Records in Ultra Low Radioactivity Measurements with Neutron Activation Analysis

R. v.Hentig [a], T. Goldbrunner, G. Angloher [a] and F. v.Feilitzsch [a]

[a]Physik Department E15, Technische Universität München,
James Franck Straße, D-85748 Garching

Neutron Activation Analysis has emerged to be an analytical method sensitive enough to detect fg/g traces of unstable primordial nuclides in complex matrices. Especially low count rate experiments in the field of solar neutrino physics and dark matter search can profit from the detection capabilities of this method which had been unattained so far. This gain in sensitivity has been achieved by combining neutron activation, radiochemical separation methods, and efficient low level counting systems at the new underground laboratory of the accelerator laboratory in Garching.

Recent improvements which have been made in the purification and analysis of the liquid scintillator, as foreseen for the solar neutrino experiment BOREXINO, are being presented as an example in this paper

1. INTRODUCTION

The upcoming generation of low background experiments in the fields of dark matter search, double beta decay and neutrino physics has triggered considerable efforts to identify sources of background which affect the detection of the interesting type of event [1, 2, 3]. One class of background usually originates from primordial, radioactive elements which are ubiquitous in the environment and thus may influence the detection of the signal by their decay due to their presence in the detector. Even traces of these elements in detector components may lead to a contamination of the experiment which makes it desirable to monitor the radioactivity of every part which is being used for the construction for such kind of experiment.

Due to the long half lives of primordial elements, trace elements can not only be detected by their radioactive decay but also by trace analytical methods. In order to give an overview, the specific activity of some primordial radioactive elements which may affect low count rate detectors is compiled in table 1.

Table 1
Activity of primordial elements in the upper earth crust. Calculated from data given in [4].

Element	Activity [Bq/kg]
Cadmium	$3.2\cdot10^{-7}$
Indium	$2.5\cdot10^{-5}$
Lanthanum	$2.5\cdot10^{-2}$
Lutetium	$2.6\cdot10^{-2}$
Potassium	$7.7\cdot10^{2}$
Rubidium	7.9
Thorium (^{232}Th)	$3.0\cdot10^{1}$
Uranium (^{238}U)	$2.2\cdot10^{1}$

One method for the determination of trace elements is neutron activation analysis. It is especially suited for the detection of trace impurities in matrices with low neutron capture cross sections or short half lives of isotopes produced from the matrix. Whereas primordial trace elements have a low specific activity, most of them are converted into isotopes with a high specific activity by neutron irradiation. The isotopes produced via neutron capture can then be detected by their decay. This leads to an increase in sensitivity for most isotopes compared to conventional radioanalytical methods which are based upon the detection of the decay of the primordial isotope.

2. EXPERIMENTAL

The presented work was initiated by the solar neutrino experiment BOREXINO which imposes stringent demands on the radiopurity of certain components. The requirements for the innermost part of this detector are compiled in table 2 to show which limits of detection analytical methods have to compete with. These requirements might be comparable for other experiments although each one has special requirements due to the sensitivity for different types of particles and different rejection capabilities.

The development of the analytical methods was guided, besides the sensitivity, by the reproducibility of the analyses. The strategy applied now is to combine and optimise all factors which contribute to these demands.

Table 2
Required limits for the BOREXINO liquid scintillator. For the U, Th decay chains, secular equilibrium is assumed [5].

Element	Limit [g/g]
Cadmium	$2.8 \cdot 10^{-8}$
Indium	$2.4 \cdot 10^{-11}$
Lanthanum	$1.2 \cdot 10^{-11}$
Lutetium	$3.7 \cdot 10^{-14}$
Potassium	$7.8 \cdot 10^{-14}$
Rubidium	$3.2 \cdot 10^{-13}$
Thorium	$2.4 \cdot 10^{-15}$
Uranium	$9.4 \cdot 10^{-17}$

Methods for the analysis of ultra pure aluminium, nylon, water, organic liquids and components of liquid scintillators have been developed.

For an analysis large samples of ultra pure material, being suited representatives of the materials investigated, are prepared in a clean environment and enclosed in synthetic quartz containers for the irradiation at the research reactor in Munich.

They being exposed to the highest neutron flux available (~10^{13} $s^{-1}cm^{-2}$) for up to 150 hours and kept in the reactor pool for the decay of the short lived radio nuclei for typically one day.

The irradiation time is limited due to the radiolysis of samples consisting of organic components as liquid scintillators or plastics. The separation of matrix activity which is produced from the matrix or other trace elements contained in the sample, is done in a radiochemical laboratory next to the reactor. Highly selective ion exchange and ion adsorption techniques with high quantitative yields are employed therefore.

By splitting up the activity of the sample into different groups, the sensitivity is considerably improved, as the activity contributing to the background during counting can be separated and each radioisotope of interest can be prepared for counting in a specific way.

After separation one obtains samples containing the activity of the indicator radionuclides in the µBq to mBq range which are counted by effective low background Ge detectors in the underground laboratory as the sensitivity after the radiochemical separation is often limited by the background of the Ge spectrometer itself.

The underground laboratory in Garching, were the detectors are operated, has an overburden of approximately 1500g/cm², shielding the detectors effectively from the hadronic component of the cosmic radiation. The three HPGe spectrometers used, are shielded by 150mm of lead with an inner layer of 50mm low activity lead, which has an intrinsic ^{210}Pb of 27mBq/g. As an example for the background reduction by the improved shielding two examples are given in figure 1 and figure 2. On the outside the lead shields are surrounded by 50mm plastic scintillator panels serving as an active veto against cosmic muons.

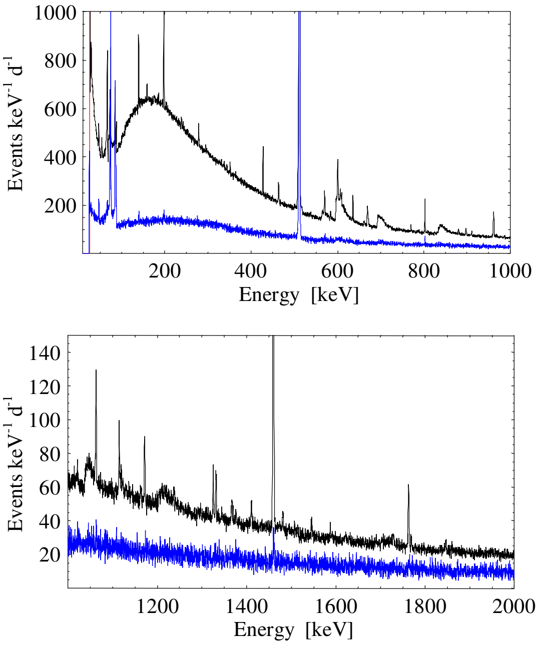

Figure 1 Background of a 3309g p-type HPGe detector (152% rel. eff.) with 200g/cm² overburden with a lead shield (upper line). The lower line is the spectrum of 1500g/cm² overburden with an additional low activity lead shield.

The counting method chosen for the detection of the indicator radionuclide can be adapted to the decay of the radionuclide leading to a further increase in sensitivity. Counting methods employed are β-γ coincidences for ^{233}Pa and β-γ-conversion electron coincidences for ^{239}Np [6, 7], the indicator radio nuclides for ^{232}Th and ^{238}U, which can improve the signal/noise ratio considerably and lead to an essentially background free measurement of the activity.

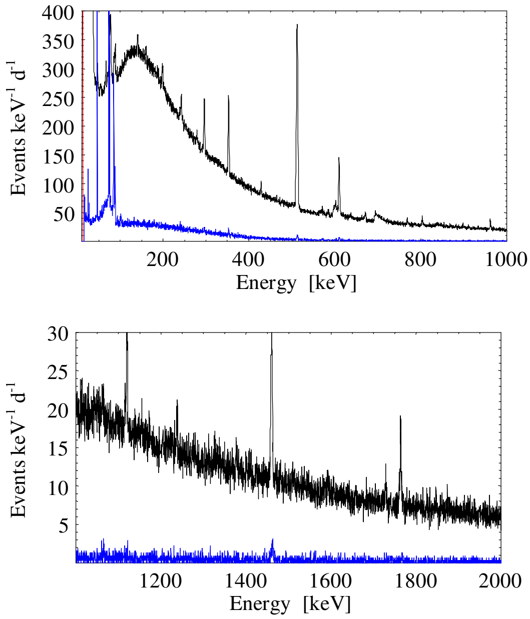

Figure 2 Background of a 600g n-type HPGe detector with 200g/cm² overburden and a lead shield (upper line). The lower line is the spectrum of 1500g/cm² overburden with an additional low activity lead shield. The detector is surrounded by an active μ-veto which has an efficiency >95%.

Besides this the correlation of photons and delayed events, including conversion electrons, X-rays and Auger electrons, can be used for a further reduction of background in the β-γ-conversion electron counting mode. The muon correlated background in these measurements can be reduced effectively by the muon veto which discriminates more than 95% of the particles traversing the detector.

3. RESULTS AND DISCUSSION

The investigated detector components included different kinds of nylon, aluminium, water and organic liquids which underwent different analytical procedures due to their manifold properties.

The most sensitive limits were achieved for an organic liquid which serves as a constituent of liquid scintillators. Water which has a much higher solubility for ionic impurities, usually contains also an increased amount of trace elements compared to the organic liquids investigated. Besides the primordial elements mentioned above, the samples were also analysed by their trace content of stable elements. These elements can give an indication to contaminations of dispensed particulate matter although they do not affect the experiment.

Table 3
Limits (90% C.L.) of trace element contaminations in Phenyl-ortho-xylylethane.

Element	Concentration [g/g]
Cadmium	$< 8.3\cdot10^{-15}$
Indium	$< 1.2\cdot10^{-13}$
Potassium	$< 6.1\cdot10^{-12}$
Lanthanum	$< 4.3\cdot10^{-16}$
Lutetium	$< 3.8\cdot10^{-16}$
Rubidium	$< 1.1\cdot10^{-13}$
Thorium	$< 1.8\cdot10^{-16}$
Uranium	$< 1.0\cdot10^{-17}$

Table 4
Limits (90% C.L.) on the decay rates of primordial nuclides in Phenyl-ortho-xylylethane.

Element	Activity [1/(kg a)]
Cadmium	$< 5.4\cdot10^{-3}$
Indium	$< 1.6\cdot10^{-3}$
Potassium	< 3.8
Lanthanum	$< 1.0\cdot10^{-4}$
Lutetium	$< 1.2\cdot10^{-4}$
Rubidium	< 19
Thorium (^{232}Th)	$< 2.5\cdot10^{-3}$
Uranium (^{238}U)	$< 3.7\cdot10^{-4}$

This makes the analytical method described an independent tool for the analysis of background events in low count rate detectors [8, 9]. The sensitivity to ^{238}U and ^{232}Th is another important characteristic of the analysis as some low background detectors can not measure all isotopes in the decay chain and secular equilibrium not always can be assumed. This makes it often indispensable to determine the activity of the parent nuclides independently in order to understand the residual background.

The exceptional purity of the investigated organic liquids shows the suitability of these components for the use in low background experiments. There they can serve as a shield or as a target [10].

The analytical method presented was used for the selection of materials and detector components foreseen for BOREXINO as well as for the test of cleaning procedures. The purification capabilities of liquid-liquid and liquid-solid extraction were also revealed for organic liquid scintillators which were under investigation in the 4t BOREXINO prototype detector [11, 12].

With the exception of potassium the limits of detection achieved for the primordial elements are in the range requested by BOREXINO. An improvement of this detection limits would be desirable as it is ubiquitous and occurs in a relatively high abundance in the environment which makes it, besides uranium and thorium, the most critical elements for the considerations of radiopurity.

4. OUTLOOK

In the near future one of the HPGe detectors will be equipped with an additional Compton-suppression system which will be used for the analysis of solids. It will improve the detection capabilities for plastics and organic solids as these matrices often contain trace amounts of Na and Br which impair the detection limits for other elements. The Compton-suppression system will not only suppress the Compton background but it will also reduce the background originating from isotopes decaying by the emission of two or more photons.

AKNOWLEDGEMENT

We are indebted to Prof. F. Baumgärtner and Prof. M. Schuster for their advise. We thank the accelerator laboratory and the Sonderforschungsbereich 375 for the financial support. One of us (R.v.H.) appreciates the grant he received from the Freistaat Bayern.

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