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Ultra-sensitive radioanalytical technologies for underground physics experiments

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Abstract
Assessment of radioactive contamination of construction materials used in deep underground experiments has been carried out using ultra-sensitive analytical methods such as radiometrics, inductively coupled plasma mass spectrometry (ICPMS), accelerator mass spectrometry (AMS), and neutron activation analysis. The lowest detection limits, < 1 nBq g^{-1}, has been obtained with ICPMS and AMS techniques.

Keywords Underground experiments · Radiopurity measurements · ^{238}U and ^{232}Th decay series · AMS · ICPMS · HPGe gamma-spectrometry

Introduction

Ultra-sensitive radioanalytical technologies have been playing an essential role in assessment of radioactive contamination of construction materials used in deep underground physics experiments, such as investigations of rare nuclear processes and decays—neutrinoless double beta-decay experiments (e.g. SuperNEMO [1], GERDA [2], MAJORANA [3], LEGEND [4]), neutrino physics experiments (e.g. BOREXINO [5]), as well as in searches for dark matter (e.g. in CRESST [6], EURECA [7]).

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Double beta-decay experiments represent the most frequently carried out underground experiments. Already 12 isotopes have been identified with half-lives ranging from 10^{19} to 10^{24} y, including ^{48}Ca, ^{82}Se, ^{96}Zr, ^{100}Mo, ^{116}Cd, ^{130}Te and ^{150}Nd which were studied in the NEMO-3 experiment ([8]). The double beta-decay is a second order process which is allowed by the Standard Model. As this is a very complex topic, we shall focus here specifically on the SuperNEMO experiment, which will search for neutrinoless double beta-decay of ^{82}Se, and its first module (called Demonstrator) is presently under construction in Modane underground laboratory [1, 9].

The search for neutrinoless double beta-decay represents a new challenge for underground experiments. If confirmed, the process would violate the lepton number conservation, requiring a Majorana neutrino (to be identical with its antineutrino), and giving unique information on the neutrino mass hierarchy. Neutrinoless double beta-decay experiments represent thus a new physics behind the Standard Model. With more than ten experiments which have been going on in several underground laboratories, only upper half-live limits in the 10^{24}–10^{26} y range have been reported till now (e.g. [2, 8]).

The aim of the present paper has been to review the sources of background in deep underground experiments and to discuss ultra-sensitive technologies for assessment of radioactivity of construction materials. At great depths

(about 4000 m of water equivalent, w.e.) contributions of cosmic rays to the detector background have been mostly negligible, therefore radioactive contamination of construction materials of detectors and surrounding laboratory walls have been dominant sources of background.

Sources of background in deep-underground experiments

The background in deep-underground experiments is determined by external and internal radiation sources [10–12]. The external background sources are represented mainly by:

1. Cosmic rays—primary protons and light nuclei interacting with atmosphere and producing secondary particles (mainly muons, neutrons and gamma-rays). Muons can penetrate even deep underground where they may interact with detectors or produce tertiary neutrons and gamma-rays in their interactions with detector shielding materials.
2. Gamma-rays and neutrons from (alpha, n) reactions originating in the laboratory environment (underground rocks, laboratory walls) due to its radioactive contamination (mainly ^{40}K, and ^{238}U and ^{232}Th decay series products).
3. Radon contamination of the laboratory air.

The internal contamination sources are represented mainly by:

1. Gamma-rays and neutrons originating in construction parts of the detector.
2. Radon contamination of the internal parts of the detector.

In very deep underground laboratories, such as Modane (4800 m w.e.) and Gran Sasso (3800 m w.e.), a contribution of the muon (hard) component to the detector background is usually negligible (e.g. [13–16]). The dominant background contribution is therefore from radioactive contamination of the shielding and construction materials of the detector itself [17, 18]. However, in experiments studying rare nuclear processes, even rare interactions of muons e.g. with shielding materials producing only a few neutrons per year, may results in important background contributions.

The primordial radionuclides in construction materials and 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. Their decay products (e.g. the ^{214}Bi and ^{208}Tl gamma-emitters, and others) are significant background sources of detectors. The Ge crystals are free of radioactive contaminations, however, surrounding materials (cryostat, window, front electronics, cables, connectors, shielding materials, etc.) are always contaminated by radionuclides.

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 of different parts of the detector. Usually a radon-free air flushing through the isolated box shielding with detector is a must in ultra-low-level counting [15].

The acceptable radionuclide contamination levels differ between various experiments. They should be usually close to 1 μBq kg^{-1} of a construction material [19], close to the present detection limits of HPGe detectors. For example, radiocontamination limits for the SuperNEMO detector, which are based on the goal to reach for the neutrinoless double beta-decay a half–life of 1×10^{26} y [1] are 0.15 mBq m^{-3} for ^{222}Rn in the tracker chamber, and 2 μBq kg^{-1} for ^{208}Tl and 10 μBq kg^{-1} for ^{214}Bi in the detector hardware (isotope sources, supporting foils, wires and walls of the tracker chamber). All construction materials must be therefore well screened for radioactive contamination before final assembly of the underground detector.

Radioanalytical technologies for radiopurity measurements

We shall focus in this paper on radiopurity measurements carried out by underground gamma-spectrometry, inductively coupled plasma mass spectrometry (ICPMS), accelerator mass spectrometry (AMS) and neutron activation analysis (NAA).

Underground radiometric technologies

Gamma-spectrometry has been the most frequently used method for screening of radioactive contamination of construction materials because of availability of large efficiency HPGe detectors (up to 200% relative to 7.6 cm in diameter × 7.6 cm long NaI(Tl) detectors), and their excellent energy resolution (2 keV @ 1.33 MeV gamma-rays of ^{60}Co). Typical detection limits are about 1 mBq kg^{-1}, however, special installations in deep underground laboratories with large HPGe detectors in ultra-low background assemblies could reach for a few kg samples detection limits of about 1 μBq kg^{-1} [20, 21] (Table 1).

Table 1 A comparison of detection limits

Radionuclide	Detection limit (μBq)					
	AMS[ah]	ICPMS[i]	BiPo-3[ch]	NAA[dh]	Alpha- spectrometry[ei]	Underground gamma-spectrometry[h]
^{232}Th	0.0002	0.003[b]	1.4	0.08	100	5000[f]
						2400[g]
^{238}U	0.0001	0.010[b]	1.6	0.2	100	4000[f]
						2000[g]

[a]Estimated from [31]
[b]Estimated from [28]
[c]Estimated from [24]
[d]Estimated from [38]
[e]Estimated from [39]
[f]Estimated from [14]
[g]Estimated from [20]
[h]Without pre-concentration chemistry
[i]With pre-concentration chemistry

A comparison of experimental background gamma-spectra with Monte-Carlo simulated ones for large volume HPGe detectors operating in Gran Sasso (depth of 3800 m w.e.; [17]) and in Modane (depth of 4800 m w.e., [18]) underground laboratories has shown that the experimental spectra are by more than two orders of magnitude above the Monte Carlo simulated cosmic-ray background spectra. This has been due to radioactive contamination of the detector environment (cryostat, detector holder, cold finger, lead shield) by ^{40}K and by decay products of ^{238}U and ^{232}Th. For example, for the Modane HPGe detector of relative efficiency of 160%, the Monte Carlo model predicted as the most important radionuclides ^{214}Bi and ^{208}Tl present in the lead shield (about 9 mBq kg^{-1}), in the cold finger, cryostat, and detector holder (< 4 Bq kg^{-1}) [18]. Radionuclides present in the lead shield because of its large mass dominated in the background of the HPGe detector. On the other hand, radionuclides present in the cryostat, cold finger, and the detector holder because of their smaller masses had smaller contributions to the detector background, although they were much closer to the HPGe crystal.

The lowest background continuum was simulated from ^{40}K, followed by the ^{232}Th chain (Fig. 1). The ^{238}U chain had a main contribution to the detector background, except for the energies above 2000 keV, where the ^{232}Th chain dominated due to ^{208}Tl (2.6 MeV gamma-rays). Further improvements in the design of ultra-low level HPGe spectrometers operating deep underground are therefore required; otherwise the benefits of operating HPGe detectors in deep underground laboratories would be lost.

Figure 2 documents that even materials situated far from the sensitive volume of the detector (e.g. an iron shielding) should also be free of radioactive contamination [22]. Monte Carlo simulations carried out for the SuperNEMO Demonstrator indicate that 100 Bq kg^{-1} of ^{208}Tl present in the external iron shielding will have very negative impact on the detector background, as the environmental gamma-flux in the Modane underground laboratory with iron shielding of 18 cm thickness will be by about seven orders of magnitude above the acceptable limit [22].

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 [23, 24]. This is the third generation of the detector [25], 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 low-radioactivity photomultipliers to detect the beta- and alpha-particles, and to measure time delays between the particle emissions for radionuclide identification. The BiPo-3 detector with surface area of 3.6 m^2 has been operating since 2013 in the Canfranc underground laboratory in Spain. It represents the most sensitive radiometric detector available at present for radiopurity measurements of large surfaces (Table 1) [23, 24].

New generations of underground experiments will require, however, further developments of ultra-sensitive radioanalytical technologies. The big problem of the

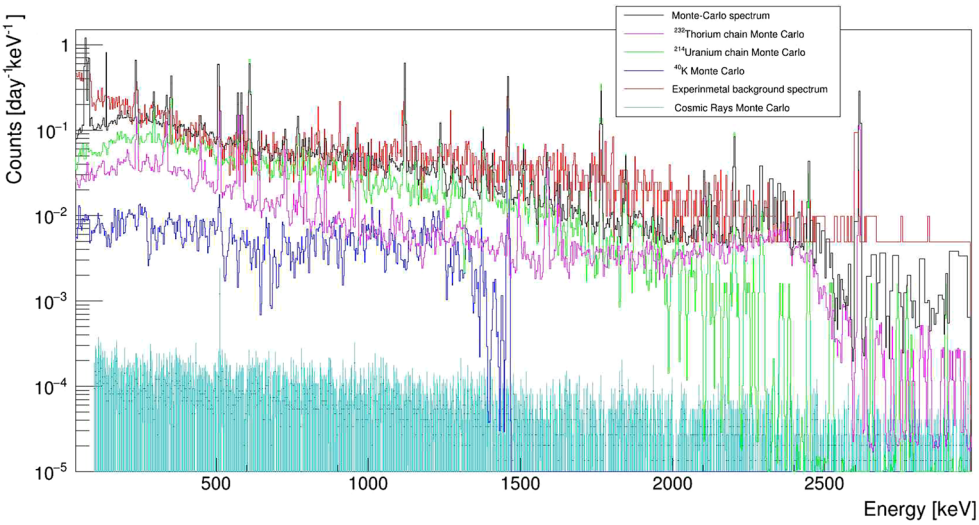

Fig. 1 Experimental and Monte Carlo simulated gamma-spectra for HPGe detector of 160% relative efficiency operating in the Modane underground laboratory (4800 m w.e.)

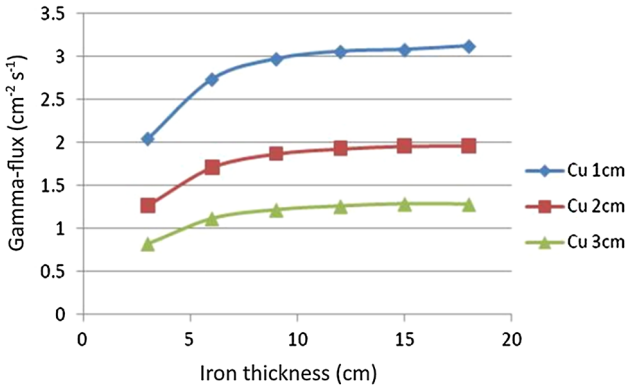

Fig. 2 Monte Carlo simulation of the gamma-flux (0–4 MeV) in the SuperNEMO Demostrator shielded with of up to 18 cm of iron with additional copper layers. The iron was intentionally contaminated by 100 Bq kg^{-1} of ^{208}Tl

gamma-ray spectrometry for ultra-low-level activity screening is the fact that the same radionuclides, which should be analyzed in construction materials, are also found in the detector background. New developments in analytical techniques with detection limits < 1 nBq g^{-1} (we prefer to use this activity unit as usually samples available for analysis have only a few grams) are required to meet background constrains for radiopurity of construction materials used in underground experiments for investigation of rare nuclear processes. Therefore, alternative methods for analysis of primordial radionuclides (mainly ^{238}U and ^{232}Th and their decay products) in construction materials have been suggested, changing radioactive-decay counting to direct mass-spectrometry measurements (as radionuclides under investigations would be stable ones). New technologies for analysis of these primordial radionuclides based on AMS and ICPMS are under development which should improve detection limits by several orders of magnitude when compared with underground gamma-spectrometry.

Developments in ICPMS technologies

ICPMS is one of the mass-spectrometry techniques for analysis of medium- and long-lived radionuclides which has made great development in recent years, both in sensitivity and operational capacity. The main advantages of ICPMS are in capability to determine different classes of radionuclides to fg levels, to analyze even very small samples rapidly in a few minutes and at the low cost. New generation of sector field instruments with double-focusing and even multi-collector systems has considerably improved sensitivity and precision over traditional quadrupole machines. With introduction of the present state of the art new machines, the ICPMS has been developing into a powerful technique competing well with AMS in many respects. ICPMS has been used in both higher-resolution and lower-resolution modes. The higher-resolution mode has the advantage of addressing polyatomic interferences, although it cannot solve all the problems with isobaric interferences, which may be caused by incomplete separation chemistry. On the other hand, maximum sensitivity can be reached in the lower-resolution mode. Thus, a combination of the two modes appears to be the best compromise for reaching maximum sensitivity and controlling interferences. ICPMS requires careful purification procedures as it is not free of matrix and isotopic effects. A steady increase over the past decade in radioanalytical applications using ICMPS has resulted in a decrease in both the price of instruments and detection limits. The ICPMS has proved to be powerful analytical technology because of its high sensitivity and multi-isotopic capability. Nevertheless, there could be problems with molecular, isobaric and isotopic

interferences even if careful purification procedures are used (e.g. [26, 27]). ICPMS measurements of ^{238}U and ^{232}Th in ultra-clean copper samples were recently reported with detection limits < 1 nBq g^{-1} [28].

Developments in AMS technologies

Similarly, as in the case of ICPMS, the AMS technology is based on direct counting of atoms. The sensitivity is achieved by ionizing sample atoms efficiently, accelerating them to high energies and counting individual isotopes using detection techniques developed for nuclear and particle physics. Accelerating ions to energies above 1 MeV and using very effective methods of background reduction, made the AMS technology the most sensitive for analysis of long-lived radionuclides. Isobar problems and formation of hydrates, which represent serious limitations in ICPMS applications, have better solutions in AMS measurements. The greatest achievement in the AMS sector occurred from the technical point of view when small tandem accelerators with nominal voltage < 1 MV became available, which replaced large accelerators (5–10 MV), previously used in nuclear physics experiments. Generally, the most sensitive technique available at present for analysis of long-lived radionuclides is AMS which gives the lowest detection limits, several orders of magnitude lower than the radiometric methods [29, 30]. In the case of radiopurity measurements 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. From this point of view the AMS could be the preferably technique as samples such as copper, steel, etc. can be directly used as targets in ion sources. Nondestructive AMS measurements of ^{238}U and ^{232}Th in copper samples were recently reported with detection limits down to 1 nBq g^{-1} [31].

One of the goals of recently established Centre for Nuclear and Accelerator Technologies (CENTA) at the Comenius University in Bratislava has been development of the AMS technique for radiopurity measurements at levels < 1 nBq g^{-1} [32–34]. We focused in radiopurity measurements on production of suitable targets, on optimization of the sputtering ion source, on optimized acceleration of ions in the Pelletron tandem and the post-acceleration ion analyses. Copper has been chosen for first radiopurity investigations as this material is usually closest to underground detectors, and it may have dominant impact on the detector background. Copper samples can be directly accommodated as targets in the ion source of the tandem accelerator, therefore non-destructive analyses could be possible. Uranium and thorium oxides or uranium and thorium compounds with copper were considered as possible negative ions extracted from the copper targets in the ion source of the tandem accelerator. The UO and ThO_2 ions with masses of 254 and 264, respectively, should be expected in the first mass peak, they would be masked, however, with ion clusters of ^{63}Cu and ^{65}Cu ($^{63}Cu_3^{65}Cu$, etc.) with masses of 254 (256), 319 (325) and 374 (388). A more favorable case should be therefore a formation of negative molecules of UCu^- or $ThCu^-$ which would fall into the mass windows of 301 and 295, respectively, where they would be free of copper cluster interferences (Fig. 3). Further studies are on the way to establish best conditions for AMS analysis of ^{238}U and ^{232}Th in copper.

Neutron activation analysis (NAA)

Another promising radioanalytical technique is based on NAA of long-lived primordial radionuclides by neutron irradiation of samples in a nuclear reactor. The NAA technique was used in the past for analysis of ^{129}I, ^{238}U, ^{232}Th and other long-lived radionuclides in environmental samples [35–37], however, after the invention of AMS and ICPMS these new technologies took over the NAA. Due to radiopurity problems in ultra-low-level gamma-spectrometry and necessary pre-concentration chemistry required for ICPMS (and sometimes also for AMS), a renaissance of NAA may be expected as an alternative method for analysis of primordial radionuclides in detector construction materials or in isotopic sources used for double beta-decay experiments [38]. This technique may be especially advantageous as no pre-irradiation chemistry is required, and on the other hand, a post-irradiation chemistry can improve detection limits. The NAA method shows promising results, however, more work is needed, especially in the post-irradiation chemistry to reach detection limits comparable with AMS and ICPMS.

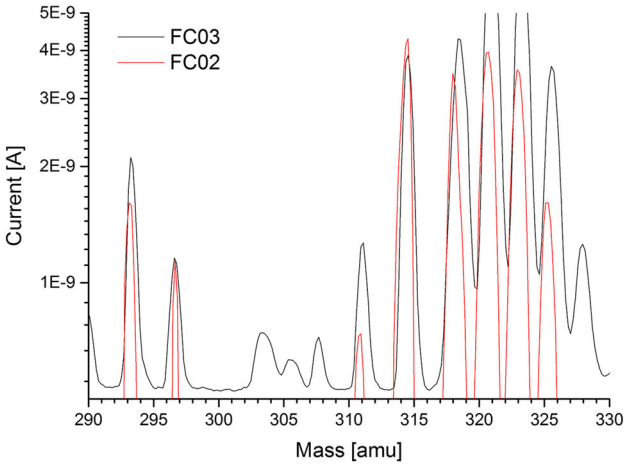

Fig. 3 Mass scan of uranium and thorium compounds produced at the CENTA facility. UCu^- (301) and $ThCu^-$ (295) compounds could be a good choice

Table 2 Comparison of results in recent radiopurity measurements (in nBq g^{-1})

Radionuclide	Electrolytic copper	^{82}Se enriched A + PVA glue	^{82}Se enriched B
^{232}Th	< 2000 GS	24 ± 20 BiPo[b]	7700 ± 1200 RNAA
	< 1000 GSU		
	90 ± 20 ICPMS		
	300 ± 40 RNAA		
	1600 ± 200 RNNA		
^{238}U	< 2000 GS	< 300 BiPo[b]	< 12,000 RNAA
	< 1000 GSU		
	160 ± 50 ICPMS		
	250 ± 60 RNAA		
	< 1600 RNAA		

GS gamma-spectrometry, *GSU* gamma-spectrometry underground, *RNAA* radiochemical neutron activation analysis, *BiPo*[b] [24]

The present state-of-the-art detection limits of the ultra-sensitive detection methods are compared in Table 1. It can be seen that the leaders are AMS, ICPMS and BiPo-3. In the mass-spectrometric sector, the lowest detection limits for ^{238}U and ^{232}Th analysis in construction materials have been reached by AMS (down to 0.1 nBq) [31]. It should be stressed that this method in analysis of specific samples (e.g. copper) can be a non-destructive one. AMS is also profiting from analysis of very small, even sub-gram samples. The isotope dilution ICPMS, supported by ultra-clean sample preparation, combined with anion exchange pre-concentration of U and Th, and with removal of matrix interferences (developed in the framework of the Majorana neutrinoless double beta-decay experiment [3, 4]) has detection limit about 1 nBq [28].

The BiPo-3 detector, as documented in Table 1, is the most sensitive method in the radiometric sector for analysis of ^{238}U and ^{232}Th in thin samples (isotopic sources, foils) deposited on large surfaces ($\sim$ 1 m^2). Detection limits about 1 nBq have been obtained for both radionuclides [23, 24]. The semiconductor alpha-spectrometry is a traditional radiometric method applied for analysis of ^{238}U and ^{232}Th samples of various origin, giving medium detection limits (0.1 mBq), requiring, however, a heavy radiochemical treatment of samples. Mass-spectrometry methods, ICPMS and AMS, have, however, detection limits by 10^4–10^6 times better.

The most frequently non-destructive technique for analysis of ^{40}K and ^{238}U and ^{232}Th (e.g. via their decay products ^{214}Bi and ^{208}Tl, respectively, assuming thus radioactive equilibrium, which need not be always fulfilled), is the underground HPGe gamma-spectrometry as it can benefit from accommodation of large volume samples (> 1 kg) around the detector. The detection limits of about 1 μBq kg^{-1} have been obtained in the Gran Sasso [16, 20] and Modane [14] underground laboratories when analyzing large mass samples. As already mentioned, the main problem of gamma-spectrometry is, however, that the same radionuclides to be analyzed in unknown samples represent the main contributions to the spectrometer background.

Results of recent radiopurity measurements

Table 2 lists a few results of recently carried radiopurity measurements of isotopic sources and construction materials. Results of ^{238}U and ^{232}Th analyses in electrolytic copper indicate that a reasonable agreement has been obtained between the ICPMS and INAA results, while both surface and underground laboratories produced only detection limits. The analyzed electrolytic copper has acceptable radioactive contamination for internal shielding of underground detectors.

Large activity variations have been observed between enriched selenium samples, which require more analytical work.

Conclusions

In the mass spectrometry sector, represented by AMS and ICPMS, we have seen the most exciting, really a revolutionary breakthrough in radiopurity measurements of long-lived ^{238}U and ^{232}Th radionuclides with detection limits < 1 nBq g^{-1}, requiring only small samples of gram size. Specifically, AMS systems have been operating at the highest sensitivities with small matrix and interference effects. In the radiometric sector the most sensitive instrument has been the BiPo-3 spectrometer (developed in the framework of the SuperNEMO collaboration) with detection limits about 1 nBq g^{-1}. Underground gamma-spectrometers (which have been dominating in the analysis

of short- and medium-lived radionuclides emitting gamma-rays) could compete with the mass spectrometry techniques only when analyzing large mass samples (> 1 kg). Specific attention was given to Monte Carlo simulations of background of HPGe detectors, which indicated that more careful selection of their construction parts should be carried out.

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