



Update on Inorganic Scintillator Development

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Fast & Radiation Hard Scintillators



- Supported by the DOE ADR program we are developing fast and radiation hard scintillators to face the challenge for future HEP experiments at the energy and intensity frontiers.
- **Fast and radiation hard scintillators at HL-LHC with 3000 fb⁻¹ will face the following radiation:**
 - **Absorbed dose: up to 100 Mrad,**
 - **Charged hadron fluence: up to 6×10^{14} p/cm²,**
 - **Fast neutron fluence: up to 3×10^{15} n/cm².**
- **Ultra-fast scintillators with excellent radiation hardness is also needed to face the challenge of unprecedented event rate expected at future HEP experiments at the intensity frontier, such as Mu2e-II, and the GHz X-ray imaging for the proposed Marie project at. Y:BaF₂ with sub-ns decay time and suppressed slow scintillation component is a leading candidate for both applications.**



Fast Inorganic Scintillators (I)



	LYSO:Ce	LSO:Ce, Ca ^[1]	LuAG:Ce _[2]	LuAG:Pr ^[3]	GGAG:Ce ^[4,5]	CsI	BaF ₂ ^[6]	BaF ₂ :Y	CeBr ₃	LaBr ₃ :Ce ^[7]
Density (g/cm ³)	7.4	7.4	6.76	6.76	6.5	4.51	4.89	4.89	5.23	5.29
Melting points (°C)	2050	2050	2060	2060	1850 ^d	621	1280	1280	722	783
X ₀ (cm)	1.14	1.14	1.45	1.45	1.63	1.86	2.03	2.03	1.96	1.88
R _M (cm)	2.07	2.07	2.15	2.15	2.20	3.57	3.1	3.1	2.97	2.85
λ _i (cm)	20.9	20.9	20.6	20.6	21.5	39.3	30.7	30.7	31.5	30.4
Z _{eff}	64.8	64.8	60.3	60.3	51.8	54.0	51.6	51.6	45.6	45.6
dE/dX (MeV/cm)	9.55	9.55	9.22	9.22	8.96	5.56	6.52	6.52	6.65	6.90
λ _{peak} ^a (nm)	420	420	520	310	540	310	300 220	300 220	371	360
PL Emission Peak (nm)	402	402	500	308	540	310	300 220	300 220	350	360
PL Excitation Peak (nm)	358	358	450	275	445	256	<200	<200	330	295
Absorption Edge (nm)	170	170	160	160	190	200	140	140	n.r.	220
Refractive Index ^b	1.82	1.82	1.84	1.84	1.92	1.95	1.50	1.50	1.9	1.9
Normalized Light Yield ^{a,c}	100	116 ^e	35 ^f 48 ^f	44 41	40 75	4.2 1.3	42 5.0	1.7 5.0	99	153
Total Light yield (ph/MeV)	30,000	34,800 ^e	25,000 ^f	25,800	34,700	1,700	13,000	2,100	30,000	46,000
Decay time ^a (ns)	40	31 ^e	981 ^f 64 ^f	1208 26	319 101	30 6	600 0.6	600 0.6	17	20
Light Yield in 1 st ns (photons/MeV)	740	950	240	520	260	100	1200	1200	1,700	2,200
Issues					neutron x-section	Slightly hygroscopic	Slow component	DUV PD	hygroscopic	



Fast Inorganic Scintillators (II)



- a. Top line: slow component, bottom line: fast component;**
- b. At the wavelength of the emission maximum;**
- c. Excited by Gamma rays;**
- d. For $\text{Gd}_3\text{Ga}_3\text{Al}_2\text{O}_{12}:\text{Ce}$**
- e. For 0.4 at% Ca co-doping**
- f. Ceramic with 0.3 Mg at% co-doping**

[1] Spurrier, et al., *IEEE T. Nucl. Sci.* 2008,55 (3): 1178-1182

[2] Liu, et al., *Adv. Opt. Mater.* 2016, 4(5): 731–739

[3] Hu, et al., *Phys. Rev. Applied* 2016, 6: 064026

[4] Lucchini, et al., *NIM A* 2016, 816: 176-183

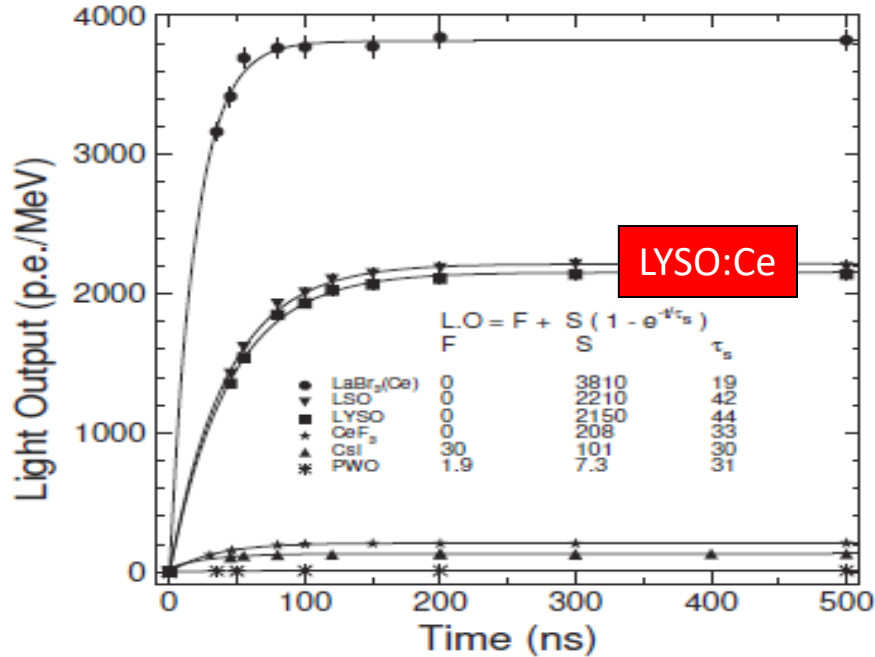
[5] Meng, et al., *Mat. Sci. Eng. B-Solid* 2015, 193: 20-26

[6] Diehl, et al., *J. Phys. Conf. Ser* 2015, 587: 012044

[7] Pustovarov, et al., *Tech. Phys. Lett.* 2012, 784-788



LYSO:Ce and LSO:Ce,Ca Crystals

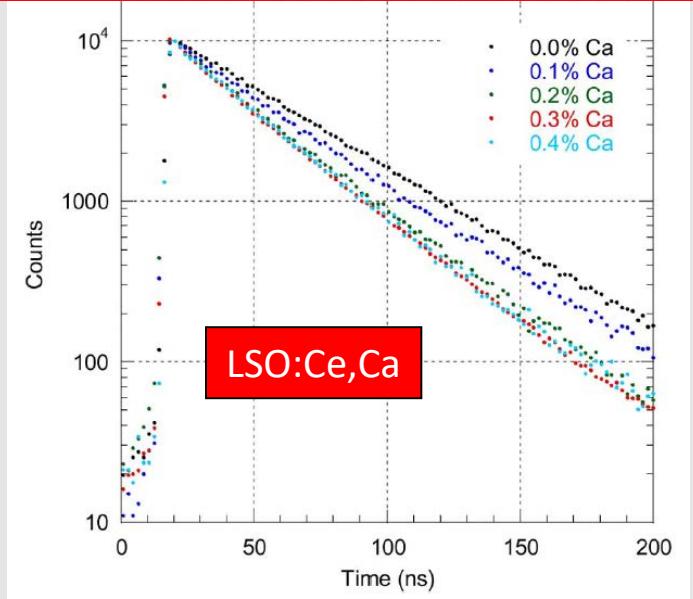


Light output measured by using a XP2254b PMT is shown as a function of integration time for six fast crystal scintillators^[1].

References.:

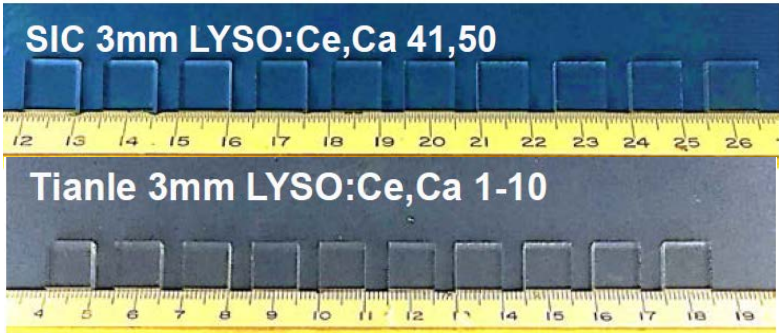
- [1] R. Y. Zhu, *Phys. Proc*, 37 2012: 372-383.
- [2] Merry A. Spurrier, et al., *IEEE TNS*,55 (3) 2008: 1178-1182.

Ca²⁺ co-doping increase the light output in 1st ns



Scintillation decay of LSO:1at%Ce crystals with 0, 0.1, 0.2, 0.3, and 0.4% Ca doping^[1]

Properties of LSO:0.1at%Ce,Ca crystals^[1]

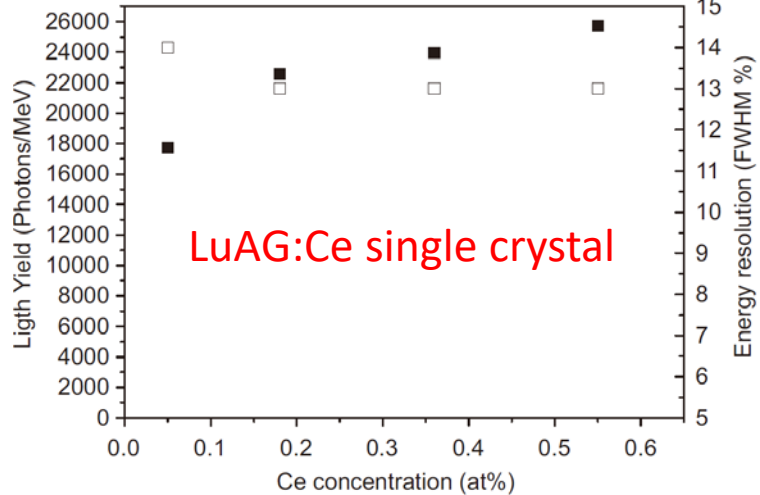
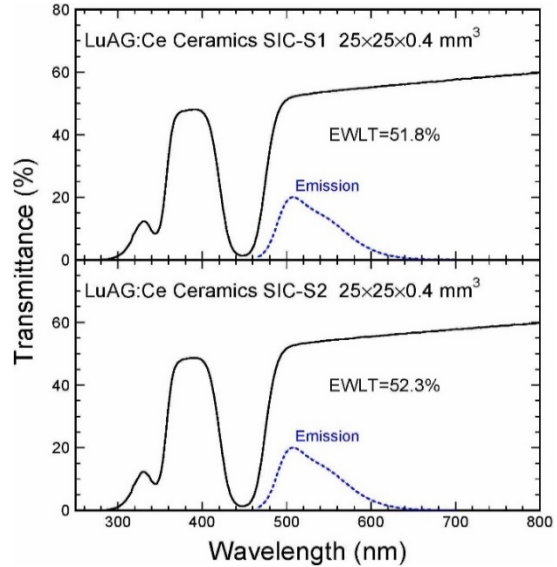
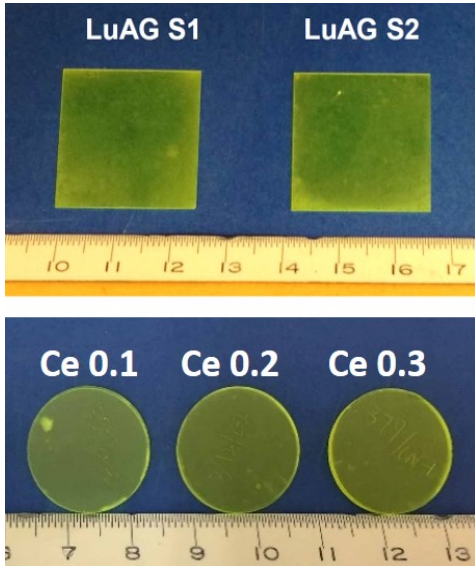


Ca concentration (%)	Light output (photons/MeV)	Decay time (ns)
0.0	30900	43.0
0.1	38800	36.7
0.2	36200	33.3
0.3	32400	31.3
0.4	34800	31.0

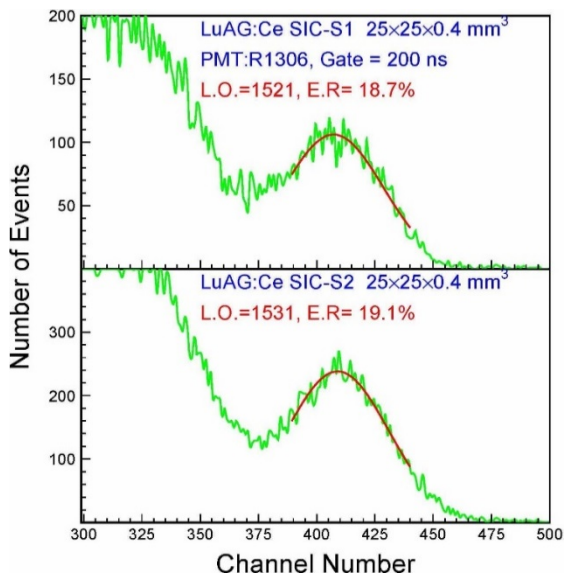
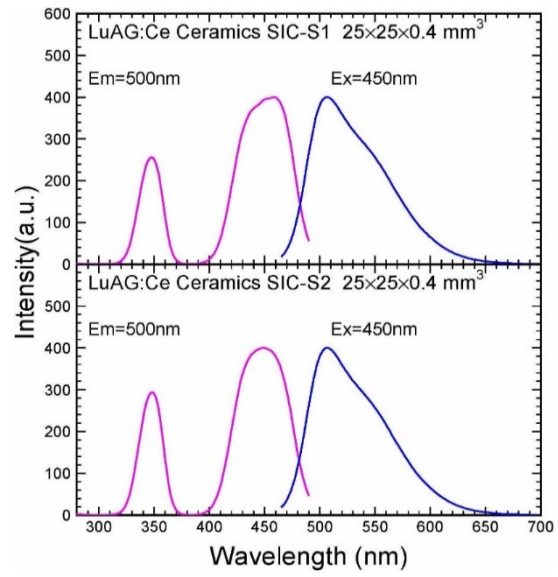
Light output and decay time values are the average of multiple samples.



LuAG:Ce Ceramic Samples



[1] A.G. Petrosyan et al., *J. Cryst. Growth*, 312 (2010) 3136–3142



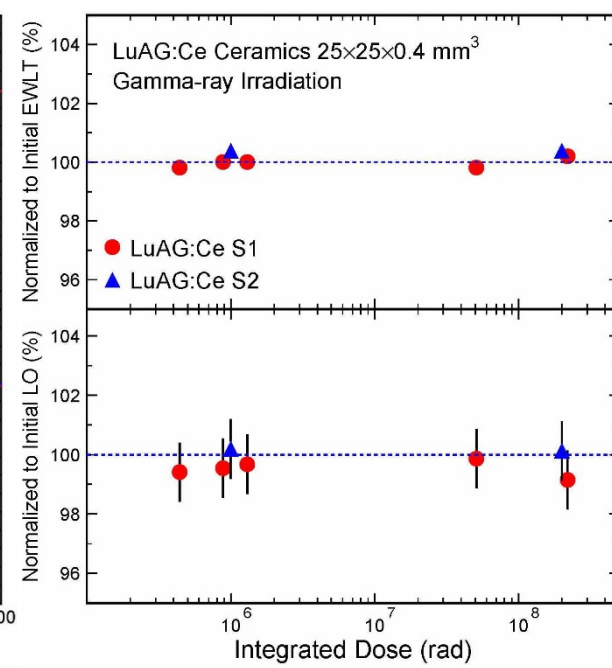
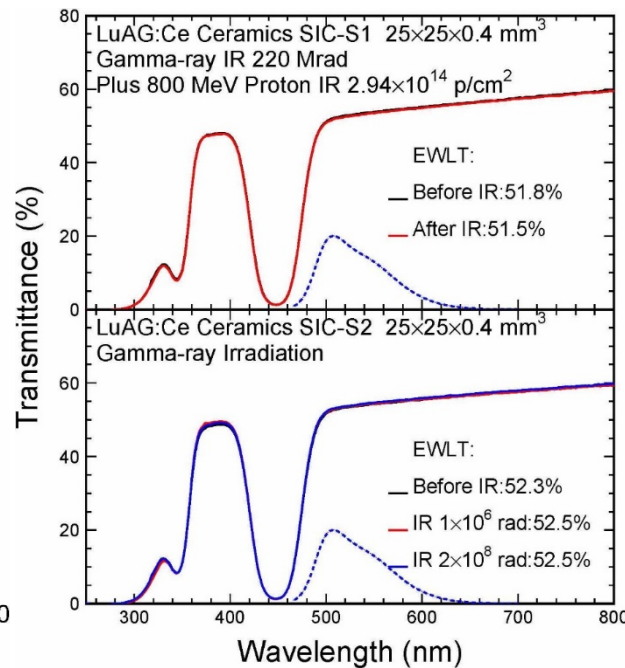
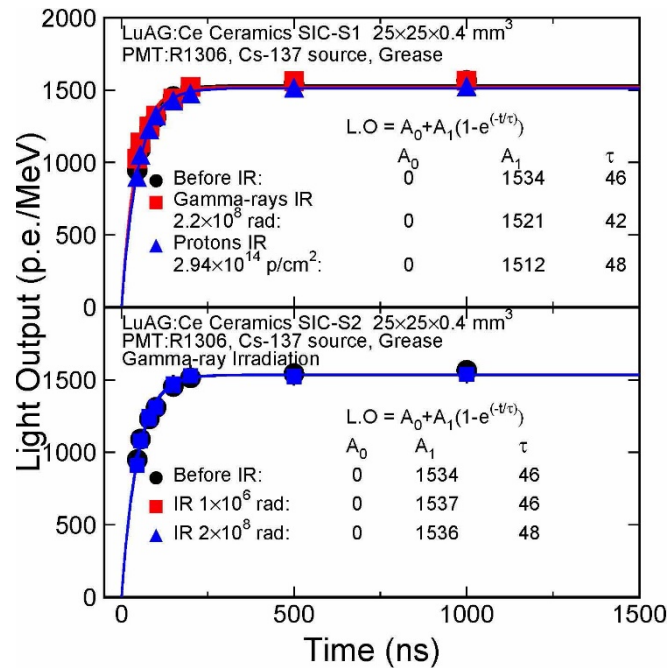
LuAG ceramic scintillators with both optical and scintillation performance matching their single crystal counterpart are investigated at Caltech.



Excellent Radiation Hardness



No damage observed in both transmittance and light output after 220 Mrad ionization dose and 3×10^{14} p/cm² of 800 MeV



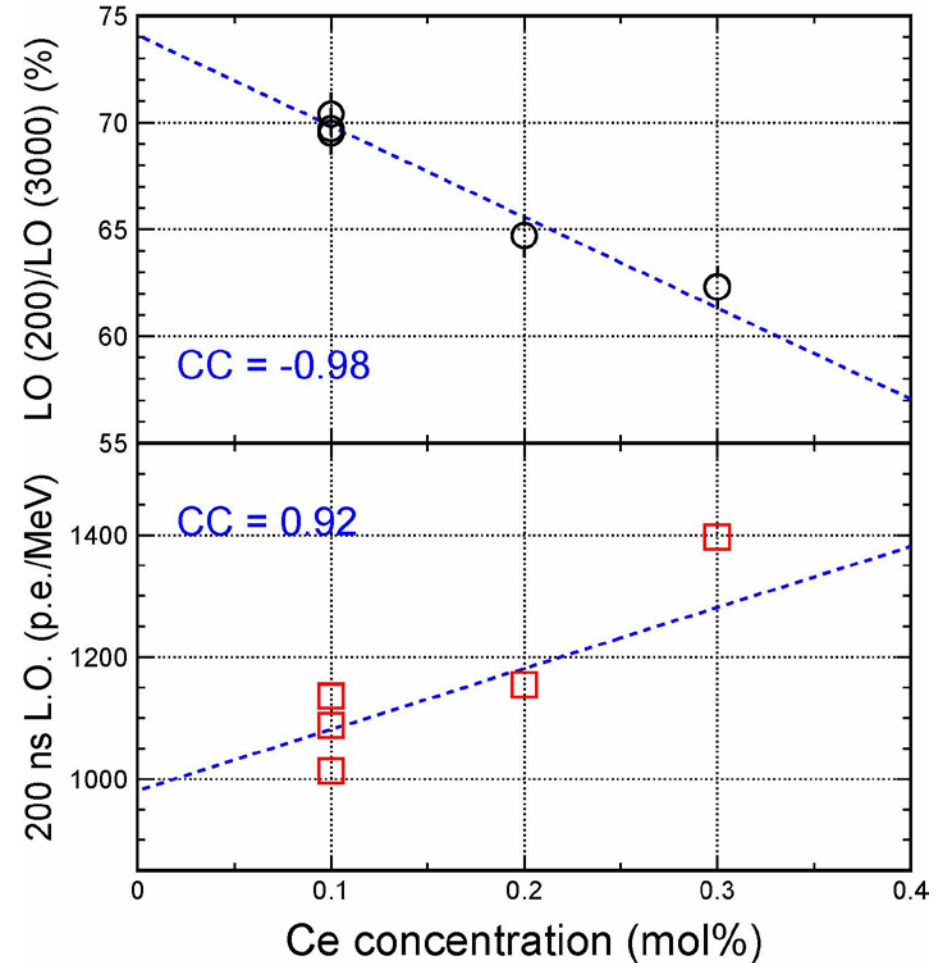
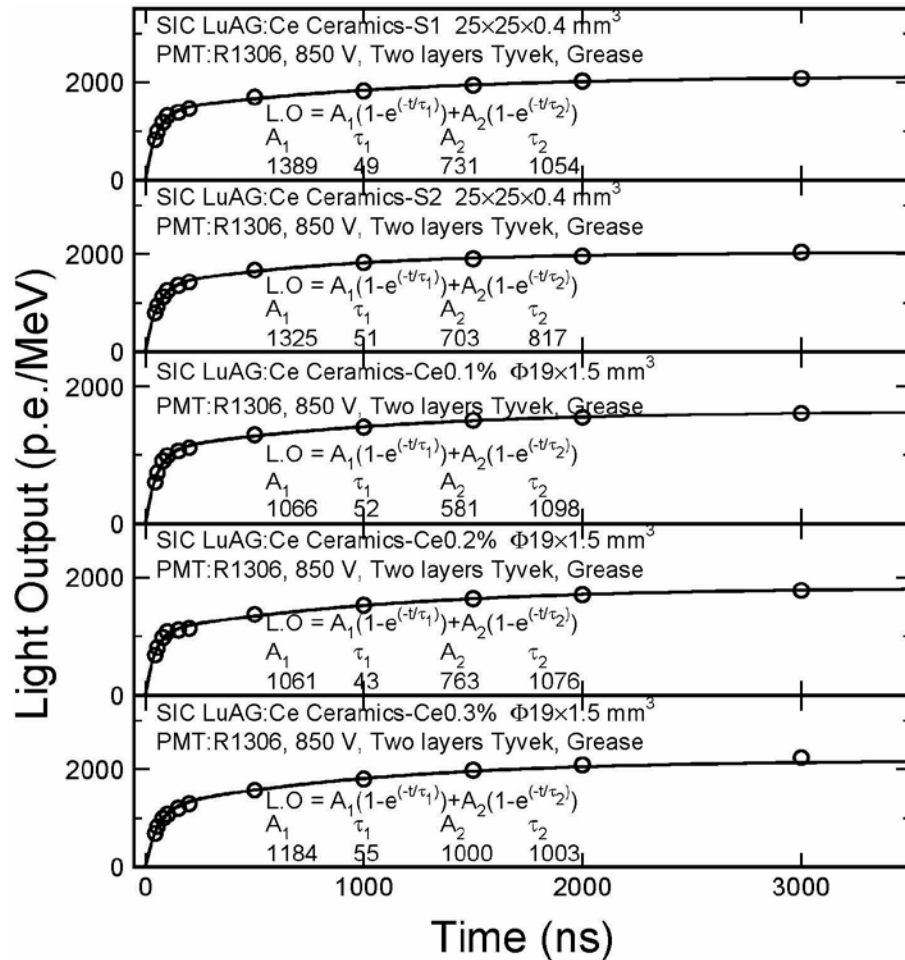
Very promising for a scintillating ceramics based calorimeter
 Further investigation: slow component and LuAG:Pr



LO & F/T ratio vs. Ce Doping



F/T ratio improves as the Ce concentration decrease

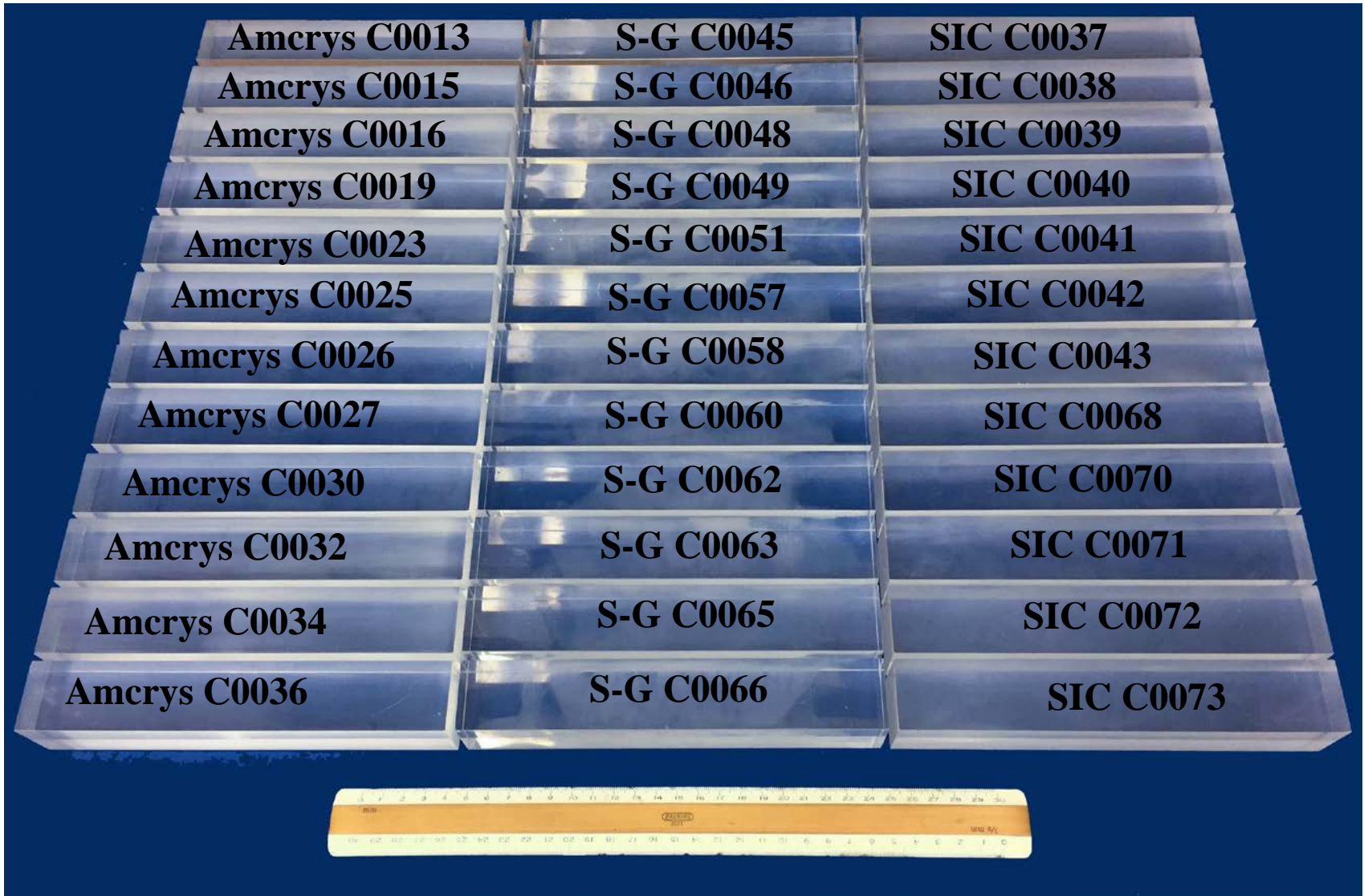




Mu2e Preproduction Csl

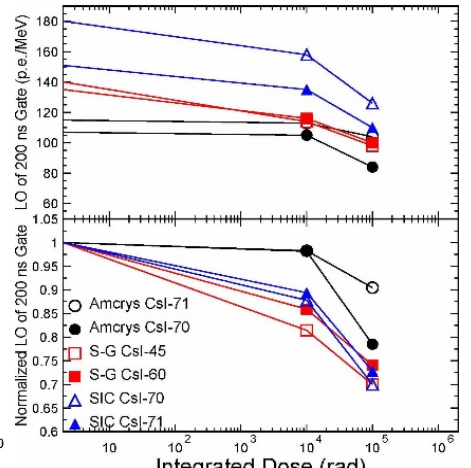
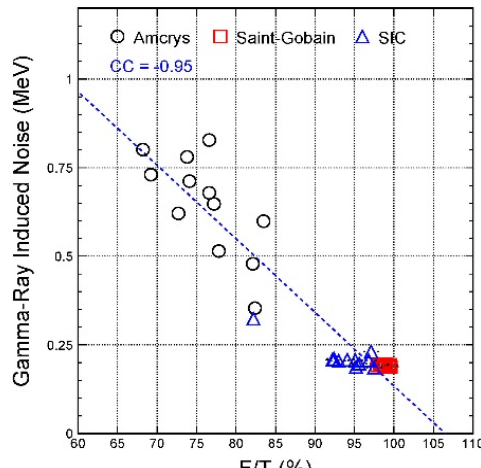
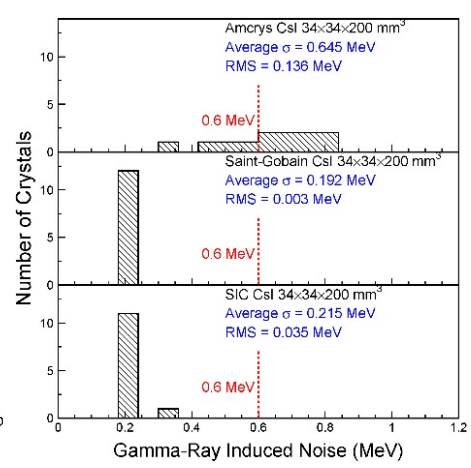
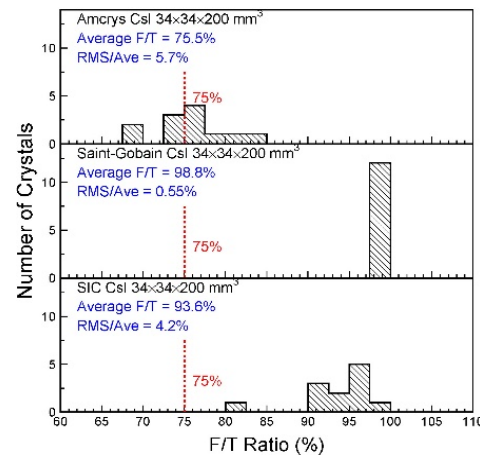
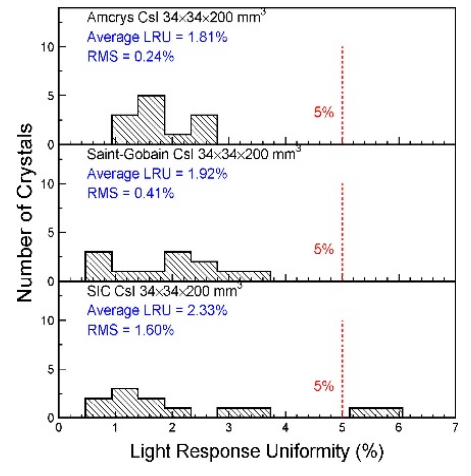
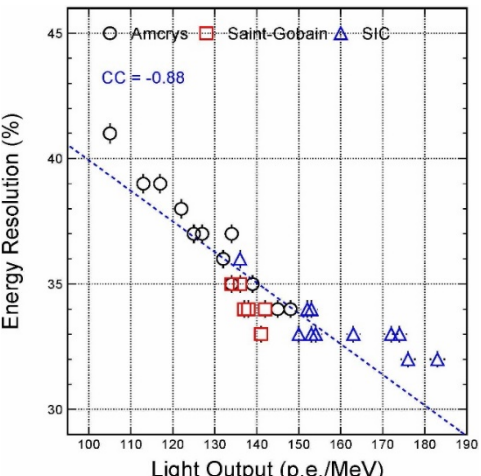
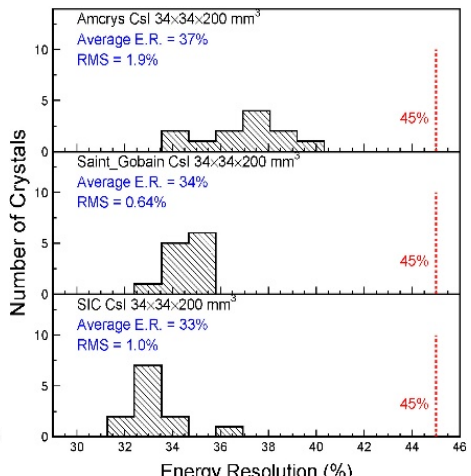
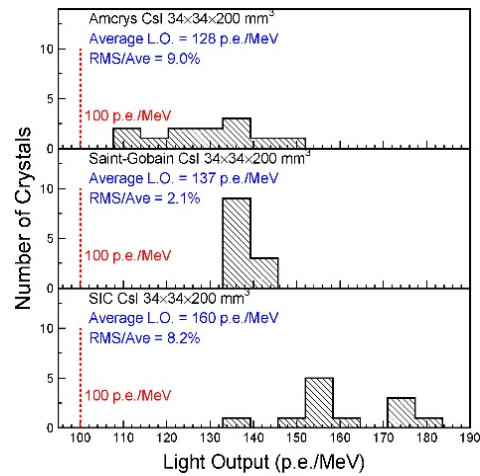
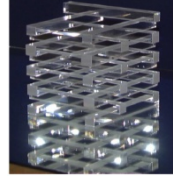


Arranged according to crystal ID in the Mu2e database





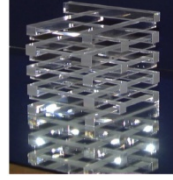
Quality of Pre-Production CsI



Most preproduction crystals satisfy specifications, except a few crystals from SICCAS fail the LRU spec and about half Amcrys crystals fail the F/T ratio and RIN



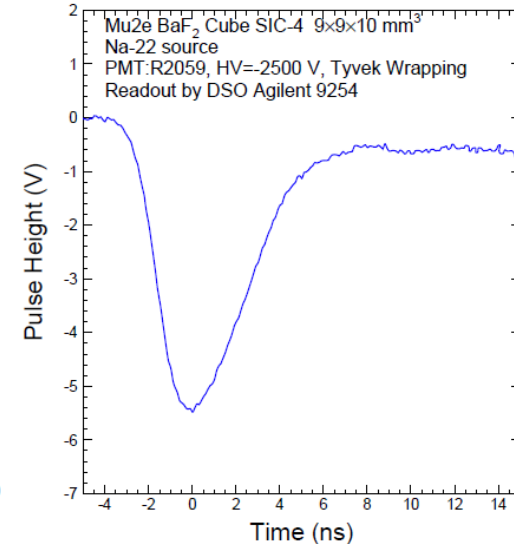
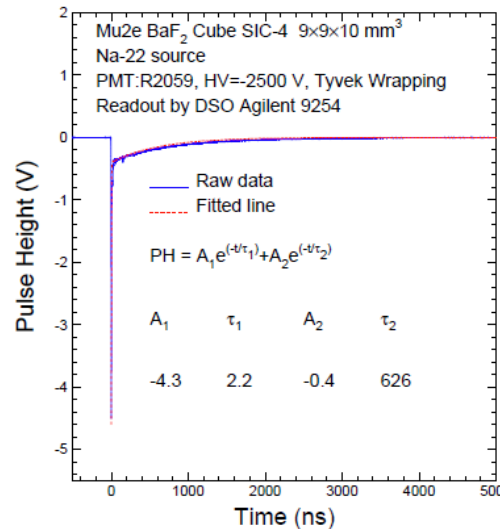
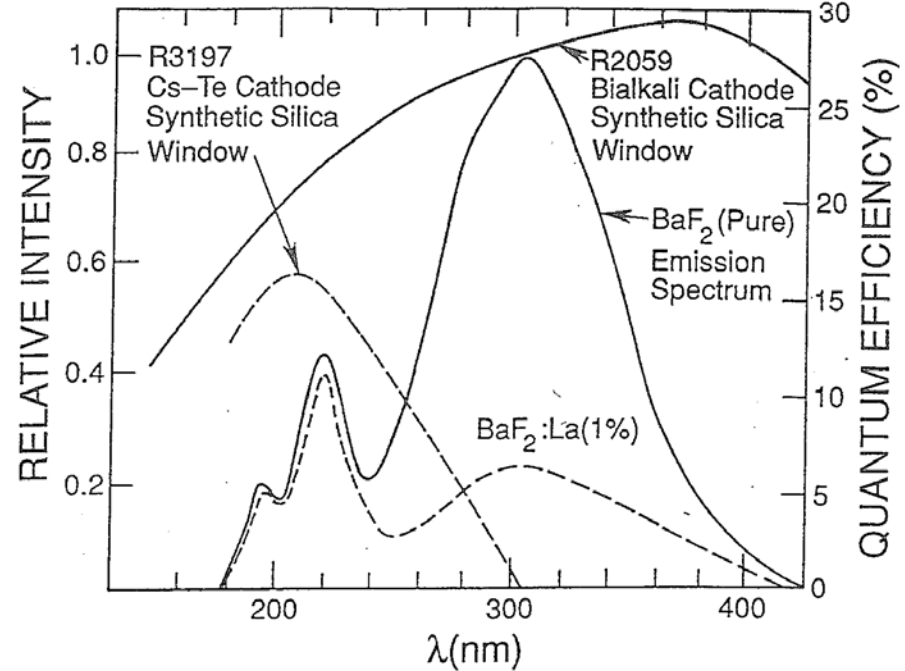
Fast and Slow Light from BaF₂



A radiation level exceeding 100 krad is expected at the proposed Mu2e-II, so BaF₂ is being considered.

The amount of light in the fast component of BaF₂ at 220 nm with sub-ns decay time is similar to CsI.

Spectroscopic selection of fast component may be realized by solar blind photocathode and/or selective doping.



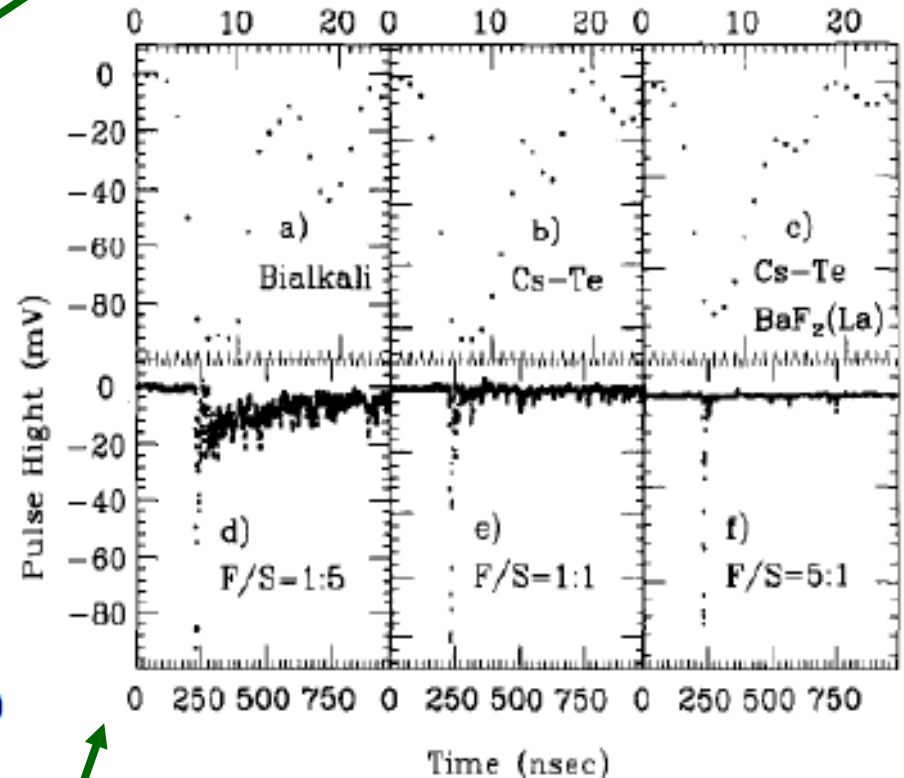
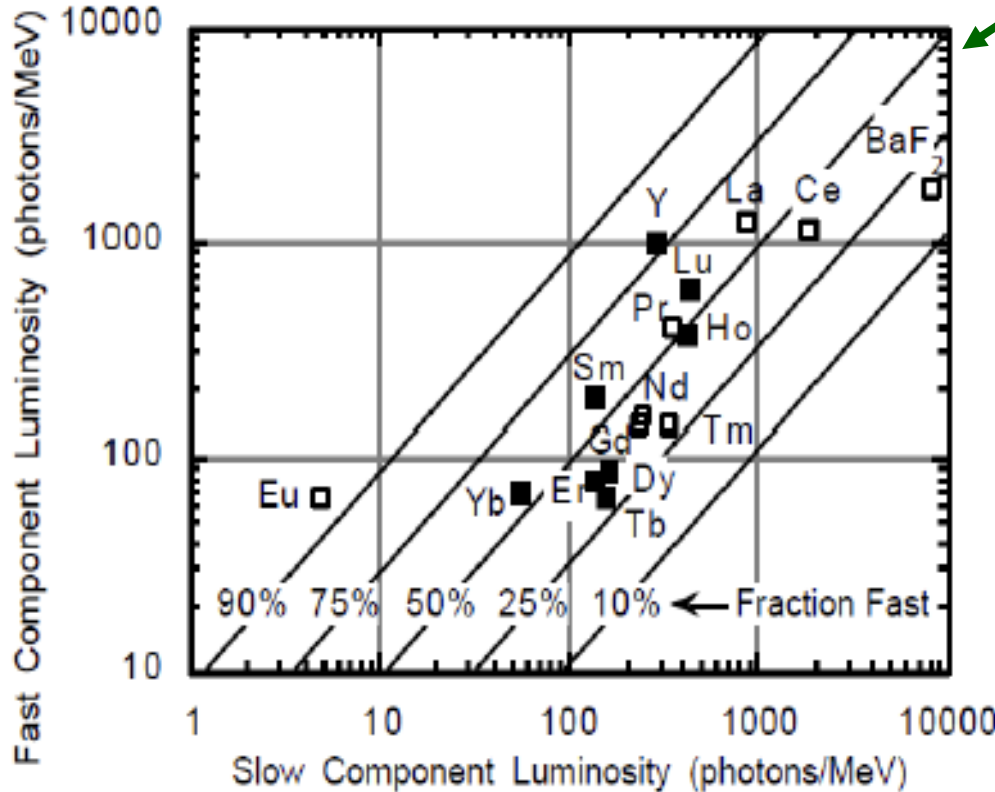


Slow Suppression: Doping & Readout



Slow component may be suppressed by RE doping: Y, La and Ce

B.P. SOBOLEV et al., "SUPPRESSION OF BaF₂ SLOW COMPONENT OF X-RAY LUMINESCENCE IN NON-STOICHIOMETRIC Ba_{0.9R0.1}F₂ CRYSTALS (R=RARE EARTH ELEMENT)," *Proceedings of The Material Research Society: Scintillator and Phosphor Materials*, pp. 277-283, 1994.



Solar-blind cathode (Cs-Te) + La doping achieved F/S = 5/1

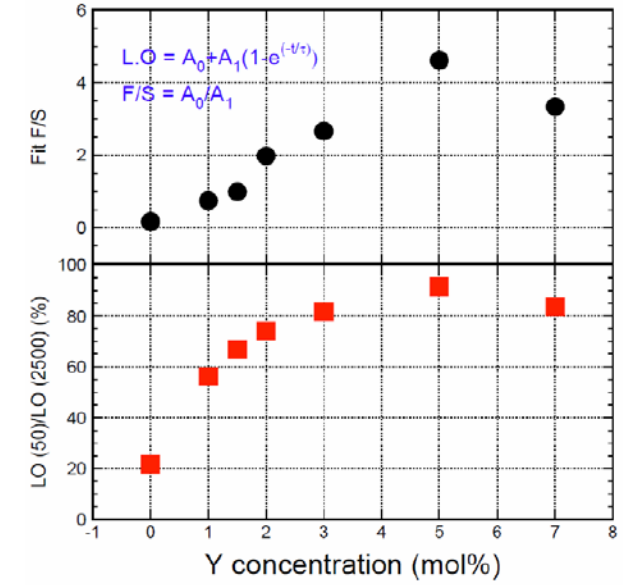
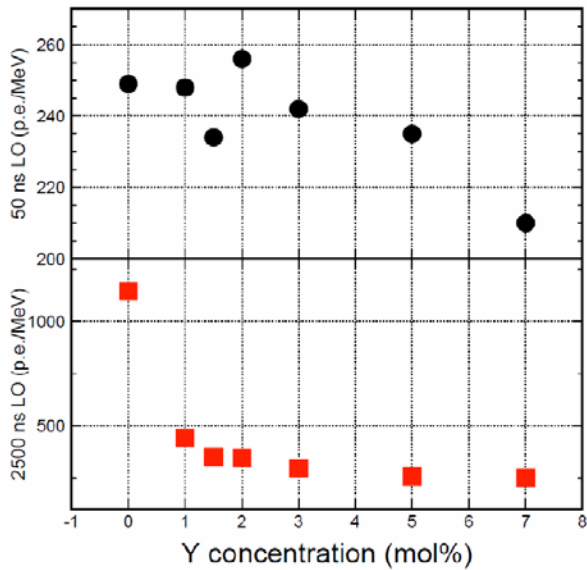
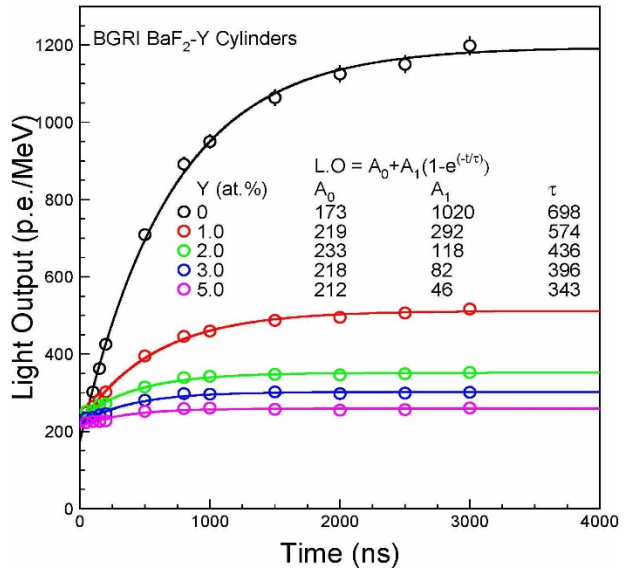
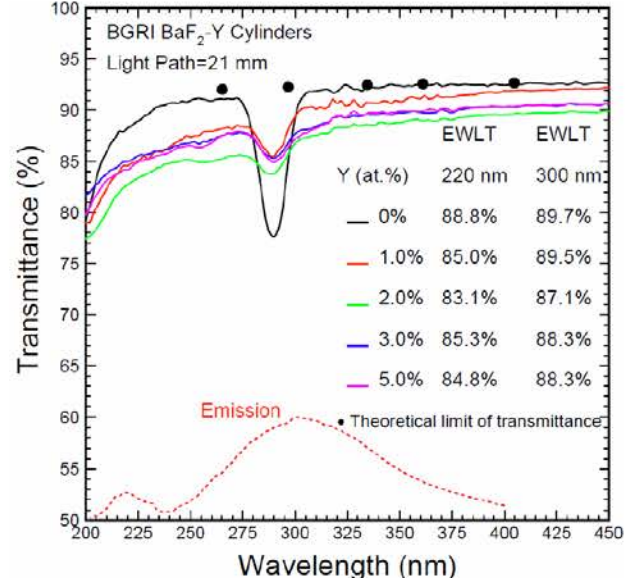
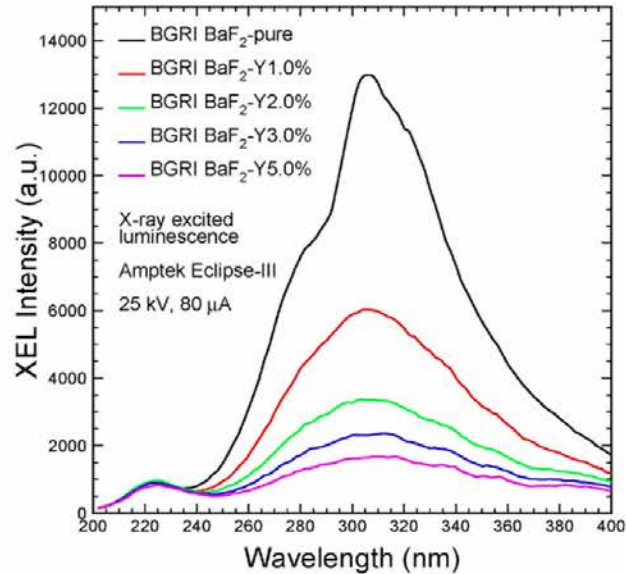
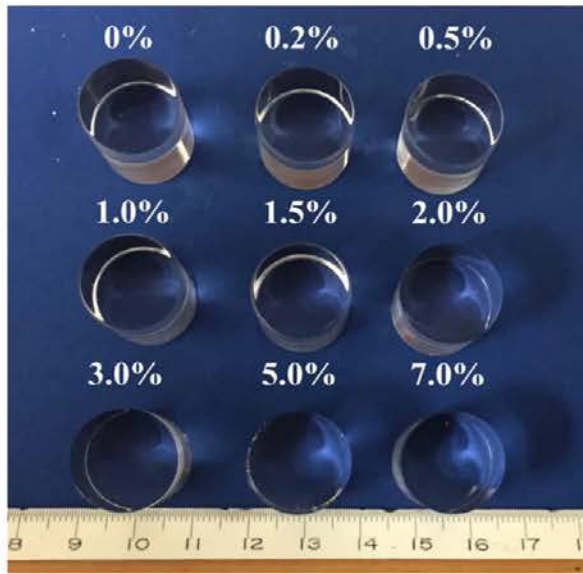
Z. Y. Wei, R. Y. Zhu, H. Newman, and Z. W. Yin, "Light Yield and Surface-Treatment of Barium Fluoride-Crystals," *Nucl Instrum Meth B*, vol. 61, pp. 61-66, Jul 1991.



Yttrium Doping in BaF₂



F/S ratio from 1/5 to 5/1: very effective slow suppression





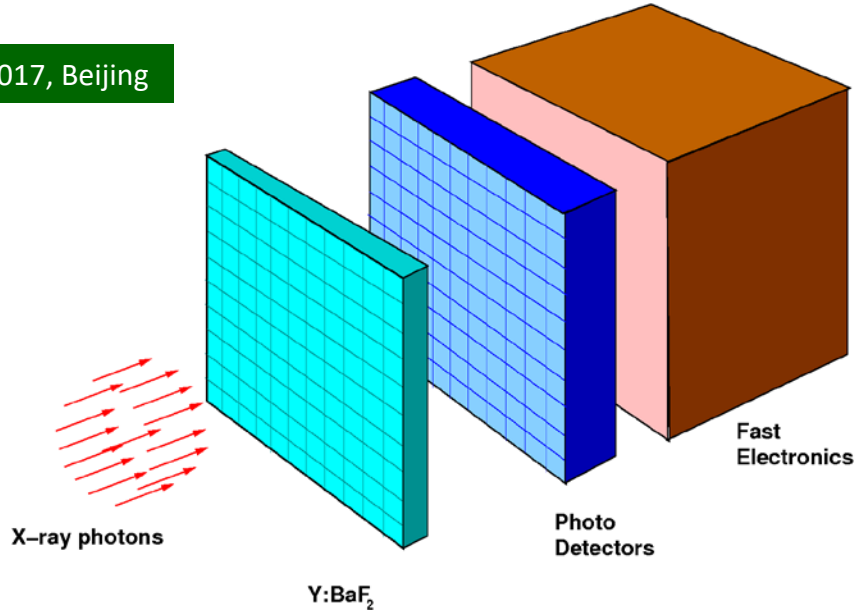
A Y:BaF₂ Crystal Based Imager



- BaF₂ has good efficiency for hard X-rays.
- Its fast scintillation with sub-ns decay time provides bright light in 2 ns with very little tail.
- Yttrium doping in BaF₂ suppresses its slow scintillation by a factor of 25 and maintains its fast light.

R-Y Zhu, Talk presented in TIPP2017, Beijing

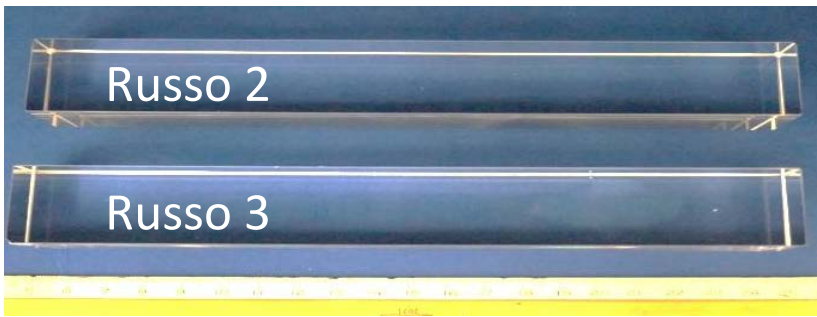
- A detector concept:
 - Pixelized Y:BaF₂ screen;
 - Pixelized fast photodetector;
 - Fast electronics readout.



- To be developed:
Crystals, DUV photodetectors and readout.



BGRI/Incrom/SIC BaF₂ Samples



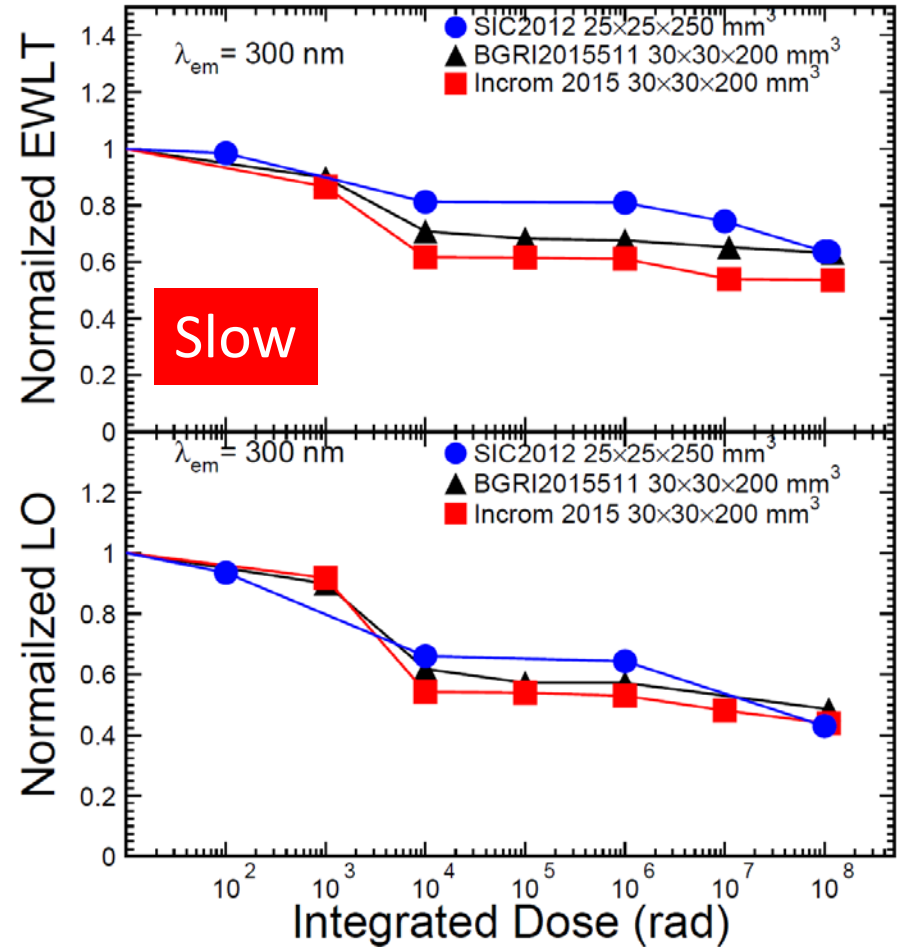
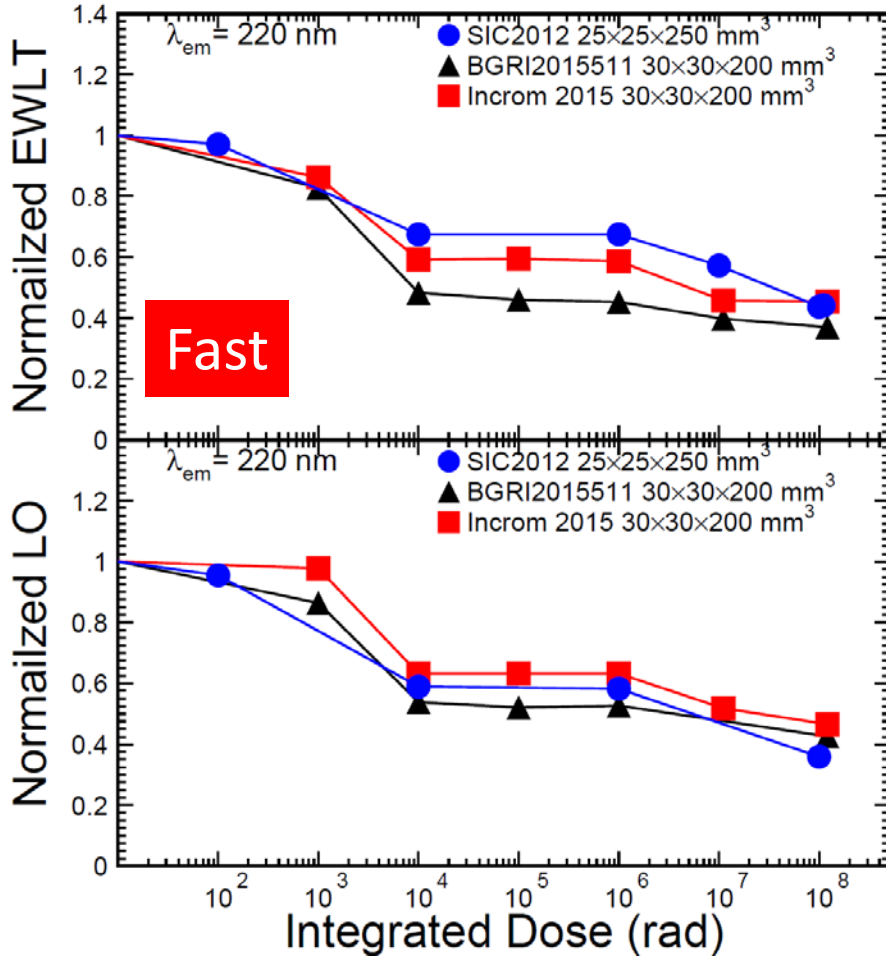
ID	Vendor	Dimension (mm ³)	Polishing
SIC 1-20	SICCAS	30x30x250	Six faces
BGRI-2015 D, E, 511	BGRI	30x30x200	Six faces
Russo 2, 3	Incrom	30x30x200	Six faces



BaF₂: Normalized EWLT and LO



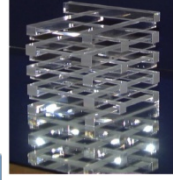
Consistent damage in crystals from three vendors



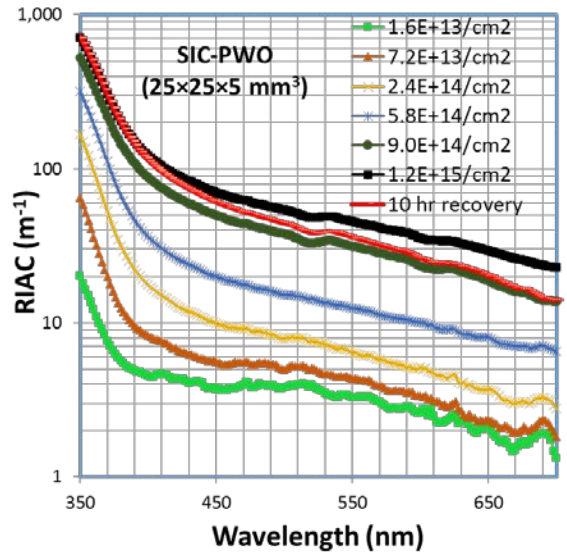
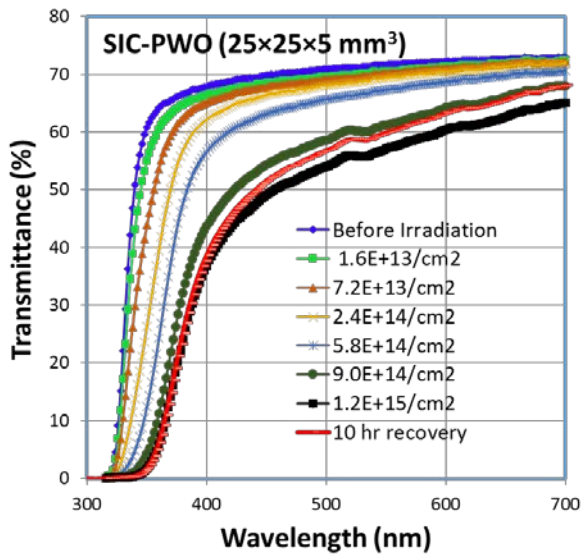
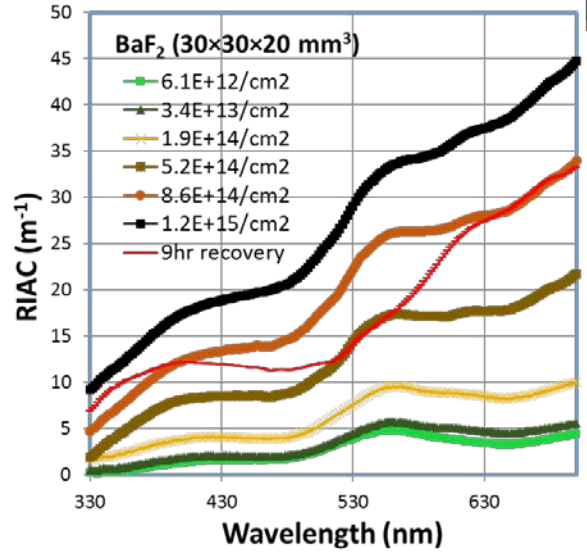
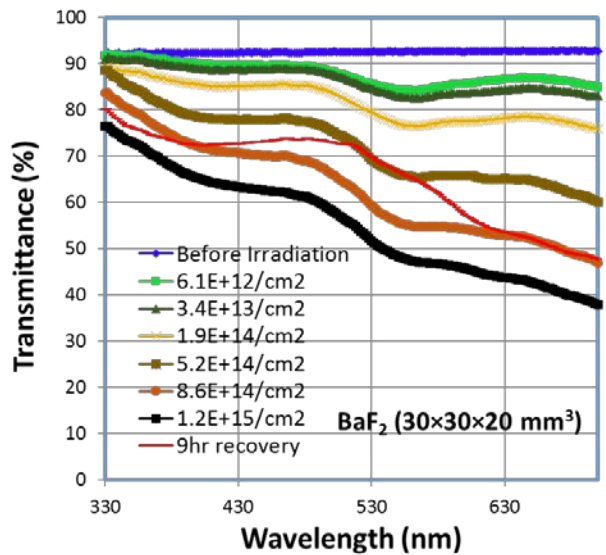
Remaining light output after 120 Mrad: 40%/45% for the fast/slow component



800 MeV Proton Damage in BaF₂ & PWO



A Hellma BaF₂ of 2 cm was irradiated from 6.1×10^{12} to 1.2×10^{15} p/cm² in six steps with transmittance (330-650 nm) measured *in-situ*. The sample will be measured at Caltech for 200 – 650 nm.

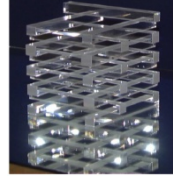


A 5 mm thick SIC PWO plate was irradiated from 1.6×10^{13} to 1.2×10^{15} p/cm² with transmittance (300-700 nm) measured *in-situ*. The RIAC at 420 nm was measured to be 13.1 / 92.2 cm⁻¹ after 2.4×10^{14} / 1.2×10^{15} p/cm².

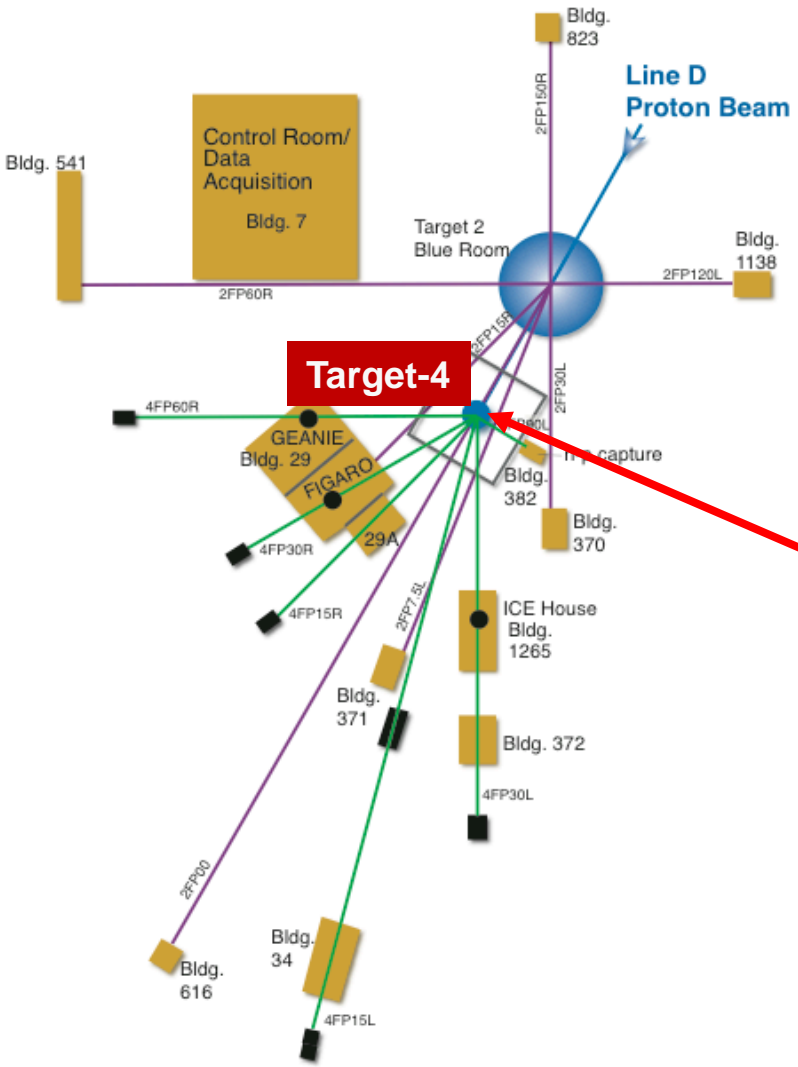
R.-Y. Zhu, "Preliminary Report on the Experiment 7324 with 800 MeV Protons at Los Alamos," http://www.hep.caltech.edu/~zhu/talks/ryz_1601207_LANL.pdf



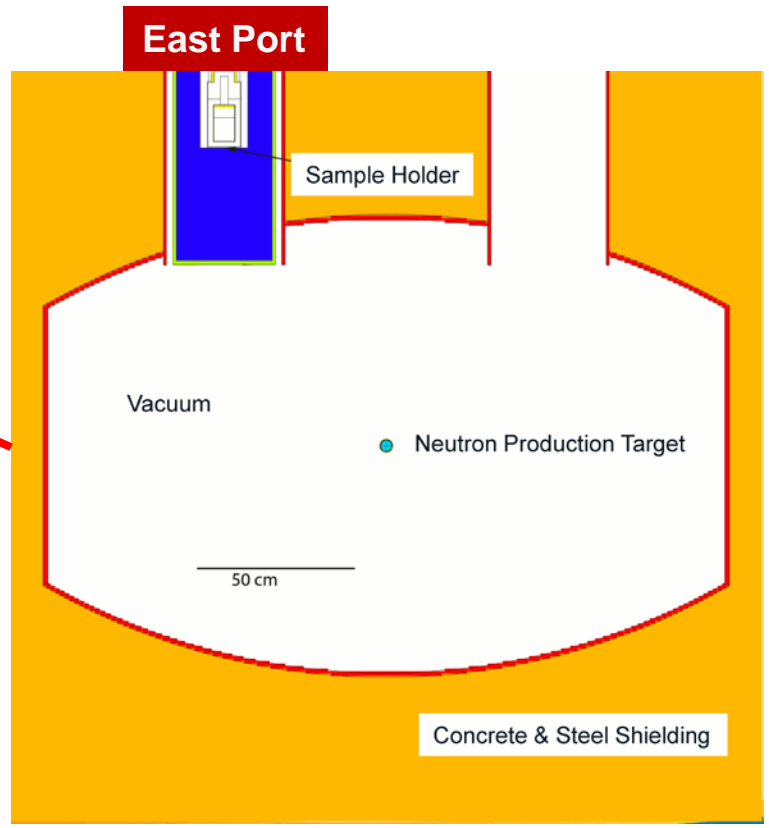
Neutron Irradiation Test at LANL



Los Alamos Neutron Science Center (LANSCE)

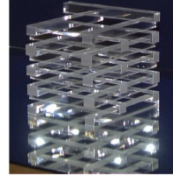


Samples are placed at Target-4 East Port, about 1.2 m away from the neutron production target.

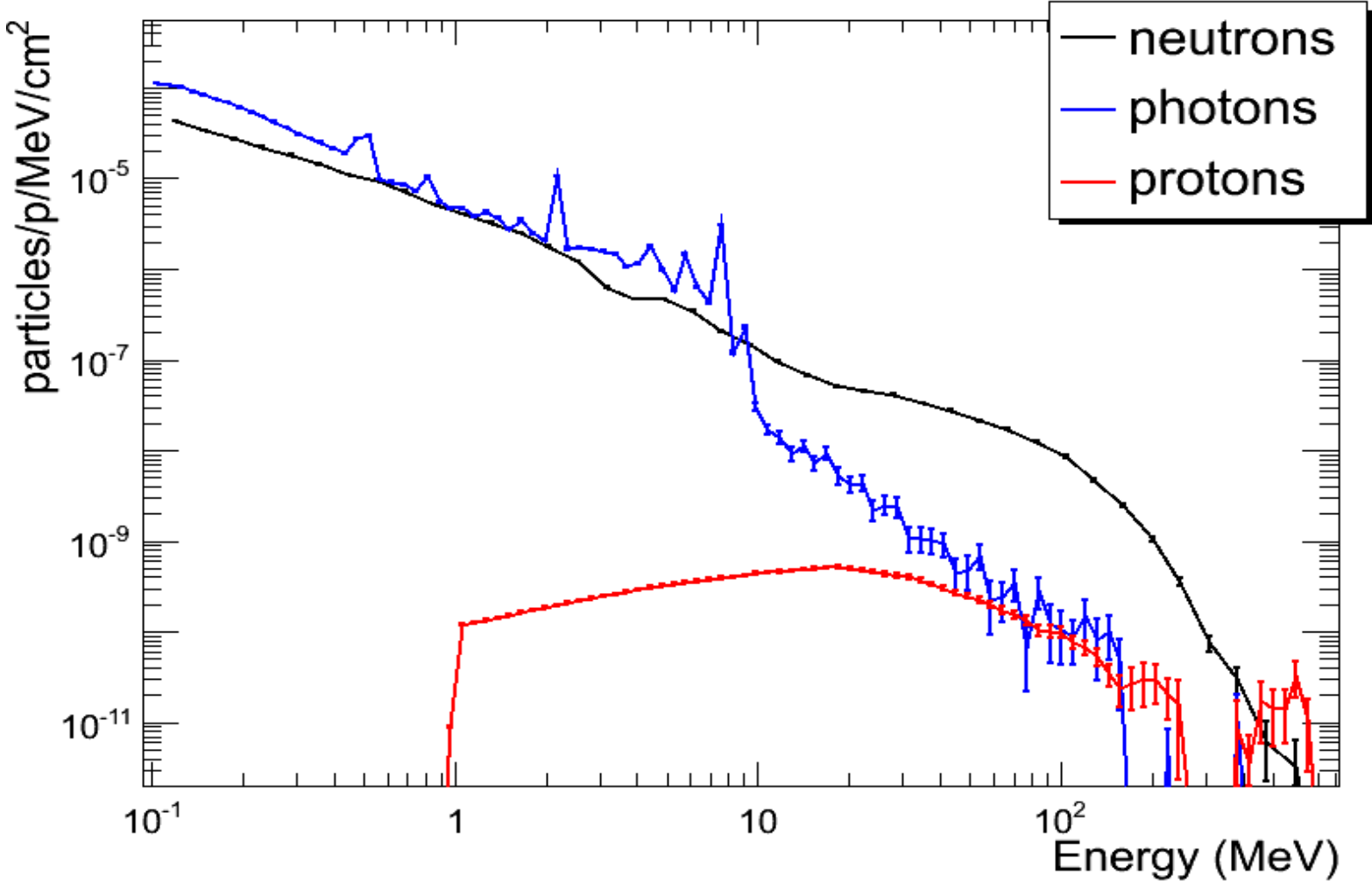




Neutrons/Photons/Protons Fluxes



Neutrons/Photons/Protons fluxes are calculated by using MCNPX (Monte Carlo N-Particle eXtended). Plotted spectra are tallied in the largest sample volume (averaging)





Estimated Fluence and Dose

3 groups of samples were irradiated for 21.2, 46.3 and 119.8 days respectively with the neutron and proton fluences and ionization dose calculated by using the 800 MeV proton beam data.

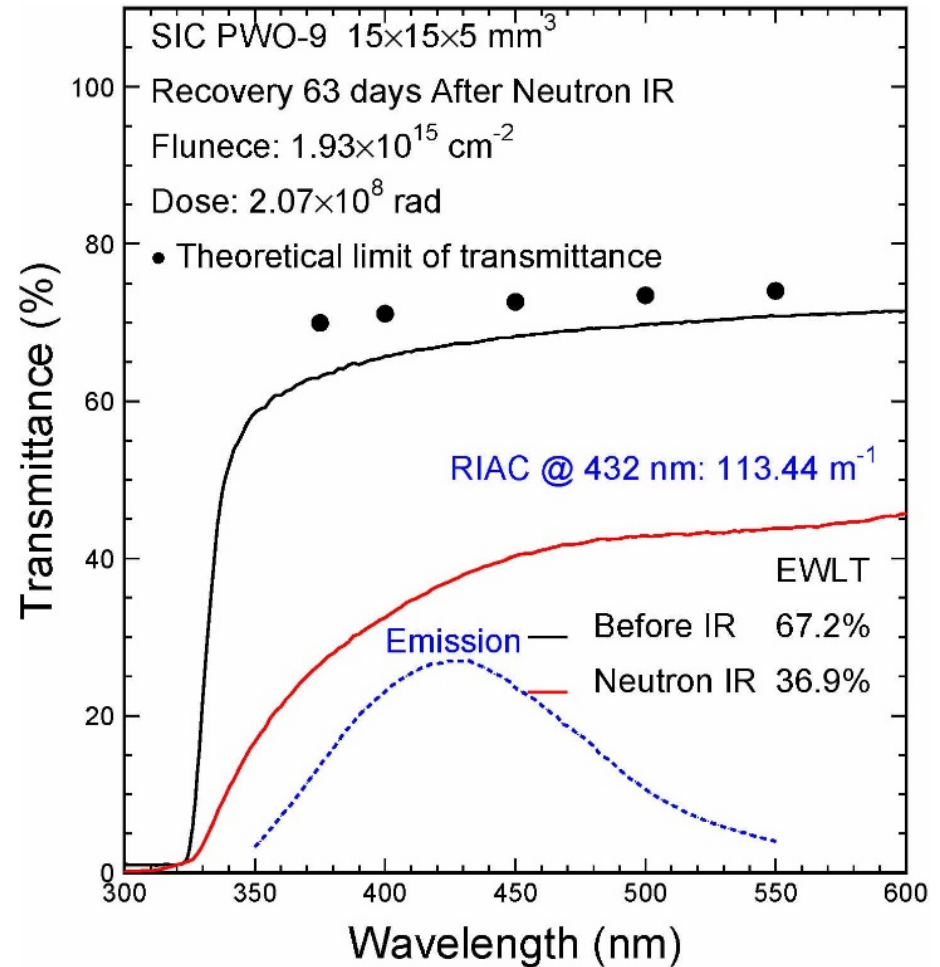
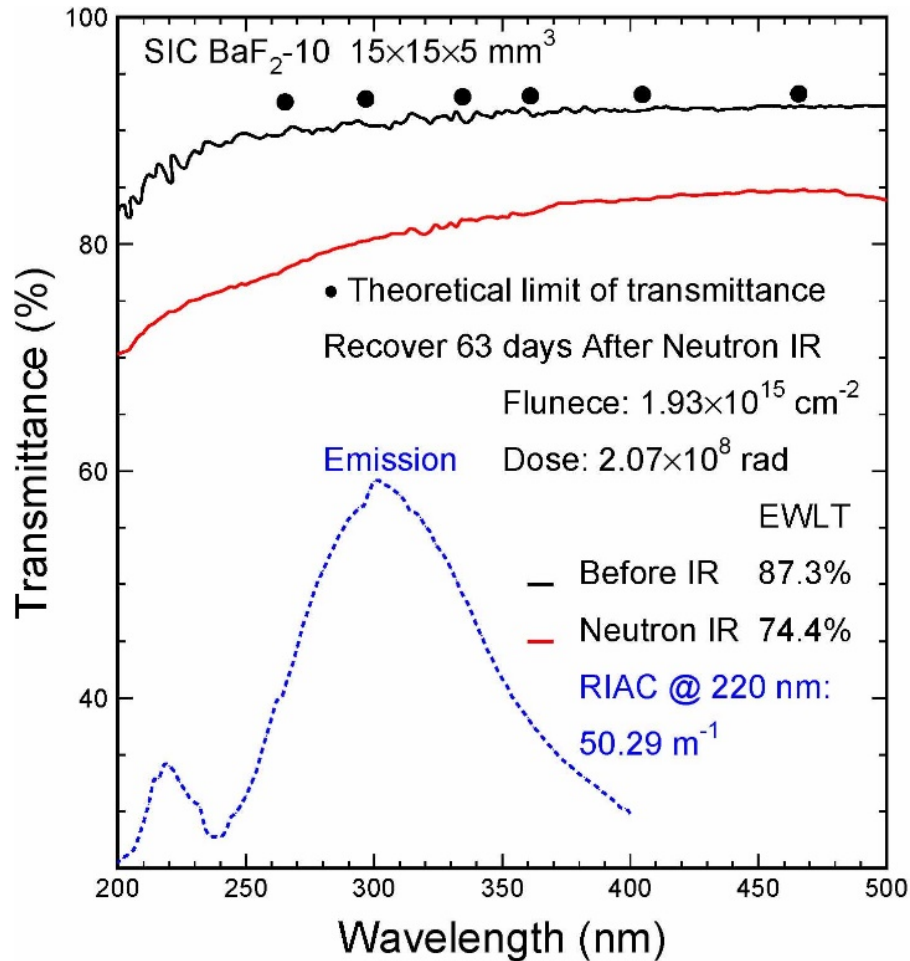
Particles	Group-1 Fluence (cm ⁻²)	Group-2 Fluence (cm ⁻²)	Group-3 Fluence (cm ⁻²)
Thermal and Epithermal Neutrons (0 < E _n < 1 eV)	1.23E+15	2.69E+15	6.04E+15
Slow and Intermediate Neutrons (1 eV < E _n < 1 MeV)	4.50E+15	9.80E+15	2.20E+16
Fast neutrons Fluence 1: (E_n > 1 MeV)	3.94E+14	8.58E+14	1.93E+15
Fast neutrons Fluence 2: (E_n > 20 MeV)	7.64E+13	1.66E+14	3.74E+14
Protons (E _p > 1 MeV)	9.34E+11	2.03E+12	4.57E+12
Protons Dose (rad)	2.44E+04	5.32E+04	1.20E+05
Photons (E _g > 150 KeV)	1.18E+15	2.57E+15	5.78E+15
Photons Dose (rad)	4.22E+07	9.21E+07	2.07E+08
Photons Dose (rad) with 5 mm Pb shielding	3.00E+07	6.54E+07	1.47E+08



Transmittance Damage in BaF₂ & PWO



Samples in three groups irradiated at the East Port of LANSCE

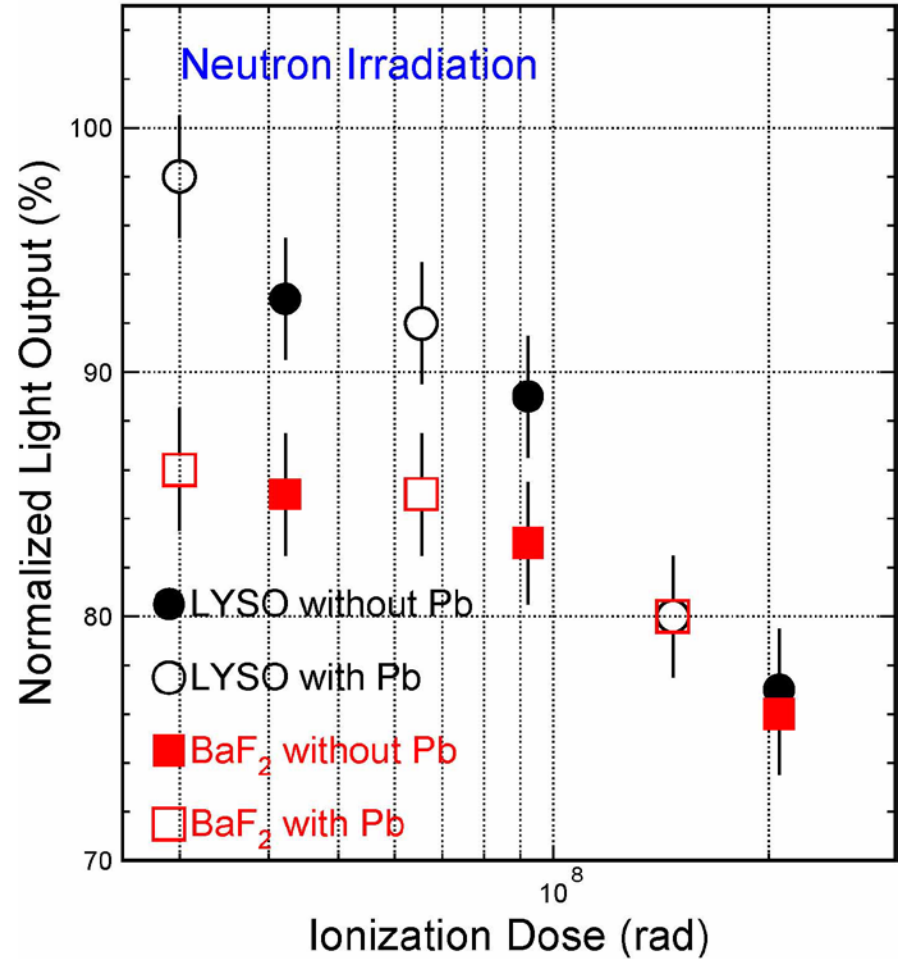
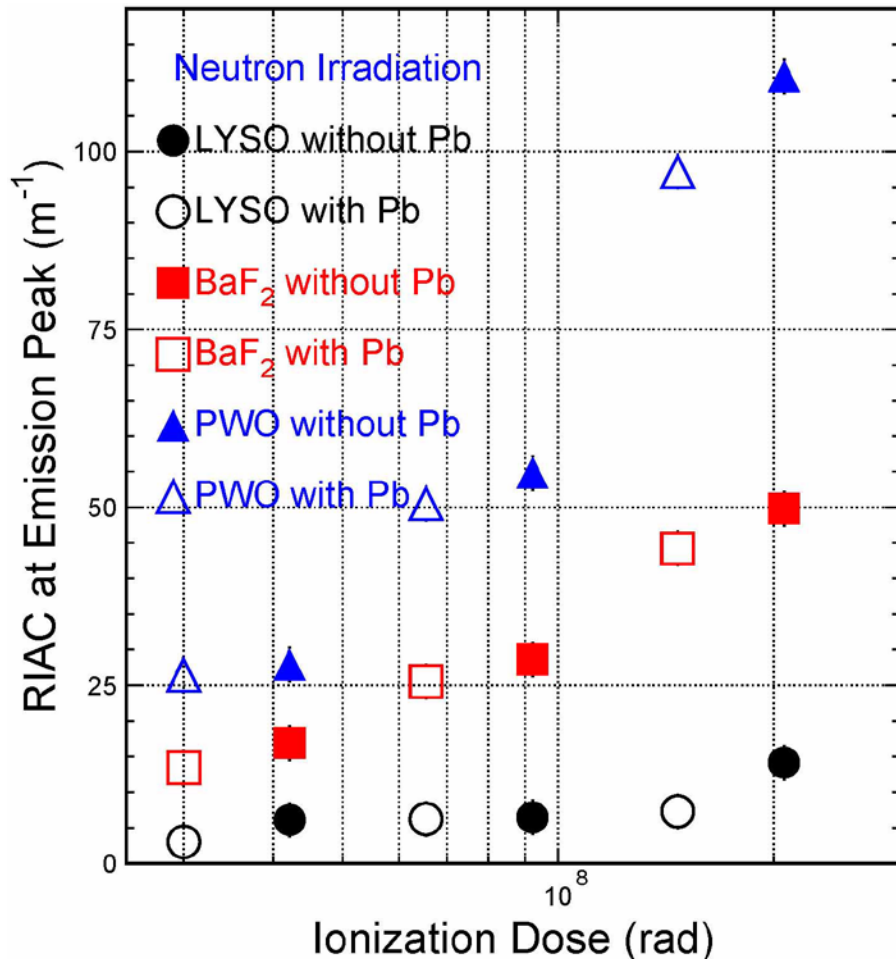




RIAC & LO Vs. Ionization Dose



Results consist with ionization dose induced damage, indicating no neutron specific damage in LYSO, BaF₂ & PWO





Summary



- ❑ **LYSO crystals and LuAG ceramics are robust scintillators against ionization dose as well as charged and neutral hadrons.**
- ❑ **Commercially available undoped BaF₂ crystals provide sufficient fast light with sub-ns decay time and excellent radiation hardness beyond 100 Mrad and 1×10^{15} p/cm². They promise a very fast and robust calorimeter in a severe radiation environment.**
- ❑ **Without using a selected readout yttrium doping in BaF₂ crystals increases the F/S ratio from 1/5 to 5/1 while keeping the intensity of the sub-ns fast component unchanged. The slow contamination at this level is already much less than commercially available undoped CsI.**
- ❑ **Results of the experiments 6991 and 7332 at LANL show fast neutrons up to 2×10^{15} n/cm² do not damage LYSO, BaF₂ and PWO crystals.**
- ❑ **Our plan is to investigate LYSO:Ce,Ca crystals, LuAG:Ce and LuAG:Pr ceramics, and radiation hardness of Y:BaF₂ crystals. Will also pay an attention to photodetector with DUV response: LAPPD, Si or diamond based solid state detectors.**



Diamond Photodetector



E. Monroy, F. Omnes and F. Calle, "Wide-bandgap semiconductor ultraviolet photodetectors, IOPscience 2003 Semicond. Sci. Technol. 18 R33

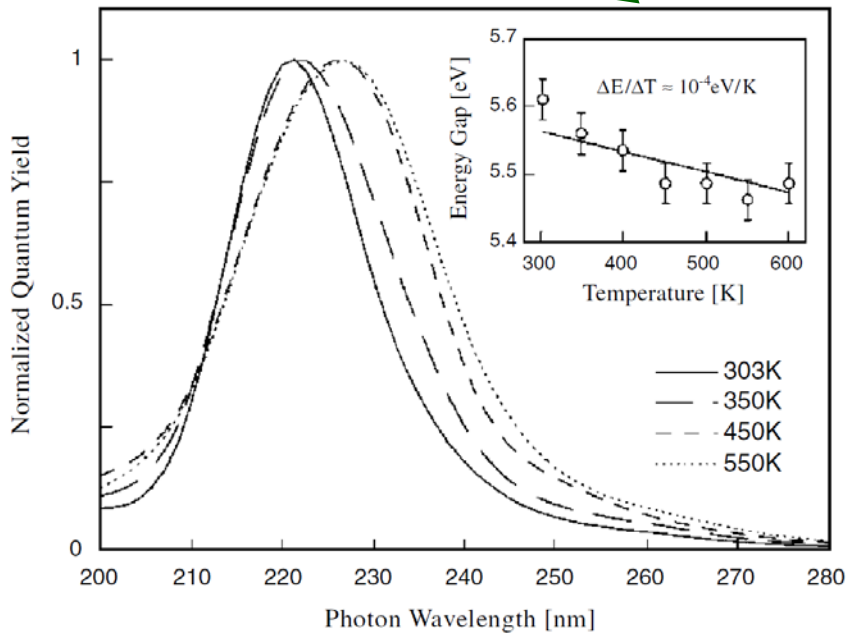


Figure 6. Quantum efficiency of diamond photoconductors at different temperatures and Arrhenius plot of the peak value (inset). (From [Sal00].)

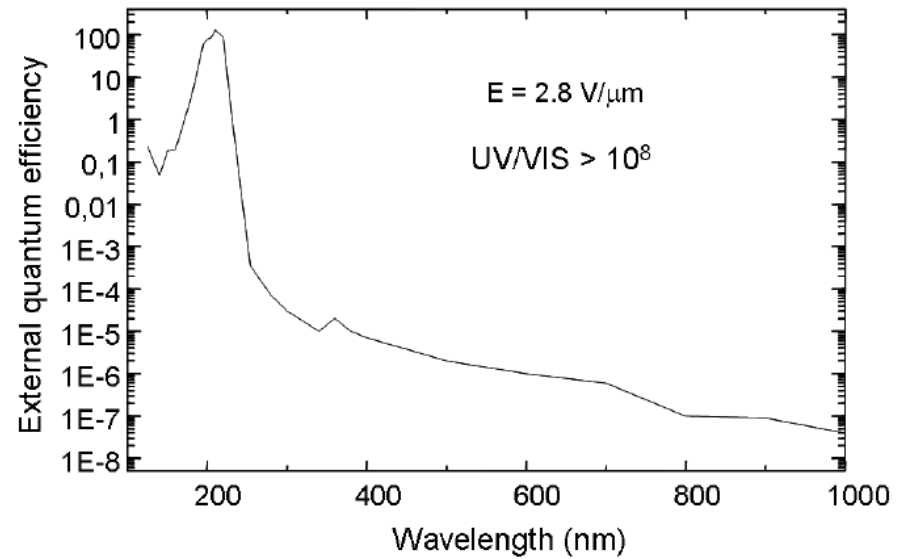
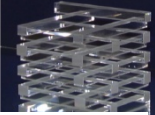


Fig. 4. External quantum efficiency extended to visible and near infrared wavelength regions. The

E. Pace and A. De Sio, "Innovative diamond photo-detectors for UV astrophysics", Mem. S.A.It. Suppl. Vol. 14, 84 (2010)

GHz Hard X-Ray Imagers



High-Energy and Ultrafast X-Ray Imaging Technologies and Applications

Organizers: Peter Denes, Sol Gruner, Michael Stevens & Zhehui (Jeff) Wang¹
 (Location/Time: Santa Fe, NM, USA /Aug 2-3, 2016)

The goals of this workshop are to gather the leading experts in the related fields, to prioritize tasks for ultrafast hard X-ray imaging detector technology development and applications in the next 5 to 10 years, see Table 1, and to establish the foundations for near-term R&D collaborations.

Table I. High-energy photon imagers for MaRIE XFEL

Performance	Type I imager	Type II imager
X-ray energy	30 keV	42-126 keV
Frame-rate/inter-frame time	0.5 GHz/2 ns	3 GHz / 300 ps
Number of frames	10	10 - 30
X-ray detection efficiency	above 50%	above 80%
Pixel size/pitch	$\leq 300 \mu\text{m}$	$< 300 \mu\text{m}$
Dynamic range	10^3 X-ray photons	$\geq 10^4$ X-ray photons
Pixel format	64 x 64 (scalable to 1 Mpix)	1 Mpix

2 ns and 300 ps inter-frame time requires very fast sensor