Cluster emission in the radioactive decay of ^{223}Ac

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Abstract
The branching ratio of ^{223}Ac decay by spontaneous ^{14}C emission was measured and a search for ^{15}N clusters was performed. After exposure of a hemispherical array of solid-state nuclear track detectors, 347 ^{14}C events were identified and no ^{15}N events. $B(^{14}C) = \lambda(^{14}C)/\lambda(\alpha) = (3.2 \pm 1.0) \times 10^{-11}$ is consistent with a favoured ground state to ground state transition. As no nitrogen tracks were found, only an upper limit could be inferred for ^{15}N emission, $B(^{15}N) = \lambda(^{15}N)/\lambda(\alpha) \leq 2.2 \times 10^{-13}$ (confidence limit 90%), consistent with an unfavoured transition. Intense ^{227}Pa sources were produced for this study, using the reaction ^{232}Th(p,6n)^{227}Pa. This offered an opportunity to compare the measured source strength with predictions based on published excitation function data.

1 Introduction

Since the discovery of ^{14}C radioactivity of ^{223}Ra by Rose and Jones [1] about 25 years ago, the spontaneous emission of neutron-rich clusters, ranging from ^{14}C to ^{34}Si, have been observed in the radioactive decay of various trans-lead nuclei [2]. A rather intense period of searching for suitable cases of this rare phenomenon lasted about 15 years, given that 18 combinations of "parent nuclei/observed clusters" were known by circa 1993, 22 cases by 1999 and 23 cases to date. The sharp decrease in new results during the last decade is largely because of the increasing difficulty to perform new experiments aimed at discovering transitions with continually decreasing branching ratios. While the use of solid-state nuclear track detectors (SSNTD) revolutionized the detection of rare heavy clusters in a background dominated by an alpha particle flux that is many orders of magnitude higher, a practical limit of detection does exist with SSNTD techniques. Although the "threshold" characteristics of these detectors cause them to be immune to the tracks made directly by the low-ionizing alpha particles, they are nevertheless weakly sensitive to the recoils produced when heavier atoms are being struck by alpha particles [2]. This sets an upper limit to the integrated alpha particle flux that can be tolerated, beyond which the detector loses sensitivity. This complication can be largely avoided and/or compensated for by increasing the total detector surface area, which, however, increases the size of the experiment and the human effort required to scan the larger detector surface for cluster events. Also, there are practical difficulties producing the very intense sources that prospective new experiments may require. For these reasons, the lowest branching ratio relative to alpha decay that could be determined experimentally to date is between 10^{-17} and 10^{-18}.

The predicted cluster radioactivity of ^{223}Ac has, ironically, eluded experimental verification for many years. The reasons for this will be discussed in due course. In principle, it should be one of the "easier" cases as the expected branching ratio for ^{14}C emission is relatively high, of the order 10^{-11}.

*Deceased

Furthermore, the reasons why ^{223}Ac should be a particularly interesting nucleus to study for its cluster radioactive decay modes are compelling. Firstly, two modes of cluster radioactivity for this nucleus are predicted: $^{223}Ac \rightarrow {}^{14}C + {}^{209}Bi$ (Q = 33.08 MeV) and $^{223}Ac \rightarrow {}^{15}N + {}^{208}Pb$ (Q = 39.49 MeV). Secondly, it is the isobaric analogue of ^{223}Ra, the nucleus first studied and probably the most studied for its cluster radioactive decay. The search for ^{15}N may be particularly interesting as two opposing factors will affect its emission. On the one hand, the decay energy to the ground state of the doubly magic nucleus ^{208}Pb is particularly large, resulting in a relatively large transmission coefficient. On the other hand, a ground state to ground state transition is expected to be hindered by the configuration of the unpaired proton before and after the decay, which is significantly different in ^{15}N and ^{223}Ac. Note that transitions to excited states of ^{208}Pb should be severely hindered energetically as those states are lying so high above the tightly bound ground state (E* = 2.61 MeV for the 1st excited state, reducing the barrier penetrability by more than 4 orders of magnitude), thus, such transitions are not expected to be experimentally attainable. In contrast, some predictions for a ground state to ground state transition seem to be experimentally quite attainable.

There is a further compelling reason to be curious about the cluster radioactivity of ^{223}Ac. Until a number of years ago, it was generally believed that a ground state to ground state transition from an odd-*A* nucleus should always be hindered [3], such as has been known to be the case for alpha decay for many years. Indeed, such unfavoured decay has been found in heavy cluster emission from various odd-*A* emitters [2] such as ^{221}Fr, ^{221}Ra, ^{223}Ra, ^{231}Pa and ^{233}U. A study published in 1993, however, found an unexpectedly high value for the branching ratio of ^{14}C emission from ^{225}Ac, consistent with a ground state to ground state transition [4]. This branching ratio was re-measured in 2001 and confirmed the 1993 result [5]. To date, the only isotope of Ac that has successfully been studied for its ^{14}C radioactivity is ^{225}Ac. Although the fact that its ^{14}C emission, which rather reflects that of even-even emitters, is now believed to be understood, this result still remains somewhat of an anomaly. In particular, if the transition hindrance is determined by the change of the odd (unpaired) nucleon configuration before and after the decay (an odd proton in this case) then one can expect similar behaviour from ^{223}Ac.

It is instructive to reflect on the experimental method used to measure the emission of heavy clusters from ^{225}Ac (amongst other cases) [4,5] and why that particular method was not successful in the case of ^{223}Ac. In any such experiment, obviously, one needs a source of the desired parent radioisotope. Typically, the SSNTD will be arranged in either a spherical array around the source *(i.e.* a 4π geometry) or a hemisphere (*i.e.* a 2π geometry). As the source is placed in the centre of the sphere, it should ideally have as close to a point geometry as possible, deposited on a backing which is thin enough not to significantly degrade the emitted ions. (The latter condition on the backing is largely relaxed for a hemispherical detector geometry). Both the array of detectors and the source have to be pumped down to vacuum during the exposure. In several previous studies, radioactive beams of the desired species were obtained at the ISOLDE facility of CERN by magnetically separating the ions of interest from the spallation products induced in the bombardment of a thick ThC_2 target with protons (either 600 MeV or 1 GeV). The extracted ions (of ~ 60 keV) were collected on thin carbon or aluminium catcher foils placed at the appropriate positions in the focal plane of the ISOLDE magnetic separator. If the half-lives were sufficiently long, the sources produced in this way could be removed after an appropriate collection time and the SSNTD exposed *off-line*. If the half-lives were short relative to the required collection time, *e.g.* in the cases of ^{221}Ra ($T_{1/2}$ = 28 s) and ^{221}Fr ($T_{1/2}$ = 4.9 min), the detector exposures had to be performed *on-line*, at the same time as the source collections [6].

Actinium, however, proved to be a very difficult case. Efforts to produce sufficiently intense beams of ^{223}Ac and ^{225}Ac were not successful as the diffusivity of Ac in the ThC_2 targets proved to be extremely low, even at highly elevated temperatures. Efforts to increase the diffusivity were also not successful. Sources of the longer-lived ^{225}Ac ($T_{1/2}$ = 10.0 d) could, however, still be produced at ISOLDE for off-line experiments, by collecting its β^- precursors ^{225}Ra and ^{225}Fr with high intensity, followed by an appropriate waiting period (about 20 days) for the ^{225}Ac to grow in as the precursors decay [4,5]. In contrast, ^{223}Ac is a very different case as its half-life is only 2.1 minutes, which means that even if a beam of this ion could have been produced, an on-line experiment would have been required. It also does not have any β-precursors. An experiment was therefore designed to produce sources of ^{227}Pa, which has a much longer half-life (38.3 min) and would continuously feed ^{223}Ac (in secular equilibrium with its precursor) by means of alpha decay. Aspects relevant to the production of the sources are presented in Sec. 2, details of the search for heavy clusters are presented in Sec. 3, the results are discussed in Sec. 4 and, finally, a summary and conclusion are presented in Sec. 5.

2 Source production

Targets of Th were irradiated with a 66 MeV proton beam delivered by the separated sector cyclotron of iThemba LABS, the relevant reaction being ^{232}Th(p,6n)^{227}Pa → ^{223}Ac + α. The Pa was chemically separated from the Th target matrix using an ion exchange chromatographic technique. Details of the radiochemical investigation of this study have already been published elsewhere [7].

During the planning stages, yield calculations were performed using the rather old excitation function data of Suk *et al.* for ^{232}Th + p [8]. Here we would like to briefly look at all the available EXFOR data sets [9] as some doubt exists about the quality thereof for this reaction. Four relevant EXFOR entries exist but two are from the same authors and are combined into a single set, shown in Fig. 1. The excitation function of Suk *et al.* [8] (EXFOR entry B0037) shows a prominent peak with a maximum of about 42 mb at 47 MeV. The data of Lefort *et al.* [10] (EXFOR entries O0044 and P0006) also show a peak in the relevant energy region but somewhat shifted towards higher energies, reaching a maximum of about 15 mb at 54 MeV. The excitation function of Meinke *et al.* [11] (EXFOR entry P0047) has a very different shape, not reaching a maximum at all in the relevant energy region. A peak is expected for the (p,6n) reaction, however, as shown by a standard Geometry Dependent Hybrid (GDH) model calculation performed using a very recent version of the computer code ALICE/ASH [12]. Based on this prediction, the data of Ref. [11] were disregarded. It is interesting that the GDH calculation gives a result located largely between the two other experimental excitation functions. Based on the shape of the excitation function, it was decided to prepare targets with an effective energy window 62 → 40 MeV, indicated by the dashed lines in Fig. 1. The lower value is just above the expected reaction threshold while the higher value is the practical upper limit for encapsulated targets, using standard iThemba LABS targetry and a 66 MeV proton beam, which is regularly available at the laboratory for its routine radionuclide production programme.

Ten Al-encapsulated Th targets were prepared for this study, shown in Fig. 2 together with a standard iThemba LABS target holder for irradiating batch radionuclide production targets. The Th discs had a thickness of nominally 4.7 g/cm^2 and weighed 8 g each. During bombardment, a target would be completely surrounded (4π) by fast flowing cooling water. Several targets were bombarded at relatively low beam intensities for use in the development and testing of the radiochemical procedures. Three test bombardments and the final experimental bombardment were done at an average beam current of 80 μA for 2 hours.

As the half-life of ^{227}Pa is quite short, time was critical. The challenge was to complete the radio-chemical separation and prepare a dry source, ready to be put into a vacuum, within two half-lives after the end of bombardment (EOB). The total time from EOB to the start of the track detector exposure could eventually be reduced to 71 minutes (42 minutes to transfer the bombarded target from the beamline to the hot-cell complex, remove the Th from the capsule, bring the Th into solution, separating the Pa from the Th using an anion exchange column and rinsing the column; Eluting the Pa into a volume of only 2 mL took about 6 minutes; It took another 23 minutes to evaporate the Pa-containing eluate to dryness on a gold-plated copper source plate (see Fig. 2) under an infra-red lamp, transferring the source to the experimental vacuum chamber and pumping down to vacuum). Here we would only like to comment on two further aspects of the radiochemistry which affected the final source yield. In order to speed up the dissolution process in concentrated HCl, it was necessary to add a small quantity of HF. As a result, about 10% of the Th converted into an insoluble fluoride form, which had to be filtered out and discarded. Although the elution was very rapid and efficient, a loss of about 5% was nevertheless allowed in order to keep the final source solution down to 2 mL. Consequently, the radiochemical separation efficiency was ~ 85%.

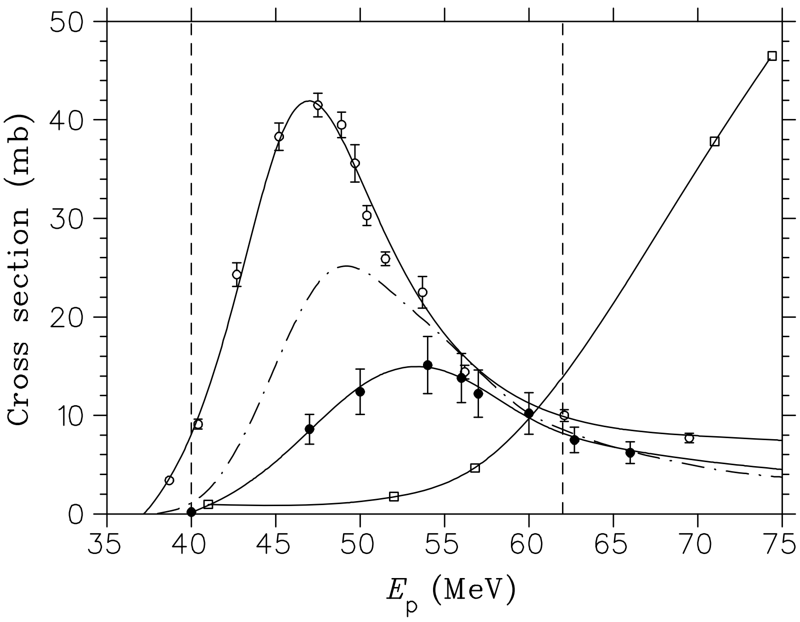

Fig. 1: Excitation functions of ^{227}Pa formed in the reaction of protons with ^{232}Th. The open circles are the data of Suk *et al.* [8], the closed circles those of LeFort *et al.* [10] and the open squares those of Meinke *et al.* [11]. The solid curves are polynomial fits used for calculating the corresponding thick-target production rates. The dot-dashed curve is a GDH calculation performed with the computer code ALICE/ASH (see text). The dashed lines indicate the selected energy window for the Th targets used in this study.

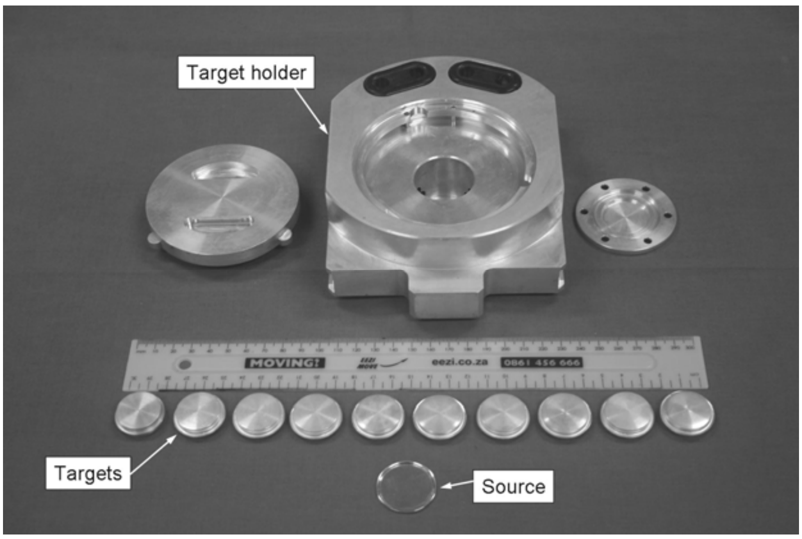

Fig. 2: FRONT: Al-encapsulated Th targets and Au-plated Cu source plate. BACK: Target holder with beam entrance window (RIGHT) and beam stop (LEFT) removed.

The strength of the source used in the final experimental run was derived from its alpha-particle emission spectrum, measured with a calibrated Si surface-barrier detector. The details of that rather involved analysis will not be given here, however, for the purposes of the discussion to follow it is convenient to give its value here: The ^{227}Pa yield at EOB was determined to be 41.1 GBq (1.11 Ci) with a total experimental uncertainty of 30%. Some of the uncertainty derives from the fact that the source geometry deviated from a point source (the diameter of the source was 25 mm) which was necessitated by a strict requirement to reduce the evaporation time (leaving no option but to increase its surface area), thus, some source imperfection was accommodated for the sake of speed. Fig. 3 shows the growth and decay curves of ^{227}Pa and its daughter ^{223}Ac for that source, taking the relevant alpha branching ratio into account: $^{227}\text{Pa} \rightarrow {}^{223}\text{Ac} + \alpha$ (85%) [13].

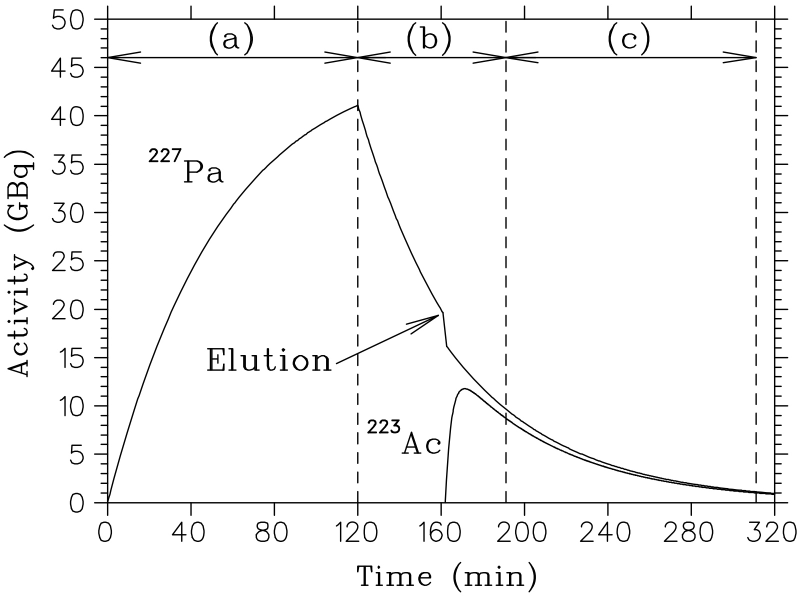

Fig. 3: Growth and decay curves of ^{227}Pa and ^{223}Ac relevant to the final experimental source. Period (a) is the target bombardment, (b) is the radiochemistry and source making stage, and (c) is the detector exposure period. Note that the exposure was stopped at a time where sufficient activity was still left in the source to measure the alpha-particle emission spectra (see text).

It is interesting to derive the *effective* ^{227}Pa cross section applicable to the full production energy window, 20 – 62 MeV, from the excitation functions shown in Fig. 1 and compare them with the corresponding value obtained from this experiment. Values of 21.68 mb and 9.45 mb were obtained, respectively, from the excitation functions of Suk *et al.* [8] and LeFort *et al.* [10]. In this study we measured a value of 7.71 ± 2.31 mb, in good agreement with the earlier work of LeFort.

3 Search for heavy clusters

A hemispherical dome with a radius of 11.75 cm was covered on the inside with BP-1 phosphate glass track detectors (Schott Glass Technologies, USA) [14] such that the geometrical efficiency was ~84% of 2π. In the case of BP-1 glass, the charge threshold is exactly 6, therefore, the enormous alpha particle flux accompanying the emission of heavier clusters was not seen. After exposure at iThemba LABS, these glasses were carefully packaged and shipped to the University of Milan, where they were first etched in 50% HBF_4 at 65°C for about 2 days to enlarge the latent tracks and make them visible under an optical microscope. The whole track detector surface (about 730 cm^2) was investigated at 200x magnification with an automated system, based on an Elbek (Siegen, Germany) image analyzer.

All the events found by the automated system were also manually inspected and the track parameters measured for those events whose identification was uncertain. Particle identification was obtained by plotting the sensitivity (S) versus the residual range (R), parameters which are proportional to the specific energy loss and energy of the ion, respectively. ($S = v_t/v_g$, where v_g is the etching velocity in the undamaged parts of the detector and v_t is the etching velocity along a particle latent track, which gives rise to a characteristic conical track that appears as a black spot in the bright field of the microscope.) This process also involved a comparison with calibration curves obtained by irradiating similar samples of BP-1 glass with ions of known mass, charge and energy, delivered by the cyclotron of LNS, Catania, Italy. The scanning of all the glasses took many months to be completed. Full details of the method used for this purpose can be found in Ref. [5].

As already mentioned in the Introduction, some predictions for heavy cluster emission from ^{223}Ac were already available many years ago. It is, therefore, interesting to first peruse some of these numbers and, scaled to the experimental source strength of this work, derive the expected numbers of clusters according to these theories. These values are presented in Table 1.

Table 1: Theoretical predictions of ^{14}C and ^{15}N decay branching ratios relative to α-particle decay and corresponding values of the expected number of emitted and detected clusters.

Cluster type	Theory	Transition	Branching ratio $B = \lambda(\text{cluster})/\lambda(\alpha)$	Expected[(a)] number of clusters emitted	Expected[(a)] number of clusters detected
^{14}C	Blendowske [3]	Favoured	3.634×10^{-11}	921 ± 276	386 ± 116
		Unfavoured	1.930×10^{-12}	49 ± 15	21 ± 6
	Poenaru [15]	—	2.514×10^{-11}	637 ± 191	268 ± 80
^{15}N	Blendowske [3]	Favoured	2.079×10^{-12}	53 ± 16	22 ± 7
		Unfavoured	8.811×10^{-14}	2 to 3	~ 1
	Poenaru [15]	—	1.001×10^{-12}	25 ± 8	11 ± 3

(a) $(2.535 \pm 0.761) \times 10^{13}$ α particles were emitted from the source during the SSNTD exposure (see text).

4 Results and discussion

The analysis of the results, based on chi-square criteria, resulted in 347 identified ^{14}C events and no ^{15}N events. Corresponding experimental branching ratios relative to alpha particle emission were derived:

$$B(^{14}\text{C}) = \lambda(^{14}\text{C})/\lambda(\alpha) = (3.2 \pm 1.0) \times 10^{-11} \quad \text{and} \quad B(^{15}\text{N}) = \lambda(^{15}\text{N})/\lambda(\alpha) \leq 2.2 \times 10^{-13}.$$

In the case of ^{15}N, only an upper limit could be inferred from inverse Poisson statistics, with a confidence limit of 90%.

The ^{14}C result is in excellent agreement with a favoured ground state to ground state transition according to the cluster model of Blendowske *et al.* [2, 3] (see Table 1), which assumes a pre-formed cluster tunneling through a potential barrier, such as traditionally assumed in theories of alpha decay. It is also in agreement with the value according to the model by Poenaru *et al.* [15], which treats the decay as a superasymmetric fission process [2,15].

The ^{15}N result is consistent with the Blendowske prediction for an unfavoured transition. It disagrees, however, with the Poenaru prediction.

The result for ^{14}C emission from ^{223}Ac is very similar to that found from ^{225}Ac and can again be interpreted in terms of the similarity between initial and final states [4,5,16]. According to this interpretation, two hypotheses should be considered, both based on the ground state configuration of the mother nucleus and, in particular, the odd (unpaired) proton. If ^{223}Ac and its sister radionuclide ^{225}Ac have *reflection-symmetric* shapes resulting from quadrupole and/or hexapole deformations, then according to older studies (see *e.g.* [17] and references therein) the ground state is described by the $3/2^-$[532] Nilsson configuration, which originates from the $1h_{9/2}$ spherical shell at zero deformation. The 83rd proton of the near spherical daughter nuclei, ^{209}Bi and ^{211}Bi, is also described by the $1h_{9/2}$ shell of the spherical shell model, consequently, the ^{14}C emission may be expected to be favoured by a ground state to ground state transition. This is the first hypothesis. The second hypothesis assumes an intrinsic ground state shape of the mother nucleus which is *reflection-asymmetric*, resulting from significant octupole and higher-order odd-multipole deformations, according to the later work by Ćwiok *et al.* [18]. In this case, the final proton state in the mother nucleus is described by the $\Omega = 3/2$ ($n_\Omega = 13$) orbital [18], which originates from the $2f_{7/2}$ shell at zero deformation. The ^{14}C decay to the ground states of ^{209}Bi and ^{211}Bi should therefore be hindered, while decay to the first excited states in these nuclei should be favoured as $J^\pi = 7/2^-$ ($E^* = 0.896$ MeV for ^{223}Ac and $E^* = 0.405$ for ^{225}Ac). Transitions to the first excited states will, however, be energetically hindered, by factors of about 40 in ^{223}Ac and 6 in ^{225}Ac relative to the respective ground states, due to lower barrier penetrability. Thus, the experimental results do not agree with the second hypothesis as the measured ^{14}C decay rates are too high, by roughly those same factors mentioned above. In this respect, the new result is especially significant as the difference between the transmission coefficients for a transition to the ground state and a transition to the first excited state is significantly larger for ^{223}Ac than for ^{225}Ac (due to the higher excitation energy of the first excited state in the former nucleus).

Another way of looking at these results is to make a deliberate assumption, for the sake of the argument, that the experimentally observed events correspond to the population of both the ground and first excited states, and to ask what the upper limit is of the fraction belonging to the first excited state. This upper limit should correspond to a unity hindrance factor, $HF = 1$ (see *e.g.* Ref. 6 for a formal definition). This yields relative intensities of 89% [11%] and 74% [26%] for the population of the ground state [first excited state] in the ^{14}C decay of ^{223}Ac and ^{225}Ac, respectively. Clearly, most of the observed events can only belong to the population of the ground state. Indeed, it is unlikely that transitions to the first excited of the daughter nucleus will not be hindered (*i.e.* it is expected that the hindrance factors will be significantly larger than unity), thus, the available information even more strongly supports the first hypothesis than these conservative relative intensities would suggest.

It is immediately evident that high-quality fine-structure measurements of these decays would be very useful, which the use of SSNTD does not allow as its energy resolution is too low to distinguish between transitions to the ground state and first excited state. It is noteworthy to reflect on one attempt to measure the fine structure in the ^{14}C decay of ^{225}Ac [5], using the superconducting magnetic spectrometer SOLENO of the IPN-Orsay, France, with which the fine structure of ^{223}Ra was successfully measured some years earlier [19]. In contrast to the ^{223}Ra experiments, the ^{225}Ac investigation with SOLENO was not successful as the ^{14}C events were swamped in the focal plane by a background of alpha particles with the same magnetic rigidity, originating from multiple scattering within the spectrometer. An experiment with a more favourable signal-to-noise ratio would be desirable but may experimentally be extremely demanding.

Lastly, we wish to report that during one of the test runs (mentioned earlier) a hemispherical dome containing only a small subset of the full complement of BP-1 glass track detectors was exposed to the emissions of a prototype ^{227}Pa source. From this trial experimental run, which was performed to test the feasibility of the experiment, a total of 47 ^{14}C events were eventually collected, in excellent agreement with the full experiment performed afterwards. Preliminary results of this work have been reported at the Nuclear Cluster Conference (Cluster'07) held in Stratford-upon-Avon, U.K. [20].

5 Summary and conclusion

An experimentally determined branching ratio for the emission of ^{14}C clusters in the radioactive decay of ^{223}Ac is consistent with a favoured ground state to ground state transition. The explanation for this rather unusual decay of an odd-*A* nucleus, which is reminiscent of even-even ^{14}C emitters in the trans-Pb region, can be based on similar arguments as those reported previously for the ^{225}Ac nucleus, related to the state to which the 83rd (unpaired) proton belongs before and after the decay.

In the case of ^{15}N emission, the non-observance of any events is compatible with an unfavoured transition. This is not unexpected as the unpaired odd proton state is very different in the heavy nucleus ^{223}Ac and the light fragment ^{15}N, resulting in a large hindrance for such a transition.

A secondary result of this work highlights the large discrepancy that exists in the available literature concerning the excitation function data for the ^{232}Th(p,6n)^{227}Pa reaction (as compiled in current EXFOR data files), with only one set of previous data found to be in good agreement with the ^{227}Pa source yield determined from an analysis of alpha-particle spectra measured in this study.

IN MEMORIAM

It is with sadness that we remember the friendship and inspiring talent, knowledge and skill of Svetlana Tretyakova and Roberto Bonetti, who tragically passed away during the course of this work.

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