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SOLVENT EXTRACTION OF ^{99m}Tc/VII/ WITH METHYLENE BLUE INTO NITROBENZENE

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The extraction of ^{99m}technetium/VII/ with methylene blue into organic solvent has been investigated. Nitrobenzene, which was found to be the most effective extractant for ^{99m}Tc/VII/, was used to separate ^{99m}Tc/VII/ from other elements. The effect of various parameters on the extraction coefficient value such as effect of pH, time of equilibration, effect of anions and cations on the E value have been evaluated. The stoichiometry of metal to reagent was determined by the slope ratio method and was found to be 1:1.

INTRODUCTION

Methylene blue has been used for the detection of molybdenum[1]. However, the extraction of ^{99m}Tc/VII/ with methylene blue has not been reported in literature. The present work describes a rapid and selective method for the extraction of ^{99m}Tc/VII/ with methylene blue into nitrobenzene and its application to the separation of ^{99m}Tc/VII/ from other elements.

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EXPERIMENTAL

Chemicals and reagents

All the chemicals, reagents and solvents used were of A.R. grade. Reagent grade /99% purity/ methylene blue of B.D.H. make was used in the extraction studies. Carrier solution was prepared by dissolving appropriate salts in double distilled water. Acid was added wherever necessary. The strength of the solution was determined by the standard method given in Vogel[2].

^{99m}Tc isotope used as a tracer in the extraction studies was obtained by irradiating MoO_3 with thermal neutrons in the CIRUS reactor, to produce ^{99}Mo by the /n,γ/ reaction on ^{98}Mo. ^{99m}Tc was separated from ^{99}Mo by the method of Gerlit[3]. The isotopes used for the interference studies were provided by the Isotope Group of BARC, Trombay, Bombay.

Instrumentation

γ-Emitters were counted on a γ-ray spectrometer in conjunction with 3.5 cm x 3.5 cm NaI/Tl/ well-type detector. β-Emitters were counted on a thin end-window type GM counter in conjunction with a decade scaler, high voltage unit and timer.

Extraction procedure

In a 150-ml separatory funnel, ^{99m}Tc was taken and the volume was made to 16 ml with water after adjusting the pH of the solution with HCl or NaOH. 1 ml of 1% methylene blue in nitrobenzene was added. The aqueous phase was equilibrated for 2 min with 15 ml of nitrobenzene. The phases were allowed to separate and a 2-ml aliquot of each phase was taken in a counting dish, evaporated to dryness and counted on a GM counter or on a γ-ray spectrometer. The extraction coefficient value was calculated in the usual way.

RESULTS AND DISCUSSION

The extraction coefficient /E/ of ^{99m}Tc/VII/ between aqueous solution and nitrobenzene showed a maximum value of E = 178 at a pH of 3.5. The percentage extraction was better than 99% over a pH range of 3.0 to 4.0. Below and above this range it decreased. The extraction coefficient was found to be maximum for an equilibration time of 2 min /Table 1/.

The extraction coefficient of ^{99m}Tc/VII/ in favour of different organic solvents was ascertained, and it was found that the E value followed a decreasing trend:

	Nitrobenzene >	chloroform >	nitromethane >	n-butanol
E =	178	2.8	2.5	0.89

O-dichlorobenzene, carbon tetrachloride, xylene, ethyl acetate, amyl acetate, cyclohexane, methyl ethyl ketone, toluene, isobutyl methyl ketone were not soluble in these solvents whereas with isopropyl alcohol and pyridine the phases were found to be miscible.

Of the salts studied 100 mg of chloride, borate, iodate; 50 mg of carbonate, phosphate, acetate, thiourea, 25 mg of sulphate, persulphate and 10 mg of EDTA, bromate, hypophosphate, fluoride, tartrate and oxalate does not decrease the extraction coefficient value of ^{99m}Tc/VII/, whereas 10 mg of nitrite, thiosulphate, citrate interfere, although percentage extraction was found to be better than 99%. 10 mg of perchlorate, nitrate, iodide decreased the extraction coefficient value.

TABLE 1

Effect of pH and time of equilibration on the extraction coefficient of ^{99m}Tc/VII/ with methylene blue into nitrobenzene. Aqueous phase: Distilled water + ^{99m}Tc/VII/ + acid or alkali. Organic phase: 1 ml of 1% methylene blue in nitrobenzene + 15 ml of nitrobenzene. Temperature: 28 ± 2 °C

pH	Extraction coefficient, E	Percentage extraction, %E	Time of equilibration, min
2.0	130	99.23	2.0
3.0	159	99.37	2.0
3.5	178	99.44	2.0
4.0	164	99.39	2.0
5.0	77	98.71	2.0
6.0	44	97.77	2.0
7.0	43	99.72	2.0
8.0	8.8	89.79	2.0
9.0	7.2	87.80	2.0
3.5	129	99.23	1.0
3.5	178	99.44	2.0
3.5	147	99.32	3.0
3.5	43	99.74	4.0

The selectivity of the method was evaluated by determining the separation factor of the elements /Table 2/ Ba/II/, Cr/III/, Mn/II/, Fe/III/, were extracted to less than 2%. K/I/, Na/I/, As/III/, As/V/, Ce/III/, Ce/IV/, Ca/II/, P/V/, S/VI/, Zr/IV/, Zn/II/, Cu/II/, Ag/I/, Tl/I/, Cd/II/, Cs/I/, Rb/I/, Eu/III/, Sb/III/, Cr/VI/, Ir/IV/ were extracted to less than 10%. Co/II/, Pd/II/, Sc/III/, Ru/III/, W/VI/, Hg/II/, Au/III/, Mo/VI/, Se/IV/, and Re/VII/ were extracted to greater than 10%. The inter-

TABLE 2

Separation factor for various elements. Aqueous phase: Water + ^{99m}Tc + isotope of the element concerned /with or without carrier/. Organic phase: 1 ml of 1% methylene blue in nitrobenzene + 15 ml of nitrobenzene. pH = 3.5. Time of equilibration = 2 min. Temperature: 28 ± 2 °C

Separation factor >	Element	
	With milligram amount of carrier	Without carrier
10^4	Ba/II/,Cr/III/,Mn/II/, Co/II/[a].	Ba/II/,Fe/III/, Co/II/[a].
10^3	Mn/II/,Fe/III/,K/I/, Na/I/,As/III/,As/V/, Ce/III/,Ce/IV/, Ca/II/,P/V/,S/VI/, Zr/IV/,Zn/II/,Cu/II/, Tl/I/,Cd/II/,Co/II/, Sc/III/[a],Ru/III/[a], W/VI/[a],Co/II/, Mo/VI/[a].	Cr/III/,K/I/,Na/I/, As/III/,As/V/, Ce/III/,Ce/IV/, Ca/II/,P/V/,S/VI/, Zn/II/,Cu/II/,Ag/I/, Cs/I/,Rb/I/,Eu/III/, Sb/III/,Co/II/, Sc/III/[a],Ru/III/[a], W/VI/[a],Mo/VI/[a].
10^2	Cr/VI/,Cd/II/,Au/III/, Pd/II/,Ru/III/,Sc/III/, Se/IV/.	Cr/VI/,Tl/I/,Hg/II/, Ru/III/,W/VI/,Se/IV/, Mo/VI/,Sc/III/.
10^1	Hg/II/,W/VI/,Mo/VI/.	Ir/IV/,Pd/II/.
10^0	Au/III/,Re/VII/.	Re/VII/.

a = After treatment.

ference of Sc/III/, Ru/III/, W/VI/, were masked by using 10 mg of NaF, whereas Co/II/, and Pd/II/ were masked by using EDTA, Se/IV/ was precipitated as metal by using SO$_2$

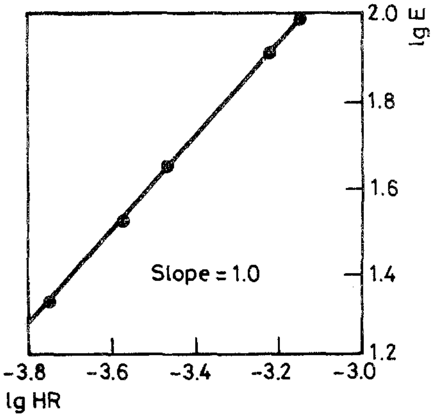

Fig. 1. Variation of extraction coefficient value with reagent concentration. Aqueous phase: distilled water + ^{99m}Tc/VII/ + acid. Organic phase: Varying amounts of methylene blue into nitrobenzene organic phase + 15 ml of nitrobenzene. Time of equilibration: 2 min. pH: 3.5. Temperature: 28±2 °C

water. The interference of Au/III/, was removed by precipitating it with SO_2 water prior to the extraction of ^{99m}Tc/VII/. Re/VII/ was found to follow ^{99m}Tc/VII/ in the extraction process. Carrier free ^{99m}Tc/VII/ was separated from mg quantities of Re/VII/ by the procedure given below.

Aqueous phase containing tracer ^{99m}Tc/VII/ was taken in a beaker along with 1 mg of Re/VII/ labelled with ^{186}Re. 100 mg of hydroxylamine hydrochloride was added and the solution was heated for 10-12 min. The solution was allowed to cool and the extraction was carried out as mentioned above. The aqueous phase contained ^{99m}Tc/IV/ and traces of Re/VII/.

The stoichiometry of metal to reagent was evaluated by the slope ratio method and was found to be 1:1 /Fig. 1/.

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