Developing measurements on activated samples in order to achieve sensitivity below ppt level

Low Radioactivity Techniques 2019
Laboratorio Subterráneo de Canfranc (LSC), Spain
Introduction

The radio purity of materials is the essential condition upon which rely the latest experiments of Rare Events Physics.

The greatest risk
radioactive background overlap the observable energy regions of interest

Fundamental a selection of the different components of the detector

Typical requirements for contamination level of detector materials

\[ ^{232}\text{Th} < 10^{-12} \text{ g/g (4 uBq/kg)} \]
\[ ^{238}\text{U} < 10^{-12} \text{ g/g (12 uBq/kg)} \]
\[ ^{40}\text{K} < 10^{-12} \text{ g/g (270 uBq/kg)} \]
Introduction

In this context is crucial to have tools able to achieve that sensitivity

For this purpose...

we have developed two detectors in ultra-low background configurations

In order to perform measurements on activated sample

This methodology of measurements allowed us to achieve high sensitivity on liquid and solid samples
Neutron Activation Analysis (NAA)

The neutron activation process consists in the production of unstable isotopes through neutrons absorption by the nuclei present in the sample.

\[
\frac{A}{Z}X + n \rightarrow \frac{A+1}{Z}X \xrightarrow{\beta^-} \frac{A+1}{Z+1}Y^* \rightarrow \frac{A+1}{Z+1}Y + \gamma
\]

The NAA technique consists of several steps:

- Sample exposure to a neutron flux
- Extraction of the irradiated sample and measurement of induced \( \gamma \) radioactivity (HPGe-detector)
- Determination of the quantity of precursor element \( \frac{A}{Z}X \)

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NAA sensitivity

High neutron capture cross section

\[ \frac{AX}{Z} + n \rightarrow \frac{A+1}{Z+1}Y + \gamma \]

Neutron flux

\[ \phi \approx 10^{12} \div 10^{13} \text{ cm}^{-2} \text{s}^{-1} \]

“Convenient” daughter nucleus (γ emission, half-life time, high BR)

\[ n + ^{41}K \rightarrow ^{42}K \xrightarrow{\beta^-} ^{42}Ca + \gamma(1524keV \text{ - BR 17%}) \]

\[ n + ^{238}U \rightarrow ^{239}U \xrightarrow{\beta^-} ^{239}Np \xrightarrow{\beta^-} ^{239}Pu + \gamma(106keV \text{ - BR 26%}) \]

\[ n + ^{232}Th \rightarrow ^{233}Th \xrightarrow{\beta^-} ^{233}Pa \xrightarrow{\beta^-} ^{233}U + \gamma(311keV \text{ - BR 38%}) \]

interferences in the matrix

Care in the sample preparation

Detector for measure
induced Y radioactivity

High Efficiency

Low background in the region of the gamma emission

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Ge-Ge HPGe detector

2x GMX detectors

Coaxial detector (n-type)

Relative efficiency for each detector: 100%

Energetic range 17-3200 keV

Selection of materials has been done for the detectors construction:

End-Cap, cryostat and holder are in high purity material

Preamplifier and the HV Filter has been remotized
Ge-Ge HPGe: Background reduction

Counts keV

GMX2200
GMX2019

Sample

Copper 15cm
Lead 20cm

Background suppression ~40%
Anticoincidence technique
Plastic scintillator veto

Shielding:

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Ge-Ge HPGe: DAQ

Dedicated acquisition system

A narrow time windows allow to acquire signals registered

GMX2200

Pre-Amplifier

Amplifier

OR

DAQ

GMX2019

Pre-Amplifier

Amplifier

γ

Strong Background Reduction

High Efficiency measurements

Coincidence measurementes suitable study γ cascade

Single spectrum from each detector

GeGe sensitivity: 0,5mBq/kg on $^{232}$Th

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In the last months we have been developing a new detector in order to perform $\beta - \gamma$ coincidence measurements. Liquid sample activated is mixed with liquid scintillator.

$$\frac{A}{Z}X + n \rightarrow \frac{A+1}{Z+1}Y + \gamma$$

GeSparK: $\beta$-y detector

Liquid sample activated is mixed with liquid scintillator. Developing measurements on activated samples in order to achieve sensitivity below ppt level.

HPGe Detector
- HPGe P-Type
- Relative Efficiency: 30%
- Cryostat: L configuration
- Carbon Window Input
- Low Background configuration
Ge-SparK: electronic chain

PMT → Trigger → Waveform → ADC Board: 2Gs/s; 2 Channel; 64MB/ch

Channel 1 → Pre-Amp → Amplifier → ADC/MCA → PC

Channel 2 → Spectroscopy

Liquid Scintillator

HPGe

10cm Copper

20cm lead

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Samples

Acrylic and Linear Alkyl Benzene (LAB) are widely used materials in many experiments in the physics of rare events.

Solid sample: Acrylic

Liquid sample: LAB scintillator

Acrylic Vessel

- Optical properties
- Mechanical properties

Liquid Scintillator based on LAB

- Chemical compatibility with acrylic
- Good purity
Samples preparation

Acrylic samples preparation

To avoid the risk of contaminating the acrylic samples, we used the laser cutting technique.

Acrylic samples washed several times in US bath (30°C) with MilliQ water in clean room 1000 atmosphere.

LAB samples preparation

LAB samples were prepared in clean room 1000 atmosphere.

Irradiation containers cleaned with ultra pure nitric acid solution (1%) in clean room 1000 atmosphere.

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NAA irradiation campaigns

Sample and a reference Standard are prepared and irradiated

STD liquid and samples were in different channels to avoid contamination

TRIGA Mark II

NAA is a comparative method

STD containing a known amount of the element to be measured

The activities of samples and standards are compared

Reactor reactor
(250 kW) - Pavia, Italy

LAZY SUSAN facility is a rotary specimen rack in a circular well within the radial reflector:

Flux of neutrons: $\approx 10^{12} cm^{-2}s^{-1}$

Irradiation Time: 6 hours

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<table>
<thead>
<tr>
<th>Sample</th>
<th>Acrylic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irradiation</td>
<td>Feb2019</td>
</tr>
<tr>
<td>Channel</td>
<td>Lazy Susan</td>
</tr>
<tr>
<td>T irradiation</td>
<td>6 hours</td>
</tr>
<tr>
<td>Detector</td>
<td>Ge-Ge</td>
</tr>
<tr>
<td>T measurement</td>
<td>27d</td>
</tr>
<tr>
<td>Sample mass</td>
<td>8.2g</td>
</tr>
<tr>
<td>Waiting Time</td>
<td>3h</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Element</th>
<th>Limit @ 90% C.L.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{40}\text{K}$ [ppt]</td>
<td>&lt;0.016</td>
</tr>
<tr>
<td>$^{238}\text{U}$ [ppt]</td>
<td>&lt;0.3</td>
</tr>
<tr>
<td>$^{232}\text{Th}$ [ppt]</td>
<td>&lt;0.5</td>
</tr>
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</table>
A slight $^{40}\text{K}$ contamination is present in LAB

**Presence of interferences** are a limit for the sensitivity on $^{238}\text{U}$

For $^{232}\text{Th}$ sensitivity is limited by cosmic muons

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Conclusions

A methodology based on neutron activation analysis (NNA) combined with high sensitivity measurements has been developed.

Methodology is suitable to determine K, Th and U content in material that usually make up central detector in experiments of physics rare events.

By this methodology:

\[ 10^{-13} \frac{g}{g} \] level has been achieved for \(^{238}\text{U},^{232}\text{Th}\) and \(^{40}\text{K}\)
Backup slides
Neutron Activation Analysis
Comparative method

Sample and a reference Standard are prepared and irradiated

STD containing a known amount of the element to be measured

Time of irradiation is the same

Neutron Flux

$T_{irr-Sample} = T_{irr-STD}$

Same position in the irradiation channel

$\phi_{Sample} = \phi_{STD}$

Geometry $Sample = Geometry_{STD}$

Gamma spectroscopy - HPGe

Efficiency $Sample = Efficiency_{STD}$

Geometry and size must be similar

The activities of samples and standards are compared

Sample trace analysis

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STD solid

Al-Co wires

Integral flux

\[ \Phi = \frac{R}{N\sigma_{eff}} \]

\( \sigma_{eff} \): calculated from MCNP

Activation Rate

\[ R \sim n_{dec} = \frac{C_{meas}}{C_{sim}} n_{sim} \]

Verification of the uniformity of the flux in LS Channels

<table>
<thead>
<tr>
<th>Channel</th>
<th>Mass (mg)</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>34.25</td>
</tr>
<tr>
<td>2</td>
<td>29.92</td>
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<tr>
<td>3</td>
<td>31.44</td>
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<tr>
<td>4</td>
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<td>5</td>
<td>32.30</td>
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<tr>
<td>6</td>
<td>34.25</td>
</tr>
<tr>
<td>7</td>
<td>31.43</td>
</tr>
</tbody>
</table>

Reference: "Measurement and simulation of the neutron flux distribution in the TRIGA Mark II reactor core" Corresponding author: D.Chiesa

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Efficiency: corrective factor

Sample and STD liquid have a different geometry

\[ \text{Efficiency}_{\text{Sample}} \neq \text{Efficiency}_{\text{STD}} \]

Corrective factor via Monte Carlo simulation

\[ E_{\text{Sample}/\text{STD}} = \frac{\text{Efficiency}_{\text{Sample}}}{\text{Efficiency}_{\text{STD}}} \]

\[ C_{\text{Sample}} \left( \frac{g}{g} \right) = I.A. \frac{\text{Counts}_{\text{Sample}}}{\text{Counts}_{\text{STD}}} \frac{C_{\text{STD}} M_{\text{STD}}}{M_{\text{Sample}}} \frac{e^{-\lambda T_{\text{Wait-STD}}}}{e^{-\lambda T_{\text{Wait-Sample}}}} \frac{(1 - e^{-\lambda T_{\text{Mis-STD}}})}{(1 - e^{-\lambda T_{\text{Mis-Sample}}})} E_{\text{Sample}/\text{STD}} \]

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