“Co-doped CeBr₃ Demonstration”

Project funded under the Innovation Triangle Initiative

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Outlines

1) Background and justification (B&J)

2) Objective(s)

3) Achievements and status

4) Benefits

5) Next steps
Background and justification (B&J): CeBr$_3$ and space

Based on previous ESA project *Low-noise scintillator detectors for planetary remote-sensing:*

A CeBr$_3$ crystal is integrated in BepiColombo as the gamma-ray sensor of the Mercury Gamma-ray and Neutron Spectrometer (MGNS).

IKI Department №63 Nuclear Planetology: “We study space objects using the methods of nuclear physics.”
B&J: Gamma-ray spectroscopy

Gamma-ray spectroscopy is a nuclear experimental technique / analytical method to identify and quantify gamma-ray emitters elements and hence elementary composition.

One of the earlier development is from the 1940’s for application in oil prospecting by Bruno Pontecorvo (one of the youngest colleagues of Enrico Fermi when in Rome) when working at Well Surveys Inc., Tusla.

Routinely employed in nuclear physics and radio-chemistry, in nuclear medicine, for geophysical surveys, in radiation protection, border safeguards and nuclear / radiation inspections.

\[ ^{137}\text{Cs} \rightarrow ^{137}\text{Ba} \]

\[ ^{40}\text{K} \rightarrow ^{40}\text{Ar} \]

\[ ^{208}\text{TI} \rightarrow ^{208}\text{Pb} \]
B&J: gamma-ray spectroscopy in space

Gamma-ray signatures of K and Th allow characterize ratio between volatile and refractory elements, fundamental as well in geology: for oil and mineral resources research and exploitation.

B&J: gamma-ray spectrometers

Two main category of gamma-ray spectrometers

**Semiconductors** e.g. HPGe, CdZnTe...
- Mars Odyssey GRS,
- SELENE Kaguya GRS

**Scintillators** e.g. CeBr₃, NaI(Tl)
- Lunar prospector
- NEAR (in collaboration with oil industry Schlumberger)
- BepiColombo

Modern high resolution scintillators offer key advantages when constraints on mass and power are strict as in the case of BepiColombo and high detection efficiency is needed.

**Messenger GRNS**
(NASA/JHU/APL)
- 9.2 Kg and 16.5 W
- plus Neutron Spectrometer
- 3.9 Kg and 6.0 watts

**MGNS IKI/ESA**
- 5.5 Kg and 6.5 W
Ongoing improvements

**Semiconductors** GeMini, highly mignaturized HPGe (?? cm³) only <3 kg and <15 W (without ACS)

**Scintillators**: further improve energy resolution, however for large crystals 2-3% is close to the fundamental limit (1-1.5% statistics).
B&J: how a scintillator works

A scintillator converts gamma-ray energy in up to several tens of thousands of low energy photons which can then be recorded using a photodetector.
**B&J: non proportionality at low gamma-ray energy**

For low gamma-ray energies (x-rays) and mainly in conjunction with the characteristics fluorescence lines (electron binding energy) the response of scintillator is non proportional. Since the gamma to scintillation photons conversion implies a stochastic energy transfer to a large number of lower energy electron, this “quenching” ultimately worsen the energy resolution of scintillators at gamma-ray energy. LaBr$_3$ (3% en. res.) vs CeBr$_3$ (4% en. res.).
One of the earlier observation about co-doping and energy resolution is by M. Moszynski (2005-2006): “[...] selective co-doping of scintillators may improve the non-proportionality and their energy resolution. No doubt that further studies are necessary.”

Also based on the above early experiments in collaboration with TU Delft and Uni. Bern with few mm$^3$ and 1 cm$^3$ showed effectiveness of Ca and Sr co-dopant.

Other tested co-dopants include: Zr, Hf, Zn, Cd and Pb (from Harrison, Doty et al. IEEE TNS 56 (2009) 1661).

Scintillators and codoping is currently a rather trendy subject.
Objectives: apply co-doping to $10 \text{ cm}^3 \text{ CeBr}_3$ to enhance its energy resolution

More specifically:
To grow and develop an innovative low-noise scintillator by demonstrating that co-doping technology enhances the energy resolution of $\text{CeBr}_3$ scintillator with volume of about $10 \text{ cm}^3$ (1 in. x 1 in.) while maintaining unaltered the other performances.

We started about 1 year ago...
Three different co-doping compositions for a total of six samples grown fabricated and characterized plus two more samples of the most performing composition. All composition showed improved nPR and three samples showed clearly improved energy resolution: 3.5% and 3.7% (two samples).
## Achievements

4 crystal compositions grown, 8 scintillator samples fabricated and characterized

<table>
<thead>
<tr>
<th>Sample</th>
<th>Dimension</th>
<th>LY</th>
<th>nPR</th>
<th>Energy resolution</th>
<th>Decay</th>
<th>Emission</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>ph/keV Fract. of STD % at 662 keV ns nm</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ref.CeBr3</td>
<td>typical values</td>
<td>40-45</td>
<td>1.0</td>
<td>3.9-4.3</td>
<td>24</td>
<td>390</td>
</tr>
<tr>
<td>Ref Sr-codoped</td>
<td>12x10x4 (0.5 cm³)</td>
<td>39.0</td>
<td>0.4</td>
<td>3.4</td>
<td>30</td>
<td>390</td>
</tr>
<tr>
<td></td>
<td>15x25 (4.5 cm³)</td>
<td>39.0</td>
<td>0.6</td>
<td>4.1 / 4.0</td>
<td>31</td>
<td>395</td>
</tr>
<tr>
<td></td>
<td>15x25 (4.5 cm³)</td>
<td>39.0</td>
<td>0.5</td>
<td>3.9 / 4.1</td>
<td>33</td>
<td>397</td>
</tr>
<tr>
<td>B1</td>
<td>~23x25 (~10.6 cm³)</td>
<td>42.0</td>
<td>0.6</td>
<td>3.6 / 3.5</td>
<td>33</td>
<td>402</td>
</tr>
<tr>
<td>B2</td>
<td>25x25 (12.9 cm³)</td>
<td>39.0</td>
<td>0.3</td>
<td>4.1 / 4.2</td>
<td>34</td>
<td>403</td>
</tr>
<tr>
<td></td>
<td>25x25 (12.9 cm³)</td>
<td>42.0</td>
<td>0.6</td>
<td>3.9 / 3.9</td>
<td>31</td>
<td>398</td>
</tr>
<tr>
<td>C1</td>
<td>25x25 (12.9 cm³)</td>
<td>42.0</td>
<td>0.4</td>
<td>4.2 / 4.2</td>
<td>34</td>
<td>402</td>
</tr>
<tr>
<td>C2</td>
<td>25x25 (12.9 cm³)</td>
<td>40.0</td>
<td>0.6</td>
<td>3.8 / 3.7</td>
<td>31</td>
<td>396</td>
</tr>
<tr>
<td>B1 rep.</td>
<td>25x25 (12.9 cm³)</td>
<td>44.0</td>
<td>0.5</td>
<td>3.9 / 3.7</td>
<td>31</td>
<td>396</td>
</tr>
<tr>
<td>B2 rep.</td>
<td>~25x21 (~10.6 cm³)</td>
<td>44.0</td>
<td>0.5</td>
<td>3.9 / 3.7</td>
<td>31</td>
<td>398</td>
</tr>
</tbody>
</table>
Achievements

All co-doped samples show a substantially improved nPR compared to standard CeBr$_3$. The stronger nPR improvement is that of sample B2, however B2 sample show a gradient of performance along the Z-axis which is thought to be responsible of its lack of energy resolution enhancement.

Measurements with alpha particles (high ionization density) reveal that sample B2 with a nPR better than that of LaBr3, still presents an alpha / gamma ratio of only 90% of that of LaBr3.
Summary of achievements

- Three cooped CeBr3 composition grown developed into spectrometers and characterized
- One composition has been established which enhances CeBr3 energy resolution
- No detrimental effects found within other CeBr3 scintillation properties up to 10cm³, neither envisaged to even larger sizes: e.g. decay time of ~30 ns applies to the smaller and larger sizes of codoped CeBr3
- The ultimate energy resolution improvement cannot be determined and 3% remain possibly achievable goal.

![Graph showing FWHM at 662 keV](image-url)
Benefits

Scintillation detectors are extensively used almost in every context where gamma-ray emissions need to be detected and/or monitored. CeBr$_3$ is already a successful scintillator. Co-doped CeBr$_3$ is a further improvement in energy resolution and sensitivity and will enables more specific energy resolution-driven applications compared to standard CeBr$_3$ all without drawbacks.

Mission(s) to Venus under Nasa’s Discovery program
A.M. Parsons et al. IEEE NSS-MIC 2016 (N46-2)
M.L. Litvak et al. NIMA 822 (2016) 112

Indian Space Research Organisation
D.K. Panda et al. GRS development for planetary missions

Korea Pathfinder Lunar Orbiter
KGRS (KPLO Gamma Ray spectrometer)
Next steps

Two more crystal growth runs, fabrications and characterizations to be performed.

Based on the final outcomes of the activity — final iteration of cost/benefit analysis for feasibility of industrialization taking into account production aspects and larger dimensions of co-doped CeBr$_3$ for commercial applications including space.

Co-doping CeBr$_3$ preserve compatibility with standard CeBr$_3$ allowing integration with novel solid state photosensors and related electronics including in the next generation of gamma-ray spectrometer instruments for space missions.

Follow-on activity not yet funded (envisaged originally as ESA’s ITI Technology Adoption).
Thank you!