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Precise measurements of the absolute γ-ray emission probabilities of ^{223}Ra and decay progeny in equilibrium

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HIGHLIGHTS

- Discrepancies found within currently published γ-ray emission probabilities.
- Absolute γ-ray emission probabilities of decay series in equilibrium determined.
- Significant improvement in precision of measured values.
- Closer agreement between deduced and experimental α transition probabilities.
- Correlation coefficients presented for γ-emissions of ^{223}Ra, ^{219}Rn and ^{211}Pb.

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ABSTRACT

Precise measurements of the absolute γ-ray emission probabilities have been made of radiochemically pure solutions of ^{223}Ra in equilibrium with its decay progeny, which had been previously standardised by 4π(liquid scintillation)-γ digital coincidence counting techniques. Two high-purity germanium γ-ray spectrometers were used which had been accurately calibrated using a suite of primary and secondary radioactive standards. Comparison of the activity concentration determined by the primary technique against γ-ray spectrometry measurements using the nuclear data evaluations of the Decay Data Evaluation Project exhibited a range of $\sim$18% in the most intense γ-ray emissions ($>$ 1% probability) of the ^{223}Ra decay series. Absolute γ-ray emission probabilities and standard uncertainties have been determined for the decay of ^{223}Ra, ^{219}Rn, ^{215}Po, ^{211}Pb, ^{211}Bi and ^{207}Tl in equilibrium. The standard uncertainties of the measured γ-ray emission probabilities quoted in this work show a significant improvement over previously reported γ-ray emission probabilities. Correlation coefficients for pairs of the measured γ-ray emission probabilities from the decays of the radionuclides ^{223}Ra, ^{219}Rn and ^{211}Pb have been determined and are presented. The α-transition probabilities of the ^{223}Ra have been deduced from $P_{(\gamma+ce)}$ balance using the γ-ray emission probabilities determined in this work with some agreement observed with the published experimental values of the α-emission probabilities.

1. Introduction

Radium-223 is a naturally occurring radionuclide, occupying the later stages of the primordial decay series of ^{235}U (see Fig. 1), that makes up approximately 0.7200(51)% of naturally occurring uranium (Rosman and Taylor, 1998). With a half-life of 11.4354 (17) d (Collins et al., 2015), ^{223}Ra has undergone investigations for use as a radiopharmaceutical, with successful clinical trials for targeted radiotherapy of bone metastases and bone palliation that occur from late-stage castration resistant prostate cancer (Michalski et al., 2013; Nilsson et al., 2007; Parker et al., 2013). As a group II element, radium shares many chemical properties with calcium and exhibits a high level of incorporation into metabolically active osteoblastic bone and tumour lesion sites (Bruland et al., 2008; Nilsson et al., 2007). Coupled with the relatively short dose deposition range of α-emissions this allows a highly targeted cytotoxic dose of ionising radiation to a specific cancer site with reduced damage to the bone marrow and other surrounding healthy tissue, giving this treatment obvious advantages over the relatively long energy deposition range of β-emitting bone-targeting radiopharmaceuticals e.g. ^{89}Sr, ^{166}Ho and ^{153}Sm, that have been used historically. Additionally, as a naturally occurring radionuclide it is of interest as a potential radiotoxic hazard from

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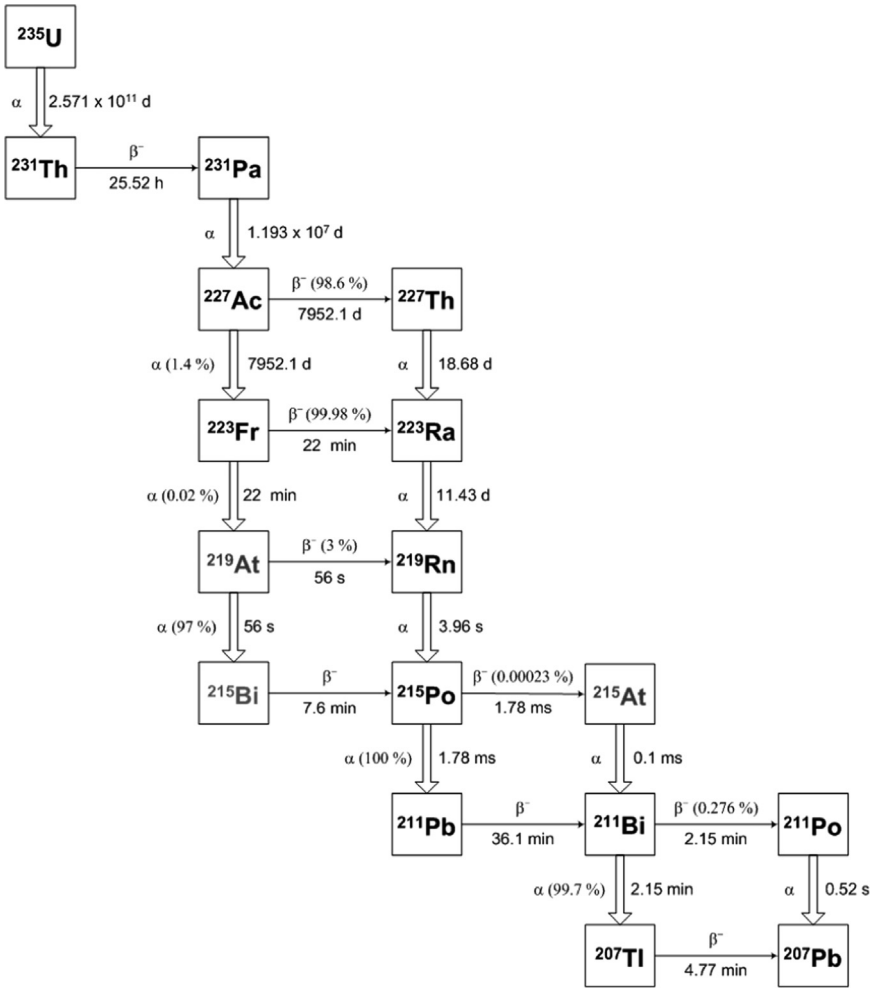

Fig. 1. Decay series of ^{235}U.

naturally occurring radioactive material (NORM) and technologically enhanced NORM (TENORM) (Kathren, 1998).

The ^{223}Ra nucleus decays by 100% α-emission to excited states of ^{219}Rn (Chechev, 2011c), the decay scheme of which is shown in Fig. 2, with a small branch (4.7×10^{-10} relative to the α-decay branch (Kutschera et al., 1985)) decaying via the spontaneous emission of a ^{14}C nuclear cluster to ^{209}Pb (Rose and Jones, 1984). The ^{219}Rn further decays via a series of relatively short-lived ($T_{1/2}<37$ min) α- and β-emitting decay progeny each decaying via their respective excited states (Chechev, 2011a, 2011b, 2011c; Nichols, 2011; Kondev, 2013a, 2013b; Luca, 2010; Luca, 2011) with associated γ-ray emissions, with the series terminating at the stable nucleus of ^{207}Pb. The ^{14}C cluster decay mode of ^{223}Ra has not been investigated in the scope of this work.

In view of the growing importance of ^{223}Ra in these applications the National Physical Laboratory (NPL) undertook a course of work to provide an absolute standardisation of ^{223}Ra (Keightley et al., 2015) that would provide traceability to the SI unit of the Becquerel. Two solutions of ^{223}Ra were independently standardised, henceforth referred to as S1 and S2, using liquid scintillation (LS) absolute standardisation techniques, the first performed in 2013 and the second in 2014. Initial measurements of the solutions were made by high purity germanium (HPGe) γ-ray spectrometry of the thirteen most intense γ-ray emissions ($>1\%$) of the ^{223}Ra decay series, listed in Table.1. The activity of each selected γ-ray of the ^{223}Ra decay series was determined using the nuclear data from the evaluations of the Decay Data Evaluation Project (DDEP) (Chechev, 2011c; Nichols, 2011; Kondev, 2013a; Luca, 2010). Whilst the weighted mean activity of the individual γ-ray emissions were in good agreement with the activity determined by the LS standardisation techniques (see Fig. 3), the activity concentration calculated from the various γ-ray emissions had a range of $\sim$18%, as shown in Table 1 and Fig. 4. We considered this indicated a statistically significant discrepancy in the existing published γ-ray emission data, and that further measurements were required. This conclusion reflected the findings of Kellett and Nichols (Kellett and Nichols, 2013) who highlighted a disagreement between the α-transitions and values deduced from the $P_{(\gamma+ce)}$ balance within the ^{219}Rn excited levels.

A review of the currently published normalised γ-ray emission values (Blaton-Albicka et al., 1976; Briançon and Leang, 1968; Davidson and Connor, 1970a; Hesselink, 1972; Kossert et al., 2015; Krien et al., 1970; Sheline et al., 1998) for the decay of ^{223}Ra was performed. In all cases, with the exception of Krien et al. (1970), no specific information was provided regarding the full-energy peak (FEP) efficiency calibration of the γ-ray spectrometers used. Such FEP efficiency calibrations are critical to the accuracy of γ-ray emission probability measurements. Comprehensive details of the methodology used and the resulting FEP efficiency calibration curve are therefore presented in detail in this article.

Maintaining a chemical separation of the different radionuclides present in the decay series of ^{223}Ra is problematic due to the short half-lives of the decay progeny and evolution of ^{219}Rn; therefore the measurements were made of the decay series in equilibrium within an aqueous solution. Investigation of the γ-ray emissions from the decay of ^{223}Ra and its decay progeny indicated