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Adsorption of Uranium from Dilute Aqueous Solution on Inorganic Adsorbents

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NOTE

Adsorption of Uranium from Dilute Aqueous Solution on Inorganic Adsorbents

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Abstract

The adsorption of uranium from a dilute aqueous solution by a large number of inorganic adsorbents has been investigated. A mixture of aluminum hydroxide, ferric hydroxide, and activated carbon in the weight ratio 1 : 3 : 4 has shown a high adsorbability for uranium. The separation of uranium from a dilute aqueous solution by this mixed adsorbent under various temperatures and pH values has been studied. The adsorbability was found to exhibit a maximum at pH 4.0 to 5.5 and to decrease with increasing temperature. A number of eluting solutions for the desorption of uranium from the mixed adsorbent were also tested: 1 *N* $(NH_4)_2CO_3$ was found to be the most suitable eluting solution (93% recovery of uranium).

Adsorption has become an established separation technique. It has found application as an effective and convenient technique not only in unit operations for a wide range of solute–solvent separation, but also in the separation and isolation of elements. The greatest advantage of adsorption is for the separation of a small amount of substance from a very large volume of solution. No universal adsorbent has yet been found, nor has a perfect adsorbent for any given purpose been developed. Knowledge of the adsorptive properties of inorganic materials has become of considerable importance in many fields, and the adsorptive properties of hydroxides are well known (*1*). Adsorption of trace amounts of ions from aqueous solution on metal oxide surfaces has been studied by numerous investigators (*2*–*4*). We have investigated the feasibility of using an adsorption process with various inorganic adsorbents for recovering uranium from dilute aqueous solutions. Our experiments have shown that the most satisfactory adsorbents are the alkaline earth oxides, hydroxides, and their salts (such as sulfates). This communication is concerned with our development of an inexpensive new adsorbent which has shown a high adsorbability for uranium. The separation of uranium from dilute aqueous solutions by this new adsorbent under various temperatures and pH values has been studied, and a number of eluting solutions for the desorption of uranium from this adsorbent were also tested. The main purpose of this investigation was a preliminary exploration of the subject.

EXPERIMENTAL

Chemical Reagents

All chemical reagents used in our experiments were of C.P. grade without further purification. The 300 to 400 mesh solid adsorbents (except activated carbon) were also C.P. grade. They were obtained from Wako Pure Chemical Industries, Japan. The 16-mesh IONAC P-50 activated carbon was obtained from IONAC Chemical Corp., U.S.

Preparation of Solutions

Aqueous reagents were prepared in deionized water. The standard uranium feed solution was prepared by dissolving 99.9% pure U_3O_8 in nitric acid and diluting with deionized water.

Apparatus and Methods

The concentration of uranyl ion was determined by polarography. A METROHM HERISAU polarecord E261 was used. Well-defined waves for the reduction of uranyl ion in a supporting electrolyte of 0.5 *M* $H_2C_2O_4$ were obtained for concentrations ranging from 10 to 10^4 ppm, and the results were accurate and reproducible.

Measurements of Adsorption Capacity

The measurements were carried out by batch techniques. One gram of adsorbent was brought into contact with uranyl solution in a closed flask and stirred for 2 hr with a magnetic stirrer to equilibrium at 25°C. Since adsorption of ions in aqueous solution follows the Langmuir- or Freundlich-type isotherm, the equilibrium concentration of $UO_2(NO_3)_2$ used was 100 mg/50 ml. The adsorption capacity was determined from the change in concentration of the solution. The capacity measurements under various temperatures and pH values were carried out in a 3 × 16 cm glass column, and a mixture of aluminum hydroxide, ferric hydroxide, and activated carbon in the weight ratio 1: 3: 4 was used as adsorbent. The pH value of the uranium solution was adjusted by nitric acid or ammonia solution. The flow rate was adjusted to 50 ml/hr. In measurements of the temperature effect on adsorption capacity, the column was provided with a water jacket for varying the temperature which was controlled to within ±0.1°C of the established operating temperature.

Desorption of Uranium from the Mixed Adsorbent

A number of eluting solutions at different concentrations was tested. The elution rate was adjusted to 0.15 ml/(min)(cm)^2. The effluent was continually collected for each fraction of 5 ml, and then the uranium concentration in each fraction was determined.

RESULTS AND DISCUSSION

A series of alkaline earth oxides, hydroxides, and sulfates was studied for the adsorbability of uranium. Nine adsorbents that showed a high adsorption capacity are listed in Table 1. In each experiment a blank test was performed (the solution was stirred in the absence of the adsorbent) to allow for adsorption on glass and for the possible effect of atmospheric gases (CO_2, O_2, etc). Aluminum hydroxide and ferric hydroxide are amorphous and thus have a very large effective surface area. Chemical compounds on the surface of hydroxides of aluminate and ferrite types are known to adsorb and to exchange ions from the solution with which they are in contact (*5*, *6*). These two inexpensive adsorbents were selected for further studies. Since the adsorption capacity of a column depends on the accessibility of the "surface" of the solid adsorbent to the molecules of the mobile phase, activated carbon was mixed with the hydroxides in order to increase the percolation rate of the column. Through a large number of systematic tests, we found that a mixture of aluminum hydroxide, ferric hydroxide, and activated carbon in the weight ratio 1: 3: 4 showed the highest adsorbability for uranium in aqueous solution. For a mixed

TABLE 1

Adsorbability of Different Adsorbents for Uranium[a]

Adsorbent	Adsorption capacity (mg U/g)
Lead monoxide	13.8
Beryllium oxide	17.6
Aluminium hydroxide	18.1
Activated carbon	19.7
Barium sulfate	20.5
Manganese dioxide	20.7
Zinc oxide	29.9
Ferric hydroxide	34.6
Magnesium oxide	45.0

[a] Equilibrium concentration used: 100 mg U/50 ml. Temperature: 25°C. pH value of uranium solution: 4.1.

adsorbent of 0.5 g aluminum hydroxide, 1.5 g ferric hydroxide, and 2 g activated carbon in the column, the capacity is 112 mg/g uranium at 25°C.

The temperature effect on the capacity of this mixed adsorbent is shown in Fig. 1. The curve shows that the capacity of adsorbent decreases with increasing temperature, indicating that the adsorption process is exothermic.

The effect of pH value on the capacity of this mixed adsorbent is shown in Fig. 2. The curve shows that the maximum capacity is in the pH range from 4.0 to 5.5. Since the pH value of the uranyl solution was adjusted by ammonia, at pH higher than 5.5 a yellow, insoluble precipitate of $(NH_4)_2U_2O_7$ was formed.

Different kinds of eluting solutions, i.e., HNO_3, HCl, H_2SO_4, NH_4OH, Na_2CO_3, $(NH_4)_2CO_3$, etc., were tested. Owing to the formation of the water-soluble complex of ammonium dioxotricarbonate uranate, $(NH_4)_2CO_3$ was found to be the most successful eluting solution. Figure 3 illustrates the elution curves of different concentrations of $(NH_4)_2CO_3$. The percent recovery of uranium with 1, 3, and 6 *N* of $(NH_4)_2CO_3$ is 93, 94, and 96%, respectively. From the economic point of view, 1 *N* $(NH_4)_2CO_3$ is the most suitable eluting solution. Since complete elution is very difficult, the elution curves show a small tailing. The uranium adsorbed in activated carbon is more difficult to elute than that in aluminum hydroxide or ferric hydroxide.

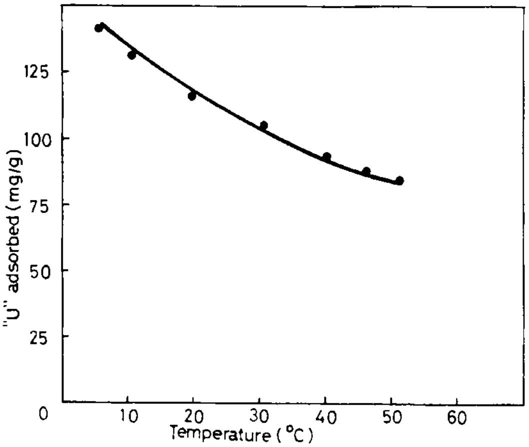

FIG. 1. Effect of temperature on adsorbability. Adsorbent: 2 g activated carbon + 1.5 g ferric hydroxide + 0.5 g aluminum hydroxide. pH value of uranium solution: 4.1.

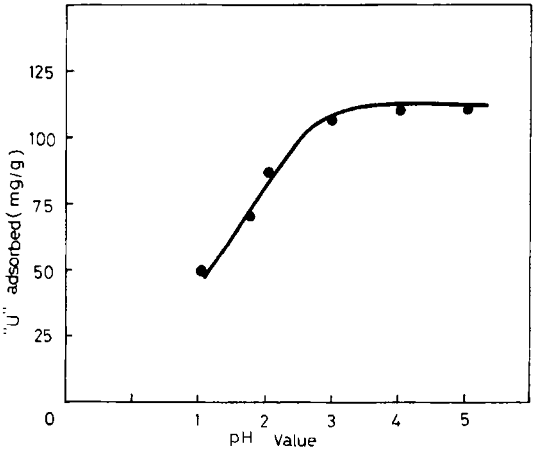

FIG. 2. Adsorbability as a function of pH value. Adsorbent: 2 g activated carbon + 1.5 g ferric hydroxide + 0.5 g aluminum hydroxide. Temperature: 25°C.

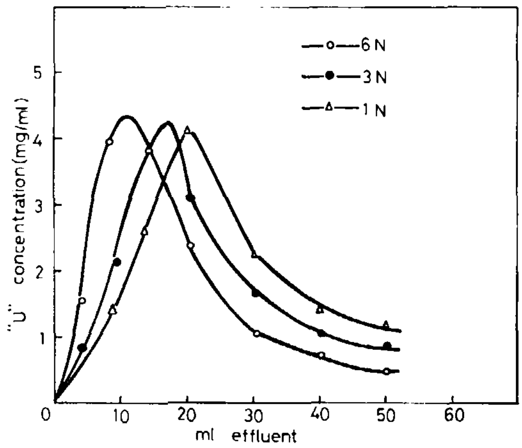

FIG. 3. Desorption curves of uranium with different concentrations of ammonium carbonate. Adsorbent: 2 g activated carbon + 1.5 g ferric hydroxide + 0.5 aluminum hydroxide. Bed dimension: 3.15 cm^2 × 6 cm. Elution rate: 0.15 $ml/(min)(cm)^2$.

REFERENCES

1. H. B. Weiser, *The Hydrous Oxide*, McGraw-Hill, New York, 1926.
2. E. Herczyńska, *J. Inorg. Nucl. Chem.*, *26*, 2127 (1964).
3. K. A. Kraus, H. O. Phillips, T. A. Carlson, and J. S. Johnson, "Ion Exchange Properties of Hydrous Oxides," in *Basic Chemistry in Nuclear Energy* (Volume 28 of the Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, September 1 13, 1958), 1958, Session C-9, P/1832.
4. S. Ahrland, I. Grenthe, and B. Noren, *Acta Chem. Scand.*, *14*, 1059 (1960).
5. E. Herczyńska, *Adsorption of Ions on Metal Oxide Surfaces in Aqueous Solution*, AEC-tr-6776, 1965, p. 23.
6. D. I. O'Connor, P. G. Johansen, and A. S. Buchanan, *Trans. Faraday Soc.*, *52*, 229 (1956).

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