

DETERMINATION OF NEPTUNIUM-239 FROM FALLOUT SAMPLES.

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ABSTRACT

An analytical procedure is described for the separation of neptunium-239 from fallout samples. Neptunium is separated from uranium, plutonium and fission products by solvent extraction method. Np (IV) is extracted from 1 M nitric acid by shaking with equal volume of 0.5 M thenoyl-tri-fluoro acetone (T.T.A.) solution in xylene and back-extracted with 8 M nitric acid. Finally, Np is co-precipitated with zirconium mandelate, and counted in a low background beta-counting set-up. The average chemical recovery by this procedure is 82.6%. Under the conditions described, very good separation of neptunium is effected from aluminium, iron, fission products, thorium, uranium and plutonium.

1. INTRODUCTION

NEPTUNIUM-239 is one of the important isotopes in the early fallout from the nuclear weapon tests. It is produced by the interaction of neutrons with the uranium used in the weapon, by (n, γ) reaction. The neutron capture cross-section for this reaction is fairly large and therefore the Np^{239} is present in large quantities in the fresh fallout. The rapid determination of Np^{239} in the presence of fission products, uranium and plutonium, requires a quantitative chemical separation from these elements, and generally solvent extraction methods are advantageous for such analyses.

Magnusson *et al.*¹ studied the extraction of neptunium from hydrochloric acid solution by thenoyl-tri-fluoro acetone (T.T.A.) and determined the distribution coefficients for neptunium as a function of acidity and valence state. Moore² demonstrated the possibility of quantitative extraction of Np (IV) with T.T.A. from nitric acid solution. The procedure described by Moore was tried for the fallout samples but it did not give consistent chemical recovery of Np^{239} . This was presumably due to incomplete reduction of neptunium to the fourth valence state or due to a change in the sample matrix. For a complete reduction of neptunium to the fourth valence state a combination of hydroxylamine hydrochloride and ferrous sulphamate was found more suitable. Fallout samples contain varying amounts of silica and iron, and as these elements interfere in the extraction of neptunium, one has to take special care during the chemical analysis.

Present paper gives the analytical procedure for the separation of Np^{239} from fallout samples. The separated neptunium is completely free from uranium and fission products.

2. EXPERIMENTAL

Sample Preparation.—(i) The high altitude dust samples are collected from the surfaces of commercial aircrafts. The various parts of the aircraft are swiped with cotton wool soaked in white petrol. After gamma-spectral analysis, the samples are leached with a mixture of 3 M hydrochloric acid and 0.1 M hydrofluoric acid. More than 95% radioactivity gets leached by this leaching mixture.³ The leached solution is made to a definite volume and known amounts of inactive carriers like Sr, Cs, Zr, Ce and Ru are added to it.

(ii) Rain-water samples are evaporated to dryness and the residue is leached in the same way as swipe samples.

(iii) Air filter samples are leached directly with the same leaching mixture as above.

Chemical Procedure and Results.—A suitable aliquot of the leached solution is taken for the neptunium analysis with 5 mg. of iron carrier. 1.0 gm. of hydroxylamine hydrochloride, boric acid + nitric acid mixture are added to the solution. The solution is then warmed and ammonium hydroxide is added slowly to the solution to precipitate hydroxides. The hydroxide precipitate is dissolved in dilute nitric acid and the solution is evaporated to dryness. (If silicic acid appeared while evaporating, it should be centrifuged in the presence of hot concentrated nitric acid to avoid the losses of neptunium due to adsorption on silicic acid. Silicic acid also interferes in the extraction of neptunium in the next steps.) The residue is then dissolved in 15 ml. of 1 N nitric acid and 0.25 gm. of hydroxylamine hydrochloride are added to it. The solution is warmed for ten minutes and 2 ml. of 3 M ferrous sulphamate are added to it. After cooling to room temperature the solution is transferred to a 50 ml.

final step is more convenient for counting the beta-activity of Np^{239} .

TABLE II

Co-precipitation of Np^{239} with Zr-mandelate

No.	c.p.m. of Np^{239} added	c.p.m. of Np^{239} carried with Zr-mandelate
1	925	915
2	1063	1060
3	808	810
4	1320	1305

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X-RAY PATTERNS OF MOLLUSC SHELLS FROM INDIAN WATERS

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1. MATERIALS

IT is well known that the shells of molluscs consist of layers of calcium carbonate interspersed with protein layers and that the calcium carbonate occurs mostly as calcite or aragonite, and more rarely as vaterite.¹ Some studies have been made on this subject and are summarised by Jean Bouillon.² These, however, appear to have been made on specimens obtained from the temperate latitudes. The author is not aware of any reports of X-ray studies of the nature of the inorganic component in mollusc shells occurring in Indian waters. A study was therefore made of the X-ray diffraction patterns of a number of typical specimens obtained from the beaches of Madras State. The specimens studied are listed serially in Table I giving their class,

TABLE I

List of the shells studied with their identifications

Class—Gastropoda
Sub-class—Prosobranchia
(A) Order—Megagastropoda
(a) Series—Strombacea
Specimens 1 and 2 Strombidae, <i>Strombus</i> (A) (Two different species)
(b) Series—Cypraeacea
Specimens 3 and 4 Cypraeidae, <i>Cypraea</i> (A) ✓ (Two different species)
(c) Series—Cerithiacea
Specimen 5—Turritellidae, <i>Turritella</i> (A) ✓

TABLE I—Contd.

(B) Order—Stenoglossa
(a) Series—Buccinacea
Specimen 6—Volemiidae, <i>Hemifusus</i> (A)
Specimen 7—Fasciolaridae, <i>Fasciolaria</i> (A)
(C) Order—Archaeogastropoda
(a) Series—Zeugobranchia
Specimen 8—Haliotidae, <i>Haliotis</i> (A)
(b) Series—Trochacea
Specimen 9—Trochidae, <i>Trochus</i> (A)
(c) Series—Patellacea
Specimen 10—Patellidae, <i>Patella</i> (C) ✓
Class—Lamellibranchiata
(A) Order—Eulamellibranchiata
Sub-Order—Heterodonta
(a) Series—Veneracea
Specimen 11—Veneridae, <i>Meretrix</i> (A)
Sub-Order—Schizodonta
(a) Series—Unionacea
Specimen 12—Unionidae, <i>Unioninae</i> , <i>Lamellidens</i> (A)
(B) Order—Anisomyaria
(a) Series—Pectinacea
Specimen 13—Pectinidae, <i>Spondilinae</i> , <i>Spondylus</i> (C) ✓
Class—Cephalopoda
Sub-Class—Tetrabranchiata
Order—Nautiloidea
Specimen 14—Nautilidae, <i>Nautilus</i> (A) ✓

(C) = Calcite; (A) = Aragonite; ✓ = Also listed in Ref. 2.

sub-class (if any), order, family and genus. A fair number of specimens contained in this list have also been studied by Jean Bouillon.² These are indicated by a mark (✓) in Table I.