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On the variation of the ^{210}Po Half-Life at low temperature

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Abstract

Experiments were performed to verify the possible influence of the temperature and source matrix on the half-life of ^{210}Po. Since the precise measurement of the activity of an α-emitting source at cryogenic temperature is far from trivial, a simpler approach was used: the activity of a ^{210}Po source was measured at ambient temperature, but in between the measurements, the source was cooled down during a few weeks in a liquid helium Dewar. A ^{210}Po solid source was prepared by electro-deposition on a silver plate. The activity of this source was first measured at room temperature, using the defined solid angle α measurement method. The source was then placed in a specific gastight container inside a liquid helium bath at 4 K during 28 days. Then the source was heated up and measured again using the same instrument in the same geometric conditions. The same experiment was repeated with the source coated with an electrodeposited thin layer of silver, in order to be sure that the radioactive material was fully embedded inside the metallic matrix. Our experiments showed no evidence of ^{210}Po half-life change at low temperature. A reduction of the half-life of ^{210}Po by 6.3 % at low temperature, as claimed by Raiola et al., would have lead to a decay-corrected activity reduction of about 1 % after 29 days, which would have been easily detectable.

The paper describes the steps of this experiment and gives a detailed uncertainty budget for the measurements. The half-life of ^{210}Po obtained in each measurement is compared with the evaluated value of (138.3763 ± 0.0017) d.

1 Introduction

The assumption that the decay constant is an intrinsic property of a radionuclide is implicitly assumed by most of the people involved in the metrology of radioactivity and also fully supported by a lot of measurements. As a consequence, nuclear data tables generally mention the values of half-lives, without any reference to the chemical state of the nuclide, nor the temperature and pressure conditions. A well-known exception is the case of some radionuclides decaying through electron capture transitions, for which the probability of disintegration, thus the decay constant, is related to the atomic electron density function. This is the case for the decay of ^{10}Be, of which the half-life is known to be slightly dependant on the chemical state of the source.

This was the situation before recent publications from the group of C. Rolfs from the University of Bochum, claiming a dependence of the α– and β–decay constant for various nuclides on the temperature and source matrix (Raiola *et al*, 2005, Limata *et al*, 2006, Rolfs, 2006). In 2007, F. Raiola *et al.* reported an increased activity for the α-decay for a ^{210}Po source when embedded into a palladium matrix at low temperature. They claimed that the ^{210}Po ambient temperature half-life is reduced by (6.3 ± 1.4) % when the source is cooled down to 12 K. Previously, in 2006, K. U. Kettner *et al.* predicted that in a metal cooled at 4 K, the half-life of ^{210}Po is shortened to 0.5 days. The authors supported these surprising results by proposing a theoretical explanation based on electron screening effects in a metallic matrix. These publications received a strong echo in our national media, some politicians suggesting that this phenomenon could be used to process radioactive nuclear wastes. This is why we undertook a precise experiment to evaluate the veracity of this phenomenon.

2 Methodology

The purpose of this study is to check if the half-life of ^{210}Po deposited on a metallic substrate or included in a metallic structure is dependant on the temperature. This radionuclide was chosen because this temperature dependence has been previously reported and is easy to measure by alpha counting, with a high precision. Surprisingly, most of the experiments reported so far were conducted using a system allowing the temperature change of the source, but this approach is not optimum as one can anticipate that these temperature changes could induce mechanical variations in the device and modify the detection solid angle.

Our approach is much simpler: we first measured the activity of a solid source at ambient temperature using a defined solid angle alpha counting device. Then the source was placed in an air-tight metallic container immerged into liquid helium during a few weeks and eventually, the source was heated up and re-measured at ambient temperature in the same conditions as before. As the activity of a source is proportional to the number of radionuclide atoms, any significant change in the radionuclide half-life during the cooling period would modify the expected number of radioactive atoms during this period and would lead to a detectable change in the decay corrected activity of the source between the two measurements. This two steps measurement thus allows a very simple detection of any significant change in the half-life of the radionuclide during the cooling period.

3 Source preparation

We prepared a ^{210}Po source by spontaneous deposition on a 25-mm diameter and 2-mm thick silver disk. This disk is placed in a PTFE (Polytetrafluoroethylene) cell, originally designed for alpha source electrodeposition.
The PTFE cell was filled with 4,10 ml of ^{210}Po standard solution with an activity of (4.1 ± 0.1) kBq/g in HCl 2M at the reference date of 28/04/09.
The deposition was carried out for 60 min at room temperature with agitation. The source was then allowed to dry at room temperature, and quickly measured to determine the order of magnitude of its activity. The deposition yield of ^{210}Po on the silver disk was estimated from the activity measurement of the solution before and after deposition, using liquid scintillation counting.

4 Absolute activity measurement method

The detector used is an alpha counter with a defined solid angle of detection. Our equipment consists of a vacuum chamber equipped with a silicon semiconductor detector (Canberra PD 2000-40-300 AM), and a stainless steel collimator. The distance between the diaphragm in front of the detector and the source is measured with a relative standard uncertainty of 0.02 %. The radius of the diaphragm is measured with a relative standard uncertainty of 0.01 %. The activity of the source is calculated from the following equation:

$$A = \frac{n}{t * G * I} \qquad \text{[Eq 1]}$$

n: net counting rate
G: detection solid angle divided by 4 π sr
t: time of measurement
I: Alpha-particle emission probability

G, the geometry factor, is calculated by an analytic program, based on Curtis' method (Curtis, 1955). The detector is connected to a spectrometry amplifier via a charge preamplifier. The output of the amplifier is connected to an MCA. The counting rate is deduced from the surface of the characteristic alpha peak. The MCA software is Genie 2000 (Canberra).

Typical uncertainty budgets are given in Tables 1 and 2. It must be pointed out that, if the source is placed in a reproducible position, the uncertainty components due to the definition of the solid angle largely cancel for the relative comparison of two measurements, and the dominant uncertainty to consider, in this case, is the statistical fluctuations of the counting rate.

4.1 Initial measurement

Firstly, we calculated the activity using the recorded spectrum. We measured the source during 20 days and the uncertainty budget is given in Table 1.

The activity of the source was (2 355 ± 5) Bq at 15:24 (UT) on 17 November, 2008.

In order to validate this activity measurement, we also made a measurement using a single channel analyzer followed by a scaler. This gave an activity of the source of (2 186 ± 6) Bq at 15 h 21 (UT) on 2 December, 2008. There is an excellent agreement between these two measurements, which give an initial activity value of (2 185 ± 4) Bq at the latter date.

4.2 Source cooling

The source was placed in a copper box (cf. Fig 1) and immersed in liquid helium at 4.2 K during 29 days.

In order to avoid any kind of liquid condensation during the cooling phase, the box was closed under helium atmosphere and hermetically tightened using an indium seal.

4.3 Final measurement

Exactly the same method was used to measure the activity of the source after cooling. We measured simultaneously with the scaler and the multi-channel analyser during 18 days, and found (1834 ± 5) Bq, and (1835 ± 4) Bq, respectively. The uncertainty budget is shown in Table 2.
The measurement was started at 14:51 (UT) on January 6th. If we calculate the radioactive decay between the initial and final measurements using a half-life of 138.3763 (17) d (Bé, 2008), the initial activity of (2 185 ±4) Bq decays to (1 834 ± 4) Bq which agrees with the final activity measurement. The difference between the final activity and the decay-corrected initial activity is (0 ± 0.8) Bq, considering that common factors in initial and final results cancel in the uncertainty on the difference. We can conclude that there is no evidence of half-life change due to the cooling of the source. To be more specific, if we consider a Gaussian distribution of the results, the relative half life change which would cause a change of activity of 1.6 Bq (i.e. mean difference plus 2 standard deviations), is 0.6 %. Thus, this experiment shows that, within a confidence interval of 95 %, there is no relative change of the half-life of ^{210}Po in these specific conditions, due to cooling at 4.2 K, of more that 0.6 %.

5 Measurements of the source embedded in silver

5.1 Addition of silver

In the first part of our experiment the ^{210}Po source was deposited on the surface of the silver support. Raiola *et al*. (2007) argued that an appreciable effect on the half-life may only be observed when the alpha emitting nuclei are situated deep enough (more than a few nm) in a metallic host material.
In order to fully embed the ^{210}Po in silver, we covered the previous source with a silver foil of silver, thin enough to only slightly decrease the energy of the alpha particles. The thickness of this silver foil was evaluated to be (170 ± 40) nm, from the shift of the alpha peak using the SRIM software. To be sure that the ^{210}Po was well embedded in silver, the foil was pressed onto the silver disk. This was done by pressing a polished silicon wafer onto the foil while baking the ensemble (silver disk - ^{210}Po source – silver foil – Si wafer) under vacuum at 320 °C during 1 h.

5.2 Initial measurement

As the activity of our source had decreased, we decided to carry out our further measurements in a different chamber with a larger detection solid angle. The advantage was that the distance between the detector and the source was smaller, so sufficient statistical accuracy could be achieved more quickly. This equipment does not allow absolute measurements, as previously. This is not a problem, because it suffices to compare the results before and after the cooling of measurements being made in exactly the same conditions.

The initial measurement was started on 6 July. We found a counting rate of (13.550 $\pm$ 0.009) s^{-1}. This counting rate was corrected for the decrease of activity during the measurement.

5.3 Source cooling

The source was cooled in the same way as described in Section 4.2. The source was immersed in liquid helium during 11 days.

5.4 Final measurement

We measured the source in the same chamber as in Section 5.2, starting from 20 July 2009. The counting rate (H) takes into account the correction due to the decrease of activity during the measurement. The expected counting rate (G), for July, 22nd, was calculated from the initial measurement (Section 5.2) using the half-life value of 138.3763 (17) d. So we can directly compare. The relative difference between the measured and calculated rates is shown in Table 3.

The difference of (0.18 $\pm$ 0.10) % between these two results cannot correspond to a relative modification of the half-live of ^{210}Po at 4.2 K superior to (3$\pm$2) %.

6 Conclusion

For measuring the activity of a ^{210}Po source deposited on a silver disk, before and after cooling, we used an absolute activity measurement method. This device allowed us to measure the source activity with a relative uncertainty better than 0.3 %. We have not observed a decrease of the half-life of ^{210}Po at 4.2 K, larger than 0.6 % within a confidence interval of 95 %.

After this, a silver foil was added on top of the source. Measurements were repeated in a geometrically less precise set-up. Nevertheless, we have observed that, if a change of the half-life of ^{210}Po occurred during the cooling time, this relative change could not be superior to (3$\pm$2) %. If necessary, better precision can be achieved for this kind of measurements using a more active source, a longer cooling period and/or a longer measurement time

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Uncertainty component	Relative standard uncertainty
Net counting statistics	$2{,}6\ 10^{-4}$
Spectrum extrapolation	$1{,}4\ 10^{-4}$
geometrical factor	$6{,}0\ 10^{-4}$
backscattering	$1{,}0\ 10^{-3}$
dead time	$1{,}0\ 10^{-3}$
alpha scattering in the detector	$1{,}0\ 10^{-5}$
Temperature	$6{,}0\ 10^{-5}$
Reproducibility	$1{,}0\ 10^{-3}$
quadratic combination	**$2\ 10^{-3}$**

Table 1: Standard uncertainty budget of the initial measurement.

Uncertainty component	Relative uncertainties

Net counting statistics	$3{,}1\ 10^{-4}$
Spectrum extrapolation	$1{,}2\ 10^{-4}$
geometrical factor	$6{,}0\ 10^{-4}$
backscattering	$1{,}0\ 10^{-3}$
dead time	$1{,}0\ 10^{-3}$
alpha scattering in the detector	$1{,}0\ 10^{-5}$
Temperature	$6{,}0\ 10^{-5}$
reproducibility	$1{,}0\ 10^{-3}$
quadratic combination	**$2\ 10^{-3}$**

Table 2: Standard uncertainty budget of the final measurement.

Measurement condition	number of counts	real time t (s)	measured counting rate H	decreased calculated rate G	**Relative difference between measured and expected rate**	Uncertainty on difference
before 2nd cooling	2329546	172779	13.550			
after 2nd cooling	2175108	172780	12.652	12.629	**$1.8\ 10^{-3}$**	$1\ 10^{-3}$

Table 3: Measurement before and after cooling of the ^{210}Po source in silver.

Figure 1: Container for cooling the source.