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Separation of Technetium from Neutron Irradiated Molybdenum

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Summary

A separation procedure of ^{99m}Tc using tetraphenylarsonium chloride has been studied. It is found that the separation of ^{99m}Mo is better carried out below 5 % ammonia-0.1M tetraphenylarsonium chloride chloroform solution system. By this method, ^{99m}Tc can be separated from the neutron irradiated molybdenum quantitatively in radiochemically pure form. Tracing the decay of the extract proves to be in good agreement with the decay of ^{99m}Tc.

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中性子照射をしたモリブデンよりテクネチウムの分離

要　旨

塩化テトラフェニルアルソニウムを用いる^{99m}Tcの分離法を研究し，つぎのことがわかった．^{99}Mo-^{99m}Tcを含む5%以下のアンモニヤ水溶液-0.1M塩化テトラフェニルアルソニウムのクロロホルム溶液系で，溶媒抽出をすれば，^{99m}Tcは^{99}Moを含まずに，よく有機層に抽出される．この方法で，中性子照射をした^{99}Moより，^{99m}Tcを定量的に，放射化学的に純粋に抽出することができる．抽出されたテクネチウムは6.0時間の半減期で減衰し，^{99m}Tcの減衰によく一致している．

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^{98}Mo, present with an abundance of 24% in the naturally occurring molybdenum, when irradiated with thermal neutrons yields technetium according to the following decay modes,

$$^{98}\text{Mo}(n,\ \gamma)\ 67\text{h}\ ^{99}\text{Mo} \xrightarrow{\beta} 6.0\text{h}\ ^{99m}\text{Tc} \longrightarrow 2.12\times10^{5}\text{y}\ ^{99}\text{Tc} \xrightarrow{\beta} {}^{99}\text{Ru}$$

Up to now, many attempts have been made for the separation of technetium from its parents. However, coprecipitation, ion exchange, solvent extraction and paper chromatography are the most commonly employed(1−3). Extraction of technetium into various organic solvents in acidic, neutral and basic media has been extensively studied(4−6). Tribalat and Beydon extracted technetium into chloroform using tetraphenylarsonium chloride at pH 10~11(7) ; however, their experimental procedure was somewhat troublesome. The authors, in the course of studying solvent extraction behaviors of inorganic ions, found that quantitative extraction of technetium with tetraphenylarsonium chloride could be achieved by dissolving the irradiated ammonium molybdate directly in ammonia and extracting into chloroform. This method is also applicable to the milking of technetium from molybdenum.

Experimental and Results

Reagents Ammonium molybdate of C. P. grade was used as the target material for neutron irradiation. Tetraphenylarsonium chloride of C. P. grade was recrystallized from distilled water and dehydrated before preparing 0.05M chloroform solution, which was standardized by Volhardt method. ^{99}Tc was imported from Oak Ridge National Loboratory, U. S. A.

Instruments A well type RCL 256 Channels Pulse Height Analyzer with 5″ NaI was employed for checking the γ-ray spectra, and an RCL proportional counter and a Philips well type scintillation counter were used for β- and γ-counting, respectively.

Extraction Technique Two and half ml of 0.05M tetraphenylarsonium chloride chloroform solution were added to 2.5 ml of 2~35% hydrochloric acid and 0.5~20% ammoniacal solutions. After adding the radioisotopes with more than 10^4 cpm/100 μg specific activities and shaking vigorously for 2 minutes, the organic and aqueous layers were transferred separately into tubes and centrifuged at about 4000 rpm. Aliquots of each phase were taken for counting. The distribution ratios, as a measure of extractability, is easily obtained by dividing the activity of the organic layer over that of the aqueous layer. The results, as illustrated in Fig. 1, show that in both ammonia and hydrochloric acid systems technetium is extremely extractable, while molybdenum is not.

Separation of Technetium from the Irradiated Molybdenum

The irradiated ammonium molybdate from the JRR-1 reactor was dissolved after putting aside for several hours that radiometric equilibrium between ^{99}Mo and ^{99m}Tc might be attained. The basicity was adjusted approximately at 0.5% ammonia solution, and in case of the presence of large excess of molybdenum larger amounts of ammonia were added to prevent molybdenum from precipitation. As seen from Fig. 1, the Kds of technetium are most independent from of ammonia concentration, molybdenum is not extracted in the ammoniacal solution. Therefore, a strict adjustment of basicity was by no means necessary.

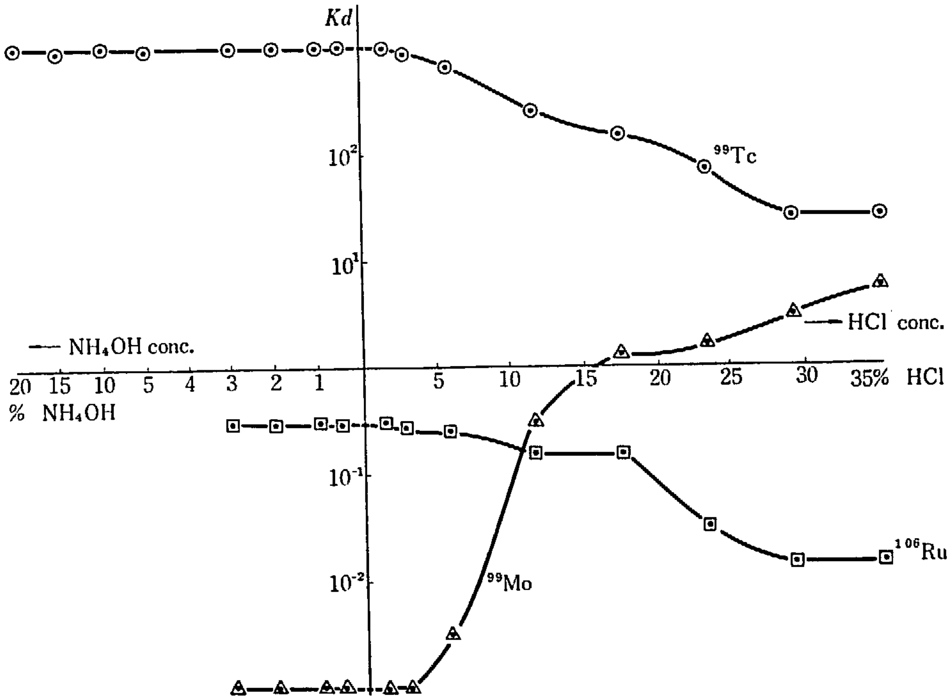

Fig. 1 The Kd Values of Molybdenum, Technetium and Ruthenium

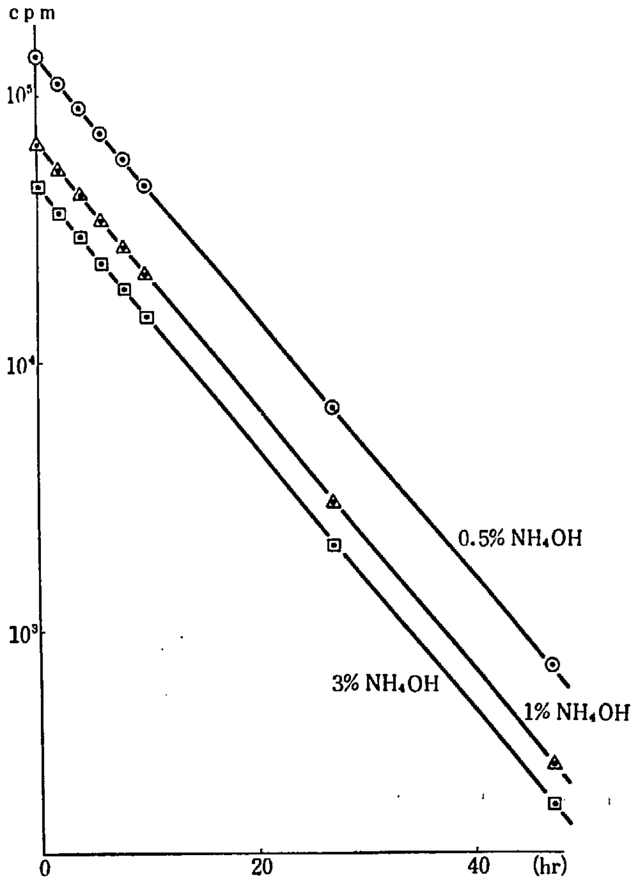

Fig. 2 Decay Curves of the Extracted ^{99m}Tc

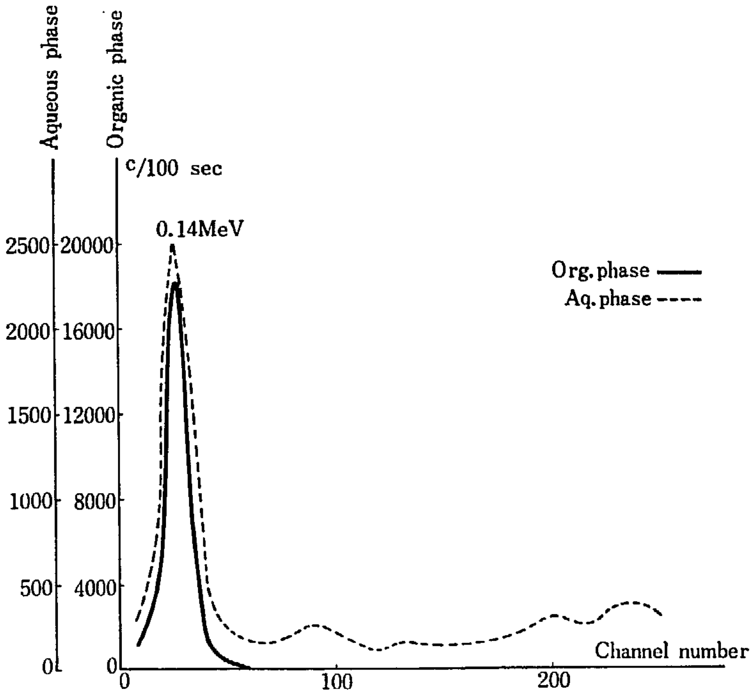

Fig. 3 γ-ray Spectra of the Organic and Aquaous Phases

Fig. 2 shows the decay curves of the extracts from 0.5, 1 and 3% ammoniacal solutions. They were traced over three days and were in good agreement with the decay of ^{99m}Tc. γ-ray spectrum, **Fig. 3**, for the extracts from 0.5% ammonia solution also confirmed that actually no radionuclides other than technetium were extracted into the chloroform layer.

Extraction behavior of molybdenum and technetium is extremely different both in ammoniac and hydrochloric acid solutions of lower concentrations so complete separation of technetium in the acid solution is also possible. However, extraction in the ammonia system has the advantage that complete dissolution of molybdenum takes place so that contamination due to other neighbouring radionuclides such as ruthenium and niobium could not be allowed for. As for the recovery of the extracted ^{99m}Tc, the chloroform layer was evaporated to dryness and then dissolved in distilled water. The solution was passed through an ion exchange colum filling with Dowex 50×2, the effluent contained only technetium and the tetraphenylarsonium ion was absorbed by the exchanger.

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