

PRODUCTION OF PLUTONIUM, YTTRIUM AND STRONTIUM TRACERS FOR USING IN ENVIRONMENTAL RESEARCH

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Abstract

Summary of cyclotron production methods of ^{237}Pu (45,2 d), ^{88}Y (106,65 d) and ^{85}Sr (64,84 d) tracers via nuclear reactions with protons and alphas on ^{235}U , ^{88}Sr and ^{85}Rb targets in wide energy range is given. Chemical methods of separation and purification of the tracers from the irradiated uranium, strontium and rubidium targets are described. The tracers were used for determination of Pu (239 – 240), Sr-90 and Am-241 in the samples (soil, plants, underground waters) from Semipalatinsk Test Site. Obtained results are discussed.

1 INTRODUCTION

One of the world largest Semipalatinsk Nuclear Test Site (STS) is located in Kazakhstan. There were produced more than 470 different nuclear explosions. In Kazakhstan there were produced also 40 nuclear explosions in peaceful applications.

At the time being research on radioecology situation of STS is one of the most important field of research for some institutions in Kazakhstan. For success of such kind research it is necessary to have the opportunity for obtaining of reliable analytical data on content of main doseproducing radionuclides-isotopes of Plutonium and Strontium-90 in different samples of environment. Such technique must be universal and must allow to determine radioisotopes in different samples such as water, soil, biomaterials. Significant stage of this technique is set of tracers. Tracers are isotope marks, that are absent in samples, to be analysed. Accuracy of measurements with application of tracers is about 2 – 5%. In the case when model radioactive solution is not available radioactive solutions of correspondent isotopes can be used. These solutions can be produced for example after dissolving of cyclotron targets in hydrochloric or nitrogen acid and purified from corresponding chemical impurities. Cyclotron isotopes are very specific according to nuclear characteristics and it is impossible to confuse them with controllable isotopes and products of their decay.

Exact determination of plutonium in general is impossible without tracer application, due to its complicated chemical behaviour, multiplicity of its oxidization state, inclination to colloidal formation.

Traditional methods of plutonium determination are based on application of radioactive alpha-emitting plutonium isotopes, mainly ^{236}Pu and ^{238}Pu as tracers for chemical yield control.

But these tracers can not be used for monitoring of all stages of radiochemical extraction including alpha sources preparation. Application of Pu-237 as radioactive indicator can solve this problem and it is especially effective at the development of this technique due to the peculiarities of its decay (gamma-radiation, characteristic X-ray radiation of neptunium daughter nucleus and appropriate half-life) Plutonium-237 with half-life of 45,2 days is decaying by electron capture on the base (79%) and excited states (21%) of neptunium daughter nucleus; gamma and X-ray radiation are observed in this case: $E_{\gamma}=59,5$ keV ($I_{\gamma}=3,3\%$); $K_{\alpha 1} - 101,1$ keV (19,45%); $K_{\alpha 2} - 97,1$ keV (12%); $K_{\beta 1} - 114,2$ keV (4,6%); $K_{\beta 2} - 117,7$ keV (92,4%) and others.

The traditional methods of strontium-90 content determination are based on the determination of the product isotope yttrium-90 after its accumulation during two weeks for achievement of equilibrium and stable yttrium application for determination of chemical yield.

At the same time ^{90}Sr and ^{90}Y are in the equilibrium state in the soils so by applying method based on the strait yttrium-90 extraction it is possible to reduce essentially analysis period. It is especially effective to use yttrium-88 as tracer for determination of chemical yield of yttrium-90. Additionally application of yttrium-88 makes it possible to avoid the application of traditional tedious and nonselective chemical methods for yttrium content determination.

2 KAZAKHSTAN ISOCHRONOUS CYCLOTRON AND RADIOISOTOPES PRODUCTION

Kazakhstan variable energy isochronous cyclotron with $K=50$ MeV was commissioned in 1972 [1].

At the cyclotron protons can be accelerated in the range from 6 MeV to 30 MeV, deuterons – from 12,5 MeV to 25 MeV, alpha-particles – from 25 MeV to 50 MeV, Helium-3 ions – from 18 MeV to 62 MeV.

It is a universal facility used both for nuclear science research and applied works in different adjoining fields. At present time one of the main applications of the

cyclotron is radioisotope production to meet the needs of the Republic of Kazakhstan. The main users of radioisotope products are Hospitals of Healthcare Ministry and also enterprises of metallurgy, mining, oil-chemistry, scientific institutes etc. For radioisotope production both external and internal targets, when high beam current is needed, are used.

The Institute has sufficient experience for production and delivery of "Thallium-210-chloride" radiopharmaceutical effectively used in cardiology. For the last years more than 5,5 Ci of the pharmaceutical was delivered to the medicine centers of the Republic and nearby regions and about 2,5 thousand patients were treated.

Technique of "Gallium-67-citrate" radiopharmaceutical production was developed. It is used for early diagnosis of oncology tumors and now it is prepared for clinical trials.

Radioisotopes of Co-57 and Cd-109 are also produced in the Institute and are used for check up of medicine cameras (cobalt) and in elemental analysis (cadmium). In the present work isotopes of plutonium were obtained in the reaction $^{235}\text{U}(\alpha, 2n)^{237}\text{Pu}$. Target of UO_2 with enrichment of U-235 (3,3%; 2,4%; 95%) was irradiated with α -particles of 50 MeV and 30 MeV. Yield of Pu-237 was equal to 11,2 kBq/ $\mu\text{A}\cdot\text{h}$ and 5,2 kBq/ $\mu\text{A}\cdot\text{h}$ in the case of maximal enrichment of the irradiated target. Isotope of yttrium was obtained in the reaction $^{88}\text{Sr}(p,n)^{88}\text{Y}$. Yield of Y-88 was equal to 2,8 MBq/ $\mu\text{A}\cdot\text{h}$. Target of SrO of natural isotope content was irradiated with 22MeV protons.

3 PU-237 RADIOCHEMICAL EXTRACTION FROM IRRADIATED URANIUM TARGET

Comparing different radiochemical techniques of plutonium separation from accompanying radionuclides (uranium, thorium and their decay products) for their selectivity and expressness it was concluded that the most reliable is extraction-chromatography method [2,3].

The method of plutonium fraction radiochemical extraction from irradiated uranium target includes irradiated target solution and plutonium fraction extraction with chromatography extraction method. It is presented in figure 1.

Pu-237 extraction from uranium target is carried out according to one of two variants: in the first one threeoctylamine (TOA) plated on Teflon with 0,5 – 1 mm fraction is used as extragent. TOA is the selective extragent for plutonium and it extracts anion complex of four-valence plutonium from 3M nitric acid solutions.

According to the second variant thenoyltrifluoroacetone (TTA) plated on polychrome with 0,5 – 1 mm fraction is used. Selective plutonium (IV) extraction is known to be in the form of intercomplex compounds with TTA.

Application of this extragent provides the efficient separation of plutonium and uranium because $\text{U}^{(\text{IV})}$ is slightly extracted if the acidity is more than 0,1 N nitric and hydrochloric solutions and at the same time maximal plutonium extraction by 0,15 – 0,4 M TTA solution in benzyl is observed in the range of 0,5 – 1 M HNO_3 concentration. The sorbents are prepared by extragent plating as much as 1 – 3 % to the carrier weight according to common method of stable phase plating on the carrier.

Thus the irradiated target is dissolved in the minimal volume of concentrated nitric acid, then the solution is evaporated up to dry, the precipitate is dissolved in 3M or 1 M solution of nitric acid depending on the type of applied extragent (TOA or TTA). For plutonium stabilization in the form of Pu (IV), Pu (III) is oxidized up to Pu (IV) by sodium nitride before sorption on the column (quantity as much as 30mg of the salt on 1 cm^3 of the solution) and is blown up with air during 10 – 15 min. After that the prepared solution is transmitted through the chromatography column with velocity of 1,5 – 2,0 cm^3/min (sorbent's weight is 1,5 – 1,8 g, diameter of the tube is 6 – 7 mm, working area height is 80 mm). Then the column is rinsed with three portions of nitric acid of correspondent concentration (3M or 1M HNO_3). Plutonium is eluted by using of recovering reextraction by transmitting fresh 1N solution of H_2SO_4 through the column. The elution containing Pu-237 is evaporated up to wet salt and is dissolved in 1 – 1,5 M HNO_3 . Concentration of plutonium alpha-isotopes in elution is determined by alpha-spectrometry sources; Pu-237 concentration is determined straightly in elution on gamma-spectrometry according to characteristic spectral lines. The technique of plutonium radioisotopes radiochemical extraction from UO_2 target, irradiated with alpha-particles, provides $\approx 90\%$ yield of desired isotopes from both variants.

4 Y-88 RADIOCHEMICAL EXTRACTION FROM IRRADIATED STRONTIUM TARGET

Yttrium fraction is extracted from irradiated strontium target by chromatography extraction method with the use of bis(2-ethylhexyl)-phosphate acid ($\epsilon\text{G-Nr}$ 206054, MERCK Schuhadat, 98%) [4]. Chromatography column, filled with polychrome-1 coated by 1M solution of bis(2-ethylhexyl)-phosphate acid in chlorinated carbon, is used. The irradiated target is dissolved in 15 cm^3 of hydrochloric acid (1:2), evaporated up to dry and diluted up to 20 cm^3 by 0,5 M solution of hydrochloric acid and then the solution is transmitted through the column at the velocity of 0,8 – 1 cm^3/min . After finishing the process the column is rinsed with three portions (each is 20 cm^3) of 0,5 M of hydrochloric acid and Y-88 is eluted with 20 cm^3 of 6 M of hydrochloric acid. Y-88 concentration in

elution is determined with the help of γ -spectrometry sources and yttrium-88 precipitation in 5 N fluorine acid with addition of stable yttrium. This method of yttrium-88 radionuclide extraction from SrO target irradiated with protons provides $\approx 95\%$ yield.

In the case of strontium preliminary extraction, for example when water sample is analysed, strontium-85 is

especially effective. The technique of this tracer production was developed using Sr-85, being obtained via reaction $^{85}\text{Rb}(p,n)^{85}\text{Sr}$. RbO and $\text{Rb}(\text{NO}_3)_2$ of natural content were used as targets. Yield of Sr-85 was equal to 2,1 MBq/ $\mu\text{A}\cdot\text{h}$ and 0,7 MBq/ $\mu\text{A}\cdot\text{h}$.

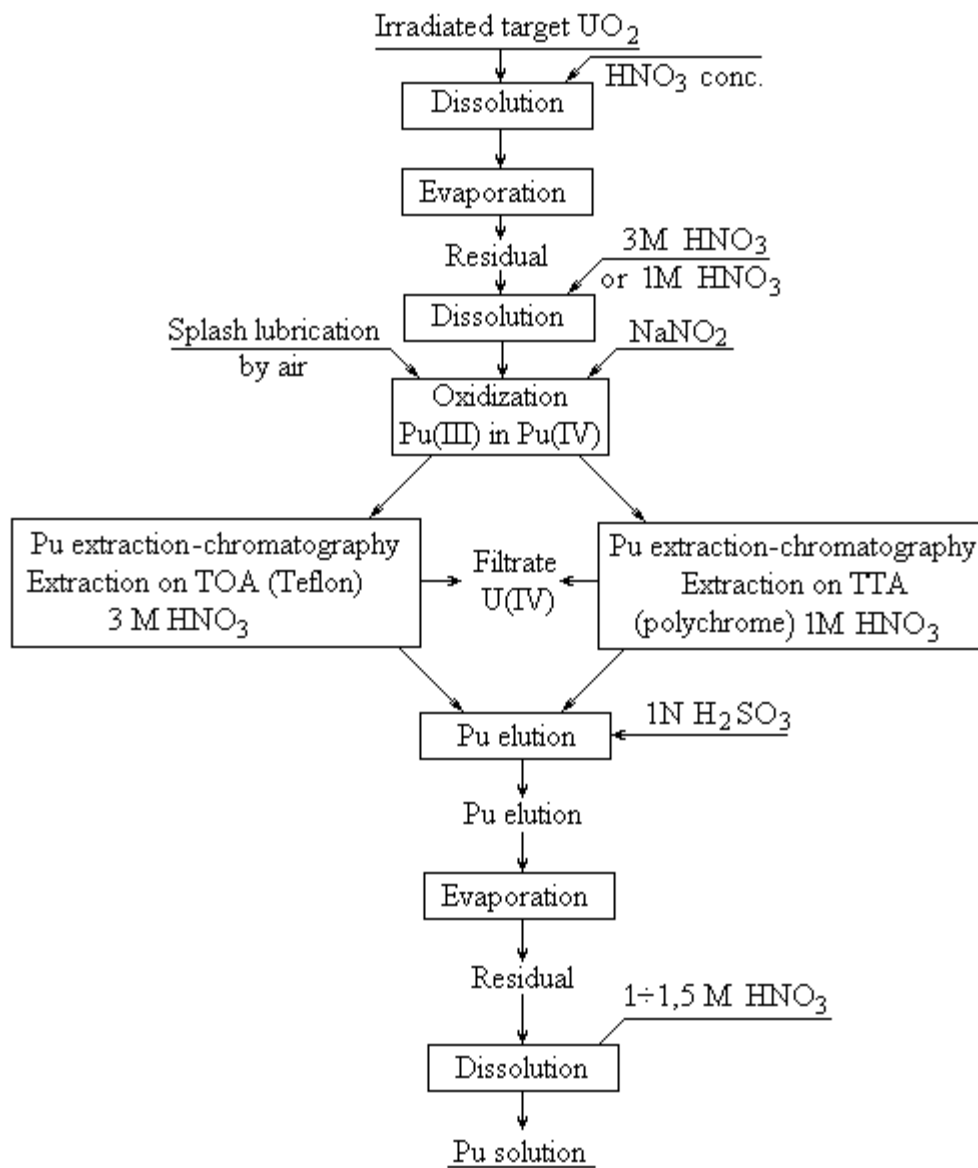


Figure 1: The scheme of uranium target radiochemical processing with plutonium fraction extraction.

5 CONCLUSIONS

Obtained Pu-237, Y-88 and Sr-85 tracers were used for development of technique for determination of Pu(239 – 240), Sr-90 and Am-241 in samples from Semipalatinsk Test Site (soil, plants, water) and also for determination of

plutonium and strontium in soil near underground cavities, produced with nuclear explosions.

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