Adsorption of Uranium (VI) from aqueous solution using silica gel

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Abstract

The recovery of uranium from nuclear industrial effluent has been studied by a Column filled with silica gel. The industrial effluent, at pH around 7, contains uranium (890 mg/L) and nitrate (30 g/L) and cannot be discharged without treatment. The effect of the flow rate and the temperature was studied. Adsorption was carried out at a flow rate of 50, 75 and 100 mL/hr, which corresponds to a retention time of 37, 25 and 16 min, respectively and a temperature of 298, 313 and 333 K. Elution was carried out with nitric acid at a concentration of 1 and 2 M. The breakthrough and regeneration experiments showed that the removal of U(VI) from a contaminated solution can be achieved by using a column packed with silica gel. The solution eluate might be recycled back into the process with the advantage of saving this valuable metal.

Key words: Adsorption, Uranium, Silica gel

1. Introduction

There is a continuous need for new separation techniques which selectively extract metal ions from dilute waste waters and industrial process streams. Tightening of environmental legislation and possible reuse of extracted metals have enhanced the search for new processes [1]. Usual techniques for metal removal are precipitation [2,3], solvent extraction [2], electrolysis [4] and adsorption and ion exchange [3,5]. Precipitation is not selective and not suitable for low-concentrated effluents due to a low metal recovery. Solvent extraction is non economical because of the loss of extracting agent and the production of large amounts of organic waste. Moreover, the effluent might be polluted with the solvent used for the extraction. Electrolysis is not appropriate for low concentrations. Adsorption and ion exchange has been widely studied for the recovery of metal ions from diluted streams [3,5–7]. Commercially available ion-exchange resins show high performances but generally poor selectivity towards different metal ions [8]. A high selectivity can be observed in some cases [9], but kinetics are slow due to the hydrophobic character of the polymeric backbone [10]. An appealing alternative method is the use of chelating agents grafted on a hydrophilic support in a solid–liquid extraction process [1].

Silica gel can be used to adsorb the uranium from waste water; it can also be used in the solutions with a high nitrate concentration. Silica gel is much more efficient for U(VI) adsorption than Amberlite® IRC718 iminodiacetic acid resin used in the same conditions. The low accessibility of uranyl to complexation sites is certainly due to the hydrophobic organic backbone of such commercial resins. In contrast, the hydrophilic nature of silica gel as well as its remarkable resistance to swelling is the main advantages of silica over organic polymer-based ion exchangers [1].

In acidic solutions, some U(VI) species adsorb mainly via an outer sphere mechanism on some silicates. Behavior between U polyhedra and silica tetrahedra as shown in Fig. 1 [11-13].

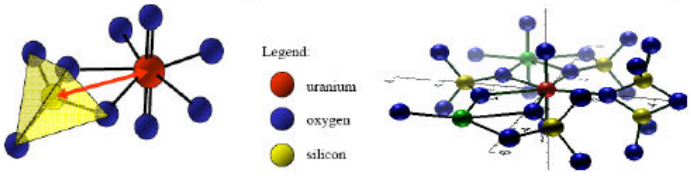

Fig. 1. Uranyl sorption to silica gels, the U-Si distance is ~2.71 Å (red arrow), for polymeric uranyl species, long U-Si distances are a signature for structural incorporation of U (i.e., longer bonds are required for a lattice arrangement as opposed to a sorption-type (i.e., amorphous) environment.

In this paper, we reported the properties of the silica gel for the recovery of uranium from nuclear industrial effluent. Various parameters involved in extraction efficiency, such as flow rate, and the temperature were studied in extraction experiments.

2. Experimental

2.1. Chemicals

The specification of silica gel adsorbent is given in table 1. The industrial effluent, at pH around 7, contains uranium (890 mg/L) and nitrate (30 g/L) and cannot be discharged without treatment. The pH of the solutions used in experiments was adjusted to 7 with NH_4OH solution. Diluted nitric acid was used for regeneration experiments.

Table 1. Specification of silica gel adsorbent

Appearance	Particle size(mm)	Real density(g/cm^3)	Bulk density(g/cm^3)	Average pore size(μm)	Specific area(m^2/g)
white	0.25-1.5	1.86	0.64	4	578

2.2. Adsorption and regeneration experiments (Fig. 2)

Adsorption experiments carried out using a glass column (D=10mm, *L*=400mm). A weighed amount of silica gel (20 g) was packed in the column. The silica gel was ready for use after washing with deionized water. The U(VI) solution was eluted downward at a flow rate of 50, 75 and 100 ml/hr and a temperature of 298, 313 and 333K, at pH=7. The regeneration experiments were carried out by downward elution of 1 and 2 M nitric acid at flow rate of 50 ml/hr.

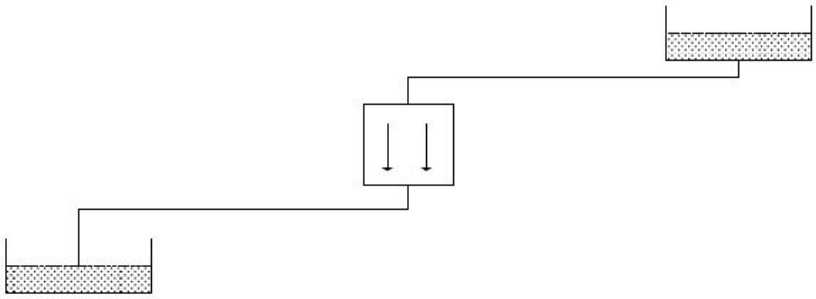

Fig. 2. Schematic drawing for adsorption and regeneration experiments. (1) Contaminated solution (a) or nitric acid (b). (2) Column filled with silica gel. (3) Decontaminated solution (a) or concentrated solution (b).

3. Results and discussion

3.1. Effect of flow rate

The adsorption performance of the silica gel was evaluated by means of adsorption or loading curves as shown in Fig. 3. These curves describe the breakthrough profiles for uranium at different flow rates in which the uranium concentration of the column effluent (mg/L) is plotted against time (hr). Although the profiles seem similar, the time which corresponds to the breakthrough point is different. The breakthrough point is usually defined as the time that the uranium concentration equal to 5% of that in the feeding solution. In this study it was set at 45 mg/L of uranium according to the industry quality criteria. As shown on Fig. 3, the breakthrough point was reached at approximately 13 hr for flow rate of 50 mL/hr, 11 hr for flow rate of 75 mL/hr and 8 hr for flow rate of 100 mL/hr. Therefore, the recommended operation flow rate should be 50 mL/hr which corresponds to the retention time of 37 min. The calculated silica gel capacity was 29 mg of uranium/g of silica gel for a flow rate of 50 mL/hr.

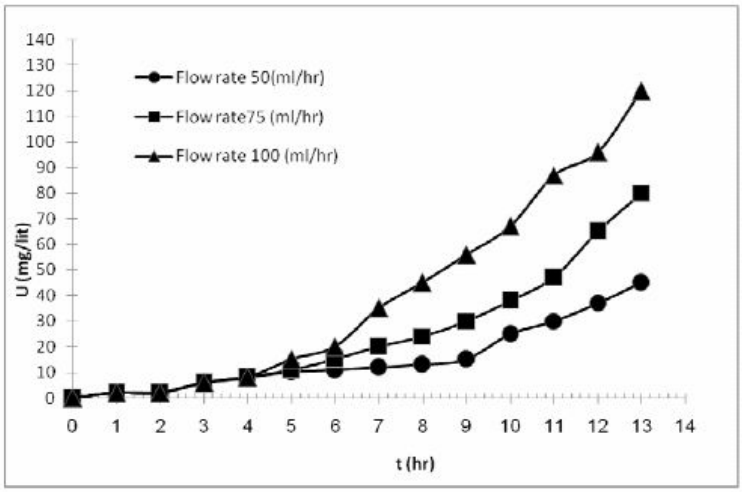

Fig. 3. Breakthrough profiles for different flow rates

3.2. Effect of the temperature

Fig. 4 shows the effect of the temperature on the uranium concentration. These curves describe the breakthrough profiles for uranium at different temperature in which the uranium concentration of the column effluent (mg/L) is plotted against time. Although the profiles seem similar, the time which corresponds to the breakthrough point is slightly different. As shown on Fig. 4, the breakthrough point was reached at approximately 13 hr for temperature of 298 K, 12 hr for 313 K and 11 hr for 333 K. Therefore, the recommended operation temperature should be 298 K.

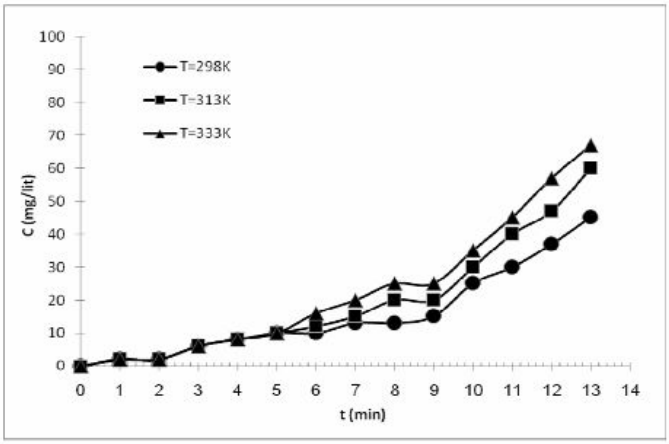

Fig. 4. Breakthrough profiles for different temperatures (flow rate of 50 mL/hr)

3.3. Regeneration experiments

The curve represented in Fig. 5 is an example of the elution profile of U(VI) using a column filled with silica gel and eluted with 1 and 2M nitric acid solution. Quantitative recovery of uranium is performed using 40 ml of nitric acid at a flow rate of 50 ml/hr. The recommended operation concentration of nitric acid should be 2 M. Although 2 M nitric acid is the better eluant, the 1 M nitric acid also presented a good efficiency. The properties of the silica gel remain unchanged after 2 adsorption / regeneration cycles.

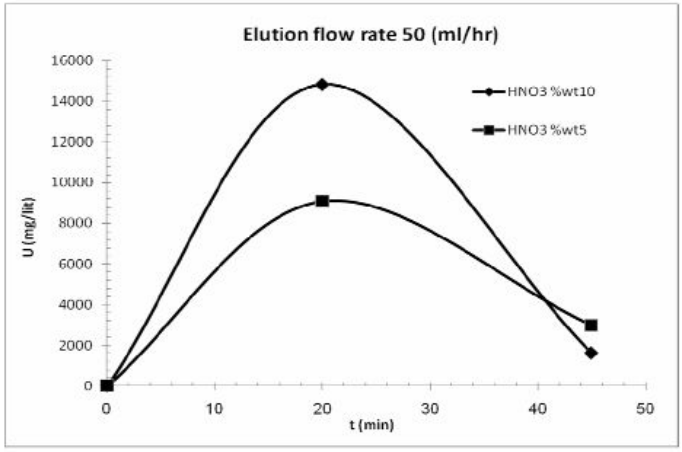

Fig. 5. Elution profile of uranium using a loaded column packed with silica gel eluted by 1 and 2 M nitric acid solution at flow rate of 50 mL/hr.

4. Conclusion

In the present investigation, the adsorption of uranium(VI) from aqueous solution by silica gel was studied. It was found that the silica gel was efficient for uranium removal from some nuclear industrial effluent. The uranium loading capacity was 29 mg of uranium/g of silica gel. The recommended operation flow rate and temperature was 50 mL/hr, which corresponds to a retention time of 37 min, and 298 K, respectively. Although 2 M nitric acid was the better eluant, the 1 M nitric acid also presented a good efficiency. Both of these eluants are technically suitable for recirculation to the industrial process. The properties of the silica gel remain unchanged after 2 adsorption / regeneration cycles.

References

[1] F. Barbette a, b, F. Rascalou a,b, H. Chollet a, J.L. Babouhot a, F. Denat b, R. Guilard b, Extraction of uranyl ions from aqueous solutions using silica-gel-bound macrocycles for alpha contaminated waste water treatment, Analytica Chimica Acta 502 (2004) 179–187.
[2] R.M.E. Diamand, Environmental Chemistry, Plenum Press, New York, 1977.
[3] G.R. Choppin, K.L. Nash, Radiochim. Acta 70–71 (1995) 225.
[4] A.T. Kuhn, Electrochemistry of Cleaner Environments, Plenum Press, New York, 1972.
[5] E.S. Pérez de Ortiz, Ion Exchange, Science and Technology, Martinus Nijhoff Publishers, Dordrecht, 1986.
[6T.A. Litovitz, P.B. Macedo, H. Hojaji, WO8803915 (1988).
[7] D.A. White, European Patent 0,266,945 (1987).
[8] S.K. Sahni, J. Reedijk, Coord. Chem. Rev. 59 (1984) 1.
[9] S.K. Sahni, W.L. Driessen, J. Reedijk, Inorg. Chim. Acta 154 (1988) 141.
[10] P.D. Verweij, S. Sital, M.J. Haanepen, W.L. Driessen, J. Reedijk Eur. Polym. J. 29 (1993) 1603.
[11] Burns, P. C. (1999). The Crystal Chemistry of Uranium. In: Uranium: Mineralogy, Geochemistry and the Environment. Reviews in Mineralogy 38, pp. 23-90, Mineral. Soc. Amer.
[12] Burns, P. C., Ewing, R. C. and Hawthorn, F. C. (1997). The crystal chemistry of hexavalent uranium: Polyhedral geometries, bond-valence parameters, and polymerization of polyhedra. Can. Mineral. 35, 1551-1570.
[13] Duff, M. C., Hunter, D. B., Barnes, M. J. and Fink, S. D. (2002). "Characterization of uranium and mercury speciation in high level waste tank 8F and 11H sludge," WSRC-TR-2001-00428, Rev. 0.