

URANIUM REMOVAL FROM CONTAMINATED WATER BY ION EXCHANGE RESINS

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ABSTRACT

In the nuclear fuel cycle, uranium containing liquid wastes can be produced in large quantities. There are several possible operations to separate this element from an aqueous solution. The classic examples are separation by solvent extraction and by precipitation when uranium is present in high concentrated solutions. In those cases where the element is present in concentrations as low as 100-200 $\mu\text{g mL}^{-1}$, ion exchange technique can be very helpful to bring the uranium concentration to less than 10-20 $\mu\text{g mL}^{-1}$. The synthetic resins used in ion exchange processes effectively remove metals from contaminated liquid wastes. This work intends to remove uranium from aqueous solutions by ion exchange technique. A column (10 cm in height and 1 cm in diameter) was used with 30 ml of dry Dowex 1-X8 strong anionic resin to fix and remove uranium from the solution. The solution with 155 mg.L^{-1} of uranium was percolated through the column with a flow of 4.0 mL.min^{-1} . Resin was conditioned at $\text{pH} = 7$. The determination of the uranium contents was performed by Optical Emission Spectrometry with Argon Plasma Source (ICP-OES). The main operational parameters, such as pH, flow and column height, were evaluated to determine the best operating condition of the system. The results indicated approximately 99.9% removal of uranium from the application of the ion exchange technique. The percentage of removal allows the effluent to be properly disposed of in a sewage collection system in accordance with the regulations in force in the country.

1. INTRODUCTION

Uranium is a natural chemical element found in low concentrations in rocks or sediments of soils. In the natural environment its average concentration is between 2 and 4 $\mu\text{g mL}^{-1}$. However, it can be found in higher concentrations due to the anthropogenic enrichment activities of uranium in nuclear installations, through the production of radioactive effluents [1].

This element is hazardous because of its intrinsic chemical and radioactive toxicity and the risks associated with materials containing this element. Risks are usually related to occupational activities and environmental contamination for the general public [2]. There are several techniques that can remove uranium from contaminated water. The classical examples are the separation by ion exchange, solvent and precipitation [3].

Whether on a large scale or in bench quantities, these operations are typically applied during the stages of the nuclear fuel cycle, the recovery of by-products, the analytical chemistry of uranium; of the actinides and also in the treatment of tailings of processes that have as intention the decrease of the pressure of the load with respect to the uranium content or the destination in sewer collection system [3].

Synthetic ion exchange resins demonstrate efficiency in this subject and tests on columns with strong anionic resins have been most effective in removing uranium with pH between 5 and 7 [2]. Ion exchange resins have been studied in uranium recovery from industrial effluents found in nuclear power plants. The ion exchange technique allows a recovery of more than 98% of contaminated water uranium [4].

During the period of operation of a pilot uranium purification plant at IPEN, several tons of pure nuclear uranium processed from Sodium Diuranate, DUS, or Ammonium Diuranate, DUA, were produced for subsequent UF₆ transformation. It produced hundreds of liters of uranyl nitrate effluent, uranium concentration of about 100 gL⁻¹ which were subsequently precipitated with ammonia, NH₃, and from this precipitation an effluent with low uranium concentrations, about 150 µg mL⁻¹.

The objective of this work is to separate uranium in trace concentrations present in aqueous solutions. To evaluate the suitability of nanostructured materials and / or ion exchangers in wastewater treatment for uranium recovery in order to comply with environmental legislation and regulatory actions.

1.1 Ion exchange

Ion exchange is a chromatographic technique for the separation of compounds based on the interaction between a mobile phase and a stationary phase. The affinities may be by adsorption or partitioning processes, this involves the relative solubility of a compound between the two phases, that involves the binding of a compound to the surface of a solid phase [5].

The ion exchange process can be carried out in columns containing a resin bed, continuously, or in batch mode, semi-continuous in a resin system in pulp. In general, the ion exchange mechanism occurs as follows: metal ions present in the solution, mobile phase, can displace the resin ions, fixed phase, taking its place, and that is carried by the mobile phase [6].

2. MATERIALS AND METHODS

2.1. Reagents

- Strong anionic resin Dowex 1-X8, 20-50 mesh, Dow Chemical;
- Hydrochloric acid, HCl, 37%, Merck Millipore;
- Sodium hydroxide, NaOH, Merck Millipore;
- Certified buffer solution pH 7, Merck Millipore;
- Certified buffer solution pH 4, Merck Millipore;
- Ethanol 96%, C₂H₅OH, Merck Millipore;

2.2. Materials

- 10 cm x 2 cm Glass column with valve;
- 15 and 50 mL centrifugal tubes in polypropylene, with lid, self-sustaining;
- 50 mL glass beaker;

2.3. Equipments

- Digital scale with four decimal places, Ohaus Pioneer;
- pHmetro, Model 420A Orion Analyser;
- Optical Emission Spectrometer with Argon Plasma Source, ICP-OES, Spectro Flame M120, Modula, Spectro Analytical Instruments;

2.4. Methods

The experiments were performed on a 20 cm high 2 cm internal glass column. The column was packaged with the resin volume established for the experiment. Prior to the use of the resin in ion exchange experiments, it was washed to remove degradation products and impurities

For the initial washing of the resin the reagents were used: 4M HCl, 2M NaOH and Ethanol. Each step of washing the resin passed with ultrasonic shaking for 20 minutes. The synthetic uranium solution with a concentration of 150 $\mu\text{g mL}^{-1}$ was obtained from a solution with a concentration of 8.618 mgU.g⁻¹ solution provided by the Nuclear and Energy Research Institute.

The ion exchange procedure was performed at room temperature with the sample at pH 7.82. The amount of resin in the column was 30 mL and the volume of solution was 120 mL. The solution was percolated through the column at a flow rate of 4.0 mL.min⁻¹ and four 30 mL aliquots were collected in 50 mL centrifuge tubes. The analysis of the samples was performed by determining the uranium content by Optical Emission Spectrometry with Argon Plasma Source, ICP-OES.

3. RESULTS AND DISCUSSION

The foundation explored in the uranium removal studies was based on the different affinities of the cations present in the starting material with the ion exchange resin. To improve the characteristics of the final product, it was adopted the strategy to apply ion exchange process until decontaminated solutions. Tests were performed by varying some parameters, such as flow rate, eluent composition and respective concentrations and the ratio of resin to cation.

Table 1 shows the characteristics of the sample contained before and after the ion exchange. The uranium concentration in the solution charged before being percolated by the column was 150 µg mL⁻¹, upon contact with the strong anionic resin the solution interacts with the resin and uranium is retained in the column. After percolation the aliquots were analyzed and the uranium concentration was found to be 0.0533 µg mL⁻¹.

Table 1 – Characteristics of the sample before and after the Ion Exchange process.

Aliquot	Volume (mL)	Concentracion U (µg mL ⁻¹)	Mass U (mg)
Start	120	150	18
Last	120	0,0533	0,0064
Efficiency: 99,96%			

The efficiency of this ion exchange process was 99.96%, as shown in Figure 1, since the uranium mass fell from 18mg to 0.0064mg. Those values are satisfactory, since uranium is a contaminant with high environmental radiological impact. It is estimated that by including a second stage of ion exchange in the process, with the same operating conditions, the effluent will comply with current legislation that determines a maximum concentration of 0.002 mg of U in wastewater.

The process flow is an important point in the art, since the elements have time of contact with the resin different from each other. In order to carry out this procedure, the diameter of the column and the height of the column were taken into account.

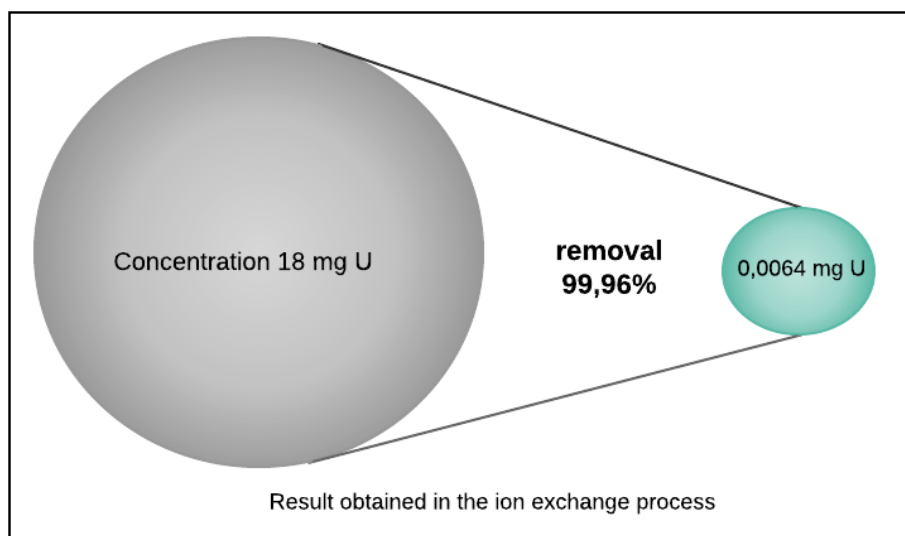


Figure 1: Result obtained in the ion exchange process.

Dowex 1X-8 resin is effective in removing uranium from contaminated effluents. Figure 1 shows the efficiency of the resin studied. Through the ion exchange process, the ions present in the resin interacted with the ionic forms of uranium in solution.

4. CONCLUSIONS

The ion exchange technique is widely used in the removal of metals, separation of chemical elements or recovery of contaminants. It is a cheap technique and it has simplified steps for its accomplishment. In addition, ion exchange is an important ally in the treatment of contaminated effluents or reduction of water hardness for drinking.

From the data presented, it is possible to conclude that the technique showed a high uranium removal efficiency, 99.96%. This value is considered satisfactory, but it is not acceptable at the level of dumping in water bodies. Therefore, the next step is to continue the ion exchange process and achieve the removal of 99.99% uranium.

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