Ba-Si-Gd:Ce and Gd-Si:Ce Glass Scintillation Materials

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Motivation

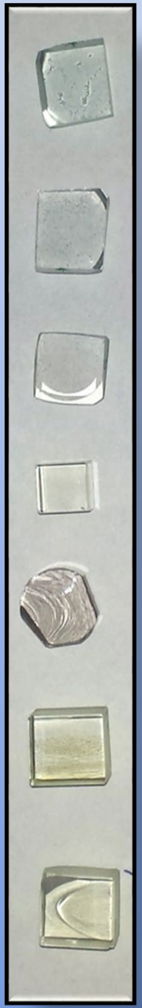
- Scintillating materials based on glasses and glass ceramics can be considered as alternatives for the crystalline scintillators currently used in radiation detectors for the detection of high-energy photons and neutrons in high-energy physics experiments and homeland security applications

- Glass material can be fabricated in various sizes and shapes, such as blocks, plates, and thin fibers. Large quantities of the detection units can be produced in a relatively short period of time

- The lead-free glass $BaO \cdot 2SiO_2$: Ce (disilicate–barium, DSB: Ce) has a density of 3.7 g/cm^3 and is found to be radiation hard. Further technology optimization showed that the loading of the DSB: Ce glass by gadolinium increases the material density up to 4.5 g/cm^3 and results in an increase in the light output by a factor of five, significantly improving the efficiency of the electromagnetic radiation registration and making the material sensitive to neutrons.

Physical properties of different heavy silica glasses

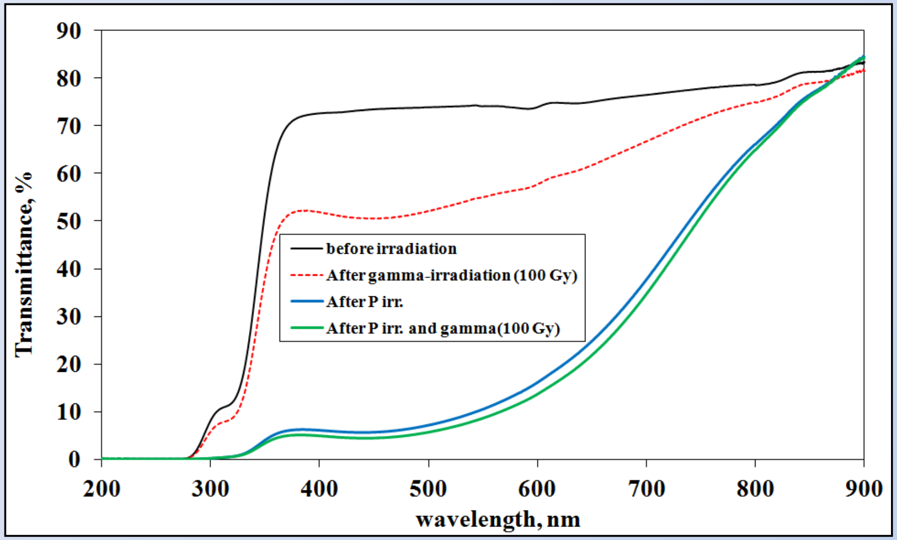
Material	ρ g/cm3	Z_{eff}	X_0 cm	λ_{max} nm	Cut-off undoped material / nm
$BaO*2SiO_2$	3.7	51	3.6	-	310
DSB: Ce	3.8	51	3.5	440-460	310
$BaO*2SiO_2$:Ce glass heavy loaded with Gd	4.7-5.4	58	2.2	440-460	318



Technology: Typical glass production technology combined with successive thermal annealing (800 – 900°C). Technological process is manageable at any glass production facility worldwide.

Samples produced in INP, Minsk in 2012-2015

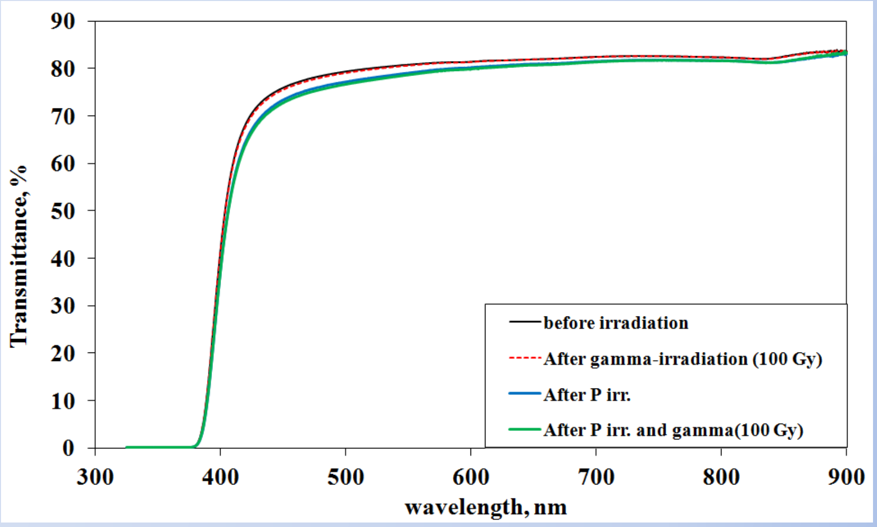
irradiation and recovery studies: with 150MeV protons



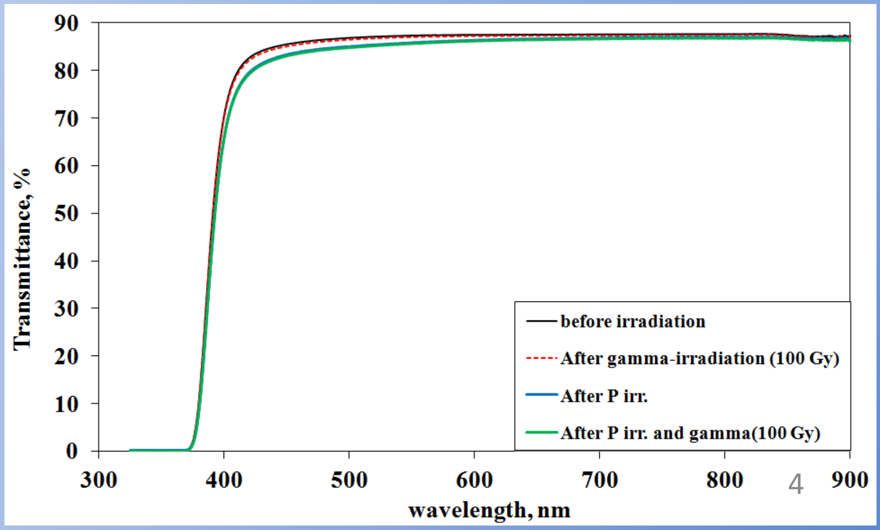
flux $\leq 2x10^{11}$ p/s cm²
integral fluence = $5x10^{13}$ p/cm²

$BaO \times 2SiO_2$
(mother glass)

$BaO \times 2SiO_2$:Ce
(without thermal treatment)

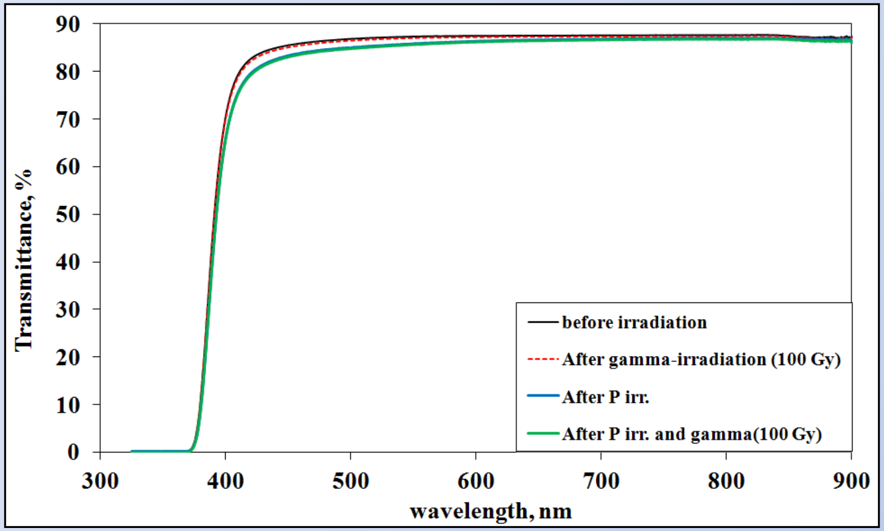


DSB:Ce
(after thermal treatment)



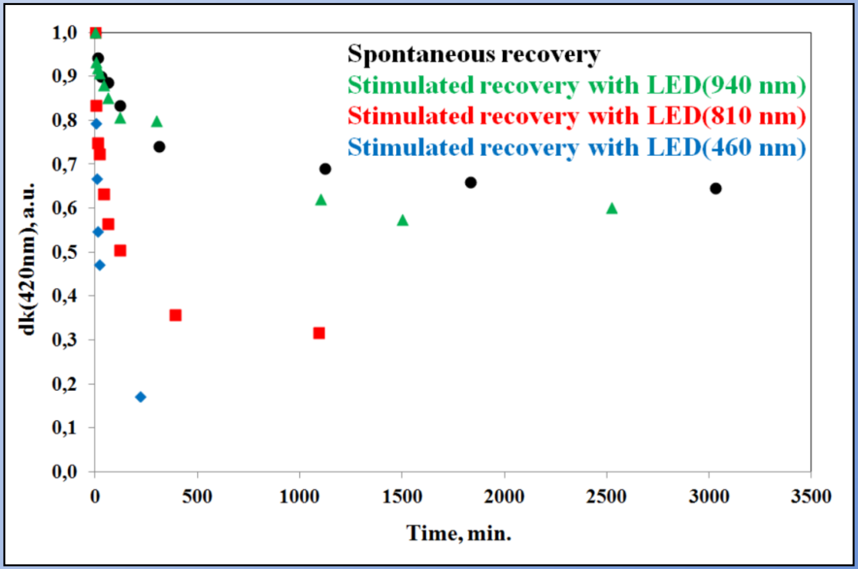
irradiation and recovery studies

DSB: Ce
(after thermal treatment)



flux ≤ $2x10^{11}$ p/s cm^2
integral fluence = $5x10^{13}$ p/cm^2

Irradiation with γ-quanta

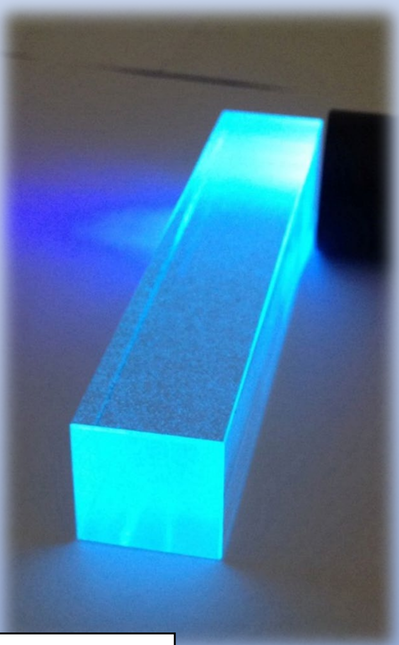


Irradiation with γ-rays and 150 MeV protons

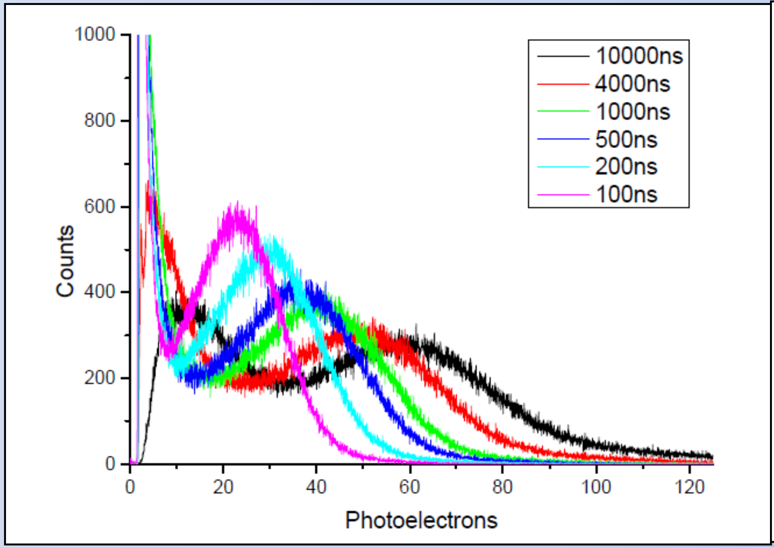
The approach developed for stimulated recovery in irradiated PWO also works for the glass family under consideration.

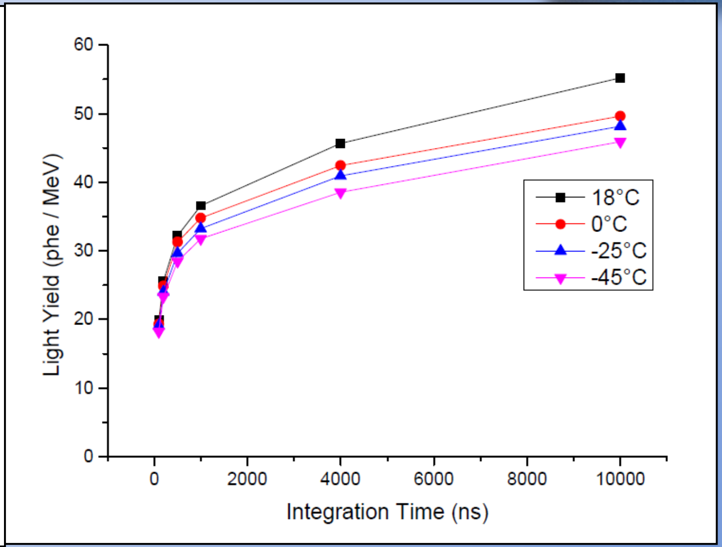
spontaneous and stimulated recovery

DSB:Ce block 23 x 23 x 120 mm^3 produced from the bulk material worked in the mold from the crucible 500 ml.



Essential problem was small bubbles inside due to an absence of agitator mechanism for glass in the crucible





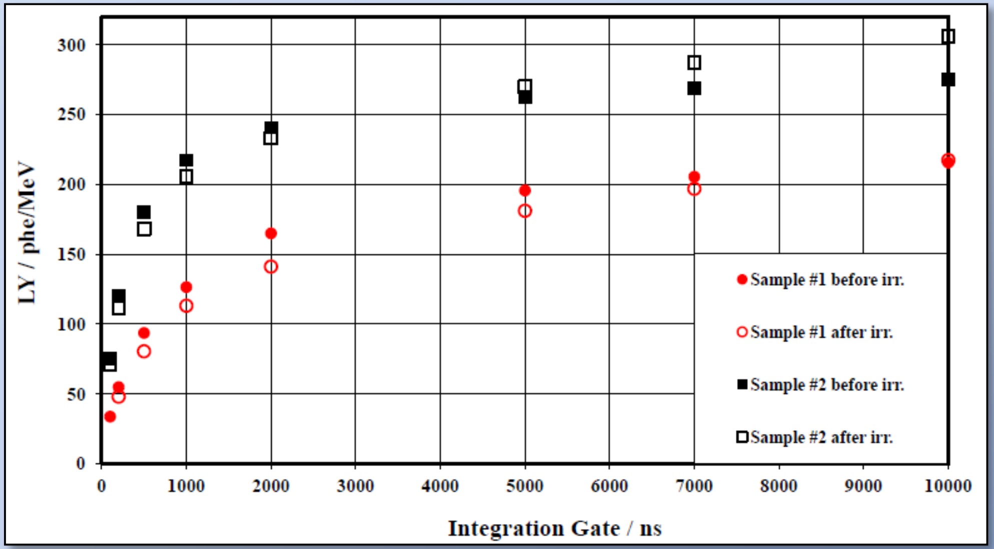
Pulse height spectra of the 662 kev (137-Cs) measured at room temperature measured in different gates and gated light yield temperature dependence

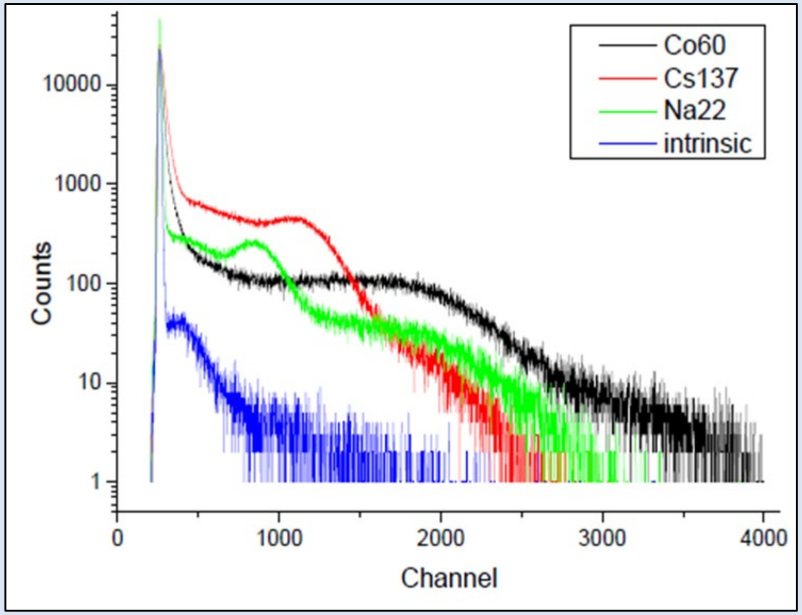
Scintillation properties of Gd-loaded samples

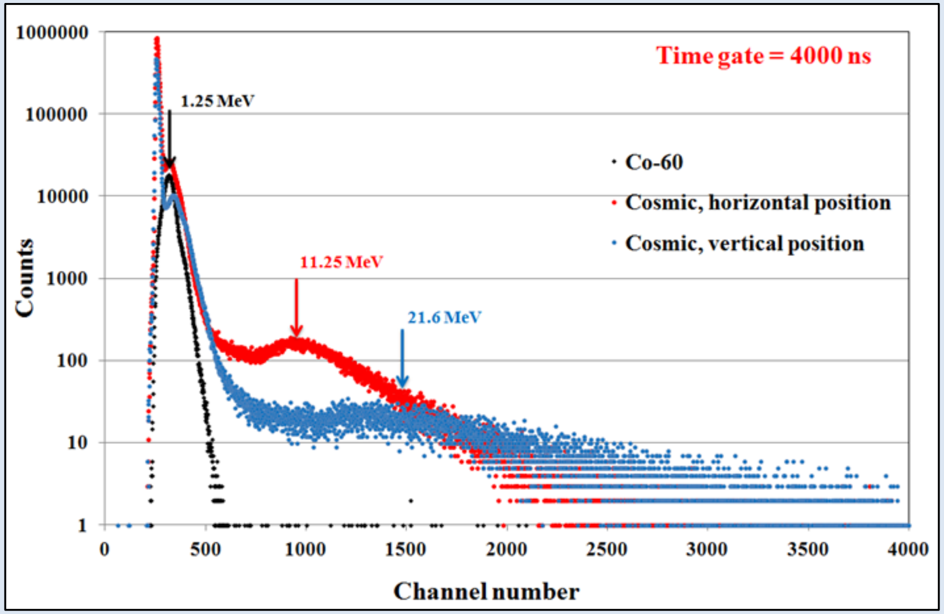
#1: 10 weight% Gd_2O_3
#2: 20 weight% Gd_2O_3
both: 0.5 weight% Ce

light yield as a function of integration time

Two samples with 5 mm thickness

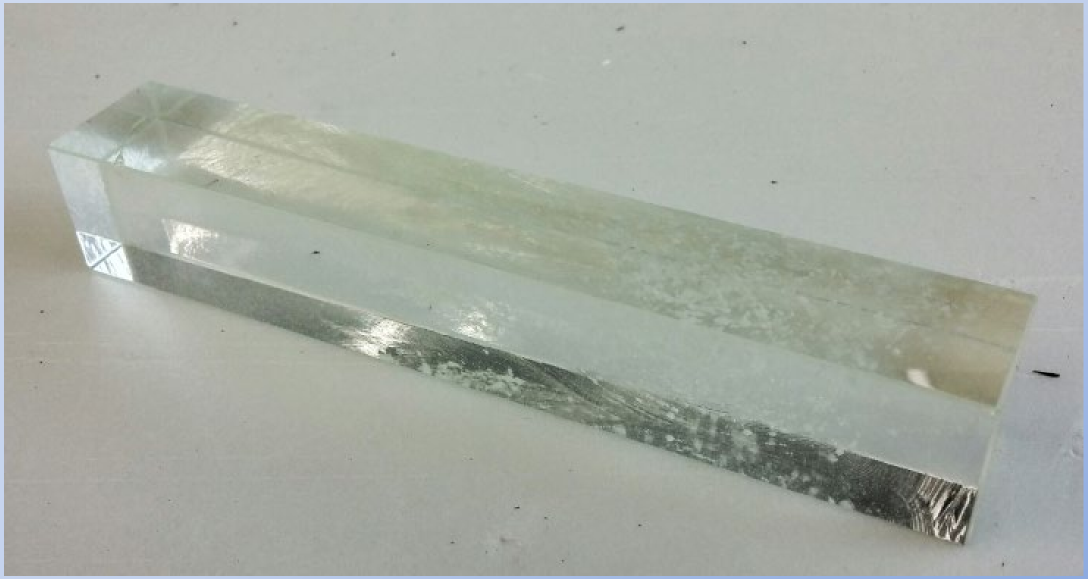






large volume samples : 25x25x125mm^3

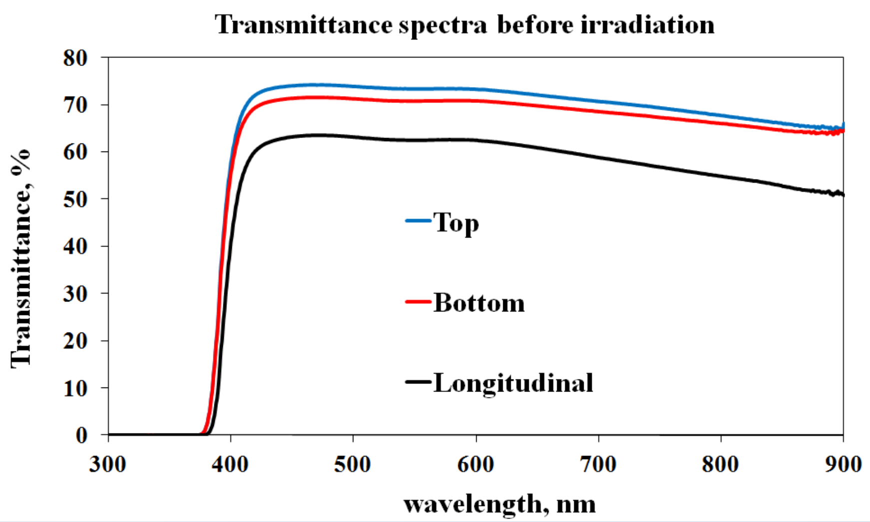
Block of DSB: Ce heavy loaded with Gd



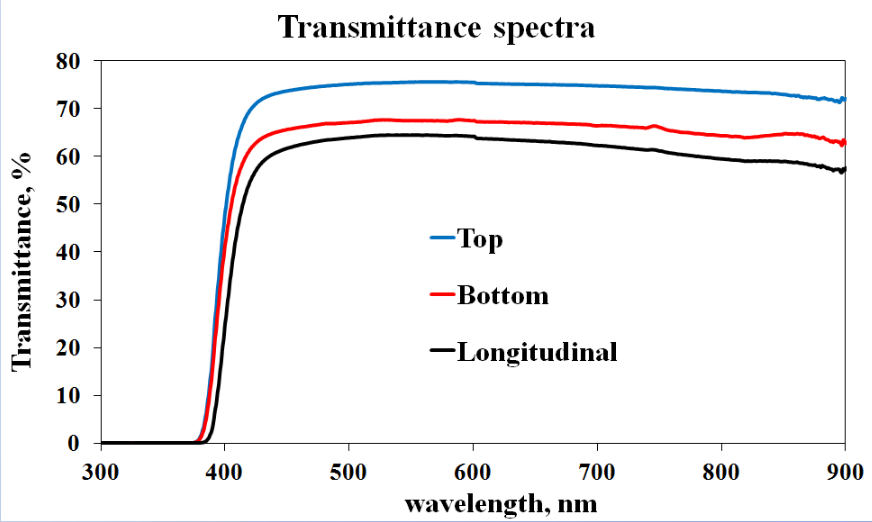
The same problem: small bubbles inside due to an absence of agitator mechanism for glass in the crucible

Irradiation tests with 190 MeV proton beam

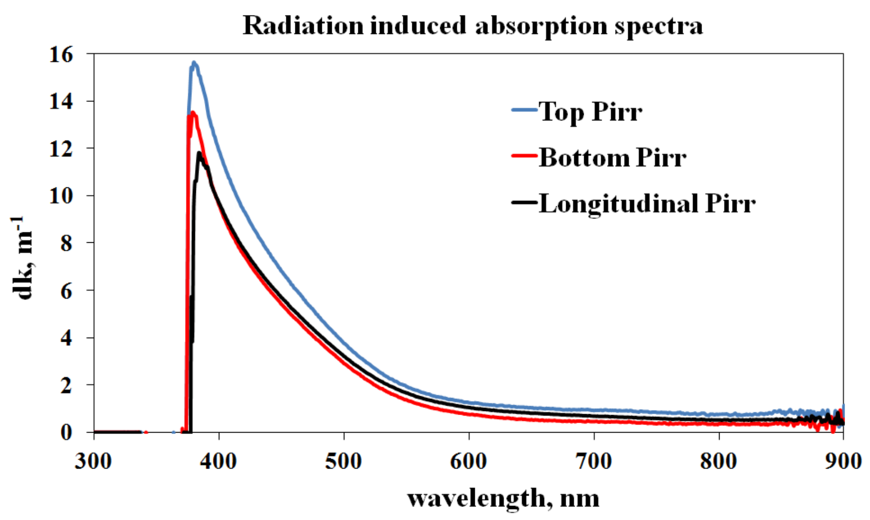
"Pure" DSB:Ce (4 cm)

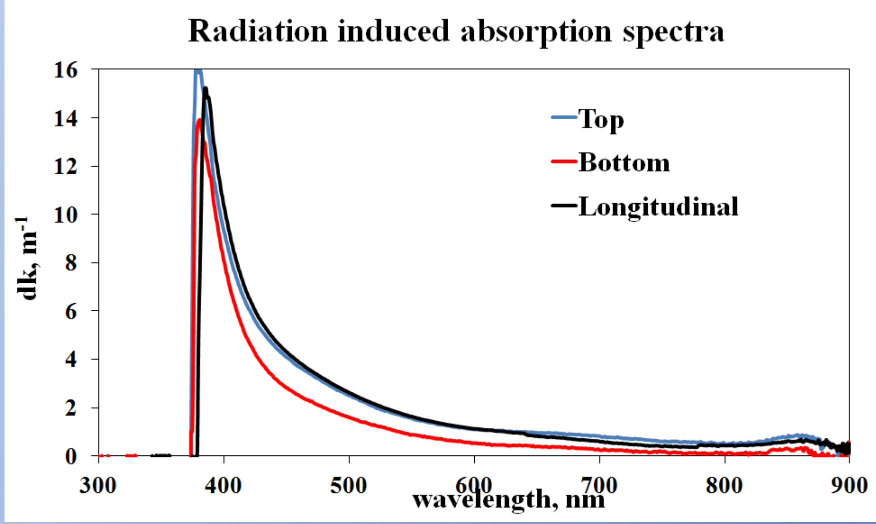


DSB:Ce loaded by Gd (4.5 cm)



Results after irradiation with fluence = 10^{12} protons/cm^2



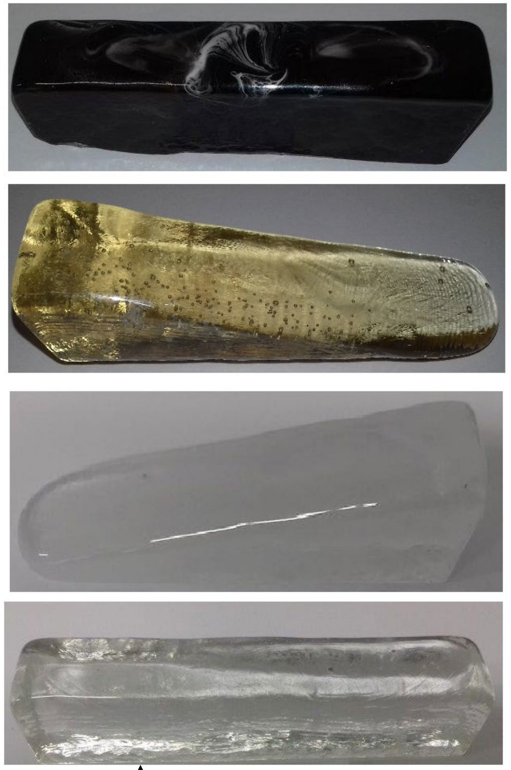


$$dk = Ln[T_{before}/T_{after}]/d$$

Next step in development with an industrial partner in frames of ATTRACT Project:

PRECIOSA

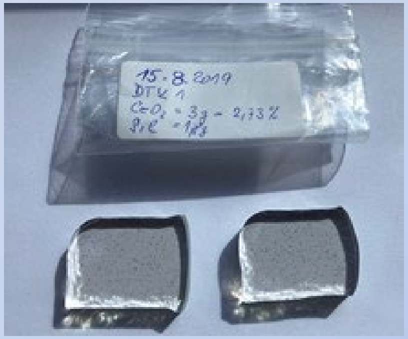
Technology evolution

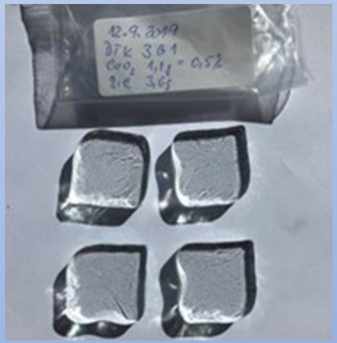


One ingot from the first probes

First ingot with reasonable Light Yield obtained in 9 month after start

www.preciosa-ornela.com
468 61 Desná, Czech Republic

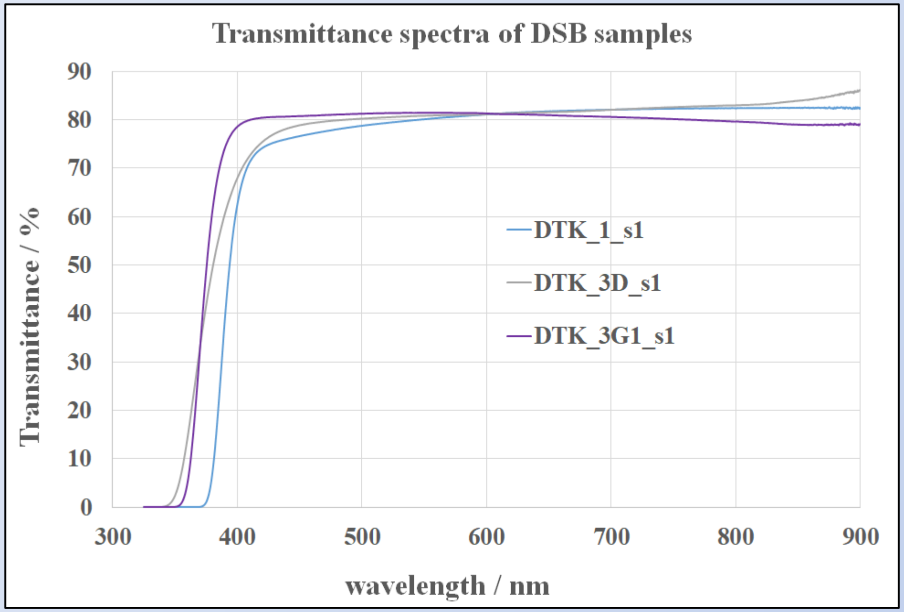


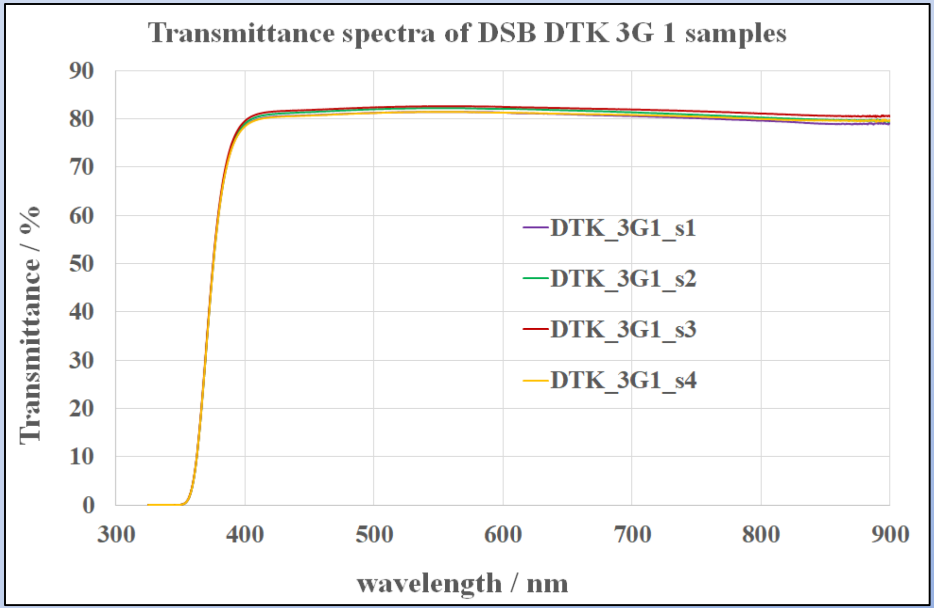


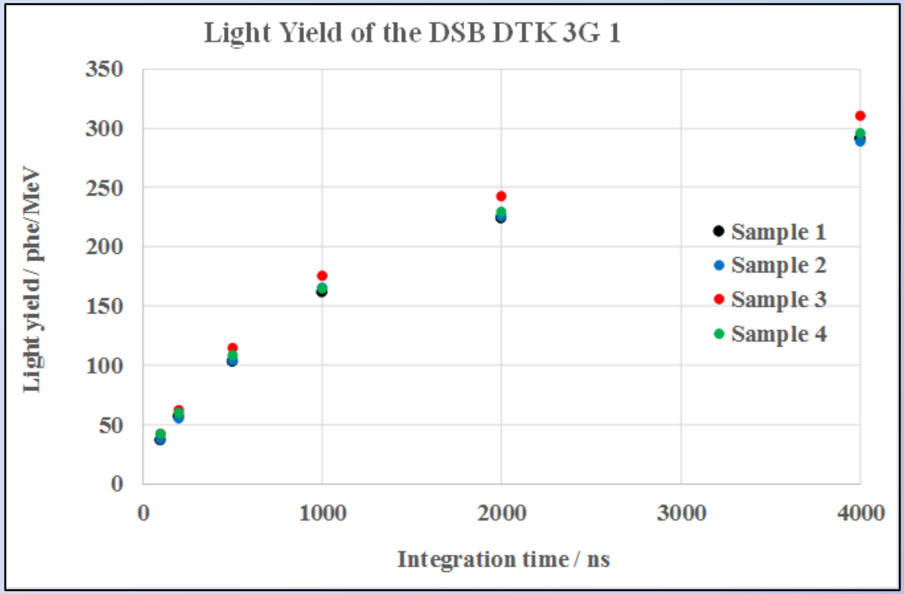
produced: Sept. 12, 2019

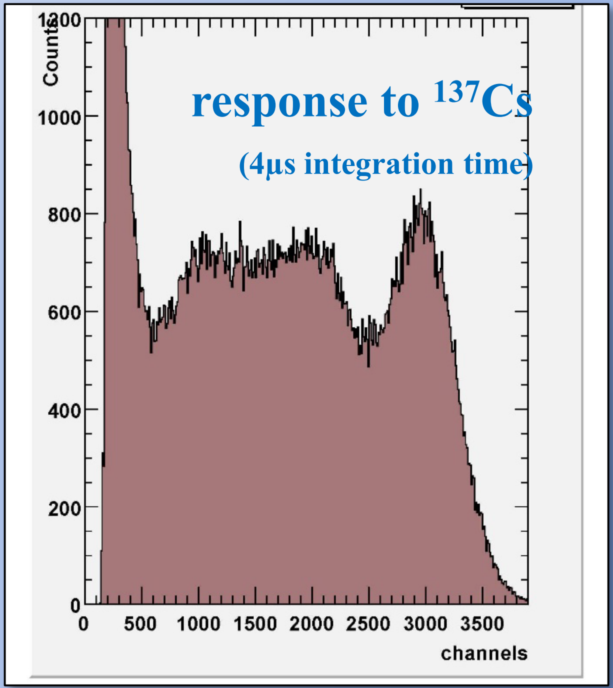
Significant reduction of the macrodefects („bubbles“) was achieved

basic properties @ start of ATTRACT



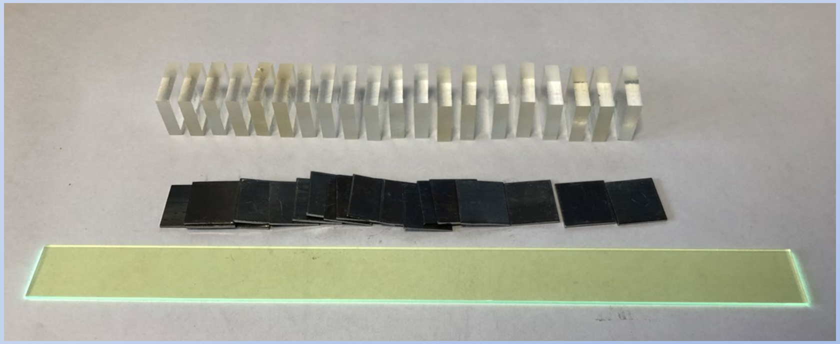




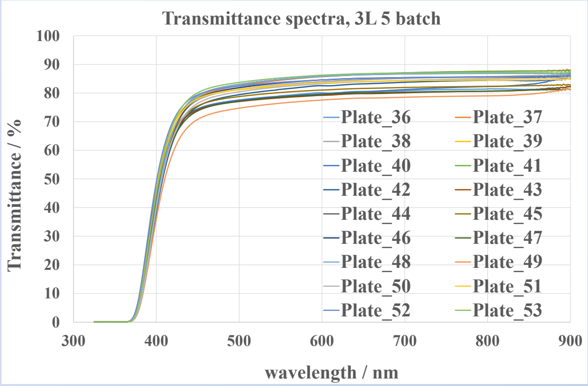


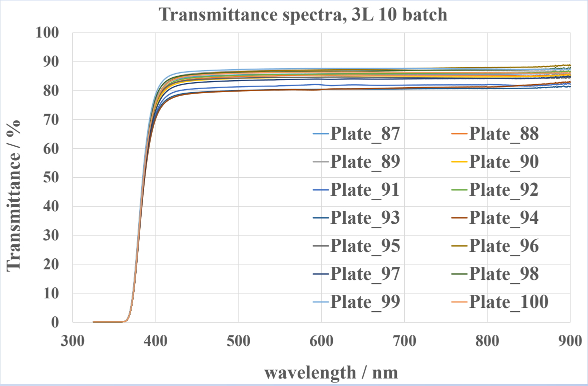
Start of the pre-production for first protypes

<u>Main task</u>: mass production of 190 plates with 17x20x5 mm dimensions to mount 3x3 sampling calorimeter array.
Each unit will consist of 20 DSB/19 Pb(1mm) plates with two WLS layers for light transportation to a photodetector.

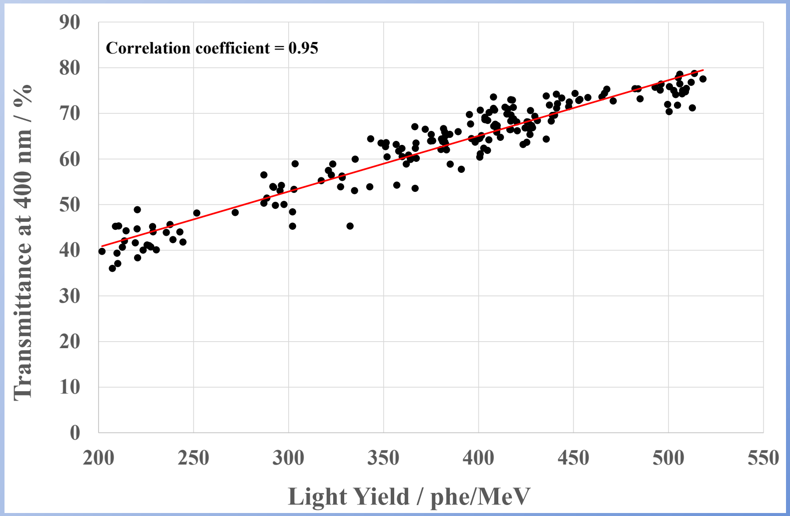


Transmittance spectra



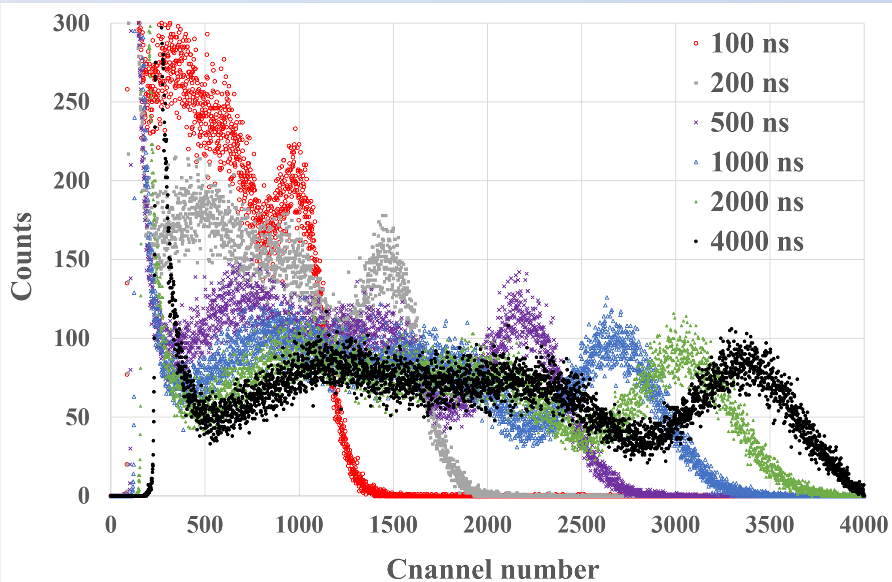


Correlation T (400 nm) vs nd the Light Yield

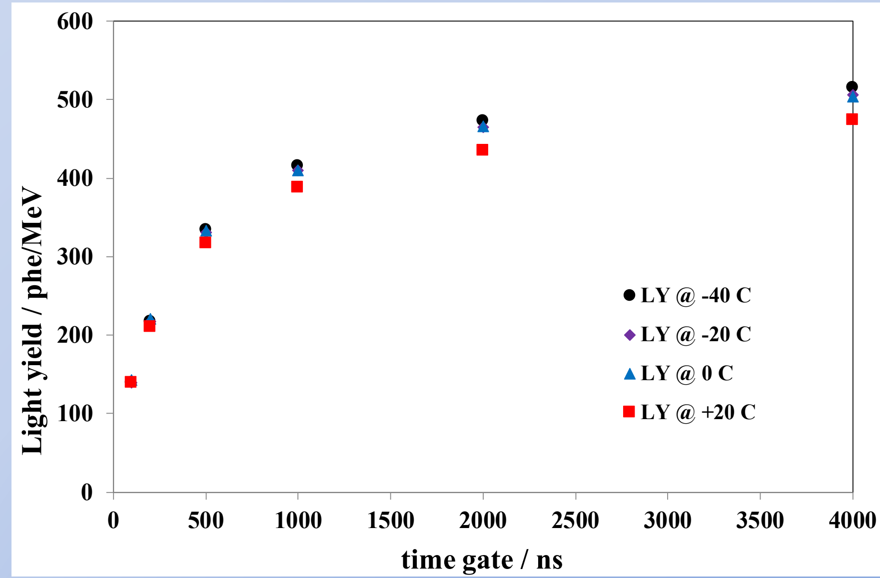


Start of pre-production for first protypes

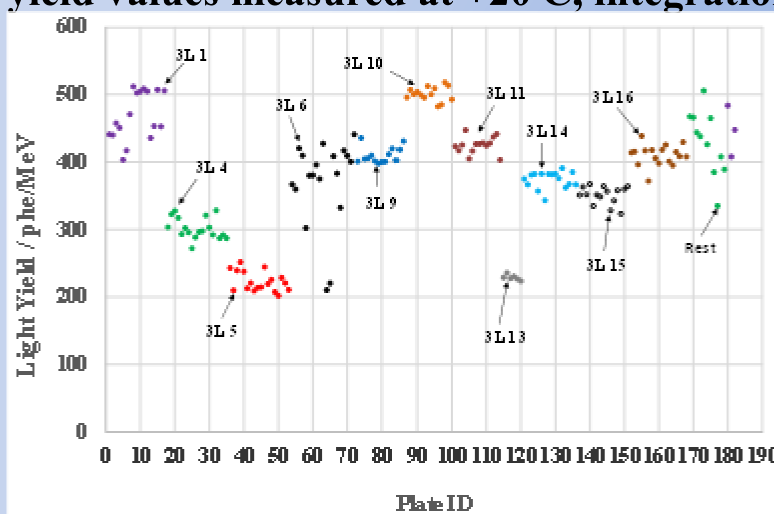
^{137}Cs spectrum measured at T= +20 C, integration time = 4 μs



Light yield vs integration time measured at different temperatures

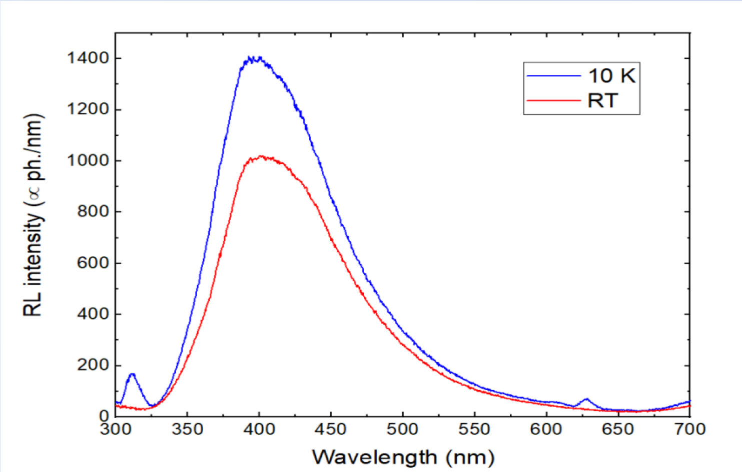


Light yield values measured at +20 C, integration time 4μs



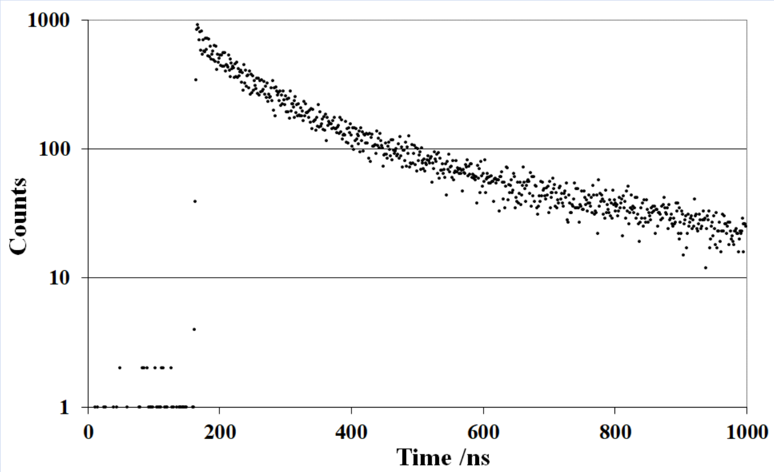
Scintillating properties characterization

Radio-luminescence spectra

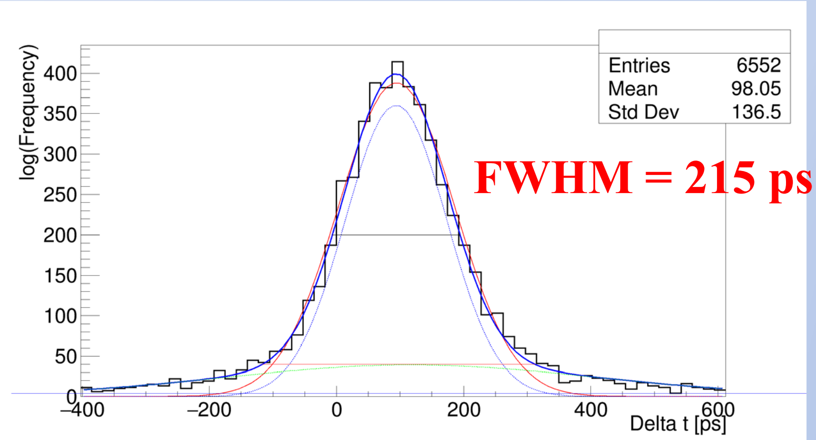


Results of Milano-Bicoca Uni

Scintillation kinetics



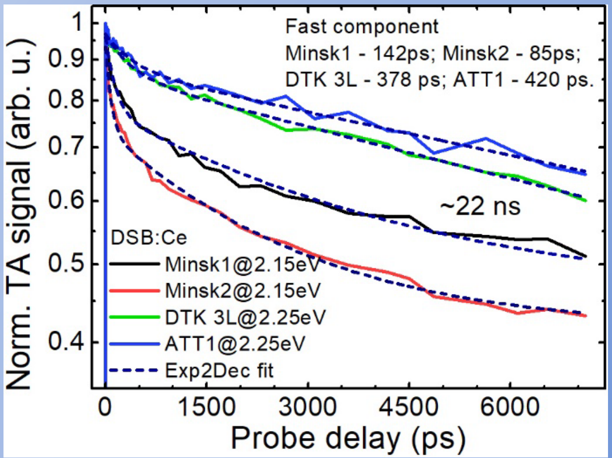
Time delay distribution between two identical channels at CTR measurement acquired at RT and 10 mV leading edge threshold



Results of CERN

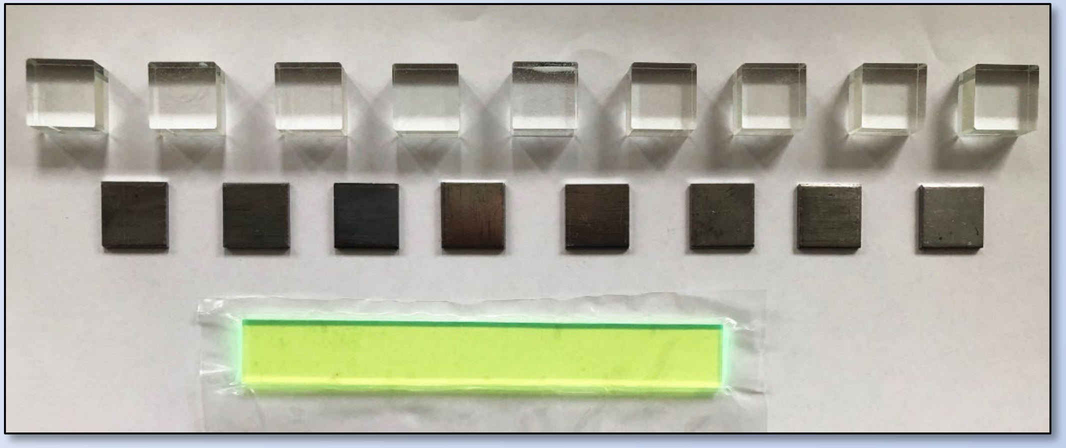
Transient absorption kinetics

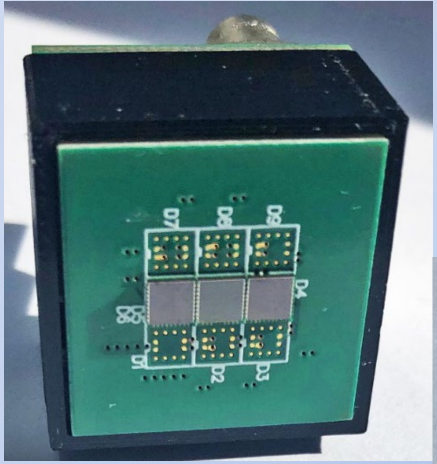
Results of Vilnius Uni



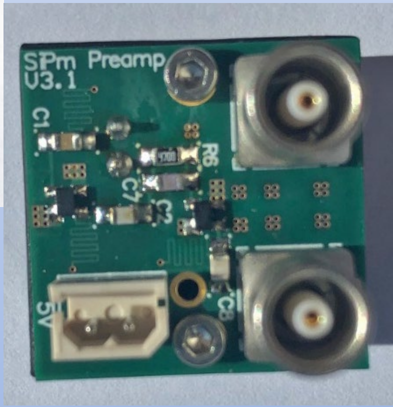
The decay time of the slow component is ~22 ns for all the samples

the first sampling prototype





photosensor
3 SiPMs

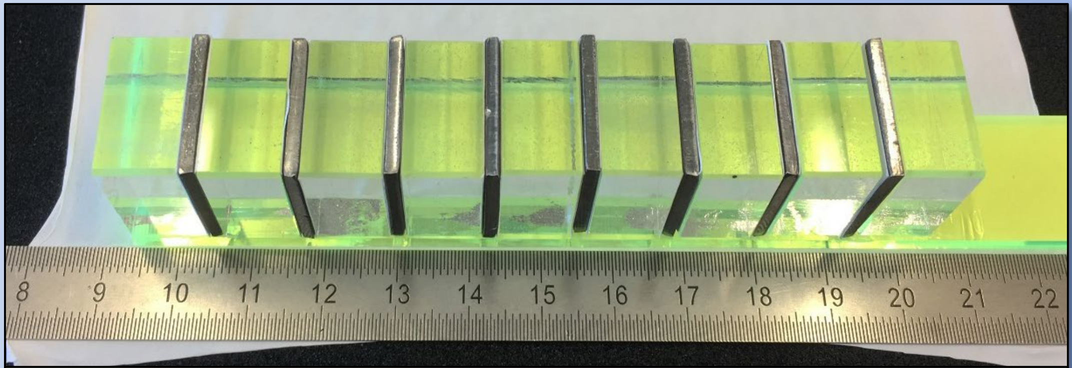


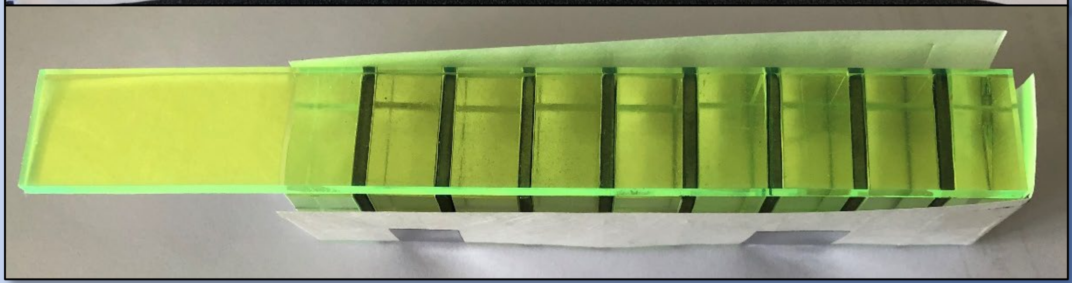
KETEK
PM3325-WB-D0
3 x 3 mm^2
25µm

9 DSB:Ce
20x20x10mm^3

WLS: EJ-280-10
Eljen
20x3x150mm^3

Pb 20x20x1mm^3

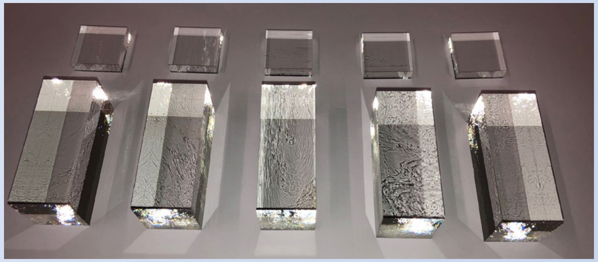


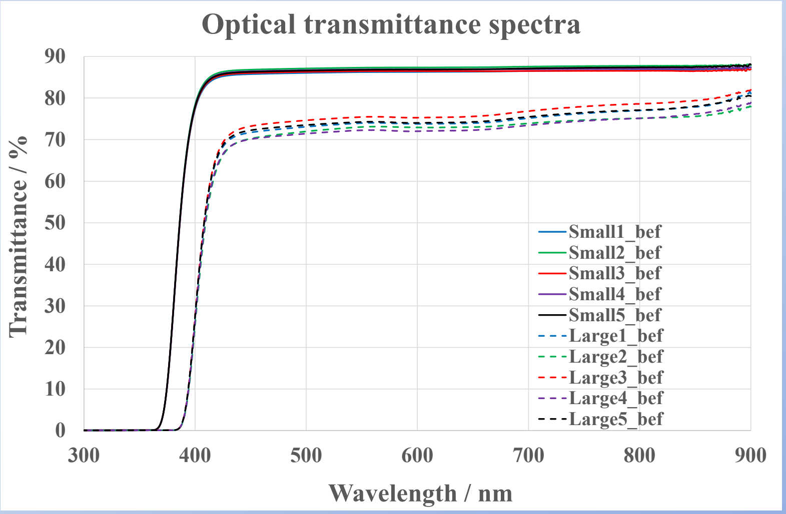


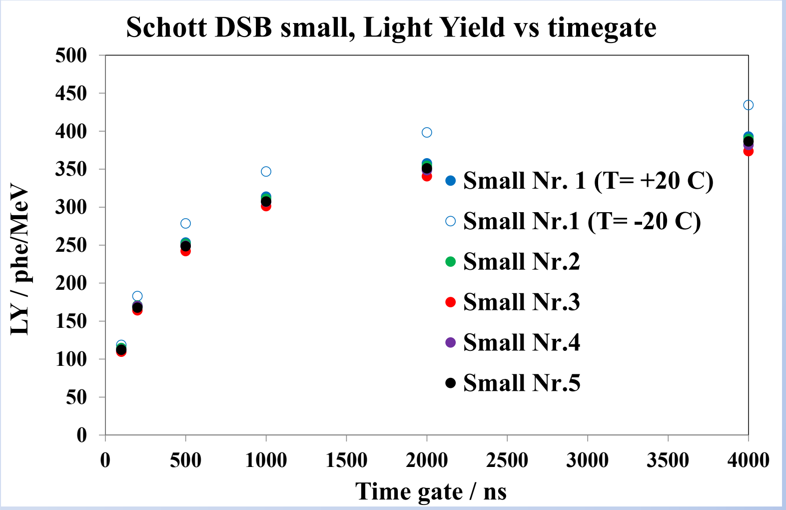
Samples produced by alternative industrial partner – Company Scott (Germany), October 2021

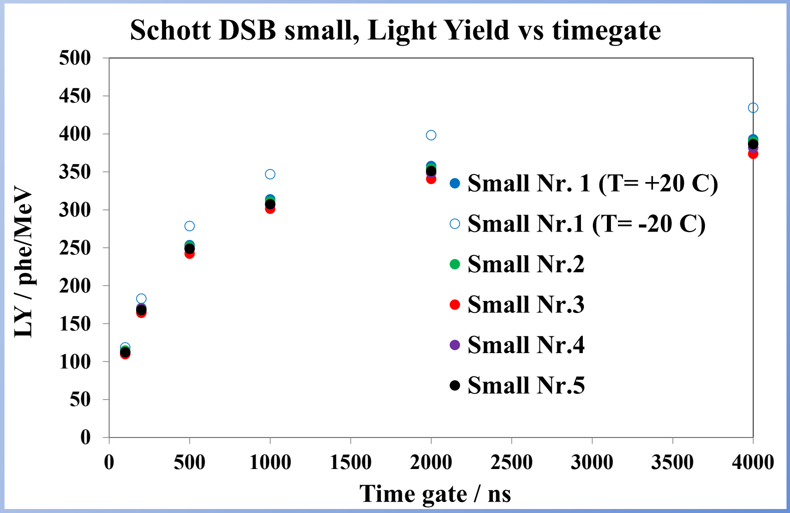
Two types of the glass materials have been delivered and tested:

- **5 samples with 20x20x5 mm^3 dimensions;**
- **5 samples with 20x20x50 mm^3 dimensions;**



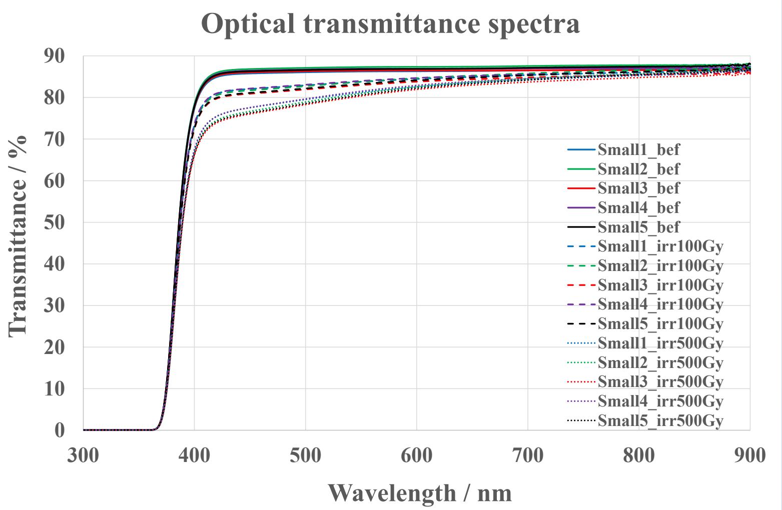


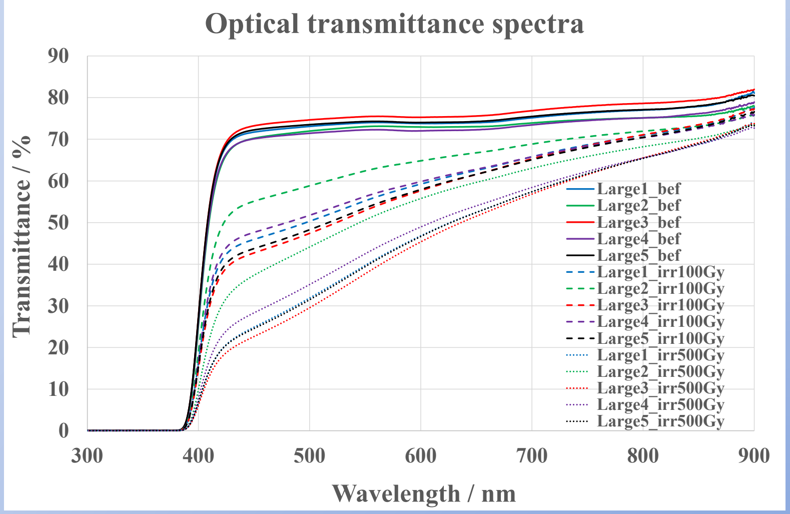


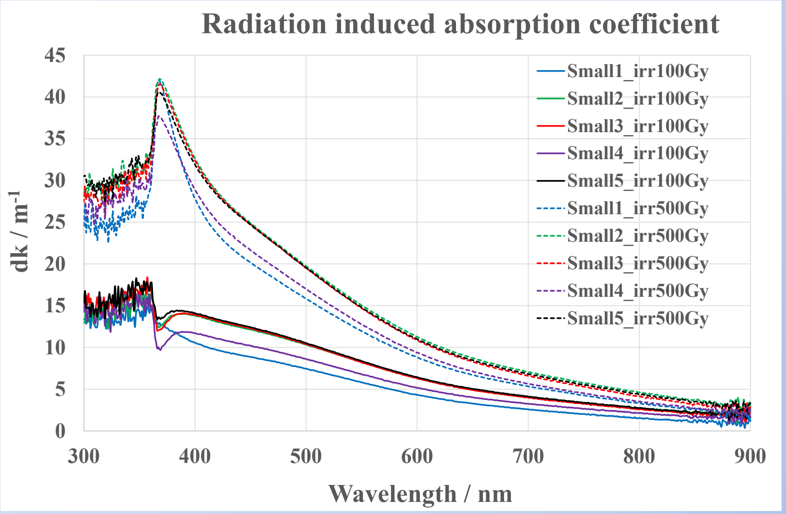


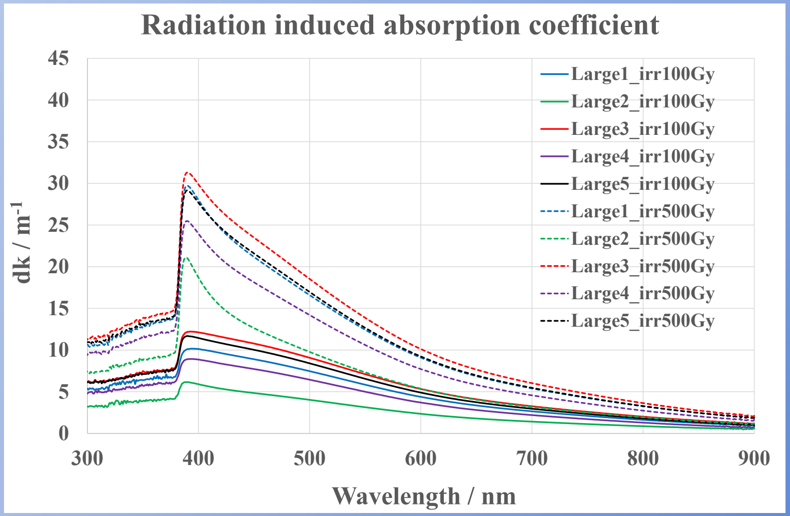
Samples produced by alternative industrial partner – Company Schott (Germany), October 2021

Irradiation with gamma-quanta ^{60}Co source, doses: 100 and 500 Gy

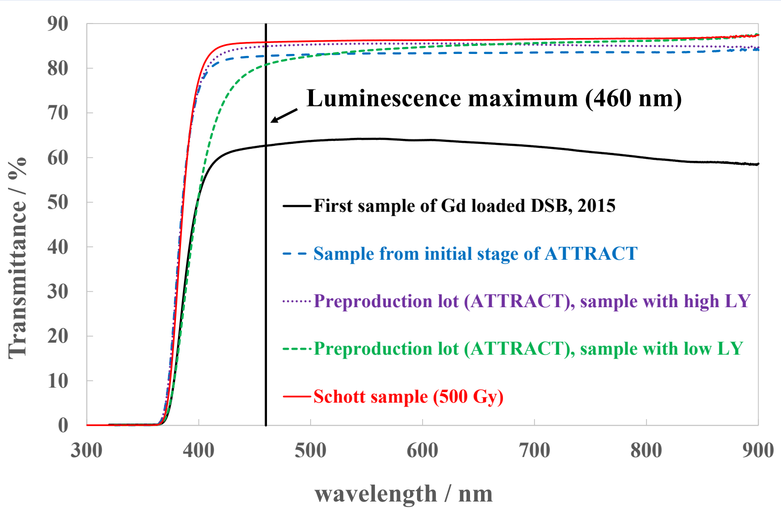


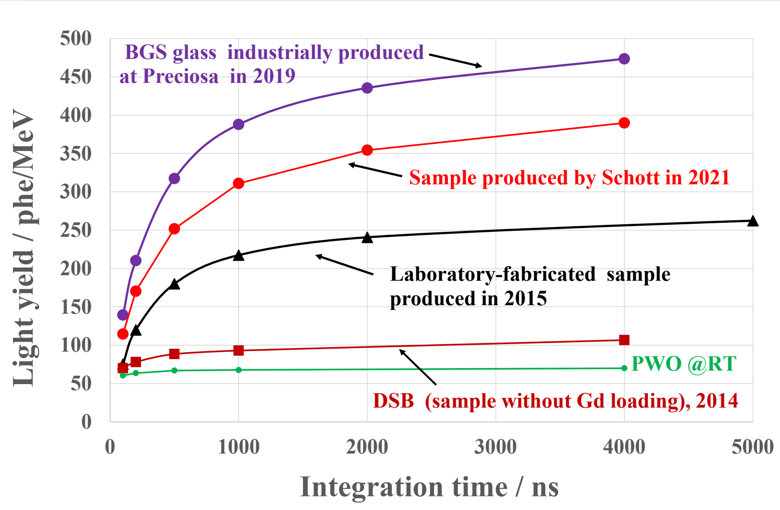


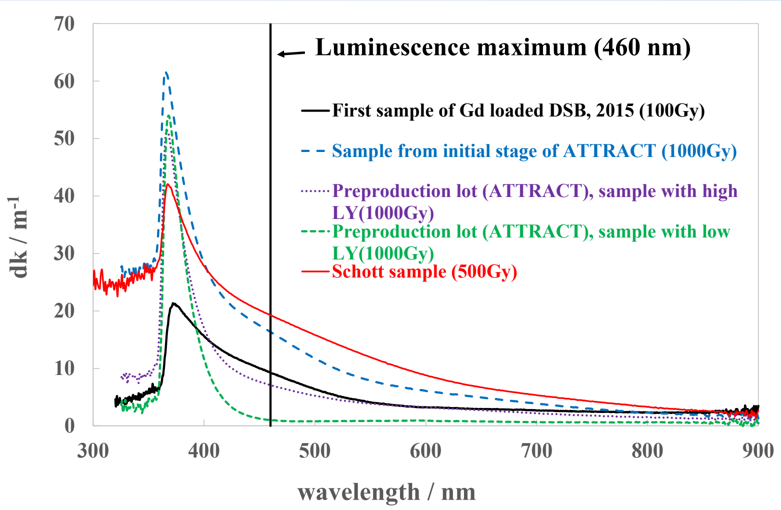


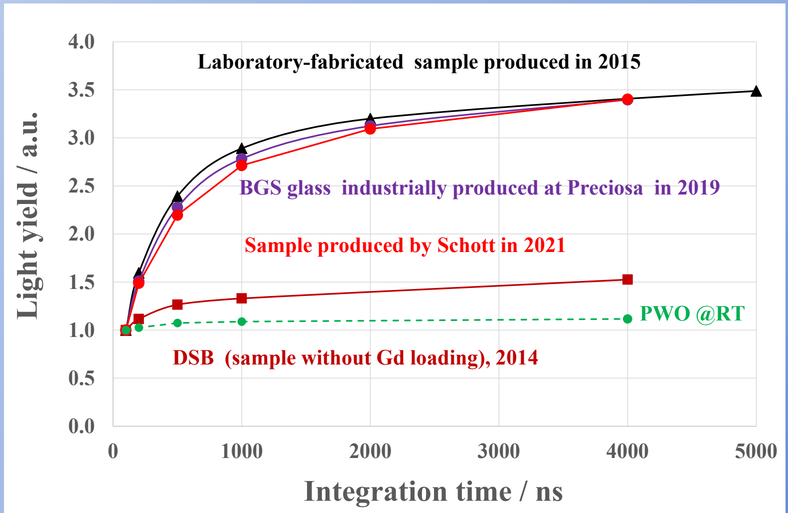


Properties evolution









$(Gd,Ce)_2O_3$-Al_2O_3-SiO_2 scintillation glass

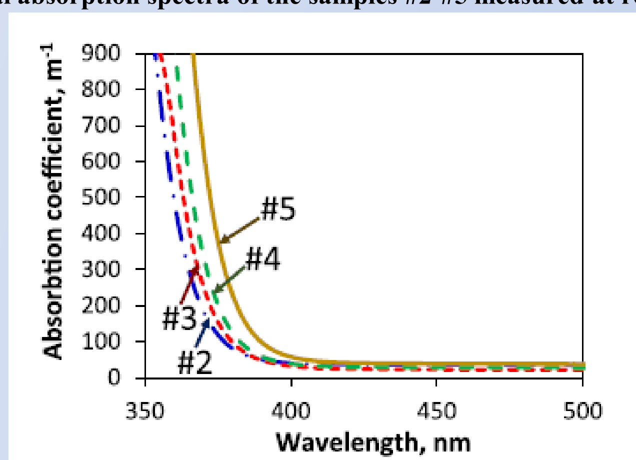
Results obtained by Kurchatov and Minsk groups. Published in Journal of non-Crystalline Solid V. 580 March 2022, https://doi.org/10.1016/j.jnoncrysol.2021.121393

Following glass composition was chosen: $2.267SiO_2$ - $0.078Si_3N_4$ - $0.6Al_2O_3$ - $0.065AlF_3$- $(Gd_{1-x}Ce_x)_2O_3$

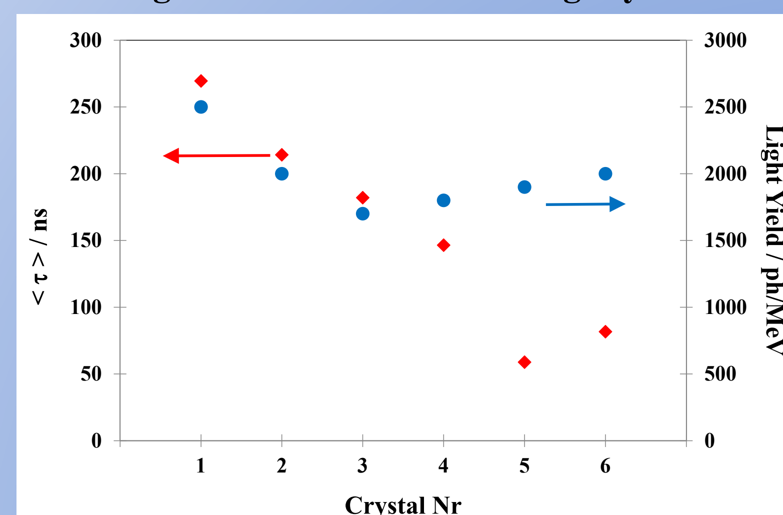
Cation composition of the glass, at. %							Kinetics parameters						Light Yield, ph/MeV	
Sample Nr.	SiO2	AlO1.5	AlF3	CeO1.5	GdO1.5	BaO	t1	f1	t2	f2	t3	f3		
1	54.82	-	2.45	1.32	22.72	18.7	-	-	90.8	45	415.6	55	2500	with Ba
2	48.38	11.12	1.26	2.16	37.09	-	-	-	98.9	58.88	379.2	41.12	2000	w/o Ba
3	48.11	11.75	1.24	3.25	35.65	-	-	-	96.6	61.32	317.5	38.68	1700	
4	48.41	11.62	1.25	4.84	33.89	-	43.3	41.5	92	28.46	340.7	30.04	1800	
5	48.41	11.62	1.25	5.81	32.92	-	40.9	64.05	90.8	35.95	-	-	1900	
6	48.41	11.62	1.25	5.81	32.92	-	30.25	19.43	64.5	60.85	185.1	19.72	2000	

The glass density of all the samples was measured to be 4.5 ± 0.1 g/cm3, which provides an effective charge Z_{eff}=52 and puts it in the line for the stopping power to ionizing radiation with alkali-halide and some oxide crystalline materials of the garnet and perovskite structure.

Optical absorption spectra of the samples #2-#5 measured at room temperature



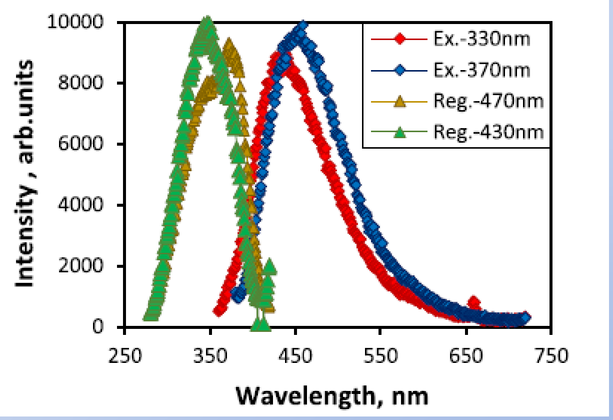
Average scintillation time and light yield

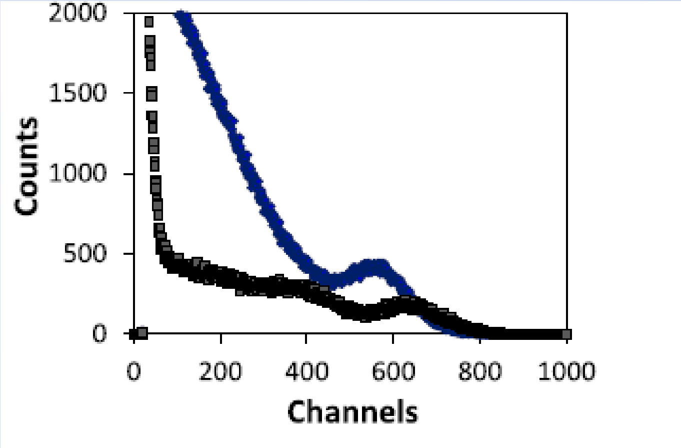


Acknowledgments to Prof. Mikhail Korjik (INP BSU, Minsk)

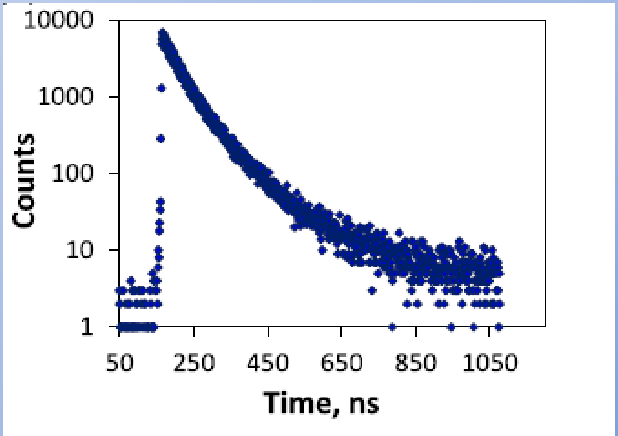
Ba-less $(Gd,Ce)_2O_3$-Al_2O_3-SiO_2 scintillation glass

Photoluminescence excitation (λ_{em} = 430, 470 nm) and photoluminescence (λex = 330, 370 nm) of Gd-Si-Al: Ce glass





Pulse height spectra under 137Cs (662 keV) source of the Gd-Si-Al: Ce glass sample #6, with dimensions 15×15×3 mm3 (◆) and Ba-Gd-Si glass sample with dimensions 20×20×5 mm3 (■)



Scintillation kinetics of the sample #6 measured at room temperature.

New scintillation Ce^{3+} -doped Gd2O3-Al2O3-SiO2 glass has demonstrated scintillation kinetics with an average decay time 60 ns and a light yield ~2000 photon/MeV.

A Amelina, A Mikhalin, S Belus, A Bondarev et al, Journal of Non-Crystalline Solids 2022, 580, 121393, /doi.org/10.1016/j.jnoncrysol.2021.121393

Cost consideration of the ”non-human” drivers

Cost driver	Examples of the prices	Comment
Raw material	Gd oxide ~50 usd/kg S- oxide –less than 50 usd/kg Ba oxide- less than 30 usd/kg Al oxide-less than 10 usd/kg	All oxides of needed purity are commercially available
Crucible material	Not necessary Pt, Carbon glass can be a good alternative	Cheaper than Pt by factor 10
Energy	Low energy consuming process per kg of the glass mass produced	High rate of the commercial glass mass production
Mechanical treatment	Lower than for single crystals	Can be worked in the mold. So, the waste at mechanical treatment may be minimised

Conclusions

- **The gadolinium based glass materials are relatively cheap and can be produced at many glass-working facilities. Properties can be adopted to application conditions. It has 3-4 times higher density in comparison to plastic scintillators allowing variable scintillator shapes**
- **Significant progress in technology optimization of the glass scintillation material was achieved**
- **The Gd loaded DSB glass has higher density in comparison with barium-silica DSB glass.**
- **Light yield value was increased due to further technology optimization:**

  2500 ph/MeV(with Gd) vs 500 ph/MeV (without Gd)
- **Further technology optimization showed possible improvement of the timing characteristics of the material without significant degradation of the light yield:**

  $< \tau > = 60$ ns, LY = 2000 ph/MeV
- **High Gd contamination opens up an opportunity for neutron detection as well**