Development of radiopure cadmium tungstate crystal scintillators from enriched ^{106}Cd and ^{116}Cd to search for double β decay

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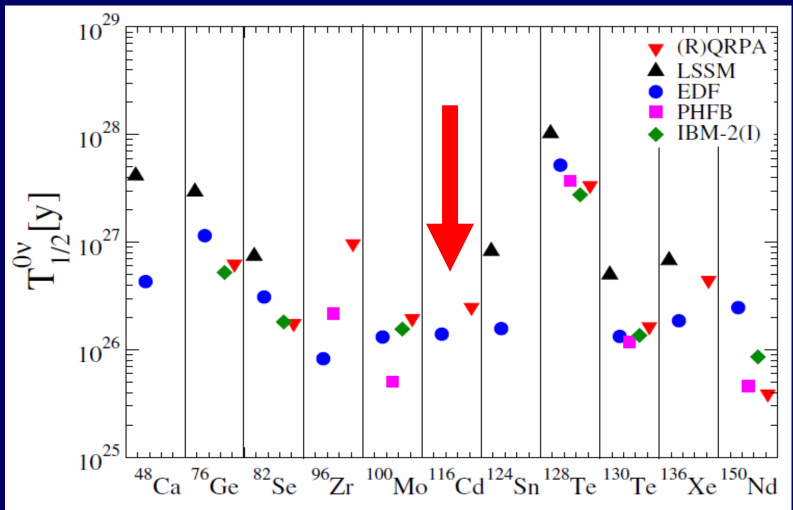
outline

- Motivation
- Development of $^{106}CdWO_4$ and $^{116}CdWO_4$
- Radioactive contamination
- Prospects

^{116}Cd is one of the most promising $0\nu2\beta$ nuclei

Motivation to study 2ε, $\varepsilon\beta^+$, $2\beta^+$ (^{106}Cd)

Theoretical calculations of $T_{1/2}$ for $\langle m_\nu \rangle$ = 0.05 eV [1]



One of the 3 nuclei with $Q_{2\beta}$ > 2615 keV

$Q_{2\beta}$ = 2813.50 ± 0.13 keV, δ = 7.5%

Test of right-handed weak current contribution

Half-lives for $0\nu\varepsilon\beta^+$ decay depend strongly on whether the decay is dominated by the mass mechanism or right-handed weak current [2]

Possibility of resonant $0\nu2\varepsilon$

$Q_{2\beta}$ = 2775.39 ± 0.10 keV, δ = 1.2%

[1] J.D.Vergados, H.Ejiri, F.Simkovic, Rep. Prog. Phys. 75 (2012) 106301

[2] M. Hirsch et al., Z. Phys. A 347 (1994) 151

Requirements to scintillators from enriched isotopes

(typically unclear for producers of scintillators)

- Maximal concentration of the isotope of interest
- High radiopurity (Th, Ra, U, K, rare earth elements)
- Minimal losses and contamination of enriched materials
- Recovery and purification of enriched materials from scraps in detectors production cycle
- Minimization of neutron & cosmogenic activation

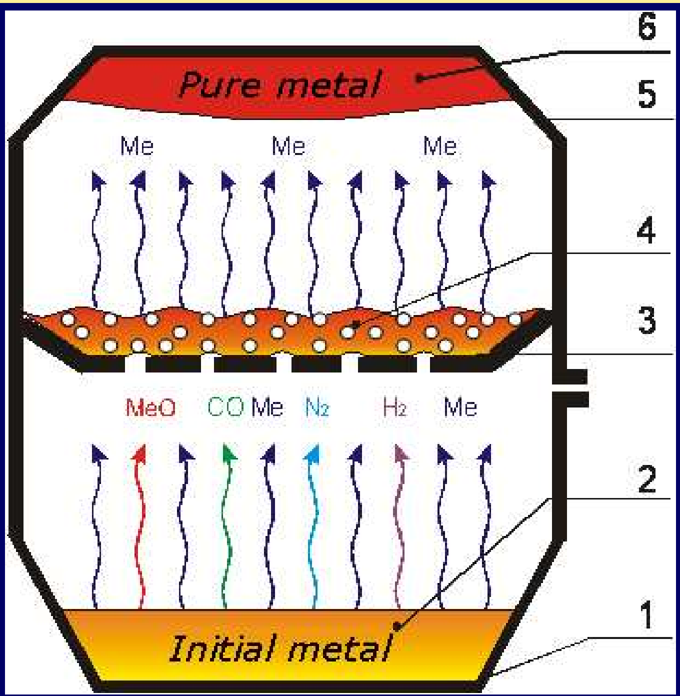
Of course, one also needs:

- High scintillation properties (energy resolution, light yield, transparency, reasonably fast scintillation decay)
- Large enough volume ~ 10^2 cm

Purification of ^{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

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**

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 $^{106,116}CdWO_4$ compounds

Joint Stock Company NeoChem, Moscow, Russia

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:

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

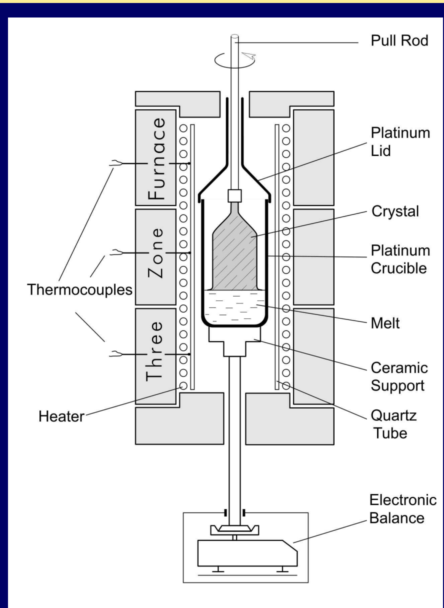
- All the operations were carried out by using quartz or polypropylene lab-ware, materials with low level of radioactive contaminations
- Reagents of high purity grade (concentration of any metal less than 0.01 ppm)
- Water, acids and ammonia were additionally distilled by laminar evaporation in quartz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

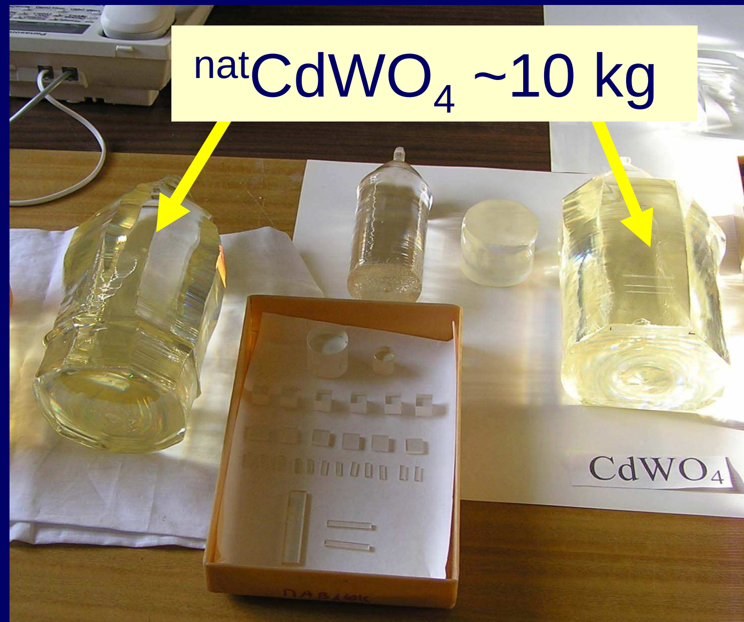
P. Belli *et al.*, NIMA 615 (2010) 301
A. Barabash *et al.*, JINST 6 (2011) P08011

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

Nikolaev Institute of Inorganic Chemistry, Novosibirsk, Russia

Low-Thermal-Gradient Czochralski (LTG-C)





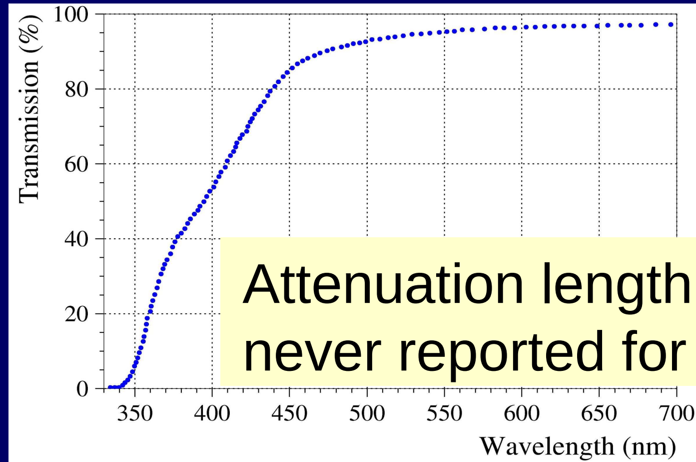
	standard	LTG-C
Output	25-30%	up to 90%
Quality		typically higher
Radiopurity		expected better
Loses of powder	2-3%	<0.3%

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

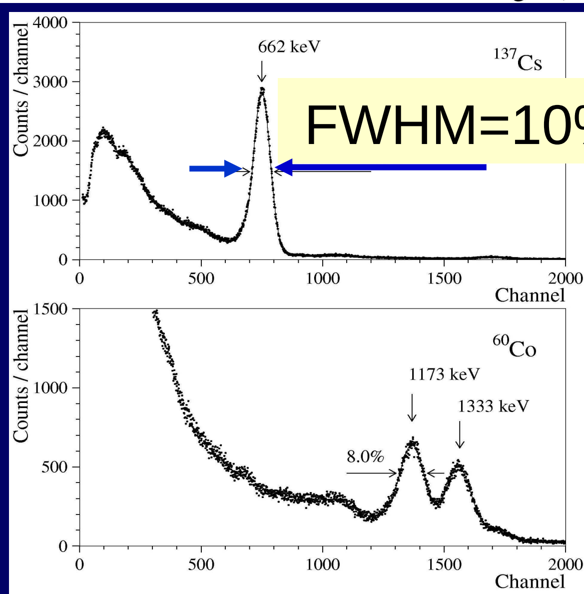
$^{106}CdWO_4$ scintillator

$^{106}CdWO_4$ boule 231 g (87.2%)

scintillator 215 g, 66% of ^{106}Cd



Attenuation length 60 cm never reported for $CdWO_4$



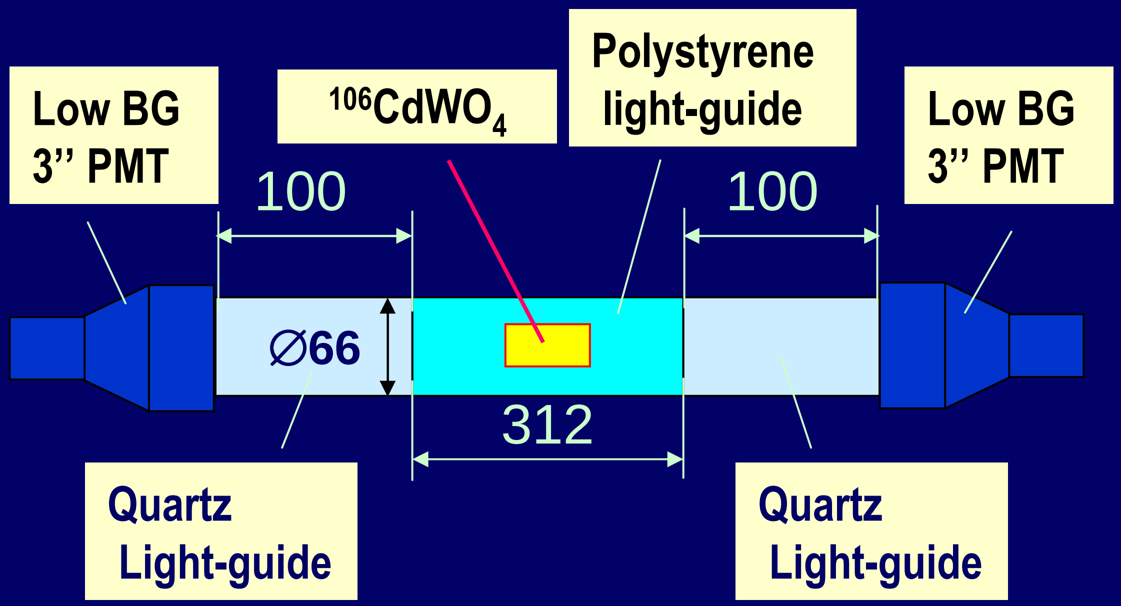
FWHM=10%

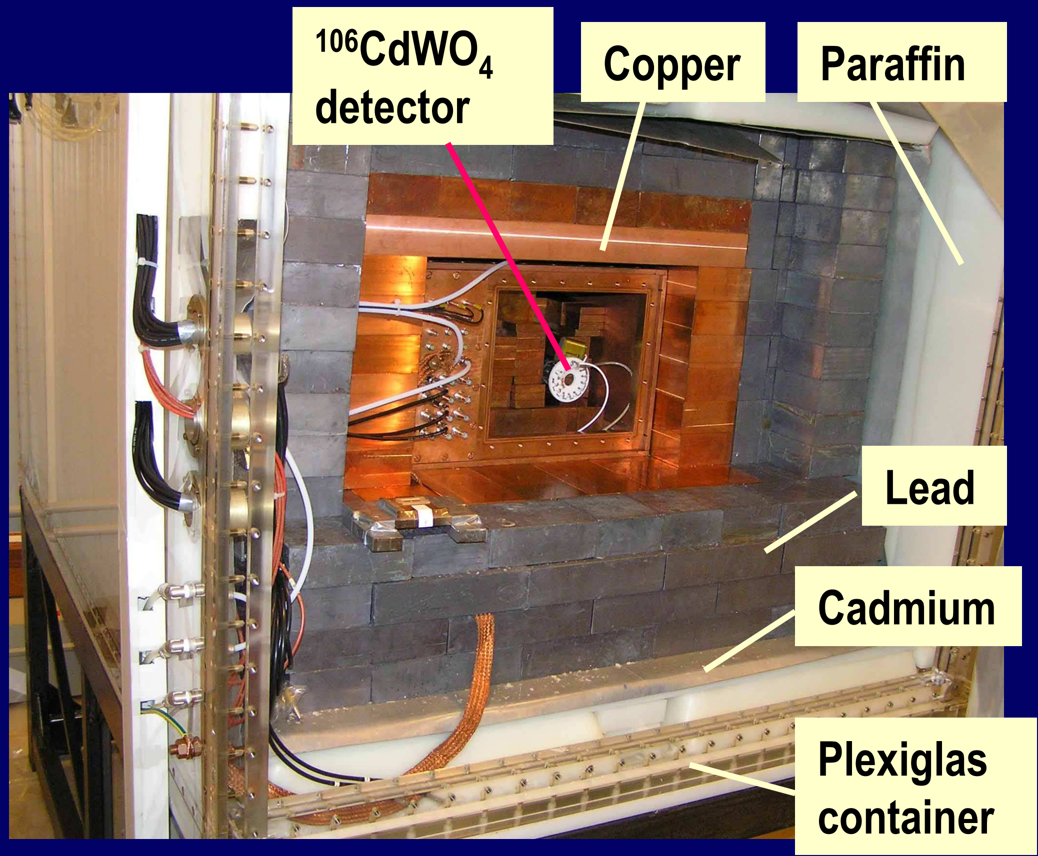
Excellent optical and scintillation properties thanks to special R&D to purify raw materials and Low-thermal-gradient Czochralski technique to grow the crystal

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

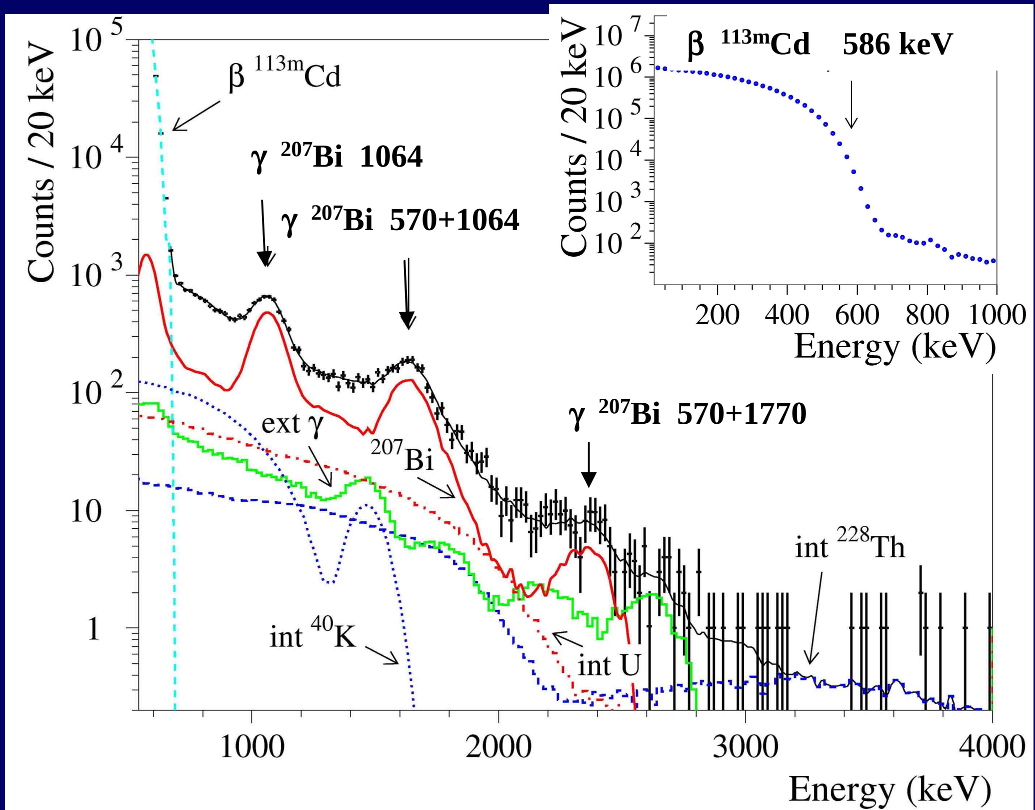
The total losses of ^{106}Cd = 2.3%

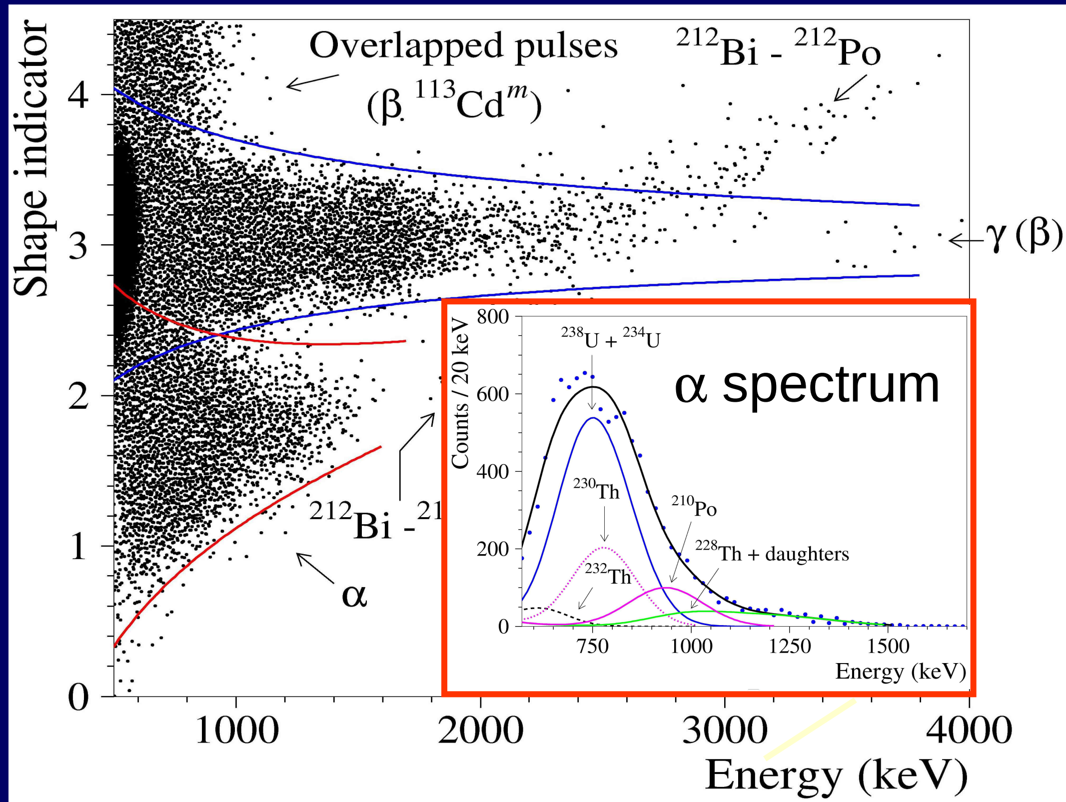
Low background $^{106}CdWO_4$ detector DAMA R&D at LNGS





$^{106}CdWO_4$: model of background, pulse-shape discrimination





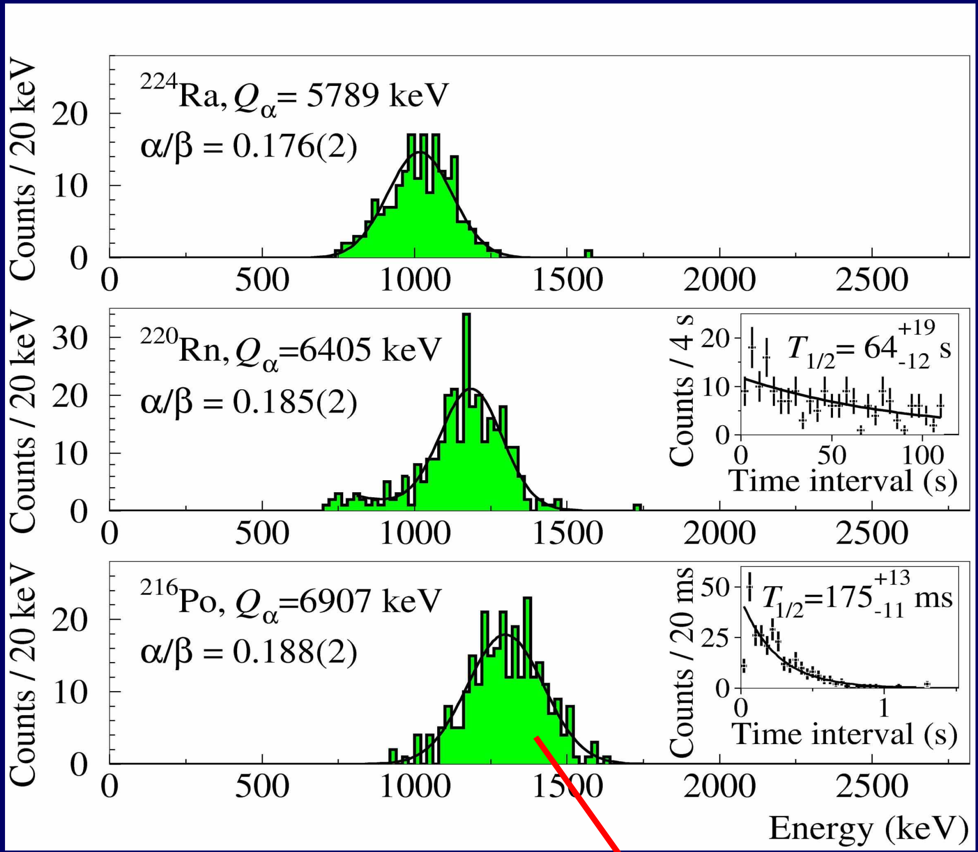
^{113m}Cd = 116 Bq/kg

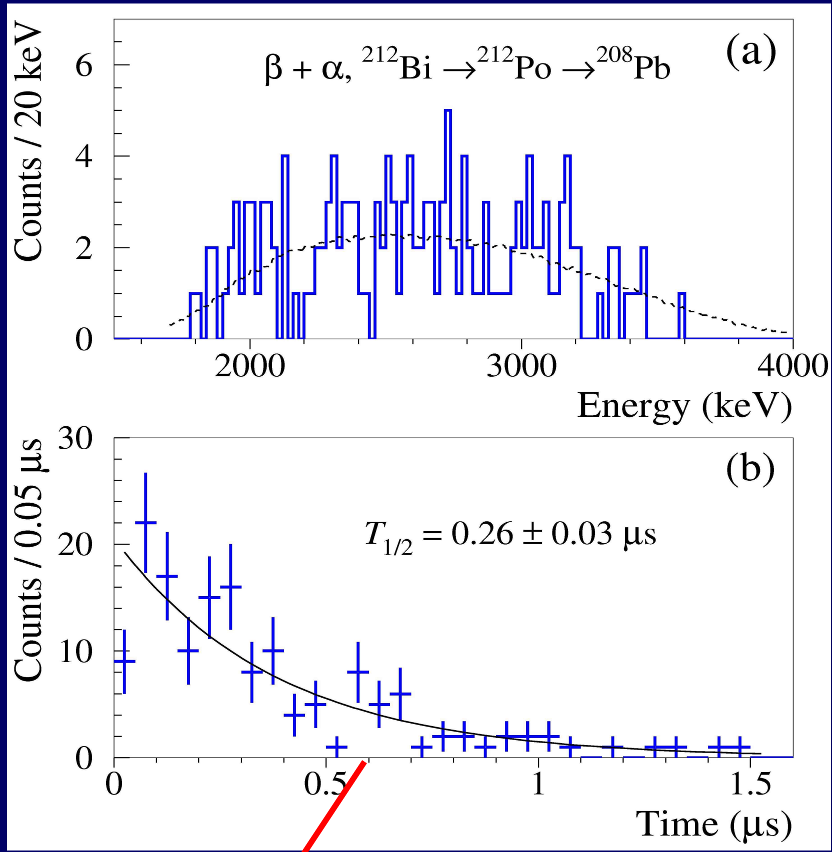
^{207}Bi = 0.06 mBq/cm^2 (on surface)

^{232}Th	<0.07
^{238}U	<0.6
^{210}Po	<0.2
Total α	= 2.1

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

$^{106}CdWO_4$: time-amplitude analysis



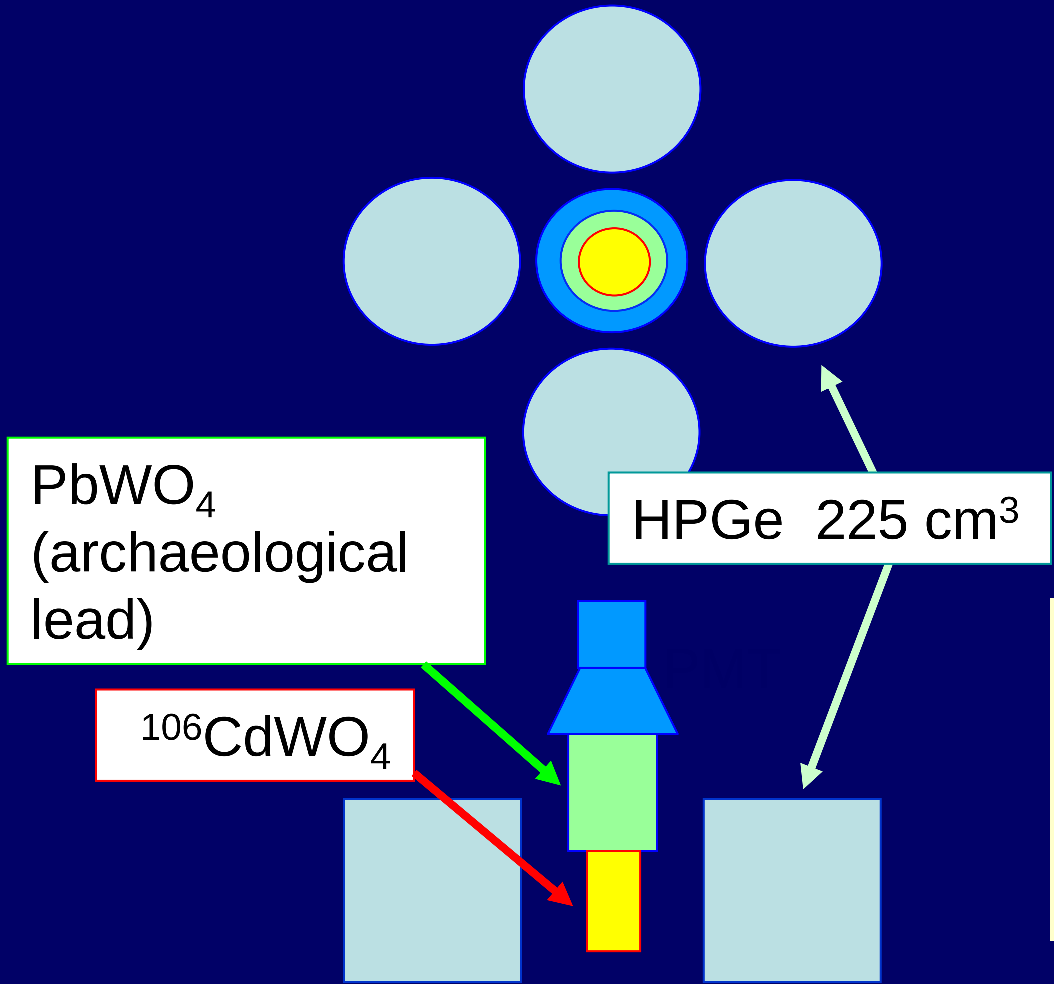


^{228}Th	= 0.042(2)	= 0.051(4)
^{226}Ra		= 0.012(3)

mBq/kg

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

$^{106}CdWO_4$ in coincidence with 4 HPGe detectors



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

Detection efficiency ~ 5–7% ($\varepsilon\beta^+$ and $2\beta^+$)

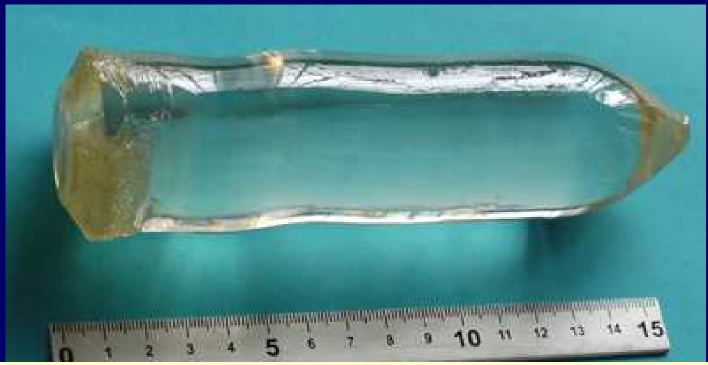
Background expected to be several events per year

Sensitivity to $2\nu\ \varepsilon\beta^+$ and $2\beta^+$ in ^{106}Cd: $T_{1/2}$ ~ 10^{20} - 10^{21} yr

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

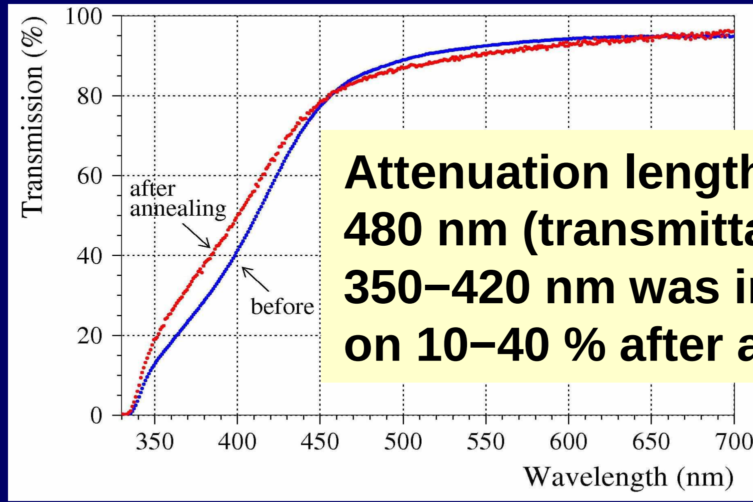
$2\nu\varepsilon\beta^+$ 8×10^{20} - 4×10^{22} yr

$^{116}CdWO_4$ scintillators

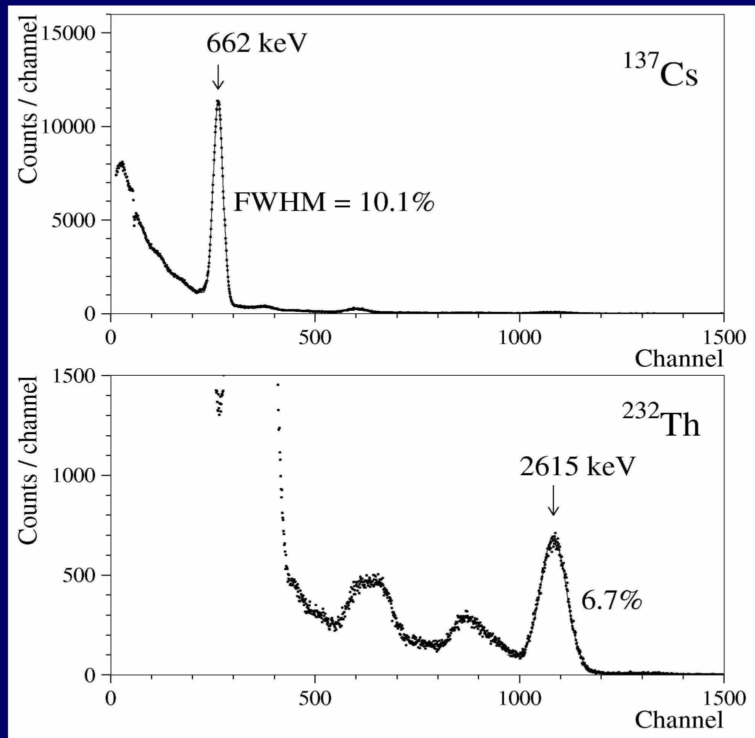


$^{116}CdWO_4$ crystal boule 1868 g
(87% of initial charge)

Scintillation elements
(82% of ^{116}Cd)

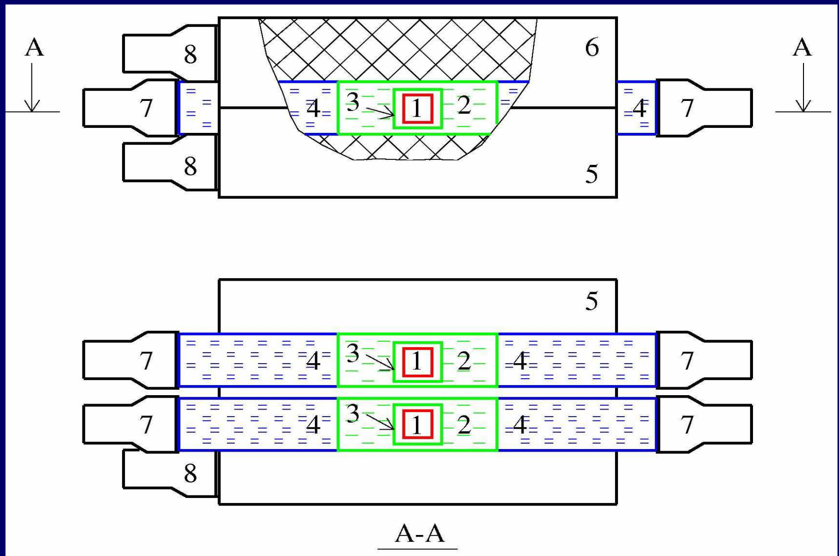


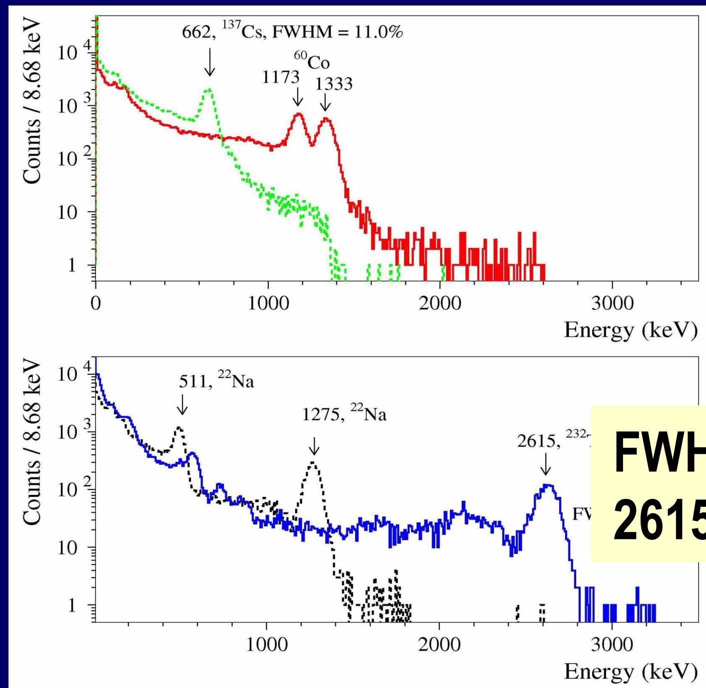
Attenuation length ≈30 cm @ 480 nm (transmittance at 350−420 nm was improved on 10−40 % after annealing)



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

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

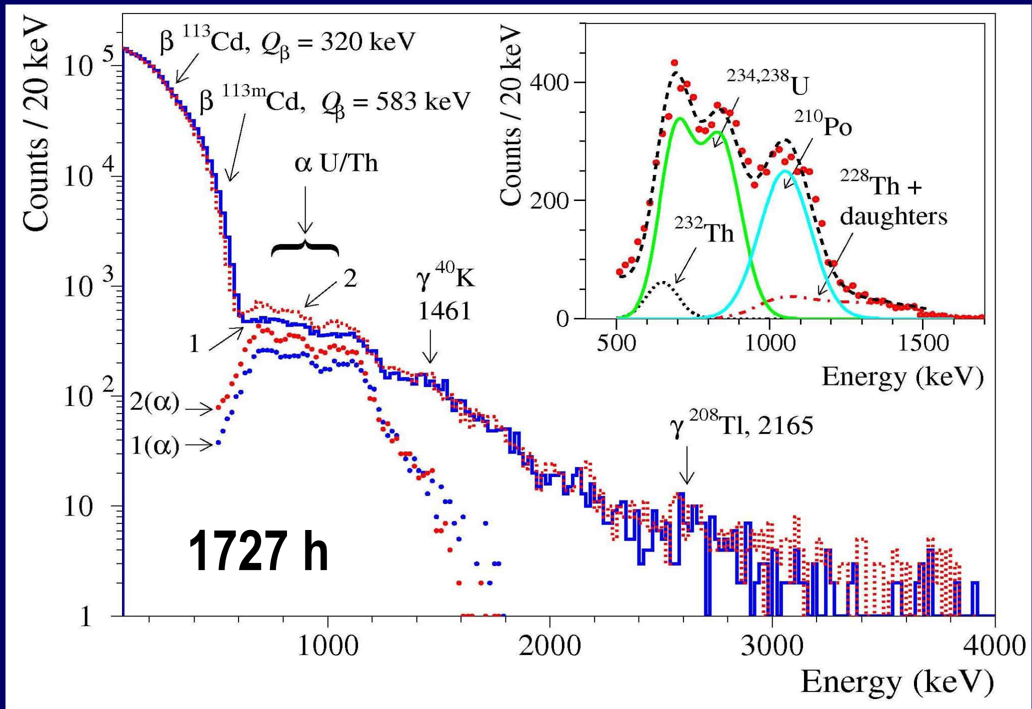


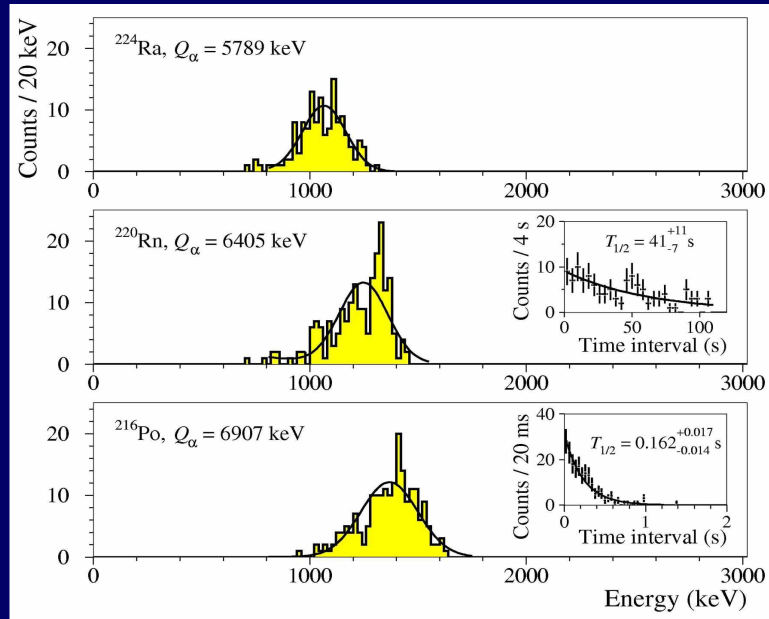


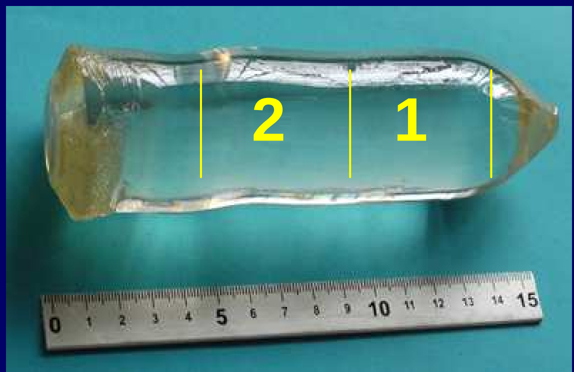
FWHM=4.5% at 2615 keV

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

Pulse-shape and t-A analyses







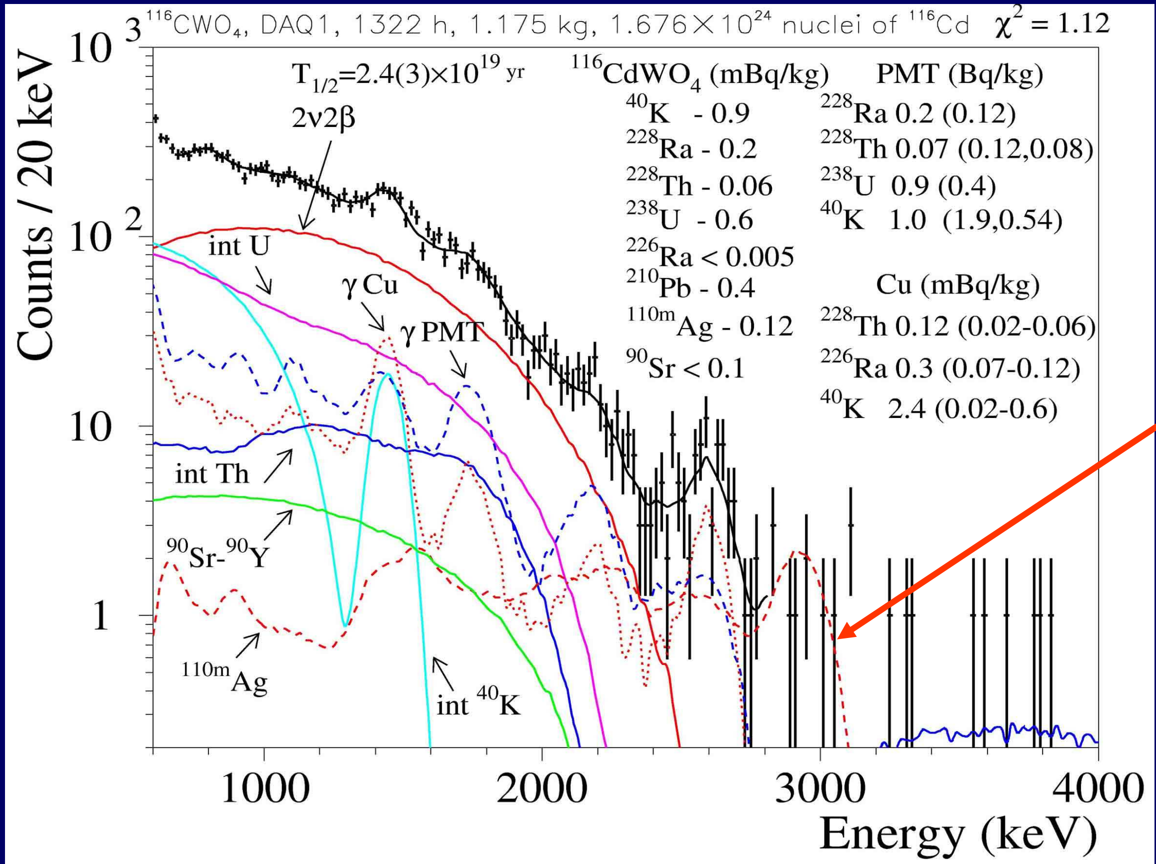
Radioactive contamination (mBq/kg)

^{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 background, cosmogenic ^{110m}Ag, $2\nu2\beta$ radioactive ^{116}Cd



Radioactive contamination (mBq/kg)

^{40}K	<1	
^{113}Cd	100(10)	
^{113m}Cd	460(20)	
^{110m}Ag	0.12(4)	(cosmogenic)

~ 1 mBq/kg of ^{116}Cd ($2\nu2\beta$)

Expected sensitivity of a 5 yr experiment (assume 2 – 5 times suppression of background after recrystallization of the crystals):

$T_{1/2}$ ~ (0.5–1.5)×10^{24} yr
$\langle m_\nu \rangle$ ~ (0.4–1.4) eV

A.S. Barabash *et al.*, JINST 6 (2011) P08011
A.S. Barabash *et al.*, Proc. NPAE 2012, Kyiv, Ukraine, 2013, p. 353
D.V. Poda et al., Radiat. Meas., DOI 10.1016/j.radmeas.2013.02.017

Comparison 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	= 0.12(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

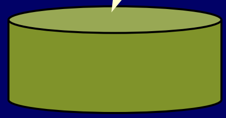
[2] A. Barabash *et al.*, JINST 6 (2011) P08011

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

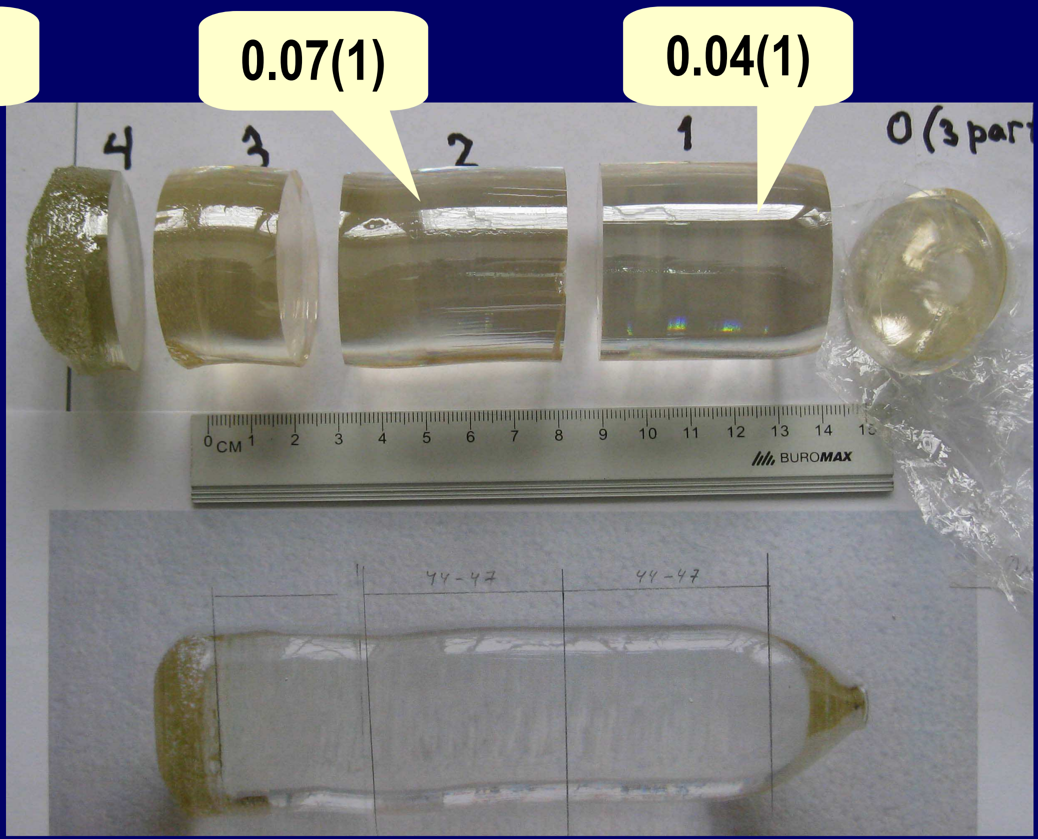
[4] P. Belli *et al.*, Phys. Rev. C 76 (2007) 064603

Possibility to improve radiopurity by recrystallization

Activity of ^{228}Th



rest of the melt



	Crystal	Rest of melt
^{226}Ra	<0.005	64(4)
^{40}K	<1	27(11)

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

Poda et al., Radiat. Meas., DOI 10.1016/j.radmeas.2013.02.017

Conclusions

- Production of crystal scintillators from enriched isotopes for high sensitivity 2β decay experiments requires an extended R&D

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

- Deep purification of initial materials is the most important issue
- Knowledge of the “history” and control of the initial materials to be used in enrichment process is important (^{113m}Cd in $^{106,116}CdWO_4$)
- Cosmogenic activation should be kept in mind (^{110m}Ag with $Q_\beta \approx 3010$ keV was observed in $^{116}CdWO_4$ with an activity of ~ 0.1 mBq/kg)
- Recrystallization should improve contamination of $CdWO_4$ by Th, U, Ra, K thanks to very low segregation of these elements

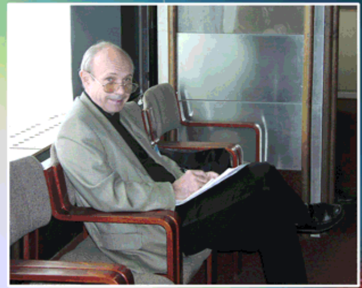
- Experiments to search for 2β decay of ^{106}Cd and ^{116}Cd by using the developed crystal scintillators are in progress at the LNGS

RPSCINT 2013

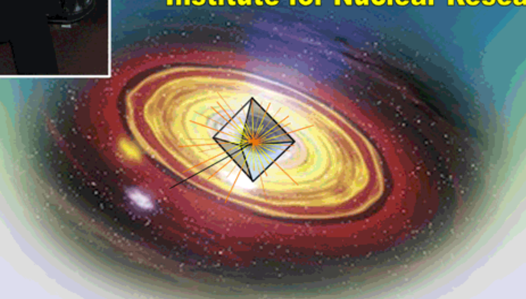
International Workshop on Radiopure Scintillators

September 17-20, 2013

National Academy of Sciences of Ukraine
Institute for Nuclear Research, Kyiv, Ukraine



The workshop is dedicated to the 70-th anniversary of Yuri Zdesenko



TOPICS

- radiopure scintillators in nuclear and astroparticle physics
- requirements of low-count rate experiments regarding radiopurity and scintillation properties
- radioactive contamination of scintillation materials
- selection and screening of input materials
- instruments and methods to test radioactive contamination of materials and scintillators
- purification of materials and preparation of raw compounds
- crystal growing, annealing and handling
- test of scintillators including scintillation, optical, luminescence low-background and low-temperature measurements
- search for and development of new scintillating materials

International Scientific Committee:

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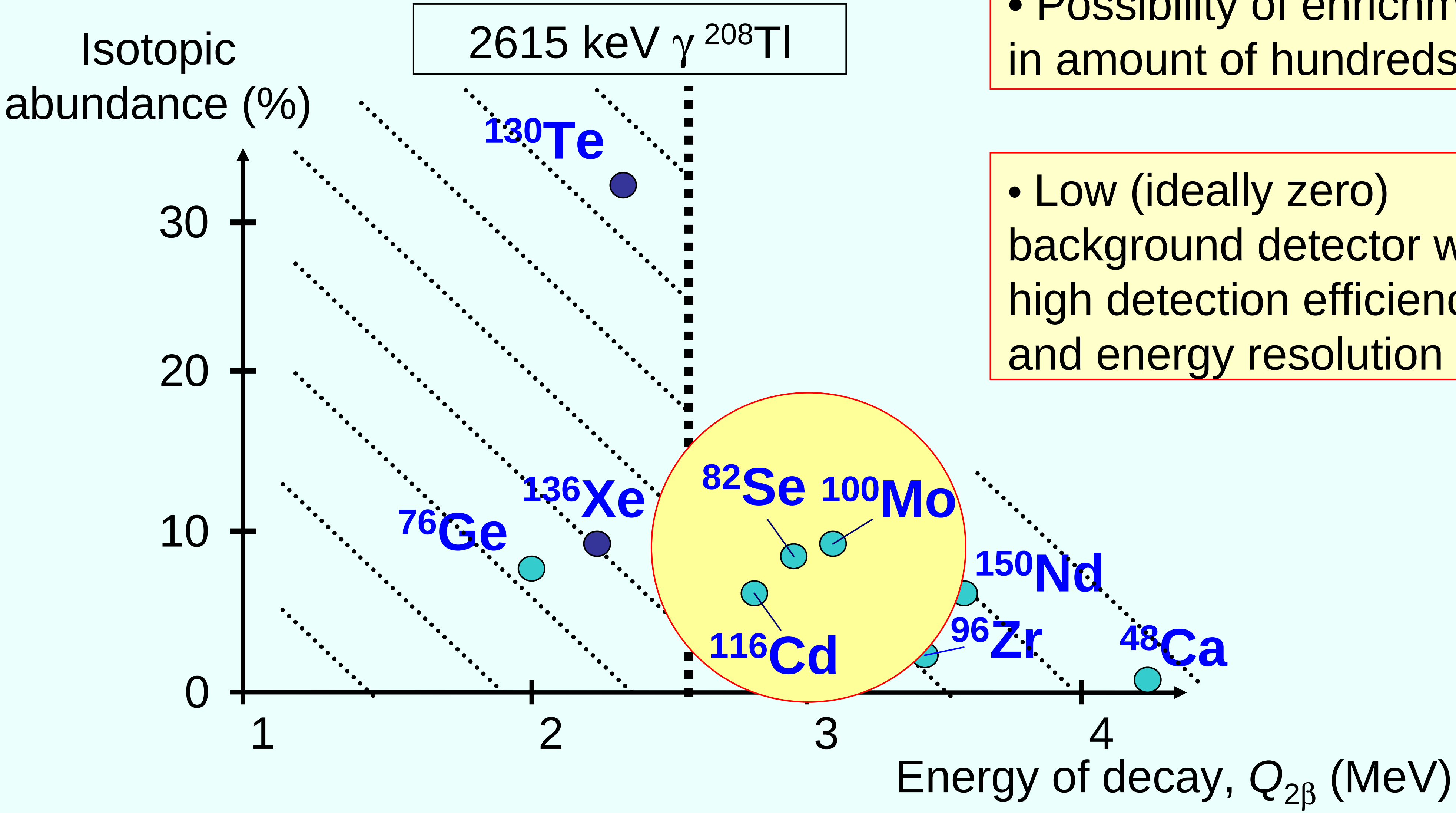
Back up slides

Most “promising” 2β nuclei

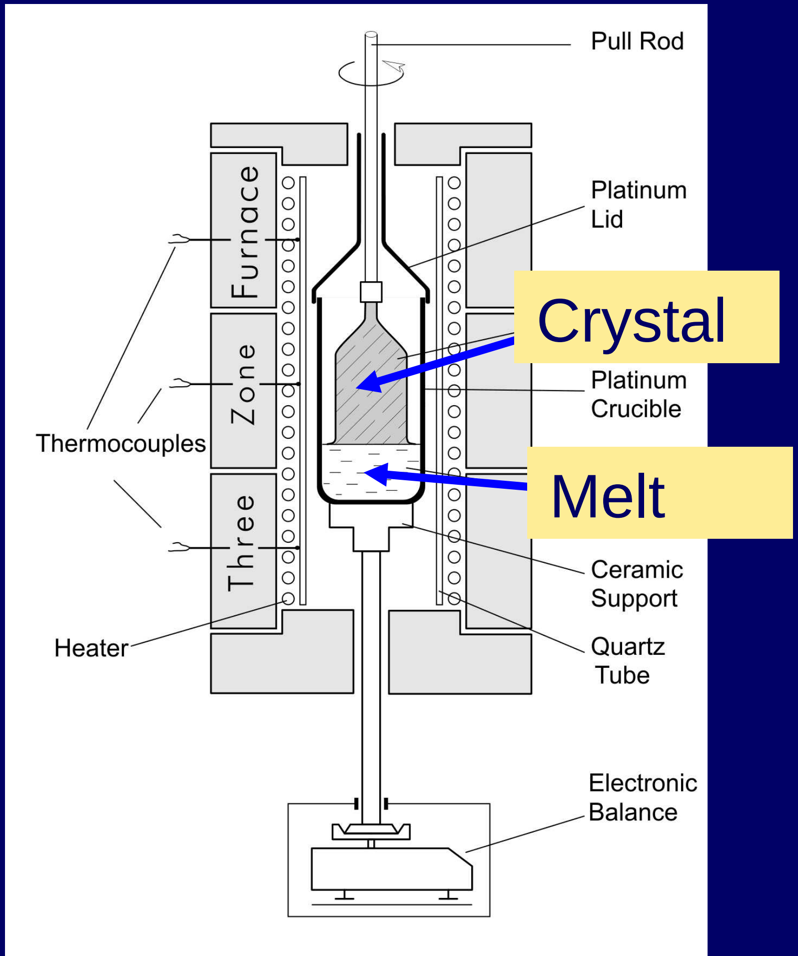
from the point of view of experiment:



- Possibility of enrichment in amount of hundreds kg

- Low (ideally zero) background detector with high detection efficiency and energy resolution

Segregation of impurities during crystal growth



Segregation of impurities

$$K = C_S/C_L,$$

where K is segregation 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 can reduce contamination of crystal