Progress towards the production of the ^{236g}Np standard sources and competing fission fragment production

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HIGHLIGHTS

- The isobaric distribution of fission residues.
- Decay analysis of thirteen isobarically distinct fission residues.
- Stoichiometric abundances were calculated via the determination of absolute activity concentrations.
- This technique was validated by computational modelling of likely sequential decay processes.
- The results were largely in agreement with previously published values for neutron bombardments on ^{238}U at energies of 14 MeV.

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ABSTRACT

The isobaric distribution of fission residues produced following the bombardment of a natural uranium target with a beam of 25 MeV protons has been evaluated. Decay analysis of thirteen isobarically distinct fission residues were carried out using high-resolution γ-spectrometry at the UK National Physical Laboratory. Stoichiometric abundances were calculated via the determination of absolute activity concentrations associated with the longest-lived members of each isobaric chain. This technique was validated by computational modelling of likely sequential decay processes through an isobaric decay chain. The results were largely in agreement with previously published values for neutron bombardments on ^{238}U at energies of 14 MeV. Higher yields of products with mass numbers $A \sim 110$–130 were found, consistent with the increasing yield of these radionuclides as the bombarding energy is increased.

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1. Introduction

The radionuclide ^{236}Np is of interest both as a long-lived odd–odd radionuclide and as a chemical yield tracer for the radiochemical analysis of ^{237}Np (Jerome et al., 2014). Neptunium-237 is significant in a number of fields of research such as nuclear forensics, environmental analysis and the nuclear fuel cycle. However, investigation of ^{237}Np is complicated by there being no known stable radionuclide of neptunium. Although various radioactive tracers have been used, including ^{235}Np, ^{239}Np and ^{236}Pu (Jerome et al., 2014), n none have been found to be satisfactory. Neptunium-236 is a potential candidate as a neptunium yield tracer, as its $1.55(8) \times 10^5$ years radioactive half-life (Chechev and Kuzmenko, 2007) allows it to be used for both radiometric and mass spectrometric measurements. One method of ^{236}Np synthesis that has shown promising results is to bombard ^{nat}U with protons (Jerome et al., 2014). This production method yields a variety of other radionuclides as a result of the high-energy proton-induced fission of ^{nat}U; however there is a scarcity of information regarding the fission yields produced by proton bombardments of this energy on natural uranium targets. In the current work, we present the mass distribution of the fission fragment residues produced following the proton irradiation of natural uranium which were formed in parallel with the production of ^{236}Np.

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2. Materials and methods

2.1. Target irradiation and sample preparation

A target containing 1 g of uranyl nitrate was produced by compacting ^{nat}U at 60 kN cm^{-2} between an aluminium target holder and a 0.025 mm titanium foil. The target was irradiated at the University of Birmingham UK cyclotron for a total duration of 18.75 h over a period of 3 days (9–11 December 2013), with an incident proton energy of 25 MeV, using an average on-target beam current of approximately 20 μA. Directly following the irradiation, the target was left for approximately 20 days, thereby allowing the short-lived fission products and their decay daughter residues (i.e., those with $T_{\frac{1}{2}} < 2$ days) to reduce significantly their specific activities. The radiochemical processing of the target was carried out at NPL with the irradiated target of ^{nat}UO$_2$(NO$_3$)$_2 \cdot$ 6H$_2$O being dissolved in nitric acid and converted to an 8 M nitric acid solution. A 1 g aliquot of the solution was then dispensed to a 2 mL glass British Standard (BS795:1983) ampoule for measurement by γ-spectrometry.

2.2. Gamma spectrometry

The LN$_2$ cooled high purity germanium (HPGe) γ-spectrometer 'BART' was used to perform a series of measurements of the prepared sample, the details of the detector specifications and the full-energy peak efficiency calibration have been previously described in Collins et al. (2014). The sample was measured twelve separate times for a typical counting time of 50,000 s over the course of several months. The net peak areas of the discrete γ-emissions associated with specific fission fragment decays were calculated using the Canberra Genie 2000 v2.1c software. The fitted photopeaks were reviewed with the Interactive Peak Fit tool and adjusted where required. The photopeak net peak areas were corrected for background, integrated radioactive decay, and pulse pile-up. No corrections were applied for true coincidence summing as the samples were measured at a source-to-detector window distance of 30 cm (solid angle=0.45 sr), reducing any coincidence summing to a level which was insignificant compared to the other sources of measurement uncertainty.

2.3. Use of the Bateman equations for elemental activity distribution simulations

The determination of the abundances of each isobaric chain produced by the proton-induced fission of ^{nat}U is complicated by the initial population of a range of elements for each isobaric chain in the fission process. The neutron-rich radionuclides produced directly following the fission events typically have radioactive half-lives in the range of seconds to hours; this means that such radionuclides have undergone sufficient radioactive decays to bring their abundances below the minimum detectable activity by the time of the initial γ-spectrometry measurement in the current work. The decay of fission products through their respective mass chains towards stability is generally accompanied by increasing half-lives as the line of stability is approached. Many of the isobaric decay chains from $A \approx 80$ to ≈ 160 decay through well-defined radionuclide species with characteristic γ-emissions following their decay by β$^-$-emission to excited states in the final, long lived daughter nucleus of that particular mass chain. These penultimate members of the isobaric fission fragment decay chains can have sufficiently long half-lives (days to months) to be a useful monitor of the decay activity in the present work.

The total number of atoms produced in a given mass chain can be estimated without prior knowledge of the initial elemental (Z) distribution of the members within that mass chain following

the initial fission using the assumption that the decay lifetime of the ultimate decay in the chain is significantly longer than each preceding decay half-life.

The basis of any such solution of a decaying nuclear chain must take into account the Bateman equations of radioactive decay (Bateman, 1910) for that chain. The number of atoms of the first radionuclide (N_1) within the series is given by the normal exponential decay equation:

$$N_1(t) = N_1(0)e^{-\lambda_1 t} \tag{1}$$

where $N_1(t)$ is the number of atoms at time t, $N_1(0)$ is the initial number of atoms of when $t=0$ and λ_1 is the decay constant. To model the system, the number of atoms present and their rate of change with respect to time (i.e. the activity) needs to be calculated.

Under the assumption that the parent radionuclide in the chain has a fixed amount of atoms, $N_1(0)$ at the effective time zero ($t=0$) and no further production of the parent radionuclide takes place, then the activity associated with this radionuclide is given by

$$\frac{dN_1}{dt} = \frac{d}{dt}N_1(0)e^{-\lambda_1 t} = -\lambda_1 N_1(0)e^{-\lambda_1 t} = -\lambda_1 N_1(t) \tag{2}$$

If N_1 decays exclusively to a daughter radionuclide (N_2) then the rate of change of number of atoms of N_2 is given by

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \tag{3}$$

Similar arguments hold for the following nuclei up to and including the terminal radionuclide (N_n):

$$\frac{dN_n}{dt} = \lambda_{n-1}N_{n-1} - \lambda_n N_n \tag{4}$$

If the nth member of the decay chain represents a radioactively stable species (i.e. $\lambda_n N_n = 0$), then $dN_n/dt = \lambda_{n-1}N_{n-1}$.

A decay simulation was produced based on the matrix based algebraic approach to the Bateman equations given by Moral and Pacheco (2003), detector efficiency and γ-emission probability so that the effect of varying the initial distribution of radionuclides on the strength of the γ-emissions could be measured. It was established that the activity of the longest-lived member in a chain could be related directly to the initial number of atoms at the effective zero time of the irradiation, provided the previous members in the chain were short-lived by comparison. This is indeed the case for most isobaric decay chains between $A \sim 80$–160 due to the time it takes for the more neutron-rich, shorter-lived ($T_{1/2} \approx$ seconds to minutes) members of a particular mass chain to decay through to the longest-lived member being negligible with respect to the half-life of the long-lived member, i.e. that closest to stability which can have $T_{1/2} \approx$ days to months. This approximation overestimates the initial number of atoms by an amount dependent on the initial distribution of the atoms within a particular mass chain. The largest overestimation occurs when the initial atoms in the mass chain are all formed in the chain member with the highest neutron-to-proton ratio (i.e. most neutron-rich). The majority of the mass chains were found to have maximum overestimations of 0.1%. Since the initial elemental distribution was not established, the current work makes the gross assumption of a 'rectangular distribution' of the initial elemental population (i.e., an equal elemental production for all isobaric chains of neutron-rich nuclei that there is half-life data for). This was fed into the computational calculation and the overestimation (or underestimation) used to calculate the correction factor to be applied to the measurements. For example, in the case of ^{95}Zr the initial ratio of ^{95}Kr:^{95}Rb:^{95}Sr:^{95}Y:^{95}Zr was assumed to be 1:1:1:1:1 and the decay of this system then modelled. This was used to calculate a