First chemical on-line separation and detection of a subsecond $\alpha$-decaying nuclide, $^{224}$Pa

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Subsecond $^{224}$Pa ($T_{1/2}=0.85\,s$) was produced via the $^{209}$Bi($^{19}$O,$n$)$^{224}$Pa reaction at the 88 inch cyclotron at the Lawrence Berkeley National Laboratory. After production it was transported via a gas-jet system to the centrifuge system SISAK.3 Following on-line extraction with triethylamine/scintillation solutions from 1M lactic acid, $^{224}$Pa was detected applying on-line $\alpha$-liquid scintillation counting. Unambiguous identification was achieved using time-correlated $\alpha$-decay chain analysis. This constitutes the first chemical on-line separation and detection of a subsecond $\alpha$-decaying nuclide, 0.85-s $^{224}$Pa with the fast extraction system SISAK.3.

Introduction

The development of automated techniques for studying the chemical behavior of very short-lived isotopes plays an important role in the investigations of the chemical properties of the transactinides. Among the problems inherent in studies of the transactinides are their small production cross sections, which range from nanobarns to picobarns and their decay predominantly by $\alpha$-emission and spontaneous fission (SF). Therefore, separation techniques are required which are very fast, highly efficient, and reach equilibrium quickly. In addition, a method for rapid transport of the separated activities to a system for measurement of $\alpha$-decay is required. On-line gas-chromatography and ion exchange chromatography have been used for rapid chemical separations for many years (see Wierczynski et al.1,4 and Trautmann2 for reviews). However, for the aqueous systems, the problem of rapid detection of the $\alpha$-emitters has not been solved. The time-determining step is drying the samples so they can then be placed in detectors for measurement. During recent years, many studies have also been carried out to explore the possibility of using the fast centrifuge system SISAK.3 (Short-lived Isotopes Studied by the AKUFVE technique)3 in combination with an on-line Liquid Scintillation System4 — which was later called LISSY — to perform on-line liquid-liquid extractions of short-lived $\alpha$-decaying nuclides and study their chemical properties.5,6 SISAK-LISSY provides the capability for chemically separating and then detecting short-lived $\alpha$-decaying isotopes in a flowing organic solution.

Before the combination of SISAK 3 and LISSY can be applied to the investigation of transactinide elements it has to be shown that the system is capable of separating and detecting short-lived $\alpha$-decaying nuclides. In a first step SISAK3-LISSY was used to identify $\alpha$-decaying $^{164}$Hf ($T_{1/2}=18\,s$).4 The next step was the positive identification of a subsecond $\alpha$-decaying nuclide. For this purpose $^{224}$Pa ($T_{1/2}=0.85\,s$) was chosen which follows an $\alpha$-decay chain to $^{220}$Ac, $^{216}$Fr, $^{212}$At and finally to $^{208}$Bi (Fig. 1). The half-life of $^{224}$Pa is comparable with the shortest-lived $\beta$-decaying nuclides, 1.0-s $^{110}$Tc and 0.8-s $^{113}$Ru, that have been

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Fig. 1. Decay chain of $^{224}$Pa

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investigated with SISAK 3 so far.\textsuperscript{8,9} The production cross section of the reaction $^{209}\text{Bi}(^{18}\text{O},3n)^{224}\text{Pa}$ is high enough to produce a reasonable number of atoms, and the decay chain allows time-correlated $\alpha$-decay chain analysis.

**Experimental**

**Target chamber and the helium gas transport system at the 88-inch cyclotron**

The target chamber and the gas transport system at the Lawrence Berkeley National Laboratory (LBNL) 88-inch cyclotron have been described previously.\textsuperscript{7,10} A Faraday cup at the beam stop measures the current and the integral beam dose is recorded. A graphite-lined recoil chamber was used in this experiment to reduce the $\beta\gamma$-background.

**Production of $^{224}\text{Pa}$**

The $^{224}\text{Pa}$ was produced at the LBNL cyclotron via the $^{209}\text{Bi}(^{18}\text{O},3n)$ reaction with $^{18}\text{O}^{5+}$ projectiles of 111 MeV (lab system). The target was 3.3 mg/cm\textsuperscript{2} $^{209}\text{Bi}$ on Be foil and was the same as that used by Wilkie et al.\textsuperscript{7} who measured the cross section and have given details of the production setup. The transport efficiency of the gas-jet system was determined with a molybdenum catcher foil. The foil was mounted directly after the target to catch the recoiling reaction products. The amount of $^{211}\text{Po}$ ($T_{1/2}=516$ ms), produced by transfer reactions of $^{22}\text{Ne}$ with trace lead impurities present in the target, in secular equilibrium with $^{211}\text{At}$ ($T_{1/2}=7.2$ h) was determined and compared with the corresponding amounts measured after transport through the system.\textsuperscript{7}

**SISAK 3 and LISSY**

SISAK 3 is an on-line liquid extraction system which has been used for many years to study the chemical and nuclear properties of $\beta\gamma$-decaying nuclides with half-lives as short as one second.\textsuperscript{3} In combination with LISSY, which is capable of measuring $\alpha$-decaying nuclides in flowing organic solution, SISAK 3 can also be used to study short-lived $\alpha$-decaying nuclides. Identification of $^{224}\text{Pa}$ was achieved by measuring time-correlated $\alpha\alpha$-decay chains. It has been shown earlier that protactinium behaves chemically similar to the group 5 elements niobium and tantalum. It can be extracted in high yield from lactic acid or $\alpha$-hydroxyisobutyric acid using trioctylamine (TOA).\textsuperscript{11} In the present experiment 1M lactic acid was used as aqueous solution and 0.046M TOA was added to a liquid scintillation cocktail containing dimethyl-POPOP and naphthalene in toluene.\textsuperscript{4} This extraction procedure requires just one extraction stage, but an additional extraction step with 1M Cyanex 471X (triisobutylphosphine sulfide)\textsuperscript{12} was performed to remove Po contamination. Figure 2 shows the experimental setup for the production, separation and measurement of $^{224}\text{Pa}$. Flow rates of about 1 ml/s were used in SISAK 3 to separate protactinium from the other reaction products. In this experiment centrifuges made of PEEK (polyetheretherketone) were run at 20,000 rpm.

After the extraction step, the scintillation solution containing protactinium entered the first detection cell of LISSY which is a 4.8 ml "meander" cell made of Teflon facing a photomultiplier tube. Altogether three detection cells were used. An energy resolution of 350 keV full width at half maximum (FWHM) for an $\alpha$-energy of 6.8 MeV was achieved.

![Fig. 2. The first centrifuge system SISAK 3 in combination with the on-line $\alpha$-liquid scintillation counting system LISSY as used for studies of $^{224}\text{Pa}$. D1 and D2 are degassing centrifuges; C1 and C2 are extraction centrifuges; LSC 1 to LSC 3 are LISSY detection cells.](image-url)