



Large Size Yttrium Doped BaF₂ Crystals for the Mu2e-II Experiment

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Introduction



- The Mu2e-I experiment is building a undoped CsI calorimeter, which has a fast scintillation at 310 nm with 30 ns decay time and survives an ionization dose up to 100 krad. A radiation level beyond 100 krad is expected by Mu2e-II, where CsI will be blackened.
- BaF₂ crystal is featured with a ultrafast scintillation at 220 nm with 0.5 ns decay time and an adequate radiation hardness. Its slow scintillation at 300 nm with 650 ns decay time, however, would cause pileup in a high rate environment.
- Two approaches are used to suppress the slow scintillation in BaF₂: selective RE doping and/or dedicated photodetector. Yttrium doping in BaF₂ crystals is found effective, promising a ultrafast calorimeter for Mu2e-II.
- Mass production capability of BaF₂ exists in industry:
 - BGRI (China), Incrom (Russia) and SICCAS (China);
 - Hellma (Germany).
- Recent progress in large size BaF₂:Y crystals for Mu2e-II is reported.



Some Fast Inorganic Scintillators



	LSO/LYSO	GSO	YSO	CsI	BaF ₂	CeF ₃	CeBr ₃	LaCl ₃	LaBr ₃	Plastic scintillator (BC 404) ^①
Density (g/cm ³)	7.4	6.71	4.44	4.51	4.89	6.16	5.23	3.86	5.29	1.03
Melting point (°C)	2050	1950	1980	621	1280	1460	722	858	783	70 [#]
Radiation Length (cm)	1.14	1.38	3.11	1.86	2.03	1.7	1.96	2.81	1.88	42.54
Molière Radius (cm)	2.07	2.23	2.93	3.57	3.1	2.41	2.97	3.71	2.85	9.59
Interaction Length (cm)	20.9	22.2	27.9	39.3	30.7	23.2	31.5	37.6	30.4	78.8
Z value	64.8	57.9	33.3	54	51.6	50.8	45.6	47.3	45.6	5.82
dE/dX (MeV/cm)	9.55	8.88	6.56	5.56	6.52	8.42	6.65	5.27	6.9	2.02
Emission Peak ^a (nm)	420	430	420	310	300 220	340 300	371	335	356	408
Refractive Index ^b	1.82	1.85	1.8	1.95	1.5	1.62	1.9	1.9	1.9	1.58
Relative Light Yield ^{a,c}	100	45	76	3.6 1.1	42 4.1	8.6	99	15 49	153	35
Decay Time ^a (ns)	40	73	60	30 6	650 0.5	30	17	570 24	20	1.8
d(LY)/dT ^d (%/°C)	-0.2	-0.4	-0.1	-1.4	-1.9 0.1	~0	-0.1	0.1	0.2	~0

a. Top line: slow component, bottom line: fast component.

b. At the wavelength of the emission maximum.

c. Relative light yield normalized to the light yield of LSO

d. At room temperature (20°C)

#. Softening point

1. <http://www.detectors.saint-gobain.com/Plastic-Scintillator.aspx>

http://pdg.lbl.gov/2008/AtomicNuclearProperties/HTML_PAGES/216.html

The 0.5 ns scintillation in BaF₂ promises a ultrafast crystal calorimeter to face the challenge of high event rate expected by Mu2e-II

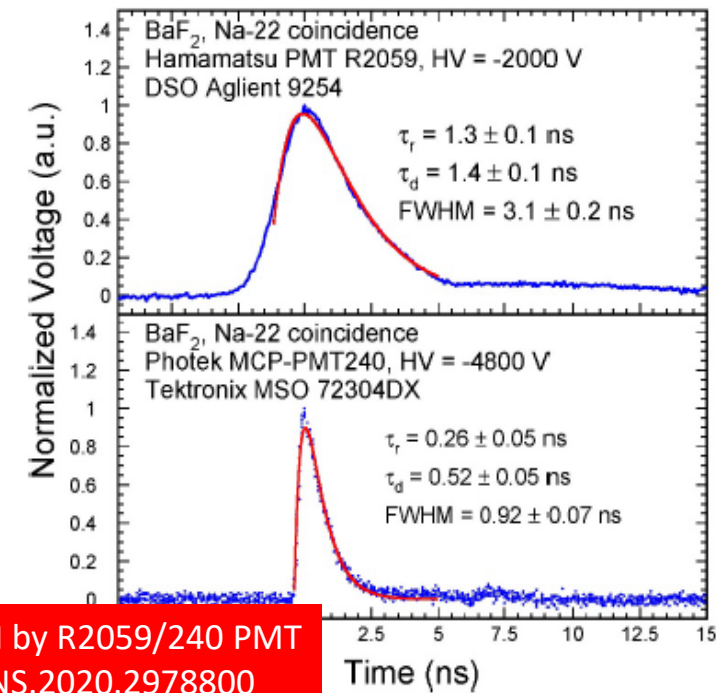
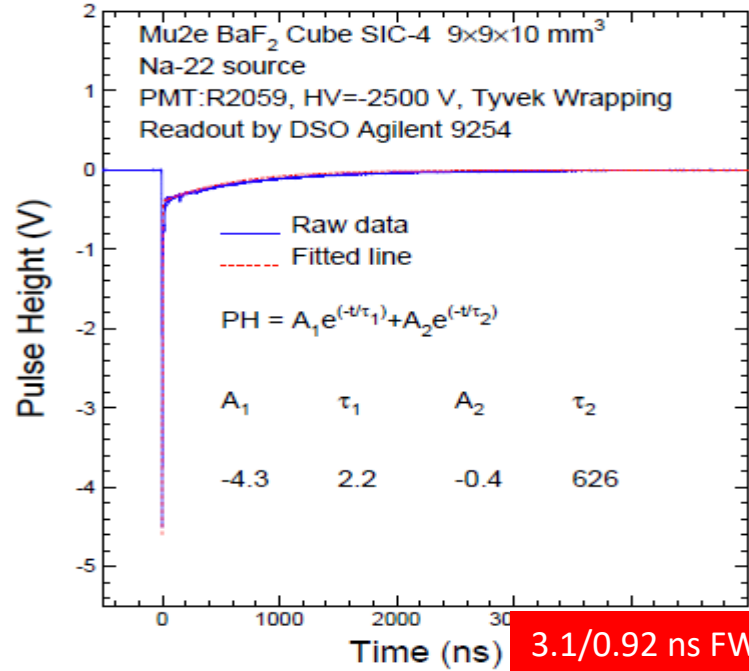
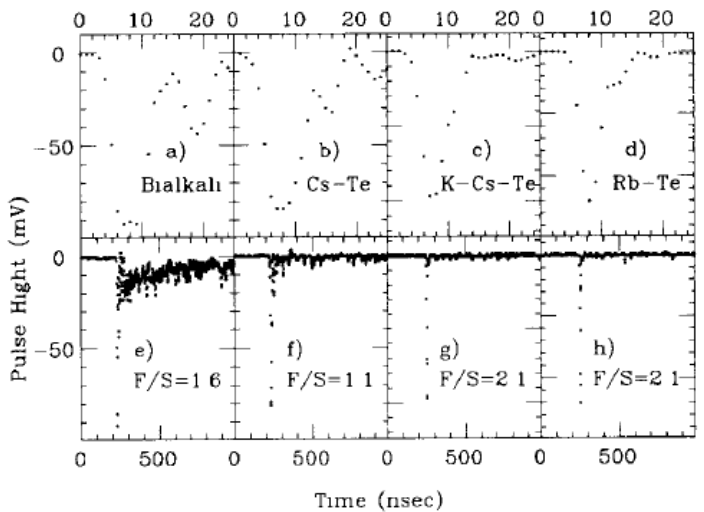
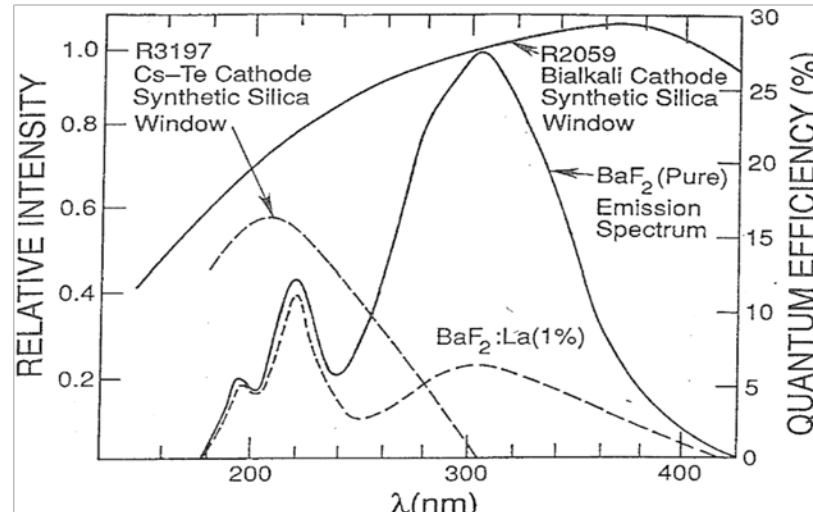


Ultrafast and Slow Light from BaF₂

BaF₂ has a ultrafast scintillation component @ 220 nm with 0.5 ns decay time and an intensity a little less than undoped CsI. It has also a factor of 5 larger slow component @ 300 nm with 300 ns decay time.

Slow suppression may be achieved by selective rare earth doping, e.g. Y, La and Ce, in BaF₂, and/or photodetectors with filters or a solar-blind cathode, e.g. Cs-Te, K-Cs-Te and Rb-Te.

NIMA 340 (1994) 442-457



3.1/0.92 ns FWHM by R2059/240 PMT
DOI 10.1109/TNS.2020.2978800

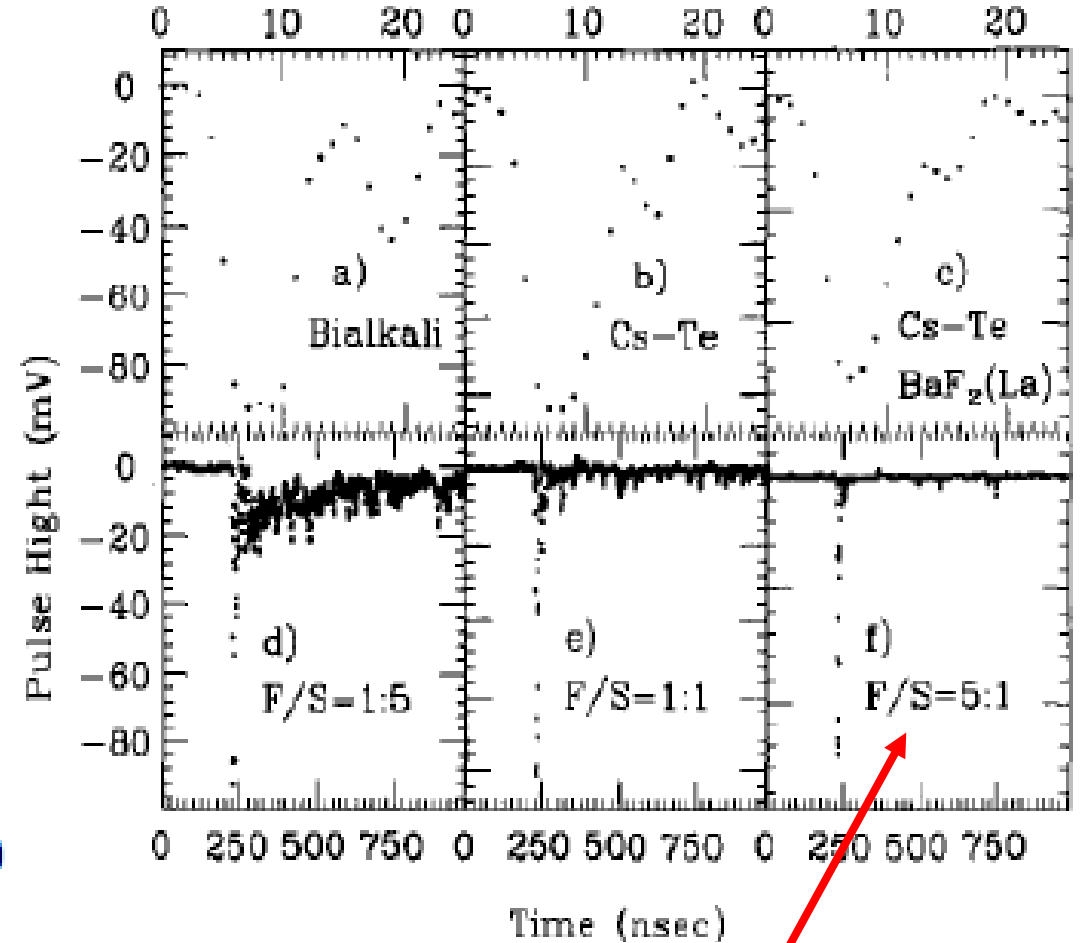
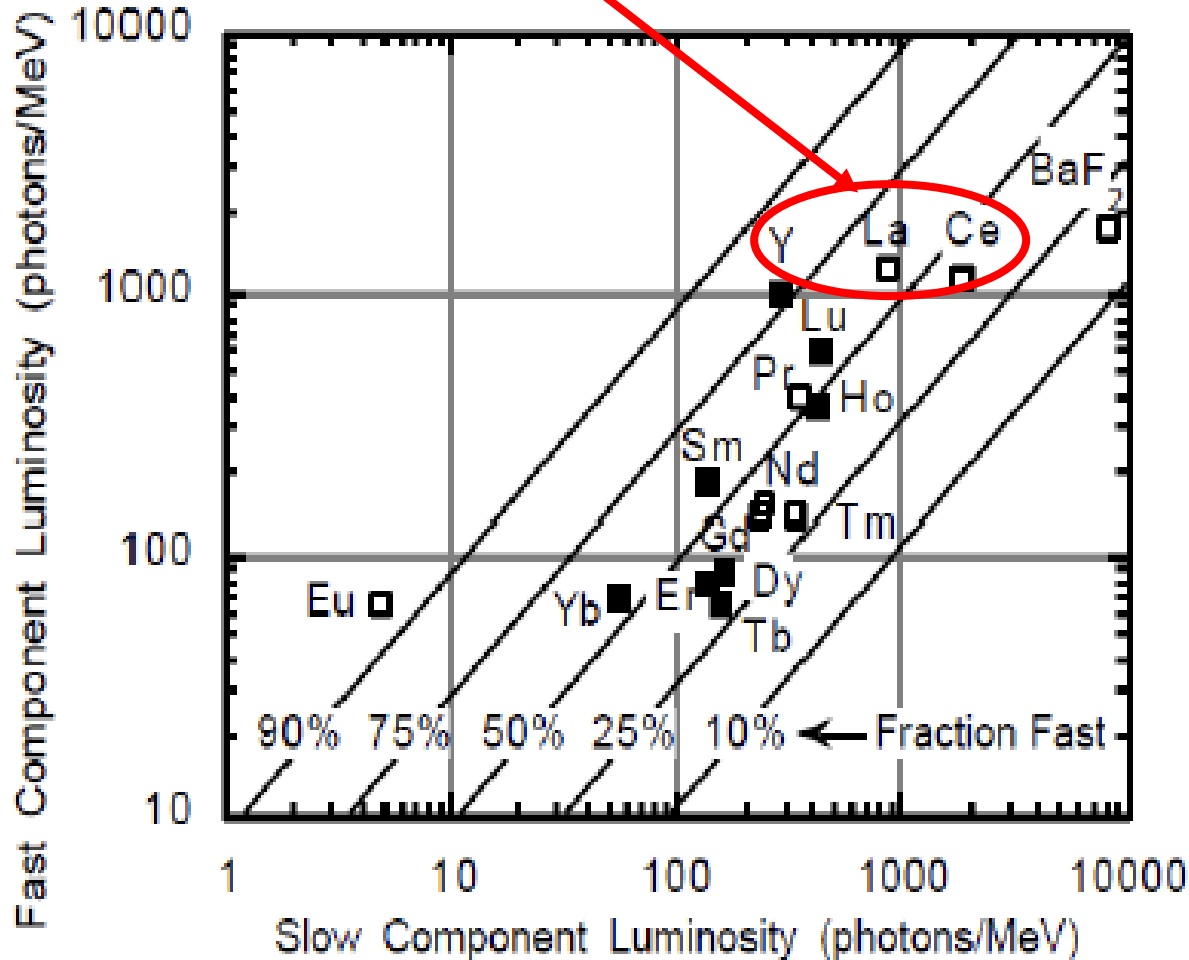


Slow Suppression: Doping & Readout



Slow suppression by RE doped BaF₂ powders: Y, La and Ce (1994)

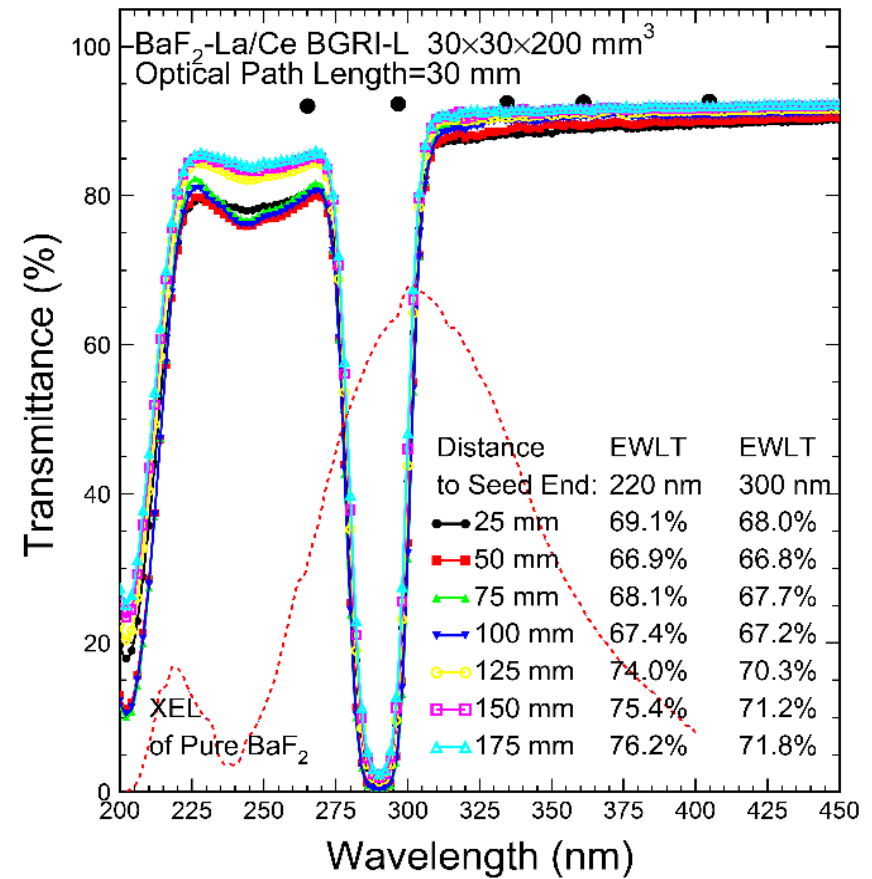
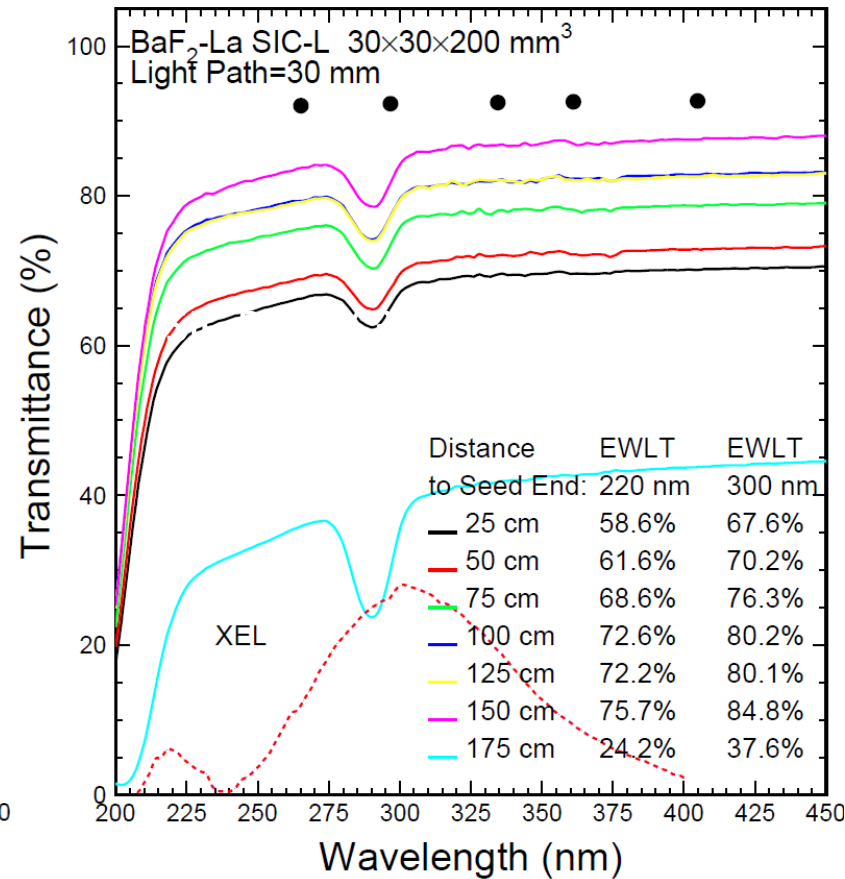
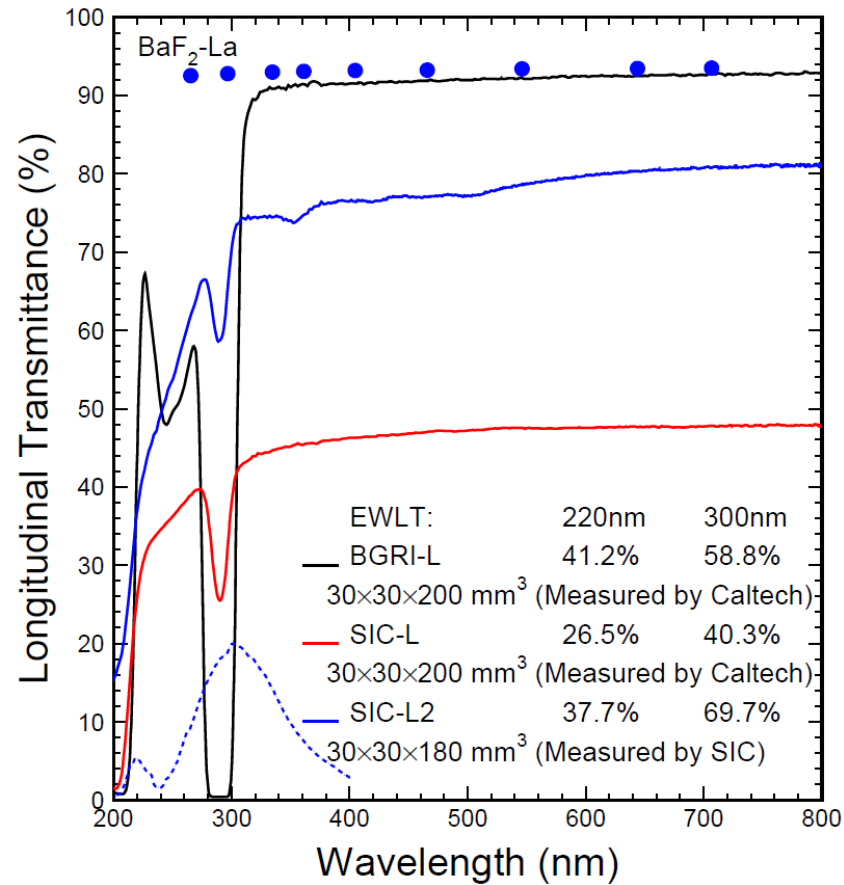
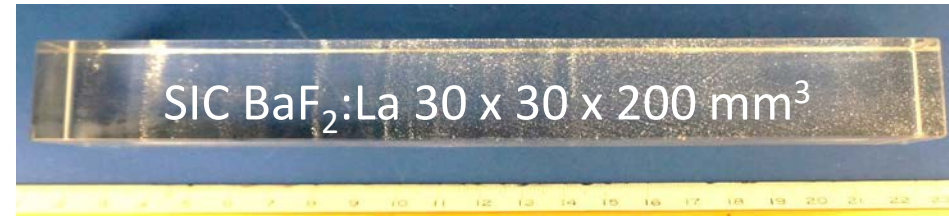
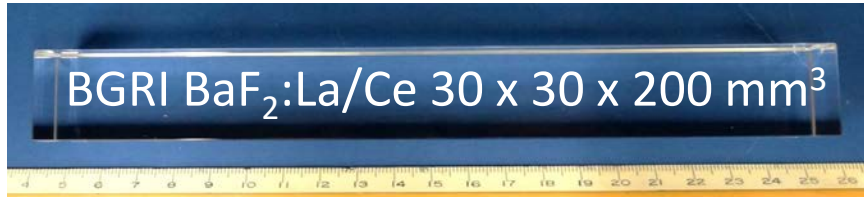
B.P. SOBOLEV et al., "SUPPRESSION OF BaF₂ SLOW COMPONENT OF X-RAY LUMINESCENCE IN NON-STOICHIOMETRIC Ba_{0.9}R_{0.1}F₂ CRYSTALS (R=RARE EARTH ELEMENT)," *Proceedings of The Material Research Society: Scintillator and Phosphor Materials*, pp. 277-283, 1994.



Cs-Te cathode plus La doping raises F/S from 1/5 to 5/1, NIMB 91 (1991) 61-66



Transmittance of BaF₂:La and BaF₂:La/Ce



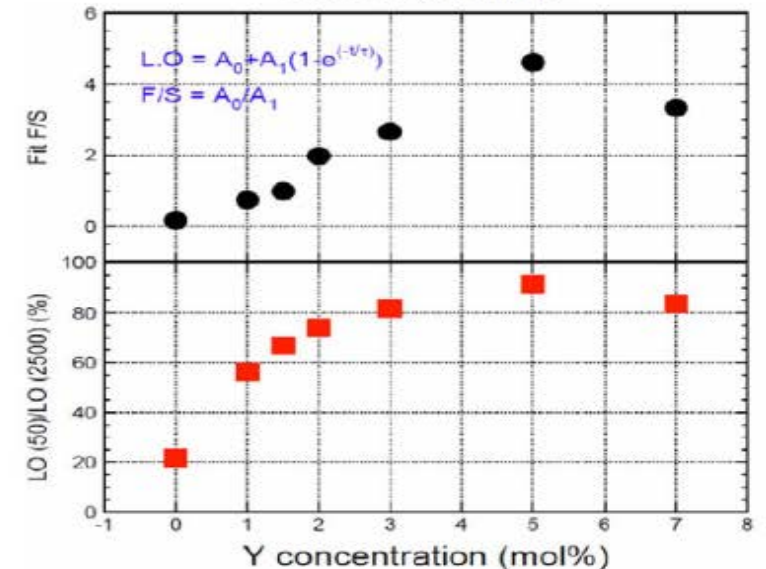
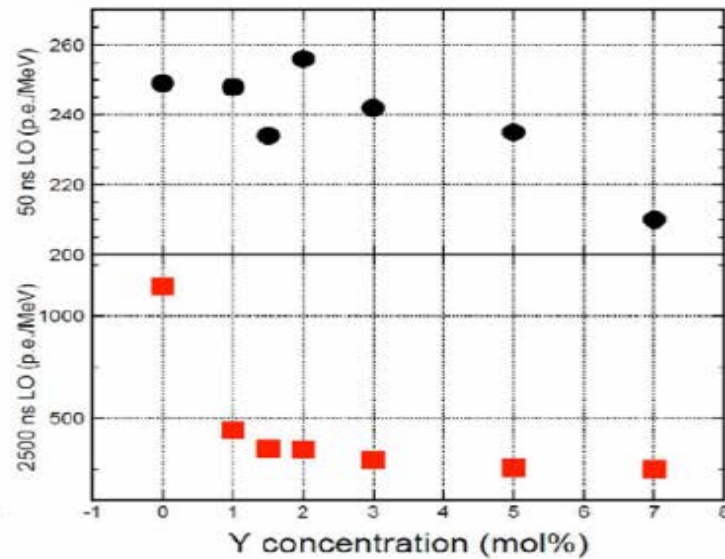
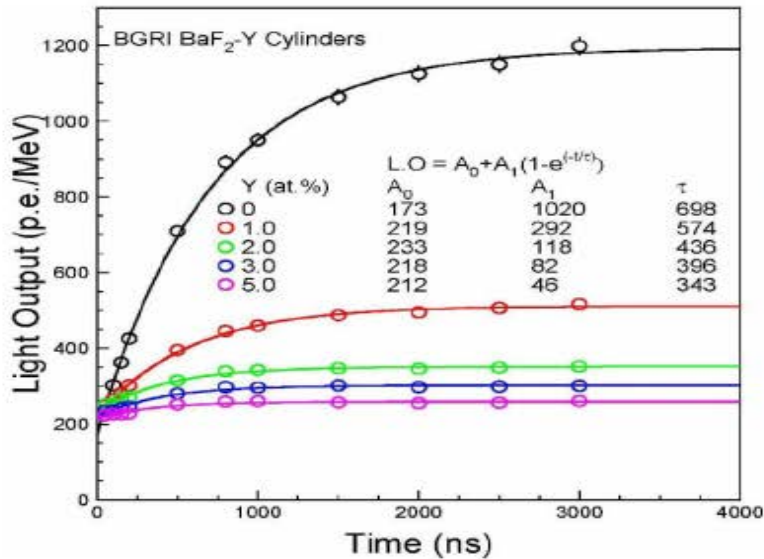
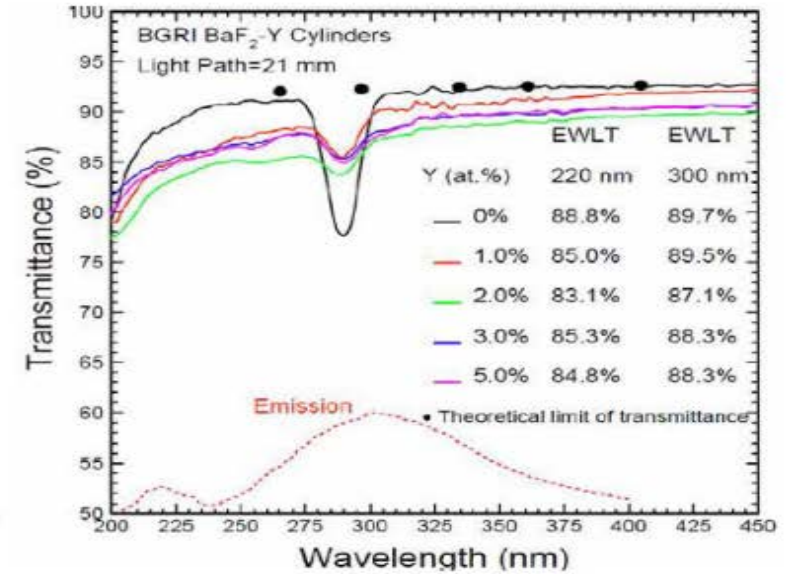
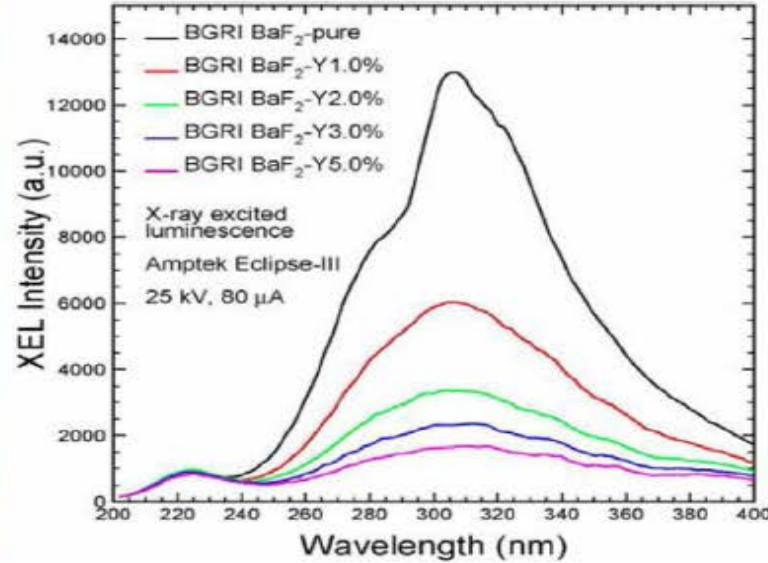
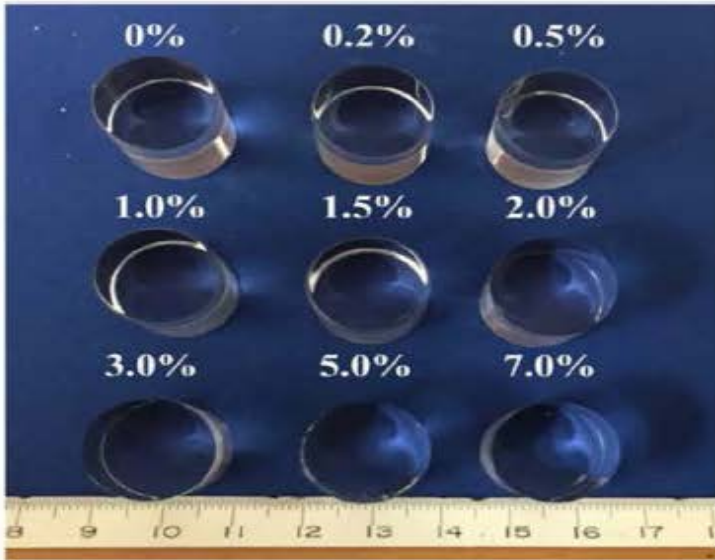
Absorptions observed in La and La/Ce doped BaF₂, published in IEEE TNS 66 (2019) 506-518



Yttrium Doped Small BaF₂ Samples



Increased F/S ratio observed in BGRI BaF₂:Y crystals, Proc. SPIE 10392 (2017)

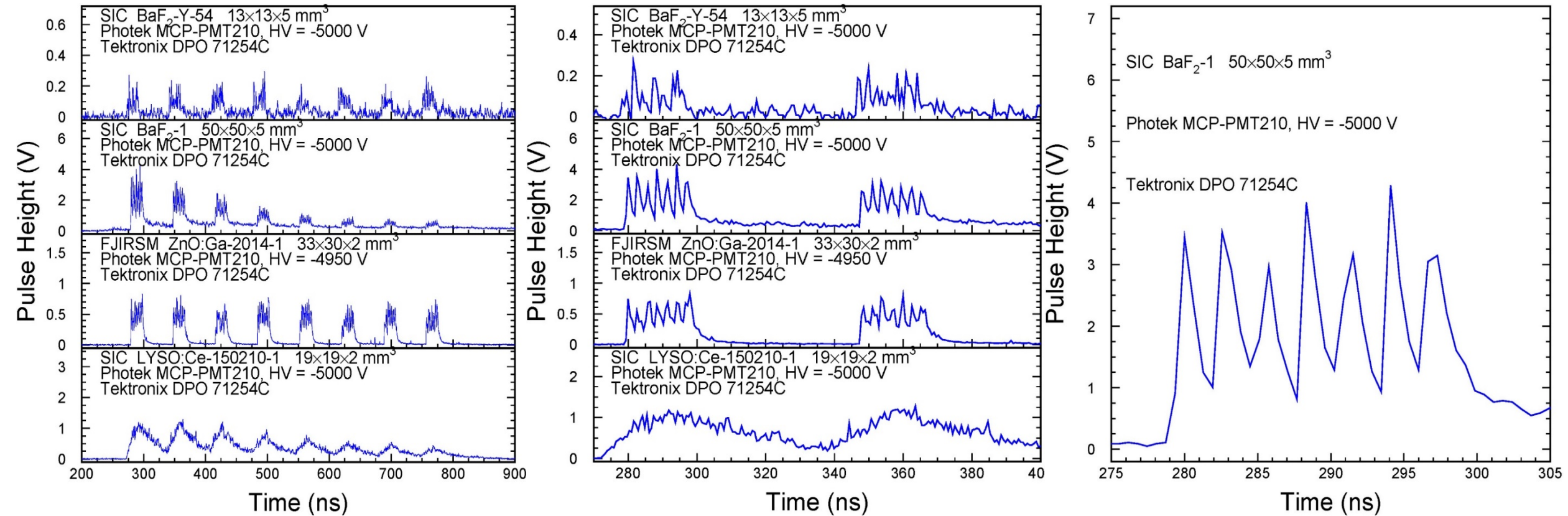




APS Beam Test: BaF₂:Y, BaF₂, ZnO:Ga & LYSO



X-ray bunches with 2.83 ns spacing in septuplet are clearly resolved by ultrafast BaF₂:Y and BaF₂ crystals, NIMA 240 (2019) 223-239



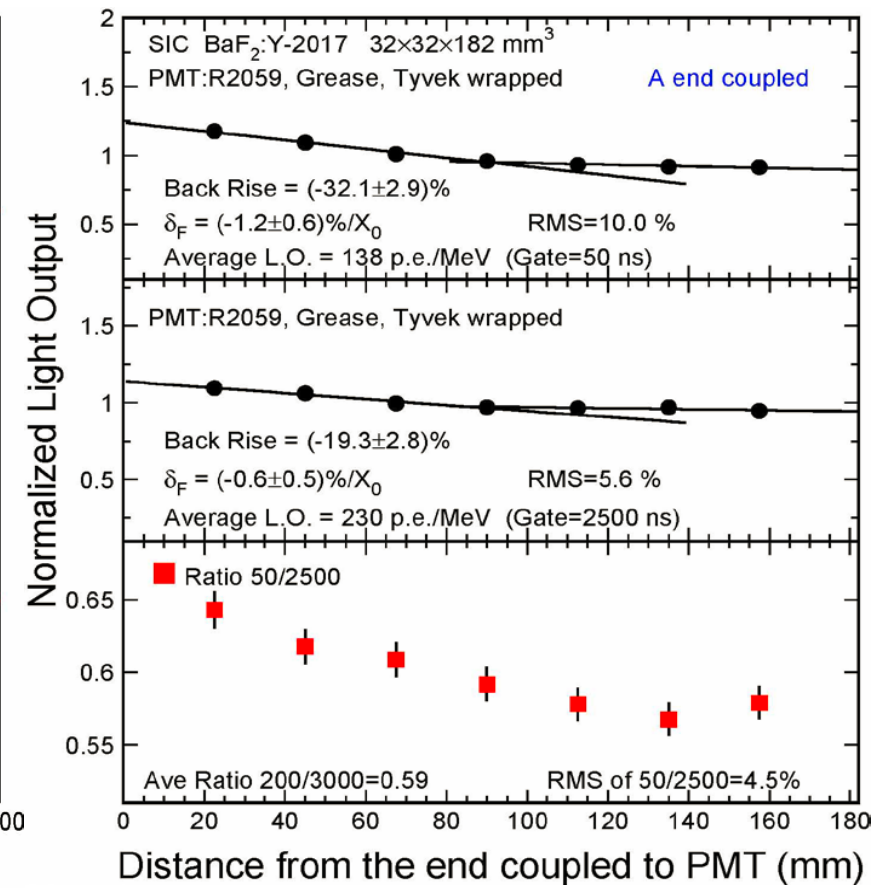
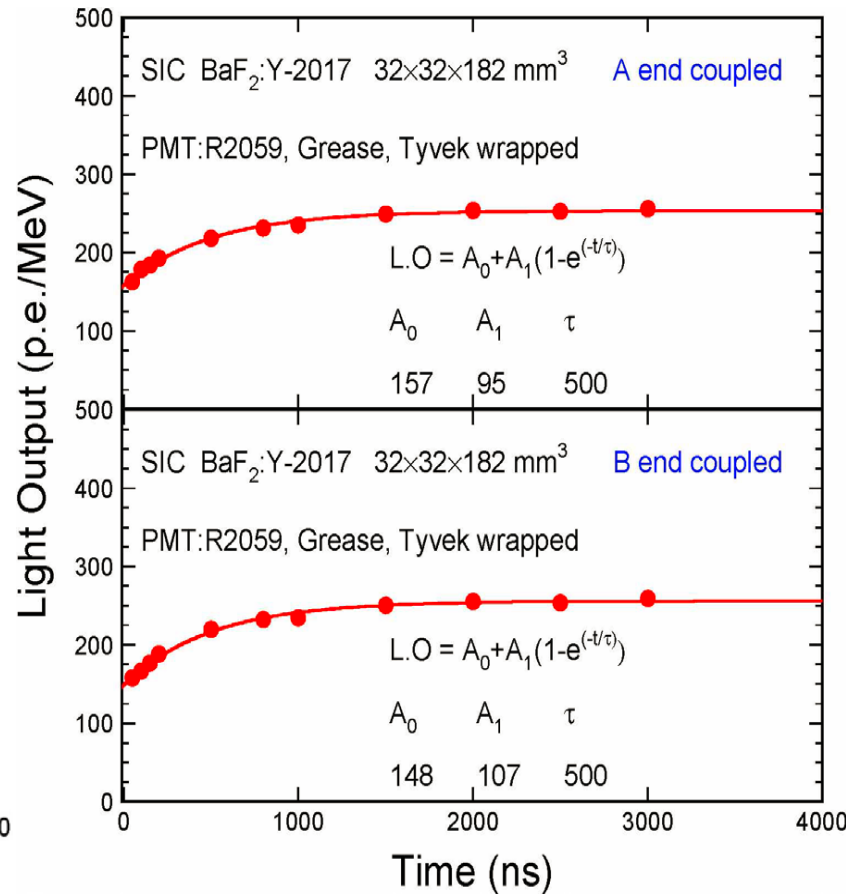
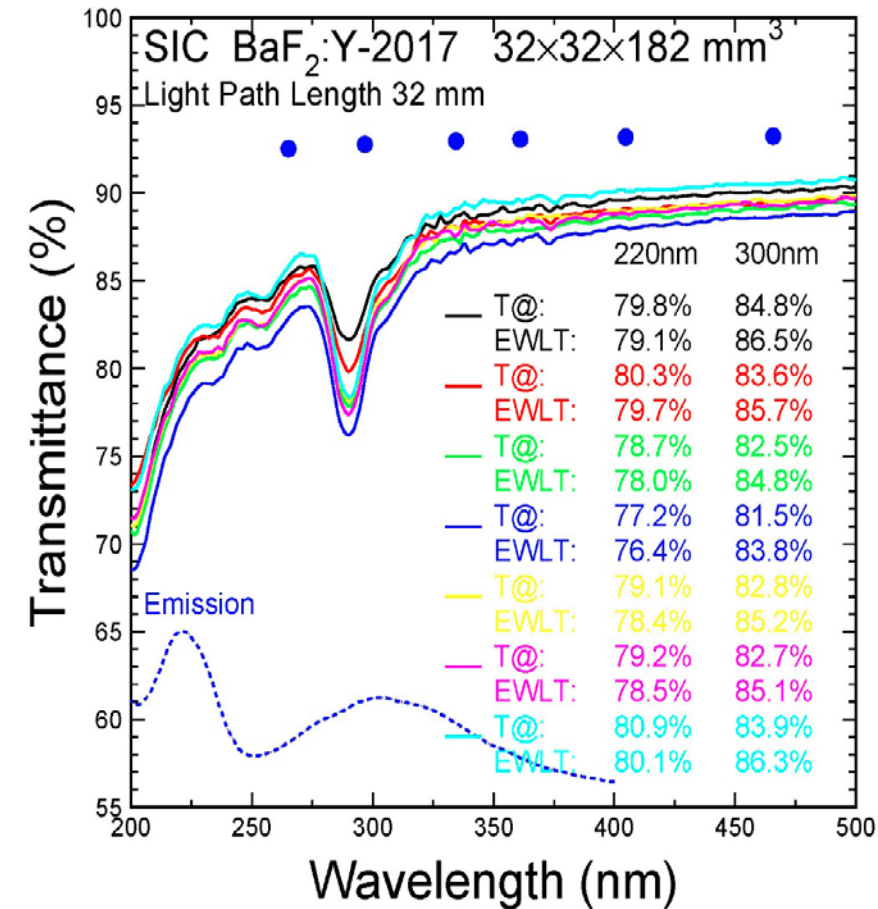
Amplitude reduction in BaF₂ and LYSO due to space charge in PMT from slow scintillation, but not in BaF₂:Y



SIC BaF₂:Y-2017

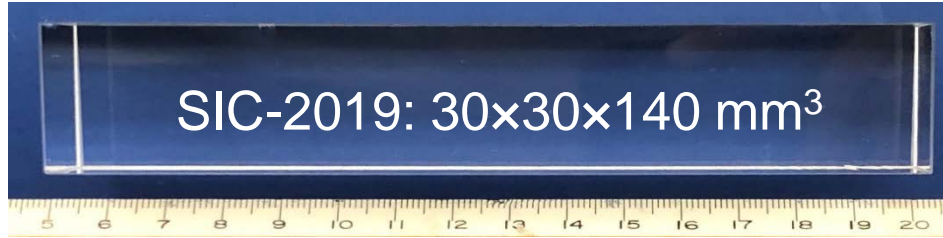


F: 150 p.e./MeV, F/S: 1.5
F/T LRU: 10%/6%, δ_F : -1.2%/X₀

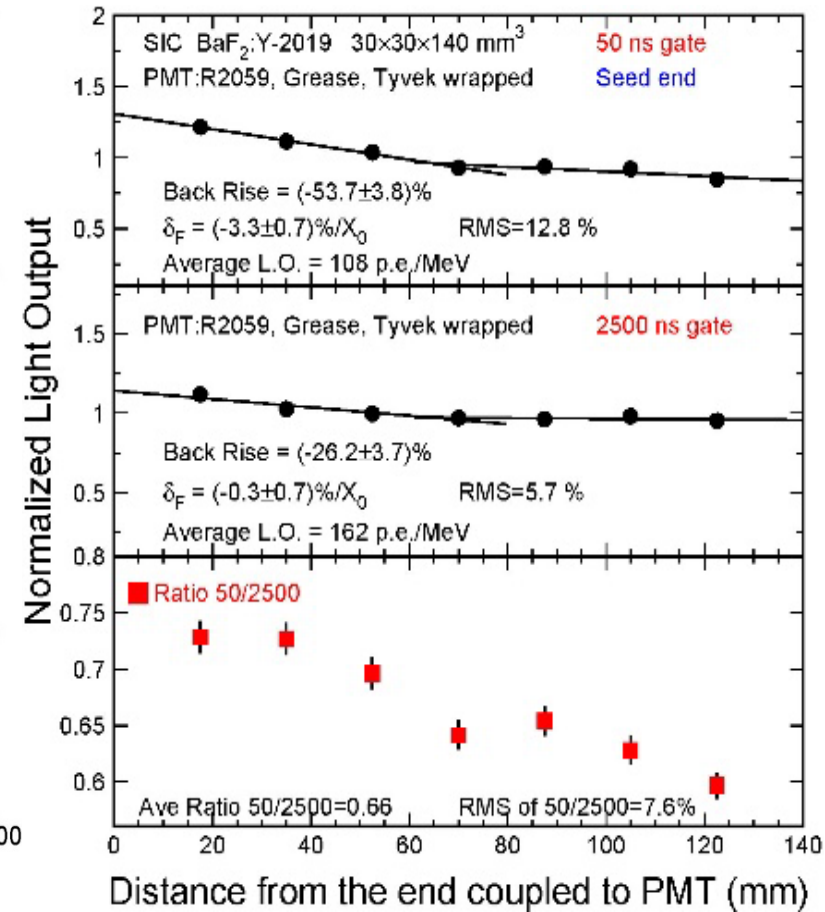
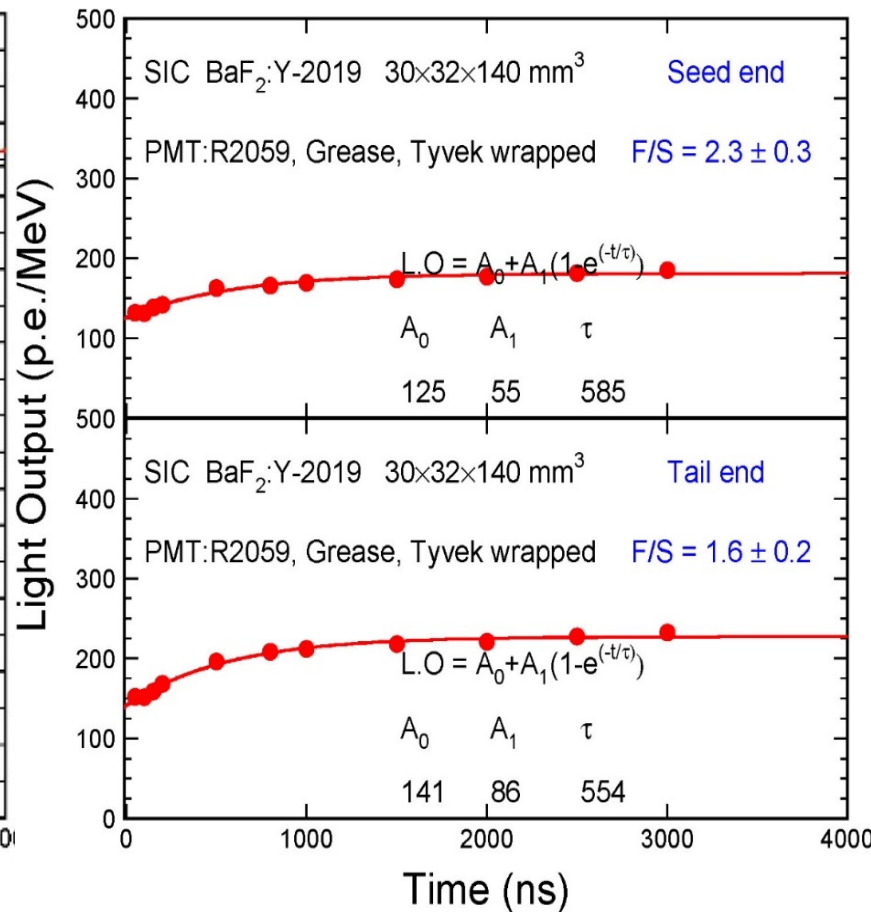
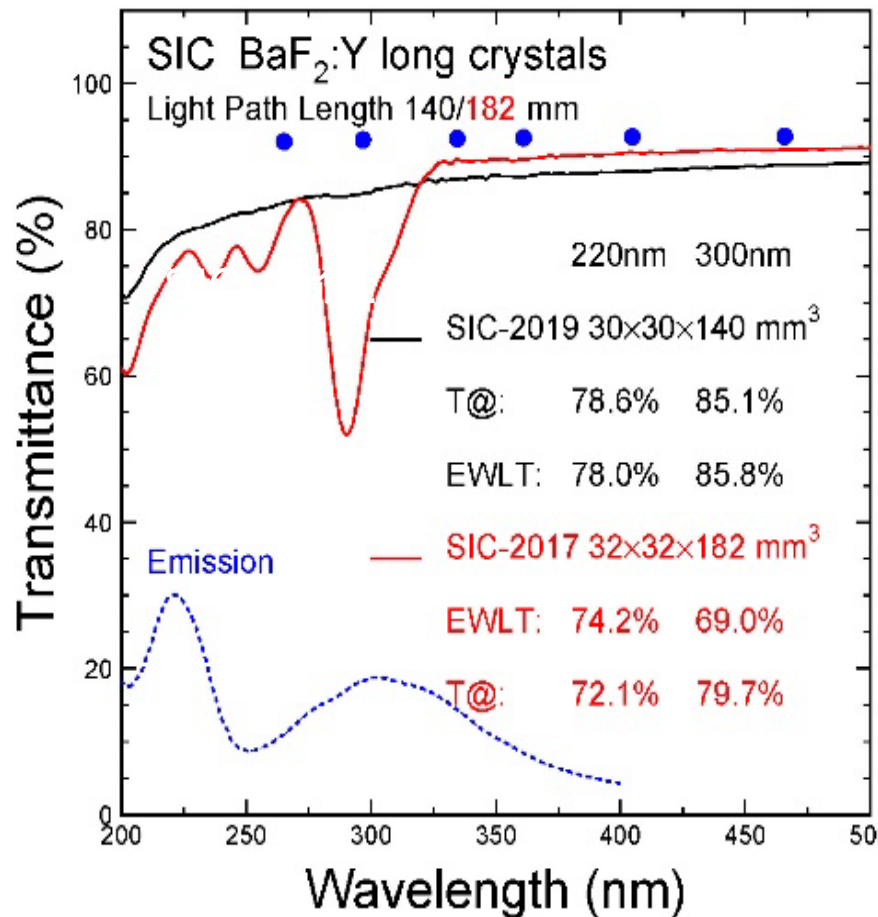




SIC BaF₂:Y-2019

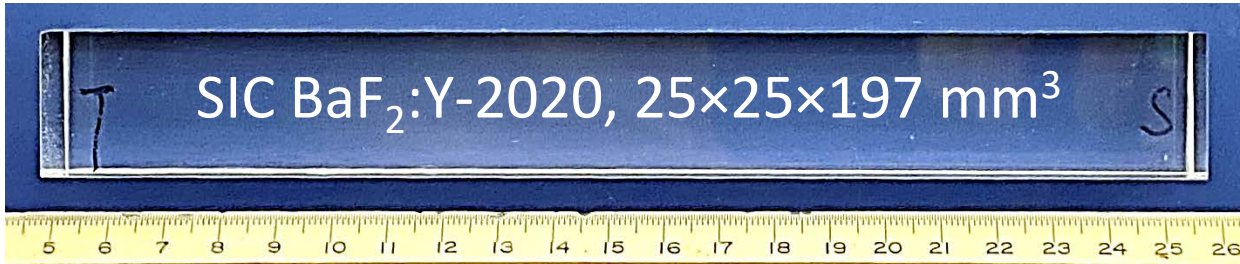


F: 130 p.e./MeV, F/S: 2
F/T LRU: 13%/6% %, δ_F : -3.3%/X₀

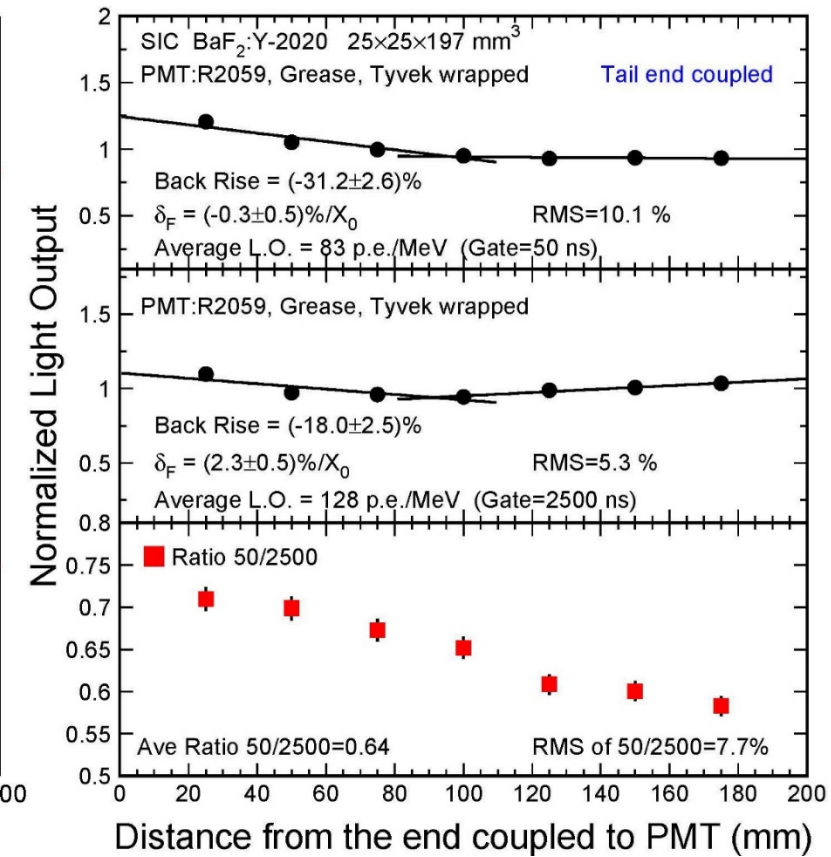
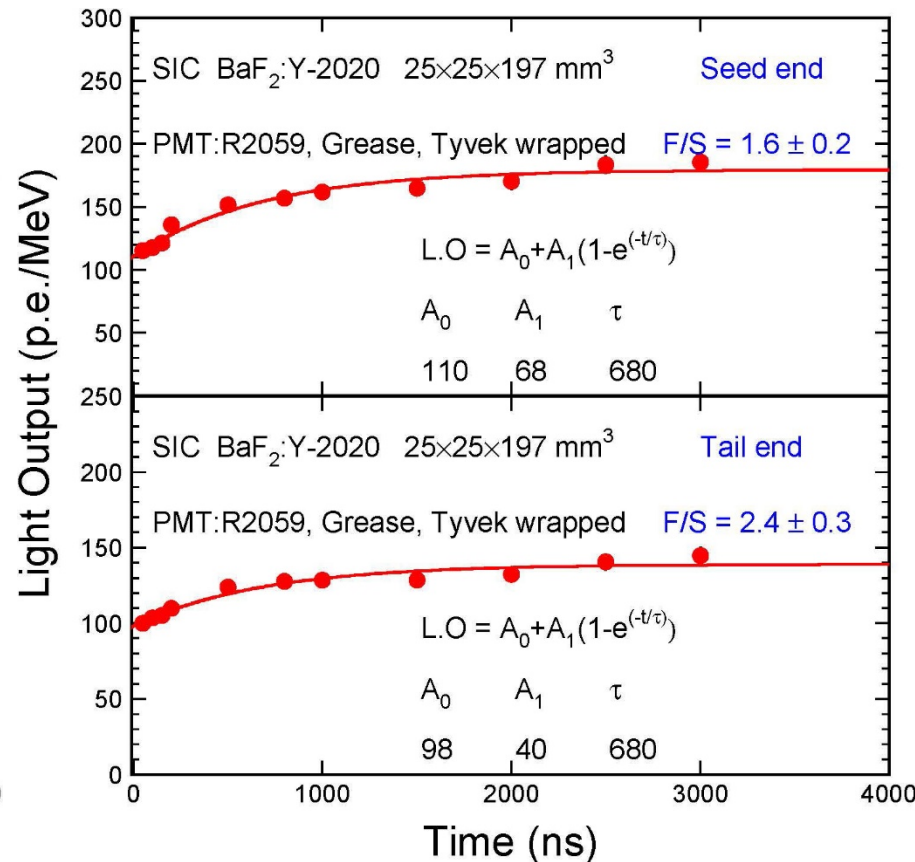
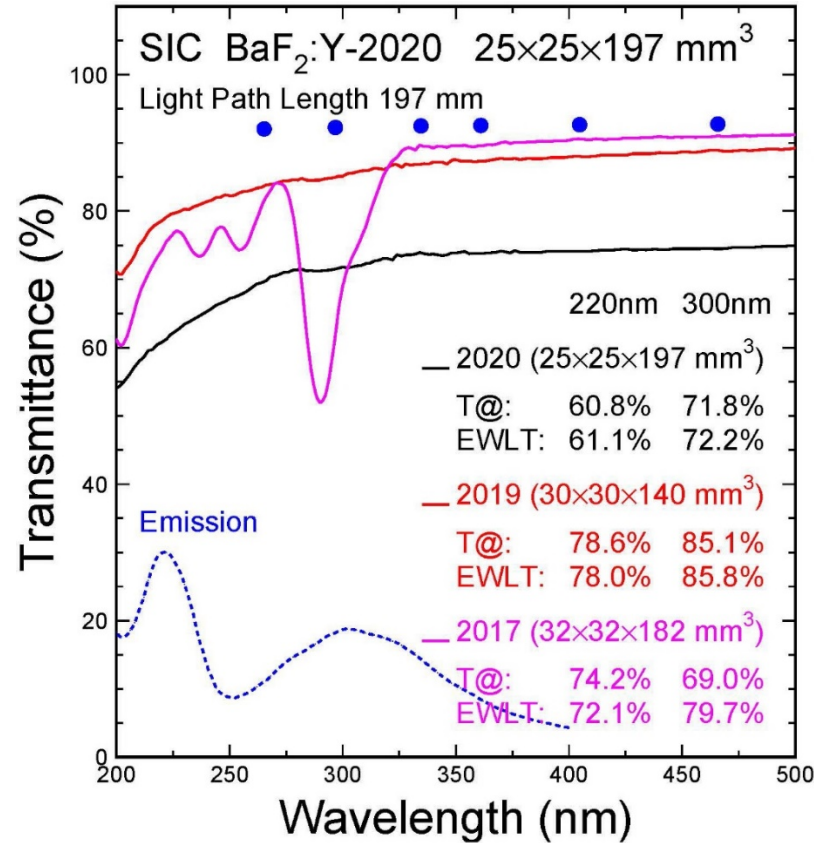




SIC BaF₂:Y-2020



F: 100 p.e./MeV, F/S: 2
F/T LRU: 10%/5% %, δ_F : -0.3%/X₀

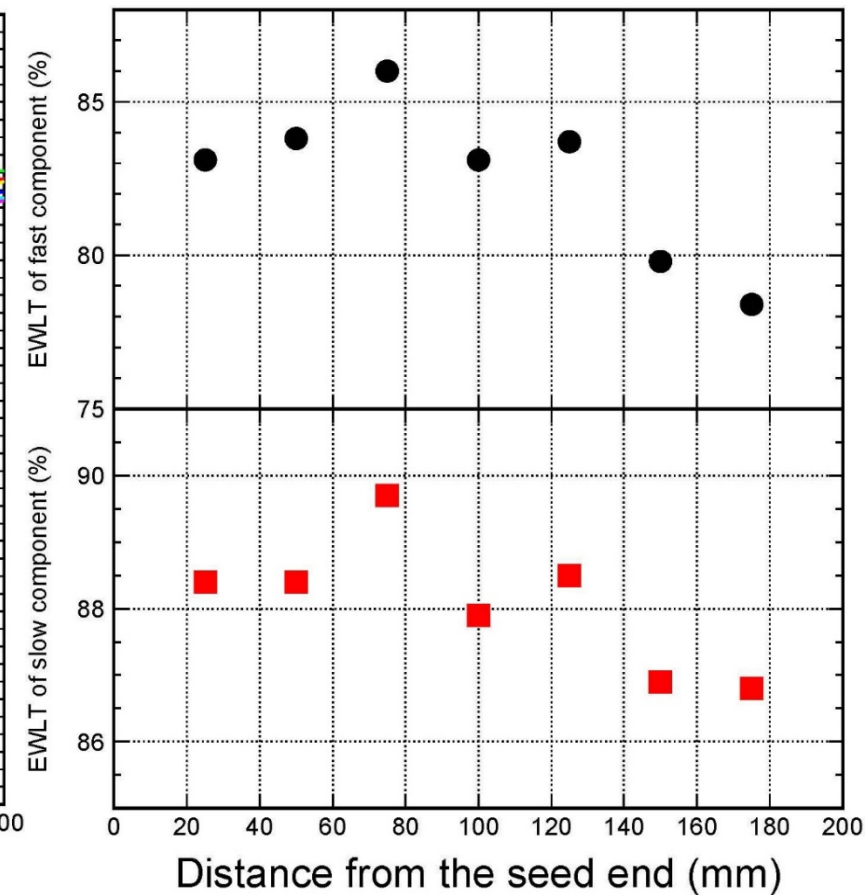
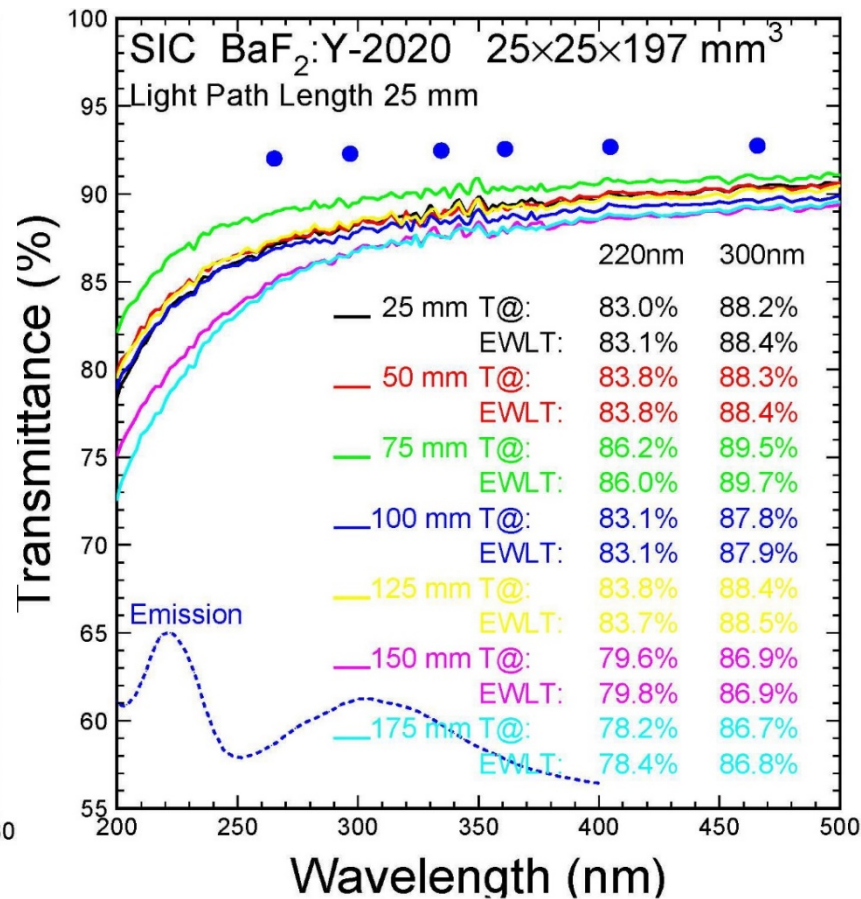
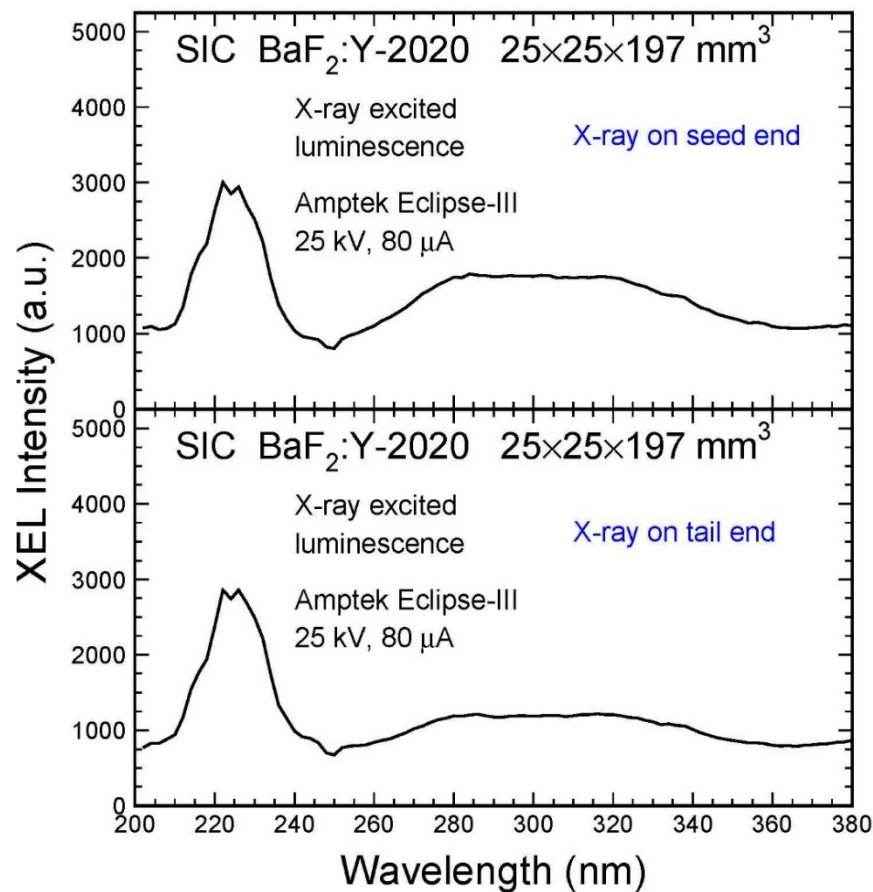




SIC BaF₂:Y-2020: Transverse T



A variation of slow emission intensity and more scattering centers starting from 15 cm from the seed



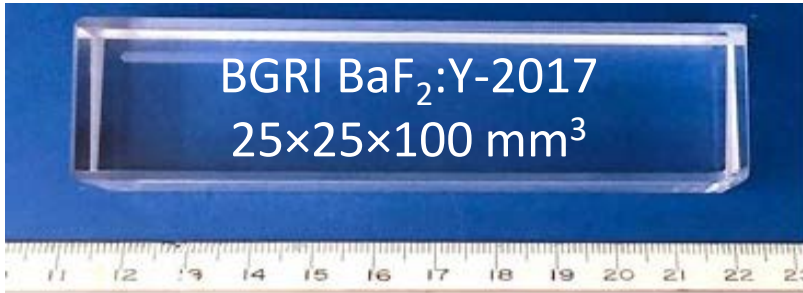


Summary: SIC BaF₂:Y Long Crystals

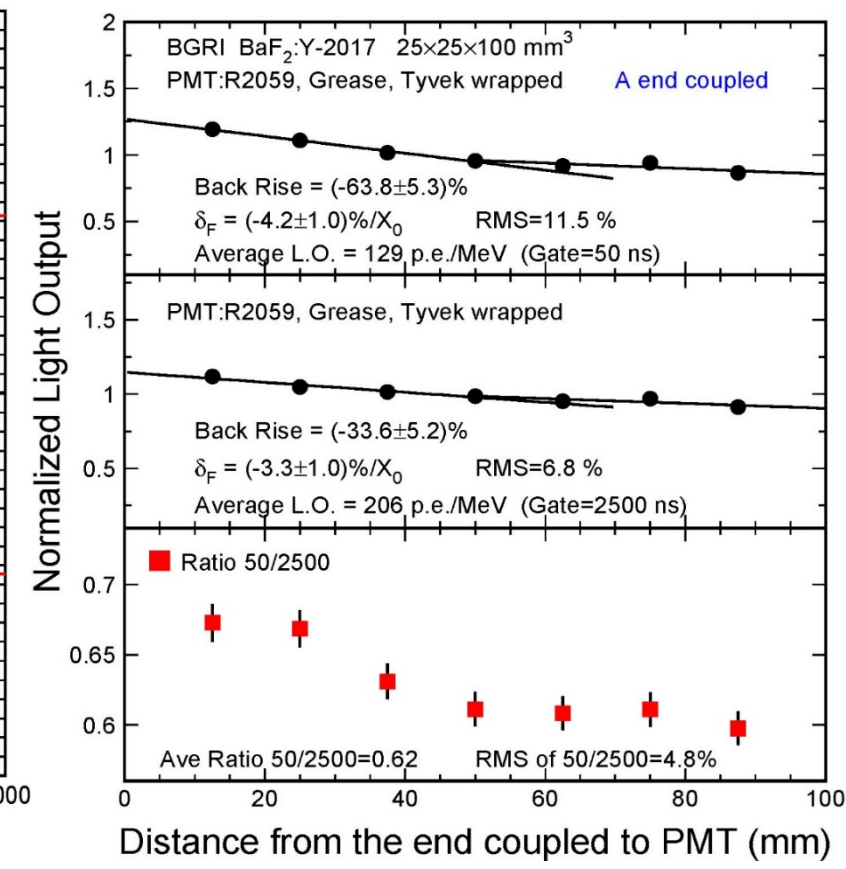
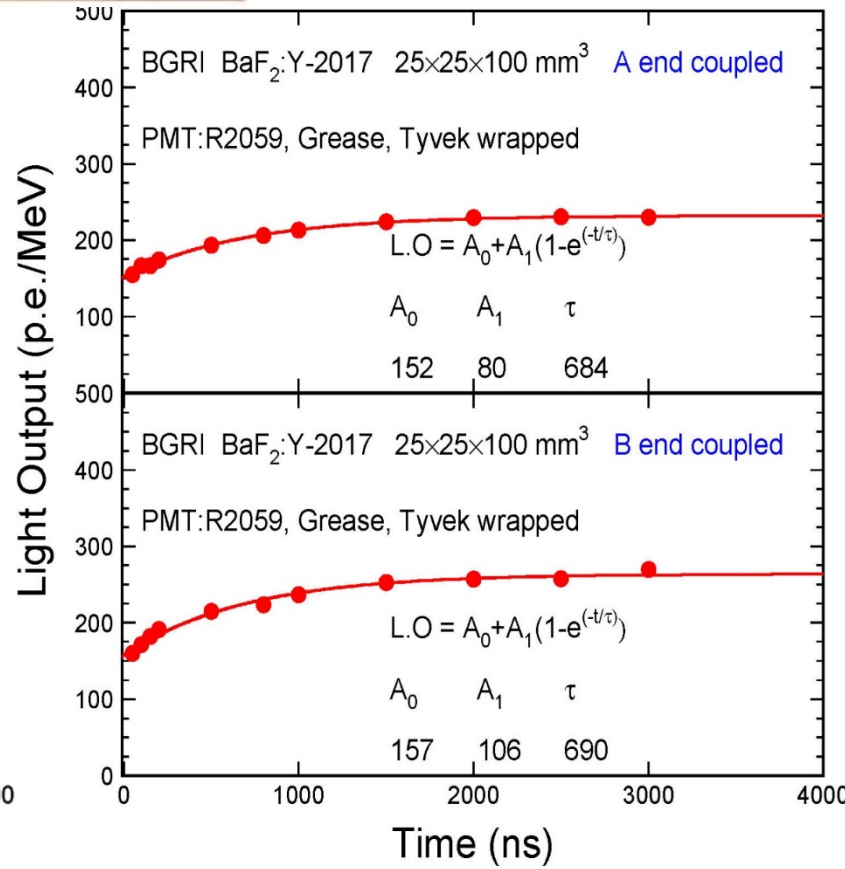
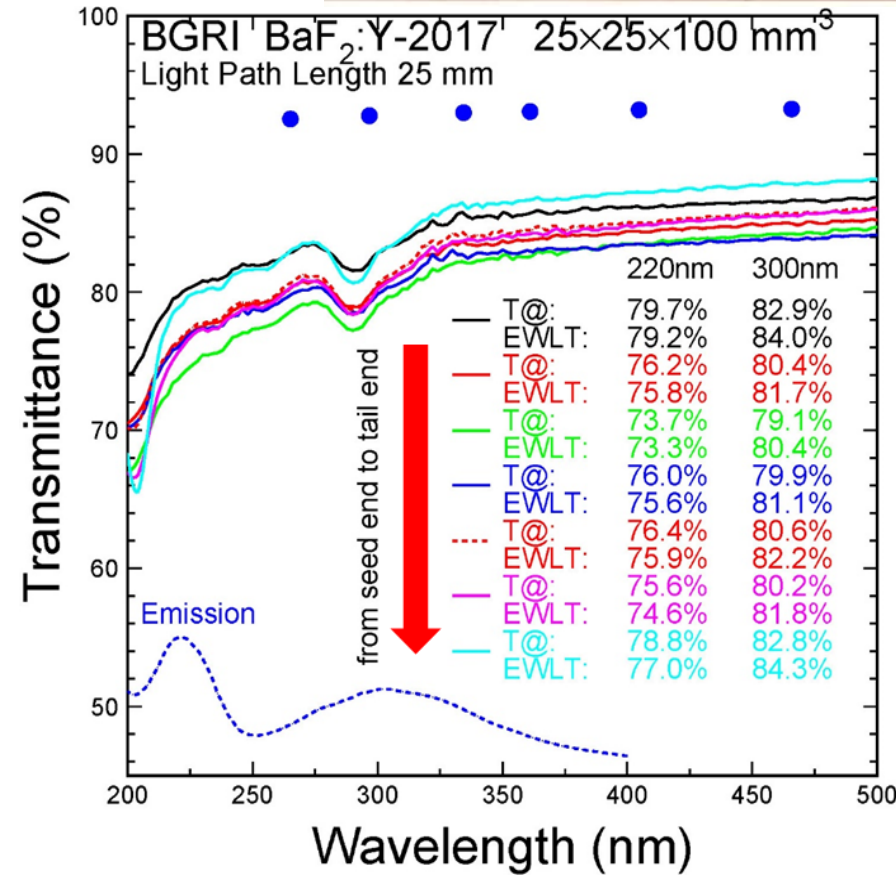
ID	Dimension (mm ³)	EWLT Fast (%)	EWLT Slow (%)	Coupling end	Basic Scintillation Performance ²² Na/ α source @ 1/8 length from the coupling end					Light Response Uniformity		
					50 ns LO (p.e./MeV)	2500 ns LO (p.e./MeV)	LO(50)/LO(2500)	F	F/S	50 ns LO	2500 ns LO	LO(50)/LO(2500)
SIC BaF ₂ :Y-2017	32x32x182	72.1	79.7	A	162	253	0.64	157	1.7	138 (10.0%)	230 (5.6%)	0.59 (4.5%)
				B	158	254	0.62	148	1.4	116 (19.1%)	200 (16.4%)	0.57 (3.7%)
SIC BaF ₂ :Y-2019	30x30x140	78.0	85.8	A	132	181	0.73	125	2.3	108 (12.8%)	162 (5.7%)	0.66 (7.6%)
				B	152	227	0.67	141	1.6	117 (15.6%)	177 (14.9%)	0.66 (1.5%)
SIC BaF ₂ :Y-2020	25x25x197	61.1	72.2	Seed	115	183	0.63	110	1.6	88 (17.7%)	136 (20.5%)	0.64 (2.8%)
				Tail	100	141	0.71	98	2.4	83 (10.1%)	128 (5.3%)	0.64 (7.7%)



BGRI BaF₂:Y-2017

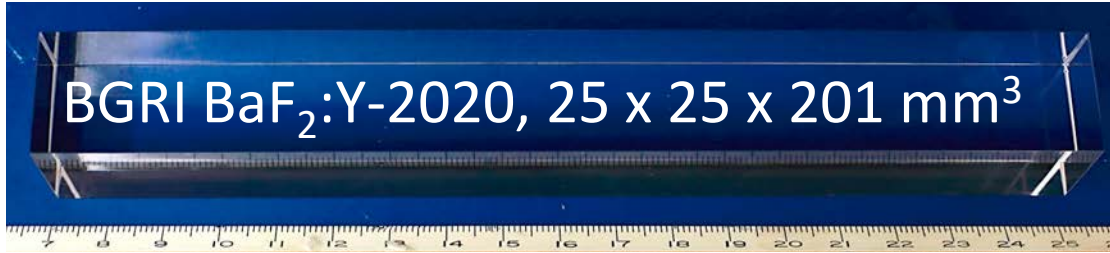


F: 150 p.e./MeV, F/S: 1.5
F/T LRU: 12%/7% %, δ_F : -4.2%/X₀

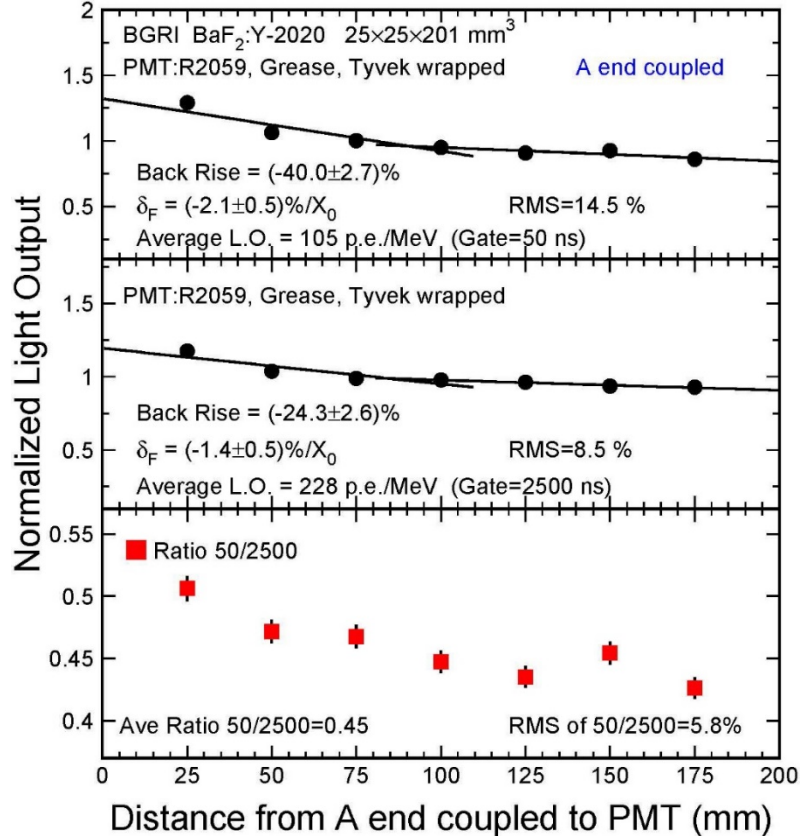
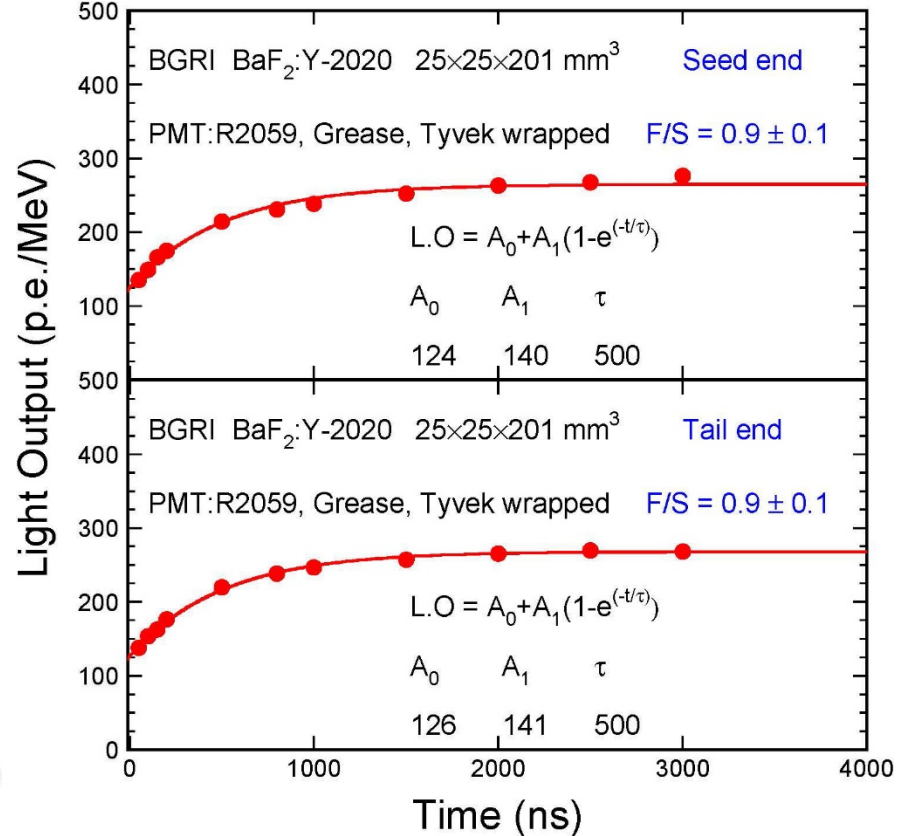
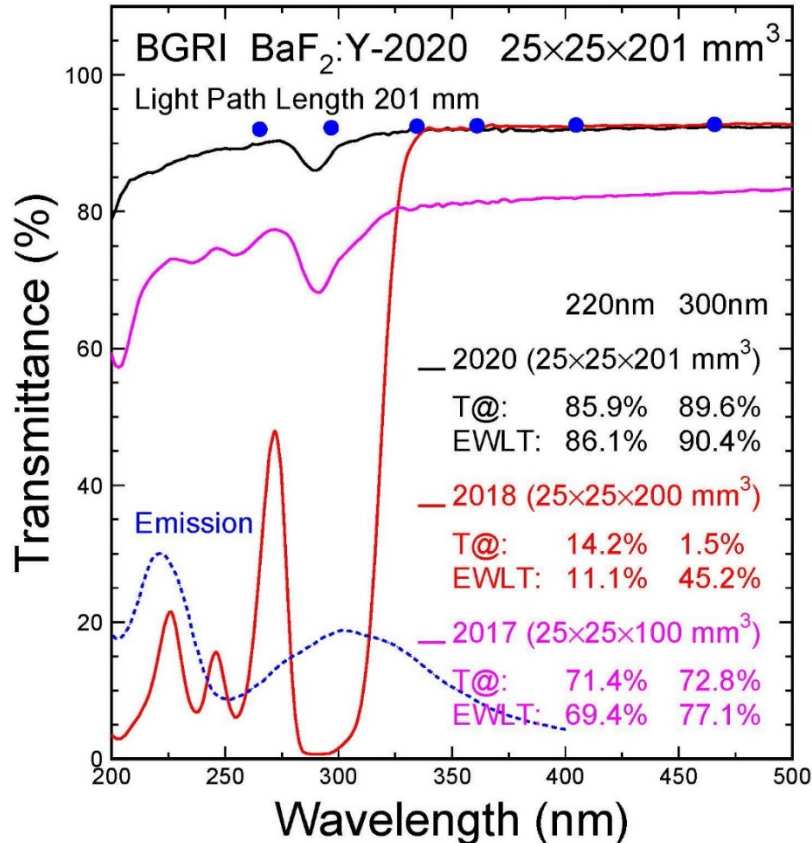




BGRI BaF₂:Y-2020



F: 125 p.e./MeV, F/S: 0.9
F/T LRU: 15%/9% %, δ_F : -2.1%/X₀

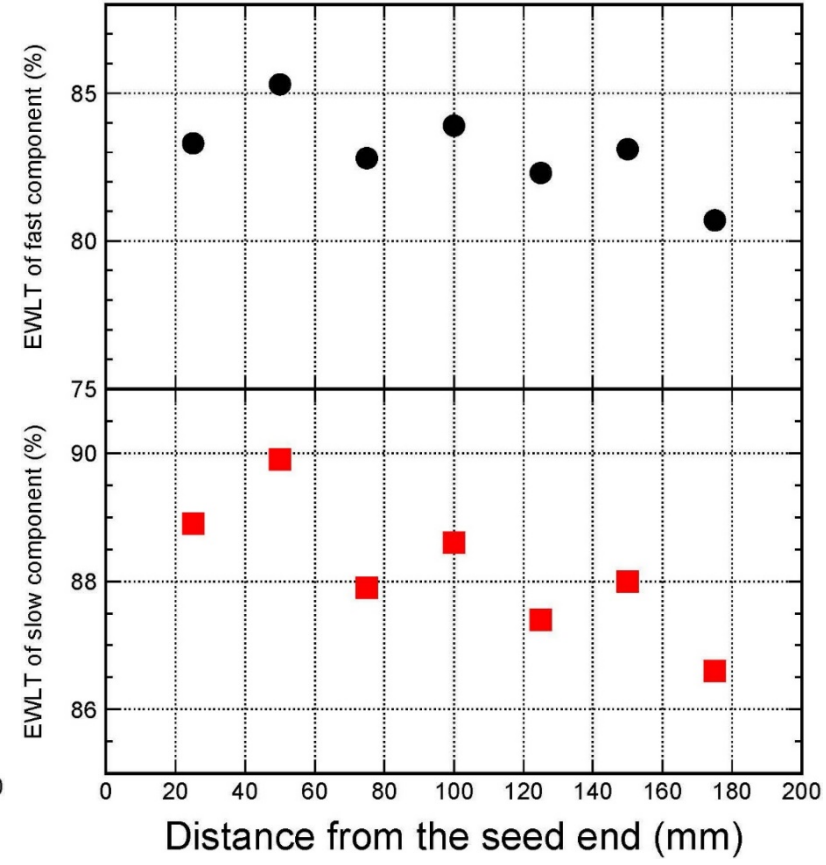
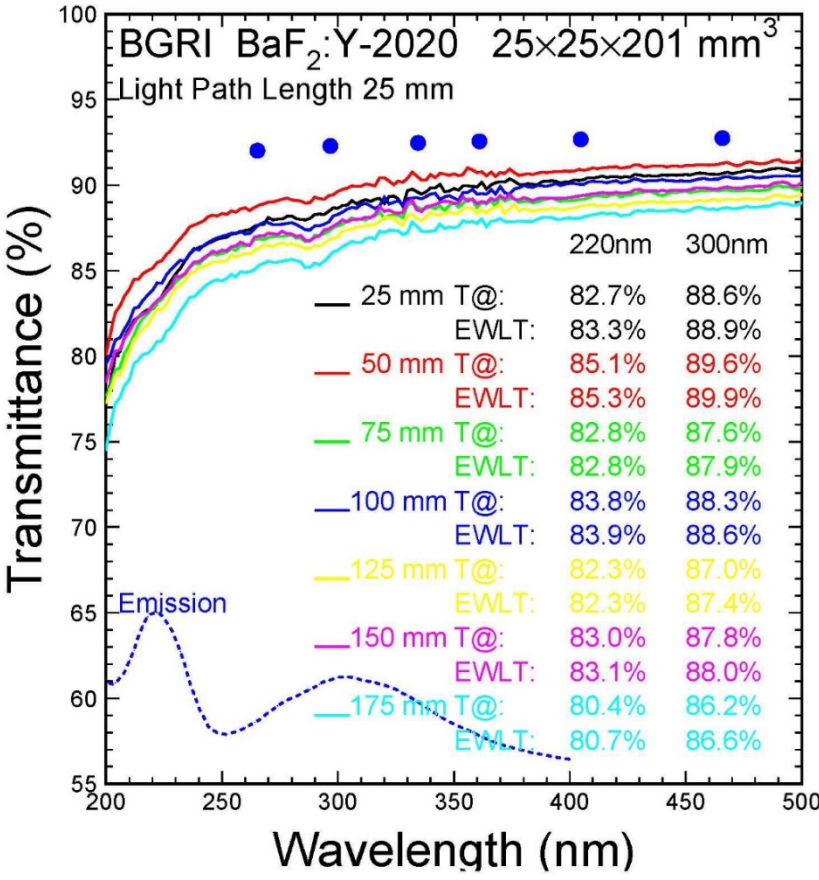
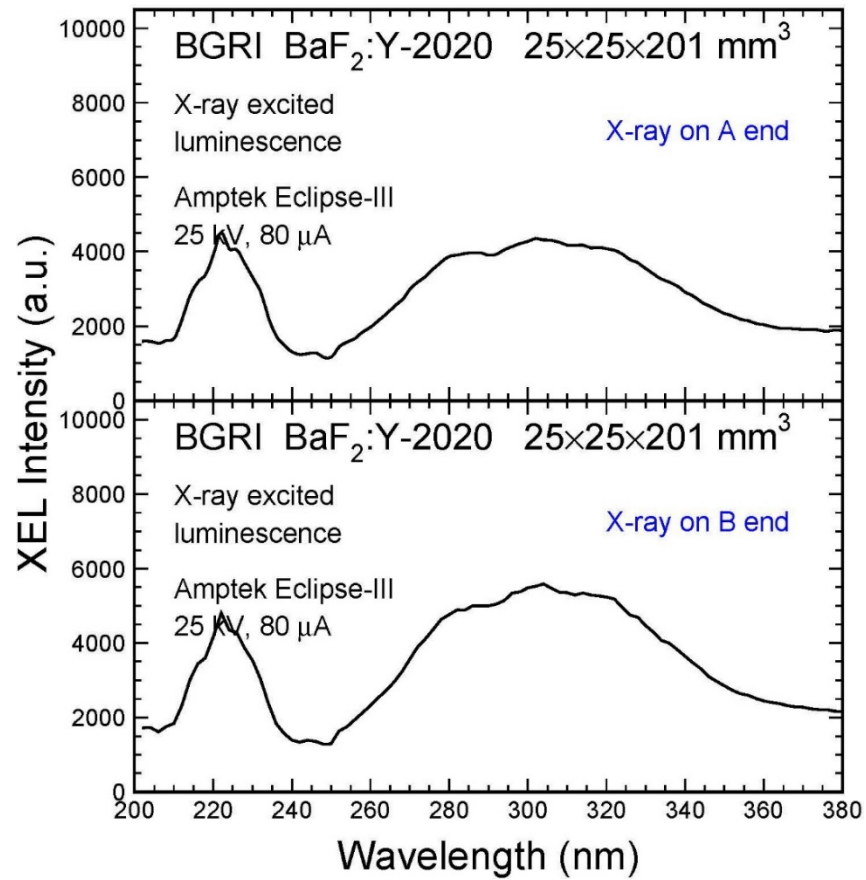




BGRI BaF₂:Y-2020: Transverse T



A variation of slow emission intensity and good optical quality along the crystal length





Summary: BGRI BaF₂:Y Long Crystals



ID	Dimension (mm ³)	EWLT Fast (%)	EWLT Slow (%)	Coupling end	Basic Scintillation Performance Source @ 1/8 length from the coupling end					Light Response Uniformity		
					50 ns LO (p.e./MeV)	2500 ns LO (p.e./MeV)	LO(50)/LO(2500)	F	F/S	50 ns LO	2500 ns LO	LO(50)/LO(2500)
BGRI BaF ₂ :Y-2017	25×25×100	69.4	77.1	A	155	231	0.67	152	1.9	129 (11.5%)	206 (6.8%)	0.62 (4.8%)
				B	160	258	0.62	157	1.5	129 (15.4%)	214 (13.7%)	0.60 (2.1%)
BGRI BaF ₂ :Y-2018	25×25×200	11.1	45.2	A	133	317	0.42	203*	NA	83 (30.6%)	229 (20.4%)	0.35 (9.4%)
				B	133	265	0.52	159*	NA	89 (26.4%)	228 (8.7%)	0.38 (17.2%)
BGRI BaF ₂ :Y-2020	25×25×201	61.1	72.2	A	135	268	0.50	124	0.9	105 (14.5%)	228 (8.5%)	0.45 (5.8%)
				B	138	270	0.51	126	0.9	106 (17.1%)	221 (14.7%)	0.47 (3.1%)

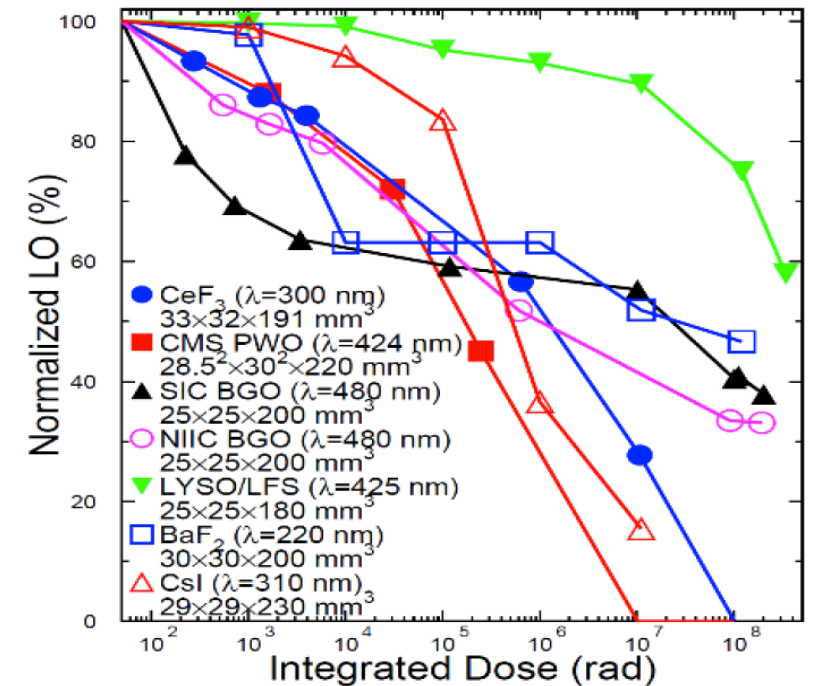
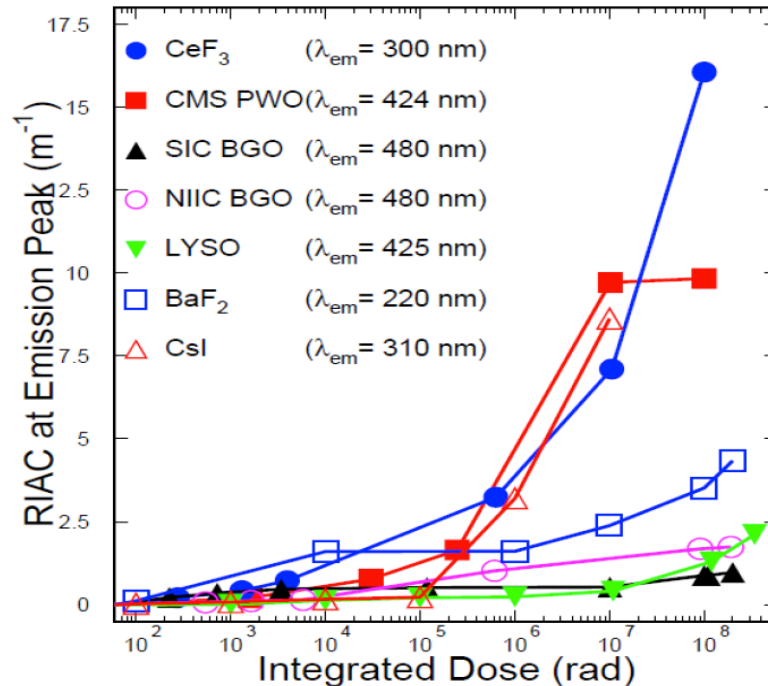
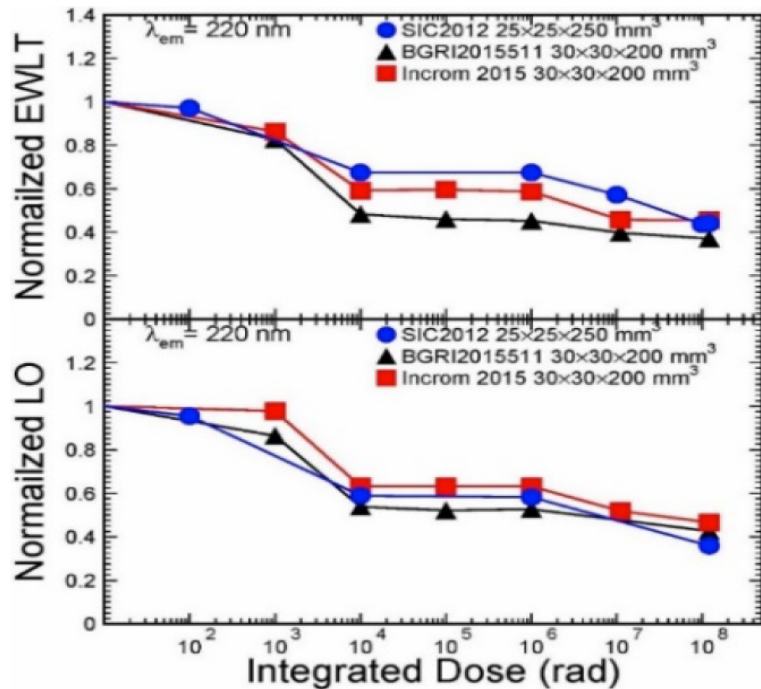
*Only one component with 30~50 ns decay time is observed, but no ultrafast component



γ -Ray Induced Damage in Large BaF₂

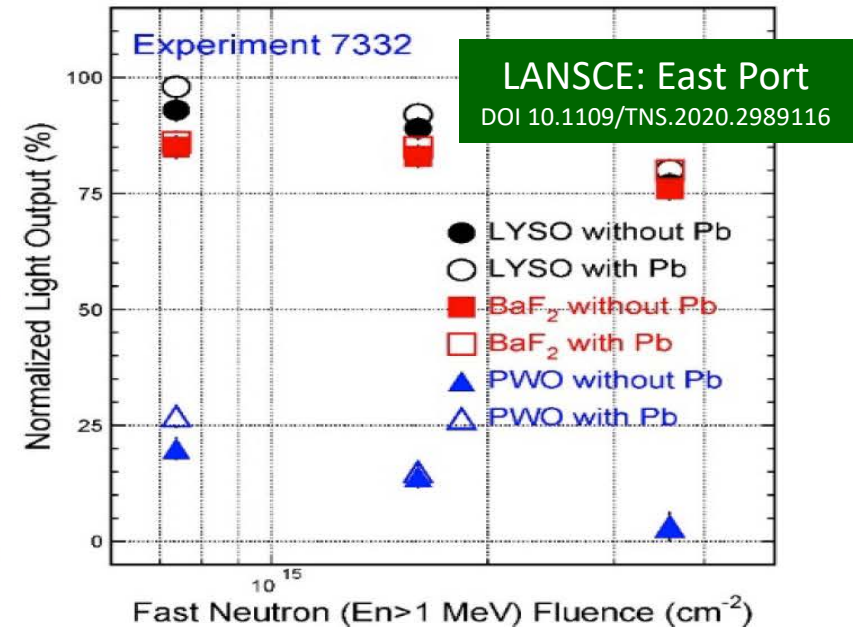
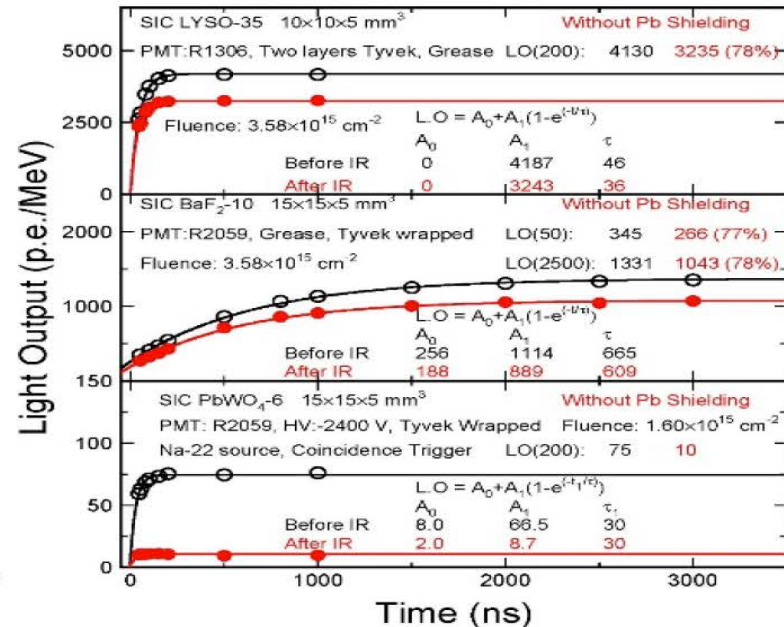
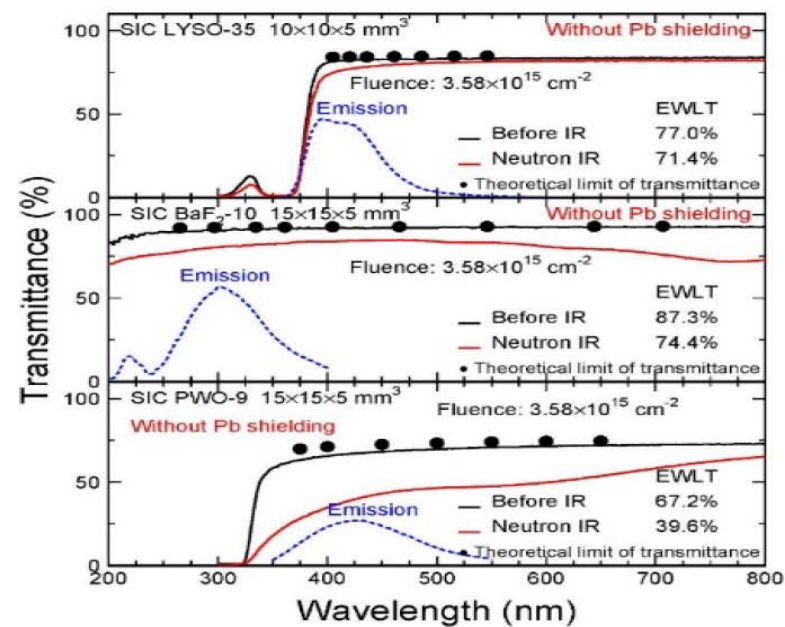
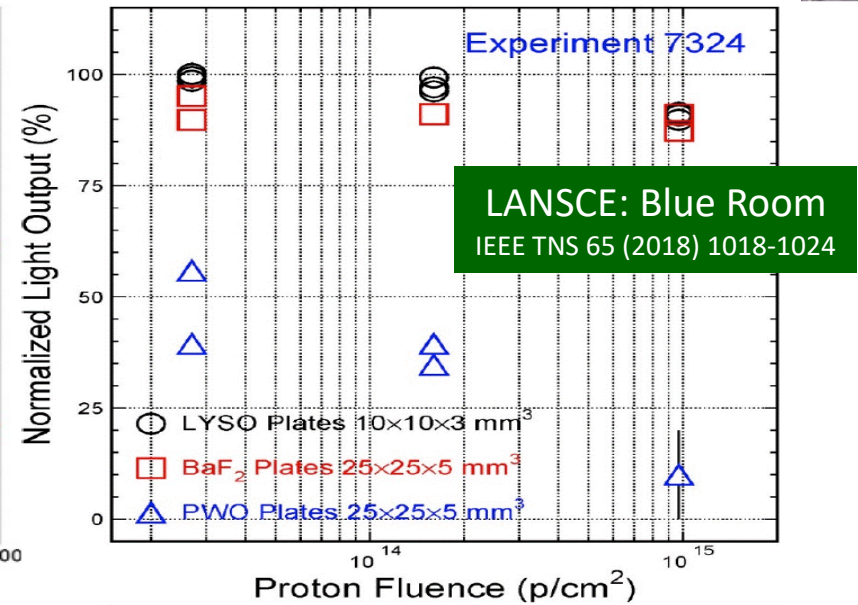
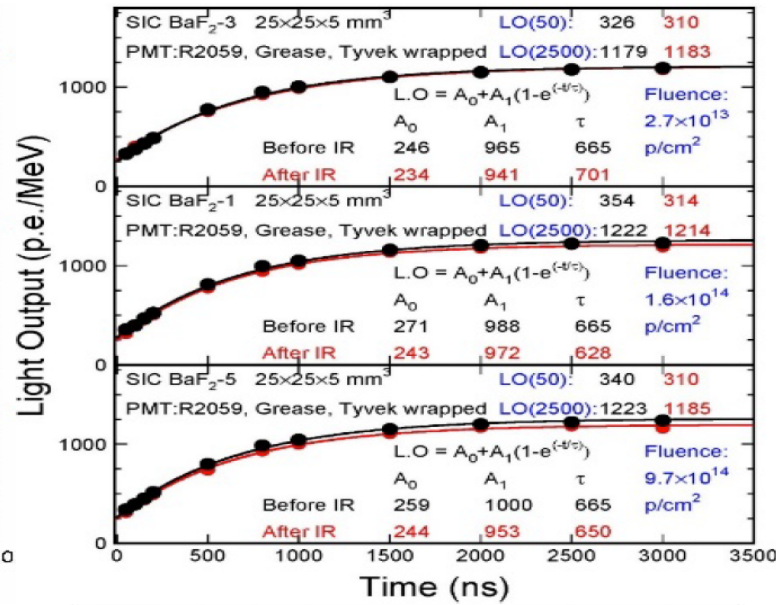
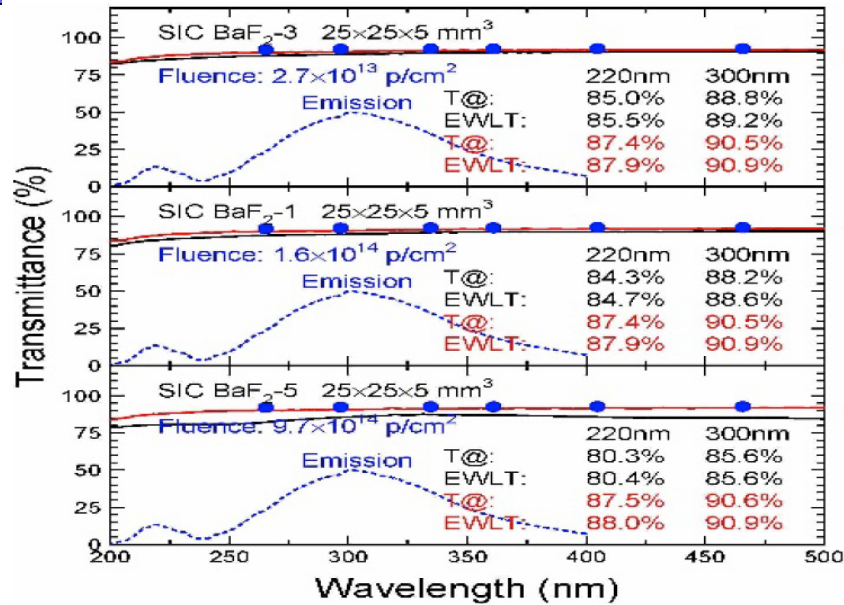
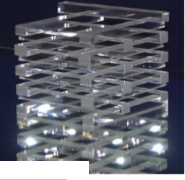


BaF₂ shows saturated damage from 10 krad to 100 Mrad, indicating good radiation resistance against γ -rays, IEEE TNS 63 (2016) 612-619





Proton and Neutron Induced Damage in BaF₂

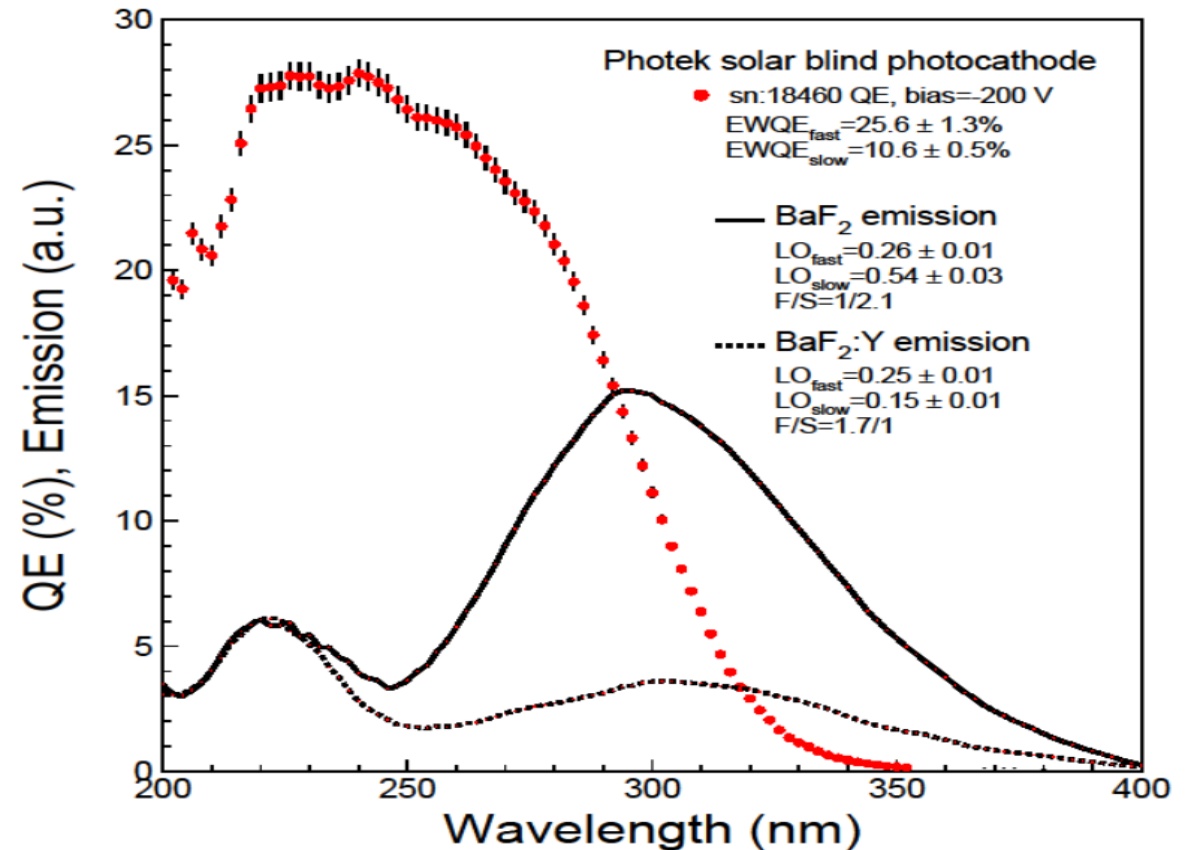
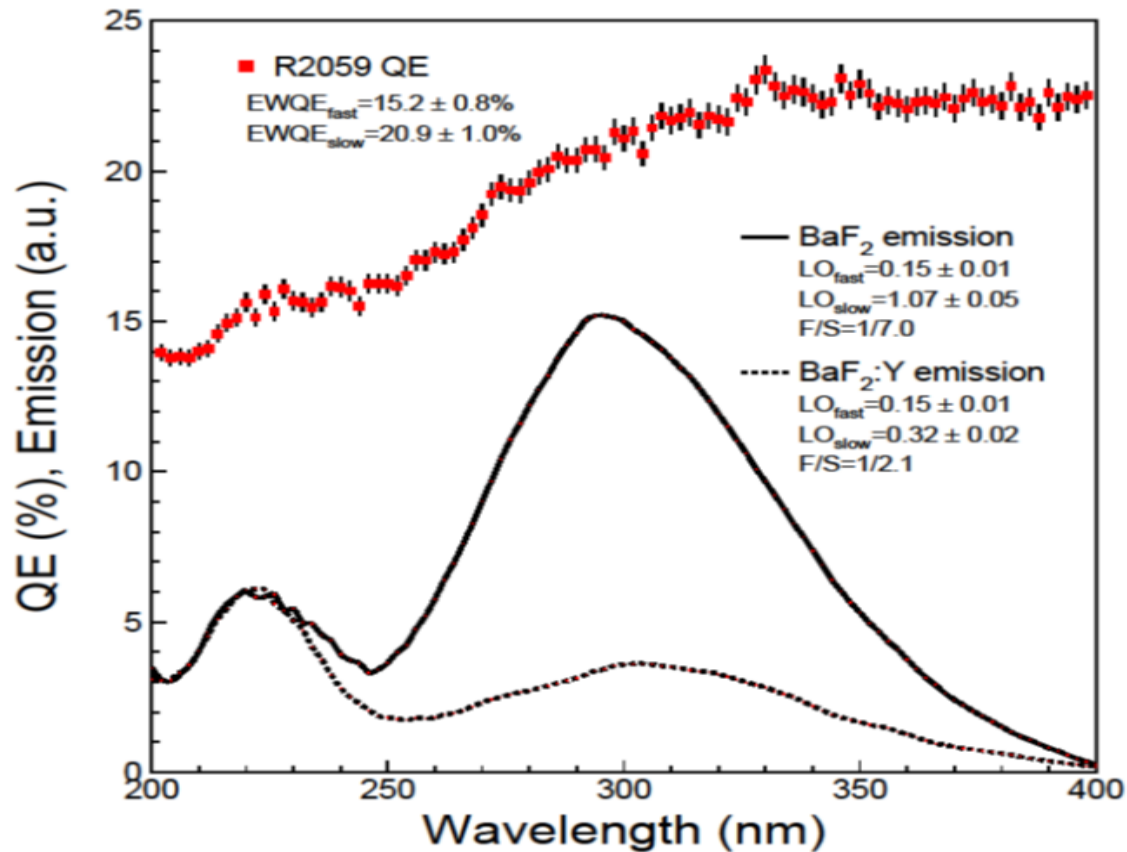




VUV PMT for BaF₂ and BaF₂:Y



Photo-detectors	EWQE _{fast} (%)	EWQE _{slow} (%)	BaF ₂ LO _{fast}	BaF ₂ LO _{slow}	BaF ₂ F/S	BaF ₂ :Y LO _{fast}	BaF ₂ :Y LO _{slow}	BaF ₂ :Y F/S
Hamamatsu R2059	15.2	20.9	0.15	1.07	1/7.0	0.15	0.32	1/2.1
Photek solar blind PMT	25.6	10.6	0.26	0.54	1/2.1	0.25	0.15	1/0.6

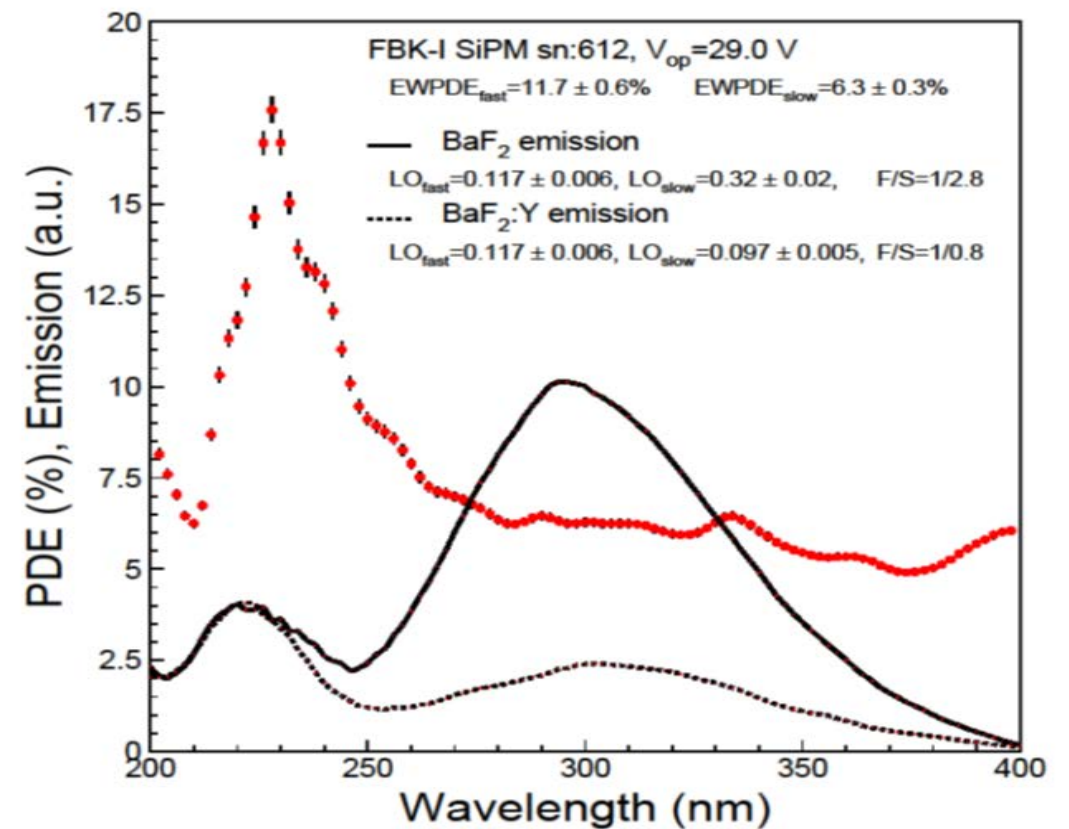
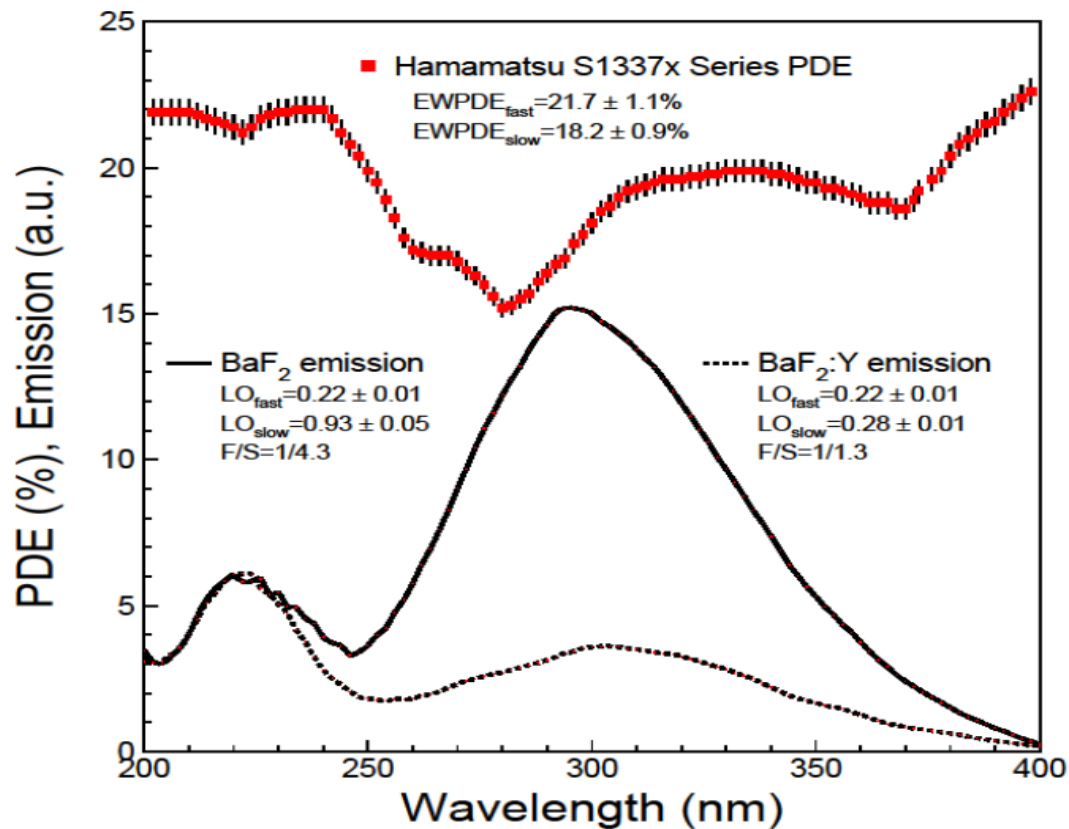




VUV SiPM for BaF₂ and BaF₂:Y



Photo-detectors	EWQE _{fast} (%)	EWQE _{slow} (%)	BaF ₂ LO _{fast}	BaF ₂ LO _{slow}	BaF ₂ F/S	BaF ₂ :Y LO _{fast}	BaF ₂ :Y LO _{slow}	BaF ₂ :Y F/S
Hamamatsu s1337x	21.7	18.2	0.22	0.93	1/4.3	0.22	0.28	1/1.3
FBK-I SiPM	11.7	6.3	0.12	0.32	1/2.8	0.12	0.097	1/0.8





Summary



- ❑ Undoped BaF₂ crystals provide adequate ultrafast light with 0.5 ns decay time. Yttrium doping increases its F/S ratio while maintaining the ultrafast intensity. With sub-ns pulse width BaF₂:Y promises an ultrafast calorimeter for Mu2e-II.
- ❑ 20 cm long BaF₂ crystals are rad hard up to 120 Mrad against ionization dose. 5 mm thick BaF₂ plates irradiation at LANSCE by 800 MeV protons up to 1×10^{15} p/cm² and fast neutrons up to 3.6×10^{15} n_{eq}/cm² did not cause significant light output loss, indicating BaF₂ may be used in a severe radiation environment.
- ❑ 20 cm long BaF₂:Y may reach LO_F>100 p.e./MeV, F/S>2, 10% LRU and $|\delta_F| < 3\%/X_0$. R&D will continue to optimize yttrium doping in large size BaF₂:Y crystals for Mu2e-II.
 - ❑ SIC plans to reduce scattering centers by refining growth parameters;
 - ❑ BGRI plans to eliminate residual cerium contamination by purifying raw material.
 - ❑ Caltech plans to investigate radiation hardness of BaF₂:Y crystals.
- ❑ Effort is also needed to develop VUV photodetector, such as solar-blind SiPM, LAPPD or diamond-based photodetectors.

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Diamond Photodetector



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

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

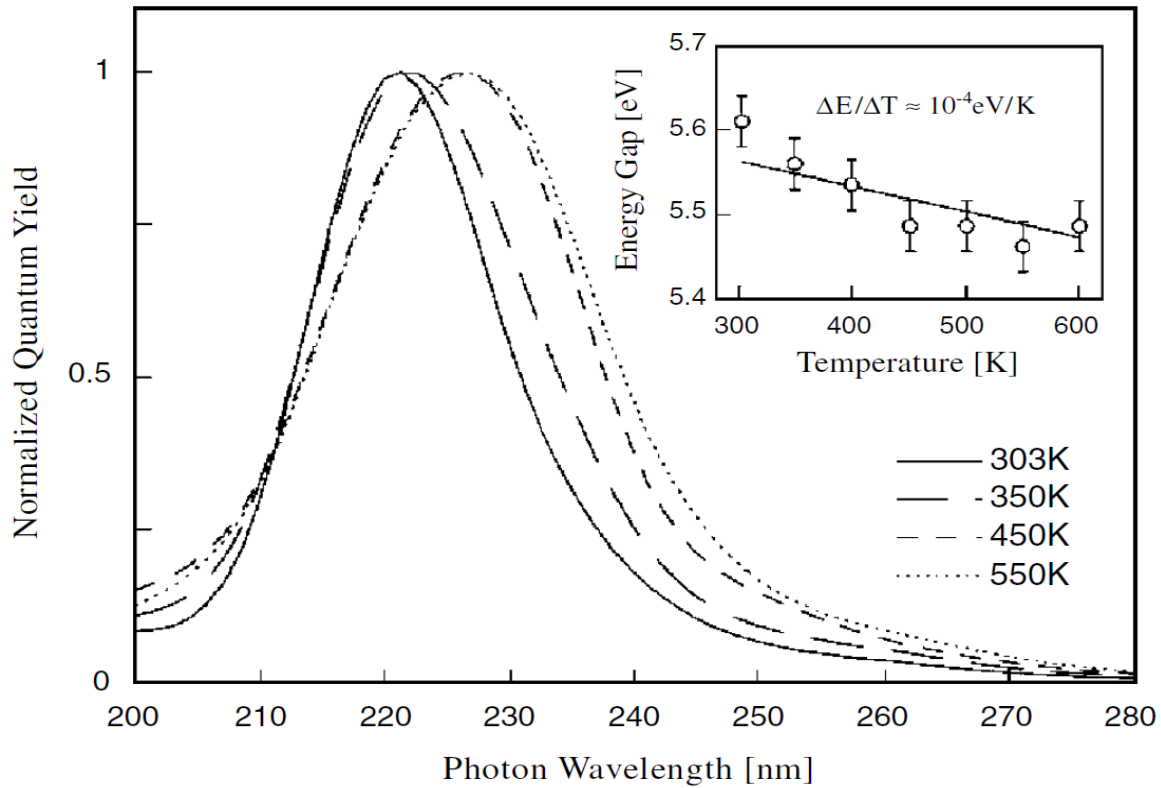


Figure 6. Quantum efficiency of diamond photoconductors at different temperatures and Arrhenius plot of the peak value (inset). (From [Sal00].)

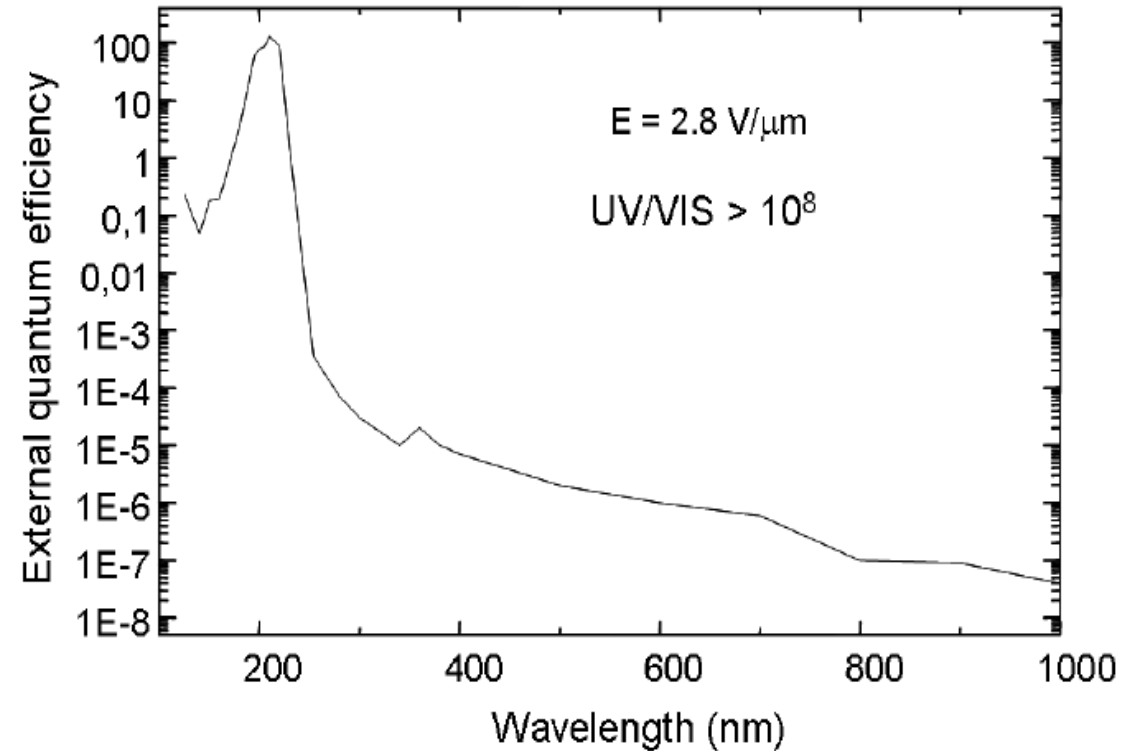


Fig.4. External quantum efficiency extended to visible and near infrared wavelength regions. The