Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Mushtaq, A.(1993) 'Sorption Behavior of Carrier-Free Technetium-99*m* on Zinc Dust', Separation Science and Technology, 28: 9, 1743 — 1751

To link to this Article: DOI: 10.1080/01496399308019039

URL: http://dx.doi.org/10.1080/01496399308019039

SEPARATION SCIENCE AND TECHNOLOGY, **28**(9), pp. 1743–1751, 1993

Sorption Behavior of Carrier-Free Technetium-99*m* on Zinc Dust

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ABSTRACT

The sorption behavior of ^{99m}Tc on zinc dust was studied as a function of pH. Optimum conditions have been found for the preconcentration of traces of Tc by zinc. The influence of surface-active substances and of complexing agents on the sorption of trace amounts of Tc on zinc dust has been also studied.

INTRODUCTION

Technetium-99 is a fission product with a long half-life (2.12×10^5 years) and for this reason should constitute a radioactive nuclide of greatest consequence for long-term nuclear waste storage (1). Consequently, this isotope represents a potential hazard to the environment should it escape the containment of the repository. The sorption of technetium either by an artificial backfill material or by the surrounding geological medium, which is the last barrier protecting man and his environment by any mechanism, is therefore of considerable importance. Many inorganic sorbents have been successfully used for the preconcentration of trace elements (1–5). For technetium, complexes are usually prepared by reducing $^{99m}TcO_4^-/ReO_4^-$ with zinc dust in the solid phase in the presence of amino acids and amines (6, 7). The main purpose of this paper is to provide information about the sorption of trace amounts of Tc on Zn dust under various physicochemical conditions in solution and to define the best conditions for the preconcentration of traces of technetium.

EXPERIMENTAL

Technetium-99*m* was obtained from a Amertec II Technetium-99*m* Sterile generator (fission product ^{99}Mo adsorbed on alumina for the production of sterile, isotonic pyrogen-free solutions of ^{99m}Tc pertechnetate), Amersham International, U.K. Zinc dust was supplied by E. Merck (Germany) (Product 8774/591829). The characteristics of the zinc dust are given in Table 1. The common salts, acids, and bases used during this investigation were of commercial A.R. Grade. The water used to prepare the various solutions was doubly distilled. The sorption studies were performed by the batch method. The radionuclide was used in the form of $^{99m}TcO_4^-$ (unless some other form is mentioned) and added to the 50-mg Zn dust. The minimum concentration of NaCl was ~0.03 M in $^{99m}TcO_4^-$ solutions. The samples each had 5 mL total volume. The pH values were adjusted with NaOH or HCl solutions with the use of a digital pH meter (Hanna Instruments). The solution was shaken in a capped vial for a prescribed length of time. No attempt was made to measure the pH of the solution after the addition of Zn dust. The supernatant was separated after centrifugation, and 2 mL was used for ^{99m}Tc. Blank experiments were conducted in parallel (i.e., the procedure as described above, except that no Zn was added to the vials) for ^{99m}Tc. The radioactivity of ^{99m}Tc was determined by means of a Capintec Inc CRC-5RH radioisotope calibrator. Mini-Assay Type 6-20 (Mini Instruments) was used for counting radiochromatography strips. The percentage of sorption was calculated from

$$\text{Sorption} = \frac{A_1 - A_2}{A_1} 100\%$$

where A_1 = total radioactivity of ^{99m}Tc
A_2 = radioactivity of ^{99m}Tc left in solution

TABLE 1
Characteristics of Zinc Dust

Particle size	<60 μm
Specification assay (complexametric, total Zn)	>97%
Assay (manganometric, metallic Zn)	>95%
Substances insoluble in dilute HNO_3	<0.5%
Lead (Pb)	<1.0%
Cadmium (Cd)	<1.2%
Iron (Fe)	<0.01%
Copper (Cu)	<0.005%
Tin (Sn)	<0.01%

RESULTS AND DISCUSSION

The concentration of ^{99m}Tc in solution was kept ~10^{-10} M during the course of this study. Paper chromatography was used to determine the different radiochemical species (heptavalent and different reduced states) of ^{99m}Tc. The time required for almost 100% sorption of ^{99m}Tc on Zn dust was ~30 minutes. It is evident from Fig. 1 that the sorption process is very fast, and within 5 minutes approximately 95% ^{99m}Tc is sorbed. The sorption time in most of the experiments was therefore kept to 30 minutes. The dependence on pH of the sorption of trace amounts of ^{99m}Tc on Zn dust is illustrated in Fig. 2. Sorption between pH 2 and pH 9 is almost complete. Dissolution of Zn dust was noted below pH 2.0. In the 10–12 pH range, sorption decreases rapidly. The results shown in Fig. 2 indicate that Zn dust sorbent can be used for the quantitative preconcentration of TcO_4^-. Desorption of ^{99m}Tc can be effectively achieved with a solution of 0.01 M NaOH.

Plots of ^{99m}Tc sorption vs pH are presented in Figs. 3, 4, and 5 on the Zn dust from 0.001 to 1 M solutions of NaCl, $NaNO_3$, and Na_2SO_4,

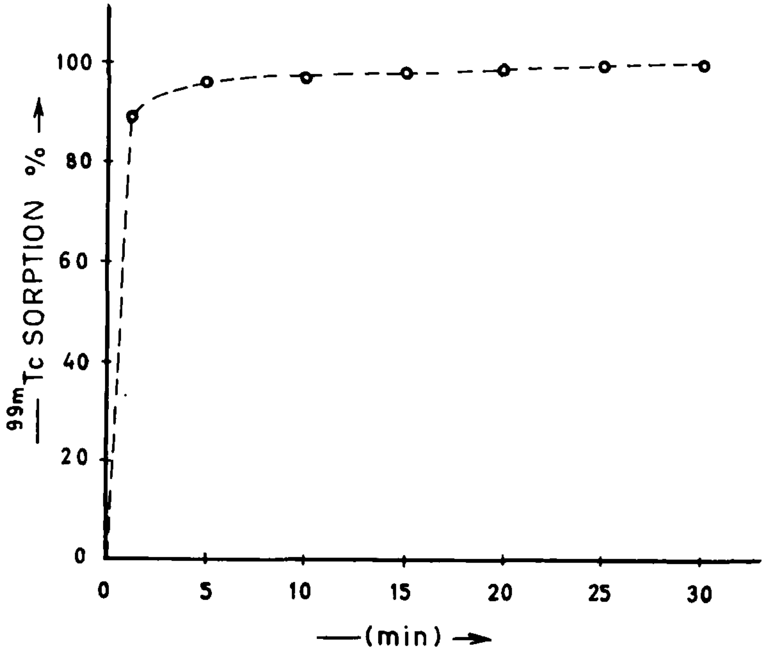

FIG. 1 The sorption of technetium-99*m* on zinc dust as a function of shaking time.

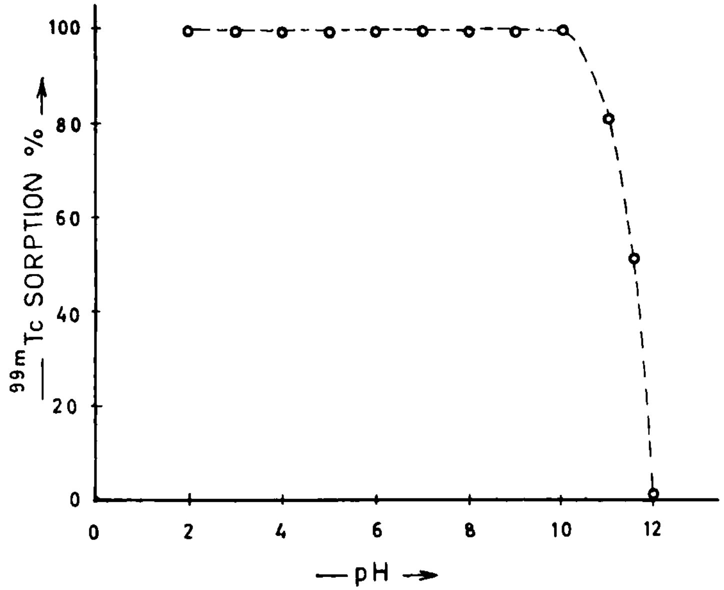

FIG. 2 The sorption of technetium-99*m* on zinc dust as a function of pH.

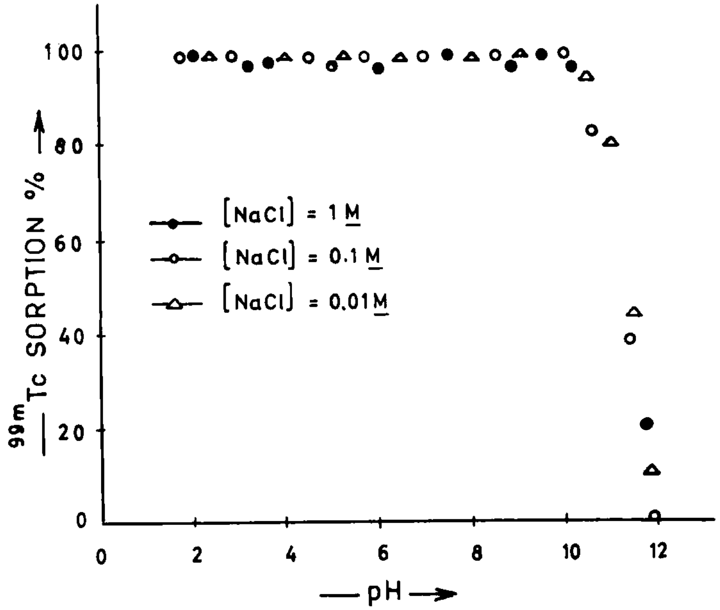

FIG. 3 The sorption of technetium-99*m* on zinc dust in the presence of sodium chloride as a function of pH.

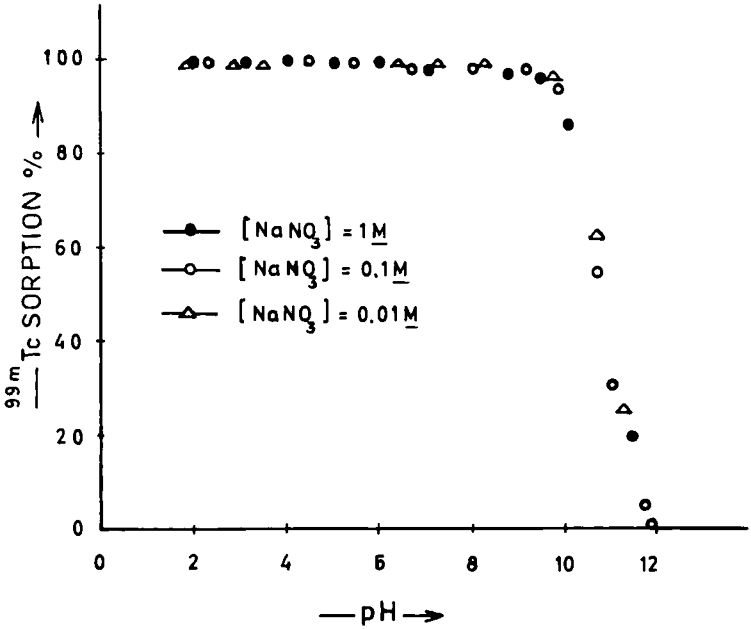

FIG. 4 The sorption of technetium-99*m* on zinc dust in the presence of sodium nitrate as a function of pH.

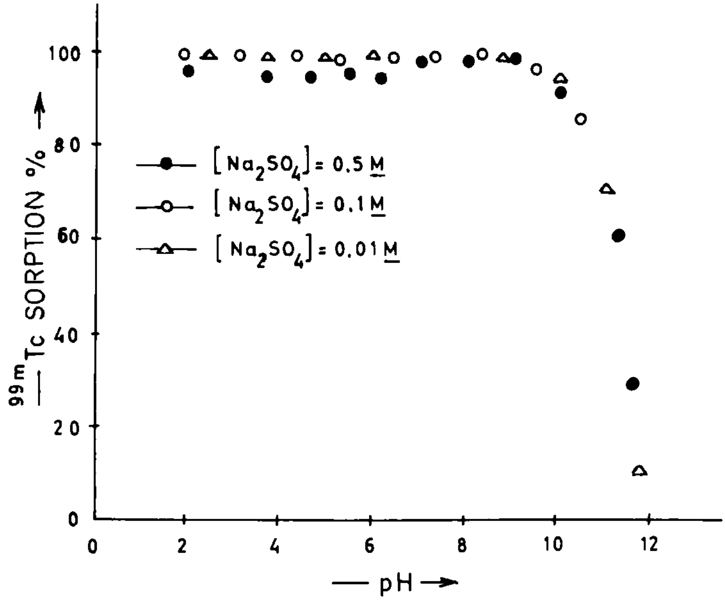

FIG. 5 The sorption of technetium-99*m* on zinc dust in the presence of sodium sulfate as a function of pH.

respectively. The shape of these curves is very similar to that observed in Fig. 2. A very small effect on sorption of ^{99m}Tc can be seen from a 1 M solution of NaCl and a 0.5 M solution of Na_2SO_4. This phenomenon establishes that the sorption of ^{99m}Tc on Zn dust is almost independent of the salts presence or absence. The effect on the sorption of $^{99m}TcO_4^-$ in 0.001–0.1 M solution of Na_2MoO_4 (parent–daughter relationship of ^{99}Mo–^{99m}Tc) was found to be negligible at pH 7. The sorption of trace amounts of ^{99m}Tc on zinc dust has also been studied as a function of pH in the presence of the disodium salt of EDTA and sodium citrate. The presence of the 10^{-4} M disodium salt of EDTA and sodium citrate has no significant effect on the pH dependence of the ^{99m}Tc sorption. The results on the influence of disodium salt of EDTA and sodium citrate on the sorption yield are shown in Figs. 6 and 7. The suppression effect of

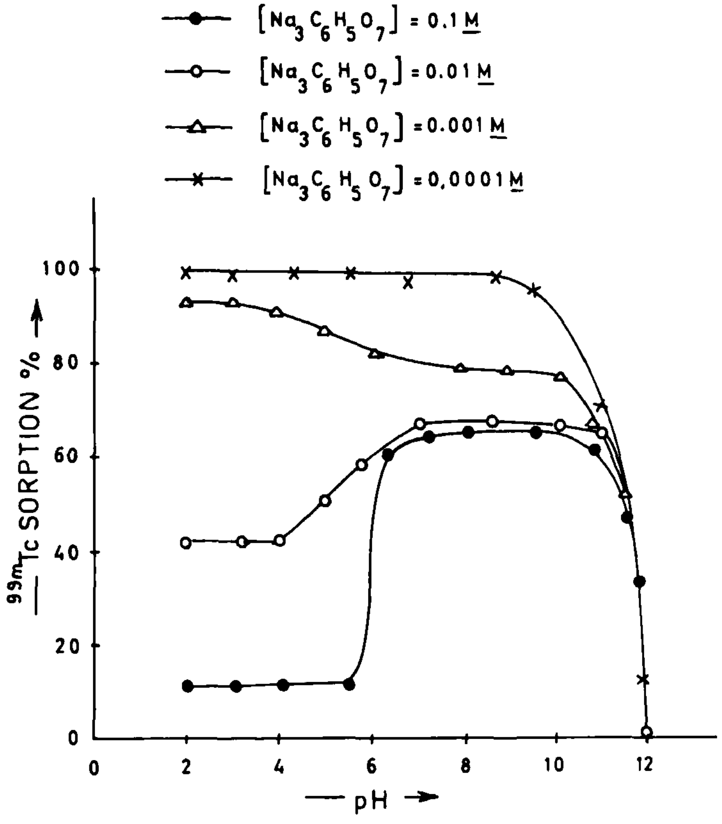

FIG. 6 The sorption of technetium-99*m* on zinc dust in the presence of sodium citrate as a function of pH.

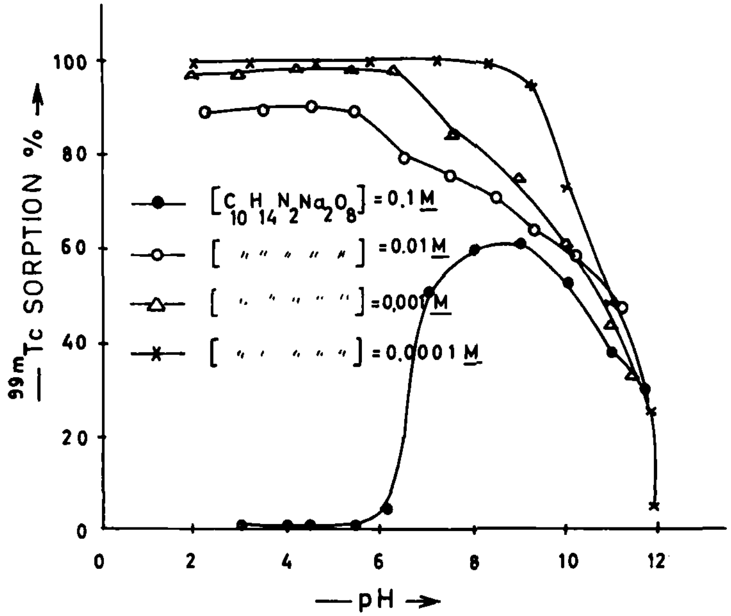

FIG. 7 The sorption of technetium-99*m* on zinc dust in the presence of EDTA as a function of pH.

sorption is less when the concentration of these complexing agents is low. At a 0.1 M concentration of sodium citrate and the disodium salt of EDTA, the surface area of Zn dust was very much reduced. From 2.0 to 5.5 pH, Zn dust coalesced into larger particles, and hence the sorption of ^{99m}Tc was almost completely suppressed. Zn dissolves in aqueous alkali to form ions such as aquated $[Zn(OH)_4]^{2-}$ zincates, and this may be the reason that the sorption of ^{99m}Tc decreases at pH values of about 10–12 for all the solutions reported. The sorption behavior of reduced ^{99m}Tc was studied in the 2–5 pH range. Eluted $^{99m}TcO_4^-$ solution was reduced by the addition of $SnCl_2$ (1 mg/5 mL $^{99m}TcO_4^-$). Reduction of ^{99m}Tc was confirmed by paper chromatography (8) (ascending type) using Whatman No. 1 paper in acetone and normal saline solution systems. That sorption of R-Tc on Zn dust was complete as in the case of $^{99m}TcO_4^-$ was established. ^{99m}Tc-tin colloid (5 mCi/5 mL) prepared in this laboratory ($SnCl_2 \cdot 2H_2O$ = 190 μg/mL, polyvinyl pyrrolidone = 250 μg/mL, NaF = 1 mg/mL, pH 5.6) was completely sorbed on Zn dust when it was shaken for 30 minutes. A freeze-dried mixture of 10 mg diethylenetriamine pentaacetic acid (DTPA) and 0.4 mg $SnCl_2 \cdot 2H_2O$, also prepared in this laboratory, was reconstitu-

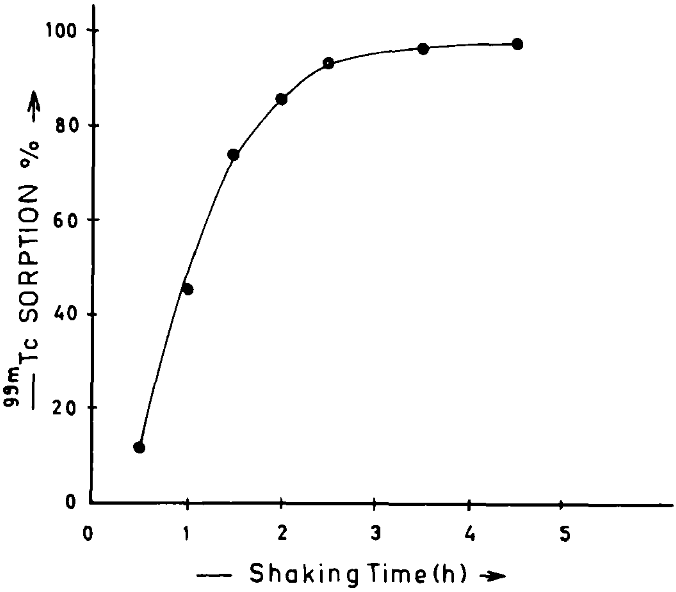

FIG. 8 The sorption of technetium-99*m* complexed with DTPA as a function of shaking time.

ted with ^{99m}Tc eluate (5 mCi/5 mL, pH 6.8) and shaken with 50 mg Zn dust. The sorption behavior of complexed ^{99m}Tc with DTPA is shown in Fig. 8. The effect of shaking time is very pronounced in this case.

Desorption of ^{99m}Tc from zinc dust was carried out with the addition of 0.01 M NaOH (^{99m}Tc was sorbed on zinc dust from $^{99m}TcO_4^-$ saline solution at pH 7). Paper chromatography (8) was used for the determination of the oxidation states of ^{99m}Tc. It was found that most of the ^{99m}Tc was sorbed as Tc(VII) (~80%) and the remaining ^{99m}Tc was reduced before sorption on zinc, evidently due to the reducing nature of zinc. Other experiments we carried out, such as the sorption of reduced ^{99m}Tc, ^{99m}Tc-Sn colloid, and ^{99m}Tc complexed with DTPA on zinc dust, also revealed that sorption is independent of the oxidation state of ^{99m}Tc. Thus, sorbent zinc dust can be effectively utilized to eliminate various species of ^{99m}Tc from different solutions, and the sorption/loss of ^{99m}Tc must be taken into account in the solid phase reduction of $^{99m}TcO_4^-$ with zinc.

ACKNOWLEDGMENT

I thank Dr. H. M. A. Karim for his active support of this research and for stimulating discussions.

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Received by editor April 3, 1991
Revised October 29, 1992