Novel approaches to produce scintillation materials

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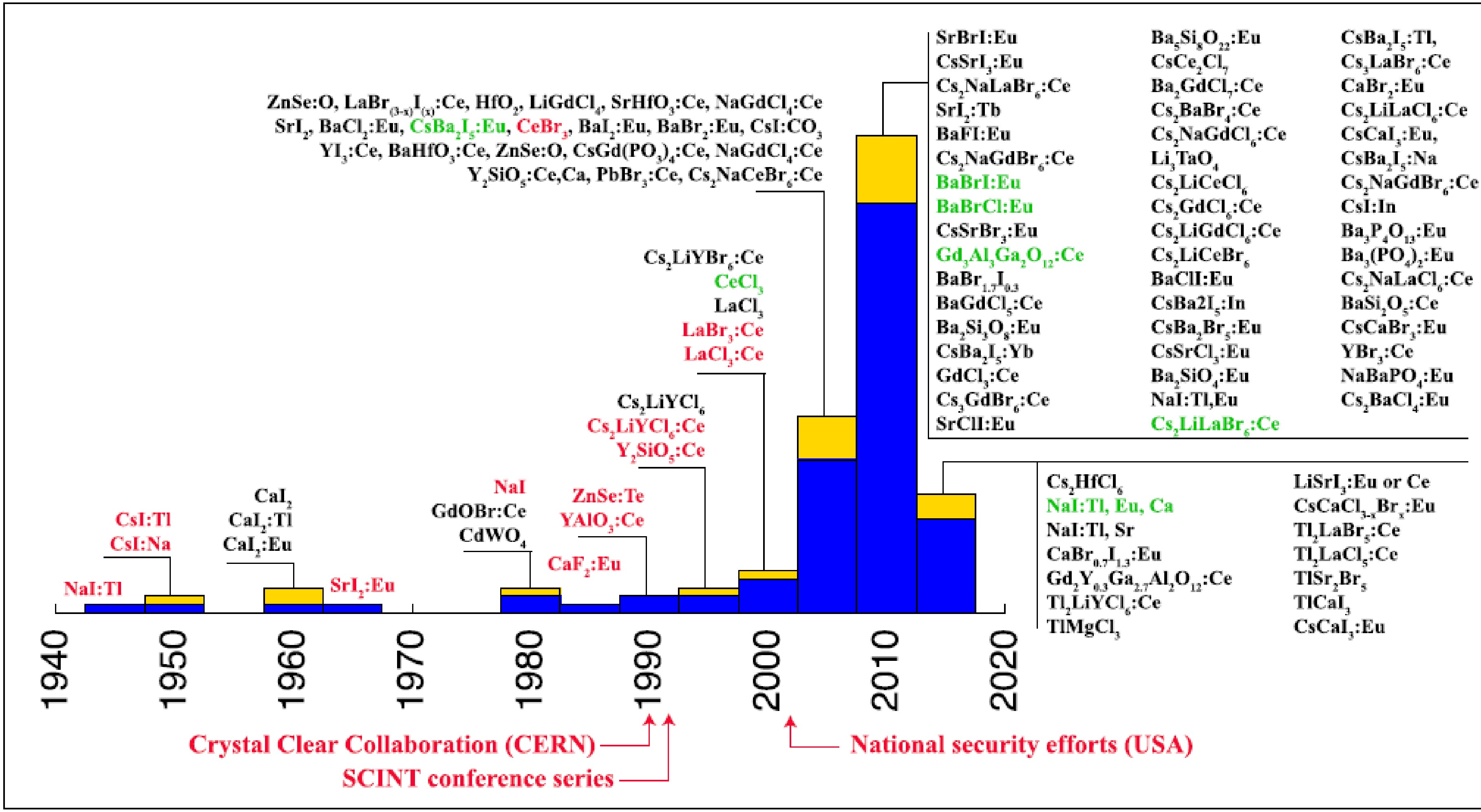
ISMART-2018, Minsk, Belarus

Result of 20 years research and development program



IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 65, NO. 8, AUGUST 2018

New or improved scintillators

Scintillator	LY, ph/MeV	R (^{137}Cs), %		Place	Cost, $/cc
$LaBr_3$:Ce	75000	2,6	new	on the market	100
SrI_2:Eu	115000	2,6	reinvented	on the market	150
GAGG:Ce	41000	6.5	new	on the market	250
CsBa2I5:Eu	102000	2.55	new	under development	-
BaBrI:Eu	97000	3.4	new	under development	-
NaI:Tl,Eu,Ca	40000	4.9	improved	under development	Potential – 3 $/cc
NaI:Tl	40000	6,2	classical	on the market	2

A reason of high price:

- **High cost of raw material and equipment for crystal growth**

- **Low productivity of crystal growth techniques**

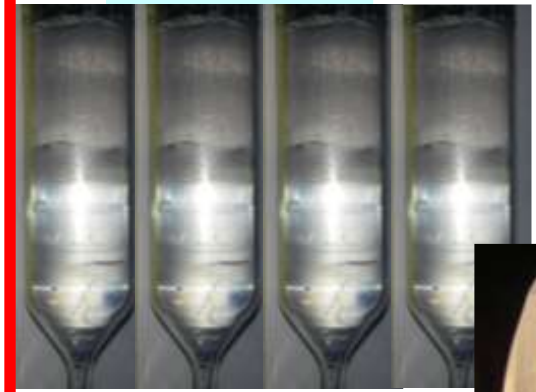
Outlines

- **Conventional crystal growth methods (Gradient freeze, Bridgman and Czochralski techniques)**

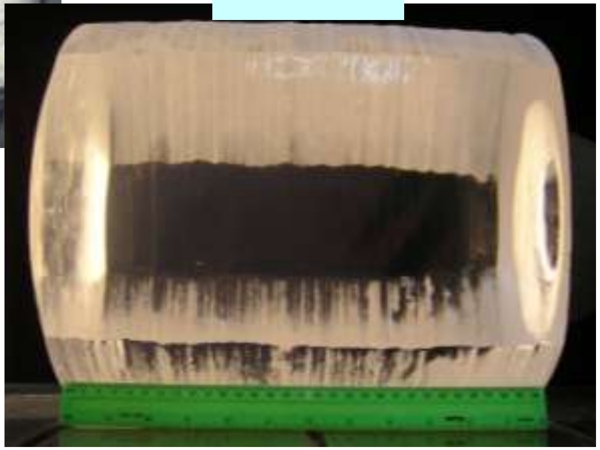
- **Alternative technology for scintillators production**

Ways of increasing of crystal growth technology efficiency

Number



Mass

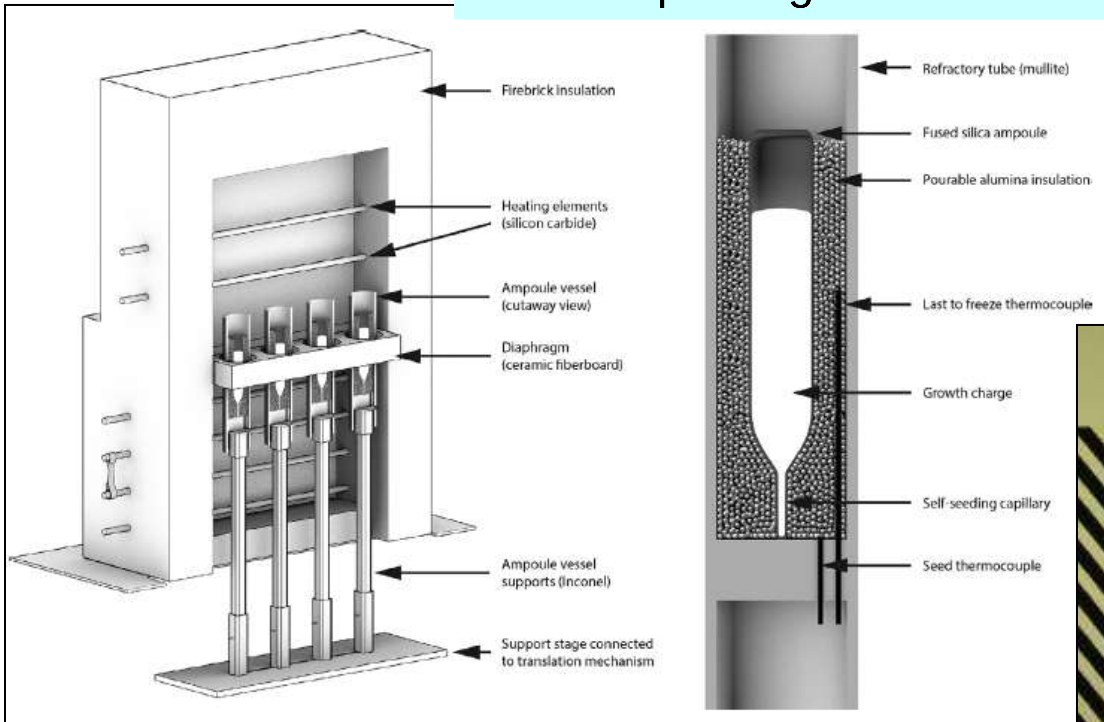


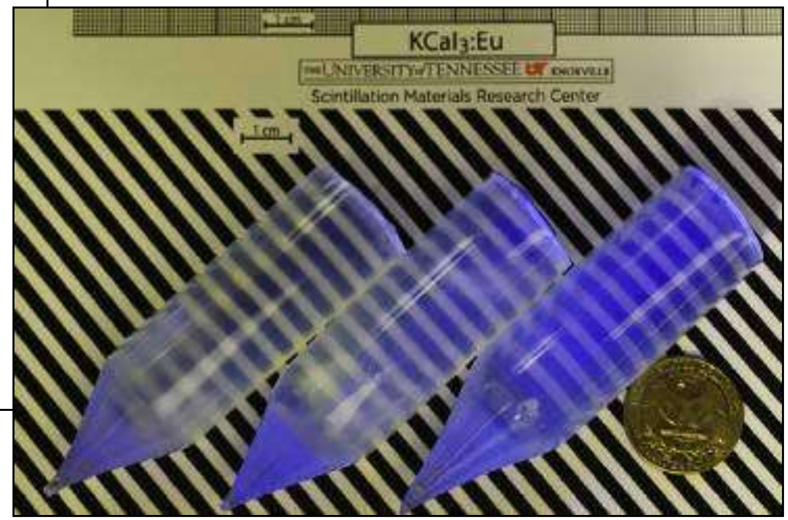
Quality of crystals

Decrease a cost of growth equipment

Scale up an equipment productivity (Bridgman technique)

Multi-ampoule growth station (MAGS)

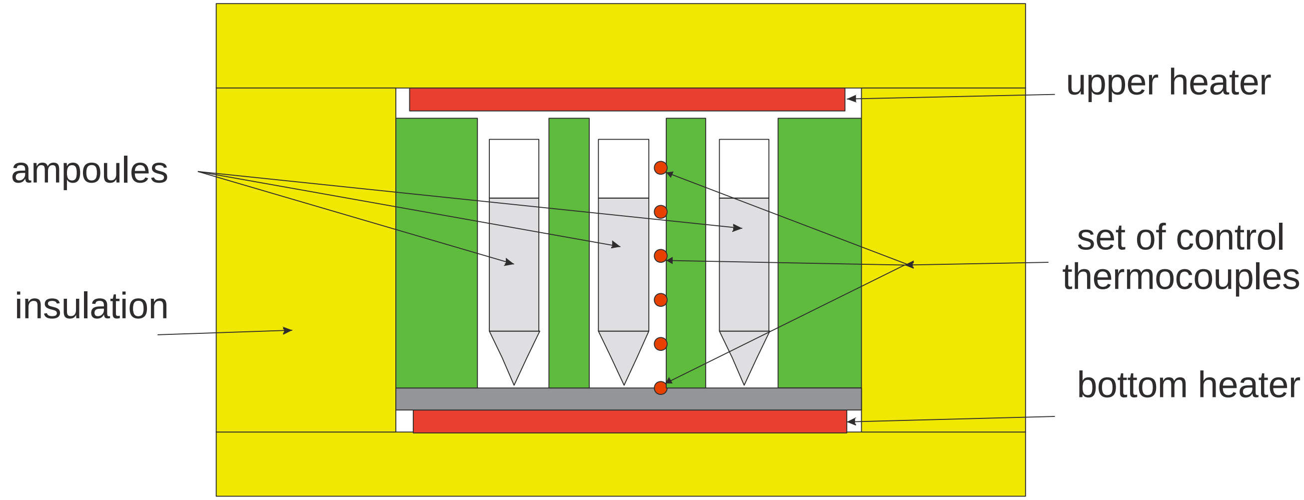


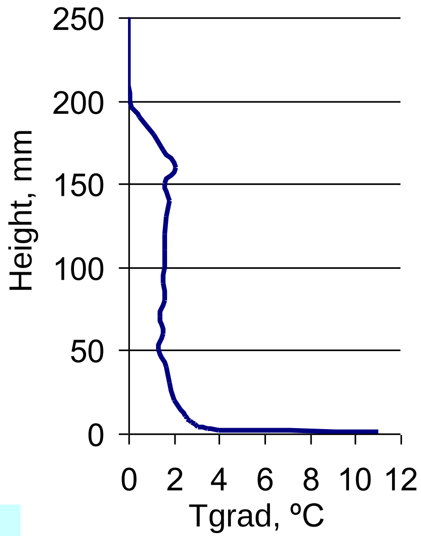


- Operating temperature - 1000 ºC
- 12 heating elements
- Diameter ampoule - Ø2“
- Up to 4 ampoules

A.C. Lindsey et al. / Journal of Crystal Growth 470 (2017) 20–26

Multi-ampoule single-zone VGF technique





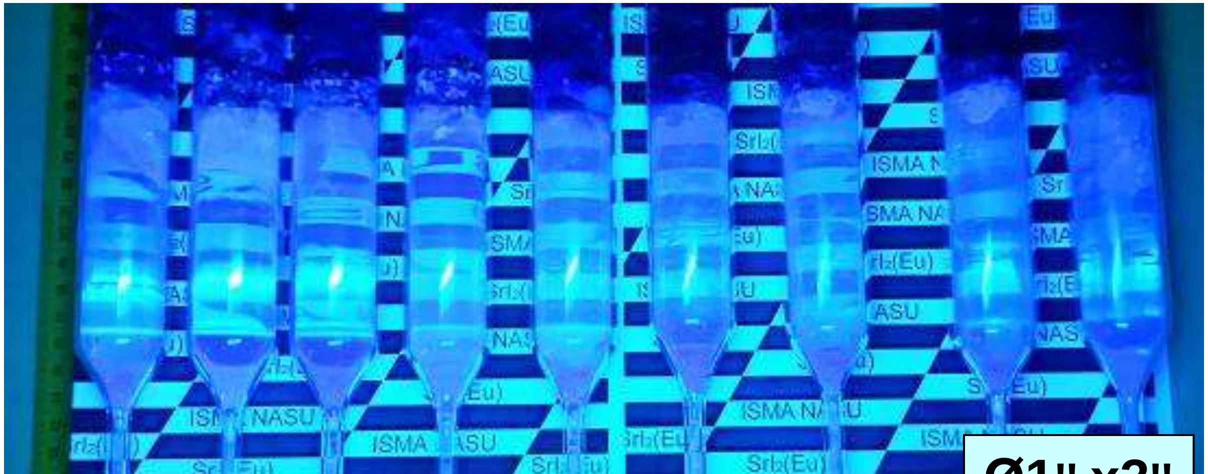
Sealed ampoule

Specification of growth equipment:

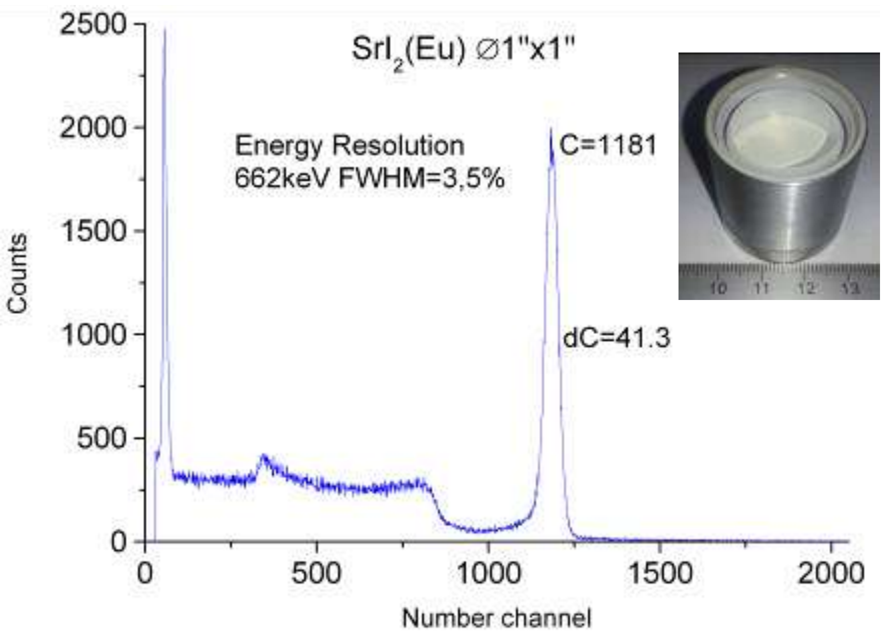
- **Simultaneously growth up to 19 crystals Ø1”x4”**
- **Ability to chose temperature gradient from 0 to 30 ºC/cm**
- **Growth rate from 0,1 mm/h.**

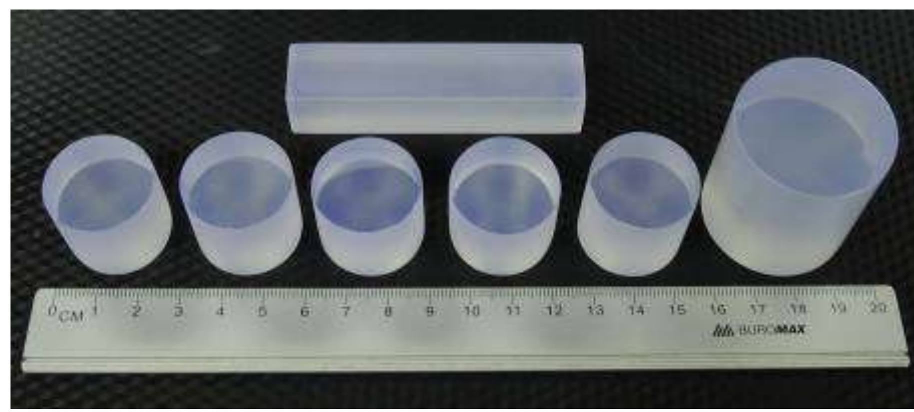
*Taranyuk V., Proc. of ICCGE-18, Nagoya, Japan 2016.

SrI_2(Eu) crystals growth


Ø1" x3"


Ø2"x3"

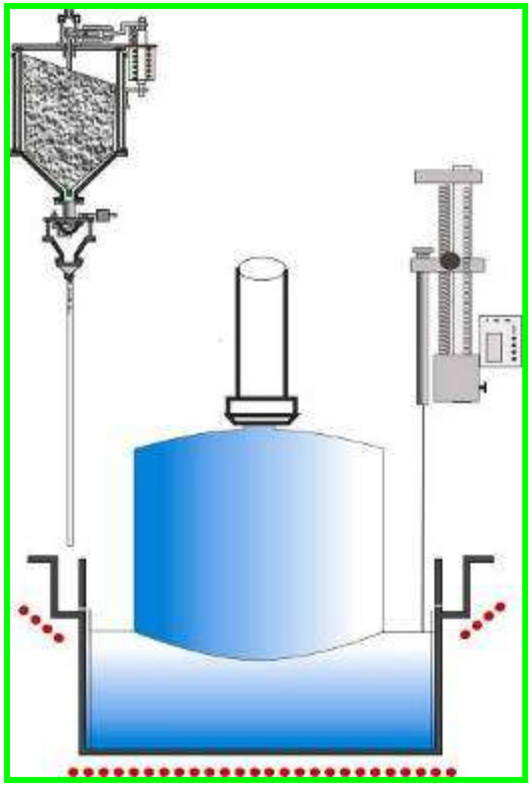




*Taranyuk V., Proc. of ECCG-6, Varna, Bulgaria 2018.

Czochralski technique – classical halide scintillators

Continuous crystal growth technique



100x100x400mm

- Stabile industrial technology
- High production yield
- Lower cost of crystals on the market (NaI(Tl) – 2$/cc)

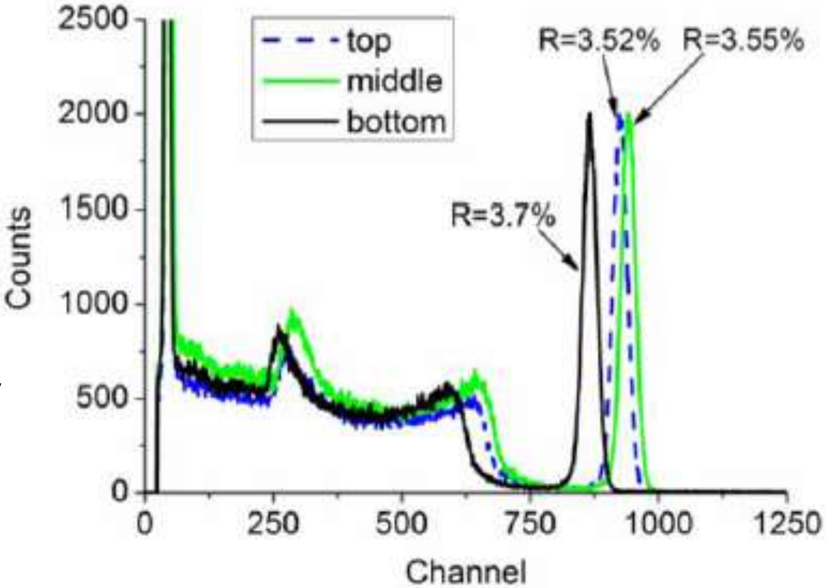
First steps. $SrI_2(Eu)$ - Czochralski technique.

Motivation –

increase of production yield

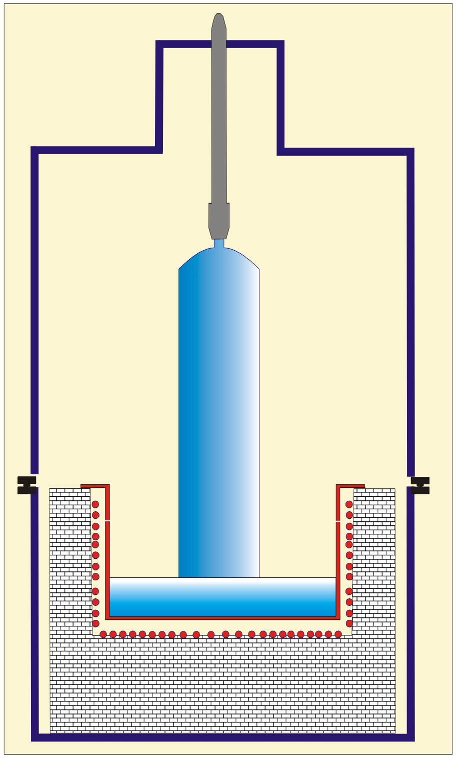
Result.

Good scintillation properties of obtained crystals (energy resolution, scintillation decay time, non-proportionality on the excitation energy).



E. Galenin, et. al. IEEE Transactions of Nuclear Science, 2018

Czochralski technique for oxide crystal growth



Scintillator	Melting point, ^oC
YAG:Ce	1940
LuAG:Ce	2020
GAGG:Ce	1850
YAP:Ce	1875
LYSO:Ce	1800
LuAP:Ce	1960
LSO	1780

Ø3" GAGG

Kamada et al., J. Cryst. Growth, 452 (2016) 81-84.

- High melting point of oxide crystals ~ 2000 C
- High cost of crystals – Ir crucible
- Induction heating
- Low productivity

Crystal cost structure

Silicon crystals (Si)	Oxide crystals	Iodide crystals (NaI)
68% - raw material	57% - raw material	60% - raw material
10% - crucible	15% - crucible	13% - crucible
8% - equipment		15% - equipment
4% - labor		3% - labor
4% - power	8% - power	4% - power
6% - other		5% - other

Iridium and Platinum crucible usage.

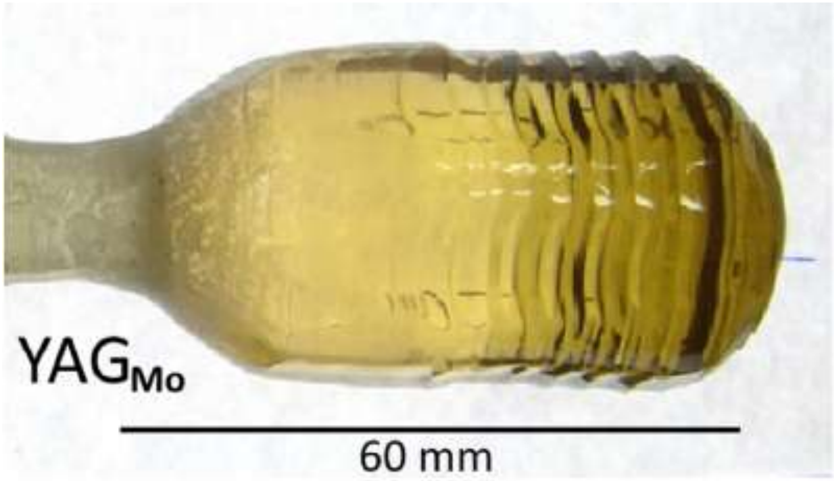
Oxide crystals

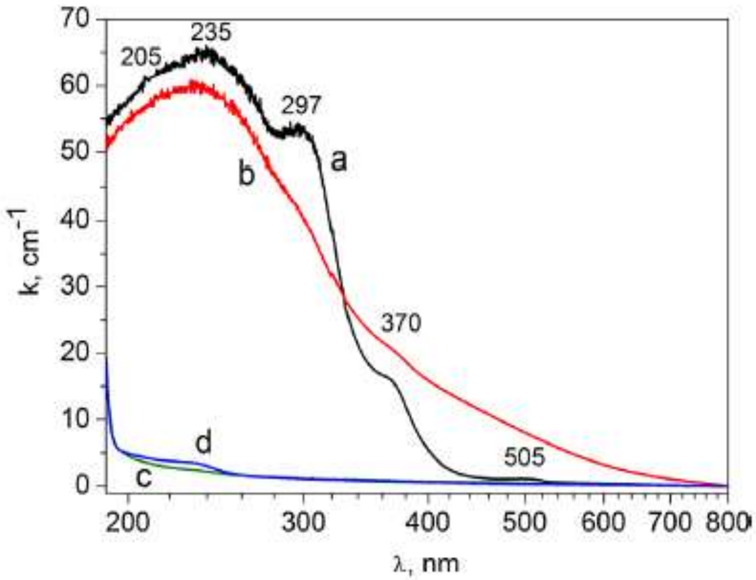
From 0.5 kg to 50 kg

Iodide crystals

Search for replacement a high cost crucibles

YAG crystal growth in Mo, W crucibles

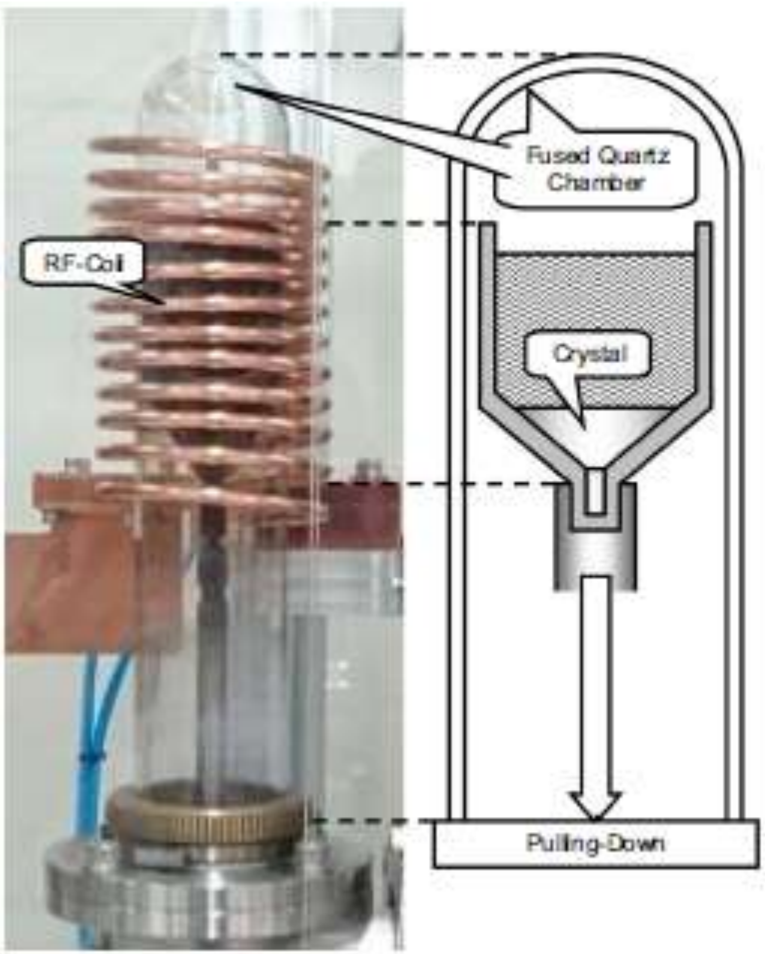


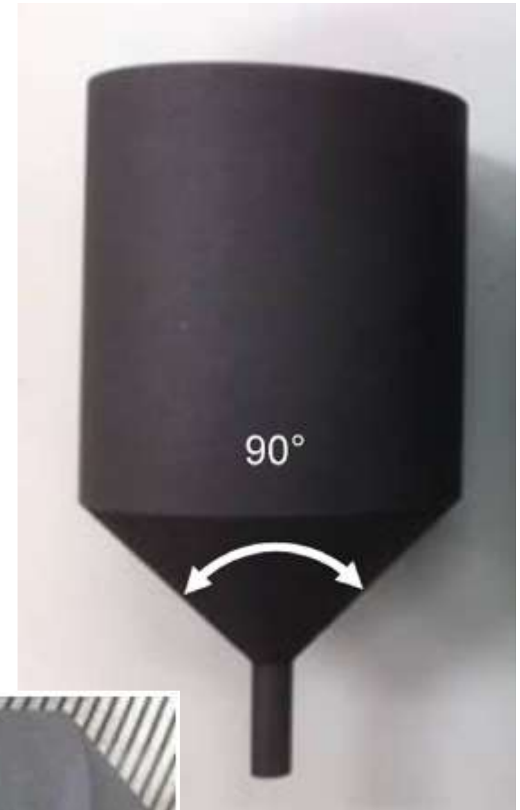


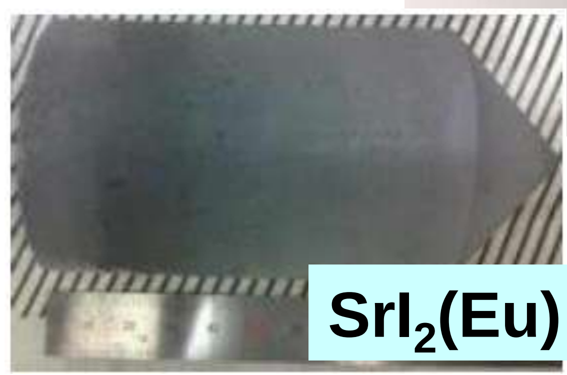
YAG$_{Mo}$ absorption spectra: a – as grown, b – after oxidizing annealing during 3 h, c – after reducing annealing during 6 h, d – after oxidizing annealing during 25 h.

S. Tkachenko et al./Journal of Crystal Growth 483 (2018) 195–199

Multiuse carbon crucible for halide crystal growth



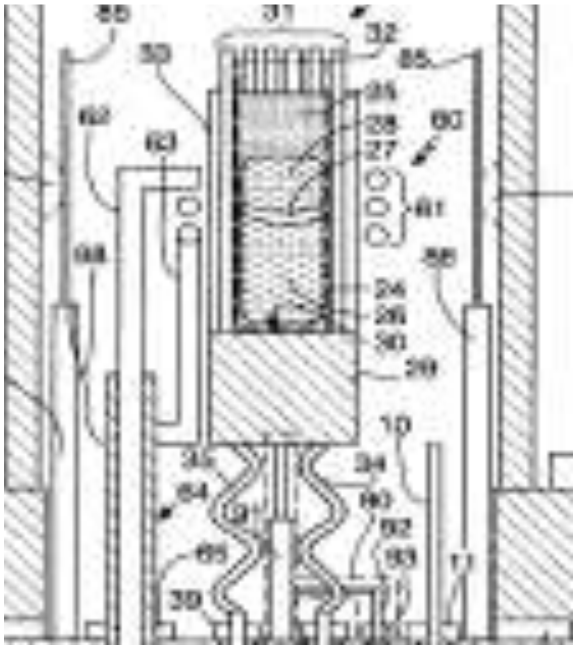




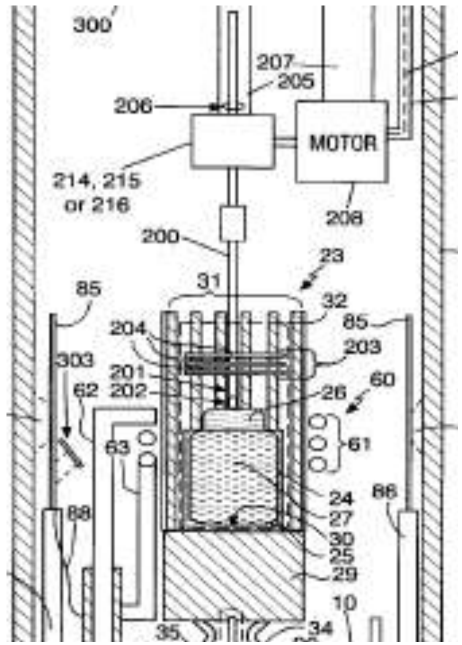
A. Yoshikawa et al. / Journal of Crystal Growth 452 (2016) 73–80

Skull method - possible ways for oxide crystal growth

Directional solidification

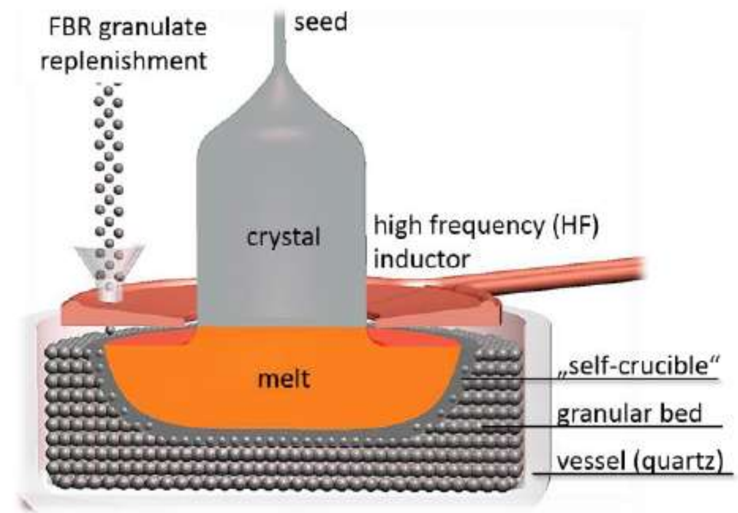


US Patent 5,900,060



US Patent 5,863,326

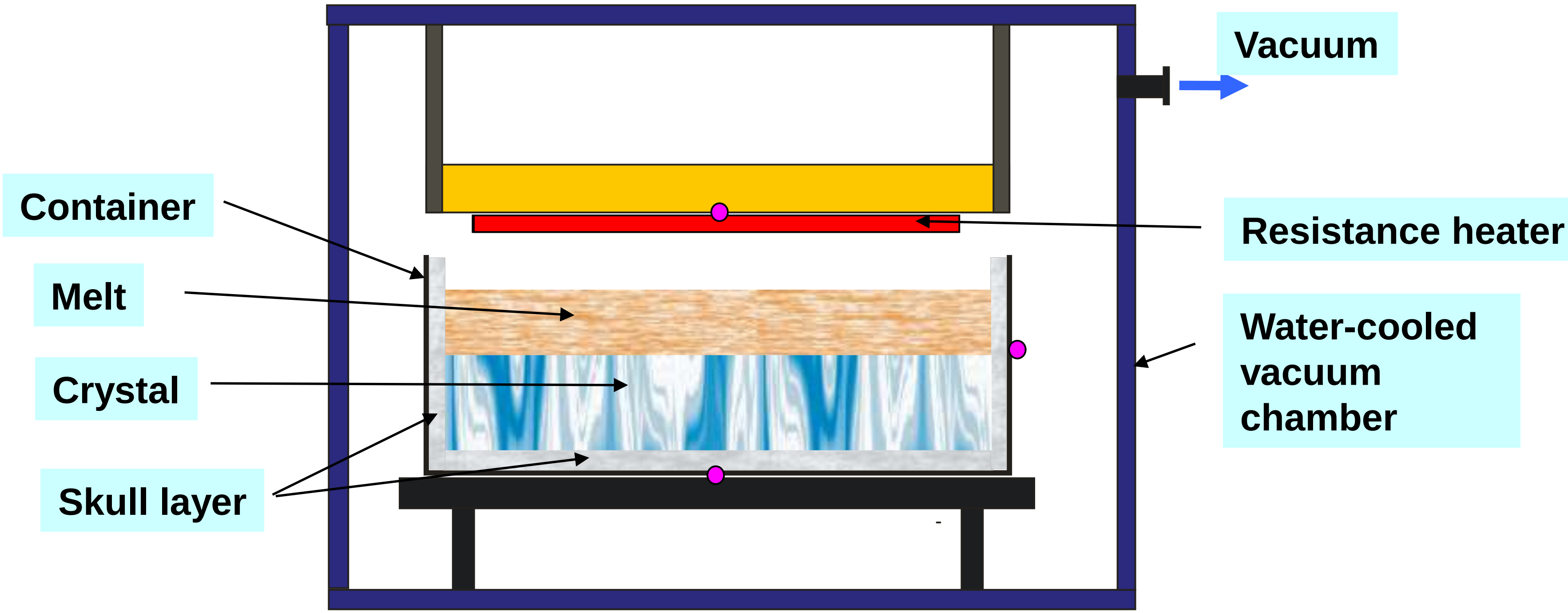
Czochralski technique



R. Menzel. Si crystal growth from granulate crucible

Perspective for oxides crystals.
First steps have done (SCINT-2017, IEEE-2018).
Yoshikawa – Skull method for GAGG:Ce, GPS:La crystals grown.

Skull technique for halide crystal growth

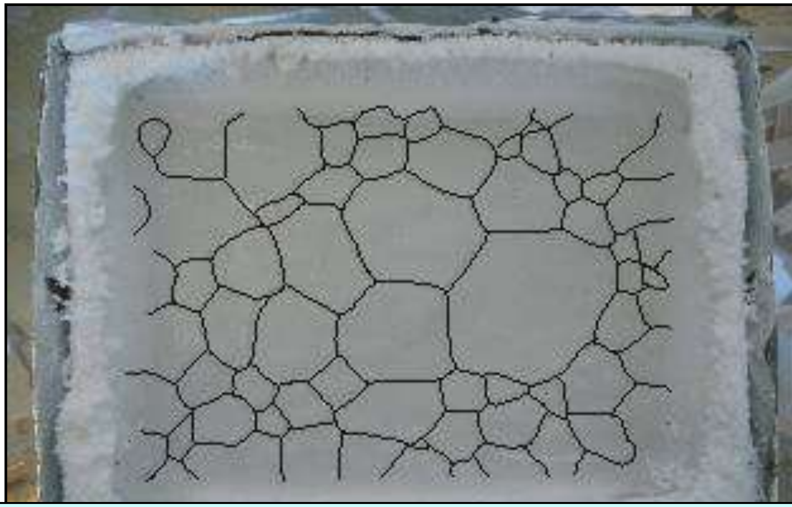


Advantages:

- No contact of the melt and crucible
- No noble metal as crucible material
- No moving elements
- – No adhesion between crystal and crucible (container)
- – Ability to grow crystals large area any configuration (circle, square)

•V. Taranyuk, A. Gektin, I. Kisil, A. Kolesnikov, NaI(Tl) and CsI(Tl) scintillation crystal growth by skull method, J. Cryst. Growth 318(1) (2011) 820–822.

Polycrystal – Skull technique



The size of block – from 3 mm to 40 mm

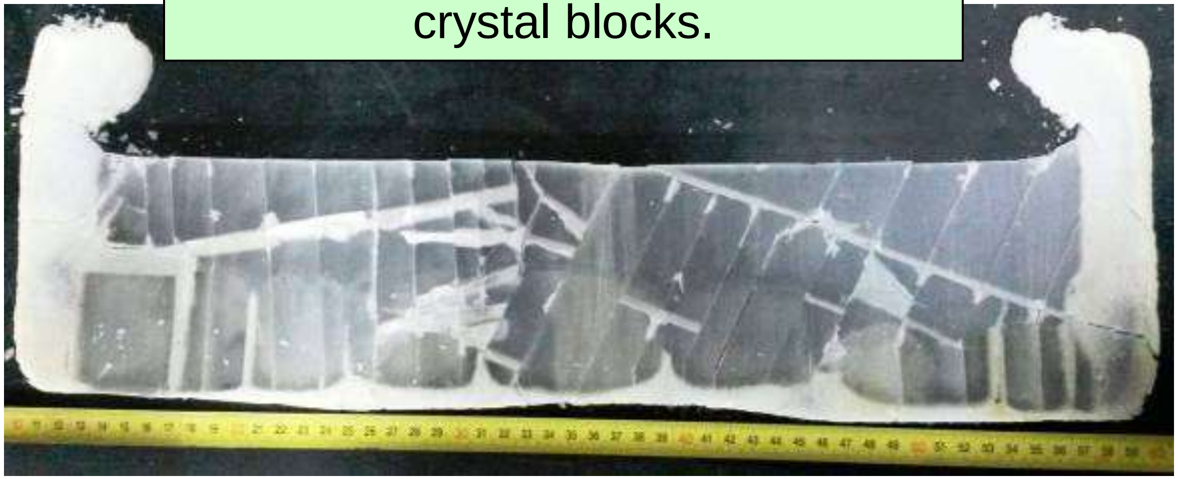
Mass of crystal – 160 kg, 470x470x200 mm

Skull technique – single crystal (NaI(Tl)).

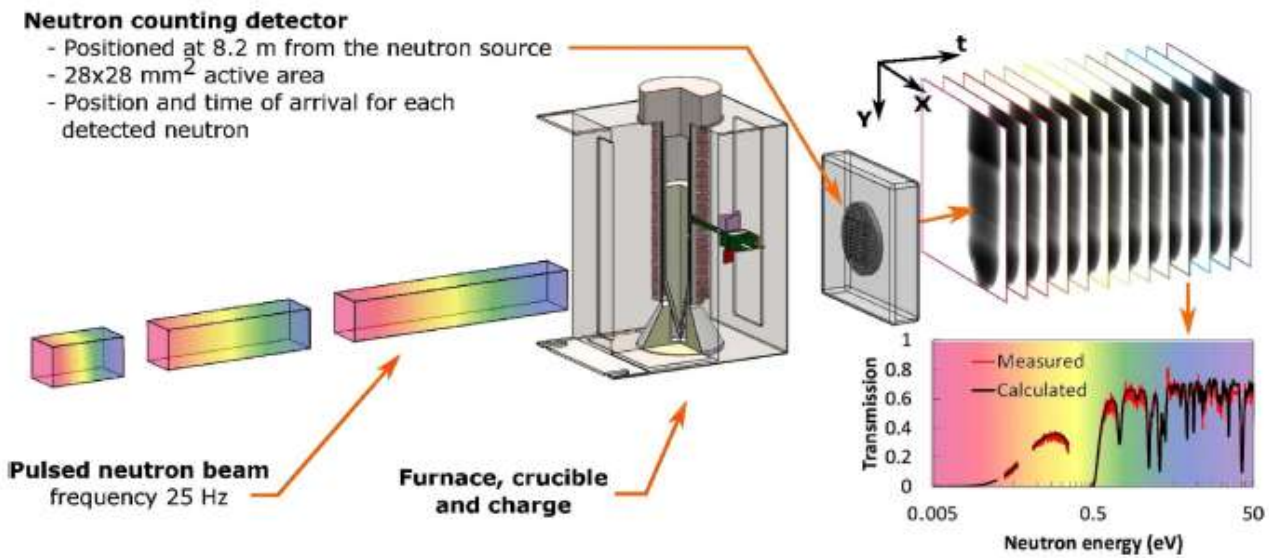

Seed crystals


450x450x90 mm

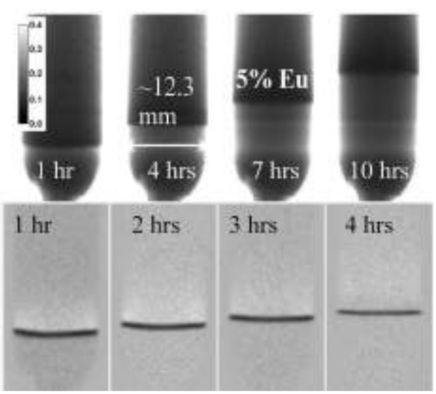
220x220x90 mm – size of single crystal blocks.



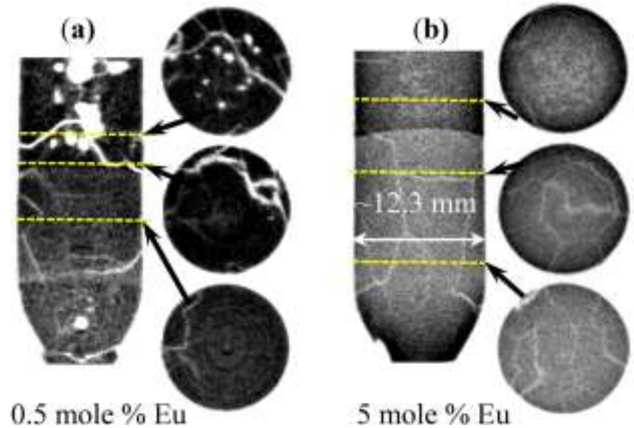
Energy-Resolved Neutron Imaging – *In situ* diagnostics of crystal growth



Innovative method for developing and improving crystal growth process!

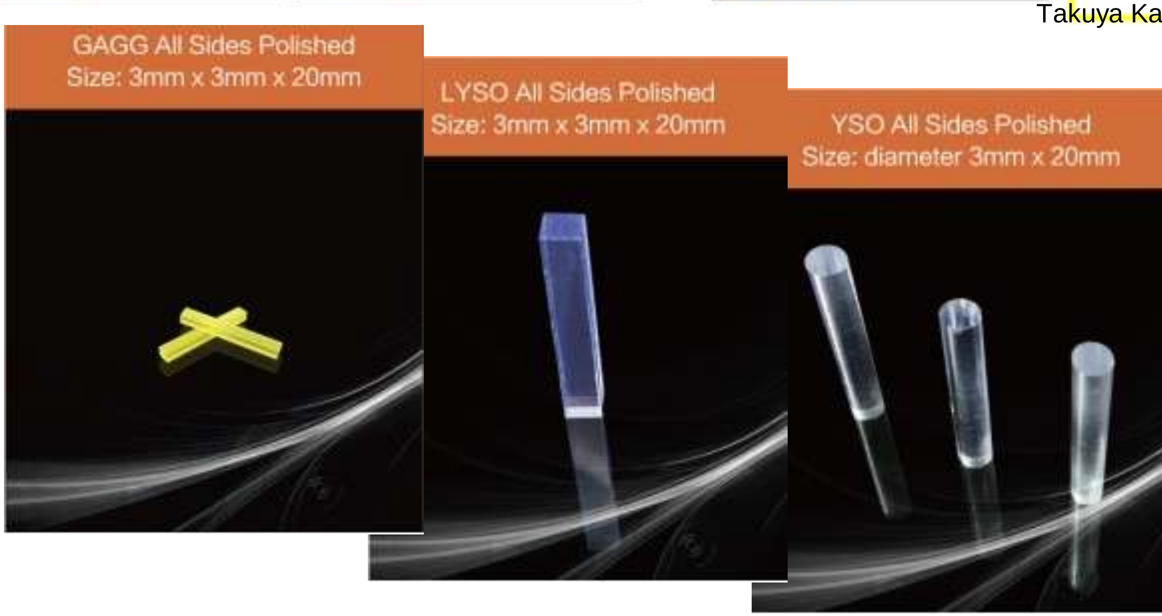


- **Visualization of location and shape of liquid/solid interface**
- **Mapping of the elemental composition**
- **Dopant concentration reconstruction**
- **Imaging of crystal defects**



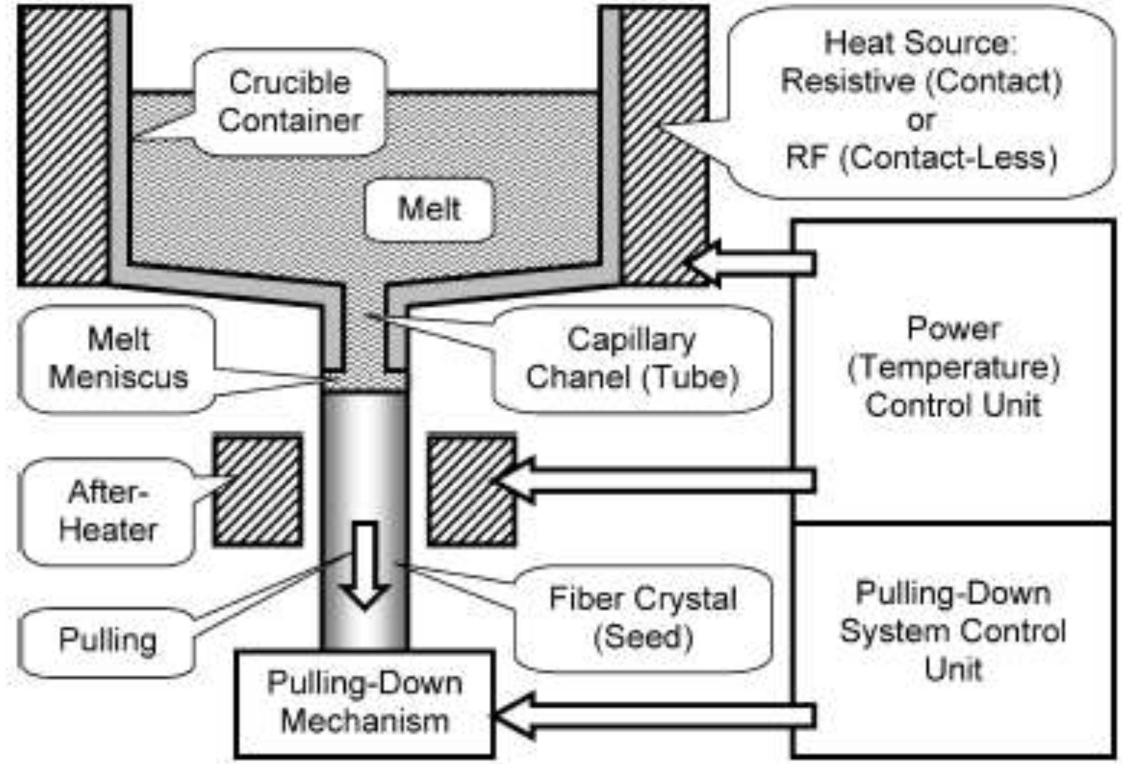
Scientific **Reports** | 7:46275 | DOI: 10.1038/srep46275

Trend – pixel matrix detectors.



Micro-pulling down technique –
a good decision for laboratory research
and small elements production

Micro-pulling down technique

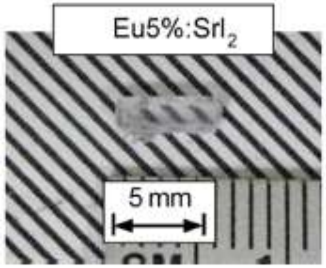


- **Fast result**
- **High growth rate**
- **Small amount of raw material**
- **Growth of fibers**

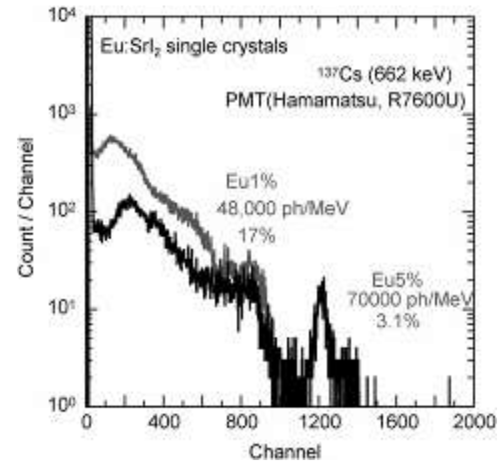
Shaped Crystals
Growth by Micro-Pulling-Down Technique
(Eds.)T. Fukuda; V.I. Chani
2007, XV, 341 p. 254 illus., Hardcover

Micro-pulling down for laboratory and mass production

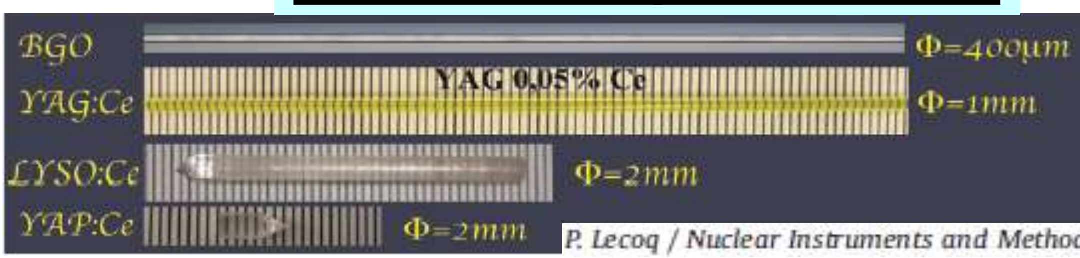
Laboratory



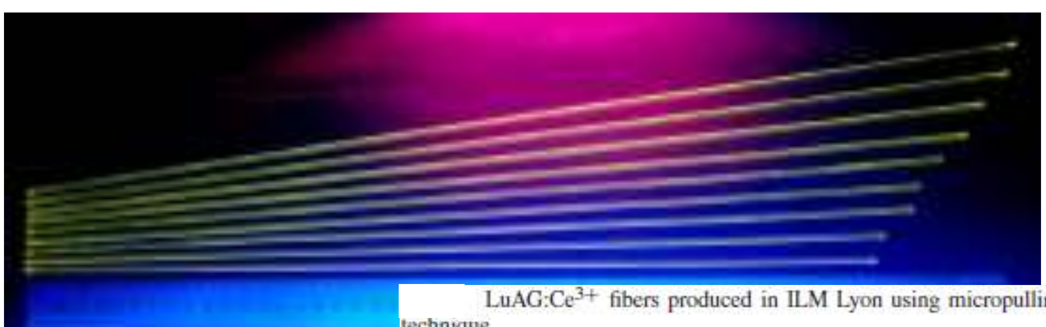
Y.Yokota et al./ Journal of Crystal Growth 375 (2013) 49–52



Fiber mass production



P. Lecoq / Nuclear Instruments and Methods in Physics Research A



LuAG:Ce^{3+} fibers produced in ILM Lyon using micropulling-down technique.

See I. Gerasimov 11[30]

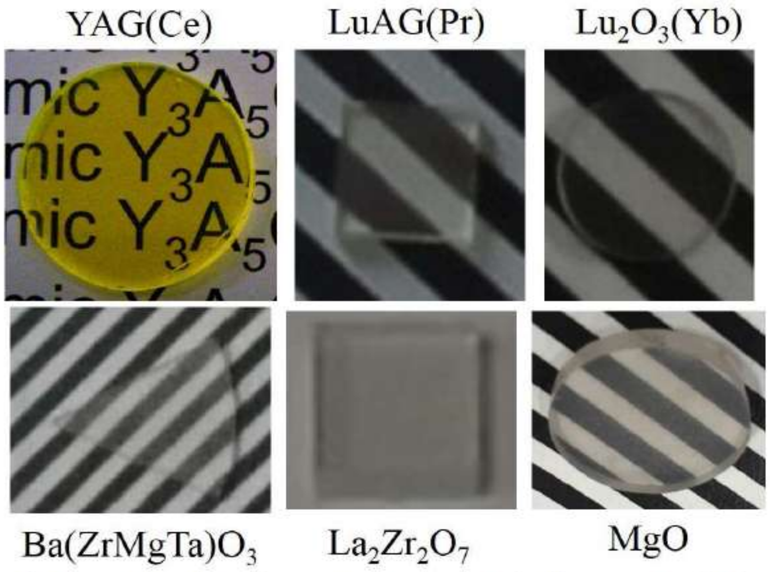
Scintillation ceramics –
serious competitor for crystals

Ceramics

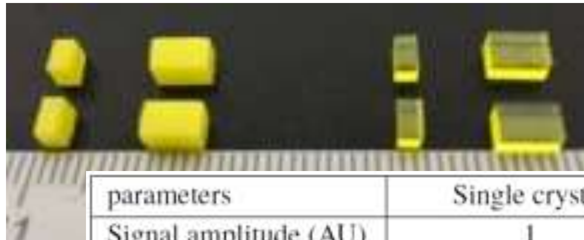
Advantages:

- Manufacturing temperature lower than melting point
- High productivity – many samples can be produced simultaneously
- High mechanical strength

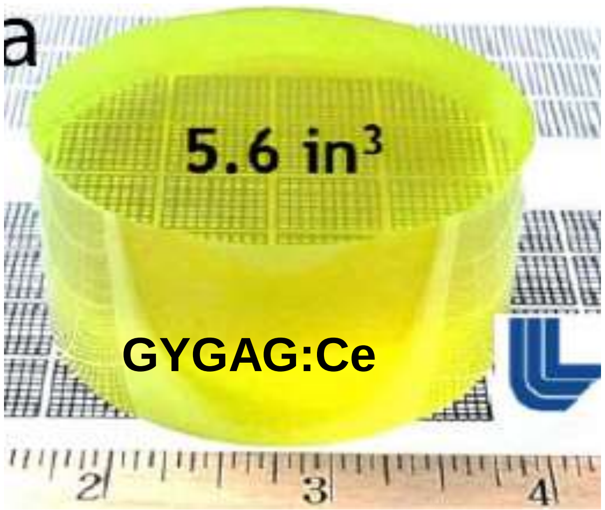
Main application area of ceramics –
materials with high melting point (oxides)



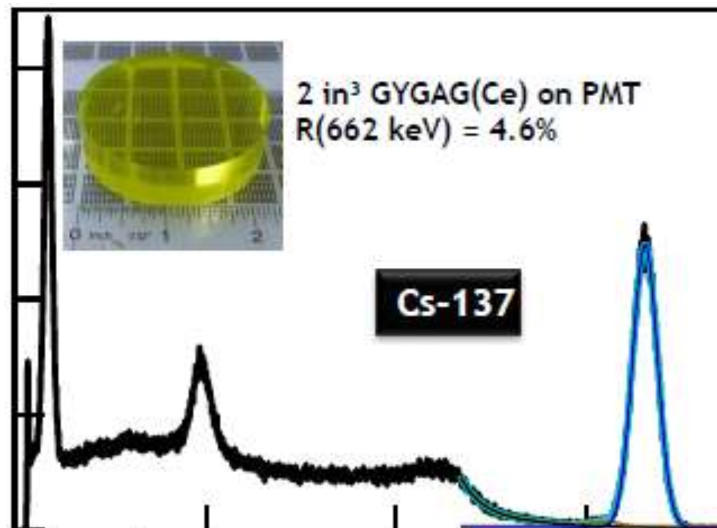
Progress in scintillation ceramics – from small samples to big transparent elements



parameters	Single crystal	Ceramic
Signal amplitude (AU)	1	0.61
Energy resolution (%)*	9.9 ± 0.2 %	13.0 ± 0.3
Time resolution (ps)*	465 ± 37	307 ± 23
Decay time (ns)*	Fast (92.5, I = 56.9%) Slow (220, I = 43.1%)	Fast (88.5, I = 72.6%) Slow (313, I = 27.4%)



N. J. Cherepy *et al.*, *Proc. SPIE, Hard X-Ray, Gamma-Ray, Neutron Detect. Phys. XVI*, vol. 9213, p. 921302, Sep. 2014



N.J. Cherepy LLNL-CONF-676780

Conclusion

By now some new perspective directions for scintillators produce have appeared and only purposeful work and time will show what will dominate in the future.

Waiting for a new progressive results!

Thank you very much!