

Development of ZnMoO_4 and Li_2MoO_4 crystal scintillators

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Content

The Low Thermal Gradient Cz Technique (LTG Cz)

The General Features of the Conventional Cz Technique

The General Features of the LTG Cz Technique

Crystals Growth BGO, CWO, ZWO, ZMO und LMO

Crystal morphology

Influence of growth rate on shaping

Advantages of BGO, CWO, ZWO, ZMO und LMO Crystals Grown by the LTG Cz

Collaboration with *Institute for Nuclear Research, Kyiv, Ukraine*

was initiated by F. Danivich visit of the NIIC at the end 2005

CWO for neutrinoless 2β decay search

2006-2008: Evaluation of CdWO_4 crystals grown by the LTG Cz as detectors for high sensitivity double beta decay experiments was performed.
CWO $\varnothing$ 20x20 mm - $\varnothing$ 70x70 mm

2009: Trial Growth of $^{106}\text{CdWO}_4$ crystals is performed
Trail growth of $^{116}\text{CdWO}_4$ crystals is planned

ZMO for neutrinoless 2β decay search

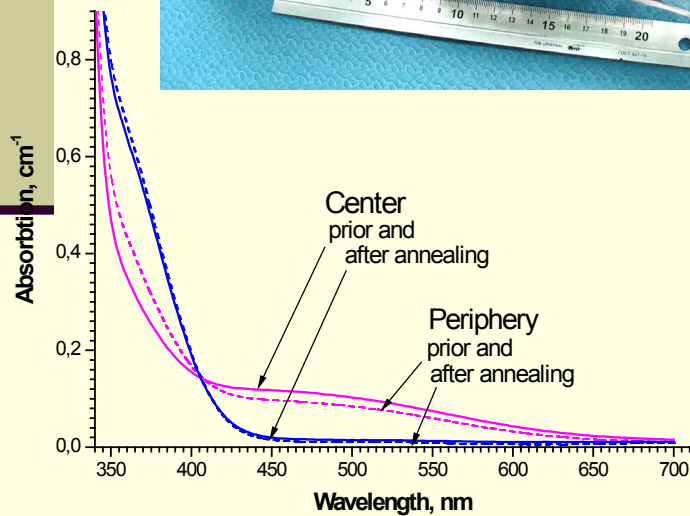
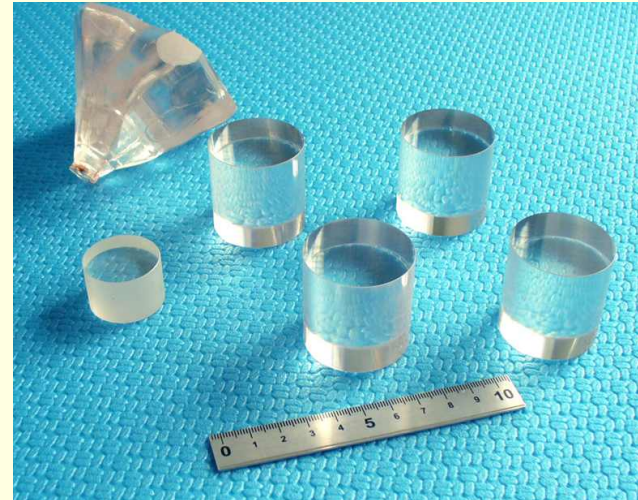
2012: Project LUMINEU (CNRS, France)

Luminescent **U**nderground **M**olybdenum **I**nvestigation for **NEU**trino mass and nature

2010 : Growth of large size radiopure ZnWO_4 crystals



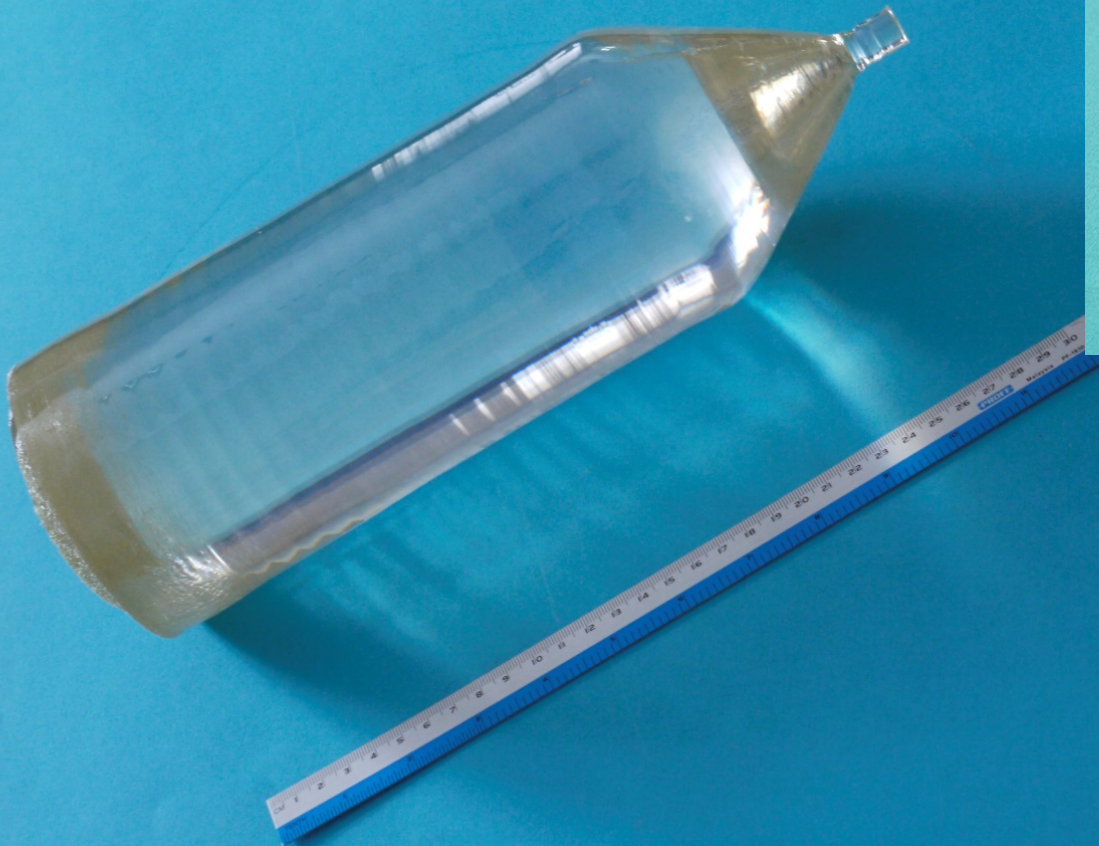
RHR $\sim 10\%$ at gammas 662 keV
for $\varnothing 40 \times 40$ mm cylinders



Collaboration with

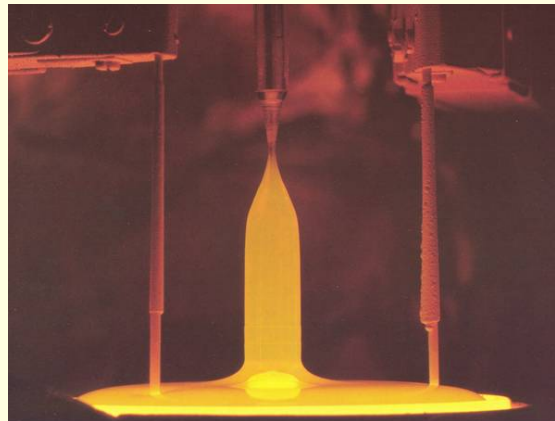
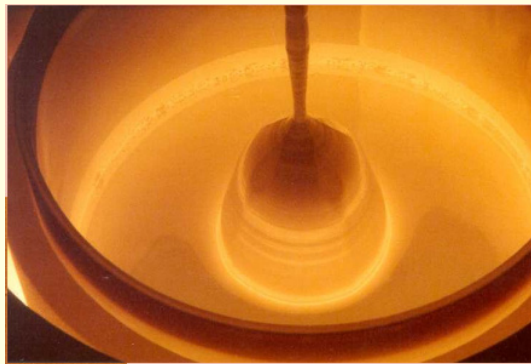
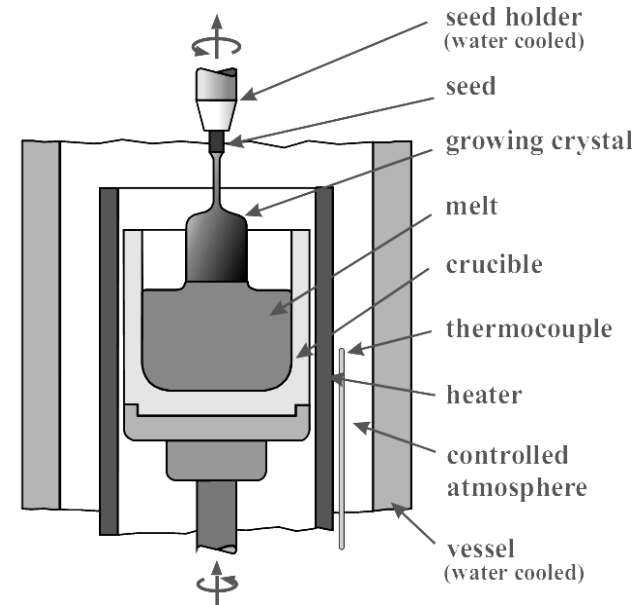
- *Institute for Nuclear Res., Kyev*
- *Max Plank Institute (Germany)*
- *INFN (Italy)*

Big size CWO und ZWO crystals

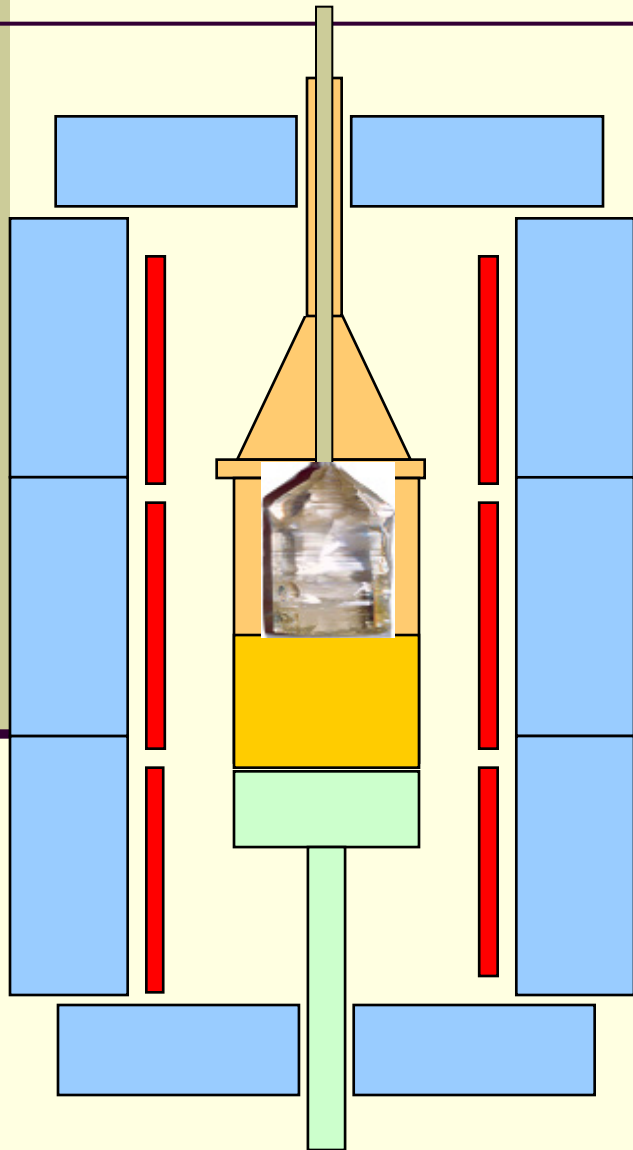


Traditional Czochralski Technique

- High temperature gradient (**$10\text{-}100\text{ K/cm}$**)
- Perfect cleavage of CWO
thermoelastic stress destroy crystal
- High local superheating of melt up to **200 K**
- Evaporation and decomposition of CWO melt

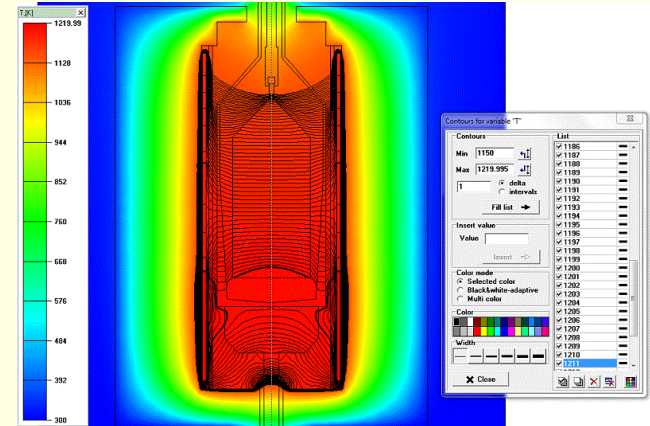
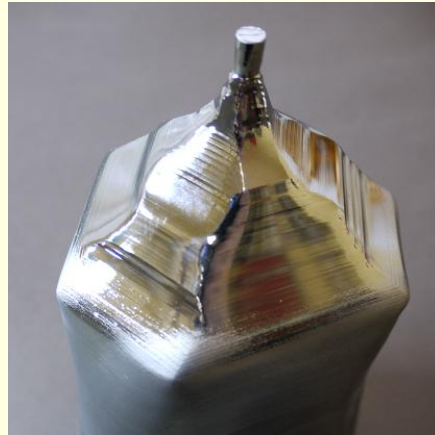


The General Features of the LTG Cz Technique

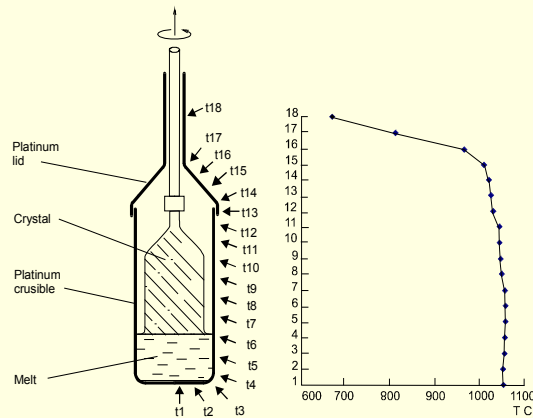


- The grown crystal inside the crucible
- Weighing control at all the stages of the process
- Temperature gradients less **1.0 deg/cm.**
- Temperature fluctuations in the melt are practically not developed
- Evaporation and decomposition processes are suppressed by the pipe socket, which works as a diffusion barrier
- The faceted interface develops and layered growth mechanism prevails

Experimental and the simulated values of temperature fields

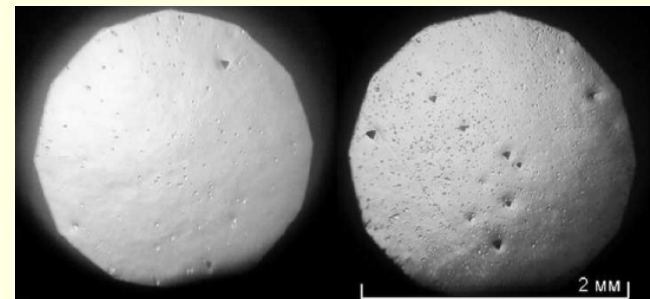


Software package CGSim was used for mathematical modeling of heat transfer in the growth furnace.

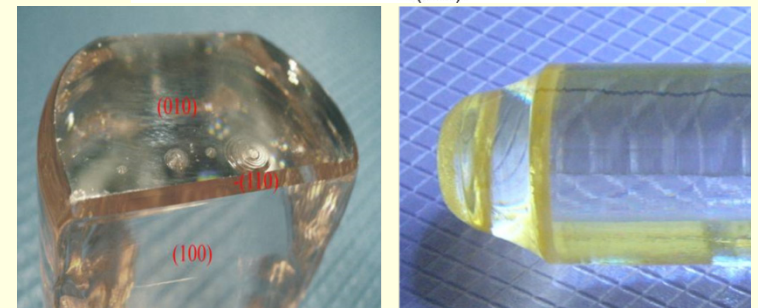
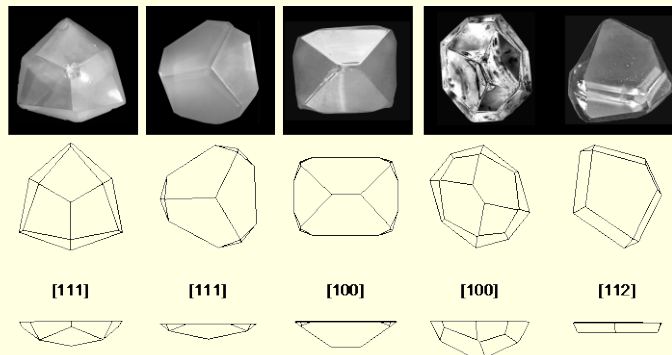
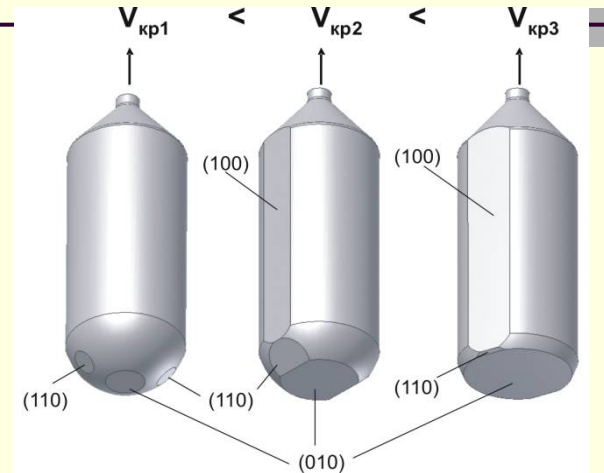
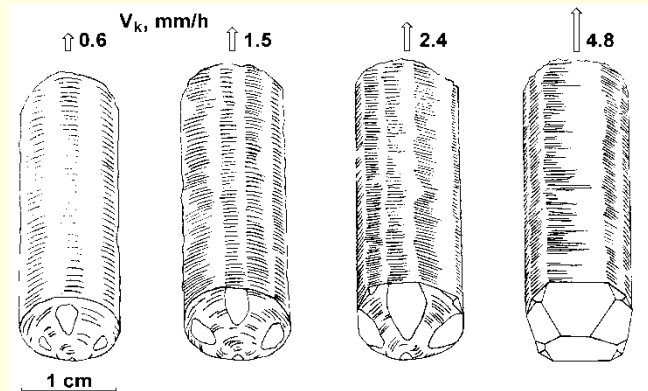


Heat conditions can be characterized experimentally by the curves of temperature distribution along the crucible

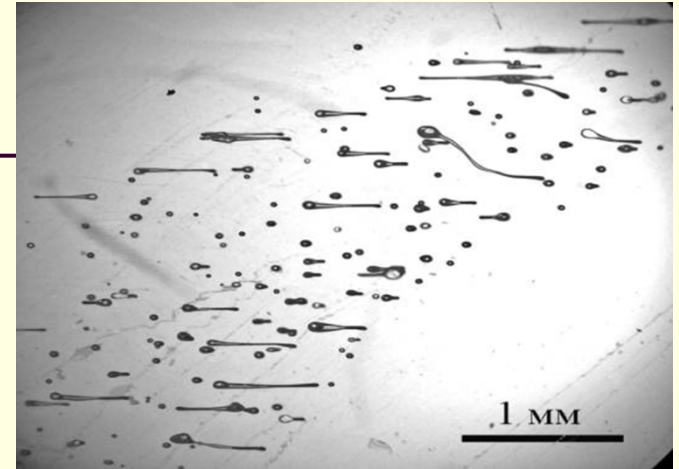
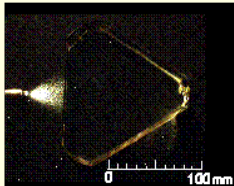
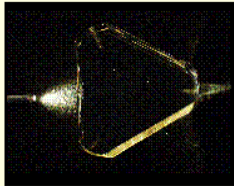
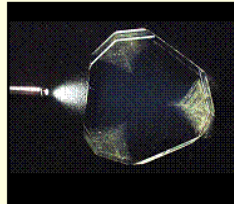
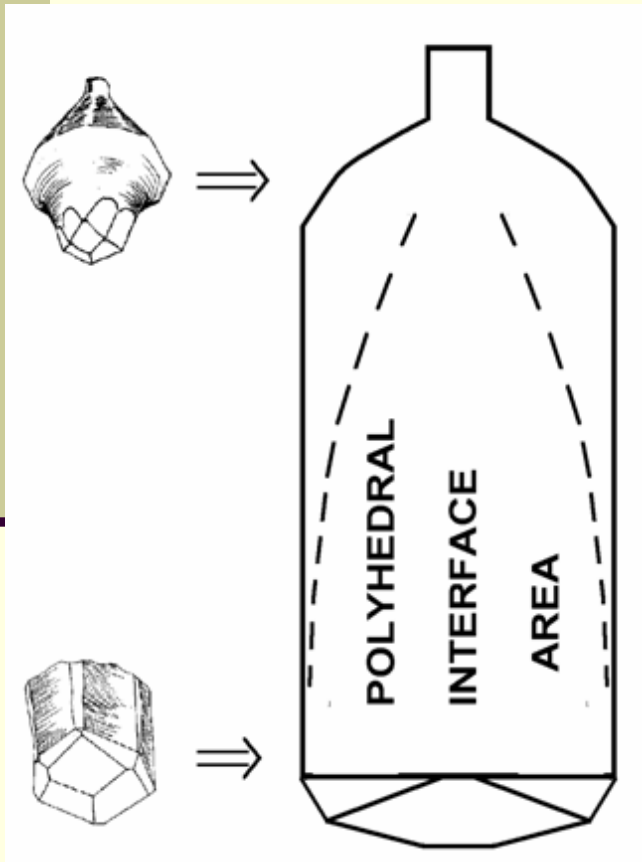
Dislocation density $10^2/\text{cm}^2$



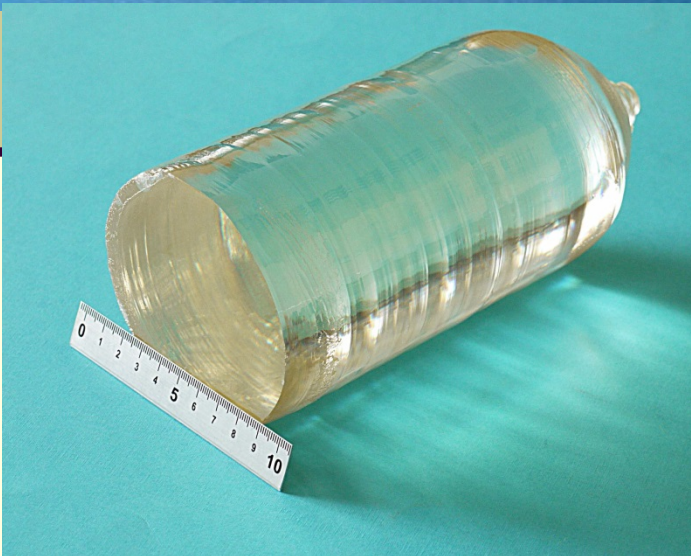
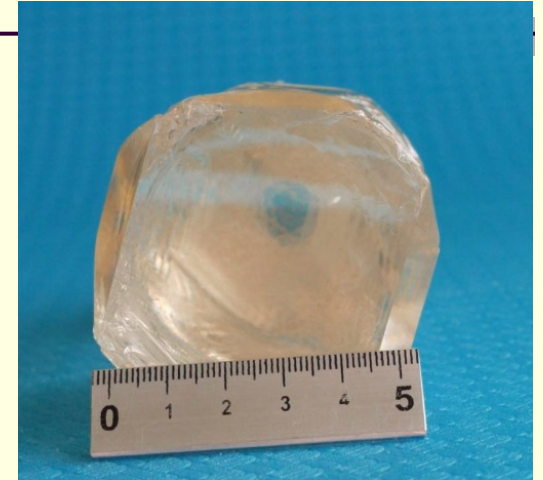
Crystal morphology LTG CZ



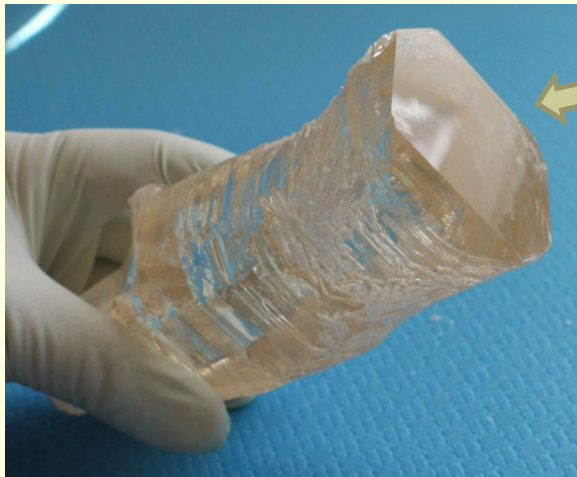
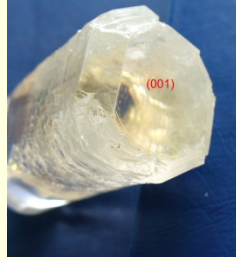
Co-existing two mechanism of growth



Layer-by-layer growth mechanism



ZnMO_4 crystals grown along [001] direction

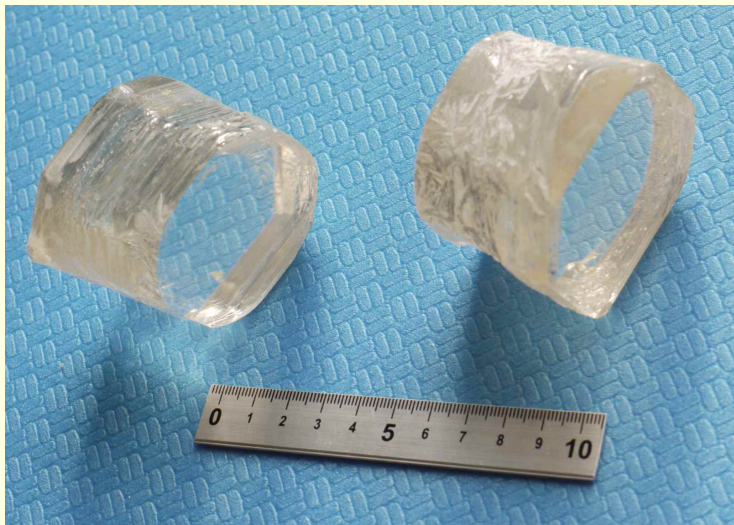


Growth
interface



2012: Project LUMINEU
(CNRS, France)

Luminescent **U**nderground
Molybdenum Investigation for
NEUtrino mass and nature



ZMO crystal grown with
the charge utilization factor ~80%

Two different ways are possible to grow high quality oxide crystals by Czochralski technique.

- **Conventional Czochralski technique is based on the use of high temperature gradients to suppress the face formation and achieve the fully rounded solidification front.**
- **By the contrast in The Low Thermal Gradient Czochralski Technique (LTG Cz) the growth conditions are selected in such a way that the temperature gradients become small and the solidification front becomes fully faceted .**

ZnWO₄ scintillation crystals

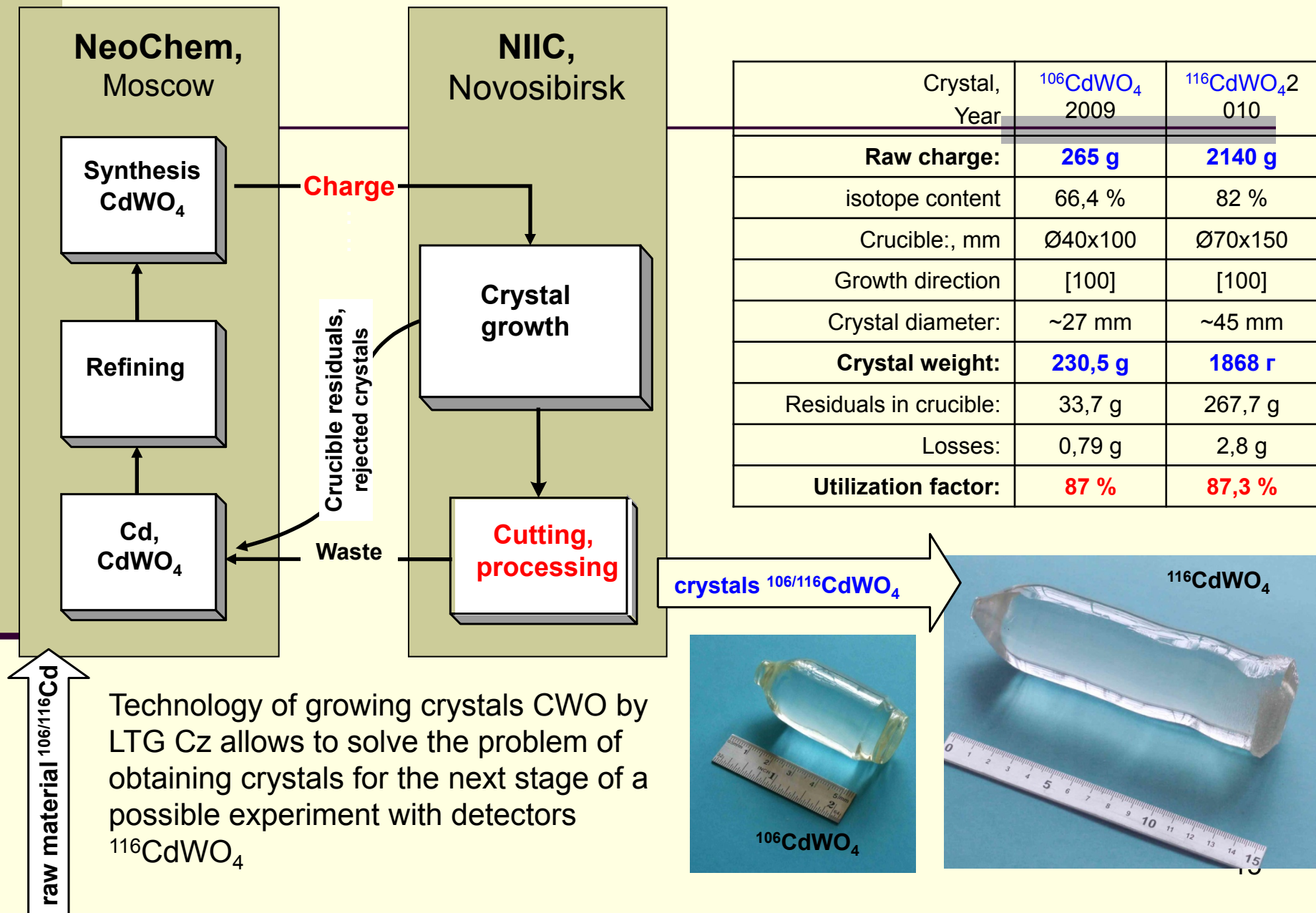


Growth direction [010]

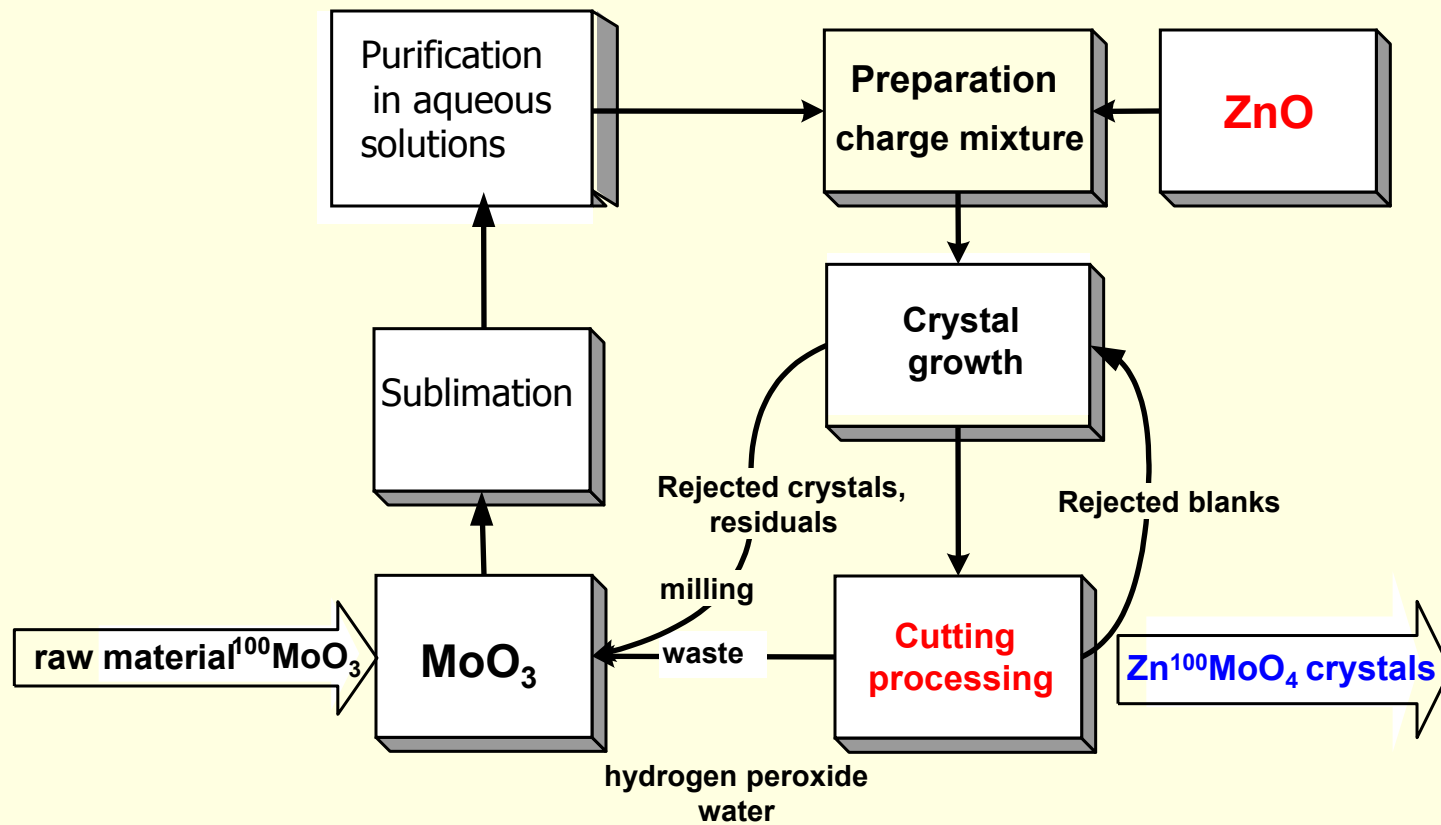
Crucible dimensions:
Ø 70 mm; H 150-200 mm

Initials:
HP ZnO from Umicore (Belgium);
HP WO₃ NIIC

Growth of isotopically enriched $^{106}\text{CdWO}_4$ and $^{116}\text{CdWO}_4$



ZMO: Full production cycle at NIIC is developing



JACKSON criterion $\alpha L/kT$

CWO			
Т пл	Q пл ккал/мол	Q пл дж/мол	
1543	33,3	133200	10,4

LMO	.		
Т пл	Q пл дж/г	Q пл дж/мол	
968	196	34068,72	4,2

ZWO			
Т пл	Q пл дж/г	Q пл дж/мол	
1475	356	111072	9,1

ZMO			
Т пл	Q пл дж/г	Q пл дж/мол	
1273	431	96975	9,2

168

BGO			
Т пл	Q пл ккал/мол	Q пл дж/мол	
1308	44,6	178400	16,4

Morphology of LMO crystals, grown under low thermal gradient conditions

■ V_{k1}



<

V_{k2}

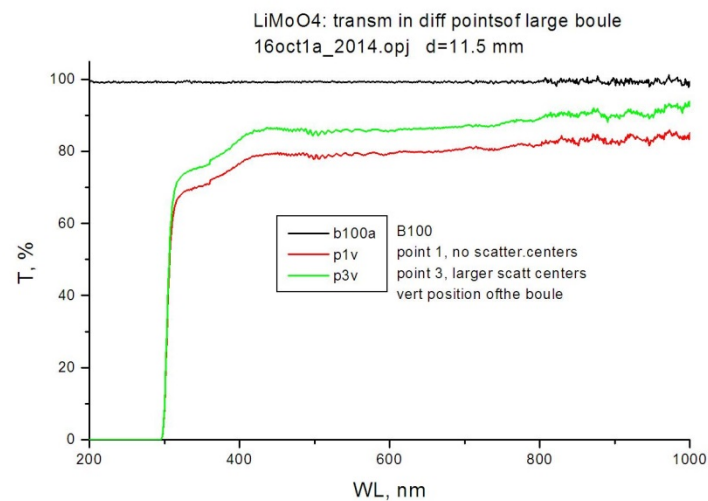


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V_{k3}



Results changing of crystallization rate in during growth process



The first crystallization and re-crystallization LMO crystals



- Diameter crystal 55mm
- Diameter crucible 68mm



Two different ways are possible to grow high quality oxide crystals by Low Thermal Gradient Czochralski technique.

- **The Low Thermal Gradient Czochralski Technique are allowed to achieve the fully rounded solidification front und good quality LMO crystal.**
- **By the contrast in The Low Thermal Gradient Czochralski Technique (LTG Cz) the growth conditions are selected in such a way that the temperature gradients become small and the solidification front becomes fully faceted .**

CONCLUSIONS

- *LTG CZ technique good variant for growing ZMO und LMO crystals high quality:*
 - *ZMO layer-by-layer mechanism of growth*
 - *LMO normal mechanism of growth*
- *The grown boules ZMO und LMO high quality is more 80% of the mass of initial powder it is real fact*

■ **LTG Cz technique with resistance-type heater is an effective method to grow oxide scintillation crystals. The grown crystals is more 80% of the mass of initial powder. The growth process and equipment are the most advanced parts of proceeding to mass scale production.**