Scintillator fundamantals and applications



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SPDAK 2021



Contents

Introduction

Organic Scintillator

Inorganic Scintillator

Radiation detection & medical imaging

High energy physics & Astrophysics

Astroparticle physics



Particle energy loss in matter



Detection Efficiency of X-ray and γ



1200 5 mall-Cs137 1000 600 400 200 200 400 600 800 1000 1200 E(keV)

CaMoO4

LYSO & TI2LaCl5:Ce



Photoelectric effect : ρZ_{eff3-4} Compton scattering : ρ Pair production : ρZ High $\rho \& Z_{eff}!$

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Principle of Radiation detection with Scintillator





Luminescence materials

- Inorganic crystal
- Organic crystal
- Inorganic/organic semiconductors
- ·
 - Organic polymers, organic Liquid
 - Glass, Ceramics



Nanoparticle, powders, QD

By LLAL group 경북대학교 물리 및 에너지학부

Scintillator

- Inorganic scintillators
 - a) Result of crystalline structure
 - b) Large band gap, insulators
 - c) High light output
 - d) Rather expensive, moderate size (~kg Ton)
- Organic scintillators
 - a) Molecular property of hidrocarbons
 - b) Moderate light output but fast (~ns)
 - c) Cheap, large size (~Ton- kTon)
 - d) Liquid scintillator (LSC)
 - Plastic scintillator (PSC)
 - Crystal (ex:anthracene, stilbene)
- Nobel liquid (gas) scintillator

Scintillator Requirement for Applications

General requirements for scintillator applications

- High γ detection efficiency : high Z, high density
 Good energy resolution : high light output (LY)
 High count rate : fast decay time
- □ Cost : low cost material, low melting temp.
- □ Handling : non-hygroscopic
- Background : internal background

None of scintillator has best performance of every aspect => Scintillator should be optimized for each application

PbWO4 : very low LY but high density and fast τ –>CMS, CALET CaMoO4, Li2MoO4 : low LY at RT, slow τ , no internal bg. -> AMoRE, CUPID LYSO:Ce : High LY, fast τ , high Z, internal background, expensive CsI:TI, NaI:TI : high LY, moderate τ , low background, cheap -> Belle, Fermi, KIMS, COSINE

Scintillator Application

- High energy physics BELLE, BES, CDF, L3, Phenix, CMS, LC, Focus ... Many more!
- Astro-particle physics Neutrino (LSND, Super-K, Kamland..), Underground(Darkmatter, neutrino, double beta), Ground array (HE neutrino, UHE cosmic ray, HE gamma)
- Nuclear Physics, Nuclear engineering.
 Neutron, Radioactive decay, heavy ion, radioactive beam.; Power reactor monitoring
- Astrophysics, Astronomy Balloon (ATIC, CREAM..), Satellite (Fermi, INTEGRAL..), Space station (AMS, CALET, ISS-CREAM..)
- Bio-science: Track radioisotopes in biology sample, Quantifying DNA and RNA
- Medical science: X-ray, PET, CT, SPECT, Track radioisotopes
- Environmental science: Monitoring of radioactivity, nuclear waste,
- More :Safety inspection, Military-> Homeland security

Scintillator supply





출처 : 서울대병원 핵의학과 이재성 교수경령태 학공 물리 및 에너지학부

Liquefied noble gases



Organic scintillator Fundamentals

Organic scintillator working principle



<u>Monocrystals</u>: naphtalene, anthracene, p-terphenyl.... <u>Liquid and plastic scintillators</u>



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Organic scintillator Solvents



 Standard Scintillator : PC + PPO(1.5-4g/l) +POPOP(10-50mg) : 65% of anthrecene, safe, Pulse shape discrimination of n/gamma

SAINT-GOBAIN & ELJEN tech

Physical Constants of SGCD Plastic Scintillators

Scintillator	Light Output % Anthracene ¹	Wavelength of Maximum Emission, nm	Decay Con- stant, Main Component, ns	Bulk Light Attenuation Length, cm	Refractive Index	H/C Ratio	Loading Element % by weight	Density	Softening Point *C
BC-400	65	423	2.4	250	1.58	1.103		1.032	70
BC-404	68	408	1.8	160	1.58	1.107		1.032	70
BC-408	64	425	2.1	380	1.58	1.104		1.032	70
BC-412	60	434	3.3	400	1.58	1.104		1.032	70
BC-414	68	392	1.8	100	1.58	1.110		1.032	70
BC-416	38	434	4.0	400	1.58	1.110		1.032	70
BC-418	67	391	1.4	100	1.58	1.100		1.032	70
BC-420	64	391	1.5	110	1.58	1.100		1.032	70
BC-422	55	370	1.6	8	1.58	1.102		1.032	70
BC-422Q	11	370	0.7	<8	1.58	1.102	Benzephenone,1%*	1.032	70
BC-428	36	480	12.5	150	1.58	1.103		1.032	70
BC-430	45	580	16.8	NA	1.58	1.108		1.032	70
BC-436	52	425	2.2	NA	1.61	0.960 D:C	Deuterium,13.8%	1.130	100

Physical Constants of SGCD Liquid Scintillators

Scintillator	Light Output % Anthracene*	Wavelength of Maximum Emission, nm	Decay Constant, ns	HC Ratio	Loading Element	Density	Flash Point °C
BC-501A	78	425	3.21	1.212		.874	26
BC-505	80	425	2.5	1.331		.877	48
BC-509	20	425	3.1	.0035	F	1.61	10
BC-517L	39	425	2	2.01		.86	102
BC-517H	52	425	2	1.89		.86	81
BC-517P	28	425	2.2	2.05		.85	115
BC-5175	66	425	2	1.70		.87	53
BC-519	60	425	4	1.73		.87	63
BC-521	60	425	4	1.31	Gd (to 1%)	.89	44
BC-523	65	425	3.7	1.74	Nat. 10B (5%)	.93	-8
BC-523A	65	425	3.7	1.67	Enr. 10B (5%)	.93	-8
BC-525	55	425	3.8	1.56	Gd (to 1%)	.88	81
BC-531	59	425	3.5	1.63		.87	93
BC-533	51	425	3	1.96		.80	65
BC-537	61	425	2.8	0.99 (D:C)	эН	.954	-11
BC-551	40	425	2.2	1.31	Pb (5% w/w)	.902	44
BC-553	34	425	3.8	1.47	Sn (10% w/w)	.951	42

ZEUS detector with Plastic +U sandwich calorimeter at HERA(ep)



Large LSC for neutrino physics

JUNO

Jiangmen Underground Neutrino Observatory Central detector (CD)

LS in acrylic vessel (35.4 m diam.)

- Requirements for JUNO LS
 - Lower background for physics: ²³⁸U<10⁻¹⁵g/g, ²³²Th<10⁻¹⁵g/g, ⁴⁰K<10⁻¹⁷g/g
 - High light yield: ~10 k ph./MeV concentration of flour need to be optimized
 - Long attenuation length: >20m@430nm
- Preliminary LS recipe (based on DYB experiment) 20 kt LS : 3g/l PPO +15 mg/l bis-MSB in LAB
 - PPO: 2,5-Diphenyloxazole
 - Bis-MSB: 1,4-di-(2-methylstyryl)benzene, p-bis(o-methylstyryl)benzene
 - LAB: linear alkyl benzene

Central detector (CD)

- Optical separation: Acrylic sphere
- Stainless Steel Latticed Shell
- 20 kton *Liquid Scintillator*
- <u>PMTs</u>: 17k 20" PMTs + 25k 3" PMTs
- Ultra-pure water buffer (2 m)

*KAMLAND kTon LSC *RENO LSC

Inorganic scintillator Fundamentals

General scheme of relaxation



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Scintillator Development Direction

Invited Article

The quest for high resolution γ -ray scintillators

Pieter Dorenbos



Fig. 2. History of scintillator discovery that distinguishes phases I to IV and phase V for future discoveries.



Optical Materials: X 1 (2019) 100021

SAINT-GOBAIN crystal scintillator



Saint-Gobain Crystals Physical Properties of Common Inorganic Scintillators

Scintillator	Light yield (photons/keV)	Light ouput(%) of Nal(Tl) bialkali pmt	Temperature coefficient of light output(%/C) 25°C to 50°C	1/e Decay time(ns)	Wavelength of max emission Im(nm)	Refractive index at Im	Thickness to stop 50% of 662 keV photons (cm)	Thermal expansion (/C)x10 ⁻⁶	Density g/cm³	Hygroscopic	Comments
LaBr ₃ (Ce+Sr)	73	190	0	25	385	~2.0	1.8	8	5.08	yes	Ultimate energy resolution (2.2% @ 662keV)
LaBr₃(Ce) BrilLanCe™ 380	63	165	0	16	380	~1.9	1.8	8	5.08	yes	General purpose, excellent energy resolution
CLLB Cs ₂ LiLaBr ₆ (Ce)	43	115		180 1080	420	~1.85	2.2		4.2	yes	Dual Gamma-Neutron detection, excellent
Nal(Tl)	38	100	-0.3	250	415	1.85	2.5	47.4	3.67	yes	General purpose, good energy resolution
Nal(Tl+Li)	35	100	-0.3	230. 1.1μs 240, 1.4μs	419	1.85	2.5	47.4	3.67	yes	Neutron-Gamma Scintillator
LaCl₃(Ce) BrilLanCe™ 350	49	70-90	0.7*	28	350	-1.9	2.3	11	3.85	yes	General purpose, good energy resolution
Csl(Na)	41	85	-0.05	630	420	1.84	2	54	4.51	yes	High Z, rugged
LYSO Lu _{1.8} Y _{.2} SiO ₅ (Ce)	33	87	-0.28	36	420	1.81	1.1		7.1	no	Bright, high Z, fast, dense, background from ¹⁷⁶ Lu activity
CdWO4	12-15	30-50	-0.1	14000	475	~2.3	1	10.2	7.9	no	Low afterglow, for use with photodioides
CaF2(Eu)	19	50	-0.33	940	435	1.47	2.9	19.5	3.18	no	Low Z, α & β detection
CsI(TI)	54	45	0.01	1000	550	1.79	2	54	4.51	slightly	High Z, rugged, good match to photodiodes
BGO	8 - 10	20	-1.2	300	480	2.15	1	7	7.13	no	High Z, compact detector, low afterglow
YAG(Ce)	8	15		70	550	1.82	2	~8	4.55	no	β-ray, X-ray counting, electron microscopy
Csl(Pure)	2	4-6	-0.3	16	315	1.95	2	54	4.51	slightly	High Z, fast emission
BaF2	1.8	3	0	0.6-0.8	220(195)	1.54	1.9	18.4	4.88	slightly	Fast component (subnanosecond)
	10	16	-1.1	630	310	1.50	1.9	18.4	4.88	slightly	Slow component
ZnS(Ag)	~50	130	-0.6	110	450	2.36			4.09	no	Coated on BC-400 or acrylic

The data presented are believed to be correct but are not guaranteed to be so.

Please see individual data sheets for full details at https://www.crystals.saint-gobain.com/products/crystal-scintillation-materials



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Crystal growing system with Czochalski method



At KNU









Bridgman Crystal Growing Methods



Crystal is growing in Bridgman furnace



At KNU









Crystals growing research at KNU over 20 years



 Nal:TI, CsI(TI, Co3, Na), BGO, BSO, BGSO, SrWO4, CaMoO4, SrMoO4 et al.
 New material : BaSrCl2, CsCe2Cl7, Cs(Rb)2Li(Na)CeCl6, Cs2LiGd(Lu)Cl(Br)6:Ce, Li6Lu(Gd,Y)(Bo3)3, NaGd(Wo4)3, LiBaF3, ZnMnTe, TI-based scintillators et al.

Discovered TI-based novel scintillators

The pioneer research work on the discovery and development of TI-based scintillators was started in 2009 by our research group and published TI₂LiGdCI₆:Ce as 1st paper in 2015.

Ternary Halides Elpasolites Tl₂LiGdCl₆:Ce [1] Tl₂LaCl₅:Ce [9-12] [9] H.J. Kim et al., J. Lumin., 186 (2017) 219-222. Tl₂LiGdBr₆:Ce [2] [10] A. Khan et al., J. Allovs Compd. 827 (2020) 154366. Tl₂LaBr₅:Ce [13] [11] Hawrami, R., et al., ., NIMA, 869 (2017): 107-109. $Tl_2LiYCl_6:Ce$ [3-6] [12] Shirwadkar, Urmila, et al. NIMA (2020): 163684. Tl₂GdCl₅:Ce [14, 15] [13] H.J. Kim et al., NIMA., 849 (2017) 72-75. $Tl_2LiLuCl_6:Ce$ [7] TlGd₂Cl₇:Ce [16] [14] Khan, A et al., J.Allovs Compd, 741 (2018) 878-882. [15] G. Rooh et al., IEEE Trans. Nucl. Sci., 65(8) (2018) 9 2157-2161. Tl₂LiScCl₆ [8] TlSr₂Br₅ [17] [16] Khan, A., IEEE Trans. Nucl. Sci., 65(8) (2018) 2152-2156. [17] G. Rooh et al., Opt. Mater 73 (2017) 523–526. TlSr₂I₅:Eu [18, 19] [18] H. J. Kim et al., Opt. Mater 82 (2018) 7–10. [19] Hawrami, Rastgo, et al., Optical Materials 100 (2020): 109624. **TlCaCl₂** [20] [20] A. Khan et al., Radiat, Meas. 107 (2017) 115-118. [21] P. Q. Vuong et al., Cryst. Eng. Comm 21(39) (2019) 5898–5904. [1] H.J. Kim et al., J. Lumin., 164 (2015) 86–89. Tl₂HfCl₆ [21-24] [22] Fujimoto, Yutaka, et al. Sens. Mater. 30.7 (2018): 1577-1583. [2] H. J. Kim et al., Rad. Measurem., 90 (2016) 279-281. [23] Hawrami, R., et al., J. Cryst. Growth, 531 (2020): 125316. [3] H. J. Kim et al., IEEE Trans. Nucl. Sci., 63 (2) (2016) 439. Tl₂ZrCl₆ [25,26] [24] Bhattacharya, Pijush, et al., ., IEEE Trans. Nucl. Sci. (2020). [4] G. Rooh et al., J. Cryst. Growth, 459 (2017) 163-166. [25] Q. V. Phan et al., J. Alloys Compd. 766 (2018) 326–330. **TIAIF**₁ [27, 28] [5] Hawrami, Rastgo, et al., Cryst. Growth Des.17 (2017): 3960-3964. [26] P. O. Vuong et al., Radiat. Meas. 123 (2019) 83-87. [6] Watts, Maya M., et al., IEEE Trans. Nucl. Sci., 67.3 (2020): 525-533 TIMgCl₃ [27] D. J. Daniel et al., IEEE Trans. Nucl. Sci 67(6) (2020) 898-903. [29] [7] G. Rooh et al., J. Lumin., 187 (2017) 347-351. [28] D. J. Daniel et al., J. Lumin. 223 (2020) 117197. [8]] M. J. Kim et al., J. Korean Phys. Soc. **TICdCl₃** [30] [29] Fujimoto, Yutaka, et al., Jpn. J. Appl. Phys. 55.9 (2016): 090301. [30] Fujimoto, Yutaka, et al., Radiat. Meas. 106 (2017): 151-154. TlCaI₃ [31] [31] Hawrami, R., et al., J. Cryst. Growth, 475 (2017): 216-219. **ORIGINAL PAPER**

Discovery, Crystal Growth, and Scintillation Properties of Novel TI-Based Scintillators

Detection efficiency comparison



- The y-rays full absorption peak detection efficiencies were calculated for 3 cm thick crystals using GEANT4 a Monte Carlo simulation package.
- These TI-based scintillators has higher detection efficiencies than commercial halide scintillators.
- Tl₂LaBr₅ and Tl₂LaCl₅ are more promising for ToF-PET application due to better detection efficiency, higher light yield and fast decay time (25 ns, 31 ns).
- TI₂HfCl₆ and TISr₂I₅ can be use potentially for y-rays spectroscopy.

Intrinsic luminescence

Bi, Pb, Tl, WO4, MoO4 BGO, PbWO4, CdWO4, CaWO4



Dopant Luminescence :NaI:Tl (CsI:Tl, LiI:Tl)



Ce3+ f-d transition



Eu2+ f-d transtion

SrCl2:Eu, Srl2:Eu, CaF2:Eu, Lil:Eu
 High light output but rather slow (~µs)



Scintillation light yield fundamentals



 $N_{ph}=S\times Q\times T\times N_{eh}=S\times Q\times T\times E/\beta E_g(N_{eh}=E/\beta E_g)$ The best case S, Q, T=1 and $\beta=2.5: N_{ph}=E/(2.5\times E_g)$

Scintillation Light yield



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Energy Resolution



Resolution of energy measurement determined by statistical variance of produced signal quanta.

$$\frac{\Delta E}{E} = \frac{1}{\sqrt{3000}} = 2\% \ r.m.s = 5\% \ FWHM$$

Energy resolution @ 662 keV





- YAIO₃:Ce, Lu₃Al₅O₁₂:Pr, LaCl₃:Ce, LaBr₃:Ce, SrI₂:Eu are reasonably close to fundamental limit.
- For LSO, NaI:Tl, CsI:Tl, R_{nonprop} dominant

By P. Dorenbos

Decay time of inorganic scintillator

Fast Scintillators

Slow Scintillators



By R.Zhu

Low energy applications Dose monitoring, Medical imaging

X-ray and gamma spectroscopy



Potable Dose Monitoring Development



Dose monitoring with flight





1.2 mSv/y at KNU0.6 mSv/y at Jakarta10 mSv /y at international Flight





Dental Computed Tomography (CT)





CsI:TI pixelated scintillator + CMOS sensors => Chest X-ray (DR)

CT (computed tomography)



Gamma Camera







B 10 16:

SPECT(single photon emission computed tomography)





By W. Moses

Inject Patient with Radioactive Drug



기업명	GE Healthcare	Philips Healthcare	Siemens Medical Solutions		
	Discovery PET/CT 710	Ingenuity TF	Biograph mCT		
모델명	C.	Ø	-		
DETECTOR Assembly					
- Number of detector rings	24	44	39, optional 52		
- Number of crystals	13,824	28,336	24,336, optional 32,448		
- Crystal material	LYSO	LYSO	LSO		
– Axial FOV, mm	157	180	162, optional 216		
CT Specifications					
 Number of slices acquired simultaneously 	64, 128	64,128	40, 64, 128		
X-ray generator					
- kW output	72	105	80, 100		
- mA range	5-800	20-500(64 CT) 20-665(128 CT)	20-800		
PET Image Acquisition					
- 2D	NO	NO	NO		
- 3D	YES	YES, 3-D RAMLA with LOR	YES, UltraHD 3-D		
- Time of flight	YES	YES	555 ps		
Gantry					
– H x W x D, cm	193 × 230 × 226	213 × 225 × 223,7	203 x 234 x 136		
– Weight, kg	6,215	4,201	3,977		
- Patient port diameter, cm	70	70(PET), 70(CT)	78		
Image Reconstruction					
- Coincidence window, nsec	4,9(3D)	3.8	4,1		
Purchase information					
- Price	\$3,722,884	Not specified	\$3,500,000-5,000,000		
- year first sold	2012	2011	2008		

자료 : ECRI Institue, Healthcare Product Comparison System

Positron Emission Tomography (PET)



Detects Pairs of Back-to-Back 511 keV Photons
 No Collimator Needed ⇒ High Efficiency



By W. Moses

RatCAP: Rat Conscious Animal PET





APD LEWINE HER (Hamamastu S8550) 4x8 array of LSO crystals (2x2x5 mm³) Actual RatCAP Ring

C.Woody, SCINT 07, 6/7/07

High energy applications High energy, Astro, Astroparticle

Example of Shower at High Energy





Fig. 3.6 'Elementary physical process' in a hadron shower.





Crystal scintillators



1.5 X₀ Cubic

Belle Csl(Tl)

L3 BGO CMS PWO(Y) Full Size Samples Belle CsI(TI): 16 X₀ L3 BGO: 22 X₀ CMS PWO: 25 X₀

경북대학교 물리 및 에너지학부

By R.Zhu

+



PbWO4 in CMS, LHC

VPT, gain

90 Ton of PbWO4

CMS Electromagnetic calorimeter



CsI:TI calorimeter for astrophysics



High energy cosmic-ray Exp.

ATIC (BGO, 22.4 Xo)



CREAM(W+plastic)





Timing Charge Detector

Cherenkov Camera

Silicon Charge Detector

> Carbon Targets

Calorimeter

Support Instrument Package

경북대학교 물리 및 에너지학부



Fig. 2. Exploded view of the ISS-CREAM Instrument.

ISS-CREAM(W+plastic)



Tungsten / Scintillating Fiber Stack



Fiber Light Guides



Tungsten Plates

High energy cosmic-ray Exp.



Direct WIMP detection technique



Dark matter Search (KIMS)

Elastic Scattering of WIMP off a nucleus in the detector

WIMP





Energy loss by ionization(scintillation) and lattice vibration

COSINE @Y2L, KOREA

New Shielding Structure



LETTER Nature

https://doi.org/10.1038/s41586-018-0739-1

An experiment to search for dark-matter interactions using sodium iodide detectors

The COSINE-100 Collaboration*



Double beta decay detection technique



AMORE-II: Mo crystals grown and tested



KNU Advanced Positronium Annihilation Experiment (KAPAE)

The Final Goal

- Positronium: C-violation & QED test & rare decay
- Invisible decay
 - \rightarrow Experimentally interesting branching ratio of the order of 10⁻⁸
 - → Extra-demensions
 - → Milli-Charged particles
 - \rightarrow Darkmatter of a mirror particle type
 - Axion
 - → Dark photon

Search for C-violation

- C-violation
 o-Ps -> 4 γ search
 - o-Ps -> 2 γ search
- Approximate calculation $10^8 \rightarrow \sim 10^{-7} (10^{-6}) : 10 \text{ times improvement}$



orthopositronium confined in a

PHYS. REV. D 97, 092008 (2018)

vacuum cavity

Searching for light dark matter through Positronium decay

Eur. Phys. J. D (2018) 72: 44

High order QED process Rare decay



Positronium decay experiment

Full Design of Detector

 <u>The trigger part is surrounded</u> by the gamma detection part with an array of <u>14 x 14 BGO</u> <u>scintillators</u> (7.5 x 7.5 x 150 mm³)



 For <u>dual readout</u> both sides of the BGO scintillators are coupled with 7 x 7 arrangement of 2 x 2 arrays for a total of 14 x 14 SiPMs



14 x 14 channels



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Calorimeter, PET and Ps decay

CalorimeterPET CameraPs decayImage: Strain of the strain of th

- Cylindrical Gamma Ray Detectors
- High Efficiency, Hermetic
- Segmented, High Density Scintillator Crystals

Thank you for your attention

Emission Spectra of Scintillators



Emission spectra of Nal(TI), BGO and CdWO₄, scaled on maximum emission intensity.



 Recent development on high light output oxide crystal
 : Gd₃Al₂Ga₃O₁₂ :Ce (GAGG:Ce by Yoshikawa group) : (46,000 ph/MeV (Yoshikawa), ~76,000 ph/MeV(TPS)
 => Column structure by Vapor deposition?



Journal of Ceramic Processing Research. Vol. 16, No. 00, pp. 1~5 (2015)

Ceramic Processing Research

NALOF

UR

Scintillation properties of the Gd₃Al₂Ga₃O₁₂ : Ce crystal

Hye-Lim Kim^a, Hong-Joo Kim^{a,*}, Eun-Jung Jang^a, Won-Guen Lee^b, Moon-Kwang Ki^b, Heun-Duk Kim^b, Gu-Sik Jun^b and Vladimir Kochurikhin^c

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Radiation hardness of Crystal Calorimeter







Crystal	Dimensions (mm ³)	ID	Emission Peak (nm)	Fluence (p/cm ²)	RIAC at EP (1/m)	@ 3E+14
BGO	25×25×200	SIC-BGO	480	1.77E+14	14.7	24.9
CeF_3	22 ² ×26 ² ×150	SIC-CeF	340	1.40E+14	17.4	37.3
LYSO	25×25×200	SG-LYSO	430	3.27E+14	0.86	0.8
LFS	25×25×180	OET-LFS	430	3.55E+14	3.7	3.1
PWO*	28.5 ² ×30 ² ×220	SIC-PWO	420	1.80E+14	> 36	> 60

LYSO is the most radiation hard among all tested at LANL

LYSO radiation hardness by by proton beam









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Proton test with Gd3Al2Ga3O12:Ce





Decay time before and after proton irradiation No significant change is observed (87, 263 nm)

Light yield change after 1000 Gy proton (12% lower)