Carbon radioactivity of $^{223}$Ac and a search for nitrogen emission

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Abstract. A very intense $^{227}$Pa source was produced in order to study the possible $^{14}$C and $^{15}$N spontaneous emission from $^{223}$Ac. After the irradiation of a hemispherical, highly efficient array of nuclear track detectors, about 350 Carbon events were found leading to a branching ratio with respect to alpha decay $B = 3.2 \times 10^{-11}$. Comparison with other $^{14}$C emitters allows the study of the influence of even-odd effects on cluster radioactivity.

1. Introduction

After more than 20 years from its discovery, due to the pioneering work of Rose and Jones [1], cluster radioactivity of heavy nuclei is now a well established phenomenon, both from the experimental and the theoretical side [2]. Twenty-three spontaneous emissions of neutron-rich clusters ranging from $^{14}$C up to $^{34}$Si have been measured and the typical, most important characteristics of the phenomenon have been established. Nevertheless, some aspects of the decay mode still remain to be clarified, among these the dependence of the decay probability on the microscopic properties of the involved nuclei, which can be better investigated by studying emissions from odd-A nuclei. In fact, in strict analogy with what happens for alpha decay, while for even-even nuclei the transitions are always favoured ground state to ground state ones, in the case of odd-A emitters the states to which the unpaired particle belongs before and after the radioactive decay can play a role, giving rise to favored or unflavored transitions according to the overlapping degree between such states [3,4].

In this context, we decided to study the possible cluster decay of $^{223}$Ac by $^{14}$C and $^{15}$N emission which could allow us to investigate the effect of the unpaired odd particle, both in the heavy residual nucleus in the case of $^{14}$C emission and in the cluster in the case of $^{15}$N decay. The last decay mode is expected to be on one side a particularly favored one, the residual nucleus being the tightly bound double magic $^{208}$Pb, and, on the other side, an extremely interesting one since, among the emissions

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discovered to date, only a single case of odd-A cluster is known, measured with the very low statistics of only one event [5].

2. The experiment
One of the most important features of cluster decay is its extremely low decay probability: partial half-lives for the emissions measured up to now range from $10^{11}$ up to $10^{30}$ s. Such a feature demands for the availability of a huge number of atoms of the decaying isotope. This is why, even if the idea of studying cluster radioactivity of $^{223}$Ac came to our mind already in 1990, we had to wait until 2005 to perform the experiment, in order to find out the best possible way to produce $^{223}$Ac.

2.1. Source Production
As the half-life of $^{223}$Ac is very short (2.1 min), we decided to produce its precursor $^{227}$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. We therefore irradiated a number of thick (4.7 g/cm$^2$ and nominally 8 g each) Th targets with a 66 MeV proton beam (I= 80 µA) delivered by the separated sector cyclotron of iThemba LABS for 2 hours: $^{227}$Pa was obtained through the reaction $^{232}$Th(p,6n)$^{227}$Pa. The chemical separation of $^{227}$Pa from the Th target material had to be completed within approximately 70 min from the end of bombardment (EOB), i.e. two half-lives of the precursor radioisotope: this was achieved by ion exchange chromatography. The chemical procedure is described in details in Ref. [6]. After the separation from the bulk material, the Pa was evaporated onto a 28mm diameter gold plated copper support and this constituted our source. The duration of the whole procedure took 71 min after EOB.

2.2 Detectors
The source was put in an irradiation chamber where it was exposed to a 23.5 cm diameter hemisphere, covered inside with BP-1 solid state nuclear track detectors (SSNTD), for 2 hours. The choice of SSNTD for measuring cluster radioactivity is almost compulsory: in fact, the branching ratios with respect to alpha decay are always very low - from $10^{-9}$ down to $10^{-17}$ for the known emissions- therefore detectors with a lower charge threshold are necessary. In the case of BP-1 glasses, this threshold is exactly 6, therefore the enormous flux of alpha particles accompanying the eventual $^{14}$C or $^{15}$N emission is not seen. The geometrical efficiency of the detecting apparatus was very high, about 84% of 2$\pi$. The BP-1 glasses were subsequently etched in 50% HBF$_4$ at 65°C for about 2 days in order to enlarge the latent tracks eventually produced by the ionizing clusters and make them visible under an optical microscope.

After the irradiation of the track detectors, the alpha activity of the Pa source was measured with a 300 mm$^2$ commercial silicon detector, placed at about 30 cm from the source. In front of both the detector and the source, two collimators of 8 and 5 mm diameter were placed, respectively, in order to better define the geometrical efficiency of the measurement. The 5 mm collimator was put in different positions on the source to overcome the ambiguity due to a possible non-uniform distribution of the source material. The alpha activity measurement was performed by doing four short (a few minutes each) acquisitions with the Si detector at different times, from about 6 to about 8 h after EOB, in order to have, on one side, a reasonable counting rate on the detector and, on the other side, a good statistics for the two more energetic alpha lines due to the $^{227}$Pa decay chain.

3. Analysis and Results
The whole surface of the irradiated track detectors (about 730 cm$^2$) was investigated under an optical microscope (at 200x magnification) with an automated system [5], based on an Elbek (Siegen, Germany) image analyzer, which allowed for a faster search of “good” candidates. Afterwards, all the automatically found events were manually inspected and track parameters were measured for those events whose identification was uncertain. This allowed the calculations of sensitivity (S) and residual range ($R_r$), two characteristic parameters of track detectors that are proportional to specific energy loss
and energy, respectively. Then a comparison with calibration curves obtained irradiating similar samples of BP-1 glasses with ions of known mass, charge and energy delivered by a Tandem accelerator finally allowed for the charge and energy identification of the events. This comparison is shown in figure 1 for some of the found events, each identified by three couples of \((R_r, S)\) measured at different etching stages of the track development, i.e. in different positions along the particle total range in the track detector.

A still preliminary analysis of the results, based on chi-square criteria, allows the attribution of about 350 events to \(^{14}\text{C}\) clusters, with energy compatible with that expected on the basis of the decay Q-value. No event was attributed, at this stage of the analysis, to the emission of a \(^{15}\text{N}\) cluster.

![Figure 1: Comparison of detected events with accelerator calibration curves. Each event is identified by three couples of \((R_r, S)\). The solid (dashed) line is the calibration curve for \(^{14}\text{C}\) (\(^{15}\text{N}\)).](image)

The analysis of the alpha spectra was much more challenging than expected. In fact, the source was not only non-uniform but also rather thick, due to a substrate of about 5-10 \(\mu\text{m}\) of FeCl\(_3\).3H\(_2\)O. It also contained a large number of isotopes with all their decay products. The substrate was a residue of the chemical procedure while the isotopes were produced during the thick Th target proton bombardment via \((p, xn)\), \((p, pyn)\), \((p, azn)\) reactions. This finally resulted in 51 elements giving rise to about 250 alpha transitions, most of which contributed to some energy spread due to the source thickness. A simple analysis of the most energetic alpha transitions due to \(^{219}\text{Fr}\) and \(^{215}\text{At}\) belonging to the \(^{227}\text{Pa}\) chain was not possible. A much more complicated analysis procedure was then undertaken, taking into account all the 250 alpha transitions. A simulation of the alpha spectrum with different substrates and different thickness distribution in the source was performed to allow a comparison with the experimental data. In the end, the number of \(^{223}\text{Ac}\) alpha particles emitted from the source during the track detectors irradiation was determined with an uncertainty not yet finalized (probably of the order of 30\% or even more) due to all the above mentioned ambiguities. From this number, the number of \(^{14}\text{C}\) clusters and the geometrical efficiency of the detecting array, it is possible to calculate the
experimental branching ratio for $^{14}$C emission, $B(^{14}$C) = $3.2 \cdot 10^{-11}$. In the case of $^{15}$N emission, only an upper limit on the branching ratio can be inferred: $B(^{15}$N) $\leq 2.2 \cdot 10^{-13}$, with a 90% confidence level.

4. Discussion

A more refined analysis of the track parameters and chi-square criteria for the charge attribution of the detected events is still in progress. Nevertheless, on the basis of our preliminary results some conclusions may be drawn. In the case of $^{14}$C emission, our result for $^{223}$Ac can be compared with other already measured cases of $^{14}$C radioactivity from even-even or odd-A emitters. This can be done in the form of a Geiger Nuttal plot, where the partial half-life $T$ is given as a function of the barrier penetrability $P$, in a Log-Log scale. This is represented in figure 2, where the results for even-even emitters are represented as circles and lie on a straight line while those for odd-A emitters are represented as squares and are above the line. For $^{223}$Ra, the decays to both the ground state and the first excited state of the heavy residual nucleus have been measured. The vertical distance of the odd-A LogT value from the line, for a fixed $-\log P$, is a simple measurement of the so-called Hindrance Factor (HF), i.e. of the lower decay probability associated only with nuclear structure effects. A more formal evaluation of the HF for $^{223}$Ac, according to the relations described in [3,4], gives about 5, not very far from unity. This means that $^{14}$C emission from this odd-A isotope is not much unfavoured but, on the contrary, resembles emissions from even-even nuclei.

![Figure 2. Geiger Nuttal plot for $^{14}$C emitters: the line interpolates the results for even-even nuclei (circles). See text for details.](image-url)
This can shed some light on the $^{223}$Ac ground state configuration which, in analogy with what happens in the decays of even-even nuclei, is expected to be similar to the ground state configuration of the heavy residual nucleus, $^{209}$Bi. A possible population of the first (0.9 MeV) excited state of $^{209}$Bi would be hindered by a factor 40 due only to energetic considerations, therefore the decay can be interpreted as a ground state-to-ground state favoured one.

In the case of $^{15}$N emission, the nonobservance of any event is compatible with an unfavoured transition: here the unpaired odd particle goes from the heavy nucleus $^{223}$Ac to the much lighter cluster $^{15}$N, whose ground state configurations are anyhow very much different.

A more detailed spectroscopic discussion of these results, once finalized, together with a comparison with the more accredited theoretical predictions will be published soon [7].

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