Measuring radioactive contamination using ICP-MS

Shintaro Ito¹, Yuichi Takaku², and The Super-Kamiokande Collaboration

¹Okayama University, Japan
²Institute for Environmental Sciences, Japan

Low Radioactivity Technique 2017
Outline

- Super-Kamiokande
- SK-Gd
- ICP-MS
- Problem
- Technique of measuring low radioactive contamination
- Conclusion
Super-Kamiokande (SK)

- 50,000t ultra pure water Cherenkov detector.
- Located 1000m underground in the mine.
- >11,000 PMTs inside the SK tank.
- Measuring Cherenkov ring of charged particle produced by the reaction of neutrino and water.

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Purpose of Super-Kamiokande

- Atmospheric neutrino
- Solar neutrino
- Supernova neutrino
- Supernova relic neutrino
- Proton decay, and so on

Atmospheric

Sun

Supernova
Supernova Relic Neutrinos (SRN)

- Neutrinos from past supernova explosion.
  ➔ Should still be in universe.

- There are several models of SRN energy spectrum.
  ➔ However, it has not been observed yet.
  ➔ Reveal the history of early universe, star-formation, and so on.
New Project “SK-Gd”

- **Super Kamiokande Gadolinium (SK-Gd) Project.**
- Gd has the largest cross section with neutron.
  - Capture neutron and emit 3~4 gamma-rays in total 8 MeV.
  - Neutron emitted via inverse beta decay ($\bar{\nu}_e + p \rightarrow n + e^+$)
  - Neutron tagging by Gd and detect $\bar{\nu}_e$ SRN
- Loading 0.2% of $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$ into SK
  - 90% of neutron capture efficiency
Problems

- As shown on p4, SK is multi-purpose detector.
- To detect solar neutrino with low energy threshold \( E = 3.5 \) MeV, ultra pure water is being used.

\[ \text{Gd}_2(\text{SO}_4)_3 \text{ should also be purified to continue solar neutrino.} \]

- Main radioactivities.

<table>
<thead>
<tr>
<th></th>
<th>Commercial Gd(_2)(SO(_4))(_3) ((\times 10^{-9}\text{ g/g}))</th>
<th>Requirement ((\times 10^{-9}\text{ g/g}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>U</td>
<td>4</td>
<td>0.4</td>
</tr>
<tr>
<td>Th</td>
<td>25</td>
<td>0.025</td>
</tr>
</tbody>
</table>

- Production of purified Gd\(_2\)(SO\(_4\))\(_3\) is in progress.
- Need to measure U/Th in Gd\(_2\)(SO\(_4\))\(_3\).
  - U/Th have long life time → less sensitivity for Ge measurement.
  - ICP-MS measurement.
Kamioka ICP-MS “Agilent 7900”

Auto-sampler is covered by clean booth. → Class 100
Kamioka ICP-MS “Agilent 7900”

- High sensitivity: U/Th ~20ppq
- Internal standard solution: estimate matrix effect.
- He gas in collision reaction cell: to reduce poly-atoms ions.

Problems

- Problems of measuring high concentration sample are
  - Significant **matrix effect** by Gd.
    - Sensitivity dropped to 30~40% by 0.2% of Gd$_2$(SO$_4$)$_3$.
  - **Spectrum interference** by remaining poly-atoms ions (e.g. GdO$_4$).
  - **Chemical extraction U/Th from Gd$_2$(SO$_4$)$_3$**

![Matrix effects.](image1)

![Typical example of spectrum interference.](image2)
U/Th Extraction from Gd$_2$(SO$_4$)$_3$

- High separation efficiency between U/Th and Gd$_2$(SO$_4$)$_3$.
- HNO$_3$ should be better for ICP-MS measurement.

**UTEVA resin** (Eichrom). Adsorbs Actinides (except Am).
How to Measure Low Radioactive Contamination

1. Load the resin into column.
2. Prewash of resin and column by HNO3.
3. Conditioning of the resin by high concentration HNO3.
4. Load Gd$_2$(SO$_4$)$_3$ + HNO$_3$ sample into column.
5. Elution by diluted HNO$_3$.
6. Measuring eluted solution by ICP-MS.
How to Measure Low Radioactive Contamination

- Test
  - Added 1~100ppt of U/Th into 0.2% of Gd$_2$(SO$_4$)$_3$ solution and estimated recovery.
  - >90% of recovery!!
  - No U/Th concentration dependence.
  - **No matrix effect.**
  - Gd could be reduced to ~1/30.
    - ➤ **No interference from poly-atoms ions.**

- Measurement
  - There are 3 purified Gd$_2$(SO$_4$)$_3$ samples in Kamioka.
  - Measured 0.2% solution with the same procedure.
## Results

<table>
<thead>
<tr>
<th>Sample</th>
<th>U ($\times 10^{-9}$ g/g)</th>
<th>Th ($\times 10^{-9}$ g/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Comercial</td>
<td>4</td>
<td>25</td>
</tr>
<tr>
<td>A</td>
<td>&lt;0.02</td>
<td>0.05</td>
</tr>
<tr>
<td>B</td>
<td>&lt;0.02</td>
<td>0.10</td>
</tr>
<tr>
<td>C</td>
<td>&lt;0.01</td>
<td>0.05</td>
</tr>
<tr>
<td>Goal</td>
<td>0.4</td>
<td>0.025</td>
</tr>
</tbody>
</table>

Preliminary

- U and Th were reduced to $<1/200$ and $1/500$ respectively.
  - Thanks to the efforts of companies!!
  - U was much purified!!
  - Th should be removed to a factor of 2 or 3!!
Conclusion

- New SK-Gd project at Kamioka in Japan.
  - Loading 0.2% of Gd₂(SO₄)₃ into SK.
  - Aims to detecto SRN.
- Necessary to purify Gd₂(SO₄)₃ to continue solar neutrino analysis.
  - Purification is in progress.
  - Measuring U/Th in purified Gd₂(SO₄)₃ using ICP-MS.
  - Chemical separation to reduce matrix effect and poly-atoms ion.
    ➡️ >90% of recovery.
- U was much purified while Th should be reduced 2 or 3 times more.
Thank you for your attention!!
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