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# ADSORPTION OF URANIUM ION IN SEA WATER ON COPRECIPITATED SILICA-TITANIA GEL

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Homogeneously coprecipitated silica-titania gel was shown to exhibit selectively high adsorptive ability for uranium ion in sea water. More than 90% of the adsorbed uranium ion on the gel was eluted with ammonium carbonate solution. The recovery percentage of the ion was found to be above 80%.

Despite extremely low concentration of uranium in sea water, the adsorption process is the most feasible among the various techniques for this recovery so far tested, whereas the development of an excellent adsorbent is prerequisite to a practical use. Hydrous titanium(IV) oxide has, at first, been most remarkable as an adsorbent for this process.[1,2] Various complex-adsorbents, e.g., hydrous titanium(IV) oxides supported on activated carbon,[3] dispersed in polyacrylamide gel,[4] and precipitated with iron oxide[5] have been studied. We report a novel adsorbent prepared by the mixing of both silica and titania gels for the improvement of adsorptive ability.

A silica-titania mixed-oxide gel was precipitated from the mixture of sodium metasilicate solution acidified with hydrochloric acid, titanium(IV) chloride, and urea by heating at 90°C for ca. 5 h until the pH of the solution became above 6. After filtration, the gel was thoroughly washed with deionized water and then dried at 110°C overnight, followed by grinding. Laboratory tests were conducted in terms of a batchwise operation by shaking 0.5 g of adsorbent with 1 $dm^3$ of sea water containing 3.0 $\mu g/dm^3$ of uranium ion sampled at the Cape of Omaezaki, Shizuoka Prefecture, Japan. Uranium ion in sea water was determined spectro-

photometrically using Arsenazo III as a reagent.[6] The surface area measurement of the gel was carried out by applying the B.E.T. method to adsorption isotherm of nitrogen at -196°C. The solid acidity of the gel was measured by titration with n-butylamine using 4-benzeneazo-1-naphthylamine ($pKa \leqq +4.0$) as an indicator.

Silica-titania gels of mole ratio in the range from 3:1 to 1:9 adsorbed more uranium ion from sea water than titania gel did, as shown in Fig. 1; the adsorption percentages for silica-titania gels were 72–85%, the values being higher in $TiO_2$-rich side of the composition, while that for titania gel was 70%. Figure 2 shows that the adsorption percentage increased gradually with an increase in temperature for the heat-treatment of the gel and reached the maximum around 400°C. This tendency was in fair agreement with the result of the solid acidity, suggesting the participation of acid sites on the surface of the gel in the adsorption. Specific surface areas of the gel, on the contrary, decreased with increasing $TiO_2$ content (Fig. 1). It has been therefore presumed that the $TiO_2$ component of the gel functions directly on the adsorption and the $SiO_2$ component contributes to the efficient adsorption resulting from the occurrence of porous solid structure.

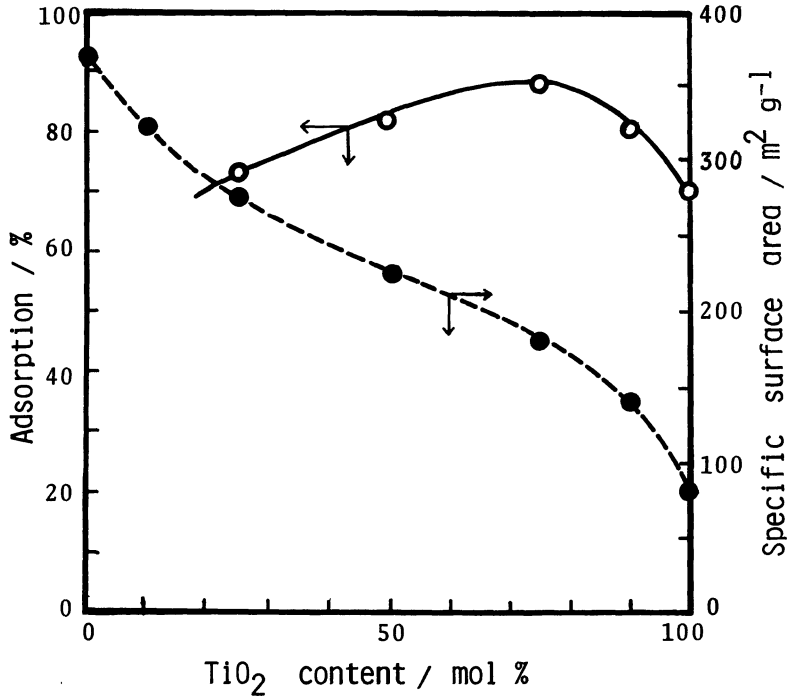


Fig. 1. Adsorption percentages of uranium ion (—○—) and specific surface areas (-●-) of silica-titania gels of various compositions heat-treated at 600°C. 5 d and 20°C.

Although this adsorption attained almost equilibrium for 5 d at room temperature, the adsorption rate was affected by temperatures of 10° to 50°C and obeyed Langmuir-type adsorption isotherm. The apparent activation energy for the adsorption was determined to be 60.5 kJ/mol so that it was attributed to chemical adsorption rather than physical one. The concentrations of the major ions contained in sea water changed little by the adsorption, as shown in the Table. The selectivity of the gel on the adsorption of uranium ion was satisfactory.

When silica-titania gel adsorbed with

Table. Change of the concentrations of the major ions in sea water.[a)]

| Ion | Concentration / g $dm^{-3}$ | |
|---|---|---|
| | Before the test | After the test |
| $Na^+$ | 10.8 | 10.6 |
| $Mg^+$ | 1.36 | 1.36 |
| $Ca^{2+}$ | 0.44 | 0.40 |
| $Cl^-$ | 20.0 | 19.3 |
| $SO_4^{2-}$ | 2.56 | 2.41 |

a) The adsorption was carried out under the conditions; $SiO_2/TiO_2$=1, 5 d, and 20°C.

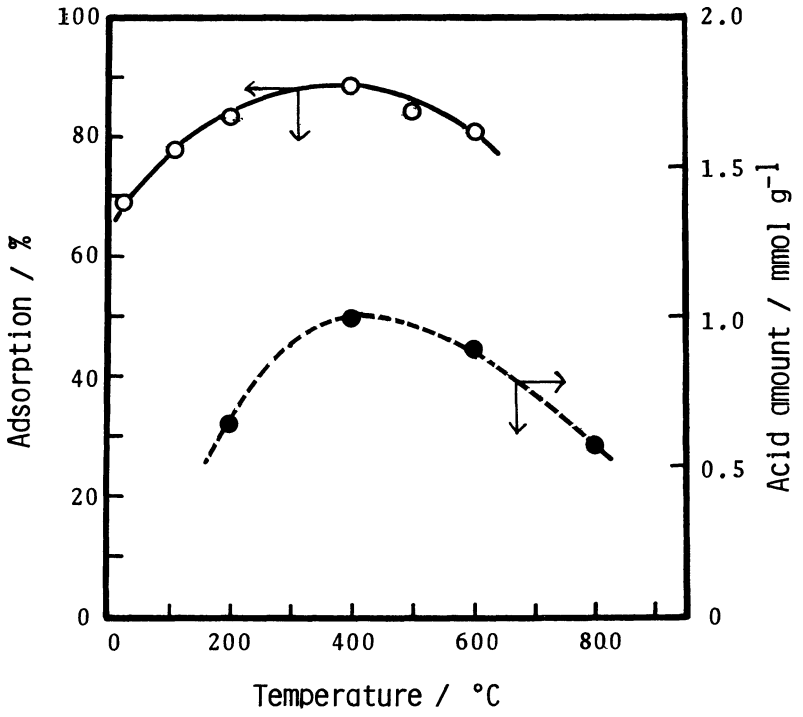


Fig. 2. Adsorption percentages of uranium ion (-○-) and acid amounts at pKa≦+4.0 (-●-) of silica-titania gels heat-treated at various temperatures. $SiO_2/TiO_2$=1, 5 d, and 20°C.

abundant uranium ion was shaken in 5% ammonium carbonate solution for 24 h at 50°C, more than 90% of the adsorbed uranium ion was eluted. Consequently, the recovery percentage of uranium ion was above 80%.

In addition, the granules of the gel were readily obtained by heat-treatment at 400–600°C, accompanied by the appearance of the high adsorptive ability. It may be expected that the granulated gel will be applicable to a column operation in a practical use.

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