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Investigation of Sorption Extraction of Uranium from Productive Solutions Obtained by Leaching Black Shale Ore



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ABSTRACT

This article presents the results of a study of sorption extraction of uranium from productive solutions obtained by leaching carbon-silicon ore (black shale ores). By optimizing the technological regimes, it was found that the sorption of uranium on highly basic sorbents increases with increasing pH of the solution (from 1.2 to 1.8). Thus, anionite SBA-800 has a better dynamic capacity for uranium compared to Ambersep 920, while uranium is sorbed 21 % more, which indicates its best efficiency. From the obtained data on the results of nitrate desorption of uranium from the anionite Ambersep 920, the extraction rate of uranium into desorbate is more than 83 % of the total amount of uranium, and the extraction rate of uranium from the anionite SBA-800 is 43 %.

INTRODUCTION

From the literature it is known that the ionic state of uranium in sulfuric acid solutions is presented in the form of complex compounds: UO_2^{2+} , $[UO_2(SO_4)_2]^{2-}$, $[UO_2(SO_4)_3]^{4-}$ and in solution, there is a mobile equilibrium [1]. As is known, in the sorption of uranium, both cation exchangers and anion exchangers can be used depending on the pH of the medium and the ionic state. Cation-exchange sorbents can extract uranyl cations, and anion-exchange sorbents can extract the anionic complex of uranium as $[UO_2(SO_4)_3]^{4-}$. As the uranyl or anion complexions are removed from the productive solution to the ion exchanger, an equilibrium shift in the solution will be observed in the direction of the extracted ion [1].

Currently, a large number of methods have been developed for the processing of uranium-containing productive solutions. Extraction of uranium from sulfate productive solutions is possible by the extraction method [2,3], as well as using biosorption [4]. However, in production, the processing of uranium-containing productive solutions using these methods is not economically feasible. In addition, there are technologies that use chelated resins in uranium technology to extract uranium-containing compounds from sulfate solutions [5].

Uranium is extracted by several methods, such as sorption [6], and solvent extraction [7]. However, methods such as precipitation and solvent extraction are expensive and inefficient for extracting molybdenum from pregnant leach solutions. Thus, in this regard, any other alternative method capable of selectively removing uranium from an aqueous solution will be of great importance. One of such methods is sorption, which is widely used because of the ease of operation, simplicity and the limited application of solvents [8]. Due to the several advantages of ion exchange processes, which include low cost, high selectivity and high efficiency [9], it becomes a suitable method for uranium extracting. Ion exchange processes also does not have issues of phase separation, third phase formation, or solvent loss, and is particularly advantageous for adsorption of metals present in low concentrations. These benefits of sorption processes therefore justify research into the technology to further develop its potential. Ion exchange resins have been developed as a major option for molybdenum sorption over the past few decades [10,11]. Selective resins allow to extract of molybdenum from pregnant leach solutions in presents of impurities.

Strongly basic ion exchangers are successfully used for the processing of sulfate productive uranium-containing solutions of borehole and underground leaching in China [12]. It is also effective to extract uranium from sulfate productive solutions on strongly basic anion exchangers [13]. Therefore, based on published data, in order to determine the possibility of sorption extraction of uranium from complex ammonium sulfate solutions, in this work, we studied sorption and desorption processes on highly basic *anion exchangers Ambersep 920, SBA-80 and weakly basic anion exchangers Purolite-A100 and WBA-301*.

MATERIALS AND METHODS

The object of the study in this study are productive solutions for the processing of polymetallic black shale ores after leaching. The composition of these solutions is as follows, g / dm³: Fe - 8.2; Al - 0.968; V - 2.89; Mo - 0.073; U is 0.062; Σ TRY - 0.064; H₂SO₄ - 240. According to the results of chemical analysis and physicochemical analyzes, it was concluded that the productive solution consists of sulfate, ammonium and hydrosulfate groups.

From the data on the composition of productive solutions, it can be said that the solutions have a complex composition and contain a significant amount of uranium, molybdenum, vanadium, and rare-earth elements in the form of sulfate, ammonium, and hydrosulfate compounds and their complexes, the complex extraction of which will reduce the cost of manufactured commercial products.

To study the sorption concentration of uranium from productive solutions under static conditions, we used high-basic anion exchangers Ambersep 920UXL and SBA-80, as well as weakly basic anion exchangers Purolite-A100 and WBA-301. For this purpose, ion-exchange resin and a productive solution were mixed in conical flasks on an orbital shaker at a ratio of 10 g of resin per 100 ml of productive solution at different pH values. Before determining the sorption properties, all the ion exchangers were preliminarily converted to the sulfate form using a sulfuric acid solution.

The study of the uranium desorption process from saturated anionites after sorption was carried out according to the following procedure. The experiments were carried out in a static mode, bringing into contact with constant stirring for 2 hours a saturated anion exchange resin with solutions of sodium hydroxide, ammonium nitrate, and soda with a concentration in the range

from 1.0 to 4.0 M. The ratio of solid and liquid phases was 1 : 10. After 30, 60, 90 and 120 minutes from the start of the experiment, a sample of the solution was taken from the reactor for analysis. The uranium content was determined in the selected sample. At the end of the experiment, the anion exchangers were separated from the desorbate, and after the resulting solutions were analyzed for uranium content. The objective of this process is to select the optimal conditions for the selective and maximum extraction of uranium from anion exchangers saturated after sorption after desorption.

RESULTS AND DISCUSSION

In the conducted studies, the influence of the acidity of the medium and the duration of the process on the extraction of uranium into the resin, weakly basic anion exchange resin of the brand WBA-301, was studied. The results of the study are presented in Figure 1.

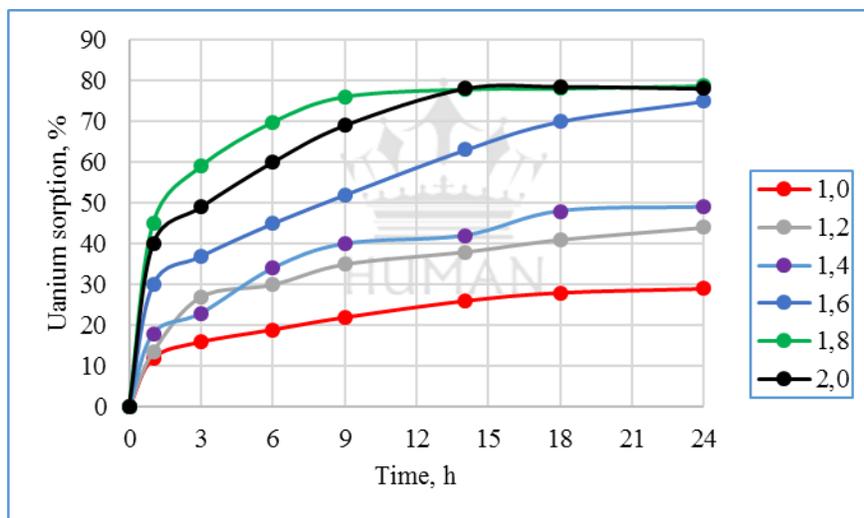


Figure No. 1: Dependence of uranium sorption on time (WBA-301) at various pH

As can be seen from the figure (Figure 1), the degree of uranium extraction is significantly affected by the acidity of the solution. This figure shows the dependence of the change in the degree of sorption of uranium on pH. The experimental results were evaluated by uranium extraction equal to the ratio of the masses of the metal in the ion exchanger and the initial solution. The influence of the pH of the medium on the sorption capacity of the ion exchanger was determined by the difference in the concentration of uranium in anion exchangers in the pH range from 1.0 to 2.0. As the results show, the best results were obtained at pH 1.8 and 2.0.

Next, a study was made of the dependence of uranium sorption on time on weakly basic Purolite-A100 anion exchange resin at various pH values (Figure 2).

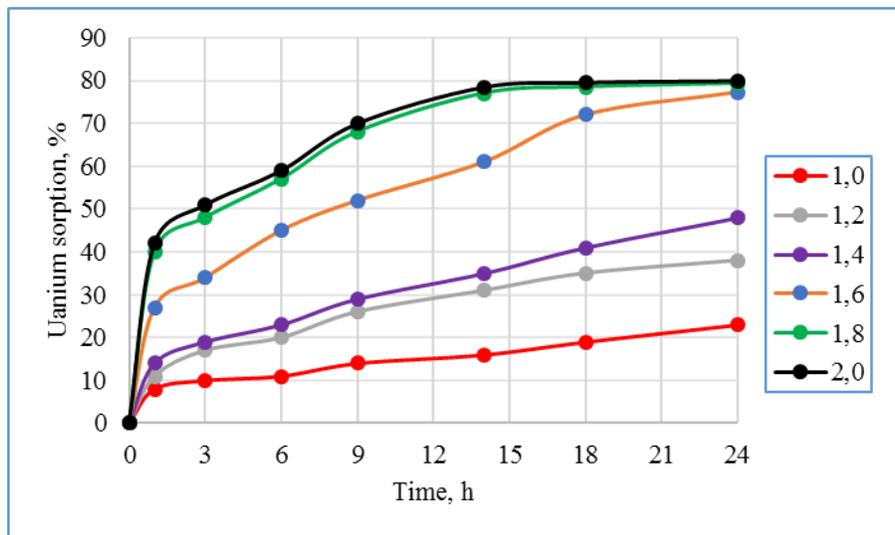


Figure No. 2: Dependence of uranium sorption on time (Purolite-A100) at various pH

From Figure 2 it follows that the sorption of uranium by Purolite A-100 brand anion exchange resin is of a similar nature with the process of sorption of uranium on WBA-301 resin. With an increase in pH in the solution from 1.0 to 2.0, the degree of uranium extraction reaches 80 % with a process duration of 24 hours.

It was also found that the sorption capacity of weakly basic anion exchangers WBA-301 and Purolite-A100 does not exceed 80 %. In this regard, in order to optimize the sorption extraction process and to search for an effective sorbent for uranium extraction, the sorption abilities of strongly basic anion exchangers were studied. A study of the process of sorption of uranium by strongly basic anionites was also carried out in a static mode in accordance with the procedure described in Section 1.1.1. The results of a study of the dependence of uranium sorption on time on SBA-800 anion exchange resin at various pH values are presented in Figure 3.

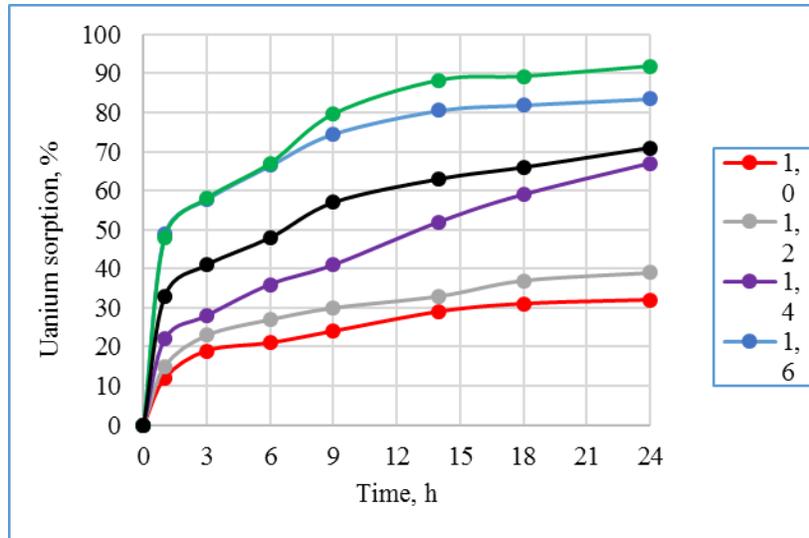


Figure No. 3: Dependence of uranium sorption on time (SBA-800) at various pH

According to the data obtained, the extraction of uranium using the strongly basic anion exchanger SBA-800 is more effective than the weakly basic ion exchangers Purolite-A100 and WBA-301). Moreover, the degree of uranium extraction reaches 92 % against 80 % on the weakly basic anion exchanger Purolite A-100. The maximum results for the sorption of uranium on this anion exchange resin were obtained at a pH of 1.8.

Uranium sorption was also studied on the strongly basic anionite Ambersep 920. A graph of the dependence of uranium sorption on time (Ambersep 920) at various pH values is presented in Figure 4.

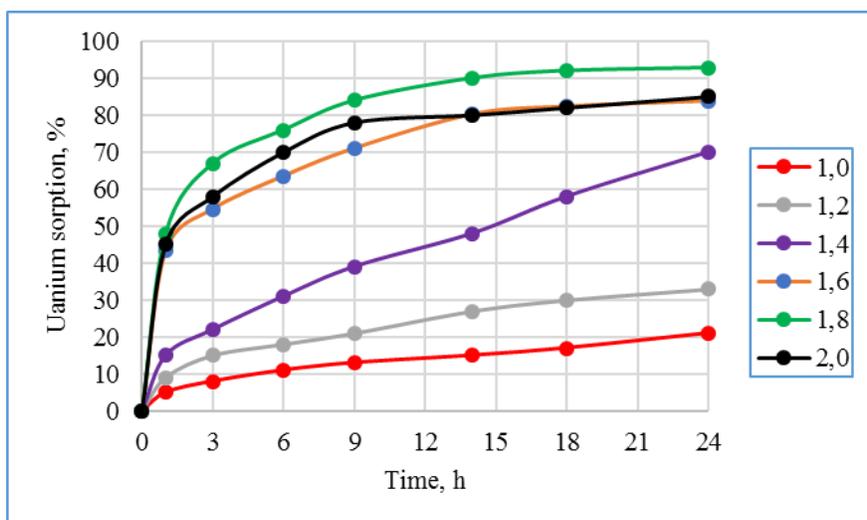


Figure No. 4: Dependence of uranium sorption on time (Ambersep 920) at various pH

It can be seen from the obtained results that the sorption of uranium on highly basic sorbents increases with increasing solution pH and on high-basic anion exchangers Ambersep 920 and SBA-80 reaches 92 %, which is 12 % more than on weakly basic anion exchangers Purolite-A100 and WBA-301.

Studies have shown that the sorption capacity of anion exchangers increases in the following order: WBA-301 > Purolite-A100 > Ambersep 920 > SBA-80. A comparison of the capacitances of the various studied ion-exchange resins shows that the strongly basic SBA-80 and Ambersep 920UXL anion exchangers have the largest capacitance.

The selected strongly basic anion exchangers were subjected to chemical analysis in order to determine the sorption capacity. The result of chemical analysis of saturated strongly basic sorbents is presented in table 1.

Table No. 1: The chemical composition of saturated ion exchangers

Ionite code	Content in air-dry ion exchanger, g / kg					
	U	SO ₄ ²⁻	Cl	Fe	Al	SiO ₂
Ambersep 920	18,6	106,0	6,1	0,92	0,017	3,9
SBA-800	21,7	114	10,6	1,01	0,024	4,6

Then saturated ion exchangers are sent to the desorption.

Study of the process of desorption of uranium

The essence of the process of uranium desorption is the use of solutions that provide direct separation of uranium from anion exchangers in the volume of the solution.

At present, in many active uranium enterprises, nitrate, chloride, and carbonate ions are most often used as a stripping solution, as well as sulfate ions, most rarely [14]. Uranium desorption occurs due to the ion exchange reaction of the desorption solution and uranyl sulfate complexes, which are associated with functional groups in the phase of the saturated sorbent ion exchanger. As the authors of [15] note, for the desorption of uranium from ion exchangers, it is effective to use a mixture of solutions of ammonium nitrate and nitric acid. The addition of nitric acid lowers

the pH of the resulting commodity desorbate and prevents the precipitation of uranium. Also, according to the authors, solutions of sodium hydroxide and carbonate can be successfully used for the desorption of uranium from strongly basic ion exchangers.

Results and discussion of desorption process

The results of the studies showed that upon desorption of uranium from the phase of saturated ion exchangers, the degree of uranium extraction increases with increasing concentration of sodium hydroxide in solution (Figure 5). In this case, for Ambersep 920 anion exchange resin, the effect of the concentration of sodium hydroxide on the degree of uranium extraction is enhanced when the NaOH concentration in the solution is more than 1.5 M. A similar situation is observed when using a solution of ammonium nitrate. The degree of uranium extraction from the saturated anionite phase increases with increasing NH_4NO_3 concentration in the range from 1 to 3.5 M (Figure 6). When using desorption solutions with a higher concentration than 3.5 M, a further increase in the degree of desorption of uranium does not occur. Moreover, the high cost of reagents significantly increase the cost of the process and make it less technological. Therefore, from the point of view of the profitability of the process, the optimal concentration of the stripping solution is a concentration value of 3.5 M.

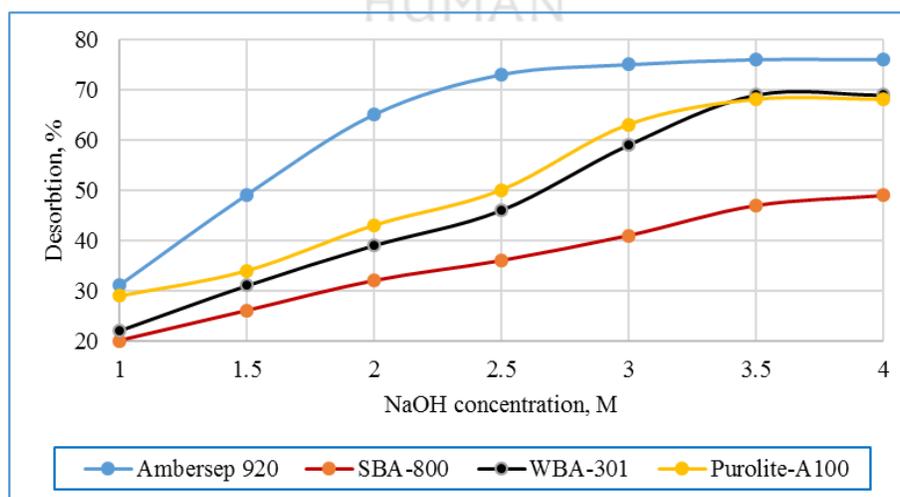


Figure No. 5: Results of the uranium desorption process with sodium hydroxide solution

When uranium is desorbed from the saturated anionite phase, the degree of uranium extraction increases with increasing concentration of the desorbing solution. According to the results of the

analyzes, the best results on the desorption of uranium were obtained using sodium hydroxide. The optimal concentration of the stripping solution is a concentration value of 3.5 M, at which the extraction of uranium reaches a maximum within 120 minutes (Figure 6).

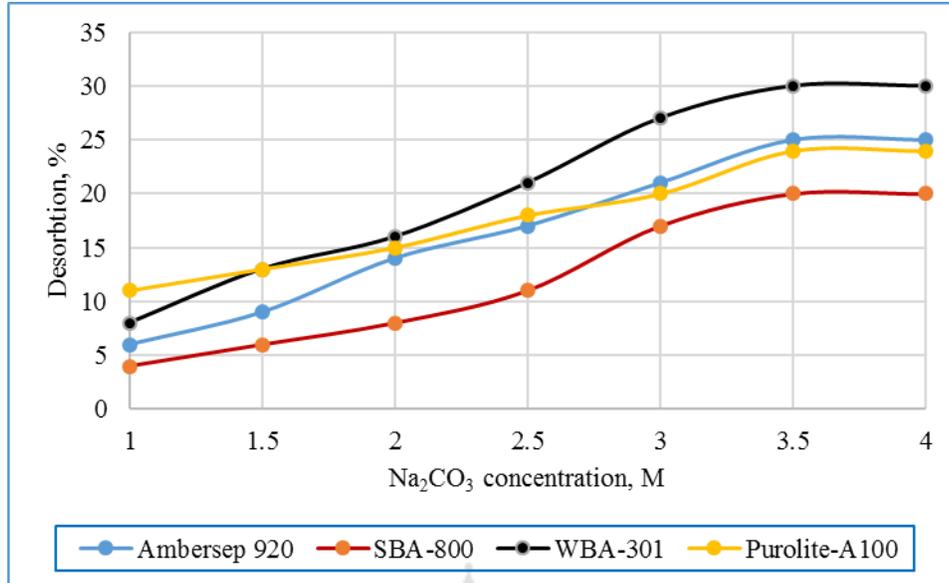


Figure No. 6: Results of the uranium desorption process with a solution of ammonium nitrate

In the process of desorption of uranium with a soda solution, the maximum degree of extraction is achieved using a 3.5 M concentration of Na₂CO₃ solution and is about 30 %. A further increase in the concentration of the stripping solution affects the extraction of uranium in the solution slightly, but significantly reduces the technological parameters of the process. Thus, it can be concluded that the use of sodium carbonate solution as a stripping solution is not of industrial interest (Figure 7).

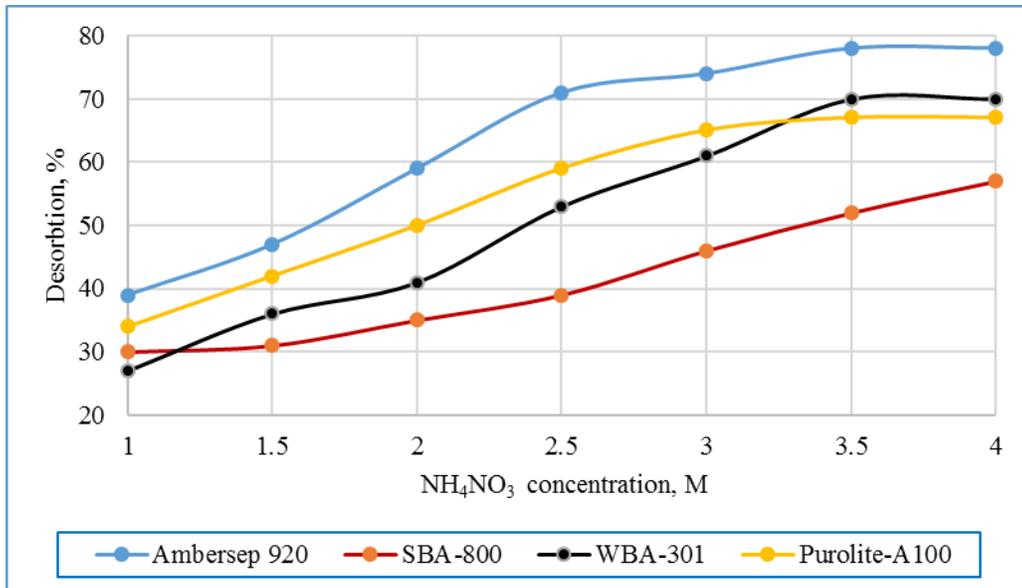


Figure No. 7: Results of the uranium desorption process of with a solution of sodium carbonate

Further studies of the nitrate desorption of uranium from Ambersep 920 ion exchanger showed that the degree of uranium extraction into the eluate is more than 83 %, and the degree of uranium extraction from SBA-80 anion exchanger is only 58 %, which indicates that the use of Ambersep 920 anion exchanger at the stage of sorption extraction uranium is the most optimal. The composition of commodity regenerates in desorbate is presented in table 2.

Table No. 2: Composition of commodity regenerates

ion exchanger	Desorbate content, g / l								
	U	Fe	Al	Si	Ca	Mg	SO ₄ ²⁻	NO ³⁻	Cl
Ambersep 920UXL	16,6	0,12	<0,01	0,002	0,102	HeT	88,0	44,2	5,2
SBA-800	9,5	0,10	0,02	0,001	0,046	0,026	81,1	71,6	5,0

Thus, the best results were obtained in the desorption of uranium by sodium hydroxide and ammonium nitrate. However, given the significant cost of sodium hydroxide and a small difference in the degree of desorption, in practice, it is advisable to use a 3.5 M solution of ammonium nitrate.

CONCLUSION

1. It was found that the sorption capacity of anion exchangers increases in the following order: WBA-301 > Purolite-A100 > Ambersep 920UXL > SBA-800.
2. Based on the analysis results, the best results on the sorption of uranium were obtained on strongly basic resins SBA-80 and Ambersep 920UXL. In addition, it was found that the optimal pH of the solution for sorption of uranium is in the range of 1.6 - 1.8;
3. The process of uranium desorption was carried out in a static mode, bringing into contact with constant stirring for 2 hours a saturated ion exchanger with solutions of sodium hydroxide, ammonium nitrate, and soda at a concentration of 1.0 to 4.0 M. The ratio of solid and liquid phases was 1: 10. Ammonium nitrate was chosen as the optimal stripping solution. The optimal stripping solution is a solution with a concentration of 3.5 M.

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