Improvement of the radiopurity level of $^{116}CdWO_4$ and $ZnWO_4$ crystal scintillators by recrystallization

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- Low background measurements
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Introduction

The radiopurity level of the crystal scintillators plays a crucial role to improve sensitivity of a double beta decay experiments.

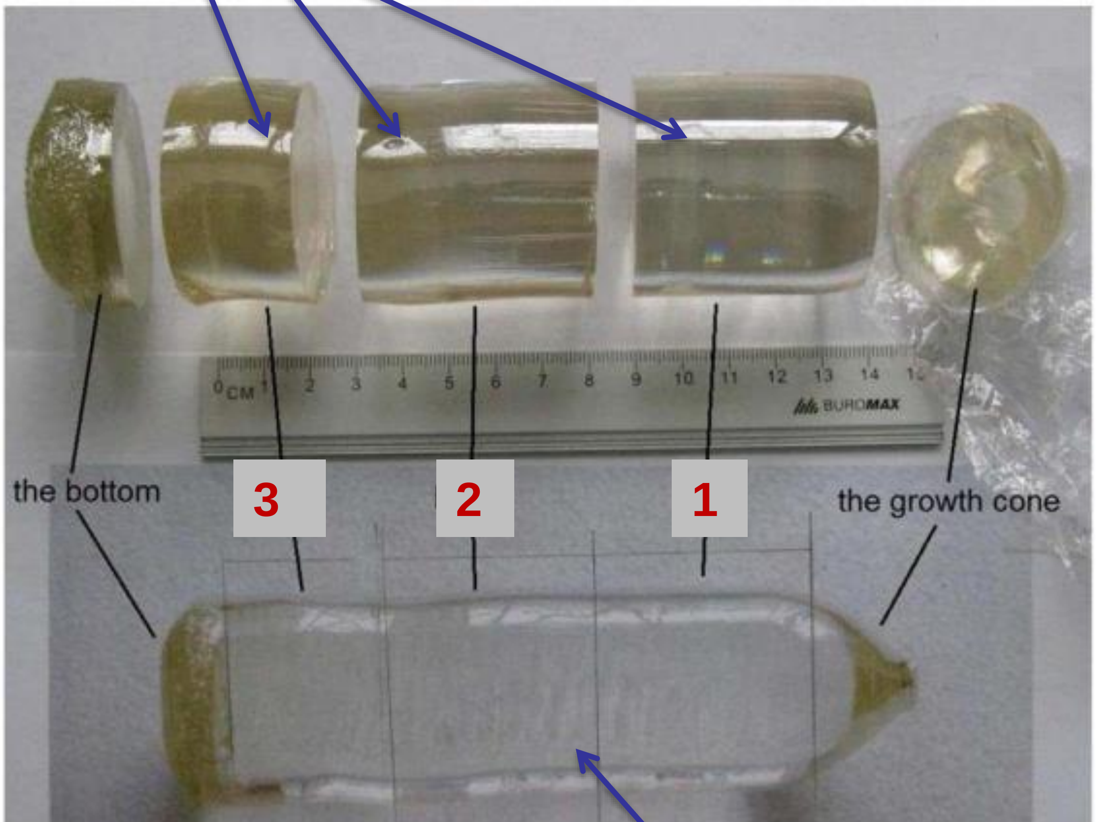
Crystal scintillators $^{116}CdWO_4$ and $ZnWO_4$:

- **Excellent scintillation and optical properties;**
- **Low radioactive contamination;**
- **Presence of ^{116}Cd($Q_{2\beta}$=2813 keV), ^{180}W and ^{186}W 2β candidates (^{180}W is of especial interest thanks to a possibility of resonant neutrinoless double electron capture [1]);**
- **Used in double beta decay experiments ($ZnWO_4$ [2] and $^{116}CdWO_4$ [3]);**
- **$CaMoO_4$, $CdWO_4$, $ZnWO_4$, $ZnMoO_4$, $LiMoO_4$, ZnSe promising scintillators for double β experiments.**

[1] NPA 859 (2011) 140
[2] J. Phys. G 38(2011)115107
[3] PRC 68(2003)035501, J. of Phys.:C.S.718(2016)062009

$^{116}CdWO_4$ crystal scintillator before recrystallization

Crystal samples



- deep purified materials;
- minimal loss of expensive isotopically enriched material;
- high optical quality thanks to the low-thermal-gradient Czochralski growth technique.

Boule of $^{116}CdWO_4$ crystal enriched in ^{116}Cd to 82% (mass=1.162kg)

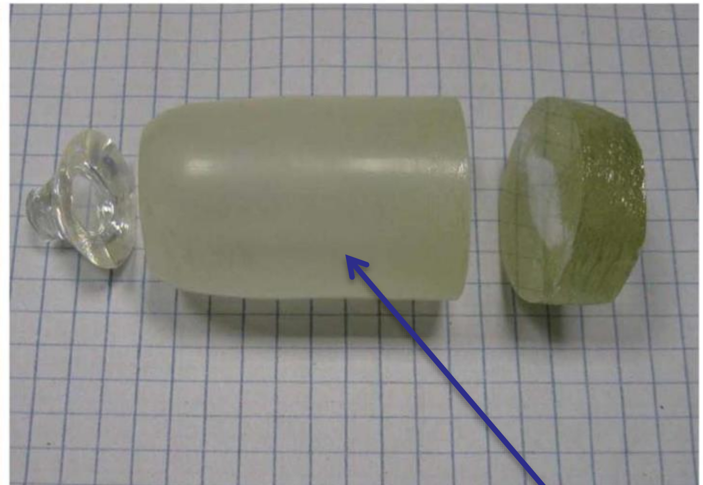
Radioactive contamination of the $^{116}CdWO_4$ crystals

$^{116}CdWO_4$ sample, mass	Radioactive contamination (mBq/kg)				Method
	^{40}K	^{226}Ra	^{228}Ra	^{228}Th	
No.1, 586g	≤0.9	≤0.005		0.02(1)	Scintillation counting
No.2, 589g	≤0.9	≤0.005		0.04(1)	Scintillation counting
No.3, 326g	≤1.2	≤0.1	≤1.3	0.10(1)	Scintillation counting
Rest of the melt after the crystal growth, 264g	27(11)	64(4)	9(2)	10(2)	HPGe γ spectrometry

Segregation of the radioactive elements in $CdWO_4$ crystal growth process?

Can we improve the radiopurity level of the $^{116}CdWO_4$ crystals by re-crystallization?

$^{116}CdWO_4$ crystal scintillator after recrystallization



Boule of enriched ^{116}**$CdWO_4$** crystal

Mass = 286 g (88% of sample before recrystallization)

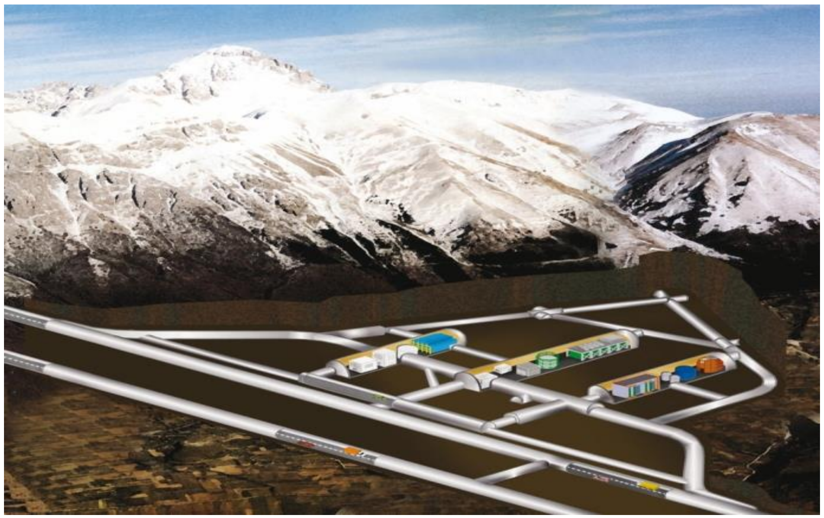
Sample of ^{116}**$CdWO_4$** crystal

Mass = 195 g (60% of sample before recrystallization)
The side surface of the sample was made opaque by grinding paper.

Recrystallized by the low-thermal-gradient Czochralski technique in a platinum crucible with 99.93% Pt purity level

Low background measurements

Laboratori Nazionali del Gran Sasso, Italy (3600 m w.e)



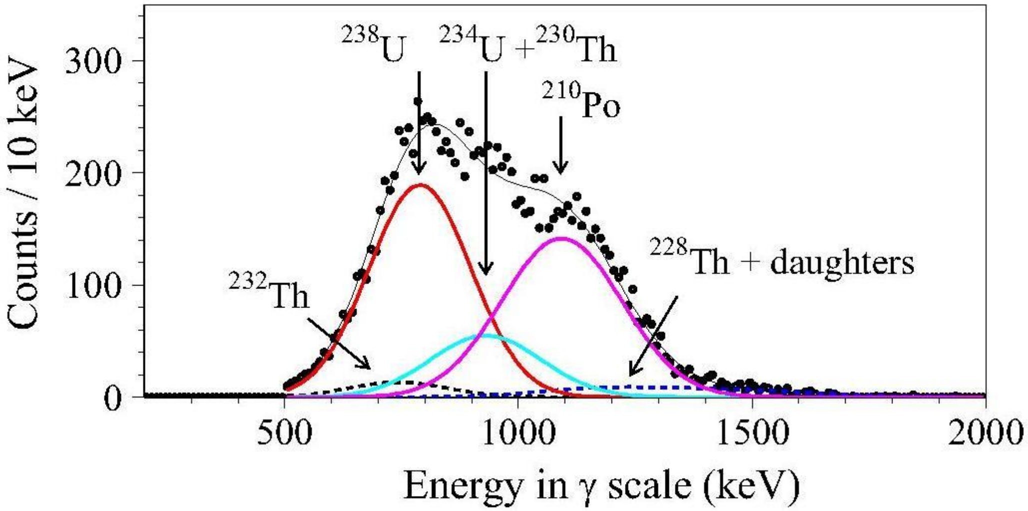
The **$^{116}CdWO_4$** crystal scintillator **after the recrystallization** was installed inside a **cavity ∅47×59mm** of a **polystyrene light guide ∅66×312mm** and viewed by a low-radioactive 3 in PMT through a high purity **quartz light guide ∅66×100mm**.

The passive shield of the DAMA/Crys set-up is made of high purity materials :

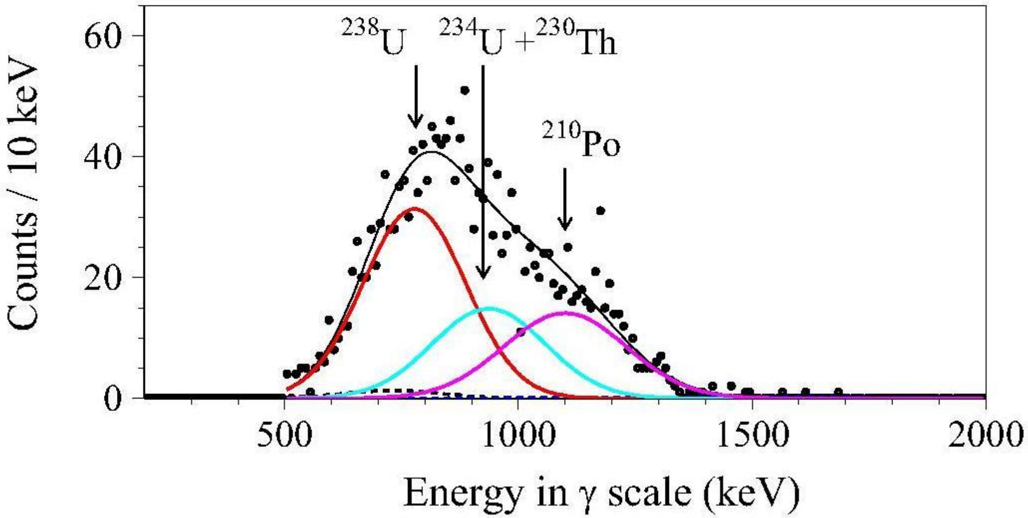
- copper (11cm)
- lead (10cm)
- cadmium (2mm)
- polyethylene (10cm)

Energy spectrum of α events

Selected by the pulse-shape discrimination



Before recrystallization
2394 hours



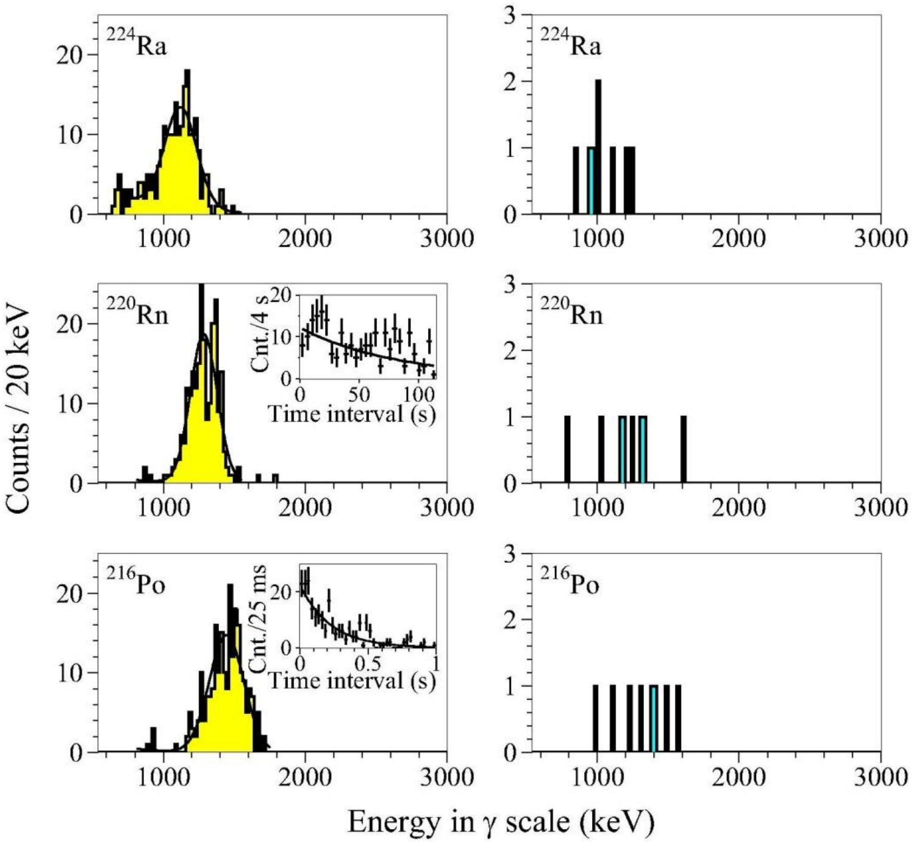
After recrystallization
1623 hours

Decrease factor ≈3

Radioactive contamination of the $^{116}CdWO_4$ crystal before and after recrystallization

recrystallization

before after



2394 hours 1623 hours

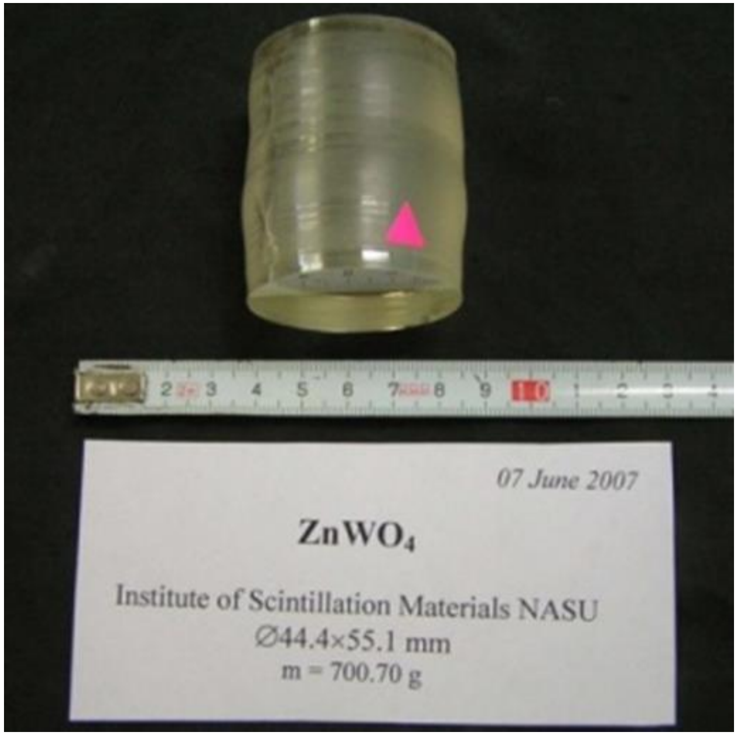
Alpha peaks ^{224}Ra, ^{220}Rn and ^{216}Po selected by the time-amplitude analysis

Chain	Nuclide	Before	After
^{232}Th	^{232}Th	0.13(7)	0.03(2)
	^{228}Th	0.10(1)	0.010(3)
^{238}U	^{238}U	1.8(2)	0.8(2)
	^{226}Ra	≤0.1	≤0.015
	^{234}U+^{230}Th	0.6(2)	0.4(1)
	^{230}Po	1.6(2)	0.4(1)
Total α		**4.44(4)**	**1.62(4)**

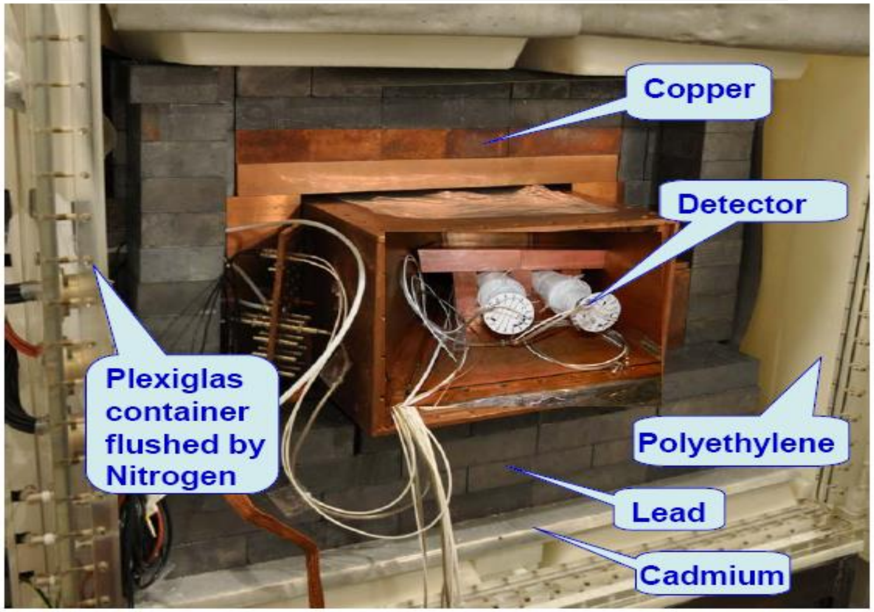
The specific activity of ^{228}Th after recrystallization decreased by ≈10 times to the level of 0.010(3) mBq/kg, which is the result of the strong segregation of thorium in the $CdWO_4$ crystal growth process.

$ZnWO_4$ crystal scintillators before recrystallization

Growth Czochralski technique, ISMA, Kharkiv, Ukraine



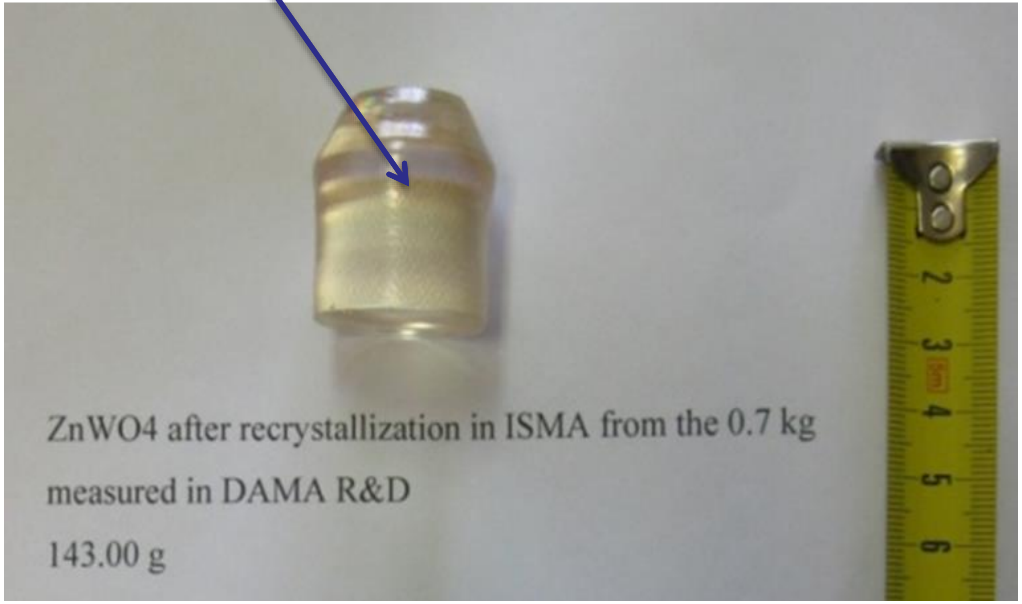
DAMA/R&D set-up, 994 hours



$ZnWO_4$ crystal scintillators after recrystallization

Sample of **$ZnWO_4$** crystal after recrystallization

Total α activity is **0.47 (7) mBq/kg** [1]



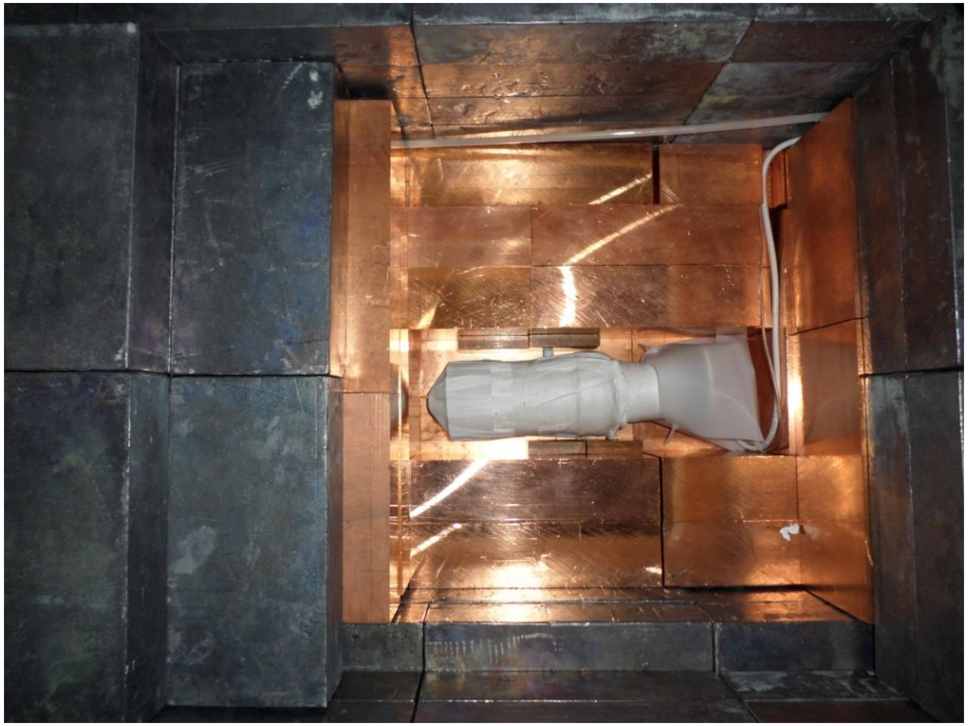
Mass = 143 g (20% of sample before recrystallization)

The side surface (~0.4 mm) layer of crystal was removed by fine grinding paper to check presence of radioactive contamination in external (surface) layer of the crystal.

[1] NIMA 626(2011)031

Low background measurements

DAMA/Crys set-up, 1445.4 hours



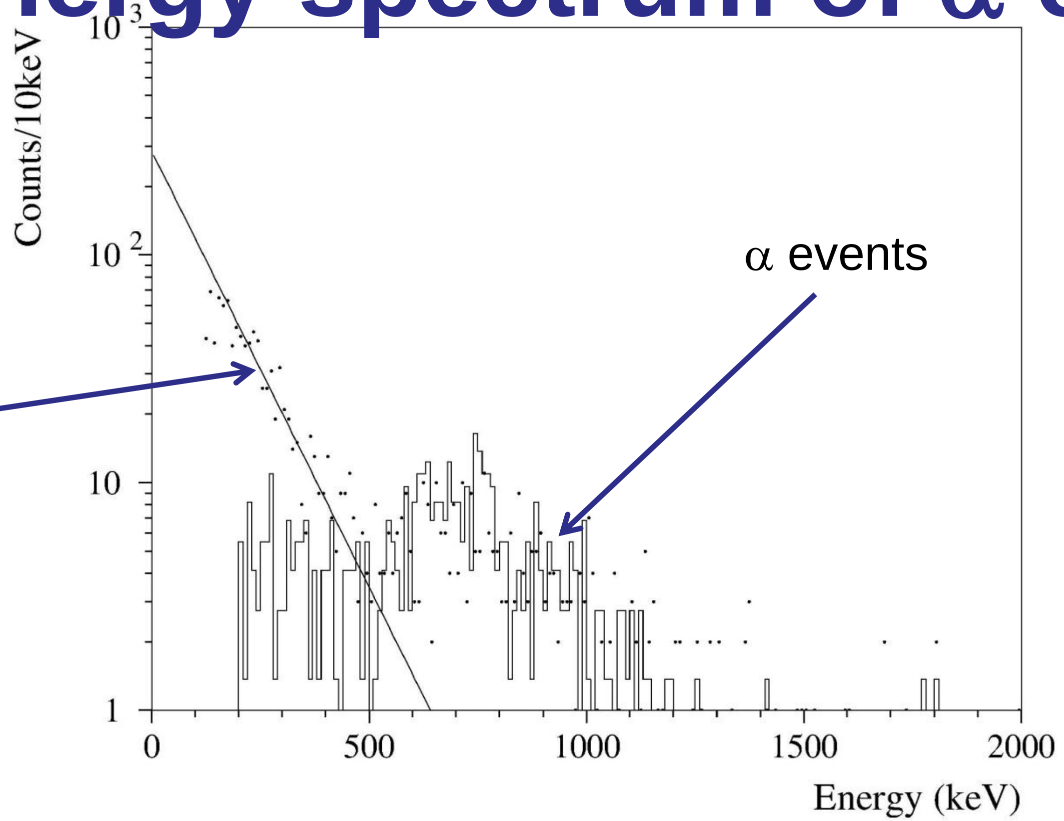
The **$ZnWO_4$** crystal crystal scintillator **after the recrystallization** and the were viewed by a low-radioactive **3 in PMT** through a high purity **quartz light guide ∅76×100mm**.

The total α activity after the surface layer treatment is **0.45 (3) mBq/kg**.

The total activity of ^{228}Th chain was estimated by using time-amplitude analysis as 0.002(2) mBq/kg (the same as before the surface layer treatment).

The energy spectrum of α events

the model of the residual γ(β) background



The energy spectrums of α events selected by the mean time method from the background data of the $ZnWO_4$ detector after its surface treatment measured during 1445.4 hours in the DAMA/CRYS set-up (dotted histogram) with the model of the residual γ(β) background (solid line) and of the $ZnWO_4$ detector before its surface treatment measured during 994 hours in the DAMA/R&D (solid histogram). The spectra are normalized on the time of measurements and mass of the crystal after the surface layer treatment.

Conclusions

- The recrystallization of a sample of cadmium tungstate crystal enriched in ^{116}Cd reduced the thorium contamination of the sample by one order of magnitude to the level of 0.01 mBq/kg (^{228}Th).
- The total alpha activity due to uranium, thorium, and their daughters decreased by a factor ≈3 to the level of 1.6 mBq/kg.
- The higher contamination of the rest of the melt remaining in the platinum crucible after the crystal growing process by potassium, radium and thorium, indicates strong segregation of the radioactive elements in the $CdWO_4$ crystals growing process.
- The concentration of α active uranium and thorium and their daughters in a thin surface layer of the enriched $^{116}CdWO_4$ crystal scintillators [1] was not due to the radioactive elements diffusion in to the $^{116}CdWO_4$ crystal during the annealing process in a ceramic oven.
- The recrystallization can be used in order to produce highly radiopure $CdWO_4$ crystal scintillators for high sensitivity double beta decay experiments.
- The possible concentration of radioactive elements (the activity of ^{228}Th and the total α activity of uranium, thorium and their daughters) in a thin ≈0.4 mm, surface layer was not observed in the $ZnWO_4$ crystal.

[1] Phys.Lett.B344(1995)72.

Thank you for attention!