THE GROWTH AND CHARACTERIZATION OF LANTHANIDE-PHOSPHATE-BASED SCINTILLATORS FOR RADIATION DETECTION APPLICATIONS

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γ- RAY SCINTILLATORS

A. 3 MAJOR PROCESSES

1. γ – ray-to-Host
2. Host-to-Activator
3. Activator-to-Light

B. MANY REQUIREMENTS!

1. High Light Output (Photons/Mev)
   NaI: Thallium → 38,000
   BGO → 8,200
2. Short Decay Time (Nano Sec)
3. Wavelength Match to Detector
4. High Density (6 g/cm^3)
5. Chemical Stability
6. Radiation Hardness
7. Cost
8. Crystal Growth

NaI: Thallium→ 1948, Hufstader

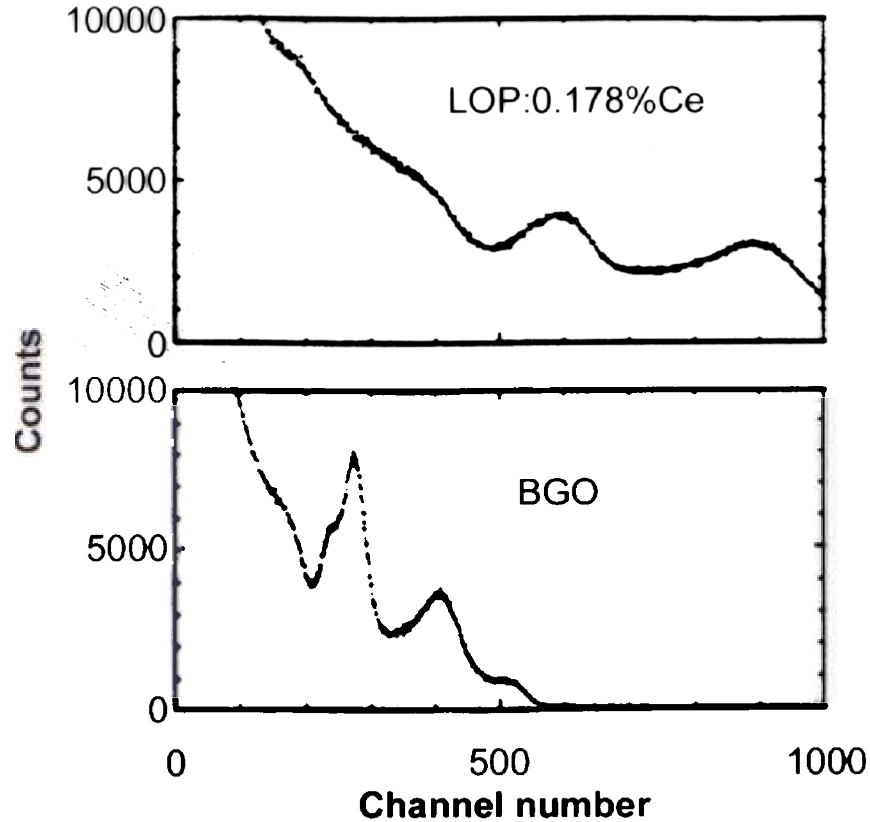
BGO → 1973, Weber & Monchamp

↓

$Bi_4Ge_3O_{12}$

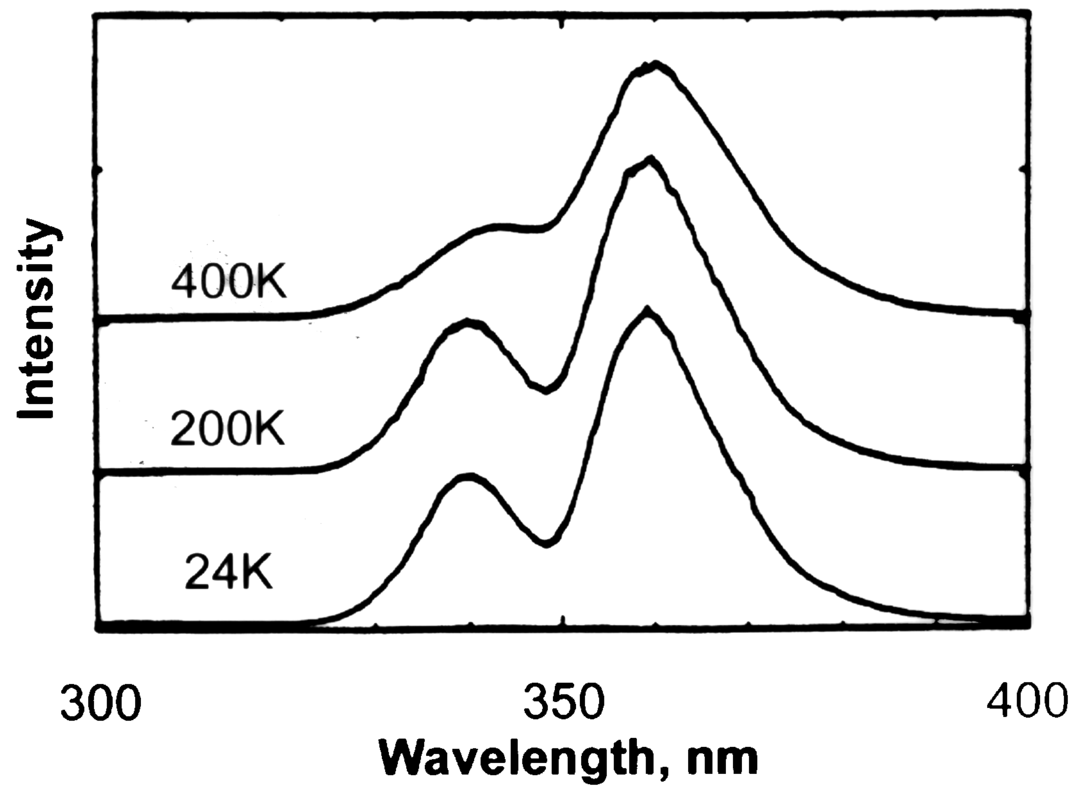
NEW: Lu_2SiO_5
MELCHER & SCHWEITZER

Energy Spectra Under ^{207}Bi Excitation. Amplifier Shaping Time 0.5 μs.



A. Lempicki, E. Berman, A. J. Wojtowicz, M. Balcerzyk, and L. A. Boatner, "Cerium-Doped Orthophosphates: New Promising Scintillators," IEEE Trans, Nucl, Sci. **40**, (4) 384—387 (1993).

Luminescence Spectra of LOP:0.898%Ce Optical Excitation at 250 nm



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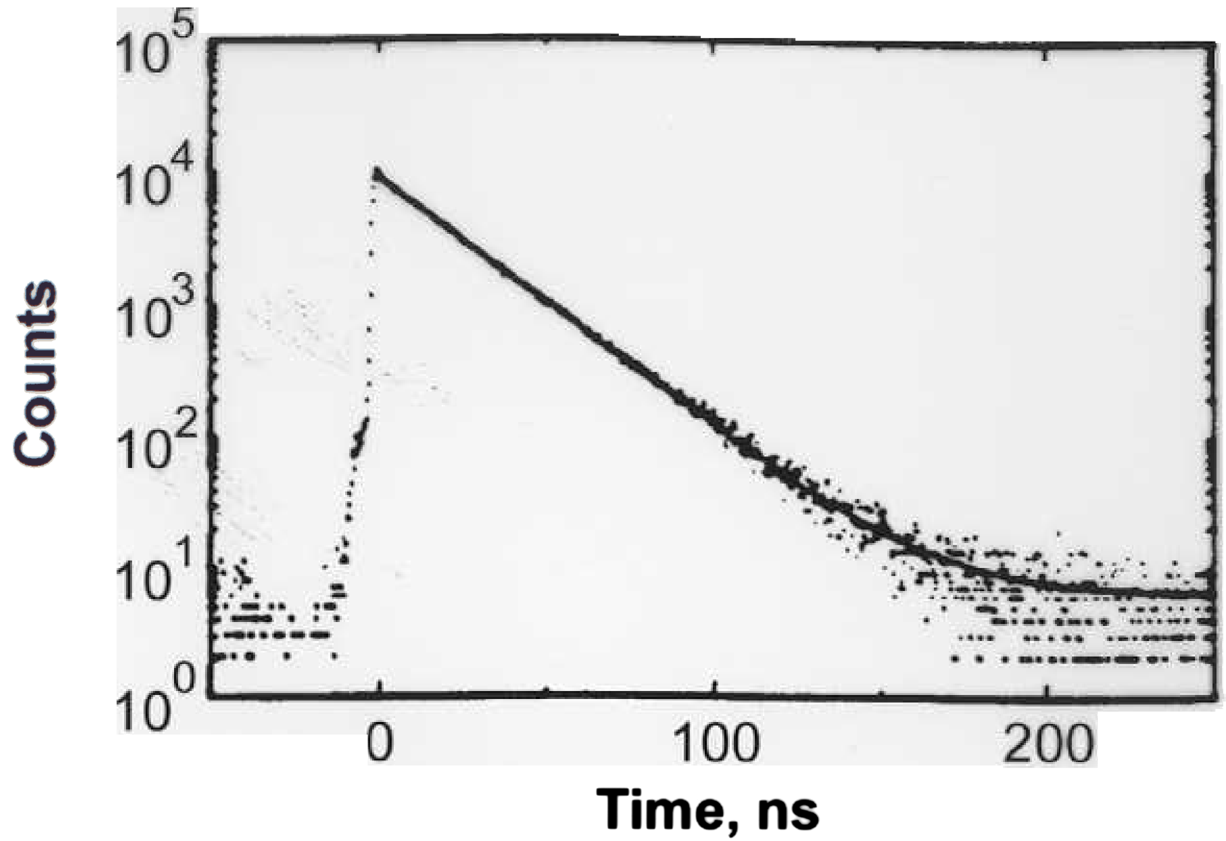
Scintillation Decay of LOP:0.178%Ce



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Comparison of Ce Doped Lutetium Compounds with BGO

	BGO	LOP	LSO
Relative light output	100	217	500
Emission wavelength (nm)	480	360	420
Decay constant (nsec)	300	24	40
Density (g/cm^3)	7.13	6.53	7.4
Attenuation length (cm)	1.10	1.43	1.22
Index of refraction	2.15	1.7 (ref.2)	1.82

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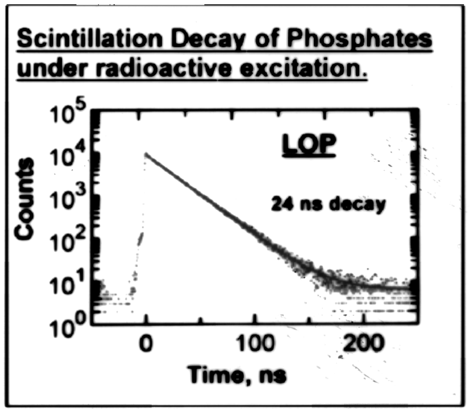
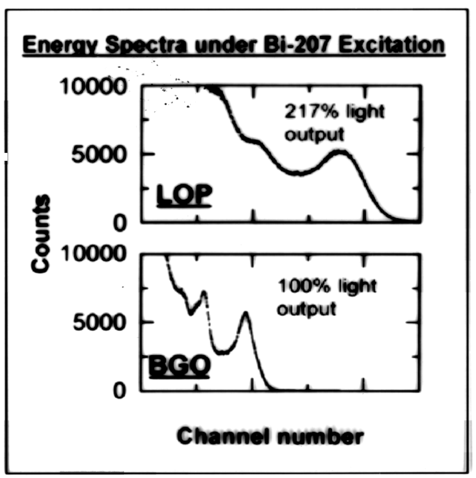
Ce-Doped Lutetium Orthophosphate Scintillators

(LOP)



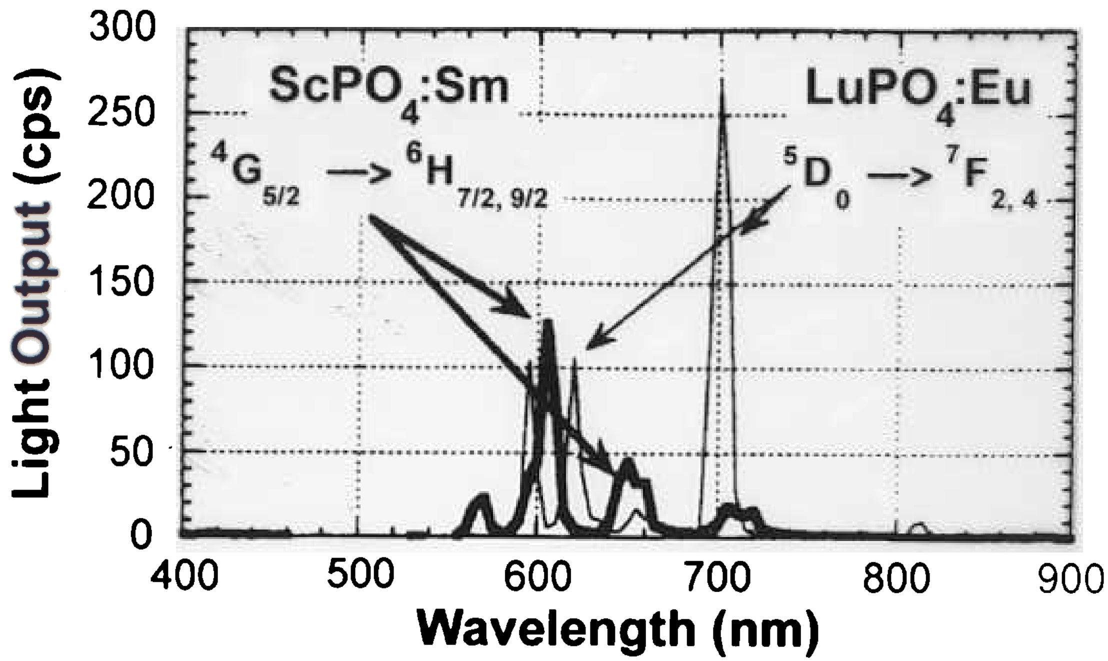
FAST:
24 ns decay



EFFICIENT:
217 % relative to BGO

New $LuPO_4$: Ce gamma-ray-scintillator crystal-growth system incorporating variable frequency acoustic crucible excitation as a means of overcoming solvent viscosity problems.
ORNL Solid State Division

Emission Spectra (Corrected for Wavelength Dependence of Monochromator Efficiency, but not for PMT Dependence) for Representative Europium and Samarium Doped Samples.



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Long Wavelength Luminosities. The Measured Luminous Intensity (in Photons per MeV) of the 600—900 nm Emissions.

Photon / MeV	Compound	Phot. /MeV	Compound
123171 *	$LuPO_4$:20% Eu	2571 *	YPO_4:5% Nd
103244 *	$LuPO_4$:30% Eu	2497	Eu_2O_3
76099 *	$LuPO_4$:~5% Eu	2468 *	$LuPO_4$:0.7% Fe
61072 *	$ScPO_4$:~1% Eu	2427	EuF_3
60316 *	$LuPO_4$:5%Eu, 1%Gd	2418 *	$BaLuYF_8$:0.2% Pr
44302 *	$ScPO_4$:10% Sm	2401 *	$Na_{0.4}Lu_{0.6}F_2$: 1% Nd
35728	YVO_4:Eu	2296	$Bi_2Al_4O_9$: 0.5% Ce
31543 *	$ScPO_4$:3% Sm	2271 *	$ScPO_4$:2% Dy
31294 *	YPO_4:2% Sm	2232	$CaMoO_4$
22095 *	YPO_4:?% Eu	2201	LaF_3:1% Pr, 1% Ba
15728 *	$LuPO_4$:2% Sm	2157	Y_2O_3:2% Tb
14589 *	$ScPO_4$:~5% Eu	2089 *	$ScPO_4$:1.7% Ni
14288 *	$LuPO_4$:10 % Sm	1806 *	BaF_2:8.4% Ho
13067	$GdTaO_4$:Tb	1799	LaF_3:0.5% Pr
13053 *	$ScPO_4$:2.9% Dy	1743 *	$PbHPO_4$:5.8% Tb
12101 *	YPO_4:1% Dy	1651 *	$ScPO_4$:0.7% Fe
10914 *	$ScPO_4$:0.7% Er	1609 *	$TbPO_4$:25% Gd
10304	BaF_2:10% Eu	1526	Y_2O_3:2% Tb, 2% Eu
9316 *	$LuPO_4$:10% Tb	1514	TbF_3
9280 *	$ScPO_4$:10% Pr	1514	$Lu_3Al_5O_{12}$:Ce
9242 *	YPO_4:2% Tm	1506 *	$LaPO_4$:?% Eu

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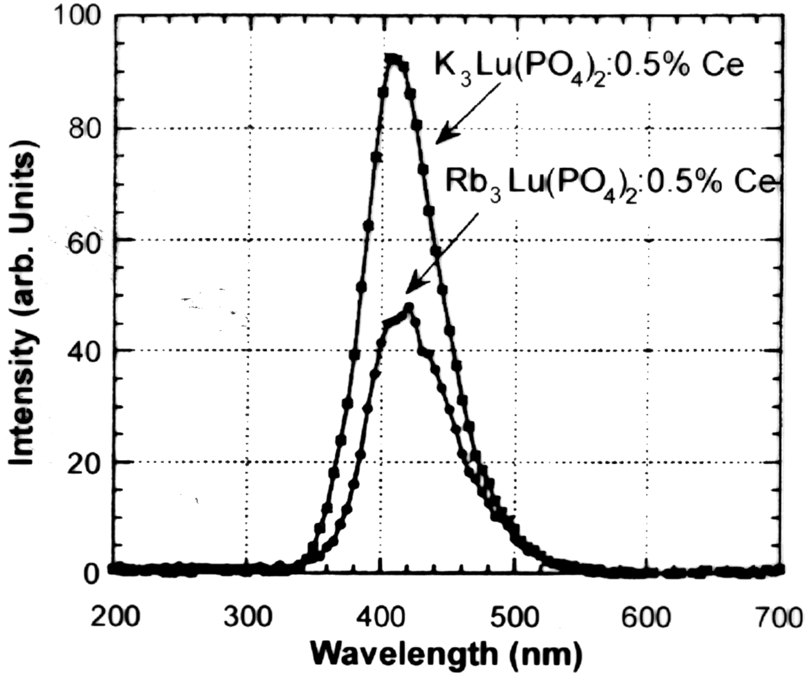
Emission Spectra of Double Phosphate Compounds, Corrected for Wavelength Dependence of Monochromator (but not PMT) Efficiency.



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The Discovery and Development of a New Class of Long Wavelength Scintillators for Gamma- and X-ray Detection

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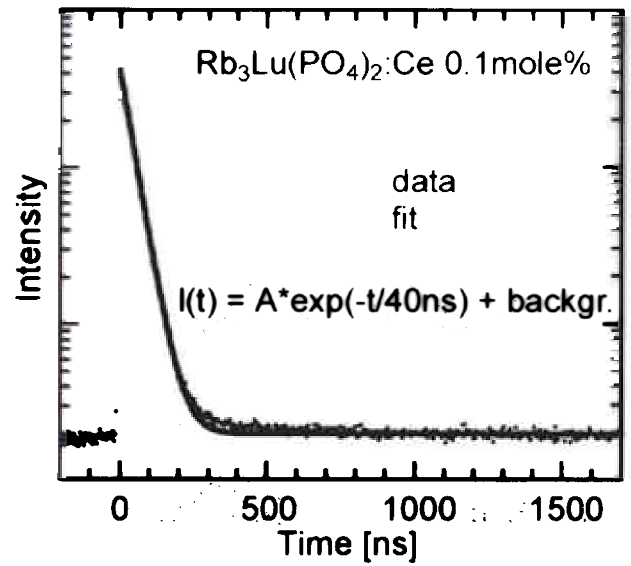
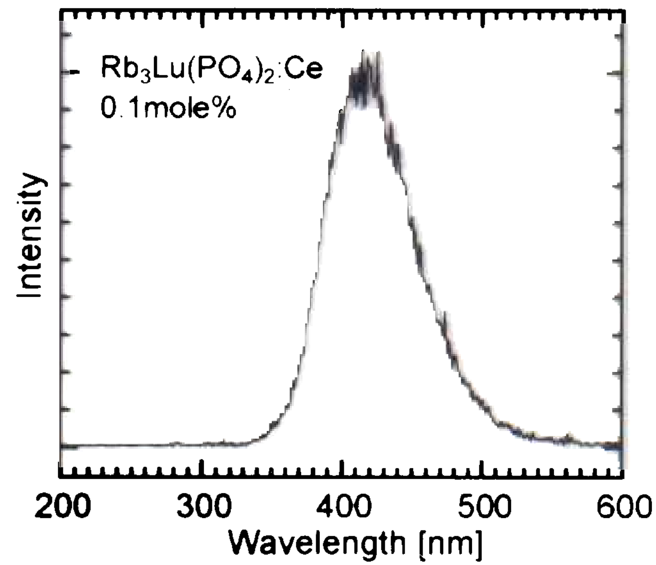
- Scintillation and properties of $Rb_3Lu(PO_4)_2$:Ce and $Cs_3Lu(PO_4)_2$:Ce

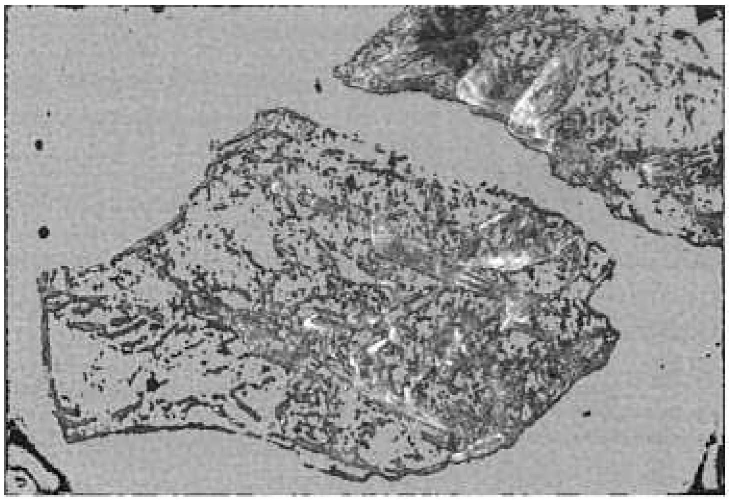


Room temperature scintillation time profile of $Rb_3Lu(PO_4)_2$: 0.1 mole % Ce excited by 662 keV gamma photons. The decay of the Ce^{3+} emission is practically single exponential and quite fast – decay time constant value is about 40 ns.

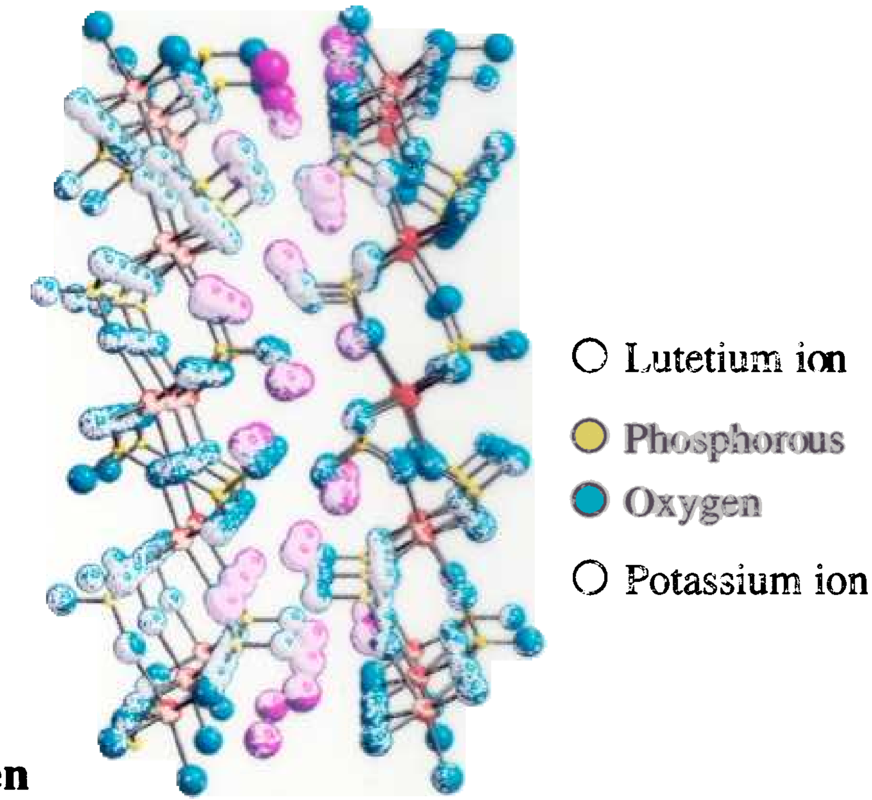


Room temperature X-ray excited luminescence spectrum of $Rb_3Lu(PO_4)_2$:0.1 mole % Ce. Scintillation properties measurements compare very favorably to those of the best commercial scintillators. Light yield at a level of about 250 % of BGO LY. The maximum intensity at about 410 nm, is suitable for use with light detectors such as photomultipliers with alkali halide photocathodes or avalanche photodiodes.

Layered structure of $K_3Lu(PO_4)_2$ seen in single crystals and crystal structure



Clear single crystal of $K_3Lu(PO_4)_2$



Potassium ions are located between the polymeric layers

Perspective view down the c axis

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Properties of Double Phosphate Compounds. The Measured Luminous Intensity (in Photons per MeV) and Decays Times of $K_3Lu(PO_4)_2$ and $Rb_3Lu(PO_4)_2$ as a Function of Cerium Concentration

Compound	Cerium Fraction			
	Undoped	0.1%	0.5%	1.0%
$K_3Lu(PO_4)_2$	14,500 phot/MeV 75% @ 44 ns 25% @ 1350 ns	46,400 phot/MeV 86% @ 39 ns 14% @ 1250 ns	52,500 phot/MeV 86%% @ 37 ns 14% @ 1080 ns	19,400 phot/MeV 74% @ 40 ns 26% @ 1060ns
$Rb_3Lu(PO_4)_2$	7,400 phot/MeV 95% @ 44 ns 5% @ 1290 ns	24,600 phot/MeV 89% @ 40 ns 11% @ 1190 ns	27,100 phot/MeV 91% @ 36 ns 9% @ 920 ns	28,200 phot/MeV 84% @ 34 ns 16% @ 990 ns

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Lanthanide-Phosphate-Based Scintillators

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New classes of scintillators with longer wavelength outputs that:

A. Match Si solid state detectors
B. Can be used in applications requiring low-loss transmission through optical fibers

Two classes of new scintillators discovered

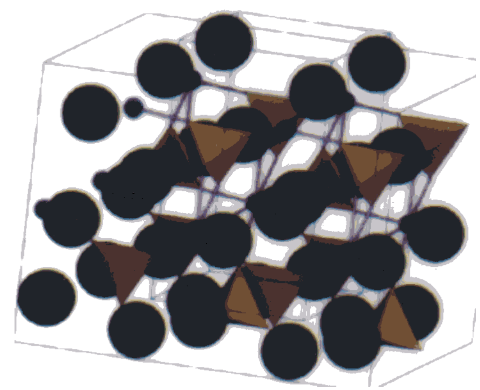
$Lu(PO_4):Eu$	123,171 photons/MeV
$ScPO_4:Sm$	44,302 photons/MeV

Ce-Doped double phosphate compounds of the type

$K_3Lu(PO_4)_2$	52,500 photons/MeV
$Rb_3Lu(PO_4)_2$	28,200 photons/MeV

Techniques for the growth of double phosphate single crystals developed

Unknown crystal structures of the double phosphates determined by single-crystal x-ray and neutron diffraction structural refinements:



$K_3Lu(PO_4)_2$ — ● K — • Lu

Discovered through ORNL collaborations involving LBL and Boston U.

New VUV Scintillators
Summary of Properties

$LuPO_4$ Nd YPO_4 Nd

- Luminescence
  - fast Nd^{3+} d f emission 192 200nm strong), 240 and 280 weaker
- LY measurements
  - Cs^{1} γ source used (662 keV
  - Photomultiplier: Philips XP2020Q
  - Integration
    - LeCroy multichannel analyser
    - 200 ns gate
    - Integration period starting about 5 ns before scintillation
  - Experimental geometry
    - Dow Corning 200 fluid used for optical coupling

Lanthanide-Phosphate-Based Scintillators
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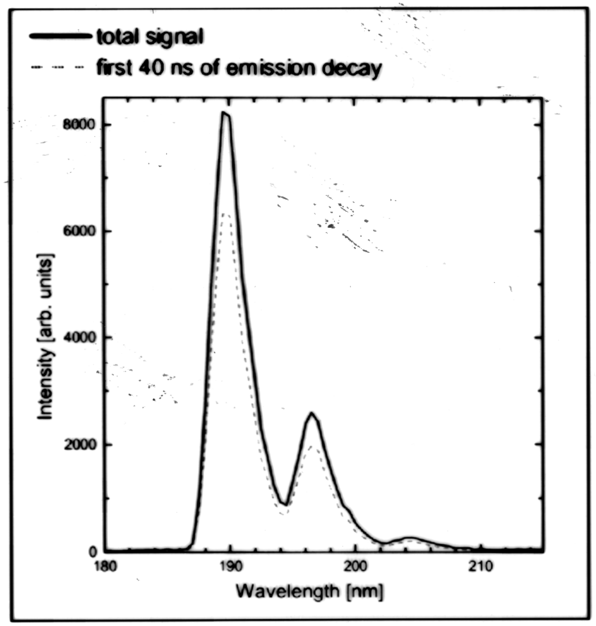
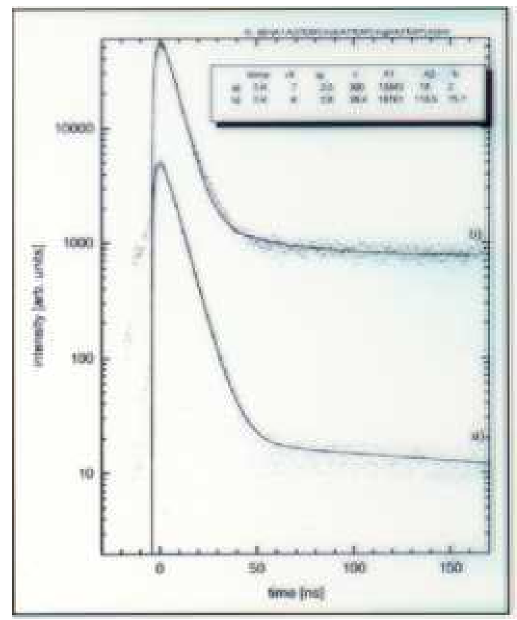
Nd-Doped $LuPO_4$

New Scintillators With Short Wavelength Outputs Appropriate for Use With Proportional Counters

Currently ongoing characterization studies of short wavelength scintillators



Luminescence Spectrum of $Lu(PO_4)$:Nd, λ_{exc} = 110 nm
Synchrotron Exp. DESY, Hamburg, 07,1999



$Lu(PO_4)$:Nd,
Pulse shapes of 190 nm emission under:
a) 80 nm Excitation, b) 110 nm excitation
Synchrotron experiments DESY, Hamburg 07, 1999

A New Class of Very Fast Short-Wavelength Scintillators for Use With Proportional Counters

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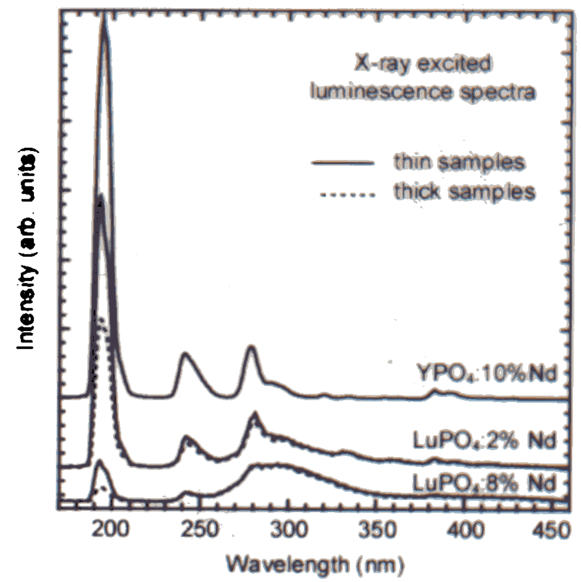
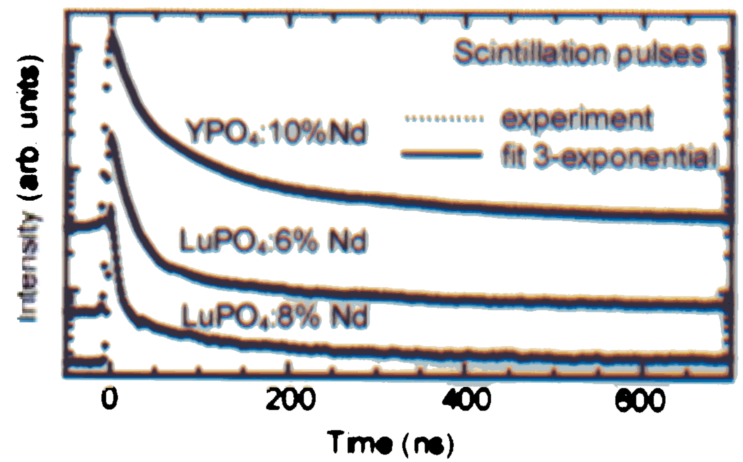
VUV Scintillation of $LuPO_4$:Nd and YPO_4:Nd



X-ray-excited luminescence spectra of YPO_4:Nd and $LuPO_4$:Nd crystals measured in "transmission" geometry. The emission spectrum of YPO_4:10%Nd is dominated by a relatively narrow band with a maximum at about 192 nm. This band can be associated with a transition from the lowest level of the $4f^25d$ configuration of excited Nd^{3+} ions down to the $^4I_{9/2,\ 11/2,\ 13/2}$ levels of the $4f^3$ configuration.



Selected scintillation pulse shapes of YPO_4:Nd and $LuPO_4$:Nd. Measurements and three exponential fits were done for a 2500 ns time range.

Scintillation Pulses Parameters

Component →	short		medium		long	
Sample ↓	τ_1 [ns]	LY_1 [%]	τ_2 [ns]	LY_2 [%]	τ_3 [ns]	LY_3 [%]
YPO_4:10%Nd	12.5	20	60	40	385	40
YPO_4:0.5%Nd	11.6	48	56	39	649	13
YPO_4:1%Nd	13.3	42	52	40	210	18
$LuPO_4$:2%Nd	10.1	29	129	21	1684	50
$LuPO_4$:8%Nd	9.0	37	109	23	1124	40

Pet "PET" Machine

- **PET machine using VUV scintillators and proportional counters with TMAE**
- **$LuPO_4$:Nd (1%) YPO_4:Nd (1%)**
- **190 mm Peak, $LuPO_4$ – 13.5 mm ext. for 511 KeV Photons**

The figure to the left shows the finished scanner, and the picture to the right shows the detector during assembly.

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IMPROVING THE PERFORMANCE OF EXISTING RADIATION DETECTOR MATERIALS

New crystal-growth methods to solve the CZT materials quality problem:

Sonic and ultrasonic methods applied to achieve
- Compositional homogeneity
- Thermal homogeneity

- **Achieve new insight into the crystal growth parameters affecting the quality of** CZT crystals for γ-ray detector applications **–**

Determine the inter-relationships between crystal-growth conditions and important materials properties through the feedback of characterization information to the crystal-growth process.