Low-level techniques applied in experiments looking for rare events

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Germanium spectroscopy
Radon detection
Mass spectrometry
Conclusions
Applications in Borexino
Applications in GERDA

<10^{-17} \text{g/g U/Th}
<3\times10^{-18} \text{g C/C}
<3\times10^{-18} \text{g K}

Borexino details: see the talks by G. Bellini and M. Pallavicini

Stainless Steel Sphere 13.7m
Stainless Steel Water Tank
Steel Shielding Plates 6m x 8m and 4m x 4m x 4m
"Holding Strings
Nylon Sphere 8.5m
Nylon film
Rn barrier
100 ton fiducial volume
Muon veto: 200 outwards pointing PMTs

Borexino Design

<10^{-17} \text{g/g U/Th}
<3\times10^{-18} \text{g C/C}
<3\times10^{-18} \text{g K}
1. Radon detection

Radon ($^{222}\text{Rn}$) and its daughters form one of the most dangerous sources of background in many experiments

- inert noble gas
- high diffusion and permeability
- belongs to the $^{238}\text{U}$ chain (present in any material)
- wide range of energy of emitted radiation (with the daughters)
- surface contaminations with radon daughters (heavy metals)
- broken equilibrium in the chain at $^{210}\text{Pb}$ level

\[
\begin{align*}
^{226}\text{Ra} & \xrightarrow{\alpha} ^{222}\text{Rn} & \xrightarrow{\alpha} ^{214}\text{Bi} & \xrightarrow{\beta} ^{206}\text{Pb} \\
^{218}\text{Po} & \xrightarrow{\alpha} ^{214}\text{Pb} & \xrightarrow{\alpha} ^{214}\text{Po} & \xrightarrow{\alpha} ^{210}\text{Po} \\
T_{1/2} & = 1622 \text{ y} & T_{1/2} & = 26.8 \text{ m} & T_{1/2} & = \text{Stable} \\
E & = 4.8 \text{ MeV} & E & = 0.7 \text{ MeV} & E & = 81 \% \\
\text{Er} & = 94 \% & \text{Er} & = 48 \% \\
T_{1/2} & = 3.8 \text{ d} & T_{1/2} & = 19.8 \text{ m} & T_{1/2} & = 5.0 \text{ d} \\
E & = 5.5 \text{ MeV} & E & = 1.5 \text{ MeV} & E & = 1.2 \text{ MeV} \\
\text{Er} & = \text{Er} & = 40 \% & \text{Er} & = 81 \% \\
T_{1/2} & = 5.1 \text{ m} & T_{1/2} & = 164 \text{ ps} & T_{1/2} & = 136.4 \text{ d} \\
E & = 6.0 \text{ MeV} & E & = 7.7 \text{ MeV} & E & = 5.3 \text{ MeV}
\end{align*}
\]
1. Radon detection

Proportional counters

- Developed for the GALLEX/GNO experiment
- Hand-made at MPIK (~ 1 cm³ active volume)
- In case of $^{222}$Rn only $\alpha$-decays are detected
- 50 keV threshold
  - bgc: 0.1 – 2 cpd
  - total detection efficiency of ~ 1.5 (0.5/$\alpha$)
- Absolute detection limit ~ 30 µBq (15 atoms)
1. Radon detection

\textbf{\( ^{222}\text{Rn in gases (N}_2/\text{Ar)} \)}

- \( ^{222}\text{Rn} \) adsorption on activated carbon
- several AC traps available (MoREx/MoRExino)
- pre-concentration from 100 – 200 m\(^3\)
- purification is possible (LTA)

\textbf{\( ^{222}\text{Rn detection limit:} \)}

\( \sim 0.5 \mu\text{Bq/m}^3 \) (STP)

[1 atom in 4 m\(^3\)]

\textbf{\( ^{222}\text{Rn emanation} \)}

- Emanation chambers
  - 20 l \( \rightarrow \) 50 \( \mu\text{Bq} \)
  - 80 l \( \rightarrow \) 80 \( \mu\text{Bq} \)
- Glass vials
  - 1 l \( \rightarrow \) \( \sim 50 \mu\text{Bq} \)

\textbf{Absolute sensitivity}

\( \sim 100 \mu\text{Bq} \) [50 atoms]
1. Radon detection

222\textsuperscript{Rn} / 226\textsuperscript{Ra in water}

- 222\textsuperscript{Rn} extraction from 350 liters
- 222\textsuperscript{Rn} and 226\textsuperscript{Ra} measurements possible

222\textsuperscript{Rn} detection limit: ~0.1 mBq/m\textsuperscript{3}
226\textsuperscript{Ra} detection limit: ~0.8 mBq/m\textsuperscript{3}

222\textsuperscript{Rn} diffusion in thin films

- Time dependent diffusion profile registered
- D reconstructed on the base of the mathematical model

Sensitivity ~ 10\textsuperscript{-13} cm\textsuperscript{2}/s
2. Mass spectrometry

Noble gas mass spectrometer

VG 3600 magnetic sector field spectrometer.

Used to investigate noble gases in the terrestrial and extra-terrestrial samples.

Adopted to test the nitrogen purity and purification methods.

Detection limits: Ar: $10^{-9}$ cm$^3$
Kr: $10^{-13}$ cm$^3$

$^{39}$Ar: $\sim 1.4 \times 10^{-9}$ Bq/m$^3$ N$_2$ (STP)
$^{85}$Kr: $\sim 1 \times 10^{-7}$ Bq/m$^3$ N$_2$ (STP)
3. Germanium spectroscopy

GeMPIs at GS (3800 m w.e.)

- GeMPI I operational since 1997 (MPIK)
- GeMPI II built in 2004 (MCavern)
- GeMPI III constructed in 2007 (MPIK/LNGS)
- Worlds most sensitive spectrometers

GeMPI I:
- Crystall: 2.2 kg, $\varepsilon_r = 102\%$
- Bcg. Index (0.1-2.7 MeV): 6840 cts/kg/year
- Sample chamber: 15 l

Sensitivity:
~10 $\mu$Bq/kg

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3. Germanium spectroscopy

Detectors at MPIK: Dario, Bruno and Corrado

Sensitivity: ~1 mBq/kg

MPIK LLL: 15 m w.e.
Radon detection
Mass spectrometry
Germanium spectroscopy
Applications in Borexino
Applications in GERDA
Conclusions

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**Germanium spectroscopy**

**Radon detection**

**Mass spectrometry**

**Applications in Borexino**

**Applications in GERDA**

**Conclusions**

- **U/Th:** 1 ppt (12 µBq/kg $^{226}$Ra)
- **1 cpd in 100 t**
- $^{39}$Ar, $^{85}$Kr, $^{222}$Rn
- **7 µBq/m$^3$** $^{222}$Rn
- 0.4 ppm Ar
- 0.2 ppt Kr

$^{10^{-17}}$ g/g U/Th

$3 \times 10^{-18}$ $^{14}$C/$^{12}$C

$<3 \times 10^{-18}$ g/g $^{40}$K

$<10^{-17}$ g/g U/Th

$3 \times 10^{-18}$ $^{14}$C/$^{12}$C

$<3 \times 10^{-18}$ g/g $^{40}$K

$<10^{-17}$ g/g U/Th

$3 \times 10^{-18}$ $^{14}$C/$^{12}$C

$<3 \times 10^{-18}$ g/g $^{40}$K

$<10^{-17}$ g/g U/Th

$3 \times 10^{-18}$ $^{14}$C/$^{12}$C

$<3 \times 10^{-18}$ g/g $^{40}$K
5. Applications in Borexino - $^{226}$Ra in the nylon foil

**Borexino nylon foil**

1 ppt U required
($\sim 12 \, \mu\text{Bq/kg for } ^{226}\text{Ra}$)

$D_{\text{dry}} = 2 \times 10^{-12} \, \text{cm}^2/\text{s} \quad (d_{\text{dry}} = 7 \, \mu\text{m})$

$D_{\text{wet}} = 1 \times 10^{-9} \, \text{cm}^2/\text{s} \quad (d_{\text{wet}} = 270 \, \mu\text{m})$

$A_{\text{dry}} = A_{\text{sf}} + 0.14 \cdot A_{\text{bulk}}$

$A_{\text{wet}} = A_{\text{sf}} + A_{\text{bulk}}$

Separation of the bulk and surface $^{226}\text{Ra}$ conc. was possible through $^{222}\text{Rn}$ emanation

Very sensitive technique:
($C_{\text{Ra}} \sim 10 \, \mu\text{Bq/kg}$)

**Bx IV foil:**

bulk $\leq 15 \, \mu\text{Bq/kg}$

surface $\leq 0.8 \, \mu\text{Bq/m}^2$

total $= (16 \pm 4) \, \mu\text{Bq/kg} \quad (1.3 \, \text{ppt U equiv.})$
5. Applications in Borexino – nitrogen

**Regular Purity Nitrogen:**
- Technical 4.0 quality, not purified
- Production rate up to 100 m³/h (STP)
- $^{222}\text{Rn}$ ~ 50 µBq/m³, Ar ~ 10ppm, Kr ~ 30 ppt

**High Purity Nitrogen:**
- $^{222}\text{Rn}$ adsorption on charcoal (LTA)
- Achieved concentration < 0.3 µBq/m³
- Production rate up to 100 m³/h (STP)
- Ar and Kr not removed

**LAK (Low Ar and Kr) Nitrogen:**
- Spec. Ar < 0.4 ppm, Kr < 0.2 ppt
- $^{222}\text{Rn}$ < 7 µBq/m³
- Purification by adsorption on different materials extensively studied (successfully!)
- Cooperation with companies on the nitrogen survey
- Tests of the nitrogen delivery chain

### Nitrogen survey

<table>
<thead>
<tr>
<th>Nitrogen sample</th>
<th>$C_{\text{Ar}}$ [ppm]</th>
<th>$C_{\text{Kr}}$ [ppt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>MESSER (4.0)</td>
<td>200 ± 30</td>
<td>1680 ± 240</td>
</tr>
<tr>
<td>Air Liquide (4.0)</td>
<td>11.0 ± 1.3</td>
<td>40 ± 5</td>
</tr>
<tr>
<td>Linde AG, (7.0)</td>
<td>0.031 ± 0.004</td>
<td>2.9 ± 0.4</td>
</tr>
<tr>
<td>SOL (6.0)</td>
<td>0.0063 ± 0.0006</td>
<td>0.04 ± 0.01</td>
</tr>
<tr>
<td>Westfalen AG (6.0)</td>
<td>0.00050 ± 0.00008</td>
<td>0.06 ± 0.02</td>
</tr>
<tr>
<td>Goal (BOREXINO)</td>
<td>&lt; 0.4</td>
<td>&lt; 0.2</td>
</tr>
</tbody>
</table>

### Tests of the delivery chains

<table>
<thead>
<tr>
<th>Supplier/setup</th>
<th>$C_{\text{Ra}}$ [µBq/m³]</th>
<th>$C_{\text{Ar}}$ [ppm]</th>
<th>$C_{\text{Kr}}$ [ppt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linde AG, 3-m³ movable tank</td>
<td>1.2</td>
<td>0.018</td>
<td>0.06</td>
</tr>
<tr>
<td>SOL, 16-m³ tank</td>
<td>8</td>
<td>0.012</td>
<td>0.02</td>
</tr>
</tbody>
</table>

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5. Applications in Borexino – nitrogen

LAK Nitrogen tank installed at Gran Sasso
6. Applications in GERDA

GERDA details: see the talk by H. Simgen

Additional inner copper shield

detectors holders

U/Th \leq 16 \mu\text{Bq/kg}

clean surface

Liquid argon

^{222}\text{Rn} \leq 1 \mu\text{Bq/m}^3

Vacuum-insulated double

wall stainless steel cryostat

U/Th \leq 5 – 10 \text{mBq/kg}

clean surface

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6. Applications in GERDA – argon purity

$^{222}$Rn in argon (GERDA goal: $C_{Rn} \leq 1 \mu Bq/m^3$)

<table>
<thead>
<tr>
<th>Quality</th>
<th>Company</th>
<th>Sample size [m$^3$]</th>
<th>$C_m$ [mBq/m$^3$]</th>
<th>$C_r$ [mBq/m$^3$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.6</td>
<td>Westfalen</td>
<td>117</td>
<td>2.9 ± 0.2</td>
<td>&gt;8</td>
</tr>
<tr>
<td>4.8</td>
<td>Air Liquide</td>
<td>80</td>
<td>0.27 ± 0.02</td>
<td>0.4</td>
</tr>
<tr>
<td>5.0</td>
<td>Westfalen</td>
<td>200</td>
<td>6.0 ± 0.1</td>
<td>8.4</td>
</tr>
<tr>
<td>5.0</td>
<td>Air Liquide (GS)</td>
<td>5</td>
<td>0.25 ± 0.04</td>
<td>0.3</td>
</tr>
<tr>
<td>6.0</td>
<td>Westfalen</td>
<td>104</td>
<td>0.11 ± 0.01</td>
<td>0.4</td>
</tr>
</tbody>
</table>

$^{222}$Rn in nitrogen

<table>
<thead>
<tr>
<th>Quality</th>
<th>Company</th>
<th>Sample size [m$^3$]</th>
<th>$C_m$ [$\mu Bq/m^3$]</th>
<th>$C_r$ [$\mu Bq/m^3$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.5 – 5.0</td>
<td>Messer/Air Liquide</td>
<td>40</td>
<td>30 – 70</td>
<td>---</td>
</tr>
<tr>
<td>6.0 (7.0)</td>
<td>Linde</td>
<td>150</td>
<td>0.7 ± 0.2</td>
<td>1</td>
</tr>
<tr>
<td>6.0</td>
<td>SOL</td>
<td>100</td>
<td>15 ± 1</td>
<td>17</td>
</tr>
</tbody>
</table>

Argon purification required. In the gas phase requested purity achieved ($C_{Rn} \leq 0.5 \mu Bq/m^3$), adsorption for liquid phase under investigations.
6. Applications in GERDA – steel screening

GERDA goal for U/Th: $\leq 5 - 10 \text{ mBq/kg}$

<table>
<thead>
<tr>
<th>No.</th>
<th>Specific activity [mBq/kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{228}\text{Th}$</td>
</tr>
<tr>
<td>1 D</td>
<td>5.1 ± 1.0</td>
</tr>
<tr>
<td>2 G</td>
<td>&lt; 0.27</td>
</tr>
<tr>
<td>3 D</td>
<td>1.1 ± 0.4</td>
</tr>
<tr>
<td>4 D</td>
<td>&lt; 2.6</td>
</tr>
<tr>
<td>5 D</td>
<td>&lt; 1.1</td>
</tr>
<tr>
<td>6 D</td>
<td>&lt; 0.8</td>
</tr>
<tr>
<td>7 G</td>
<td>&lt; 0.20</td>
</tr>
<tr>
<td>8 G</td>
<td>&lt; 0.11</td>
</tr>
<tr>
<td>9 G</td>
<td>&lt; 0.41</td>
</tr>
<tr>
<td>10 G</td>
<td>&lt; 1.0</td>
</tr>
<tr>
<td>11 G</td>
<td>1.5 ± 0.2</td>
</tr>
</tbody>
</table>

(SS 1.4571)
**6. Applications in GERDA – copper cleaning**

### 222Rn daughters on copper surface

- Screening of \(^{210}\)Po with an alpha spectrometer
  50 mm Si-detector, bcg ~ 5 \( \alpha / d \) (1-10 MeV) sensitivity ~ 20 mBq/m² (100 mBq/kg, \(^{210}\)Po)
- Screening of \(^{210}\)Bi with a beta spectrometer
  2×50 mm Si(Li)-detectors, bcg ~ 0.18/0.40 cpm sensitivity ~ 10 Bq/kg
- Screening of \(^{210}\)Pb (46.6 keV line) with a gamma spectrometer
  25 % - n-type HPGe detector with an active and a passive shield sensitivity ~ 20 Bq/kg
- Only small samples can be handled – artificial contamination needed: e.g. discs loaded with 222Rn daughters

**Copper cleaning tests**

- Etching removes most of \(^{210}\)Pb and \(^{210}\)Bi (> 98 %) but **not** \(^{210}\)Po
- Electropolishing is more effective for all elements but proper conditions have to be found (e.g. \(^{210}\)Po reduction from 30 up to 200)

**Etching:** 1% \( \text{H}_2\text{SO}_4 \) + 3% \( \text{H}_2\text{O}_2 \) **Electropolishing:** 85% \( \text{H}_3\text{PO}_4 \) + 5% 1-butanol

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7. Conclusions

- Low-level techniques have “natural” applications in experiments looking for rare events (low-energy neutrinos, neutrino-less double beta decay, search for dark matter/proton decay…)

- Several detectors and experimental methods were developed allowing measurements even at a single atom level

- Described experimental methods were very successfully applied in the Borexino experiment (very low background achieved) and can be adopted in other projects
  - material screening
  - purification and cleaning techniques
  - study of noble gases
6. Applications in GERDA – argon purification

$^{222}$Rn removal from gaseous/liquid argon
150 g- (gas phase) and 60 g-AC traps (liquid phase) used

<table>
<thead>
<tr>
<th>Quality</th>
<th>Sample size [m$^3$]</th>
<th>$C_1$ [mBq/m$^3$]</th>
<th>$C_2$ [mBq/m$^3$]</th>
<th>Red. factor [1/kg]</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.6</td>
<td>141</td>
<td>0.20 ± 0.02</td>
<td>&lt; 0.0005</td>
<td>&gt; 2700</td>
<td>Gas phase</td>
</tr>
<tr>
<td>4.8</td>
<td>80</td>
<td>0.27 ± 0.02</td>
<td>0.0007 ± 0.0003</td>
<td>2500</td>
<td>Gas phase</td>
</tr>
<tr>
<td>4.6</td>
<td>67</td>
<td>0.050 ± 0.003</td>
<td>0.0020 ± 0.0005</td>
<td>420</td>
<td>Liquid phase</td>
</tr>
<tr>
<td>4.6</td>
<td>77</td>
<td>0.056 ± 0.004</td>
<td>0.0027 ± 0.0006</td>
<td>370</td>
<td>Liquid phase</td>
</tr>
<tr>
<td>4.8</td>
<td>140</td>
<td>0.20 ± 0.01</td>
<td>0.005 ± 0.001</td>
<td>640</td>
<td>Liquid phase</td>
</tr>
<tr>
<td>4.8</td>
<td>48</td>
<td>0.14 ± 0.01</td>
<td>0.003 ± 0.001</td>
<td>700</td>
<td>Liquid phase</td>
</tr>
<tr>
<td>5.0</td>
<td>200</td>
<td>6.0 ± 0.1</td>
<td>0.60± 0.02</td>
<td>170</td>
<td>Liquid phase</td>
</tr>
<tr>
<td>6.0</td>
<td>104</td>
<td>0.11 ± 0.01</td>
<td>0.006± 0.001</td>
<td>305</td>
<td>Liquid phase</td>
</tr>
</tbody>
</table>

Required reduction factor for GERDA: O(500)