

Extraction of Technetium by Tri-n-octyl Amine Impregnated Resin

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I. 19. Extraction of Technetium by Tri-n-octyl Amine-Impregnated Resin

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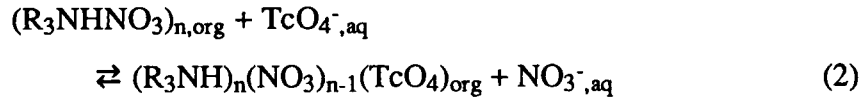
The content of long-life technetium-99 in waste solutions in nuclear fuel cycles must be kept as low as possible to prevent accumulation of ^{99}Tc in human-life environment.

Several methods such as sorption on ion exchange resins and active carbon have been employed for collection of technetium. Technetium can be sorbed on these sorbents from aqueous solutions, but some problems arise in elution of sorbed ions. If technetium sorbed can be readily desorbed, it contributes not only to recover useful isotopes but to reduce amounts of radioactive wastes containing Tc. A pertechnetate ion which is stable in acidic solutions can be highly extracted by tri-n-octylamine (TOA) and tri-n-octyl phosphine oxide (TOPO), however, these reagents extract simultaneously mineral acids and then result in lowering of the distribution ratio of Tc.^{1,2)}

In this study, liquid-liquid extraction is applied to solid-liquid extraction by impregnating TOA in macroporous resins (MR resin) for selective extraction of technetium, diminishing acid extraction.

Radioisotope $^{95\text{m}}\text{Tc}$ ($t_{1/2} = 60$ d) was made on a $^{95}\text{Mo}(d,2n)^{95\text{m}}\text{Tc}$ reaction by irradiation from an AVF cyclotron in Tohoku University. Technetium was separated from Mo target dissolved in aqua regia by solvent extraction with tetraphenylarsonium chloride-chloroform solution and then was stripped with 8 M nitric acid.

To investigate the extraction behavior of Tc, the distribution ratio was measured after shaking of equal volumes of 0.5 M TOA in toluene and a nitric acid solution for 1 h. The D_{Tc} value sharply decreases with an increase in acid concentrations above 1 M, as shown in Fig. 1. The distribution ratio of uranium are also given, for comparison, in Fig. 1. High separation factor ($D_{\text{Tc}}/D_{\text{U}}$) was obtained at low acid concentrations below 1 M. Plots of D against [TOA] in a log scale are given in Fig. 2. The D value for Tc increases with the concentration of TOA and the slopes of plots are found to be close to 2. The extraction equilibria can be expressed as³⁾,



Here, the reaction of $n=2$ is probably predominant. The pertechnetate ion can be quantitatively stripped by shaking with a NaOH solution.

A TOA-impregnated resin was prepared by soaking a macroporous resin (HP-series) in a toluene solution of TOA. This TOA-MR resin (m g) was equilibrated with the nitric acid solution ($V \text{ cm}^3$) containing ^{95m}Tc by shaking in a circle (100 rpm) at 25°C . After equilibration (24 h), the distribution coefficient K_{Tc} (cm^3/g) was determined from radioactivities of aliquots of the aqueous solution,

$$K_{Tc} = \frac{A_o - A_f}{A_f} \times \frac{V}{m} \quad (3)$$

where A_o and A_f are counting rates before and after equilibration with the MR resin, respectively.

The sorption behavior of Tc from dilute nitric acids was investigated using MR resins impregnating 1 M TOA. The K_{Tc} value decreases with increasing concentration of nitric acid from 0.1 to 2 M, as shown in Fig. 3. Using different kinds of MR resins, a similar sorption behavior was observed, indicating that MR resin serves only as an inert support for the extractant and sorption is entirely governed by the extraction reaction between the TOA solution impregnated in MR resins and aqueous solutions. The technetium sorbed was thoroughly eluted from the resin by contacting with a 1 M NaOH solution or by rinsing with an organic solvent such as acetone.

The effect of NaNO_3 of different concentrations, 0.01-2 M, on K_{Tc} is given in Fig. 4. The value of K_{Tc} is fairly high at low NaNO_3 concentration below 0.1 M, and decreases with increasing concentration above 0.5 M owing to the competitive reaction with the nitrate ion. The magnitude of the decrease in K_{Tc} value is rather small with the nitrate concentration compared to that with the nitric acid concentration, thus partial neutralization of the acid will enhance the K_{Tc} value.

Repetition of sorption on resin and desorption with 1 M NaOH is summarized in Table 1. The percent sorption and desorption are almost unchanged; the impregnated resin is effective for repetitive cycles. Thus, the solid-liquid extraction with the TOA impregnated resin is considered to be a useful and convenient means for collection of Tc from dilute acid solutions.

References

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Table 1. Repetition of sorption and desorption.

Times	Sorption(%)*	Desorption(%)**
1	58	91
2	56	89
3	56	89
4	56	86

*) 1 M TOA-impregnated HP-10
0.4 g; ^{95m}Tc in 2.16 M
 HNO_3 40 cm^3 ; 24 h

***) 1 M NaOH; 24 h

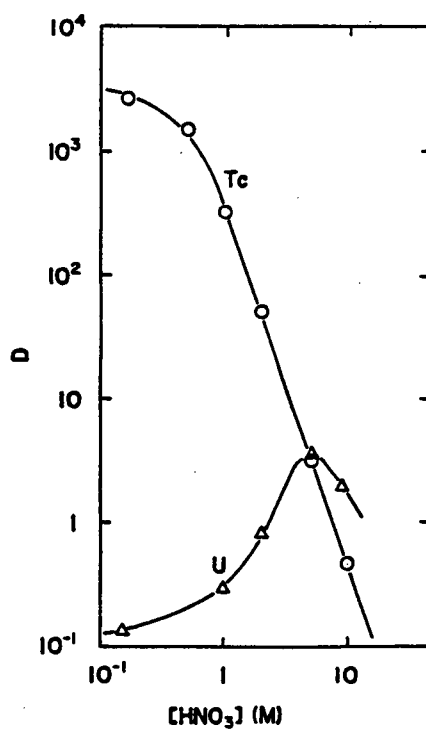


Fig. 1. Liquid-Liquid extraction of technetium and uranium by TOA. 0.5 M TOA in toluene.

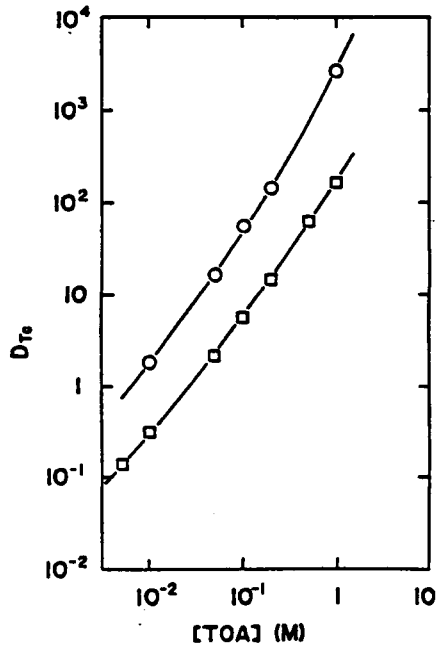


Fig. 2. Effect of TOA concentration on the distribution ratio of technetium. (○)0.5M HNO₃, (□)2M HNO₃.

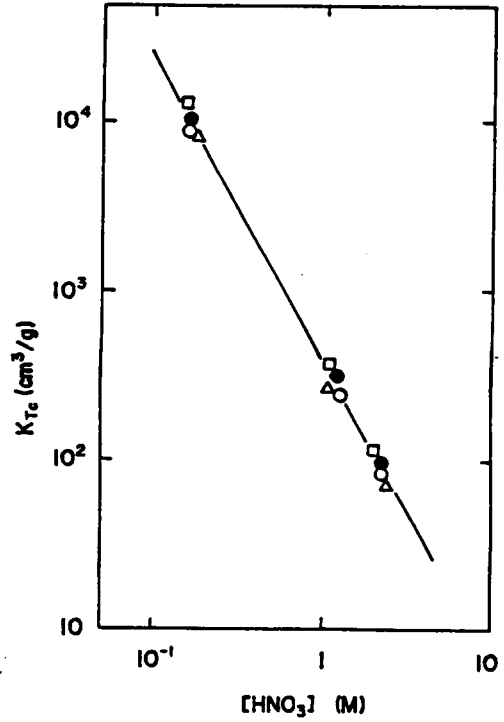


Fig. 3. Liquid-Liquid extraction of technetium by TOA-impregnated resin. 1M TOA/0.4 g MR resin; (●)HP-10, (Δ)HP-20, (□)HP-30, (○)HP-50.

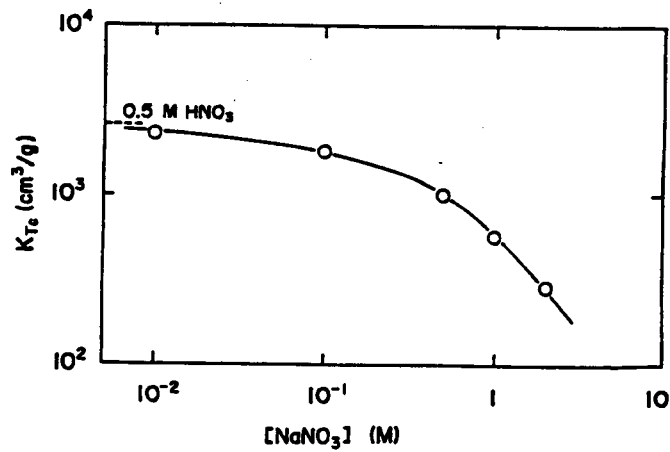


Fig. 4. Effect of sodium nitrate concentration on the distribution coefficient of technetium. 1M TOA/HP-10; 0.5M HNO₃-NaNO₃.