Radioactive contamination of enriched $^{106,116}CdWO_4$ crystal scintillators

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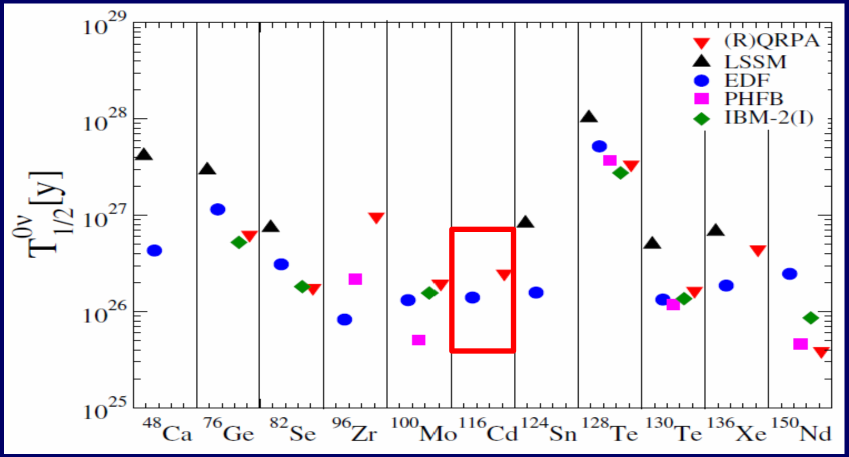
- Motivation
- Development of $^{106,116}CdWO_4$ crystal scintillators
- Experiments to search for 2β decay of ^{106}Cd and ^{116}Cd
- Radioactive contamination $^{106,116}CdWO_4$ scintillators
- Segregation of thorium, radium and potassium
- Conclusions

Motivation

^{116}Cd

One of the most promising isotopes to search for 0ν2β decay

- $Q_{2\beta}$ = 2813 keV, δ = 7.5%
- promising theoretical calculation
- the possibility of isotopic enrichment in large amount

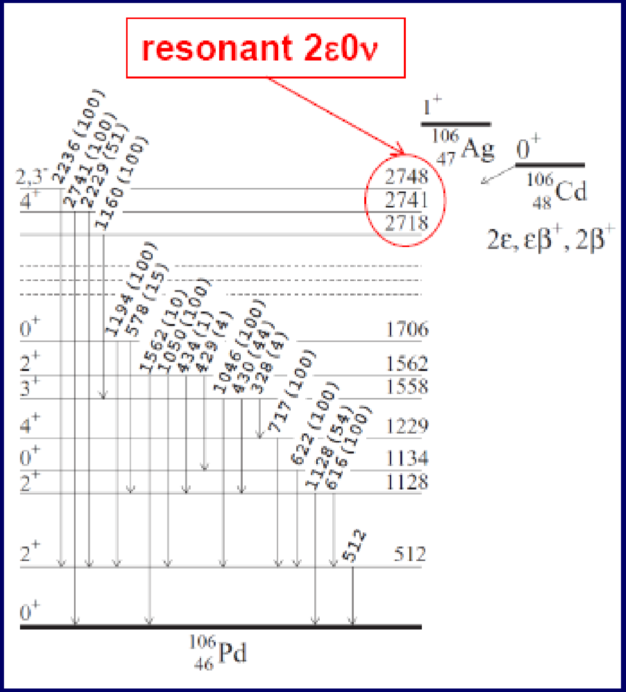


- good scintillation properties
- low levels of internal contamination
- particle discrimination ability to reduce background
- well established production

^{106}Cd

The one among six 2β$^+$ isotopes (2ε, εβ$^+$, 2β$^+$)

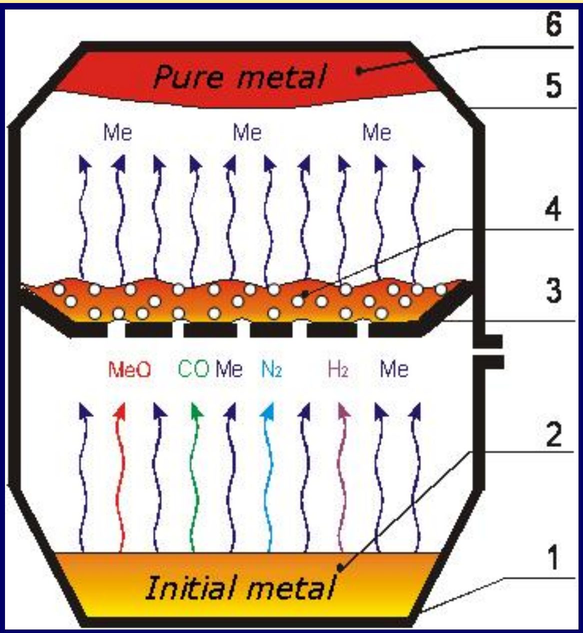
- $Q_{2\beta}$ = 2775 keV, δ = 1.3%
- possibility to distinguish mass and RCH mechanism
- resonant 2ε0ν decay on the exited levels of ^{106}Pd



Production of $^{106,116}CdWO_4$

Purification of ^{106}Cd and ^{116}Cd metal samples

Distillation through getter filters



1 – crucible; 2 – initial metal; 3 – plate with holes; 4 – getter; 5 – condenser; 6 – purified metal

result →

Concentration of impurities in ^{106}Cd (ppm)

Element	Before	After
K	11	0.04
Ni	0.6	< 0.2
Cu	5	0.5
Fe	1.3	0.4
Mg	12	<0.05
Mn	0.1	0.1
Cr	9	<0.1
Pb	270	<0.3

R.Bernabey *et al.*, Metallofiz. Nov. Tekhn. 30 (2008) 477
G.P.Kovtun *et al.*, Functional Materials 18 (2011) 121

Production of $^{106,116}CdWO_4$

Synthesis of $^{106,116}CdWO_4$ compounds

After dissolving the metallic cadmium in nitric acid, the purification was realized by co-precipitation on a collector. Solutions of cadmium nitrate and ammonium para-tungstate were mixed and then heated to precipitate cadmium tungstate:

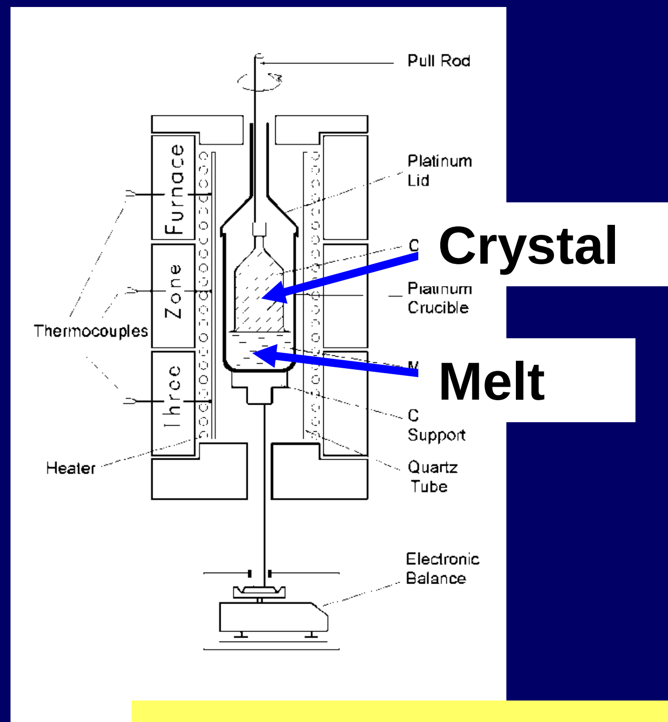
$$Cd(NO_3)_2 + (NH_4)_2WO_4 \rightarrow CdWO_4 + 2NH_4NO_3$$

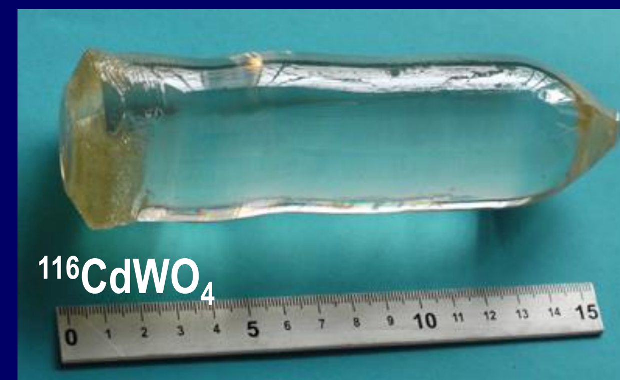
- All the operations were carried out by using quartz or polypropylene lab-ware, materials with low level of radioactive contaminations
- Reagents of high purity grade (concentration of any metal less than 0.01 ppm)
- Water, acids and ammonia were additionally distilled by laminar evaporation in quartz installation
- Additional recrystallization was performed to purify ammonium para-tungstate

P. Belli *et al.*, NIMA 615 (2010) 301
A. Barabash *et al.*, JINST 6 (2011) P08011

Production of $^{106,116}CdWO_4$

Crystal growth by Low-Thermal-Gradient Czochralski technique



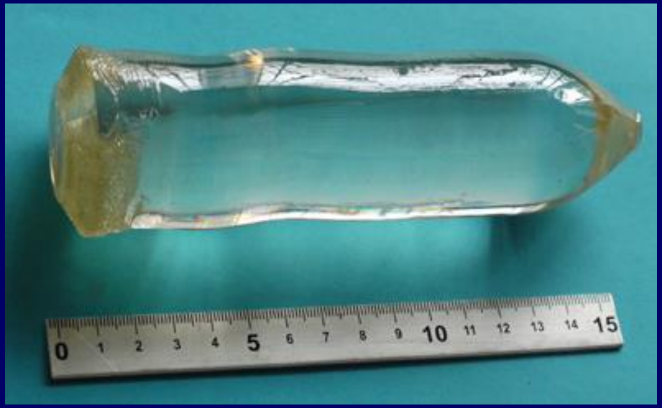


	standard	LTG-C
Output	25-30%	up to 90%
Quality		typically higher
Radiopurity		expected better
Loses of powder	2-3%	<0.3%

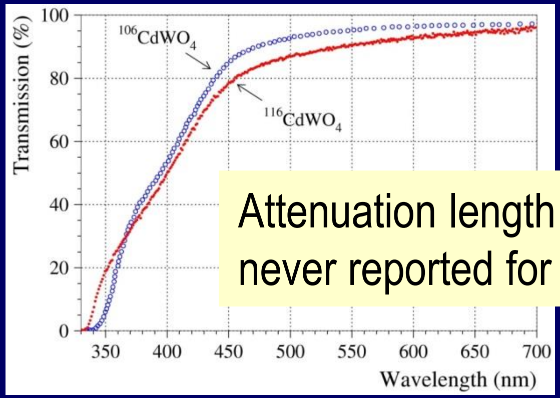
[1] A.A. Pavlyuk *et al.*, Proc. *APSAM*-92, April 26–29, Shanghai, China (1992)

Optical properties and energy resolution

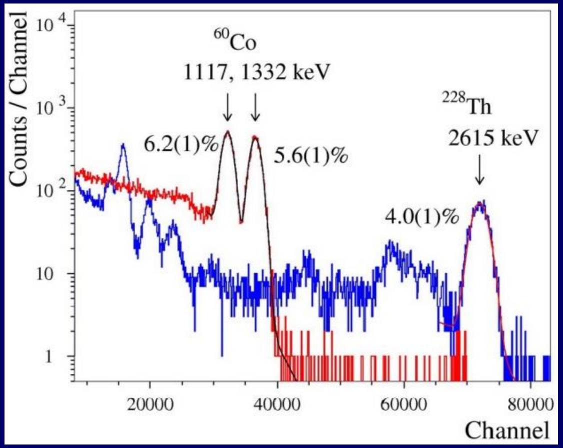
$^{106}CdWO_4$ 231 g (87%) [1]



$^{116}CdWO_4$ 1868 g (87%) [2]



Attenuation length ≈ 50-60 cm never reported for $CdWO_4$



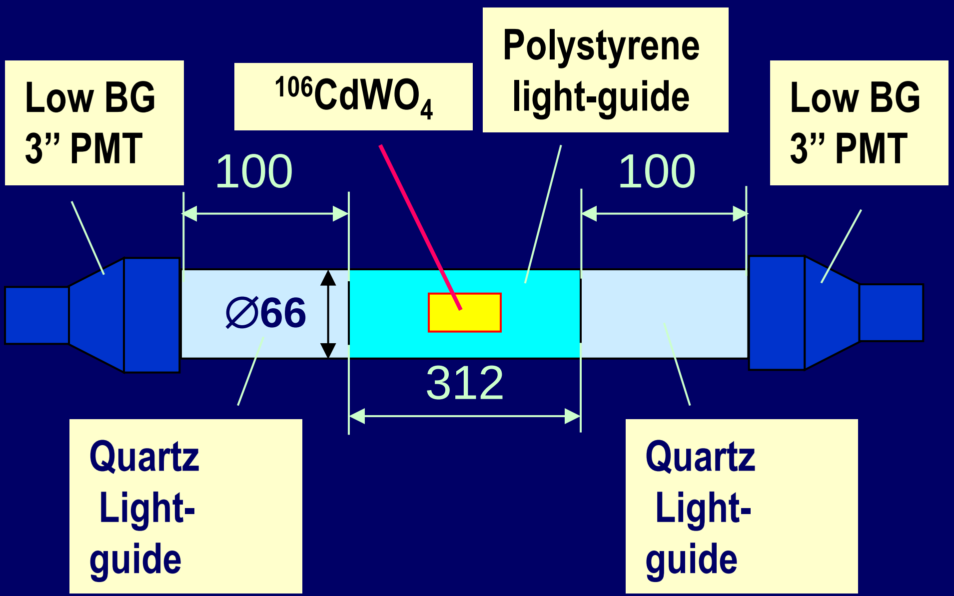
Excellent optical and scintillation properties thanks to special R&D to purify raw materials and Low-thermal-gradient Czochralski technique to grow the crystal

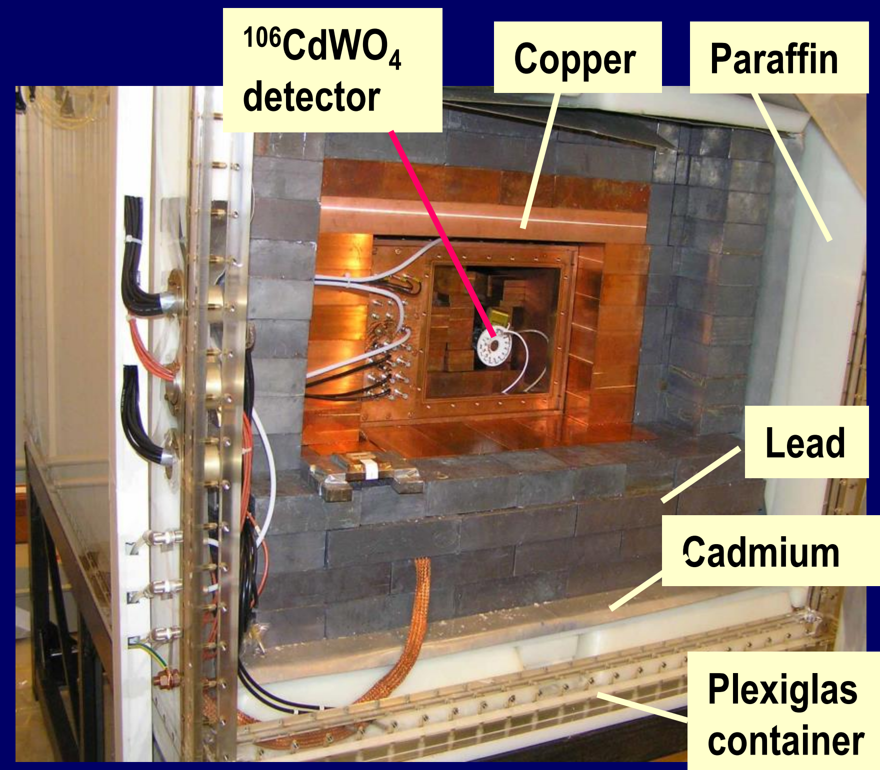
[1] P. Belli *et al.*, NIMA 615 (2010) 301

[2] A.S. Barabash *et al.*, JINST 6 (2011) P08011

The total losses of ^{106}Cd ≈ 2%

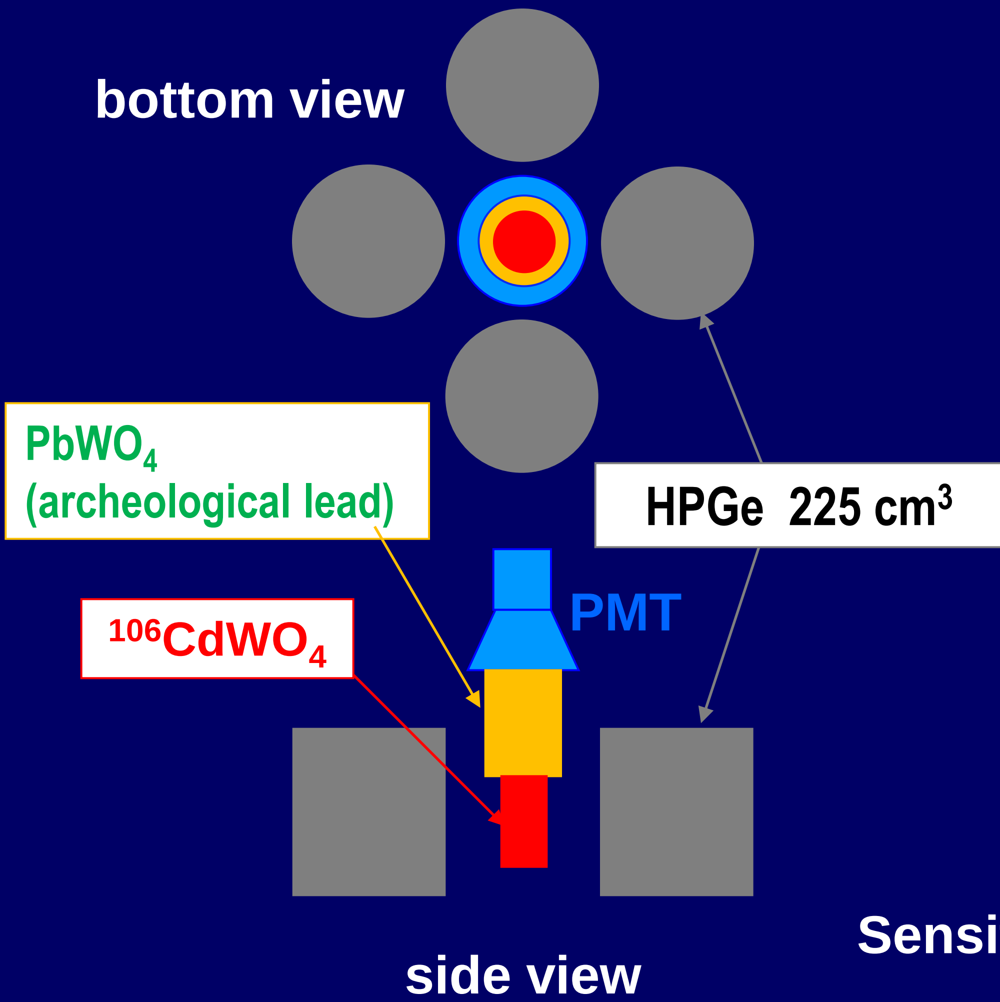
Low background $^{106}CdWO_4$ detector in DAMA R&D at LNGS





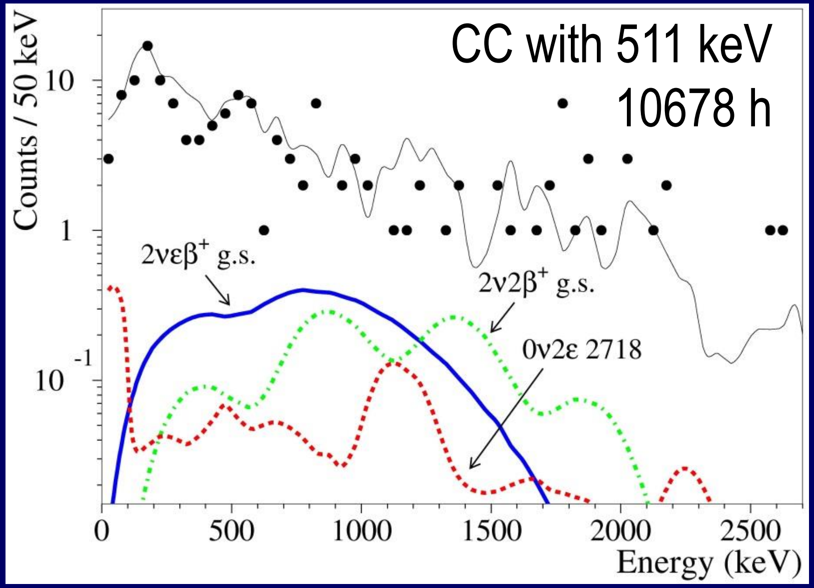
P. Belli *et al.*, PRC 85 (2012) 044610

$^{106}CdWO_4$ in the GeMulti setup with 4 HPGe



4 HPGe, ~ 225 cm^3 each, in one cryostat

$^{106}CdWO_4$ in coincidence / anticoincidence with HPGe

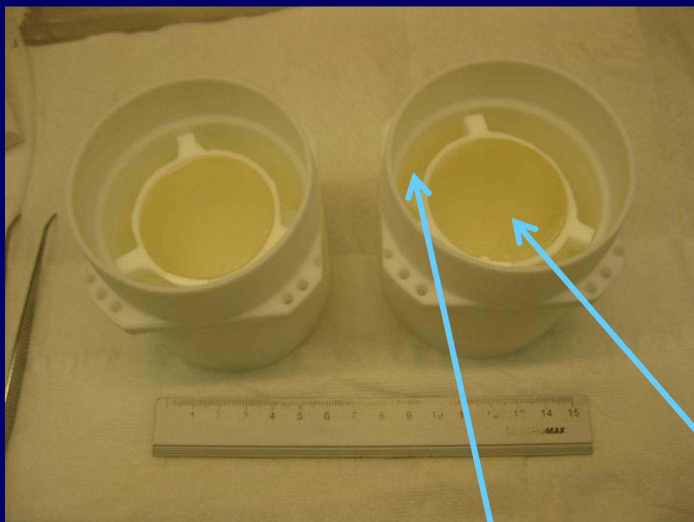


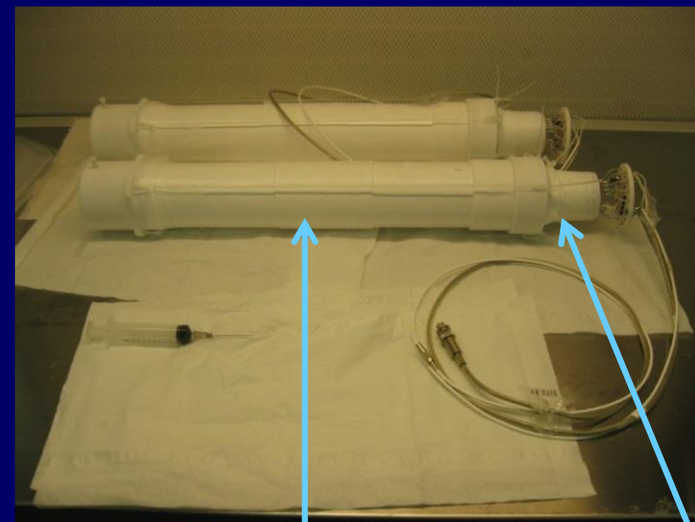
Sensitivity to 2ε, εβ$^+$ and 2β$^+$ decay of ^{106}Cd:

$T_{1/2}$ ~ 10^{19} – 10^{21} yr

V.I. Tretyak et al., EPJ WC 65(2014)01003.

$^{116}CdWO_4$ detector





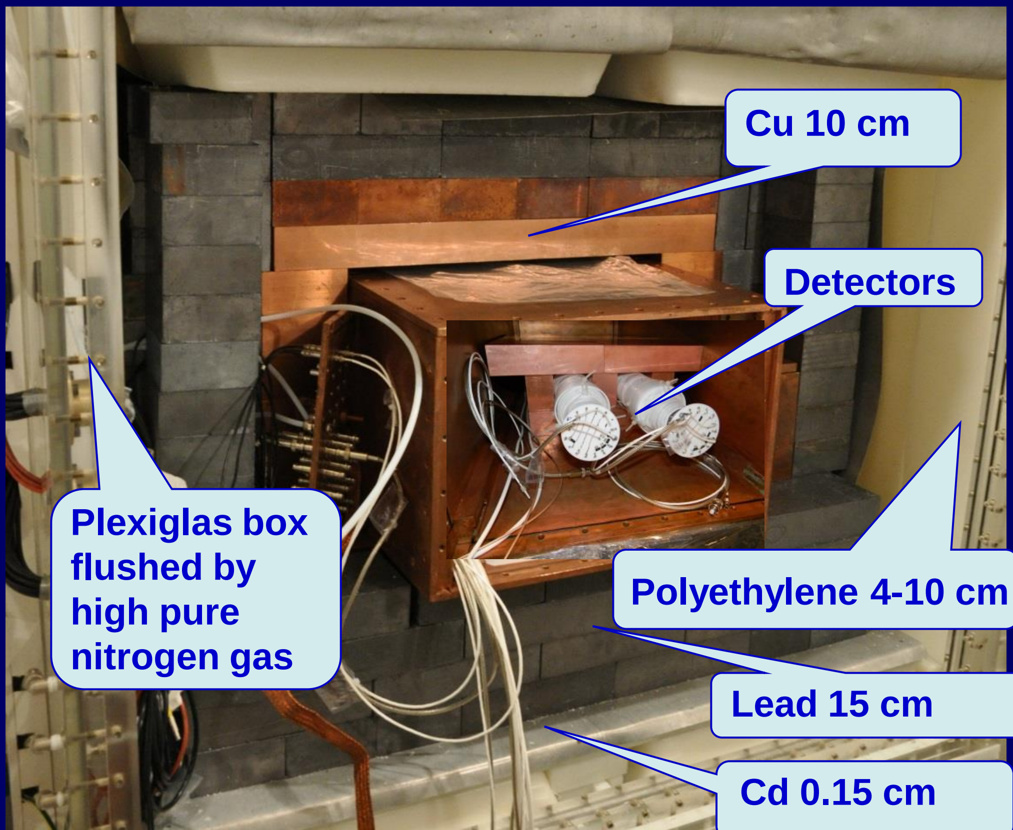
$^{116}CdWO_4$

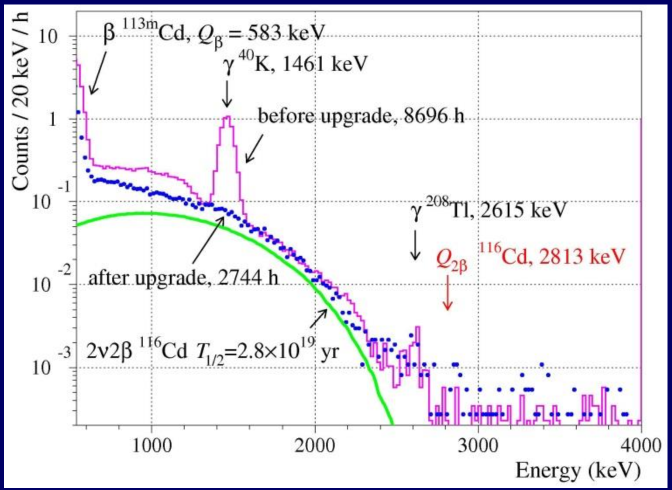
HP quartz light-guide

PMT

Borexino liquid scintillator in Teflon container

$^{116}CdWO_4$ set-up (DAMA R&D at LNGS)

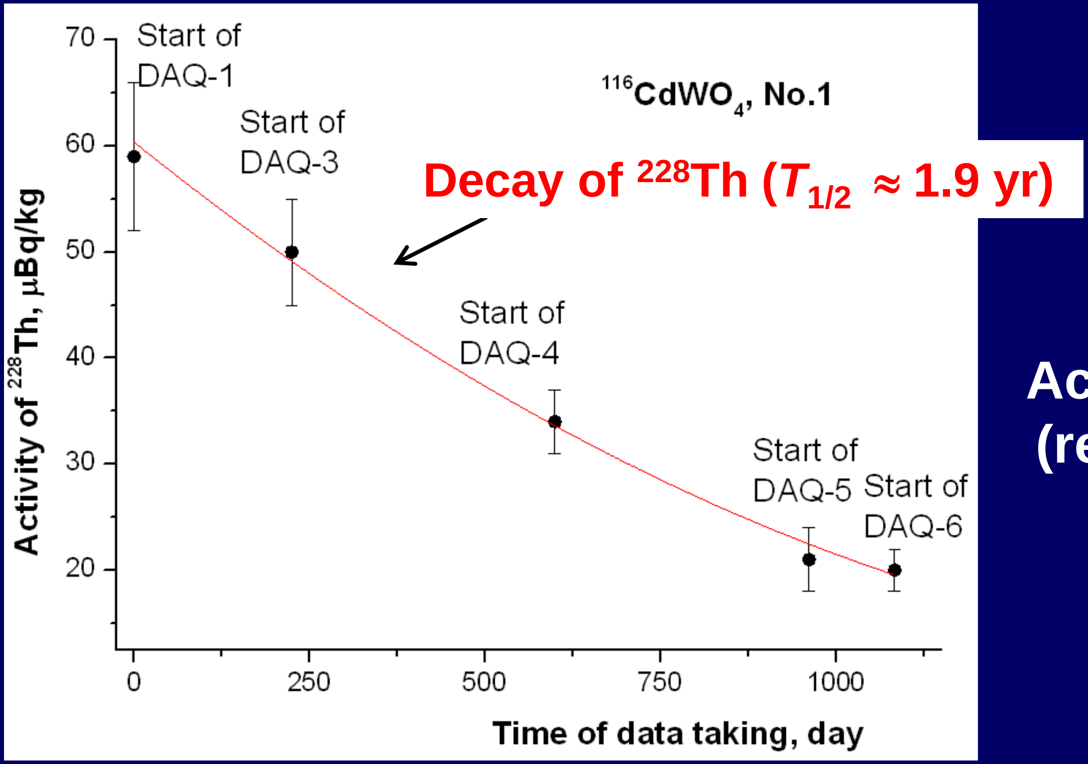




^{228}Th and ^{226}Ra by time-amplitude analysis

D.V. Poda et al., EPJ WC 65 (2014) 01005.

Decay of ^{228}Th in ^{116}CdWO$_4$



Activity of ^{228}Th (in μBq/kg)
(reference date: May 2014)

No.1	**20(2)**
No.2	**39(3)**

- **Contamination is mainly by thorium**
- **Radium is much lower**

Radiopurity of $^{106,116}CdWO_4$ and $CdWO_4$

(mBq/kg) **Ref data April 2013**

Nuclide	**$^{106}CdWO_4$** [1]	**$^{116}CdWO_4$** [2]	**$CdWO_4$** [3,4]
^{40}K	<1.4	<1	< (1.7 – 5)
^{110m}Ag	**<0.06**	**= 0.12(4)**	–
^{113}Cd	= 182(1)	= 100(10)	= 558(4)
^{113m}Cd	= 116 000(4000)	= 460(20)	< 3.4 – 150
^{232}Th	<0.07	<0.08	< 0.03
^{228}Th	**= 0.042(4)**	**= 0.060(6)**	**< (0.003 – 0.014)**
^{238}U	<0.6	<0.5	<1.3
^{226}Ra	**= 0.012(3)**	**<0.005**	**< (0.007 – 0.02)**
^{210}Po	<0.2	<0.5	< 0.06
Total α	**= 2.1(2)**	**= 1.9(2) – 2.7(3)**	**= 0.26(4)**

[1] P. Belli *et al.*, PRC 85 (2012) 044610

[2] A. Barabash *et al.*, JINST 6 (2011) P08011

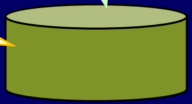
[3] F.A. Danevich *et al.*, Z. Phys. A 355 (1996) 433

[4] P. Belli *et al.*, Phys. Rev. C 76 (2007) 064603

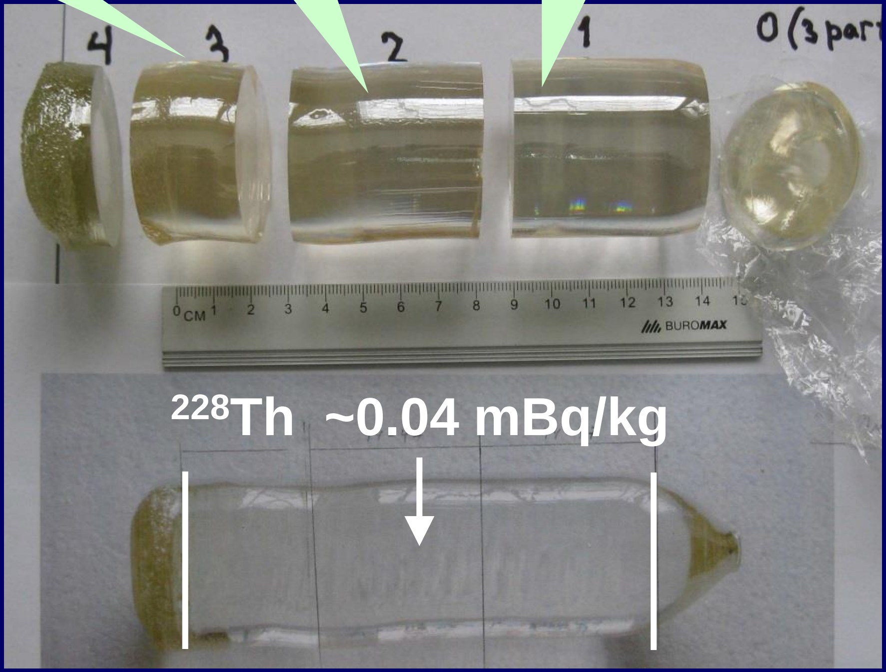
Possibility to improve the radiopurity of $^{116}CdWO_4$ by recrystallization

Activity of ^{228}Th: 10(2) | 0.09(1) | 0.04(1) | 0.02(1) **May 2014**

rest of the melt after the crystal growth 279 g



Nuclide	Crystal	Rest of melt
^{40}K	<1	27(11)
^{226}Ra	<0.005	64(4)
^{228}Th	0.02 – 0.09	10(2)



^{228}Th in charge ~1.4 mBq/kg

We expect to reduce K, Th, U and Ra contamination by recrystallization

Thorium expected to be reduced by a factor ~35 ⇒ 1 μBq/kg

conclusions

- **High quality $CdWO_4$ crystal scintillators were developed from enriched ^{106}Cd and ^{116}Cd with output 87%, irrecoverable losses 2%**
- **Radioactive contamination of the ^{106,116}CdWO4 crystals is on the level of 0.05 mBq/kg ^{228}Th, ~ < 0.01mBq/kg of ^{226}Ra**
- **Strong segregation of thorium is observed in $CdWO_4$ crystals, substantial reduction of Th (~ 35 times) may be achieved by recrystallization**