

Rapid communication

An EC-branch in the decay of 27-s ^{263}Db : Evidence for the isotope ^{263}Rf

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Summary. 27-s ^{263}Db was produced in the ^{249}Bk (^{18}O , $4n$) reaction at 93 MeV. The activity was transported by a He/KCl-jet to the laboratory where it was collected for 15 min and then subjected to a chemical separation specific for group-4 elements. The activity was dissolved in 0.5 M unbuffered α -HiB and eluted from a cation-exchange column. The effluent was made 9 M in HCl and group-4 tetrachlorides were extracted into TBP/Cyclohexane which was evaporated to dryness on a Ta disc. The Ta discs were assayed for α and SF activity. A SF activity with a half life on the order of 20 min was observed and assigned to the nuclide ^{263}Rf . It is formed by electron-capture decay of ^{263}Db with a decay branch of $3^{+4}_{-1}\%$.

Introduction

Attempts to discover the isotope ^{263}Rf date back about 10 years when K. R. Czerwinski [1] searched for it in the ^{248}Cm (^{18}O , $3n$) reaction at 92.5 MeV using liquid-liquid extraction of rutherfordium from 0.05 M HCl into 0.5 M thenoyltrifluoroacetone in benzene. In 300 3-min collections and separations, 7 spontaneous fission (SF) events were observed with a half life of 500^{+300}_{-200} s corresponding to a cross section of 140 ± 50 pb. No α particles attributable to the decay of ^{263}Rf were found.

The isotope 27-s ^{263}Db was discovered in the ^{249}Bk (^{18}O , $4n$) reaction at 93 MeV by eluting element 105 (dubnium) from cation exchange columns in unbuffered 0.05 M α -hydroxy isobutyric acid (α -HiB) [2]. ^{263}Db decays predominately by SF (57%) and by α decay (43%) and was produced with a cross section of 10 ± 6 nb. An electron-capture

(EC) decay branch could not be excluded. Therefore, a search for an EC-branch in the decay of 27-s ^{263}Db was performed in a milking experiment: ^{263}Db was again produced in the ^{249}Bk (^{18}O , $4n$) reaction and transported to the Automated Rapid Chemistry Apparatus (ARCA) [3] by a He/KCl jet. After 1 min collections, the activity was dissolved in 0.05 M α -HiB and eluted in that solution from a cation exchange column. The same column was used for 5 subsequent collection-elution cycles with ^{263}Db . After the fifth elution, an elution of tetravalent ions including those of rutherfordium, if present, was performed with 0.1 M HF [4]. In 155 of such rutherfordium fractions, a total of 22 SF events was observed. From the known α /SF ratio for Fm isotopes and from the Fm contained in the measured α -particle spectra, 8.8 of the SF events had to be attributed to ^{256}Fm . A two-component fit with the ^{256}Fm fixed yielded a half life of 600^{+300}_{-200} s for ^{263}Rf . Based on the effective production cross section and on the known cross section for production of ^{263}Db at 93 MeV, 10 ± 6 nb, an EC-branch in the decay of ^{263}Db on the order of 5% was deduced [5]. Two α particles at 7.9 MeV with life times compatible with a half life of about 10 min were discussed as possibly being associated with the decay of ^{263}Rf giving an upper limit for the α -decay branch of 30%. The effective production cross section for ^{263}Rf via EC decay of ^{263}Db is thus significantly higher than that in the ^{248}Cm (^{18}O , $3n$) reaction.

A search for ^{263}Rf in the ^{248}Cm (^{22}Ne , $\alpha 3n$) reaction at 122 MeV by Dressler *et al.* [6], involving a chemical separation of Rf as the volatile tetrachloride, yielded two α particles at 7.8 and 7.9 MeV and four SF events with very long life times, of which 1 ± 0.5 had to be assigned to ^{256}Fm . A cross section of $0.9^{+0.9}_{-0.4}$ nb was estimated. The long life times were tentatively interpreted as being associated with the decay of 95-min ^{259}Md which is formed if there is an α -decay branch in the decay of ^{263}Rf through electron capture of the daughter, ^{259}No . Another search using the same reaction and aqueous chemistry with fluoride complexes of

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Rf [7] yielded two α events near 7.9 MeV with unusually long life times. According to the empirical α half-life systematics by Hatsukawa *et al.* [8], an α energy of 7.9 MeV for ^{263}Rf correlates with a half life of 1800 s. Neither of these experiments [6, 7] was considered to present conclusive evidence for ^{263}Rf .

We have attempted to add further evidence for an EC-branch in the decay of ^{263}Db and for ^{263}Rf in a new series of experiments producing again ^{263}Db in the ^{249}Bk (^{18}O , $4n$) reaction at 93 MeV. A two-step chemical separation scheme was used consisting of the successful cation-exchange separation with α -HiB and a liquid-liquid extraction of the group-4 elements from 9 M HCl into tributylphosphate (TBP)/cyclohexane yielding an improved decontamination of the rutherfordium fraction from actinides. We report here on the results of these experiments which are consistent with our previous evidence [5].

Experimental

Chemical separations

The activity delivered by a He/KCl gas jet was collected for 15 min (sufficiently long for 27-s ^{263}Db to decay) on a Ta disc by impaction. The KCl spot was dissolved by $2 \times 20 \mu\text{L}$ of unbuffered 0.5 M α -HiB containing ^{88}Zr tracer and pipetted on top of a $3 \times 50 \text{ mm}$ glass column thermostated at 50°C containing the cation-exchange resin AG 50 W \times 8, minus 400 mesh. The column was previously preconditioned with 0.5 M α -HiB. The group-4 elements were eluted from the column in $2 \times 500 \mu\text{L}$ 0.5 M α -HiB and collected in a centrifuge cone. The eluate (1 mL) was combined with 3 mL concentrated HCl to yield a solution being 9 M in HCl. This was contacted for about 20 s with 200 μL of a 20 vol. % TBP/cyclohexane solution for liquid-liquid extraction. The phases were separated and the organic phase was evaporated to dryness on a Ta disc. The Ta discs were assayed for α and SF activity starting about 8 min after the end of collection. The chemical yields were determined *via* the ^{88}Zr activity by γ -ray spectroscopy for each sample and were 65% on the average. The decontamination factor from trivalent metal ions determined with lanthanide tracers was on the order of 15 000.

Production and transport

^{263}Db was produced in the reaction ^{249}Bk (^{18}O , $4n$) at the Philips Cyclotron of the Paul Scherrer Institute (PSI) Villigen, Switzerland.

The $478 \mu\text{g}/\text{cm}^2$ target was prepared by electrodeposition of $\text{Bk}(\text{NO}_3)_3$ on Be. After the plating process, the nitrate was converted into the oxide by heating. It was bombarded with average $^{18}\text{O}^{5+}$ beam intensities of $2.7 \times 10^{12} \text{ s}^{-1}$.

The 123.1 MeV ion beam passed through a $15.8 \mu\text{m}$ Be window, through 2 mm N_2 cooling gas, a $13.0 \mu\text{m}$ Be degrader foil, another 2 mm of N_2 cooling gas, and through the $14.8 \mu\text{m}$ Be target backing, entering the target material with a projectile energy of 93.5 MeV, and leaving the target with 93.0 MeV.

The target contained 80 to 100 $\mu\text{g}/\text{cm}^2$ Tb as upper layer to simultaneously produce Ta isotopes (notably ^{172}Ta) that

were used to perform yield checks by γ -spectroscopy. Reaction products recoiling out of the target were stopped in He gas containing KCl aerosols generated by sublimation from solid KCl at 660°C . The aerosols together with the recoil products were transported through a steel capillary (2 mm inner diameter, length 50 m) to the collection site. The transport plus deposition yield was $38 \pm 11\%$ on the average at a He flow of about 2 L/min.

Detection of Rf

α -particle and SF-decay events were detected using sixteen 450 mm^2 passivated implanted planar silicon (PIPS) detectors. At a collection and separation cycle time of 15 min, the counting time for each sample was 240 min. List mode data were stored for each sample and decay event, including energy, detector number, and time after start of the measurement. The detection efficiency for an α particle was 30%, for a SF event 60%. After the end of counting, each individual sample was monitored by γ -spectroscopy to determine the chemical yield *via* the ^{88}Zr activity.

Results

The summed α -particle energy spectrum obtained in 225 experiments is shown in Fig. 1. The data for three additional experiments were omitted as there was obviously a small Fm contamination. Two of these omitted sets of data also contained a long-lived SF event. As in the preceding experiments [1, 5–7], there is a peak at 7.68 MeV due to the decay of the naturally occurring ^{214}Po (a member of the ^{238}U decay chain) that prevents detection of a possible α -decay branch

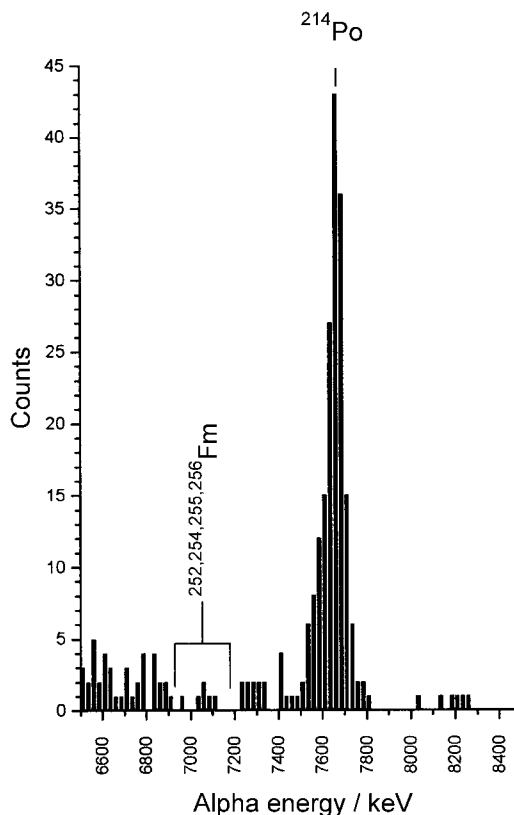


Fig. 1. Summed α -energy spectrum from 225 experiments.

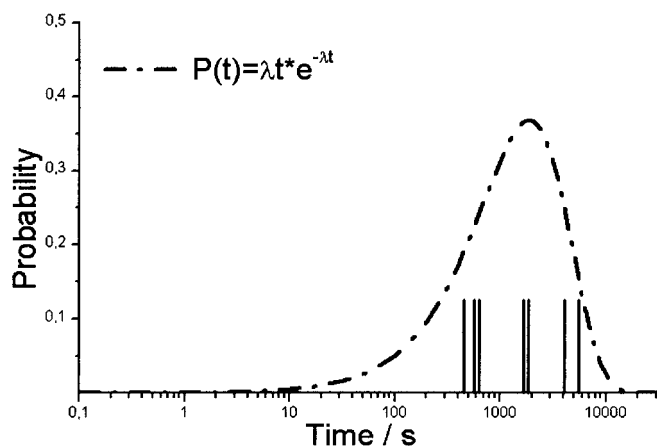


Fig. 2. Time distribution of the SF events observed in 225 experiments. The solid line is the probability distribution for a half life of 24 min.

of ^{263}Rf near that energy and also prevents detection of its potential α -decay daughter, ^{259}No (7.47–7.69 MeV) which could help to determine the α -decay branch in the decay of ^{263}Rf . No α -decay events at 7.9 MeV were observed. Above 8 MeV, a number of spurious events was observed with life times much too long to be considered as α particles.

A total of 9 SF events was registered of which 2 have to be considered a long-lived electronic background. The remaining 7 events are compatible with a half life of 24^{+19}_{-7} min. Fig. 2 shows the probability distribution for a half life of 24 min together with the experimental life times. By taking the jet yield into account, the chemical yield, the counting efficiency, the target thickness, the beam intensity, we obtain for the 7 events an effective production cross section of 290^{+140}_{-50} pb. With the measured production cross section for ^{263}Db at 93 MeV of 10 ± 6 nb, this results in an EC-branch in the decay of ^{263}Db of $3^{+4}_{-1}\%$ for a half life of ^{263}Rf of 24^{+19}_{-7} min which is fully consistent with our previous evidence [5]. This number ignores any α -decay branch in the decay of ^{263}Rf . An upper limit on the order of 30% [5] would increase the EC-branch to 4%.

From the number of α -events in the energy interval 6.9 MeV and 7.2 MeV, bracketing the decay energies of $^{252-256}\text{Fm}$ and from measurements of α spectra of unseparated samples, we can estimate a decontamination factor of 9000 for Fm. As there are no peaks at 7.0 MeV ($^{252,255}\text{Fm}$) and at 7.2 MeV (^{254}Fm) in Fig. 1, and in view of the decontamination factor of 15000 determined with lanthanide tracers, the value of 9000 might have the character of a lower limit.

Discussion

In a series of experiments in which the isotope 27-s ^{263}Db was produced and was allowed to decay, we have performed chemical separations specific for group-4 elements consisting of an elution by 0.5 M α -HiB from cation-exchange columns and a subsequent extraction into TBP/Cyclohexane giving a decontamination from fermium on the order of 10^4 . As in similar, previous experiments [1, 5], a SF activity with a relatively long half life is observed. This cannot be attributed to a long-lived isomeric state in ^{263}Db as the SF

activity follows a specific group-4 chemical separation involving an extraction step which would eliminate dubnium. Dubnium, in strong HCl, is known [9] to form anionic chlorocomplexes which would not be extracted by TBP, known to extract only neutral species. Thus, from the chemical evidence, we must conclude that the SF activity must be associated with rutherfordium. The most natural explanation is the isotope ^{263}Rf which is likely to have been produced in the $^{248}\text{Cm} (^{18}\text{O}, 3n)$ reaction in [1] and which seems to be the EC-decay daughter of ^{263}Db with an EC-branch of about 3%. Here, the previous evidence [5] and the result of this work are fully consistent. Note that, in the present experiment, the existence of an EC branch in the decay of ^{263}Db producing ^{263}Rf cannot be distinguished from the direct production of ^{263}Rf in the $^{249}\text{Bk} (^{18}\text{O}, p3n)$ reaction. This holds for our previous experiments [5] as well. However, calculations with the fusion-evaporation code HIVAP predict a $p3n$ -cross section on the order of 0.02 nb only, which is 15 times lower than the effective cross section determined in the experiment. Thus, production of ^{263}Rf in the $^{249}\text{Bk} (^{18}\text{O}, p3n)$ reaction cannot be ruled out, however, its production through EC decay of ^{263}Db appears to be more likely. According to the 1993 Atomic Mass Evaluation of Audi and Wapstra [10], the Q_{EC} value of ^{263}Db is 2.4 MeV. The ^{263}Db partial EC-decay half life of 675 s gives a $\log(ft)$ of 4.3. Thus, there is growing evidence for a small EC-branch in the decay of ^{263}Db through which the new isotope ^{263}Rf is formed. ^{263}Rf has a relatively long half life of tens of minutes and decays predominantly by SF. The long half life is presumably due to the proximity of ^{263}Rf with $N = 159$ to the deformed neutron shell $N = 162$. Quantitative statements about its possible α -decay branch are difficult to make. The absence of α events at 7.9 MeV in [1] and in the present work let the possible assignment of two events at 7.9 MeV to the decay of ^{263}Rf [5] appear unlikely. The work of [1, 5], and the present work, together, would give an α branch at 7.9 MeV of 12% only. This corresponds to a partial half life of 4 hours which is too long to be consistent with systematics [8]. If the α energy is between 7.5 and 7.8 MeV, any α events from the decay of ^{263}Rf would be masked by α events from ^{214}Po . If the α energy is between 7.5 and 7.8 MeV, a partial half life between 60000 s and 4000 s would result [8]. If the total half life of ^{263}Rf is 10 (24) min this would give α branches of 1 (2)% at 7.5 MeV and 15 (33)% at 7.8 MeV. These considerations show that an

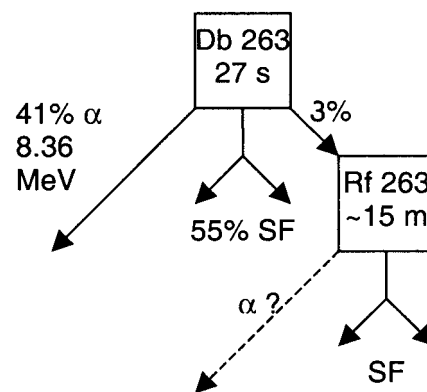


Fig. 3. Decay scheme for ^{263}Db and ^{263}Rf .

α branch in the decay of ^{263}Rf is likely to be small. The picture that emerges both from [5] and from the present work is shown in Fig. 3.

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