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Measurement of the ^{211}Pb half-life using recoil atoms from ^{219}Rn decay

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HIGHLIGHTS

- Radiochemically pure ^{211}Pb samples produced from α recoil from ^{219}Rn decay.
- Half-life measured using a 2π proportional counter.
- Measured ^{211}Pb half-life of 36.164 (13) min.
- Recommended ^{211}Pb half-life of 36.161 (17) min evaluated.

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ABSTRACT

The radioactive half-life of ^{211}Pb was measured, by α-particle counting of samples of radiochemically pure ^{211}Pb in equilibrium with its α-emitting progeny, ^{211}Bi and ^{211}Po. The samples were prepared by the collection of ^{215}Po recoil atoms from the decay of the ^{219}Rn decay progeny produced from a ^{223}Ra sample onto stainless steel discs. The radioactive decay of the ^{211}Pb was measured utilising a 2π proportional counter operating on the α plateau. A half-life of 36.164 (13) min was determined, which is in agreement with currently available literature. A full uncertainty budget is presented. A recommended half-life of $T_{1/2}(^{211}\mathrm{Pb})=36.161$ (17) min has been evaluated from the current literature values.

1. Introduction

The radionuclide ^{211}Pb is a short-lived ($T_{1/2}\sim$36.1 min) member of the primordial $4n+3$ decay series of ^{235}U (see Fig. 1), which constitutes approximately 0.72% of naturally occurring uranium by mass (Rosman and Taylor, 1998), although the activity ratio of ^{235}U to ^{238}U is approximately 21:1. The ^{211}Pb nucleus decays via 100% β-emission to ^{211}Bi ($T_{1/2}=2.15$ (2) min (Luca, 2010)), further decaying via α-emission (99.7% branching ratio) to ^{207}Tl ($T_{1/2}=4.774$ (12) min (Kondev, 2013a)) and β-emission (0.3% branching ratio) to ^{211}Po ($T_{1/2}=0.516$ (3) s (Luca, 2011)), which further decays to ^{207}Pb by α-emission.

As noted above, as a member of the ^{235}U decay series it is of some importance as a radiotoxic hazard in naturally occurring radioactive material and technologically enhanced naturally occurring radioactive material where the precursors of ^{235}U, ^{231}Pa and ^{227}Ac can accumulate through diverse industrial processes, especially during the refining of metal ores or crude oil (Kathren, 1998). In addition, ^{211}Pb is the decay progeny of ^{227}Th and ^{223}Ra, which respectively are under investigation for use as a radiopharmaceutical (Dahle et al., 2009; Heyerdahl et al., 2012) or has already been implemented as the radiopharmaceutical Xofigo® (FDA News Release, 2013; EMA/CHMP, 2013). As the longest lived decay progeny it exists in transient equilibrium with ^{223}Ra, an accurate value of the half-life for ^{211}Pb is important for dose studies and ingrowth corrections over the first hours of a chemical separation from ^{227}Th and ^{223}Ra.

There have been three previous determinations of the half-life of ^{211}Pb, by Sargent (1939), Nurmia et al. (1965) and more recently by Kossert (2015), as presented in Table 1. An IAEA Coordinated Research Project (Kellett, 2012) identified ^{211}Pb as being in need of new half-life measurements to improve the accuracy and precision of the recommended half-life. This work describes the measurement of the half-life and presents an uncertainty budget. All uncertainties are stated as standard uncertainties or combined standard uncertainties as defined in the *Guide to the Expression of Uncertainty in Measurement* (GUM) (BIPM, 2008).

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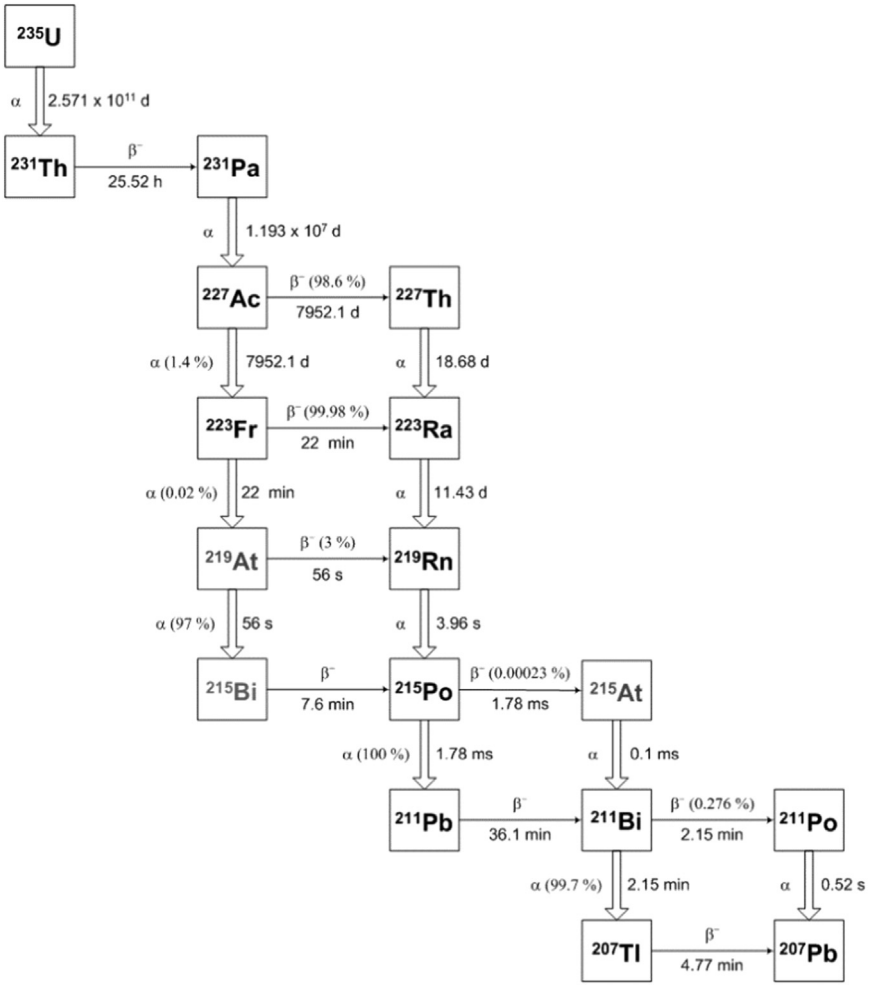

Fig. 1. Decay series of ^{235}U.

Table 1
Summary of published ^{211}Pb half-life values.

Reference	Year	$T_{1/2}$ (min)	$\sigma(T_{1/2})$ (min)
Sargent	1939	36.1	0.2
Nurmia et al.	1965	36.1	0.2
Kossert	2015	36.165	0.037
This work	2015	36.164	0.013

2. Experimental conditions

2.1. Sample preparation

A radiochemically pure ^{223}Ra solution was supplied to NPL by Bayer (Norway) as $RaCl_2$ solution with a nominal activity of 3.2 MBq g^{-1} at the start of the measurement campaign. Approximately 0.3 g (~1 MBq) of this solution was deposited on each of three stainless steel discs normally used for α-spectrometry measurements. The short half-life of ^{219}Rn ($T_{1/2}$=3.98 (3) s (Nichols, 2011)) means that it can decay to ^{215}Po before diffusing out of solution, and so this is not an efficient means of collecting recoil atoms. The strategy adopted was to evaporate the ^{223}Ra solution on to the stainless steel disc, providing a relatively thin deposit and to collect the recoil atoms onto three more stainless steel discs (recoil discs). The recoil discs were mounted approximately 2 mm above the ^{223}Ra sources using a bespoke 3D printed sample holder and placed in a fume cupboard.

A gap in the sample holder where the top and bottom sections met was apparent. This was found to result in the airflow within the fume cupboard removing ^{219}Rn from the region of the recoil discs and reducing the recoil atoms collected. This was remedied by wrapping adhesive tape around the sample holder.

Radon-219 diffuses from the ^{223}Ra sources into the air space between the ^{223}Ra source and the recoil disc, decaying to ^{215}Po. Due to conservation of momentum during α-decay, the recoiling nucleus of ^{215}Po has a kinetic energy of 0.127 MeV which was found to be sufficient to impact and implant it onto the surface of the recoil disc. Polonium-215 ($T_{1/2}$=1.781 (4) ms (Chechev, 2011a)) rapidly decays by 100% α-decay to ^{211}Pb (there is a very minor 0.00023% β-decay branch to ^{215}At ($T_{1/2}$=0.10 (2) ms (Chechev, 2011b))). Lead-211 and ^{215}At both decay to ^{211}Bi, which will reach secular equilibrium with the ^{211}Pb within 45 min.

The recoil discs were left to accumulate ^{215}Po recoil atoms for approximately 4 to 16 hours before being removed and mounted in the detector. The count rates achieved from the three recoil discs after this modification was >7000 s^{-1}, with a maximum of 16,864 s^{-1} observed, 45 min after collection. For the measurements presented in this work, a total of twelve sets of three recoil discs were produced and measured which were not found to be contaminated with ^{223}Ra. In total only three discs were found to be contaminated and the measurements rejected. This contamination was traced to inadvertent contact of the tweezers, used to remove the recoil discs, with the ^{223}Ra sources.