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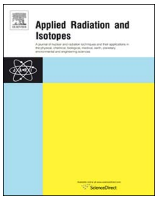

Analysis of radionuclides at ultra-low levels: A comparison of low and high-energy mass spectrometry with gamma-spectrometry for radiopurity measurements

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ABSTRACT

Recent developments in high-energy accelerator mass spectrometry (AMS) and low-energy inductively coupled plasma mass spectrometry (ICPMS) made them the most sensitive techniques for ultra low-level analysis of ^{238}U and ^{232}Th. Detection limits below 1 nBq g^{-1} for analysis of these radionuclides in construction materials were obtained. The most important development in the radiometric sector was operation of HPGe detectors in underground laboratories where detection limits of about 1 μBq kg^{-1} were obtained. Specific features of radiometric, mass spectrometry and neutron activation techniques for ultra low-level radionuclide measurements are discussed, and obtained results for analysis of radiopure materials prepared for underground experiments are compared.

1. Introduction

State of the art technologies for low-level radionuclide analyses have always been a limiting factor for underground nuclear physics experiments mainly because of radioactive contamination of construction parts of the detectors. In the field of radioanalytical technologies we moved from simple gas and solid counters to sophisticated detectors, often operating underground (Heusser, 1995; Neder et al., 2000; Neumaier et al., 2000; Laubenstein et al., 2004; Hult et al., 2006; Heusser et al., 2008; Niese, 2008; Budjáš et al., 2009; Loaiza et al., 2015). Anticosmic shielding, protecting the gamma-spectrometers against hard-cosmic radiation has been useful for their operation at shallow depths (Zvara et al., 1994; Heusser et al., 1995; Povinec et al., 2004; Povinec et al., 2008; Heusser et al., 2015). In special applications, coincidence-anticoincidence gamma-spectrometers proved to be useful mainly for analysis of small volume samples (Wogman et al., 1967; Povinec, 1982; Heusser, 1995; Povinec et al., 2005; Lutter et al., 2013; Ješkovský et al., 2015).

However, the most important break-through in the radioanalytical technology has been the change in philosophy of radionuclide analysis from the concept of counting of radioactive decays (and thus waiting for them) to the direct counting of atoms (as they would be stable) using high sensitive mass spectrometers working either with low energy ions (e.g. ICPMS (Roos, 2008), Resonance Ionization Mass Spectrometry – RIMS (Erdmann et al., 2008), Thermal Ionization Mass Spectrometry – TIMS and Secondary Ion Mass Spectrometry – SIMS (Povinec et al., 2008), or with ions accelerated up to hundreds of MeV in AMS systems (Tuniz et al., 1998; Fifield et al., 2008; Jull et al., 2008; Povinec et al., 2008; Famulok et al., 2015). This move from simple analytical techniques to the present sophisticated state of the art underground technologies (Avignone et al., 2005) has also been accompanied by a change in philosophy of nuclear physics experiments, when institutional investigations and experiments (e.g., Bellotti et al., 1992) have been replaced by global international projects (e.g., Arnold et al., 2005, 2010; Abgrall et al., 2015). These experiments focus on investigations of rare nuclear processes and decays, such as double beta-decay (e.g., Arnold et al., 2005, 2010; Agostini et al., 2016; Abgrall et al., 2015), searches for dark matter (Angloher et al., 2014), and on neutrino interactions (Alimonti et al., 2009.). As the two neutrino double beta-decay process has already been observed for several isotopes (e.g. ^{48}Ca, ^{96}Zr, ^{100}Mo, etc.), recent investigations have been focusing on neutrinoless double beta-decay experiments. Although more than ten experiments have been going on in several underground laboratories, no positive result has been obtained till now, only half-life limits of the order of 10^{25} y have been reported, (e.g., Arnold et al., 2015; Agostini et al., 2016).

Radiopurity of construction materials used in underground experiments has been crucial for background reduction. The analyses were carried out by non-destructive gamma-spectrometry with detection limit about 1 μBq kg^{-1} (Heusser et al., 2006; Budjáš et al., 2009; Povinec, in press). A new generation of underground experiments requires, however, the detection limits at least by a factor of 50 lower

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(Angloher et al., 2014; Abgrall et al., 2016). Such very low-radioactivity measurements would require essential improvements in gamma-spectrometry, determined mainly by the detector background. The big problem of the gamma-spectrometry is that radionuclides, which should be searched for (mainly the decay products of ^{238}U and ^{232}Th, and ^{40}K), are also found in the detector background.

Therefore alternative methods for analysis of primordial radionuclides (mainly ^{238}U, ^{232}Th and ^{40}K) in construction materials have been under development, e.g., AMS and ICPMS in the mass spectrometry sector, as well as the neutron activation analysis (NAA). It would be advantageous, if no pre-concentration treatment of samples would be carried out, as this process could add radioactive contamination from chemicals used during sample processing. The AMS could be the preferable technique as samples such as copper, steel, etc. can be directly used as targets in ion sources, while ICPMS would require a pre-concentration chemistry. The NAA could be used without pre-irradiation chemistry as well, but it has also an advantage that a post-irradiation chemistry can be used, which could improve detection limits. On the basis of previous experience with AMS analysis of uranium in environmental samples (Lee et al., 2008), it has been expected that detection limits for analysis of ^{238}U in construction materials below 1 nBq g^{-1} could be achieved.

In the present paper we discuss recent developments in ultra low-level radioactivity measuring techniques using radiometric, mass spectrometry and NAA methods. In the radiometric sector we focus on large volume HPGe detectors operating in shallow and deep underground laboratories. Mass spectrometry techniques, mainly developments and applications of AMS and ICPMS for radiopurity measurements are discussed as well, and finally the radiometric and mass spectrometry techniques are compared with the NAA method.

2. Underground detectors

The development of large volume HPGe detectors operating deep underground considerably improved their sensitivities because of higher detector efficiencies and a lower background (Heusser et al., 2006; Budjáš et al., 2009). The sources of background for underground detectors remain almost the same as in surface laboratories, except the fact that the cosmic-ray component can be significantly reduced, depending on the operational depth of the detector. The cosmic-ray background is mainly due to high-energy muons, which penetrate through the rock and generate in the shielding materials secondary neutrons and gamma-rays. Cosmic-ray nucleons generated in the atmosphere, similarly as electrons, positrons and gamma-rays are absorbed by several meters of rock material, therefore their contribution to the detector background is negligible. In very deep underground laboratories, such as Modane (4800 m w.e. – water equivalent) and Gran Sasso (3500 m w.e.) the muon component of the background is negligible (Laubenstein et al., 2004; Loaiza et al., 2015), and the dominant background contribution is from radioactive contamination of the shielding and construction materials of the detector itself (Breier et al., 2016a, 2016b). The primordial radionuclides in laboratory walls (^{232}Th, ^{238}U) produce neutrons in (alpha, n) reactions, plus gamma-rays, which may affect the detector background. The ^{238}U and ^{232}Th decay products include noble gas radon (^{222}Rn - radon and ^{220}Rn - thoron), which emanates from the walls and construction materials surrounding the detector, and their decay products (e.g. the gamma-emitters ^{214}Bi and ^{208}Tl, etc.) contaminate the detector environment. Although the Ge crystals are free of such contamination, surrounding materials (cryostat, window, front electronics, cables, connectors, shielding materials, etc.) are always contaminated. As radon may infiltrate into the sensitive volume of the detector, it represents usually the most dangerous source of background in underground detectors. To reduce the radon contribution requires an exhaustive control of the radon-tightness and radon-emanation from different parts of the detector, therefore a radon-free air factory, flushing the clean air around the detector should be installed in underground laboratories (Arnold et al., 2005, 2010).

The ^{40}K emits high-energy gamma-rays (1.46 MeV), and it is present mainly in glass of photomultipliers, which are usually outside of the sensitive volume of the detectors, therefore radiopurity limits for ^{40}K are not so strict as in the case of radon and its progenies. For example the main background contributions found in the NEMO-3 detector (the predecessor of the SuperNEMO) as determined during the search for neutrinoless double beta-decay in ^{100}Mo (Arnold et al., 2015) were from internal and external background sources of ^{214}Bi and ^{208}Tl. However, in the case of Ge detectors, the ^{40}K (present in construction parts of the detectors and in shielding) has proven to be the most difficult contaminant to be removed from the construction materials (Heusser et al., 2006; Budjáš et al., 2009).

The acceptable contamination levels differ between various experiments. In some cases they should be below 1 nBq g^{-1} of a material (Povinec, in press), close to the present detection limits of detectors. As an example we present radiopurity limits for the SuperNEMO detector, which are based on the goal to reach for the neutrinoless double beta-decay a half–life of $1 \cdot 10^{26}$ y (Arnold et al., 2010). The limit set for ^{222}Rn in the tracker chamber is 0.15 mBq m^{-3}, and 2 nBq g^{-1} for ^{208}Tl and 10 nBq g^{-1} for ^{214}Bi are the limits for radioactive contaminants in the detector (the isotope source, supporting foil, wires and walls of the tracker chamber). In the framework of the SuperNEMO experiment, underground HPGe gamma-spectrometry facilities in Modane (France) and in Boulby (UK), and surface ones in Bordeaux (France) and Bratislava (Slovakia) have been responsible for radiopurity measurements of construction materials (Povinec, in press). All materials (supporting foils, source frame, tracker wires, screws, glues, connectors, signal and high voltage cables, and other construction materials) have been screened using HPGe detectors with sensitivity down to about 10 nB g^{-1} for ^{238}U and ^{232}Th decay chains, and for ^{40}K.

A very sensitive detector (called BiPo-3) for measuring ultra low-levels of radionuclides on large thin surfaces (supporting foils, isotope sources) has been developed by the SuperNEMO collaboration (Barabash et al., 2016). This is the third generation of the detector (Agyriades et al., 2010), which uses the same principle to detect delayed beta-alpha coincidences of the ^{214}Bi - ^{214}Po cascades in the ^{238}U chain. The high-energy gamma-emitter ^{208}Tl in the ^{232}Th chain is analyzed via its parent, the ^{212}Bi. The detector consists of 2 face-to-face planar calorimeters made of pure aluminized polystyrene scintillators coupled to 5″ low radioactivity PMTs to detect the beta- and alpha-particles, and to measure time delays between the particle emissions, so radionuclides under investigation could be identified. The BiPo-3 detector with surface area of 3.6 m^{2} is operating since 2013 in the Canfranc Underground Laboratory in Spain. It is the most sensitive radiometric detector available at present for radiopurity measurements of large surfaces (Tables 1 and 2).

A comparison of experimental background gamma-spectra with Monte-Carlo simulated ones for large volume HPGe detectors operating in Modane (Breier et al., 2016a) and Gran Sasso (Breier et al., in press) underground laboratories has shown that the experimental spectra are by about two orders of magnitude above the simulated cosmic-ray spectra. This has been due to radioactive contamination of the detector environment by decay products of ^{238}U and ^{232}Th, and by ^{40}K. Further improvements in the design of ultra-low level HPGe spectrometers with even lower radioactive contamination should be therefore realized in future installations, otherwise the benefits of operating HPGe detectors in deep underground laboratories would be lost. Companies producing “low-background HPGe detectors” should more carefully control radiopurity of construction parts (cryostat, window, copper finger, preamplifier, cables, connectors, etc.), preferably using low-level HPGe spectrometers already installed in deep underground laboratories.

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