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Ultrasonic Intensification of Uranium Sorption from Pregnant Solutions by Ion-Exchange Resin

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Abstract: Until now, the intensification of ion exchange processes (sorption, desorption, washing of ion exchanger) remains an urgent problem in obtaining commercial strippants. This paper presents the study of ultrasonic (US) effects on the process of uranium sorption from pregnant solutions by ion-exchange resin at operating in-situ leach recovery ("ISR") uranium production. The study and evaluation of effectiveness of ultrasonic intensifying the ion exchange processes was implemented at one of the mines of NAC Kazatomprom JSC. Ultrasonic pulses periodically generated by emitters produced effects on the whole working space of the mass transfer apparatus. Thus, the whole mass of reagents is kept in continuous motion, and the whole surface of the anion exchanger grains is permanently purified during the ultrasonic device operation. The study findings showed that the ultrasonic intensification of the sorption process allows:

– increasing the sorption rate by 6.4 times at uranium concentration in the pregnant solutions of 0.003 g/m^3;
– increasing the sorption rate by 1.4 times at uranium concentration in the pregnant solutions of 0.014 g/m^3;
– achieving weighted average increasing the sorption rate by 1.3 times through applying the ultrasonic treatment;
– increasing full dynamic exchange capacity of the ion exchange resin for uranium in 1.13 times at keeping mechanical strength of the ion exchanger grains.

Keywords: uranium, ion exchange, intensification of mass transfer processes, ion exchanger, ion exchange resin, sorption, implosion, cavitation, diffusion, ultrasonic vibrations, pregnant solution.

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Интенсификация сорбции урана из продуктивных растворов на ионообменную смолу ультразвуком

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Аннотация: До сегодняшнего дня интенсификация процессов ионного обмена (сорбция, десорбция, отмывка ионита) была насущной проблемой при получении товарных десорбатов. В настоящей статье представлены материалы исследования влияния наложенного ультразвукового (УЗ) воздействия на процесс сорбции урана из продуктивных растворов на ионообменную смолу действующего рудника подземного скважинного выщелачивания (ПСВ) при производстве урана методом подземного скважинного выщелачивания. Для исследования и оценки эффективности интенсификации процессов ионного обмена наложением ультразвука в цехе переработки продуктивных растворов на одном из рудников АО «НАК «Казатомпром» были проведены работы по интенсификации процесса сорбции урана из продуктивных растворов ультразвуком. Периодически создаваемые УЗ-излучателями импульсы воздействуют на весь рабочий внутренний объем массообменного аппарата. Таким образом, вся масса реагентов находится в движении, и вся поверхность зерен анионита непрерывно очищается в процессе работы УЗ-оборудования. В результате проведенного исследования удалось установить, что УЗ-интенсификация процесса сорбции позволяет:

– при содержании урана в маточниках сорбции 0,003 г/м^3 увеличить скорость сорбции в 6,4 раза;

– при содержании урана в маточниках сорбции 0,014 г/м^3 увеличить скорость сорбции в 1,4 раза;
– за счет применения УЗ-воздействия получить средневзвешенное увеличение скорости сорбции в 1,3 раза;
– увеличить полную динамическую обменную емкость ионита по урану в 1,13 раза и при этом не уменьшить величину механической прочности зерен ионита.

Ключевые слова: ионный обмен, интенсификация процессов массообмена, ионит, ионообменная смола, сорбция, имплозия, кавитация, диффузия, УЗ-колебания, продуктивный раствор, уран.

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Definitions used in the paper

Diffusion - the process of mutual penetration of molecules or atoms of substances between the molecules or atoms of another substance, leading to spontaneous equalization of their concentrations throughout the whole occupied volume.

Intensification is the process and organization of development of production in which the most effective facilities are used, as well as expansion of the production and increasing its productivity.

Ionite is a solid insoluble substance capable of exchanging its ions for ions from the solution surrounding it. Ionites are commonly synthetic organic resins, having acidic or alkaline groups. Ionites are subdivided into cation exchangers (absorbing cations), anion exchangers (absorbing anions), and amphoteric ion exchangers with both of these properties.

Ion-exchange resin is a synthetic organic ion exchanger, a high molecular weight synthetic compound with three-dimensional gel and macroporous structure, which contains acidic or basic functional groups capable to ion-exchange reactions.

Cavitation is the process of formation, in a liquid, cavities (cavitation bubbles or caverns) filled with steam, resulting from external physical actions.

Sorption mother liquor is a pregnant solution after the sorption process.

Pregnant solution is a solution containing useful component.

Sorption is the process of absorption of various substances by a solid body (sorbent) from the environment, regardless of the absorption mechanism.

Ultrasonic transducer is a device that converts electrical oscillation created by high-frequency generator into vibrations.

Introduction

At present time uranium industry needs new technical solutions for increasing quality of products and reducing cost. One of such solutions may be the use of ultrasonic technologies in the production of uranium by the method of in-situ leach recovery (“ISR”).

The use of ultrasound technologies in various industries and the national economy has been known since the middle of the last century and has become widespread in the republics of the USSR. Relevance of ultrasound technologies has not decreased today – they are used in the oil industry, food industry, medicine, etc.

Ultrasonic technologies in uranium industry can be used in such processes as leaching, absorption, desorption, ion-exchange resin grain washing to remove impurities (phosphorus, iron, silicon, etc.), extraction, re-extraction and deposition. Intensification of ion-exchange processes by ultrasound is achieved due to decreasing thickness of the diffusion layer. Turbulent flows, implosion and cavitation, sound pressure, and some other second-order effects arising in a liquid under the action of ultrasonic vibrations change the character of the diffusion boundary layer immediately adjacent to the ion exchanger grain surface, the thickness of which limits the rate of ion-exchange processes, resulting finally in the intensification.

1. Effect of ultrasound on the process of uranium sorption

This paper describes the study of ultrasound effect on the process of uranium sorption on ion-exchange resin from pregnant solutions at the operating ISR mine.

One of the most effective methods of sorption process intensification is ion exchanger ultrasonic treatment. Second-order effects caused by ultrasound of specified frequency and intensity, first of all, implosion and cavitation, lead to more complete opening of micropores in ion-exchange resin grains and complete cleaning of their surfaces, which inevitably causes noticeable increasing the resin sorption capacity. Ultrasound not only acts on the ion-exchange resin surface layers, but also changes their capillary structure, and also increases the compensated molecular forces on the overall surface, including the surfaces of the walls of micro- and macro-capillaries.

Cavitation phenomena and the so-called ultrasonic wind, which causes intensive mixing of liquids in macro- and especially micro-volumes, leads to decrease in gradients in solutes concentrations on the boundary of solid and liquid phases, which should reduce the time of sorbent saturation with the extracted valuable components from the solutions (uranium in this case).

The rate of chemical reactions in heterogeneous systems depends on the rate of diffusion of molecules to the phase boundaries, chemical interaction, and the reaction product diffusion.

In most cases, the rate of chemical interaction is quite high compared with the diffusion rates; as a result, the rate of the entire reaction as a whole, at sufficiently developed phase contact surface is determined by the rate of diffusion of the reactants and reaction products.

The main resistance to the mass transfer process is offered by the diffusion layer located at the phases boundary, due to the fact that the process of mass transfer in it proceeds most slowly, by molecular diffusion way. The thickness and properties of the diffusion layer have strong effect on the rate of the heterogeneous processes. Turbulent flows caused by intensive stirring reduce effective thickness of the layer offering resistance to mass transfer. Increasing the stirring rate increases the rate of the entire heterogeneous process. When applying ultrasound, the process is significantly accelerated. Implosion and cavitation, acoustic pressure, sound wind cause intense turbulent flows not only in the entire mass of the treated solution, but also in close proximity to the phase boundary, resulting in significant reducing the boundary diffusion layer.

The studies on intensifying uranium sorption from pregnant solutions on ion-exchange resin under the action of ultrasound with vibration frequency of 22 kHz were conducted. The findings showed that decreasing the boundary diffusion layer thickness can accelerate the process several times. The equilibrium state under the ultrasonic treatment was reached 4–5 times faster than under identical sorption conditions without the ultrasound imposition. For testing integrity, all tests were carried out on two identical SNK-640 ion-exchange columns with the same amounts of pregnant solutions and ion-exchange resin; the solution was fed under control of electronic flowmeters. In this case, one column (with ultrasound) was for primary test, and another (without ultrasound), for control test.

Ultrasonic oscillation was generated in the tank with the ion exchanger and the pregnant solution using submerged metal ultrasonic vibrator. With this method of the solution exposure to ultrasound, the solution, in addition, was slightly heated. With increasing temperature, the time required for reaching equilibrium further decreased. Due to the temperature rise, the time required for reaching the equilibrium decreased by half on average, and, under exposure to ultrasound, by additional 3 times. The greatest acceleration of the ion-exchange process was achieved under the conditions of the resin and solution exposure to ultrasound at 40 °C.

2. Description of equipment used in the tests

The core equipment used in the tests was presented by two identical SNK-640 type ion-exchange units (columns) and the ultrasonic unit.

Each sorption column (Fig. 1) performs the function of a tank with resin through which pregnant solution goes. Technical features of the column are given in Table 1.

The ultrasonic unit (Fig. 2) is used as the main equipment causing intensification of the sorption process; its parameters are given in Table 2.

3. Experimental study of ultrasonic action on uranium sorption

The experimental study of ultrasonic action on uranium sorption process was carried out in two SNK-640 sorption columns under comparable conditions (Fig. 3).

In the experiments, ultrasonic action on uranium sorption rate and total dynamic exchange capacity of the ion exchanger in the process of uranium sorption from pregnant solutions at the operating ISR mine was investigated. The experiments were carried out in two sorption columns of the same type, one of which was equipped with an ultrasonic unit. Pregnant solution was fed to both columns simultaneously, at identical fed parameters. The ultrasonic unit position on the column is shown in Fig. 3. In the experiments, 0.297 m^3 of ion exchanger taken from the process and subjected to desorption, denitration, and tail washing were loaded into each column.

Uranium concentration in the initial ion exchanger was 2.68 kg/m^3.

The amount of pregnant solutions fed into the columns was regulated based on the readings of preliminarily calibrated electronic flowmeters.

Table 1

Technical characteristics of SNK-640 ion exchange column

Parameter	Value
Assembled column dimensions, mm	1160×1160×3700
Column diameter, mm	640
Sorbent volume, m^3	0.7
Sorbent layer height, m	2.2
Treated solution volume, m^3/h	0.2–5.0
Linear rate of solution flow, m/h	0.6–16.0
Unloaded sorbent volume, dm^3/h	75–225

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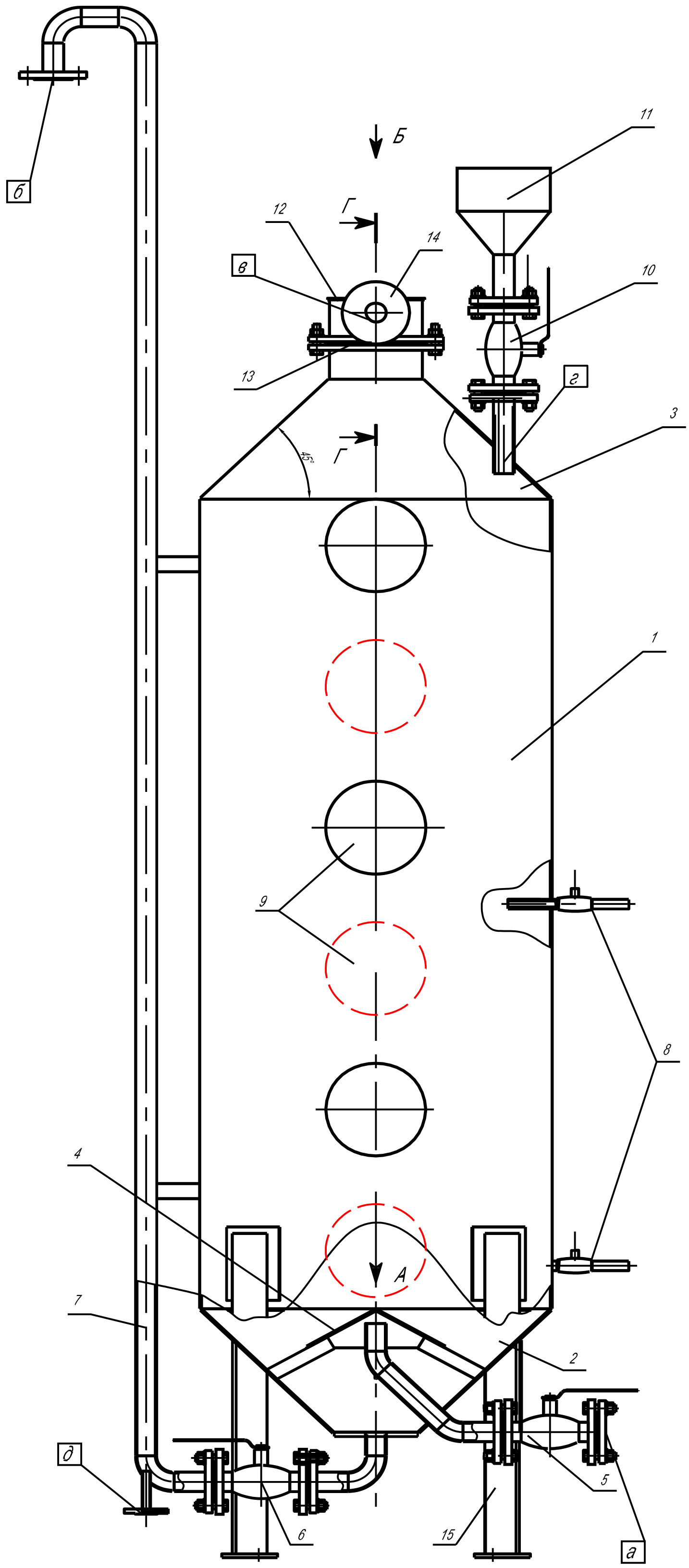

Fig. 1. SNK-640 ion exchange pressure column:
1 – body; *2* – lower cone; *3* – upper cone; *4* – solution divider; *5* – solution supply tap; *6* – saturated sorbent discharge tap; *7* – airlift; *8* – samplers; *9* – viewing windows; *10* – ion exchanger loading tap; *11* – ion exchanger receiving hopper; *12* – tip; *13* – drainage unit; *14* – sorption mother liquor outlet; *15* – support

Table 2

Parameters of the ultrasonic unit

Параметр	Value
Overall dimensions of the ultrasonic oscillatory system, mm	Ø215×750
Ultrasonic Intensity, W/cm^2	>10
Power cable length, m	25
ac power supply, voltage, V	220±22
Peak demand, VA	<3000
Power-control band, %	40–100
Continuous work time, hr	8
Electronic unit overall dimensions, mm	640×450×250

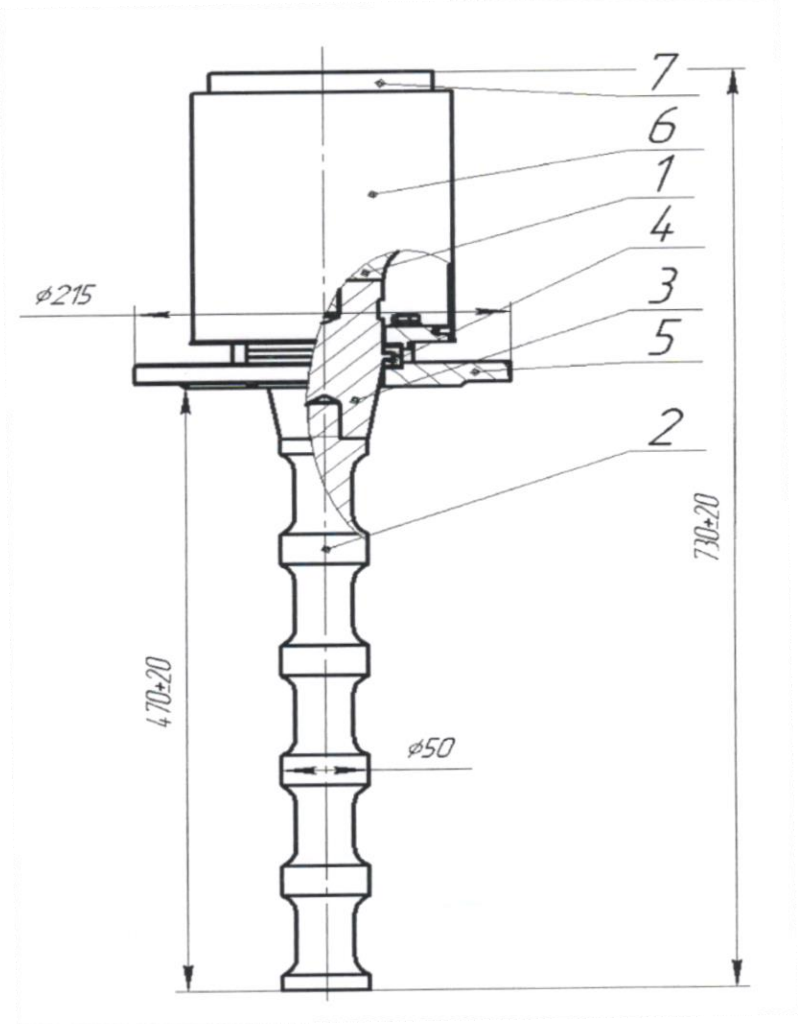

Fig. 2. Immersion ultrasonic oscillatory system (unit):
1 – ultrasonic oscillatory system (a set of piezocrystals); *2* – working emitting tool; *3* – booster link; *4* – tight seal; *5* – flange of the ultrasonic oscillatory system; *6* – body; *7* – fan

Fig. 3. Installation of the ultrasonic unit on the SNK-640 ion exchange column

Table 3

Test 1 Results

Column	Process time, min	Flowmeter readings m^3/h	Solution designation	Uranium concentration, kg/m^3
No US (control test)	120	2.4	Pregnant solution	0.048
			Sorption mother liquor	0.002
With US (primary test)	120	2.4	Pregnant solution	0.048
			Sorption mother liquor	0.001

At the initial stage of testing, the amount of pregnant solution (PS) fed into the columns was as follows:

–no US-unit – 2.4 m^3/h;

–with US-unit – 2.4 m^3/h.

The total sorption time at the initial stage was 2 hours. The experiment findings are given in Table 3.

The increased uranium concentration in the sorption mother liquor in the column with no ultrasonic treatment as compared with the sorption mother liquor in the column with the ultrasonic treatment in the first 2 hours indicates very large amount of pregnant solution fed for sorption. For instance, the volumetric ratio of the PS to the ion exchanger ($V_{ps}/V_{i\text{-}e}$) was 8.08 per hour.

The next experiment was carried out at decreased volume of PS feeding into the column with no ultrasonic treatment, equal to 5.86 $V_{ps}/V_{i\text{-}e}$ per hour. The testing results are given in Table 4.

Table 4

Test 2 Results

Test	Process time, min	Flowmeter readings m^3/h	Solution designation	Uranium concentration, kg/m^3
No US (control test)	60	1.74	Pregnant solution	0.050
			Sorption mother liquor	0.001
With US (primary test)	60	2.40	Pregnant solution	0.050
			Sorption mother liquor	0.001

Table 5

The results of the experiment on determining full dynamic exchange capacity at uranium sorption from pregnant solutions

Column without ultrasound (control test)						Column with ultrasound (primary test)					
$V_{ps}/V_{i\text{-}e}$	Uranium concentration in pregnant solution, g/l	$V_{ps}/V_{i\text{-}e}$	Uranium concentration in pregnant solution, g/l	$V_{ps}/V_{i\text{-}e}$	Uranium concentration in pregnant solution, g/l	$V_{ps}/V_{i\text{-}e}$	Uranium concentration in pregnant solution, g/l	$V_{ps}/V_{i\text{-}e}$	Uranium concentration in pregnant solution, g/l	$V_{ps}/V_{i\text{-}e}$	Uranium concentration in pregnant solution, g/l
5.860	0.050	5.860	0.050	5.860	0.050	5.860	0.050	5.860	0.050	5.860	0.050
8.260	0.048	8.260	0.048	8.260	0.048	8.260	0.048	8.260	0.048	8.260	0.048
25.400	0.048	25.400	0.048	25.400	0.048	25.400	0.048	25.400	0.048	25.400	0.048
34.180	0.049	34.180	0.049	34.180	0.049	34.180	0.049	34.180	0.049	34.180	0.049
51.180	0.049	51.180	0.049	51.180	0.049	51.180	0.049	51.180	0.049	51.180	0.049
67.950	0.048	67.950	0.048	67.950	0.048	67.950	0.048	67.950	0.048	67.950	0.048
84.810	0.049	84.810	0.049	84.810	0.049	84.810	0.049	84.810	0.049	84.810	0.049
101.48	0.049	101.48	0.049	101.48	0.049	101.48	0.049	101.48	0.049	101.48	0.049
118.69	0.049	118.69	0.049	118.69	0.049	118.69	0.049	118.69	0.049	118.69	0.049
135.52	0.049	135.52	0.049	135.52	0.049	135.52	0.049	135.52	0.049	135.52	0.049
152.39	0.049	152.39	0.049	152.39	0.049	152.39	0.049	152.39	0.049	152.39	0.049
169.39	0.051	169.39	0.051	169.39	0.051	169.39	0.051	169.39	0.051	169.39	0.051
186.33	0.050	186.33	0.050	186.33	0.050	186.33	0.050	186.33	0.050	186.33	0.050
203.13	0.049	203.13	0.049	203.13	0.049	203.13	0.049	203.13	0.049	203.13	0.049
219.93	0.048	219.93	0.048	219.93	0.048	219.93	0.048	219.93	0.048	219.93	0.048
236.70	0.048	236.70	0.048	236.70	0.048	236.70	0.048	236.70	0.048	236.70	0.048
253.40	0.048	253.40	0.048	253.40	0.048	253.40	0.048	253.40	0.048	253.40	0.048
270.40	0.049	270.40	0.049	270.40	0.049	270.40	0.049	270.40	0.049	270.40	0.049
287.74	0.050	287.74	0.050	287.74	0.050	287.74	0.050	287.74	0.050	287.74	0.050
304.41	0.050	304.41	0.050	304.41	0.050	304.41	0.050	304.41	0.050	304.41	0.050
321.01	0.050	321.01	0.050	321.01	0.050	321.01	0.050	321.01	0.050	321.01	0.050
337.71	0.049	337.71	0.049	337.71	0.049	337.71	0.049	337.71	0.049	337.71	0.049
354.38	0.050	354.38	0.050	354.38	0.050	354.38	0.050	354.38	0.050	354.38	0.050
371.75	0.049	371.75	0.049	371.75	0.049	371.75	0.049	371.75	0.049	371.75	0.049
382.32	0.050	382.32	0.050	382.32	0.050	382.32	0.050	382.32	0.050	382.32	0.050

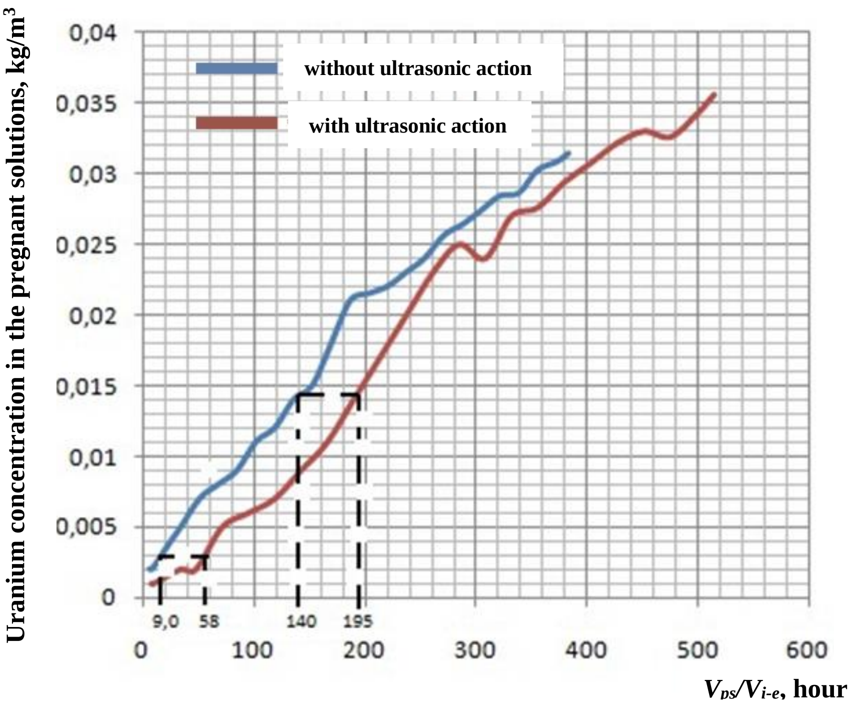

Fig. 4. Graphs of uranium sorption by the ion exchanger with and without ultrasonic action

The experiment findings indicate the optimal volumetric ratio of the PS to the ion exchanger to be 5.86 $V_{ps}/V_{i\text{-}e}$ per hour for sorption process with no ultrasonic treatment.

The sorption rates measured in the first experiment in the column with and without ultrasonic treatment were taken as the basis for the next experiment on measuring total dynamic exchange capacity (TDEC) of the resin with and without ultrasonic treatment. The results of the experiment on determining full dynamic exchange capacity of the anion exchanger in the process of uranium sorption from pregnant solutions with and without ultrasonic treatment are given in Table 5.

Graphs of uranium sorption with and without ultrasonic treatment under dynamic conditions of the experiments are presented in Fig. 4.

The tests for investigation on intensifying uranium sorption from pregnant solutions on the anion-exchange resin by ultrasonic treatment showed that the ultrasonic treatment results in:

–increasing the sorption rate by 6.4 times at uranium concentration in the pregnant solutions of 0.003 g/m^3;

–increasing the sorption rate by 1.4 times at uranium concentration in the pregnant solutions of 0.014 g/m^3;

–achieving weighted average increasing the sorption rate by 1.3 times through applying the ultrasonic treatment;

–increasing the anion exchanger TDEC for uranium by 1.13 times.

The total ultrasonic treatment time in the experiments was 63.3 hours.

The experiment findings indicated the optimal volumetric ratio of the PS to the anion exchanger to be 5.86 $V_{ps}/V_{i\text{-}e}$ per hour for sorption process with no ultrasonic treatment.

4. Tests for investigation the effect of ultrasonic treatment on the anion exchanger mechanical strength

Tests of Ambersep 920U SO_4 anion-exchange resin (used in all the experiments) were carried out in polymer tank having volume of 60 l, into which 40 l of the ion-exchange resin were loaded. The passport specifications of the anion-exchange resin are given in Table 6.

Table 6

Characteristics of the strongly basic macroporous anion exchange resin of Ambersep 920U SO_4 grade manufactured by Dow Chemical Company

Indicator	Value
Polymer matrix	Macroporous Crosslinked Polystyrene
Physical form	Non-transparent granules
Ionic form when delivered	Sulfate (not less than 50%)
Functional group	$–N+(CH_3)_2CH_2CH_2OH$
Full ion-exchange capacity	≥ 1.0 g-eq/l (Cl^--form)
Moisture Content	53–65 % (Cl^--form)
Commodity weight	680–710 g/l
Harmonic mean diameter	0.845–1.050 mm
Uniformity factor	≤1.50
Content of small granules <0.710 mm	Max 5.0 %
Content of large granules >1.180 mm	Max 4.0 %
Maximum reversible swelling $Cl^- \rightarrow SO_4^{2-}$	about 5 %
Mechanical strength	≥98 %
Manufacturer	Rohm and Haas France S.A.S. (Dow Chemical Company)
Country	France

The ultrasonic unit and the hose for supplying flowing technical water were installed on the upper part of the tank (lid). The working body (ultrasonic transducer) was placed coaxially inside the cylindrical tank. Dimensions of the working body of the transducer were as follows: length, 480 mm; diameter, 50 mm. The tank height and diameter were 640 mm and 360 mm, respectively. The anion exchanger was located between the transducer working body and the tank walls. The distance from the containing walls to the transducer working body was 335 mm.

This treatment mode is hard, since the ultrasonic wave in the heterogeneous medium (solution + anion exchanger) forms reflected wave due to small distance between the transducer and the tank wall. The reflected waves form zones of multiple increasing ultrasonic action on the anion exchanger. Notice that this effect will not take place in commercial-scale sorption columns (and other flow-type ion-exchange units). The zone of maximum ultrasonic action for this type of transducers extends 550–600 mm from the surface of the transducer. The volumetric flow rate of water was 20 l/h. The ultrasonic transducer operating mode was set as follows: 6 hours of ultrasonic treatment and 25 minutes with no ultrasonic action. Total operating time of the ultrasonic transducer in the experiment was 148 hours at frequency of 22 kHz. General arrangement of the ultrasonic assembly installation is shown in Fig. 5.

Fig. 5. Installation for the study of hard ultrasound on the ion exchanger grain mechanical strength

For the experiment, samples of the fresh (new) resin were taken and labeled No. 3 and No. 4, respectively (Fig. 6). After the experiment, resin samples that underwent ultrasonic treatment were taken and labeled No. 1 and No. 2. Figure 6 shows that samples No. 3 and No. 4 present new resin, and samples No. 1 and No. 2 are of darker color with gray tint, indicating their previous use.

All the samples were sent to an independent specialized laboratory for control tests.

The results of the samples testing for determining mechanical strength of the ion exchanger grains are as follows:

– sample No. 1: mechanical strength – 99.0%;

– sample No. 2: mechanical strength – 99.5%;

– sample No. 3: mechanical strength – 98.0%;

– sample No. 4: mechanical strength – 98.0%.

Fig. 6. Samples of the initial new ion exchanger (Nos. 3 and 4) in comparison with the samples of the previously used ion exchanger (Nos. 1 and 2)

As can be seen from the obtained data, ultrasonic treatment of the ion-exchange resin grains even under specially produced hard conditions not only did not decrease their mechanical strength, but, on contrary, caused their insignificant, but marked strengthening. This strengthening was due to complete ultrasonic cleaning of macroporous structure of the resin grains, as well as restoration effect of the so-called ultrasonic crosslinking of polystyrene matrix filaments damaged in the course of the ion-exchange resin use in the ISR uranium production process.

Conclusion

The performed tests for investigation intensifying uranium sorption from pregnant solutions on the anion-exchange resin by ultrasonic treatment showed marked potential of applying the ultrasonic treatment for intensification of other ion-exchange processes in the ISR uranium production process. It was also experimentally established that the ultrasonic treatment doesn't produce negative effect on mechanical strength of the ion-exchange resin grains. A regeneration effect was observed.

The experiments described in this paper cover only the process of uranium sorption from pregnant solutions. However, it can be expected that the ultrasonic treatment is promising for other ion-exchange processes used in the production of uranium by the ISR method.

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