ADSORPTION STUDIES OF CERIUM ON LEAD DIOXIDE FROM AQUEOUS SOLUTIONS

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The adsorption of cerium from aqueous solutions on lead dioxide has been investigated and optimized as a function of pH, equilibration time, sorbate and sorbent concentrations. The effect of other anions and cations on its adsorption has also been studied. Citrate, EDTA, tartrate, oxalate, U(VI), Th(IV), Pb(II), Cr(III) and Al(III) drastically reduce the adsorption. Adsorption of other metal ions on the same oxide has been measured under identical conditions. The distribution coefficient indicates that cerium can be separated from Fe(III), Tc(VII), In(III), Ag(I), Hg(II) and Ta(V). The data fitted very well to Freundlich as well as Dubinin-Radushkevich (D-R) isotherms. A mean free energy of sorption 11.62 ± 0.2 kJ mol⁻¹ was calculated, using the D-R equation and corresponds to an ion exchange reaction.

Hydroxides and hydrous oxides of various metals, because of their superior thermal and radiation stability, have been widely used as adsorbents over the last two decades. These materials have wide applications in industry and chemistry. Hydrous lead dioxide was used as adsorbent for the adsorption of Bi(III) and Cu(II) and was found to be a superior adsorbent for Bi(III) in acidic solutions.¹ ITO and coworkers² studied the adsorption of Bi(III) on HLD from aqueous solutions. Similarly HLD was used by Kawano and others³ for the adsorption of K, Cu, Zn, Cd, and nitrate ions. Lead dioxide, a promising inorganic adsorbent for metal ions, has been used for the adsorption of variety of elements.¹⁻⁵

Cerium, an important fission product, was the subject of several investigations of adsorption in the past, where oxides of aluminium,⁶⁻⁷ silicon,⁸⁻¹¹ iron,¹²⁻¹⁴ plutonium,¹⁵ thorium,¹⁶ and manganese¹⁷⁻²⁰ have been employed. The adsorption of cerium on manganese dioxide in the presence of complexing agents was reported earlier.²¹ Here we report the results of our investigations on the adsorption of cerium on lead dioxide from aqueous solutions.

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Experimental

Reagents and radiotracers: $^{139,141}$Ce tracer was produced by irradiating specpure cerium oxide in the PARR-1 research reactor of PINSTECH for five days at a thermal neutron flux of $2 \times 10^{13} \text{n cm}^{-2}\text{s}^{-1}$. All other isotopes were prepared by irradiating either metals or their appropriate compounds in the same reactor. Their radiochemical purity was checked by γ-spectroscopy.

Lead dioxide, a Fluka microanalytical reagent, black in color, was used as such. The BET surface area determined by adsorbing nitrogen was found to be 2.57 m$^2$/g. The porosity was found to be 0.36 cm$^3$/g and average pore diameter was found to be 0.56 μm. All chemicals used in this investigation were of analytical grade, and all the solutions were made in doubly distilled deionized water.

Buffer of pH 1–3 were made from the mixture of 0.1 mol dm$^{-3}$ HCl and KCl, and those of pH 4–7 were made from mixtures of 0.1 mol dm$^{-3}$ acetic acid and sodium acetate solutions, while buffer solutions of pH 8–10 were prepared by mixing 0.1 mol · dm$^{-3}$ ammonium chloride and ammonium hydroxide.

Procedure. Adsorption measurements were carried out radiometrically by batch technique, by shaking 3 cm$^3$ of buffer solution of known pH containing a known concentration of cerium. The details of experimental procedure and computation of distribution coefficient ($K_D$) and percent adsorption are given elsewhere. The measurement of $K_D$ was carried out at (23 ± 2) °C. The results are the average of at least triplicate runs and the error in most cases is around 5%.

Instruments. The pH measurements were made with a digital pH meter (POPE, model No. 1501). The surface area of lead dioxide was measured with a BET Quanatsorb Sorption System model No. QS-11. The pore size analysis was performed with a mercury porosimeter, Micrometrics Auto Pore 9200 model. Gross γ-measurements were carried out with the aid of a Chicago model 8725 well-type scintillation counter using a 58 cm$^2$ NaI(Tl) crystal. γ-Ray spectroscopy was done with the help of semiplanar 30 cm$^3$ Ge(Li) detector (Canberra Inc.) coupled with a Nuclear Data 4410 computerized multichannel analyzer having 8K memory.

Results and discussion

Prior to detailed studies of the adsorption of cerium, the time required to ensure an equilibrium between cerium and lead dioxide was determined. This was performed by shaking 50 mg of lead dioxide in 3 cm$^3$ buffer solution of pH 6 containing $8.3 \times 10^{-6}$ mol · dm$^{-3}$ radiocerium from 1 to 60 minutes. The adsorption equilibrium