CdWO$_4$ crystal scintillators from enriched isotopes for double beta decay experiments

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Double beta ($2\beta$) decay

- Nuclear transformations when the charge of nuclei changes by two units: $(A, Z) \rightarrow (A, Z \pm 2)$

- $2\beta$ decay was registered for only 12 nuclides (from 69 $2\beta$-candidates)

- The rarest nuclear decay ever observed (half-lives $T_{1/2} \sim 10^{18} \text{–} 10^{24}$ yr)

- Could help to clarify the fundamental problems in particle physics:
  - Lepton number violation
  - Nature of neutrino (Dirac or Majorana particle)
  - Hierarchy of neutrino mass
  - Absolute scale of neutrino mass
  - Other effects beyond the Standard Model of particles

- Scintillation counting is one of the most sensitive techniques for $2\beta$ decay search
# Enriched scintillators for $2\beta$ decay search

<table>
<thead>
<tr>
<th>Year</th>
<th>Scintillator</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1966</td>
<td>$^{48}$CaF$_2$(Eu) ($\delta$=97%, m=22 g)</td>
<td>[1]</td>
</tr>
<tr>
<td></td>
<td>$^{40}$CaF$_2$(Eu) ($\delta$=97%, m=22 g)</td>
<td></td>
</tr>
<tr>
<td>1987 – 2003</td>
<td>$^{116}$CdWO$_4$ ($\delta$=83%, m=510 g)</td>
<td>[2]</td>
</tr>
<tr>
<td>2009 – present</td>
<td>$^{106}$CdWO$_4$ ($\delta$=66%, m=231 g)</td>
<td>[3]</td>
</tr>
<tr>
<td>2010 – present</td>
<td>$^{116}$CdWO$_4$ ($\delta$=82%, m=1868 g)</td>
<td>[4]</td>
</tr>
<tr>
<td>2010 – present</td>
<td>$^{40}$Ca$^{100}$MoO$_4$ ($\delta$=96% of $^{100}$Mo, $\delta$=99.964% of $^{40}$Ca depleted on $^{48}$Ca, m=550 g)</td>
<td>[5]</td>
</tr>
<tr>
<td>2013 – 2014</td>
<td>$^{100}$MoO$_4$, $^{82}$Se</td>
<td>[6,7]</td>
</tr>
</tbody>
</table>

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Why are $^{106,116}$Cd promising for $2\beta$ search?

- **Very high $2\beta$ decay energy**
  \(Q_{2\beta} = 2770\) keV for $^{106}$Cd and 2813 keV for $^{116}$Cd)

- **Relatively large isotopic abundance**
  \(\delta = 1.25\%\) for $^{106}$Cd and 7.49\% for $^{116}$Cd)

- **Promising theoretical estimations on $2\beta$ decay of $^{106,116}$Cd**

- **CdWO$_4$ – good detector for $2\beta$ decay search** [1,2]
  - a) calorimetric “source = detector” experiment
  - b) low level of intrinsic radioactivity
  - c) good scintillation properties
  - d) pulse-shape discrimination ability
  - e) relatively inexpensive
  - f) stability for long term operation

- **Availability of raw materials enriched in $^{106,116}$Cd**

Refs.:  
Specific demands for production of enriched scintillators for $2\beta$ experiments

- Minimal loss of expensive isotopically enriched materials
- High yield of crystals
- Prevention of radioactive contamination
Steps of $^{106,116}$CdWO$_4$ productions

Raw material (W)

Production of $^{106,116}$CdWO$_4$ powder

ICP-MS
L-MS
AAS

Raw material ($^{106,116}$Cd)

Pure $^{106,116}$Cd metal

Vacuum distillation of $^{106,116}$Cd metal

Crystal growth

Treatment

$^{106,116}$CdWO$_4$
Deep purification of enriched $^{106,116}\text{Cd}$

Vacuum distillation with use of getter filters

It allows to purify the enriched $^{106,116}\text{Cd}$ isotopes on the level of 1–0.1 ppm:

<table>
<thead>
<tr>
<th>Impurities</th>
<th>Level, ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni, Cu</td>
<td>&lt; 0.2</td>
</tr>
<tr>
<td>Fe, Mg, Mn, Cr, V, Co</td>
<td>&lt; 1</td>
</tr>
<tr>
<td>Th, U, Ra, K, Rb, Bi, Pb, Lu, Sm</td>
<td>&lt; 0.1</td>
</tr>
</tbody>
</table>

Main advantages:
- high effectiveness of purification
- higher yield of end product (> 96% of original charge)
- minimum irrecoverable losses of material (< 1–2 %)

Synthesis of $^{106,116}$CdWO$_4$ powders

Choice of the methods for $^{106,116}$Cd additional purification:

- Recrystallization methods (used for Cd salts)? NO
  - Low outcome of the final product (< 85%)

- Coprecipitation of impurities in cadmium nitrate on a collector!
  + Recrystallization of ammonium para-tungstate

$$
\text{Cd(NO}_3\text{)}_2 + (\text{NH}_4\text{)}_2\text{WO}_4 \xrightarrow{t^\circ} \text{CdWO}_4 + 2\text{NH}_4\text{NO}_3
$$
Growth of $^{106,116}$CdWO$_4$ scintillators

Low-Thermal-Gradient Czochralski technique (LTG Cz)

231 g, $\varnothing 27 \times 60$ mm

1868 g, $\varnothing 45 \times 147$ mm

87% of initial charge

while for Cz only $\sim 30$

216 g, $\varnothing 27 \times 50$ mm

82%

70%

of initial charge

## Characterization of $^{106,116}$CdWO$_4$

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Attenuation length @ 480 nm</td>
<td>(60 ± 7) cm *</td>
<td>(31 ± 5) cm</td>
</tr>
<tr>
<td>FWHM (CWO on PMT) @ 662 keV of $^{137}$Cs</td>
<td>10.0%</td>
<td>10.1%</td>
</tr>
<tr>
<td></td>
<td>@ 2615 keV of $^{208}$Tl</td>
<td>8.4% **</td>
</tr>
<tr>
<td>Enrichment</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Isotopic abundance in $^{nat}$Cd)</td>
<td>66.4% of $^{106}$Cd (1.25%)</td>
<td>82.2% of $^{116}$Cd (7.49%)</td>
</tr>
</tbody>
</table>

* – Never reported for CdWO$_4$

** – FWHM that was reached in 2$\beta$ experiment

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Search for $2\beta$ processes in $^{106,116}$Cd

DAMA R&D set-up, Laboratori Nazionali del Gran Sasso (Italy)

### Radioactive contamination of $^{106,116}$CdWO$_4$ crystals

<table>
<thead>
<tr>
<th>Source</th>
<th>Activity in $^{106/116}$CdWO$_4$, mBq/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{116}$CWO [1]</td>
</tr>
<tr>
<td></td>
<td>$^{116}$CWO [1] /Scraps</td>
</tr>
<tr>
<td></td>
<td>$^{116}$CWO [2]</td>
</tr>
<tr>
<td></td>
<td>$^{106}$CWO [3]</td>
</tr>
<tr>
<td></td>
<td>scintillation mode by HPGe detector</td>
</tr>
<tr>
<td>scint.</td>
<td>scint. mode</td>
</tr>
<tr>
<td>scint.</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{232}$Th</td>
<td>$\leq 0.08$</td>
</tr>
<tr>
<td>$^{228}$Ra</td>
<td>$\leq 0.2$</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>$0.041(6) - 0.072(8)$</td>
</tr>
<tr>
<td>$^{227}$Ac</td>
<td>$\leq 0.002$</td>
</tr>
<tr>
<td>$^{238+234}$U</td>
<td>$\leq (0.4 - 0.6)$</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>$\leq 0.005$</td>
</tr>
<tr>
<td>$\Sigma \alpha$</td>
<td>$2.1(2) - 2.9(3)$</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>$\leq 0.9$</td>
</tr>
<tr>
<td>$^{110m}$Ag</td>
<td>$0.12(4)$</td>
</tr>
<tr>
<td>$^{113}$Cd</td>
<td>$100(10)$</td>
</tr>
<tr>
<td>$^{113m}$Cd</td>
<td>$460(20)$</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>$\leq 0.3$</td>
</tr>
<tr>
<td></td>
<td>$\leq 31 / \leq 38$</td>
</tr>
<tr>
<td></td>
<td>$\leq 3.1 / 64(4)$</td>
</tr>
<tr>
<td></td>
<td>$1.4(1)$</td>
</tr>
<tr>
<td></td>
<td>$2.1(2)$</td>
</tr>
<tr>
<td></td>
<td>$0.3(1)$</td>
</tr>
<tr>
<td></td>
<td>$91(5)$</td>
</tr>
<tr>
<td></td>
<td>$1.1(1)$</td>
</tr>
<tr>
<td></td>
<td>$0.43(6)$</td>
</tr>
<tr>
<td></td>
<td>$\leq 1.4$</td>
</tr>
<tr>
<td></td>
<td>$\leq 0.06$</td>
</tr>
<tr>
<td></td>
<td>$182$</td>
</tr>
<tr>
<td></td>
<td>$116(4) \times 10^{3}$</td>
</tr>
</tbody>
</table>

Results of $2\beta$-experiments with $^{106,116}\text{CdWO}_4$

$^{106}\text{CdWO}_4$, 215 g, 6590 h [1]

$^{116}\text{CdWO}_4$, 1.16 kg, 7593 h [2]

$T_{1/2}$ ($2\beta$, $^{106}\text{Cd}\rightarrow^{106}\text{Pd}$) $\geq 10^{19-21}$ yr

27 new results for $2\beta$ $^{106}\text{Cd}$

9 of them – for the first time

$T_{1/2}$ ($2\nu2\beta$) = 2.5(5)$\times10^{19}$ yr

$T_{1/2}$ ($2\beta$ to $^{116}\text{Sn}^*$) $\geq 10^{21}$ yr

6 new results for $2\beta$ $^{116}\text{Cd}$

[2] A.S. Barabash et al., Talk at Int. Conf. NPAE-2012, September 03–07, Kyiv, Ukraine
Search for $2\beta$-decay of $^{106}\text{Cd}$, stage 2

Experiment is in preparation:

$^{106}\text{CdWO}_4$ in coincidence / anticoincidence with 4-crystals HPGe detector

Registration efficiency $\sim (3\text{–}8)\%$

Expected background – a few counts/yr

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$^{106}\text{CdWO}_4$ in coincidence / anticoincidence with 4-crystals HPGe detector

Registration efficiency $\sim (3\text{–}8)\%$

Expected background – a few counts/yr

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PbWO$_4$ (from low radioactive lead)  
HPGe 225 cm$^3$  
PMT

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Sensitivity to $2\nu\, \epsilon\beta^+$ and $2\beta^+$ of $^{106}\text{Cd}$:

$T_{1/2} \sim (1\text{–}10)\times10^{20}$ yr

Theory [1-4]:

$2\nu2K$  
$1(\text{–} 50)\times10^{20}$ yr

$2\nu\epsilon\beta^+$  
$(8 \text{–} 400)\times10^{20}$ yr

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Summary

• Enriched CdWO₄ scintillators have been developed: ¹⁰⁶CdWO₄ (231 g; 66% of ¹⁰⁶Cd) & ¹¹⁶CdWO₄ (1.9 kg; 82% of ¹¹⁶Cd)

• The total irrecoverable loss of the enriched cadmium on the all stages of ¹⁰⁶,¹¹⁶CdWO₄ development does not exceed 3%

• The produced ¹⁰⁶,¹¹⁶CdWO₄ scintillators exhibit excellent optical and scintillation properties, and high level of radiopurity. Low segregation of Th and Ra by CdWO₄ was observed

• Double beta (2β) experiments using ¹⁰⁶,¹¹⁶CdWO₄ were realized at the Gran Sasso underground laboratory (Italy). Measurements with ¹¹⁶CdWO₄ are in progress. A next step experiment “¹⁰⁶CdWO₄ in 4-crystals HPGe” is in preparation

• By analyzing the accumulated data, the new half-life ($T_{1/2}$) limits on 2β decay of ¹⁰⁶,¹¹⁶Cd were set at $\text{lim} T_{1/2} \sim 10^{19-21}$ yr, $2\nu 2\beta$ decay of ¹¹⁶Cd was measured with $T_{1/2} = (2.5\pm0.5)\times10^{19}$ yr