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Analysis of time correlations for high sensitivity measurements

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Summary. — This work illustrates the development of an alpha spectroscopy system dedicated to the measurement of surface contaminations in materials used for low-background radioactive experiments. The system described is sensitive to trace levels of natural radionuclides. The operating principle is based on the simultaneous measurement of time and energy for each detected particle. A dedicated selection algorithm is developed to identify events in defined temporal relationships, so as to highlight cascade decays in natural radioactive chains. The shown results highlight the potential of the technique in reducing the radioactive background and revealing contamination otherwise not observable with usual analysis techniques.

1. – Introduction

Experiments searching for rare events require high radiopurity of the materials used in the experiment itself since it can impact its sensitivity and thus the accomplishment of its objectives. Hence the request to elaborate high-performance techniques for screening the materials, among which α-spectroscopy has some advantages that make it qualified for future developments. α-spectroscopy detectors are sensitive both to the upper and lower parts of the natural decay chains, therefore they can provide a complete characterization of material surface contaminations. Measurements of weakly contaminated samples are typically limited by the inability to isolate the small signal from background fluctuations. Therefore, the improvement in the sensitivity of the apparatus relies on exploiting the well-defined temporal laws characterizing natural radioactivity and decay chains. Combining the energetic and temporal signatures of each decay, it becomes possible to accurately label the signals produced by radioactive sample contaminations.

In this paper, we report the preliminary results obtained by applying the analysis based on the study of delayed-coincidence events to the natural radioactive chains, to improve the sensitivity of an α-spectrometer.

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TABLE I. – *Decay sequences for each natural radioactive chain analysed using the selection algorithm.*

Chain	Selected coincidences	$\tau_{\frac{1}{2}}$ expected
^{238}U	$^{214}\mathrm{Bi} \overset{\beta}{\Rightarrow} {}^{214}\mathrm{Po} \underset{[\tau_{1/2}]}{\overset{\alpha}{\Longrightarrow}} {}^{210}\mathrm{Pb}$	164.3 μs [1]
^{235}U	$^{223}\mathrm{Ra} \overset{\alpha}{\Rightarrow} {}^{219}\mathrm{Rn} \underset{[\tau_{1/2}]}{\overset{\alpha}{\Longrightarrow}} {}^{215}\mathrm{Po}$	3.96 s [2]
^{232}Th	$^{220}\mathrm{Rn} \overset{\alpha}{\Rightarrow} {}^{216}\mathrm{Po} \underset{[\tau_{1/2}]}{\overset{\alpha}{\Longrightarrow}} {}^{212}\mathrm{Pb}$	145 ms [3]

2. – Experimental technique

The analysis based on time-delayed coincidences is an efficient way to identify and measure the contaminations of the lower parts of the ^{238}U, ^{235}U and ^{232}Th natural chains. The analysis requires that the acquisition system of the α spectrometer records the time stamp of each event together with the energy information. Offline analysis is performed on the acquired experimental data to search for couples of parent-daughter events associated with the same precursor isotope. This is performed by selecting the events with energy compatible with a potential parent decay and searching for its daughter signature within a time window of the order of a few half-lives of the daughter nucleus. When a time-delayed coincidence is found, both events are tagged for subsequent analyses. The sequences of parent-daughter events researched for each natural radioactive chain and the corresponding values of the half-lives are listed in table I.

In this context, α silicon detectors are an excellent device to search for $\alpha - \alpha$ and fast $\alpha - \beta$ time-delayed coincidences thanks to their good time and energy resolution, high-efficiency values and affordable cost.

3. – Experimental set-up

The measurements described in this work are performed with an Ortec α detection system. It is an integrated spectrometer for measuring low activity samples in which the detector, a surface barrier silicon semiconductor, is located inside a vacuum chamber and provided with a complete amplifying chain. The amplification is performed by a charge sensitive pre-amplifier and an amplifier with a 1 μs shaping time. The signals are digitized with a CAEN multi-channel analyzer module (MCA N957) and, in order to study the delayed coincidences, the data acquisition is modified to save the time stamp of each event, with a time resolution of 1 ms, together with the pulse amplitude.

We use a Monte Carlo (MC) code based on the Geant4 toolkit [4] to reproduce the experimental energy spectra and to determine the detection efficiency. The detector configuration is implemented and the output is processed to record the energy deposited in the silicon active layer and the time at which the interaction occurs.

4. – Experimental measurements and data analysis

The selection technique is tested and validated on the three natural radioactive chains of ^{238}U, ^{235}U and ^{232}Th. For this purpose, copper plates are contaminated using different techniques with nuclides belonging to one of the chains. They are respectively contaminated through exposition to ^{222}Rn inside a radon-box (^{238}U chain), implantation of ^{223}Ra and its daughters (^{235}U chain) and deposition of a ^{232}Th liquid standard.

The plates are measured using the experimental set-up described in the previous section and fig. 1 shows in black the acquired energy spectra. Data are analysed offline to identify the delayed-coincidences listed in table I and the results deriving from the application of the selection criteria are displayed in red. To perform the selection on the ^{238}U decay chain, an energy window of 200 keV large is chosen for the ^{214}Po α-peak in fig. 1(a), while a ten times bigger window is imposed for the ^{214}Bi β-spectrum. To perform the energy selection on the degraded α energy spectra of fig. 1(b), energy windows of 5000 keV are used. Therefore, the imposition of time windows of $5\tau_{\frac{1}{2}}$ length represents a strong constraint for the identification of the delayed coincidences.

The distribution of the intervals between the timestamps of the events in coincidence is analysed for ^{235}U and ^{232}Th chains, to verify the goodness of the selection technique. According to the fundamental law of radioactive decay [5], it is expected to be of the form

$$N(t) = N_0 \, e^{-\frac{t}{\tau_{1/2}} ln(2)} + N_{rc},$$

where N_{rc} represents the number of random coincidences. Figures 2(a) and (b) show the interpolated distributions. The extracted values for the half-lives of ^{219}Rn and ^{216}Po, respectively of 3.96 s and 141 ms, are in agreement with the expected values summarized in table I. It is interesting to observe that a good agreement is obtained both for the plate contaminated with ^{235}U daughters and characterized by distinctive energy peaks and for the ^{232}Th contaminated plate that presents a degraded energy spectrum (fig. 1(b)).

A Monte Carlo reconstruction of the measurement set-up, consisting of the detector and radioactive source, is performed for each configuration and tuned on experimental data. Efficiency values are extracted to evaluate the activity of the sources.

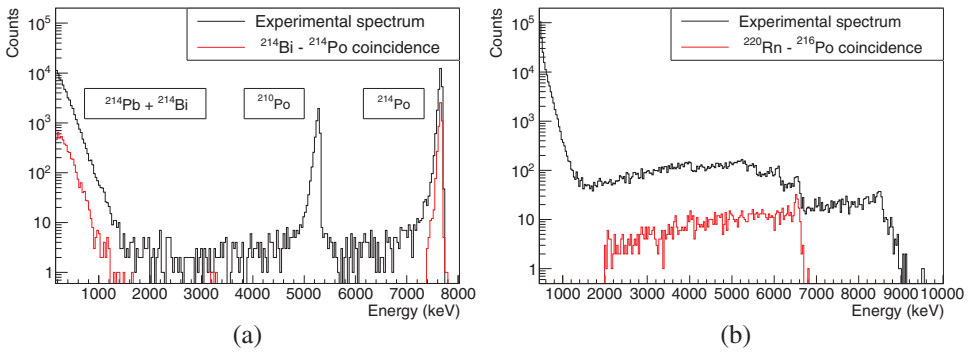


Fig. 1. – Acquired energy spectra (black) and selected coincidence events (red). (a) ^{238}U chain, ^{214}Bi-^{214}Po coincidences. (b) ^{232}Th chain, ^{220}Rn-^{216}Po coincidences.

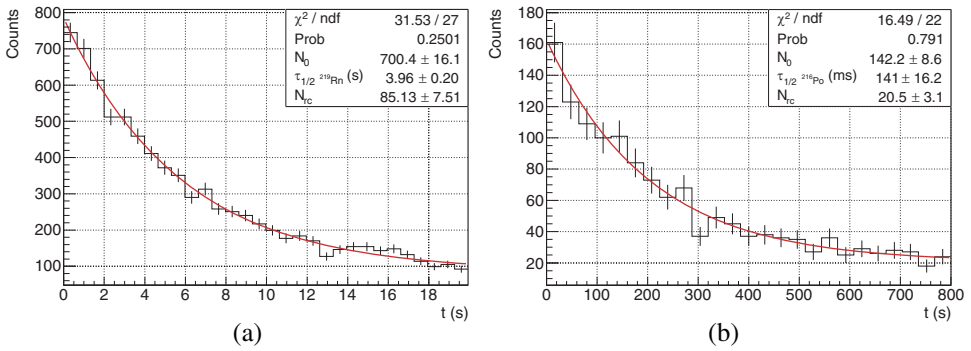


Fig. 2. – Distributions of the time intervals between the events in coincidence. (a) ^{223}Ra-^{219}Rn coincidence (^{235}U chain). (b) ^{220}Rn-^{216}Po coincidence (^{232}Th chain).

5. – Low activity sample measurement

After the testing and validation of the time-amplitude selection on different sources, the technique is applied to a weakly contaminated sample to evaluate the possible presence of ^{238}U and ^{232}Th contaminations up to a depth of implantation of 10 nm. ^{235}U contamination is not investigated due to the low natural isotopic abundance.

The source consists of a high purity copper plate treated with surface plasma ablation and measured for 32 days. The selection technique identifies one delayed coincidence for both the ^{224}Ra-^{220}Rn and ^{220}Rn-^{216}Po decays, belonging the ^{238}U chain, while no event is found belonging to the ^{232}Th chain. The resulting specific activities, calculated using Feldman-Cousins approach [6], are

$$A_{^{238}\mathrm{U}} = 7^{+17}_{-5} \cdot 10^{-7} \frac{\mathrm{Bq}}{\mathrm{cm}^2} \quad \text{and} \quad A_{^{232}\mathrm{Th}} \leq 1.6 \cdot 10^{-6} \frac{\mathrm{Bq}}{\mathrm{cm}^2} \text{ for } 90\% \text{ C.L.}$$

The probability that any of the selected coincidences is random is lower than 2%.

6. – Conclusions

This paper shows the potentiality of the delayed-coincidence selection, whose application to a naturally contaminated sample achieved a sensitivity of the order of $10^{-6} \frac{\mathrm{Bq}}{\mathrm{cm}^2}$. This value can be improved with longer measurements and higher detection efficiencies, therefore future implementations will develop detectors with larger active areas.

The main fields of application include material screening in experiments searching for rare events and environmental studies.

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