Development of $CdWO_4$ crystal scintillators from ^{106}Cd and ^{116}Cd, and of $Zn^{100}MoO_4$

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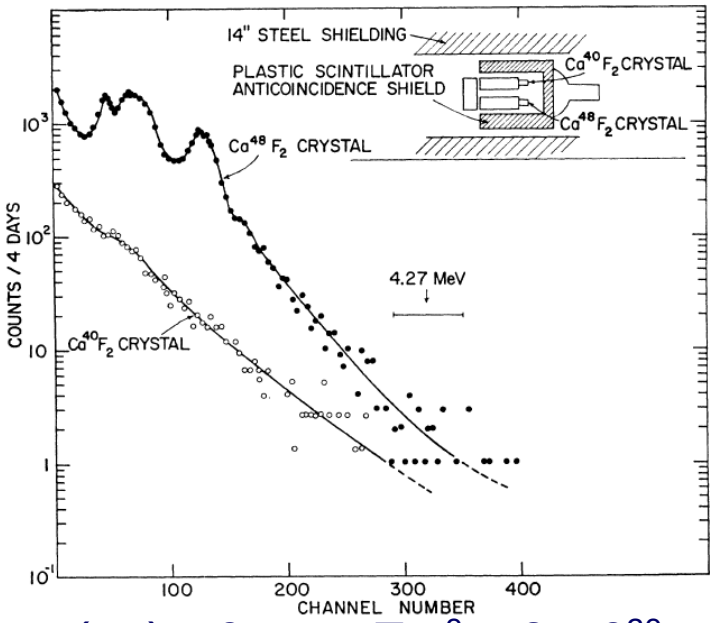
http://lpd.kinr.kiev.ua

- **Scintillators in 2β experiments**
- **Specific requirements to scintillators from enriched materials**
- **Enriched $^{106,116}CdWO_4$ crystals**
- **R&D of $Zn^{100}MoO_4$**
- **Conclusions**

2β experiments with crystal scintillators

Limits for Lepton-Conserving and Lepton-Nonconserving Double Beta Decay in Ca^{48}†

E. DER MATEOSIAN AND M. GOLDHABER
Brookhaven National Laboratory, Upton, New York
(Received 10 February 1966)



$^{48}CaF_2$(Eu), 19 g → $T_{1/2}^{0\nu} > 2\times10^{20}$ yr [1]

- **$CdWO_4$, $^{116}CdWO_4$, $^{106}CdWO_4$ →** ^{106,108,114,116}Cd, ^{180}W, ^{186}W
- **CaF_2(Eu) →** ^{40,48}Ca
- **$CaWO_4$ →** ^{48}Ca
- **GSO(Ce) →** ^{160}Gd, ^{136,138,142}Ce
- **$ZnWO_4$ →** ^{64,70}Zn, ^{180,186}W
- **$^{40}Ca^{100}MoO_4$ →** ^{100}Mo
- **CeF_3, $CeCl_3$ →** ^{136,138,142}Ce
- **BaF_2 →** ^{130,132}Ba
- **$SrCl_2$, SrI_2(Eu) →** ^{84}Sr

$T_{1/2}^{0\nu} > 1.7 \times 10^{23}$ yr ($0\nu2\beta$ of ^{116}Cd)

$T_{1/2}^{2\nu} = 3 \times 10^{19}$ yr ($2\nu2\beta$)

- **A sensitivity to explore the inverted hierarchy: $T_{1/2} \sim 10^{26} - 10^{27}$ yr**
- **Cryogenic scintillating bolometers able to provide such a sensitivity**

[1] E. der Matosian and M. Goldhaber, Phys. Rev. 146 (1966) 810

Requirements to scintillators for 2β experiments

Sensitivity (half-life $T_{1/2}$) of 0ν2β experiments:

$$T_{1/2} \propto \varepsilon \cdot \delta \sqrt{\frac{m \cdot t}{R \cdot BG}}$$

ε – detection efficiency
δ – abundance of candidate nuclei in the detector
m – mass of detector
t – time of measurements
R – energy resolution
BG – background

- Maximal concentration of an isotope of interest
- Very low (ideally zero) radioactive contamination
- High scintillation properties (energy resolution, light yield, reasonably fast scintillation decay)
- Large enough volume ~ 10^2 cm^3

Specific requirements to scintillators from enriched isotopes

(typically unclear for producers of scintillators)

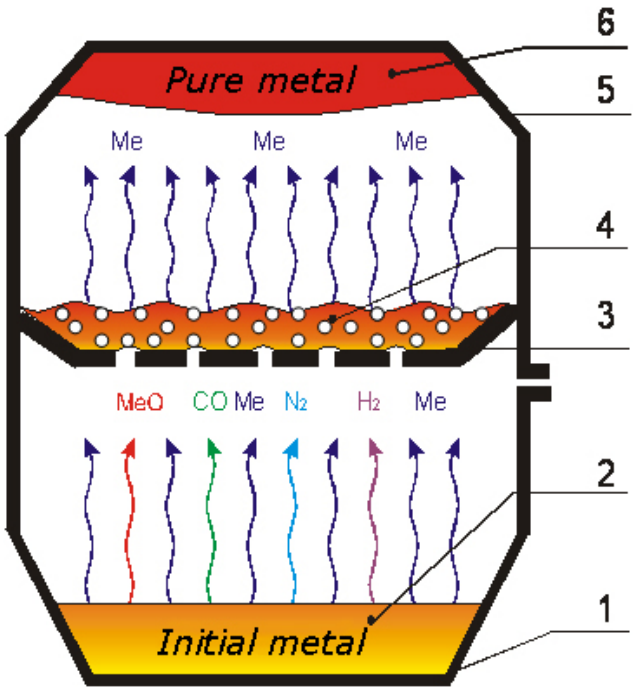
- Low contamination by Th, Ra, U, K, rare earth elements
- Minimal loses and contamination of enriched materials
- Recovery and purification of enriched material form the scraps after detectors production
- Prevention of neutrons & cosmogenic activation

Development and radioactive contamination of $^{106,116}CdWO_4$ scintillators

Purification of ^{nat}Cd, ^{106}Cd and ^{116}Cd

Kharkiv Institute of Physics and Technology, Ukraine

Distillation through getter filters



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

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**

Measured by: ICP- MS *, Laser Mass Spectroscopy **, Atomic Absorption Spectroscopy ***

R.Bernabey *et al.*,Metallofiz. Nov. Tekhn. 30 (2008) 477

G.P.Kovtun *et al.*, Functional Materials 18 (2011) 121

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

Joint Stock Company NeoChem, Moscow, Russia

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

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

- 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

Methods to recover Cd from $CdWO_4$ crystalline scraps were developed and used to recover ^{106}Cd and ^{116}Cd

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

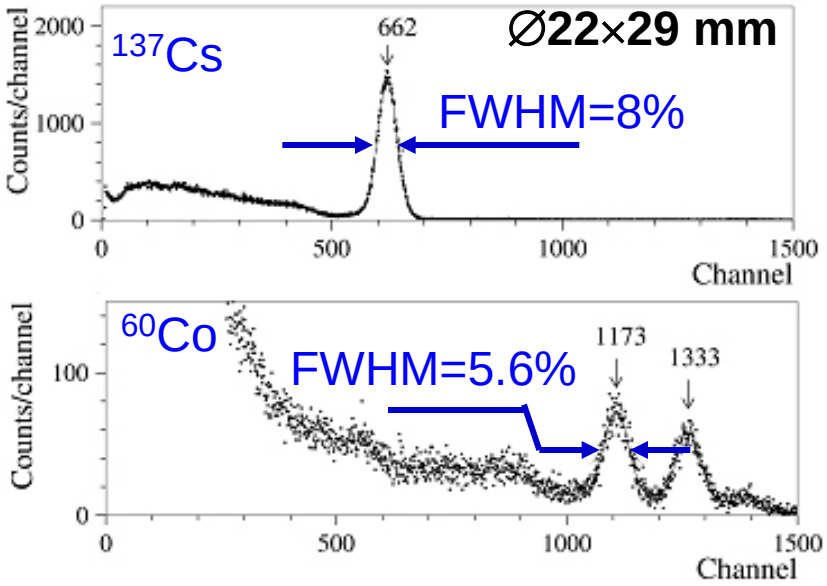
Growth of $CdWO_4$ from purified Cd

$CdWO_4$ scintillators of improved quality were grown by standard Czochralski method

in collaboration with the Institute of Scintillation Materials, Kharkiv, Ukraine

"Usual" $CdWO_4$ powder

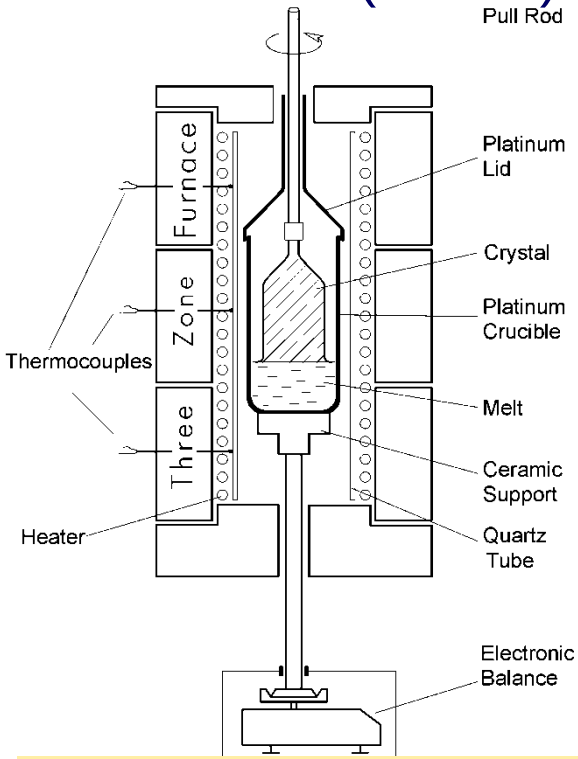
Additionally purified Cd and W

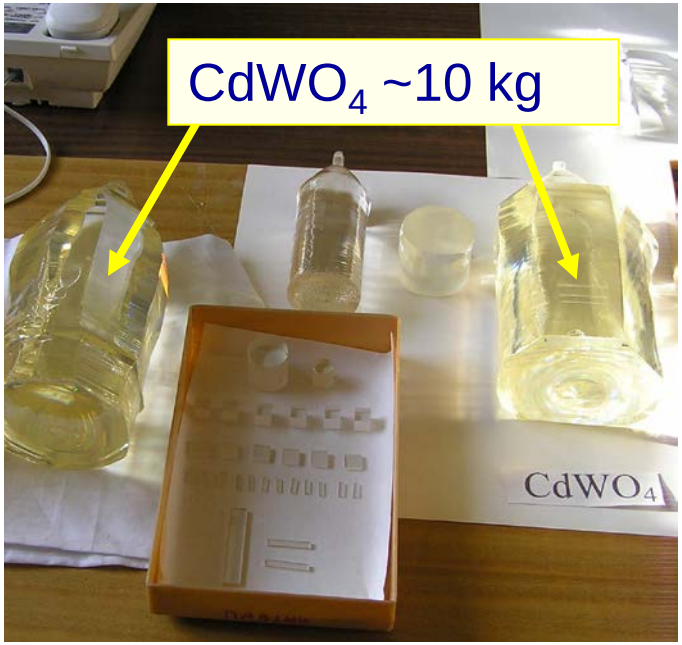


R.Bernabey *et al.*, Metallofiz. Nov. Tekhn. 30 (2008) 477

Growth of $^{106}CdWO_4$ and $^{116}CdWO_4$

Low-Thermal-Gradient Czochralski (LTG-C) technique to grow crystals





However, it works

Nikolaev Institute of Inorganic Chemistry, Novosibirsk, Russia

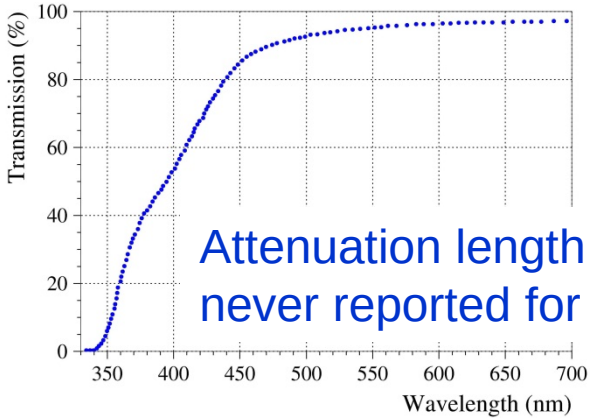
	standard	LTG-C
Output	25-30%	up to 90%
Quality		typically higher
Radiopurity		expected better
Loses of powder	2-3%	<0.3%

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

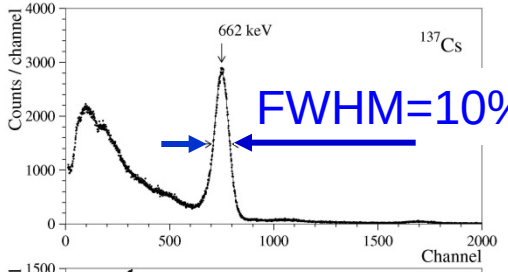
$^{106}CdWO_4$ crystal scintillator

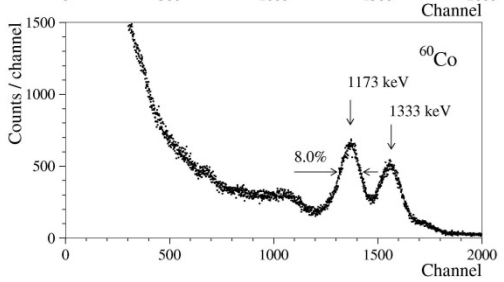
$^{106}CdWO_4$ boule 231 g (87.2%)
The total losses of ^{106}Cd = 2.3%

$^{106}CdWO_4$ scintillator 215 g,
66% of ^{106}Cd (1.25% natural)



Attenuation length 60 cm
never reported for $CdWO_4$

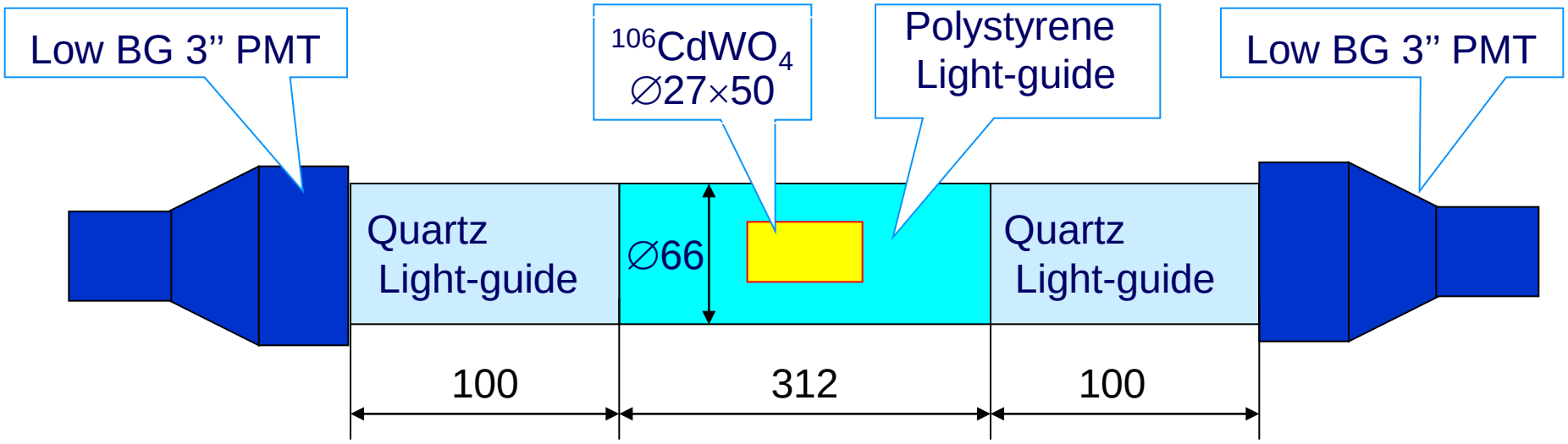


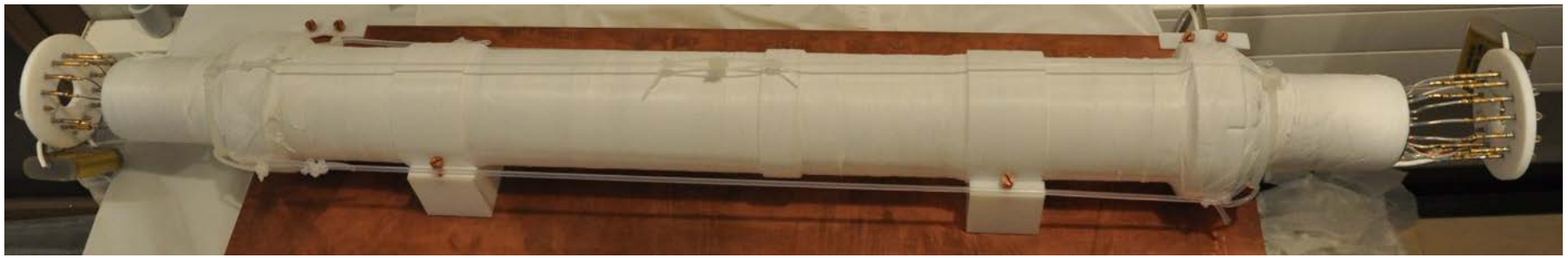


Excellent optical and scintillation properties thanks to special R&D to purify raw materials and Low-thermal-gradient Czochralski technique to grow the crystal

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

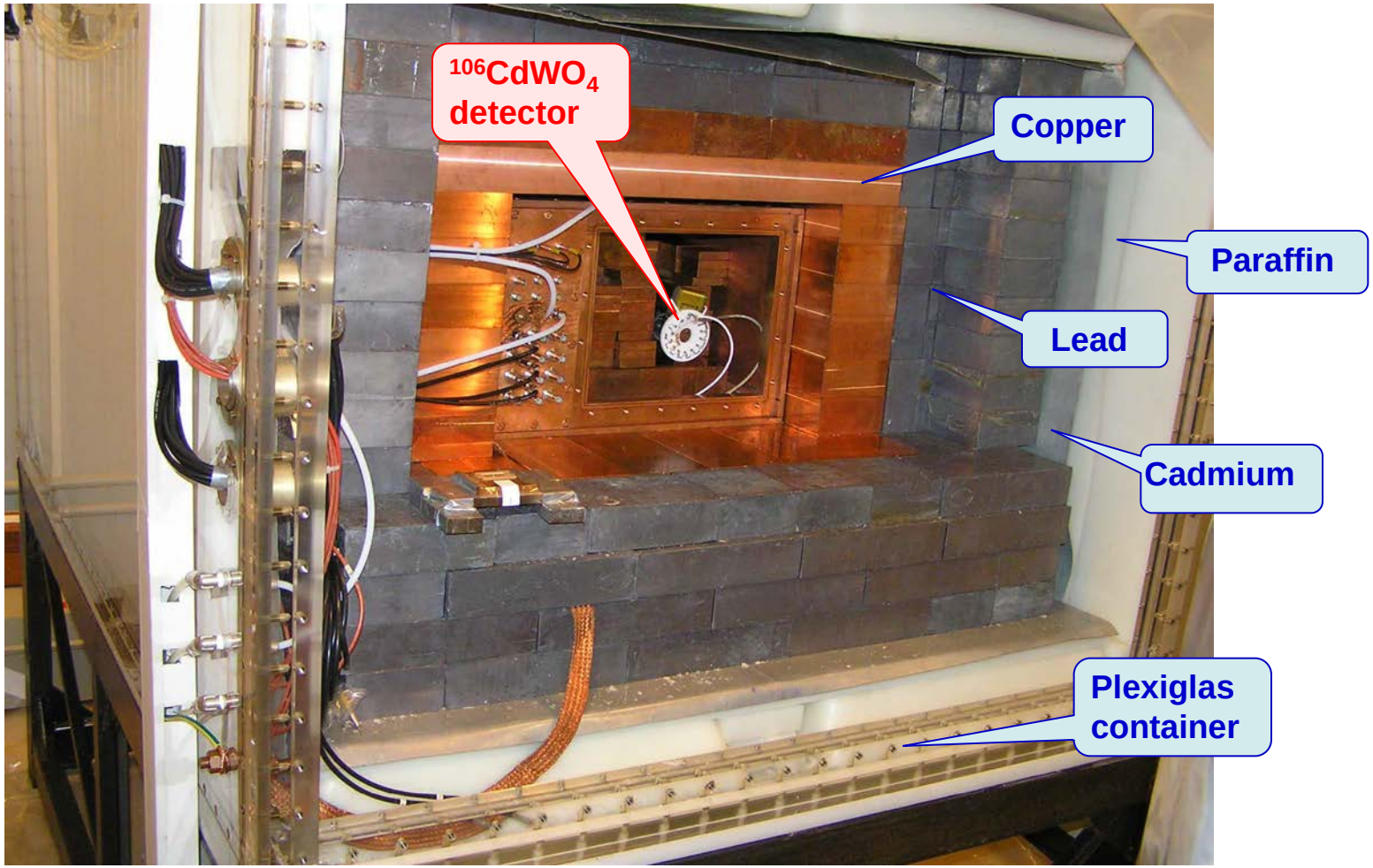
Low background scintillation detector with $^{106}CdWO_4$ crystal scintillator



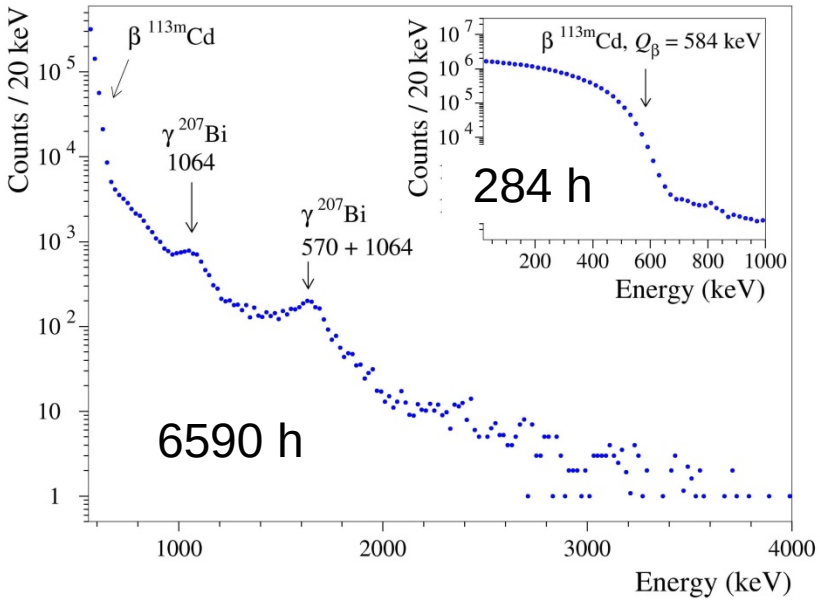


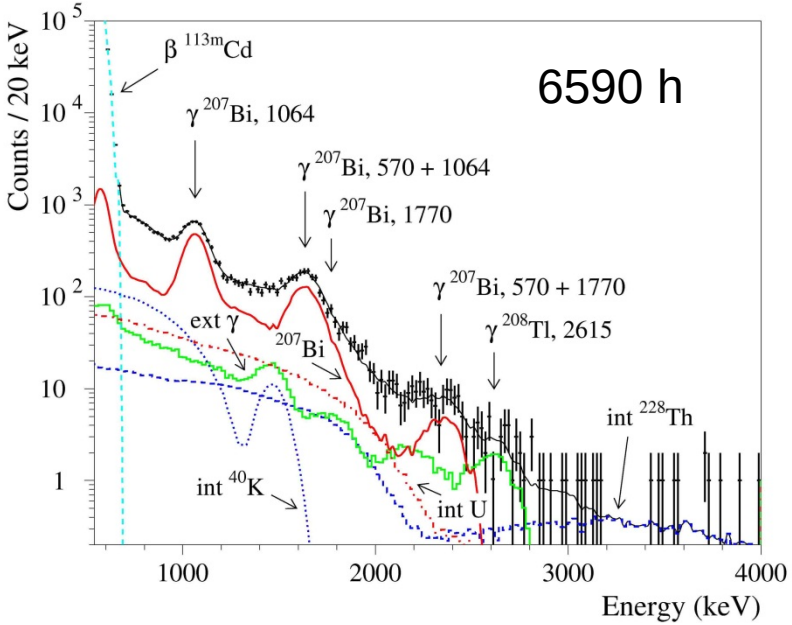
Low background scintillation set-up DAMA/R&D

Gran Sasso National Laboratories of the INFN (Italy)



Background of $^{106}CdWO_4$ detector



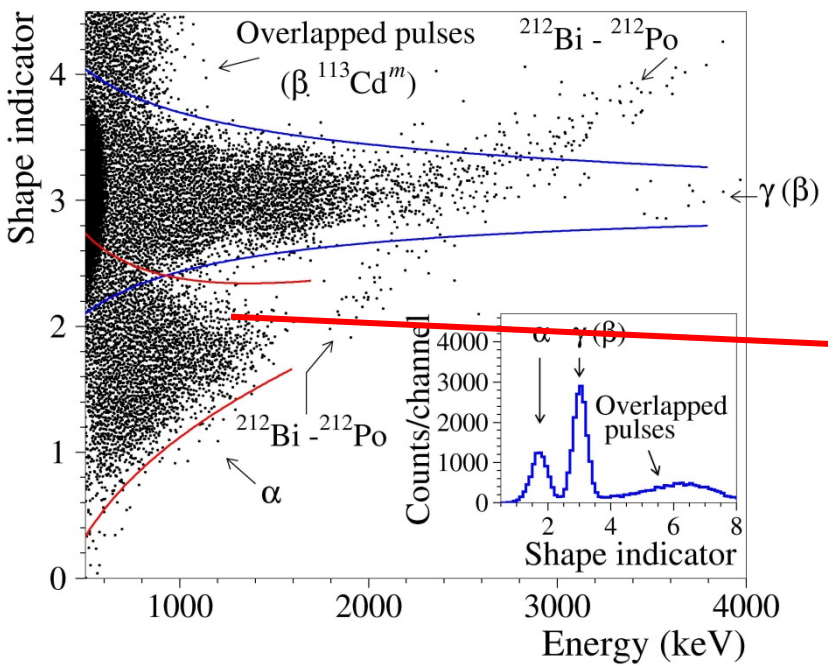


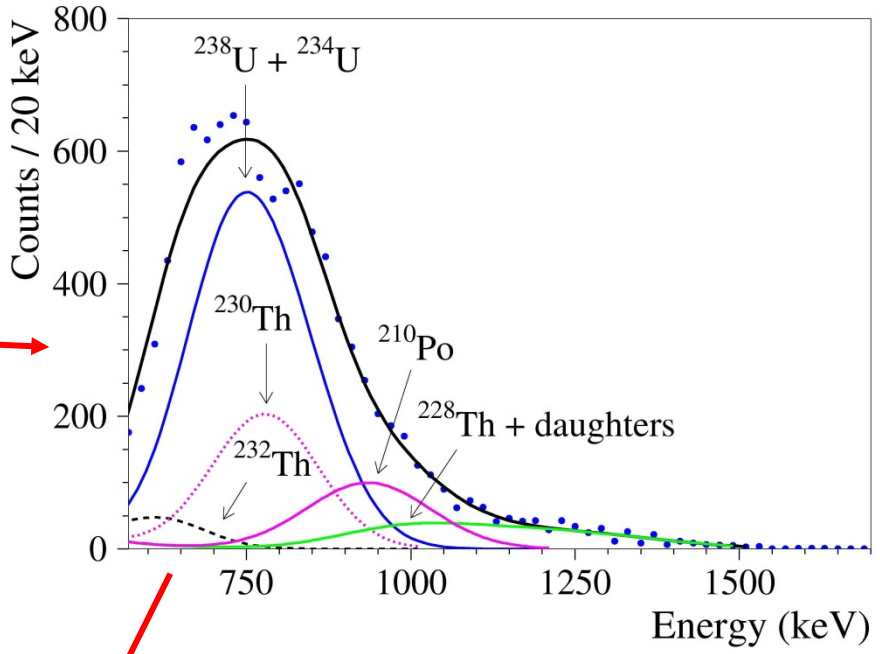
Contamination of $^{106}CdWO_4$ (mBq/kg)

^{207}Bi	**<0.7 (= 0.06 mBq/cm^2 on surface)**
^{113m}Cd	**116 000**
^{40}K	**<1.4**

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

Pulse-shape analysis

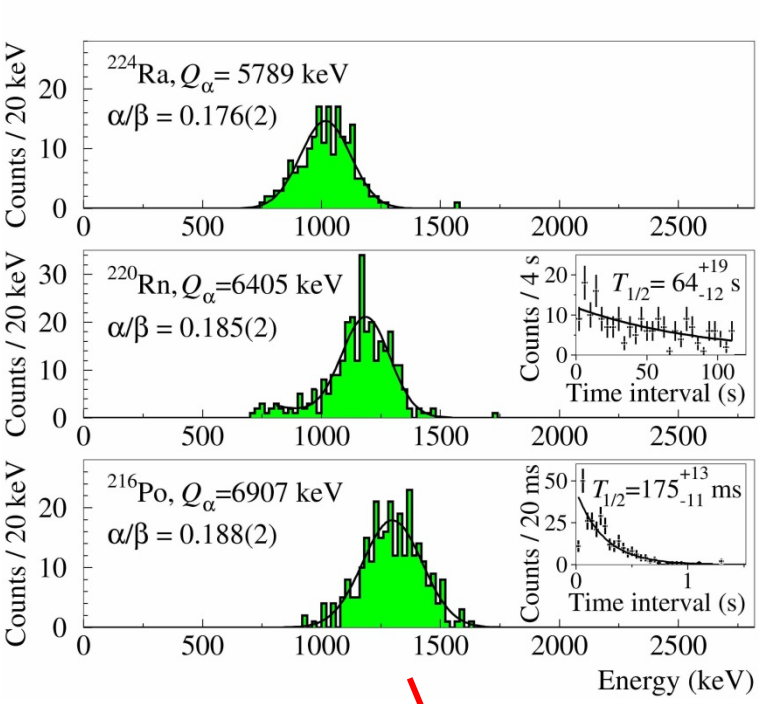


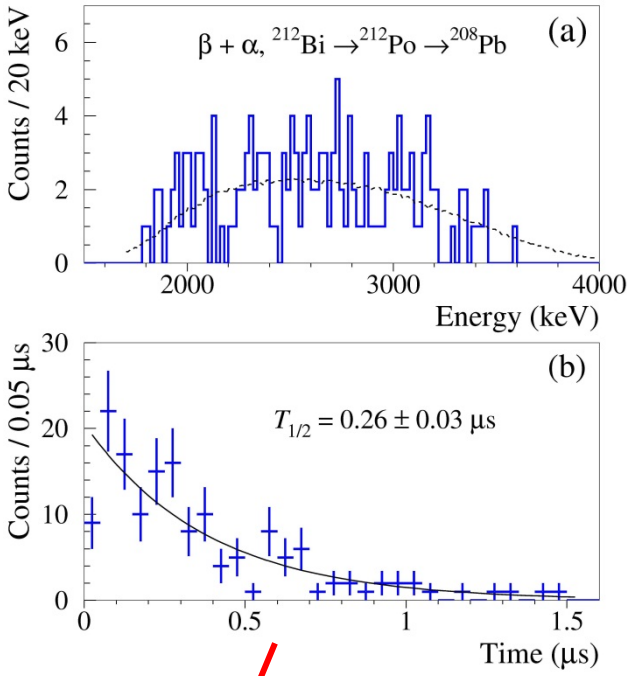


Contamination of $^{106}CdWO_4$ (mBq/kg)

^{232}Th	**<0.07**
^{238}U	**<0.6**
^{230}Th	**<0.4**
^{210}Po	**<0.2**

Time-Amplitude and Bi-Po analyses





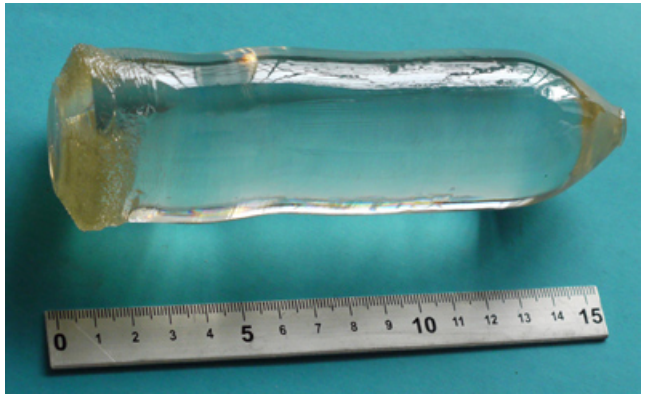
Contamination of $^{106}CdWO_4$ by **^{228}Th** (mBq/kg)

0.042(2) **0.051(4)**

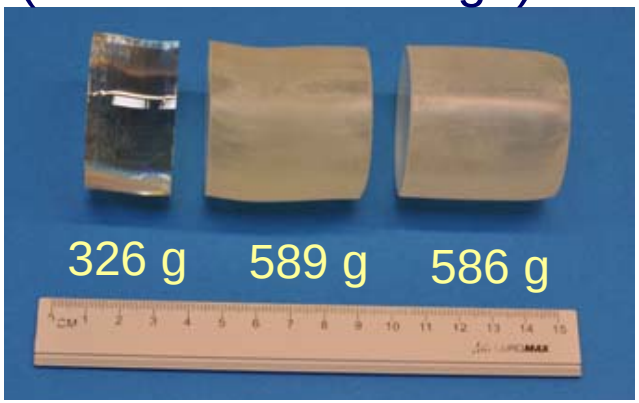
Contamination by **^{226}Ra** (mBq/kg)

0.012(3)

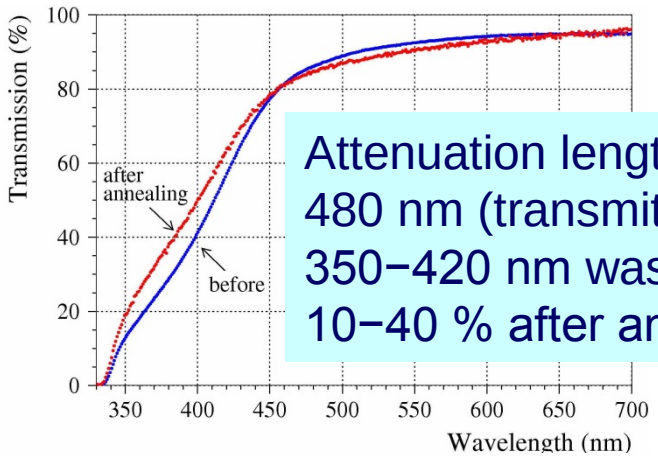
$^{116}CdWO_4$ scintillator



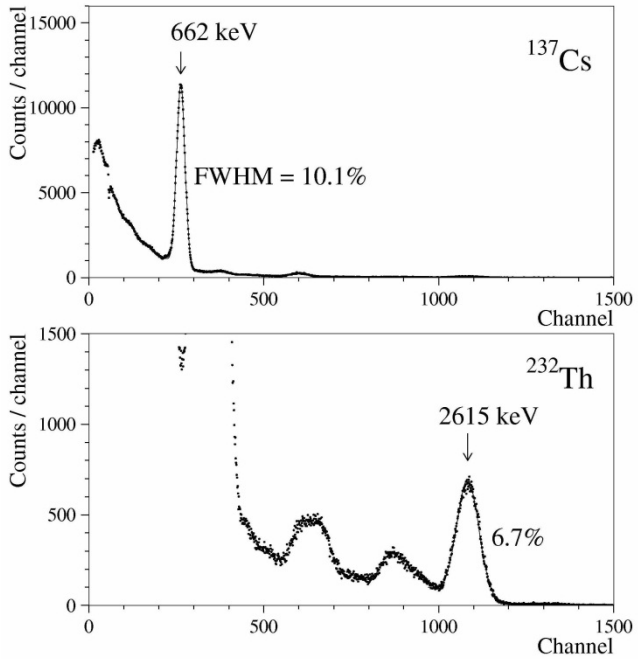
$^{116}CdWO_4$ crystal boule 1868 g (87% of initial charge)



Scintillation elements
Abundance of ^{116}Cd is 82%

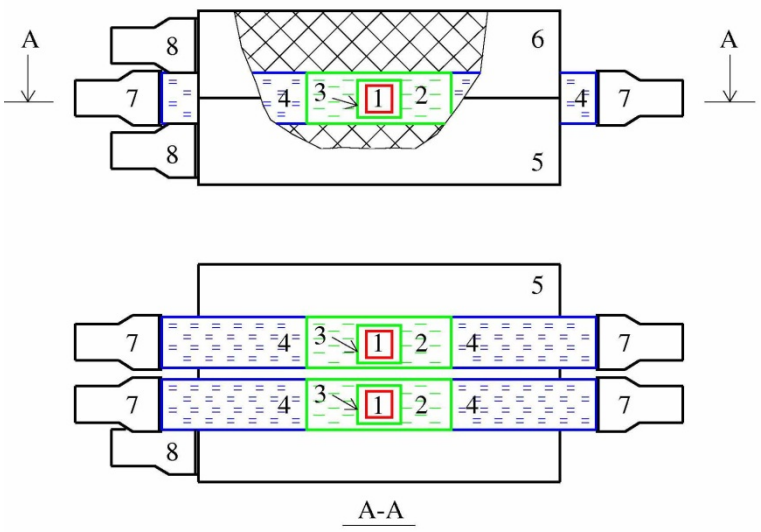


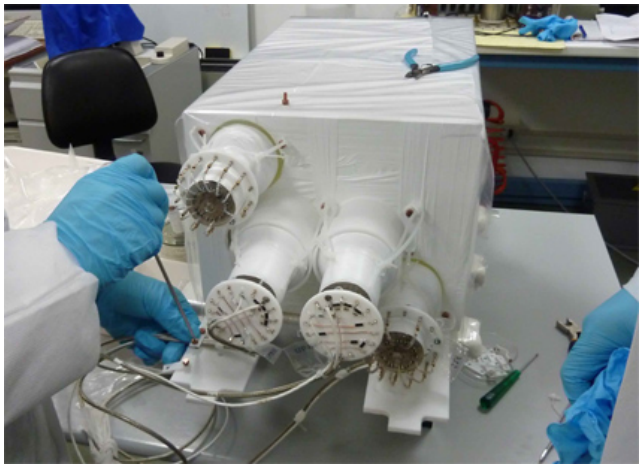
Attenuation length ≈30 cm @ 480 nm (transmittance at 350−420 nm was improved on 10−40 % after annealing)

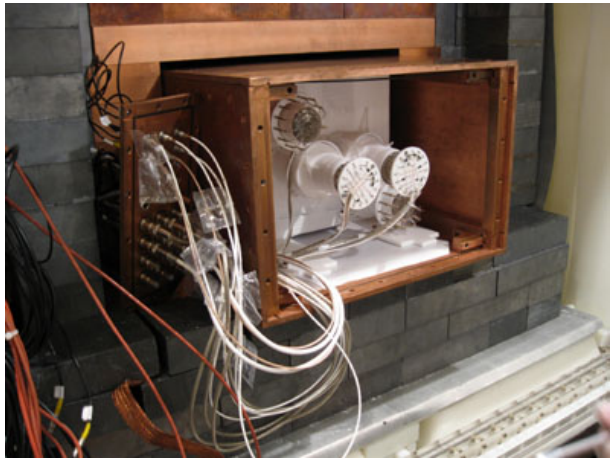


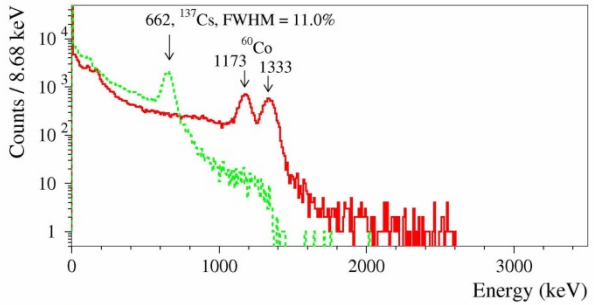
A. Barabash *et al.*, JINST 6 (2011) P08011

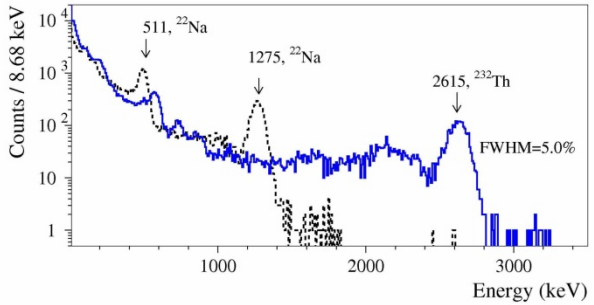
Low background detector with the $^{116}CdWO_4$ scintillators





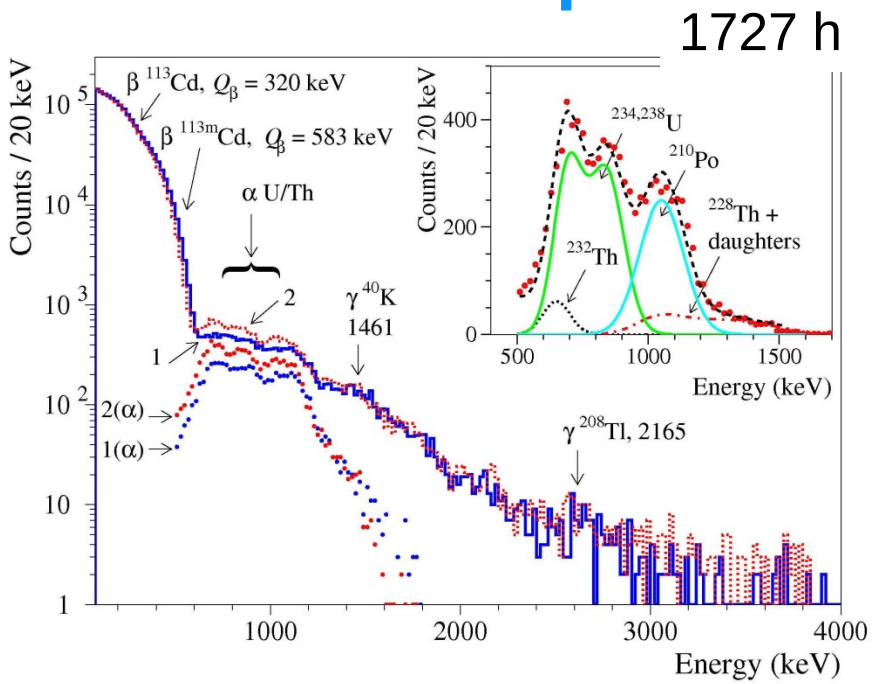


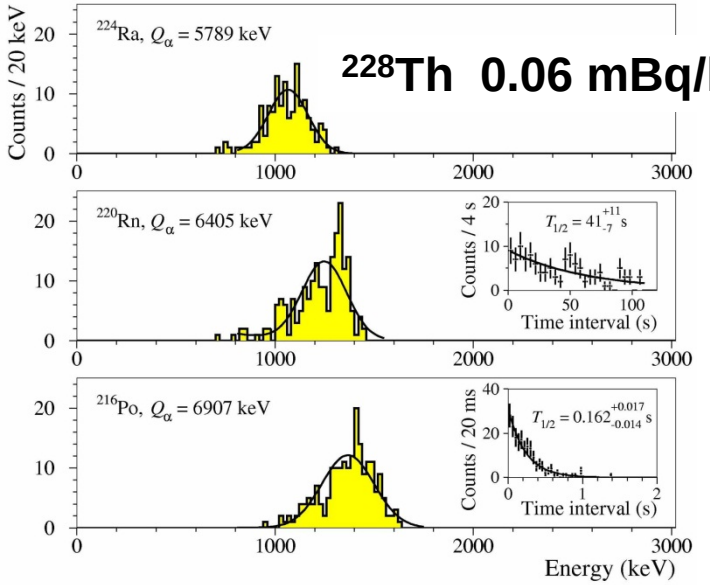




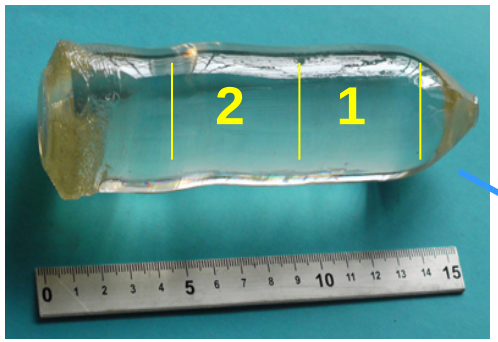
A. Barabash *et al.*, JINST 6 (2011) P08011

Pulse-shape and t-A analyses





^{228}Th 0.06 mBq/kg



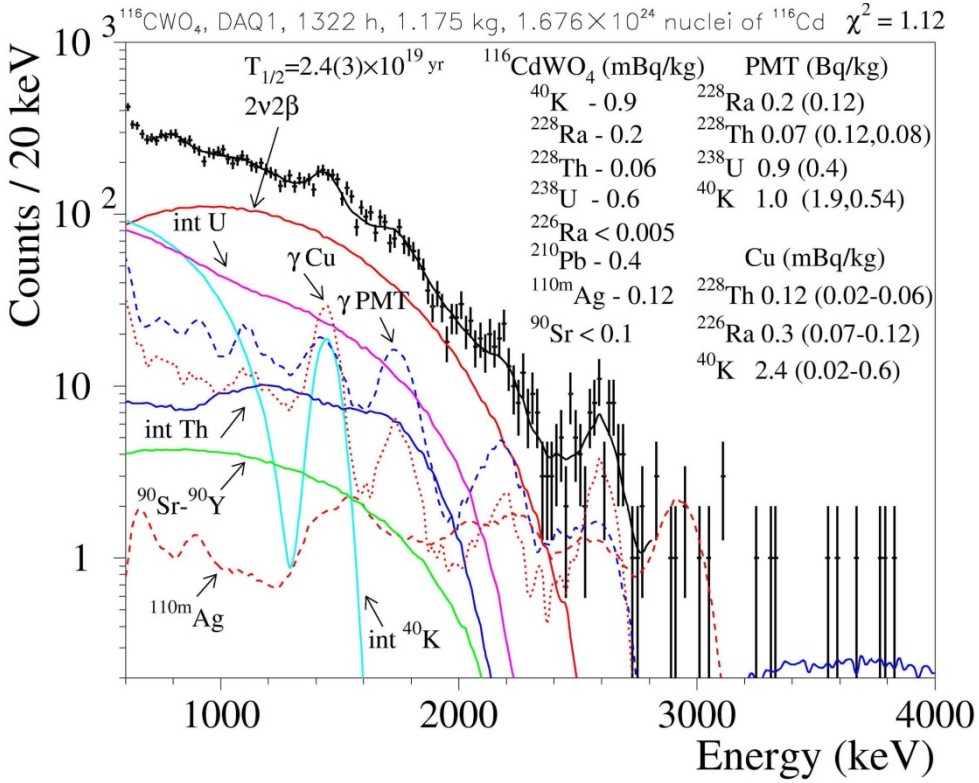
Radioactive contamination (mBq/kg)

^{232}Th	<0.08
^{228}Th	0.06
^{238}U	<0.5
^{226}Ra	<0.005
^{210}Po	<0.5
total α	1.9 – 2.7

1 2

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

Model of the background of $^{116}CdWO_4$



Radioactive contamination (mBq/kg)

^{40}K	<1
^{113}Cd	100(10)
^{113m}Cd	460(20)
^{110m}Ag	0.06 (cosmogenic)

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

A. Barabash *et al.*, to be presented at NPAE 2012, Kyiv, Ukraine

Radioactive contamination of $^{106,116}CdWO_4$ and $CdWO_4$

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	0.06(4)	-
^{113}Cd	182	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)

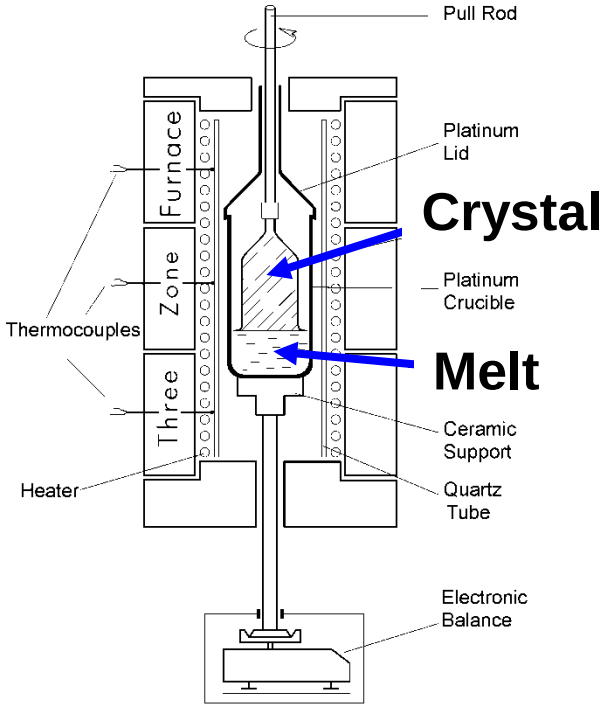
[1] P.Belli *et al.*, PRC 85 (2012) 044610

[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

Segregation of radioactive elements in $^{116}CdWO_4$



Segregation of impurities

$$K = C_S/C_L,$$

where K is segreagation coefficient, C_S is concentration of impurity in solid phase (crystal), C_L is concentration of impurity in liquid phase (melt),

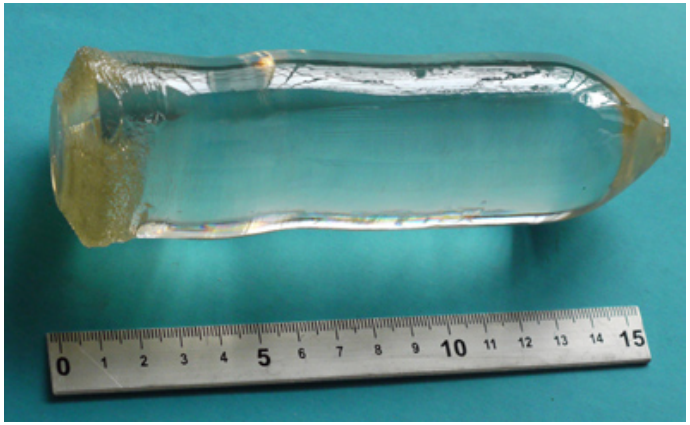
If $K < 1$, recrystallization could improve radiopurity of the crystal

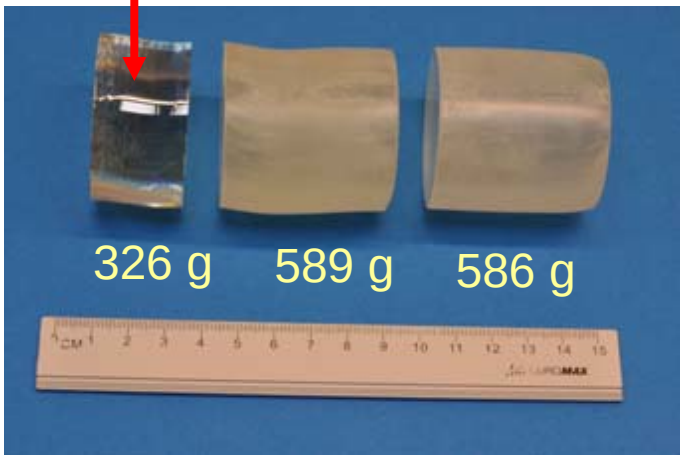
The scraps after the $^{116}CdWO_4$ crystal grown were measured by HPGe γ detector

Matthias Laubenstein (LNGS)

Radionuclide	Crystals	Scraps
^{228}Th	**0.06**	**10(2)**
^{226}Ra	**<0.005**	**64(4)**
^{40}K	**<1**	**27(11)**

Plans to test the 3rd $^{116}CdWO_4$ crystal





We are going to measure radioactive contamination of the 3rd sample in scintillation mode to estimate how concentration of Th, U, Ra, K depends on crystal growth

- **We hope to estimate how recrystallization could improve radioactive contamination of $CdWO_4$ crystal scintillators**
- **Then the $^{116}CdWO_4$ crystals can be re-crystallised**

Effect of crystal growth ?

At present we ascribe radioactive contamination of crystals mainly to contamination of raw materials (powder for crystal growth). However, there was no systematic study how crystal growth process effects radioactive contamination of crystals

- Ceramics have typically contamination a few orders of magnitude higher
- Crucible is in direct contact with the melt for crystal growth

Contamination of Ceramics vs $ZnWO_4$ crystals (mBq/kg)

Producer	Ceramics (^{228}Ac)	$ZnWO_4$ (^{228}Th)
ISMA, Kharkiv	$(<2 – 42)\times10^3$	0.002 – 0.005
NIIC, Novosibirsk	$<3\times10^3$	0.02

Producer	Ceramics (^{226}Ra)	$ZnWO_4$ (^{226}Ra)
ISMA, Kharkiv	$(<1 – 36)\times10^3$	0.002 – 0.02
NIIC, Novosibirsk	$(<0.4 – 2)\times10^3$	0.02

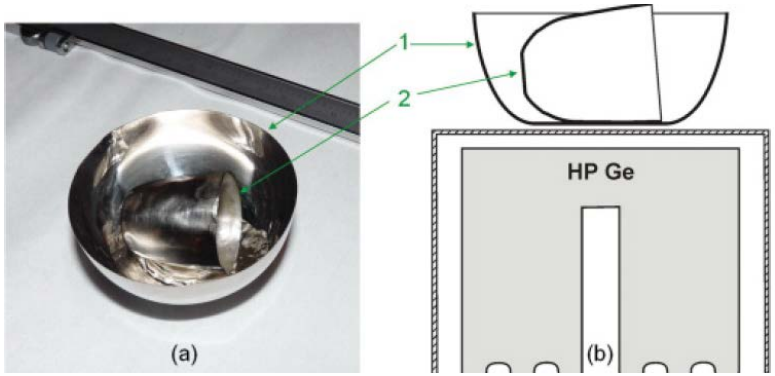
- **There is no correlation between radioactive contamination of ceramics and $ZnWO_4$ crystal scintillators**
- **We are going to grow $ZnWO_4$ crystals in the different conditions by using the same raw material**

P. Belli *et al.*, NIMA 626 (2011) 31

Contamination of platinum

- **Pt crucible is in direct contact with the melt for crystal growth**
- **One cannot grow high quality crystal in a crucible where another crystal was grown before (e.g., $CdWO_4$ after BGO)**

HPGe 468 cm^3, 42.5 g of Pt, 1815 h [1, 2]



Radioactive contamination of Pt (mBq/kg)

^{40}K	< 25
^{228}Th	< 7
^{226}Ra	< 3
^{192m}Ir (241 yr)*	= 40

* Q_β = 1460 keV, Q_{EC} = 1046 keV

- **More sensitive measurements are necessary**
- **One should study behavior of Th, Ra, K**

[1] P.Belli *et al.*, EPJA 47 (2011) 91

[2] P.Belli *et al.*, PRC 83 (2011) 034603

$ZnMoO_4$

Large $ZnMoO_4$ single crystals were developed for the first time in 2008

Idea of Stefano Pirro (during our discussion at NANP 2005 conference in Dubna, June 23)

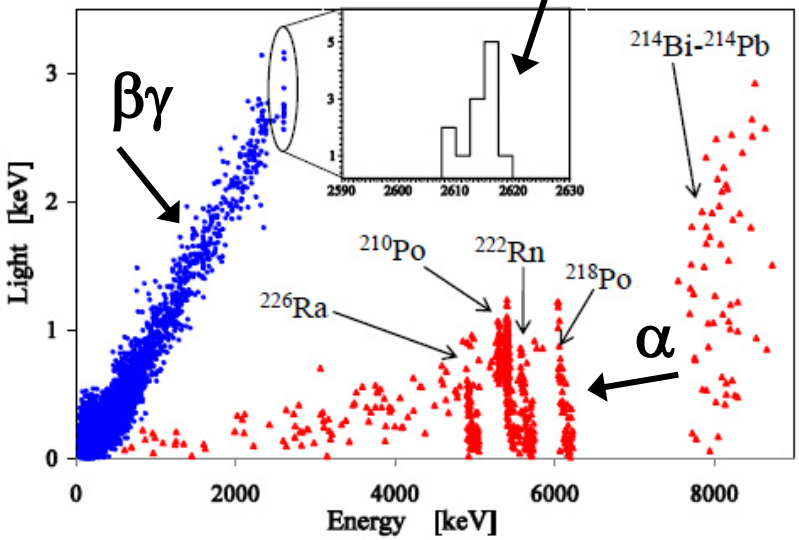
"Why nobody try to grow $ZnMoO_4$?"

2008 (IGP, Moscow, Russia) [1]

2009 (ISMA, Kharkov, Ukraine) [2,3]

2010 Low-Thermal-Gradient Czochralski (NIIC, Novosibirsk, Russia) [4]

FWHM = 5.6 keV at 2615 keV



[3] L. Gironi *et al.*, JINST 5 (2010) 11007

A high sensitivity 2b experiment can be realized with enriched $Zn^{100}MoO_4$ [5,6]

[1] LI. Ivleva *et al.*, Crystallography Reports, 2008, Vol. 53, No. 6, pp. 1087

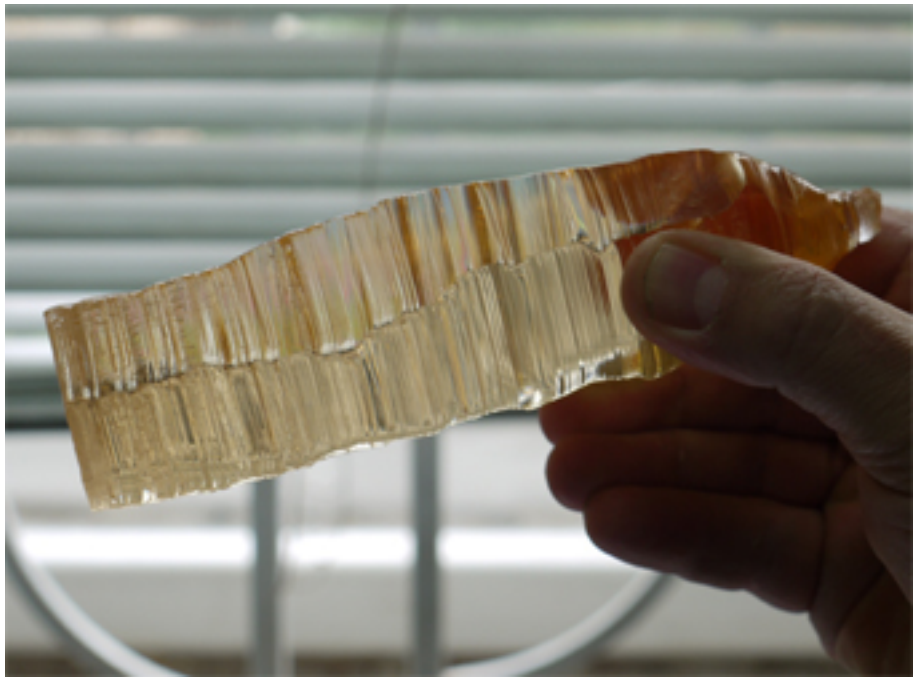
[2] L.L. Nagornaya, *et al.*, IEEE TNS 56 (2009) 2513

[4] J.W. Beeman *et al.*, J Low Temp Phys 167 (2012) 1021

[5] J.W. Beeman *et al.*, PLB 710 (2012) 318

[6] J.W. Beeman *et al.*, APP 35 (2012) 813

Large $ZnMoO_4$



2011 Low-Thermal-Gradient Czochralski
(NIIC, Novosibirsk, Russia)

R&D of $Zn^{100}MoO_4$ is in progress

- Radioactive contamination of ~1 kg of $^{100}MoO_3$ was measured by low-background HPGe detector at LNGS [1]

$^{100}MoO_3$ (mBq/kg)	
^{40}K	36
^{226}Ra	2
^{228}Th	1

- Requirements of $0\nu2\beta$ experiment to $Zn^{100}MoO_4$ crystals:

$Zn^{100}MoO_3$ (mBq/kg)	
^{40}K	<10 *)
^{226}Ra	<0.1 – 1
^{228}Th	<0.01 – 0.1
Total α activity	< 1 mBq/kg

*) $2\nu2\beta$ activity of ^{100}Mo in $Zn^{100}MoO_4$ is 8 mBq/kg

[1] P.Belli et al., NPA 846 (2010) 143

Conclusions I

- The next generation double β experiments call for large mass detectors (~100 - 1000 kg) with extremely characteristics: containing certain elements (isotopically enriched), very low (ideally 0) radioactive background, high energy resolution (< 1%), discrimination ability, long time operation (~ 10 yr)

- Cryogenic scintillating bolometeres are promising tools to search for $0\nu2\beta$ decay

- High quality $^{106}CdWO_4$ and $^{116}CdWO_4$ crystal scintillators were developed from enriched ^{106}Cd and ^{116}Cd

- R&D of $Zn^{100}MoO_4$ from enriched ^{100}Mo is in progress

Conclusions II

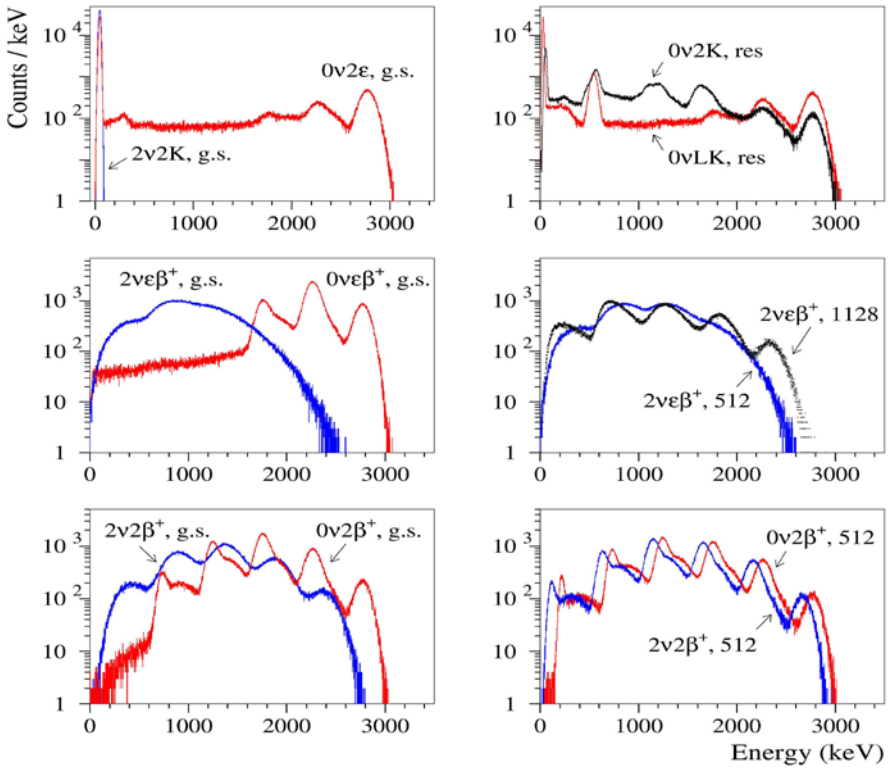
- Production of high quality low radioactive crystal scintillators from enriched isotopes for high sensitivity 2β decay experiments requires a special extended R&D: radiopurity is the most important issue

- Deep purification of initial materials looks the most important issue to be addressed
- Study of Th, U, Ra, K segregation in $CdWO_4$, $ZnMoO_4$ could allow to improve radiopurity by recrystallization
- Effect of crystal growing (contamination of ceramics & crucible) can be important at the μBq/kg level
- Knowledge of the "history" of initial materials to be used in enrichment process is important (^{113m}Cd in $^{106,116}CdWO_4$)
- Keep in mind cosmogenic activation (very dangerous ^{110m}Ag with $Q_\beta \approx 3010$ keV in $^{116}CdWO_4$ is observed with an activity of ~ 0.06 mBq/kg)

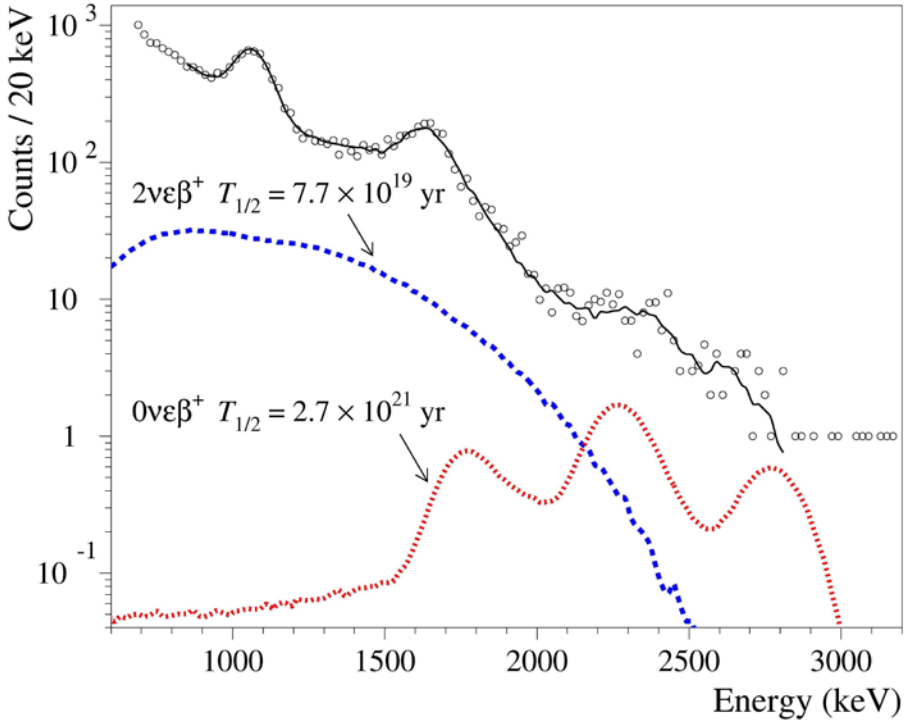
backup slides

Search for 2β decay of ^{106}Cd

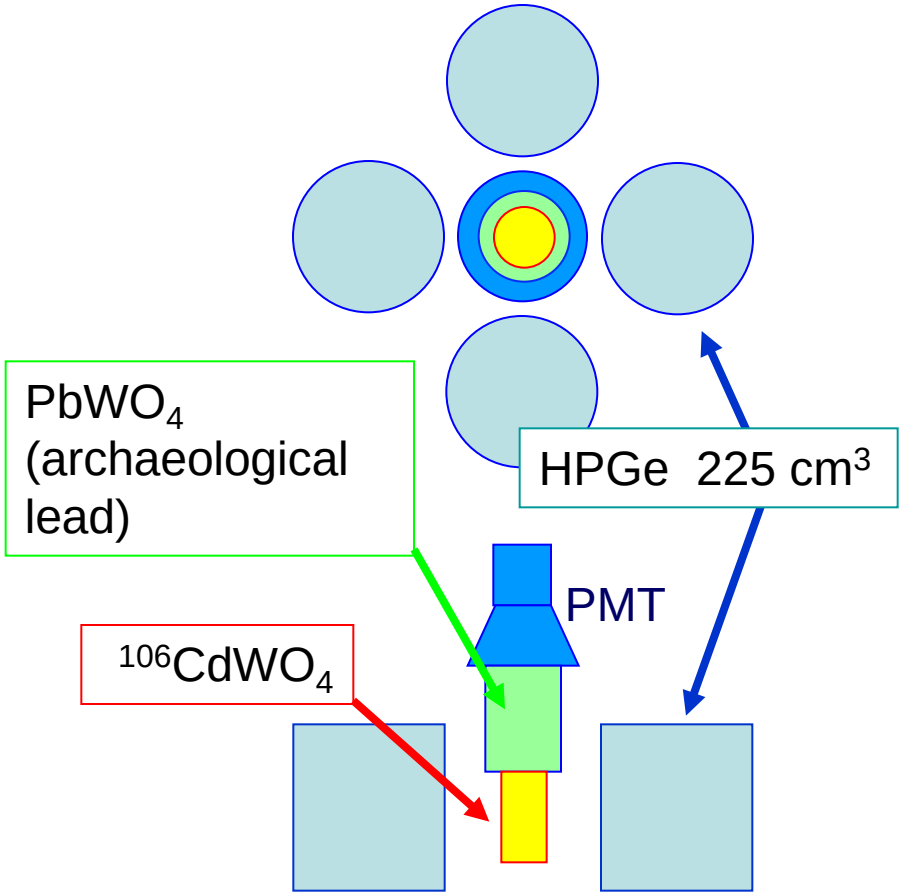
Response of the $^{106}CdWO_4$ detector to 2β processes in ^{106}Cd simulated by EGS4 and DECAY0



Fit of the experimental data to estimate $T_{1/2}$ limits on εβ+



next step: $^{106}CdWO_4$ in HPGe



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

Detection efficiency ~ 5–7%

Background expected to be several events during year

Sensitivity to $2\nu\ \varepsilon\beta^+$ and $2\beta^+$ in ^{106}Cd:

$T_{1/2} \sim 10^{20} - 10^{21}$ yr

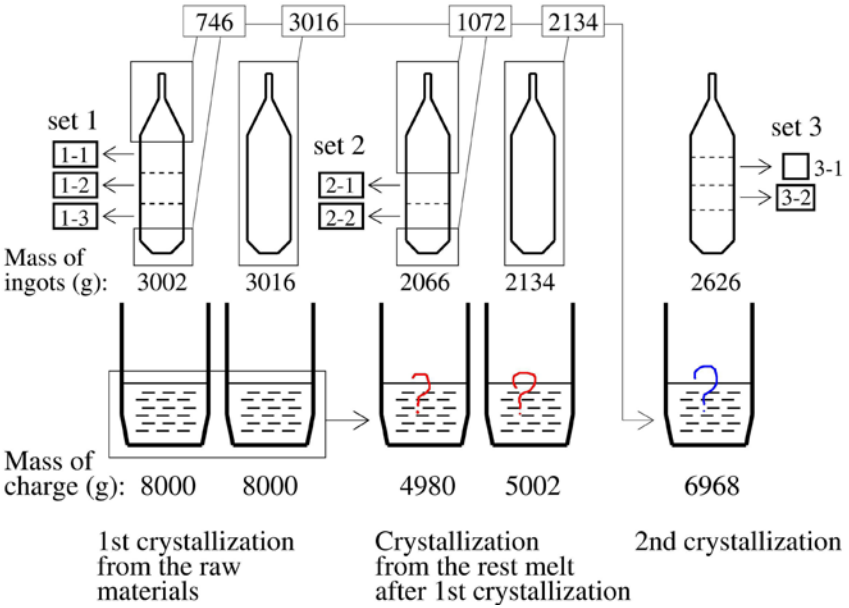
Theory: $2\nu 2K$ $10^{20} - 5\times10^{21}$ yr

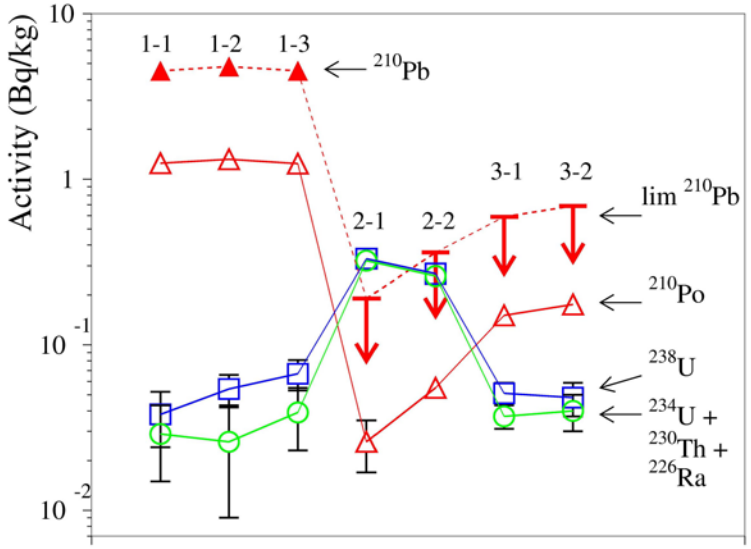
$2\nu\varepsilon\beta^+$ $8\times10^{20} - 4\times10^{22}$ yr

A possible further step:

Production of $^{106}CdWO_4$ from the ^{106}Cd depleted in ^{113}Cd to remove ^{113m}Cd

Segregation of Pb, Po, U in $CaWO_4$





F.A.Danevich et al., NIMA 631 (2011) 44