Low-level techniques applied in experiments looking for rare events

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Abstract. Techniques allowing investigation and reduction of backgrounds are crucial for all experiments looking for rare events. In this paper investigations of ^{222}Rn, germanium spectroscopy and noble gas mass spectrometry will be described. Selected measurements and their applications in the Borexino and GERDA experiments will be discussed.

1. Introduction

Low-level techniques could be defined as techniques allowing investigations of natural and man-made radioisotopes at very low activities (concentrations). One can consider here studies of radioactive noble gases (diffusion, emanation), germanium spectroscopy, mass spectrometry (ICP-MS, noble gas mass spectrometry), analysis of surface contaminations (α- and β-spectroscopy), investigation of purification techniques (gases, liquids), constructing algorithms for background events rejection (time coincidences, pulse shape analysis) and modelling of backgrounds using experimental results and Monte Carlo simulations. In the following chapters as selected topics radon studies, germanium spectroscopy and noble gas mass spectrometry will be discussed. Some measurements performed in the frame of the Borexino [1] and GERDA [2] experiments will be described.

2. Studies of ^{222}Rn

^{222}Rn is one of the most dangerous isotopes for many low-background experiments. It belongs to the ^{238}U chain and as an inert noble gas it has typically high mobility (diffusion coefficient) allowing easy escape from bulk materials and diffusion into active parts of detectors. Together with the short-living daughters ^{222}Rn generates radiations with a wide energy range. Through a broken equilibrium in the chain (^{210}Pb level) surface contamination with long-living heavy metals can also cause problems (e.g. it may appear some years after cleaning as ^{210}Po).

Activity of ^{222}Rn can be measured using for example low-background proportional counters designed originally for the GALLEX/GNO experiment [3]. They are hand-made out of very pure quartz and have an active volume of about 1 cm^3. The background and detection efficiency (for alpha particles) for energies above a 50-keV threshold is ~1 count per day and 50 %, respectively. Counters are filled with radon samples using a special gas filling line.

^{222}Rn emanation can be studied using emanation chambers [4]. Two stainless steel electro-polished and completely metal-sealed vessels of the volume of 20 and 80 l are available. For samples up to 1 l volume small glass vials could be used. The absolute detection limit obtained for emanation tests is at the level of 100 μBq (~50 atoms).

To measure radon concentration in gases a system called MoREx (Mobile Radon Extraction Unit) has been constructed [5]. It takes the advantage of the ^{222}Rn pre-concentration in charcoal traps at low temperatures. In connection with low-background counting it is possible to analyze gases contamination down to ~0.5 μBq/m^3 (STP).
To analyze ^{222}Rn and ^{226}Ra in the water STRAW (System for the ^{222}Rn and ^{226}Ra Assay of Water) can be used [6]. The sensitivities are at the level of 0.1 and 0.8 mBq/m^3 for radon and radium, respectively.
In many cases it is also interesting to know radon diffusion through different materials. This knowledge helps to design proper barriers protecting active parts of the detectors from radon moving from outside. An apparatus for this kind of research was constructed and successfully used in the frame of the Borexino experiment to measure the diffusion and solubility coefficients for radon barrier- and inner vessel foils [7], also as a function of the relative humidity of the foils vicinity [8]. The diffusion measurements could be performed at the 10^{-13} cm^2/s level.

3. Noble gas mass spectrometry

Other noble gases, which can be considered as sources of background e.g. in Borexino are ^{39}Ar or ^{85}Kr present in nitrogen as traces. In principle their concentrations could be measured as that for radon, using low-background proportional counters, however the pre-concentration does not work for them efficiently enough, and achievable detection limits would be here much poorer. Other solution is to look for natural Ar and Kr using a highly sensitive noble gas mass spectrometer. Concentrations of ^{39}Ar and ^{85}Kr can be later reconstructed taking into account their fairly known specific activities. Ar/Kr is detected using a VG3600 mass spectrometer devoted to investigate rare gases at low concentrations in terrestrial and extraterrestrial samples. Besides the mass spectrometer, the system consists of a sample preparation and a sample purification section. In the first one the volume of the investigated sample can be determined (or if necessary divided into smaller fractions), in the second one the sample is purified from reactive gases before it is introduced into the spectrometer. For final purification Al-Ti getter pumps are used. The calibration is performed with air or Ar/Kr standards. Taking into account the intrinsic background of the whole system and measuring 1 cm^3 of nitrogen we could achieve the following detection limits: 1×10^{-9} m^3/m^3 for Ar in N_2 (~1.4 nBq/m^3 for ^{39}Ar in N_2) and 1×10^{-13} m^3/m^3 for Kr in N_2 (~0.1 μBq/m^3 for ^{85}Kr in N_2) [9].

4. Germanium spectroscopy

Germanium spectroscopy is one of the most powerful tools to identify γ emitters (U/Th chain, ^{60}Co, ^{40}K). The strength of this technique comes from the excellent energy resolution of germanium diodes and from their very low intrinsic background. External background can be usually effectively shielded or vetoed. In order to reach high detection sensitivities one should assure also a large sample chamber and proper determination of efficiencies (measurements or simulations).
Max Planck Institute operates several detectors at different locations. The most sensitive world-wide are GeMPI-type spectrometers located at the Gran Sasso laboratory in Italy. Due to the extremely low background und large capacity, for high density materials it is possible to reach the detection limit at the level of 10 μbq/kg (U/Th) [10]. Some detectors are also run in the MPI underground laboratory (15 m w.e.), however because of the shallow depth the lowest measurable concentrations are in the range of 1 mBq/kg [11].

5. Applications in Borexino

5.1. ^{226}Ra in the inner vessel foil

The purity requirements for the construction materials in Borexino, especially for the scintillator and its nylon vessel were very strong. The allowed contamination for the second one with ^{238}U was at the level of 1 ppt, what corresponds to ~12 μBq/kg for ^{226}Ra (in secular equilibrium). This limit could not be reached with any Ge spectrometer (foil is a low density material) and a new technique had to be developed. It is based on the combination of the ^{222}Rn diffusion and emanation measurements and uses

the fact that the radon diffusion coefficient in nylon depends very strongly on its water content. Measuring emanation rates for dry and wet material one could determine the surface and bulk ^{226}Ra concentrations at the levels of 0.5 μBq/m^3 and 10 μBq/kg, respectively. In the frame of those investigations a proper material for the vessel was identified [12]. It was also shown that ^{238}U and ^{226}Ra in nylon was in disequilibrium.

5.2. Nitrogen

One of the most important applications of ultra-pure nitrogen in Borexino was the scintillator purification. The purity requirements for N_2 were the following: 7 μBq/m^3 for ^{222}Rn, 0.4 ppm and 0.2 ppt for Ar and Kr, respectively. Such a gas was delivered by the SOL company after thorough purity and transportation tests, where possible (re-)contamination sources were eliminated [13]. Beside mentioned ultra-pure- Borexino uses also ^{222}Rn-free- [5] and boil-off nitrogen (obtained from technical quality LN_2) produced with the rate up to 100 m^3/h.

6. Applications in GERDA

6.1. Germanium spectroscopy of steel samples

As in many other experiments germanium spectroscopy was applied in GERDA to screen construction materials like steel (for the cryostat) or copper (for detector holders and shields). The screening was done in Heidelberg and in Gran Sasso using the GeMPI detectors. One of the most important achievements was finding of steel with surprisingly low U/Th contamination of the order of < 0.1 – 1 mBq/kg [14]. Batches with the lowest concentrations (< 0.1 – < 0.4 mBq/kg) were used to construct the cylindrical part of the cryostat, the others for the top and bottom heads. Low U/Th content in steel allowed thickness reduction of the copper shield installed inside the vessel (keeping the same background index).

6.2. Surface cleaning tests

Cleanliness of surfaces located close to the Ge detectors is one of the key issues in GERDA. This concerns mainly the long-living radon daughters deposited for example on the copper support structure of the diodes. To test different cleaning procedures some Cu disc were loaded with radon daughters and then cleaned applying selected methods. Removal efficiencies for ^{210}Pb, ^{210}Bi and ^{210}Po were investigated using γ-, β- and α-spectroscopy, respectively. The main result from this research is that the electro-polishing usually removes effectively all mentioned isotopes, while etching with acids does not work for ^{210}Po practically at all [15]. Further tests are planned for steel samples.

7. References

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