reaction to proceed. Intensification of the leaching process in the column is achieved by supplying additional energy. The superposition of the vibrational (reciprocating) motion on the volume of the liquid is converted, due to the nozzle, and is evenly distributed over the cross section.

The pulse generator is located outside the device, so the cost of creating the installation is reduced, and the process becomes easier and cheaper.

The pulsating column can operate in a direct-flow and counter-current mode.

The purpose of the experiment was to determine the possibility of deep leaching of uranium when replacing the reactor with a pulsating column.

TECHNIQUE OF THE EXPERIMENT

The process of leaching of uranium was carried out in a reactor with a stirrer and a pulsating column. The nitric acid in the reactor was preheated to 70 °C. The concentrate was periodically dosed into the acid so that there was no ejection. The acid concentration was kept constant. The process time was 1 hour.

Due to the reaction heat, the temperature in the reactor rises to 90 °C. After the end of the process, the product is filtered. The solution is sent for extraction to purify uranium from impurities. The sediment is washed and sent for disposal. The reaction products were analyzed for uranium content.

According to the analysis, the dependence of the degree of leaching on the rate of mixing, temperature, nitric acid concentration, and the duration of the process was determined. After studying the physico-chemical parameters of the process, the physico-chemical properties of the chemical concentrate were studied, and the difficult-to-reveal classes that influenced extraction were determined.

Taking into account the obtained data, a second experiment was carried out on a pulsating column in a direct-flow mode. For the leaching process in the pulsation column, the following parameters were selected: residual concentration – 100 g/l; contact time – 40 min; temperature – 45 °C; contact time – 40 min.

The acid enters the product level mirror in the pulsating column and is evenly distributed. As a result, the primary reaction proceeds over the entire surface, causing a slight foaming.

The product was dosed through a calibrated opening. The acid was administered continuously, its flow rate was regulated by an acid meter.

The level in the column was maintained automatically in the zone of intensive mixing, i.e. at the first distribution plate. The residence time of the product in the column can be controlled by the product flow rate.

RESULT AND DISCUSSION

The process of uranium leaching with nitric acid is described by the reaction:

$$(NH_4)_2U_2O_7 + 6HNO_3 = \\ = 2UO_2(NO_3)_2 + 2NH_4NO_3 + 3H_2O. \quad (1)$$

The process of leaching of the chemical concentrate, as a rule, is realized in cascades of reactors. The number of reactors is determined by the formula:

$$n = (Q \cdot \tau) \ / \ 24 \cdot V \cdot \eta, \quad (2)$$

where n – number of devices; Q – daily productivity; τ-duration of the process, hour; V – volume of the apparatus, m^3; η – is the filling factor of the apparatus.

The number of reactors in the cascade is no more than 4. This does not exclude the "breakthrough" of uranium-rich particles. The increase in the number of devices is economically unprofitable. Equipment for leaching must meet the following requirements:

– provide nitric acid access throughout the surface of the solid particles;

– provide acid flow to the particle surface;

– evenly distribute the acid on the surface of the plate;

– eliminate the possibility of formation of stagnant zones;

– ensure continuity and stability of the leaching process.

Reactor with a stirrer does not provide these requirements.

Dependence of the degree of leaching on process parameters is shown in Figs. 1–3.

From the data presented, it can be seen that an increase in the speed of the stirrer from 100 to 500 rpm significantly increases the degree of uranium recovery.

An increase in the reaction temperature from 50 to 95 °C sharply reduces the time of the process, but does not affect the degree of uranium extraction. In the solution, the leaching process lasts for 5…10 min. The degree of uranium recovery in the reactor is 92…94%. The residual concentration of nitric acid is 100…120 g/l.

As can be seen from the data in Tabl. 1, the main class of chemical concentrates is 0.25 mm, large particles break down into smaller fractions in which the residual uranium content is approximately the same.

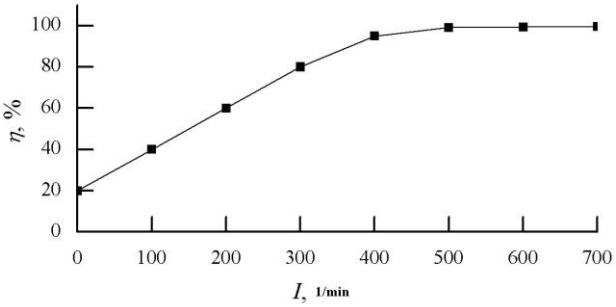

Fig. 1. Dependence of the degree of leaching on the intensity of mixing: τ = const, c = const, t = const

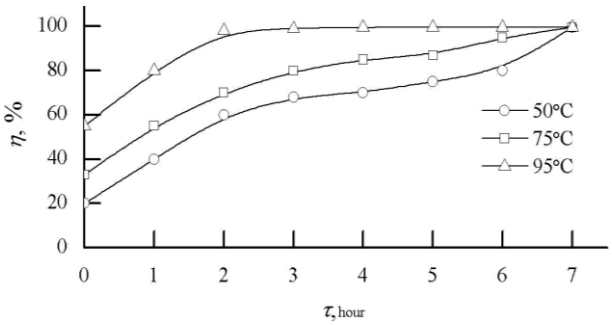

Fig. 2. Dependence of the degree of leaching on temperature and time, c = const, I = 200 rpm

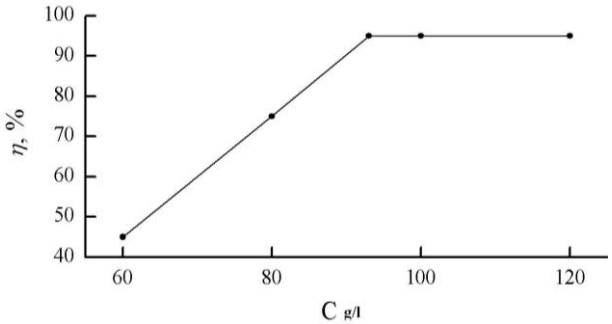

Fig. 3. Dependence of the degree of leaching on the concentration of acid

Table 1

Physico-chemical properties of uranium concentrate

Pulp until autopsy			Pulp after autopsy	
% sieving	% of uranium	Speed deposition m/hour	% sieving	% of uranium
0.1	71.6	360	–	–
0.2	73.4	100	1.1	2.9
1.3	63.5	50	6.3	3.25
98.4	68.0	0.3	92.6	4.2

To select the scheme of acid movement, you can focus on the main class, 0.25 mm. The deposition speed of smaller fractions is 0.3…0.5 m/h, therefore in the pulsating column it is possible to organize only a direct flow of the product and acid, and its specific surface will be optimal.

The reaction time (τ) is determined by the ratio of the volume of the apparatus (V) and the total volume of the product (Q_p) and acid (Q_a) per unit time. It is described by the equation:

$$\tau = V/(Q_p + Q_a). \quad (3)$$

The process of uranium leaching from the concentrate was carried out on the pulsating column of Fig. 4.

Comparative indexes of work of the pulsation column and the reactor are given in Tabl. 2.

Process shortcomings of the reactor:

– the labor input and intermittence of process does not allow to automate it and to increase efficiency;

– existence of a two-stage temperature schedule which leads to intensive gas emission and foaming that reduces efficiency of process;

– when the stirrer (mechanism) rotates, a laminar flow of liquid and solid particles is formed, which does not allow to create a motion of the solution around (relative to) a single particle;

– low extraction of uranium.

Speed of rotation of the mixing mechanism exceeds the speed of rotation of a stream of solution that increases thickness of a boundary layer. Local resistance at the same time increases therefore the gradient of strength of acid decreases and the small mass-transfer coefficient is created that does not allow to increase uranium extraction.

Table 2

Comparative indexes of work of the pulsation column and the reactor

Type of apparatus	Schedule of work	Volume of the device, m^3	Temperature, °C	Degree extractions of uranium, %
Column	Continuous	8	45.0	99.95
Reactor	Periodic	40	98.0	94.0

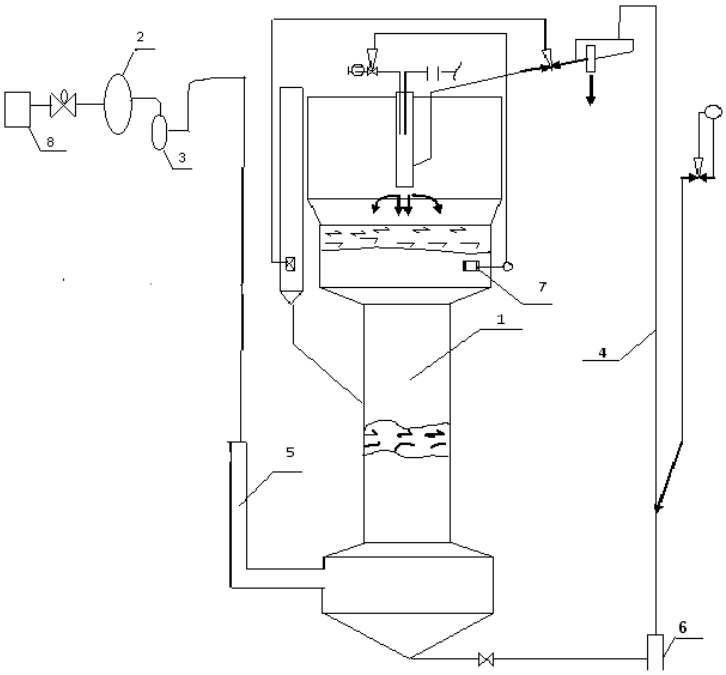

Fig. 4. Installation for a lixiviation of uranium from a concentrate: 1 – column; 2 – receiver; 3 – pulsation source; 4 – contour; 5 – the pulsation camera; 6 – level gage; 7– acidometer; 8 – compressor

During the stable work of the pulsation column the following indicators were reached:

– concentration of uranium in solution – 180…200 g/l, in a solid phase – 3%;

– residual concentration of hydrogen nitrate – 100 g/l;

– the residual maintenance of solid – 9 g/l;

– uranium extraction degree – 99.95%.

Concentration of uranium:

– solution of 180…200 g/l;

– in the solid phase – 3%;

– the residual concentration of nitric acid is 100 g/l.

Residual solid content – 9 g/l.

The degree of extraction of uranium is 99.95%.

It should be noted that the content of uranium in a solid phase after the pulsation column decreases from 3 to 0.05% that in the reactor cannot be reached.

CONCLUSIONS

Application of the pulsation column in the course of a lixiviation of uranium allowed to reduce device volume by 5 times.

Pulse column allowed to intensify the leaching process, fully automate it, reduce the loss of reagents and products, reduce the consumption of nitric acid and improve working conditions. A real reduction in capital and operating costs has been achieved.

The degree of uranium extraction in the continuous process on the pulsating column increased to 99.95%.

The temperature schedule 90 °C at a lixiviation is excluded. The temperature regime of 90 °C during leaching is eliminated, which prevents the release of nitrogen oxides and foaming.

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ИНТЕНСИФИКАЦИЯ ПРОЦЕССОВ ВЫЩЕЛАЧИВАНИЯ КОНЦЕНТРАТОВ УРАНА

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Изложены результаты экспериментов по выщелачиванию концентрата урана в азотной кислоте. Показано, что пульсирующая колонна позволяет интенсифицировать процесс выщелачивания урана, сократить его потери, расход азотной кислоты и снизить температуру процесса. Пульсационные колонны позволяют автоматизировать процесс.

ІНТЕНСИФІКАЦІЯ ПРОЦЕСІВ ВИЛУГОВУВАННЯ КОНЦЕНТРАТІВ УРАНУ

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Викладені результати експериментів з вилуговування концентрату урану в азотній кислоті. Показано, що пульсуюча колона дозволяє інтенсифікувати процес вилуговування урану, скоротити його втрати, витрати азотної кислоти і знизити температуру процесу. Пульсаційні колони дозволяють автоматизувати процес.