Advanced Materials Research Vols 418-420 (2012) pp 922-927 Online: 2011-12-06
© (2012) Trans Tech Publications, Switzerland
doi:10.4028/www.scientific.net/AMR.418-420.922

Comparison of $Lu_3Al_5O_{12}$:Ce and $LaBr_3$:Ce Scintillators in Gamma-Ray Spectrometry

Weerapong Chewpraditkul[1,a], Chalerm Wanarak [1,b],
Marek Moszynski[2,c] and Lukasz Swiderski [2,e]

[1] Department of Physics, King Mongkut's University of Technology Thonburi, Bangkok 10140,Thailand

[2] The Soltan Institute for Nuclear Studies, Otwock-Swierk, PL-05-400 Poland

[a] weerapong.che@kmutt.ac.th, [b] 52500406@st.kmutt.ac.th,

[c] marek@ipj.gov.pl, [d] l.swiderski@ ipj.gov.pl

Keywords: Energy Resolution, γ-ray detection, $LaBr_3$:Ce, LuAG:Ce, Non-proportionality, Photofraction

Abstract. The performances of Ce-doped $Lu_3Al_5O_{12}$ (LuAG:Ce) and $LaBr_3$ ($LaBr_3$:Ce) scintillators were compared for γ-ray detection using photomultiplier tube (XP5500B PMT) readout. For 662 keV γ-rays (^{137}Cs source), an energy resolution of 3.5% obtained for $LaBr_3$:Ce is much better than that of 6.7% obtained for LuAG:Ce, while the estimated photofraction of 28.0% for LuAG:Ce is higher than that of 16.2% for $LaBr_3$:Ce. The light yield non-proportionality and energy resolution versus γ-ray energy were measured and the intrinsic resolution of the crystals was calculated. The coincidence timing resolution, obtained in this work for 511 keV annihilation quanta, was 583 ps and 204 ps, respectively, for LuAG:Ce –BaF_2 and $LaBr_3$:Ce – BaF_2 detectors.

Introduction

Research and development of new scintillator materials is mainly triggered by the rapidly growing needs of medical imaging and high energy physics. During the last two decades, new types of scintillators, in particular, Ce-doped inorganic scintillators were intensively studied and some of them were successfully developed for commercial production, for recent reviews see [1- 4].

$Y_3Al_5O_{12}$:Ce (YAG:Ce) single crystal was reported in the literature as a fast oxide scintillator [5,6]. Isostructural $Lu_3Al_5O_{12}$:Ce (LuAG:Ce) has a higher density (6.67 g/cm^3) than YAG:Ce (4.56 g/cm^3), which is advantageous in the case of high energy gamma-ray detection [7,8]. Its emission spectrum at room temperature (RT) is peaked around 525 nm. The scintillation light yield within 1 μs time gate is about 12,500 ph/MeV and 22,000 ph/MeV, respectively, for LuAG:Ce and YAG:Ce crystals [9].

New Ce-doped $LaCl_3$ [10] and $LaBr_3$ [11] scintillators appeared with attractive properties due to high light output and very good energy resolution. $LaBr_3$:Ce has a density of 5.29 g/cm^3 and an emission spectrum at RT is peaked around 370 nm. $LaBr_3$:Ce exhibits a very high light yield above 60,000 ph/MeV and an excellent energy resolution of about 3% for 662 keV γ-rays. The excellent energy resolution of $LaBr_3$:Ce crystal is confirmed by good proportionality characteristics and corresponding excellent intrinsic resolution [12].

In this paper, we present the comparative study on energy resolution and timing properties of LuAG:Ce and $LaBr_3$:Ce crystals. The light yield non-proportionality and energy resolution versus γ-ray energy were measured and the intrinsic resolution of the crystals was determined. The estimated photofraction in the pulse height spectra of 320, 662 and 835 keV γ-rays was determined for both crystals and compared with the ratio of the cross-sections for the photoelectric effect to the total one calculated using the WinXCom program.

Methodology

The LuAG:Ce crystal with size of 10×10×5 mm^3 was supplied by Crytur Ltd (Czech Republic). The $LaBr_3$:Ce crystal encapsulated in an aluminum can with size of ∅13×13 mm^2 was supplied by Saint-Gobain (France).

Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonis XP5500B PMT using silicone grease. The signal from the PMT anode was passed to an ORTEC 113 preamplifier and was sent to a Tennelec TC244 spectroscopy amplifier. The measurements were carried out with 3 μs shaping time constant in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8k [13] was used to record energy spectra.

The number of photoelectrons was evaluated by Bertolaccini method [14,15], which is based on a comparison of the position of a full energy peak of γ-rays detected in the crystals with that of the single photoelectron peak from the PMT photocathode. The measurements of light yield non-proportionality and energy resolution were carried out for a series of γ-rays emitted by different radioactive sources (^{93}Mo, ^{145}Pm, ^{241}Am, ^{133}Ba, ^{109}Cd, ^{57}Co, ^{51}Cr, ^{137}Cs, ^{54}Mn and ^{22}Na) in the energy range from 16.6 keV to 1274.5 keV.

Coincidence timing measurements were performed using 511 keV annihilation quanta from a ^{22}Na source. Each crystal was coupled to a Photonis XP20D0 PMT. A BaF_2 crystal coupled to a Photonis XP20Y0Q/DA PMT was used as the reference detector. Its time resolution of 128 ps for 511 keV full energy peak selection in side channel was reported [16]. Upon irradiation of the crystals coupled to each PMT with 511 keV annihilation quanta, a signal from each PMT was processed with and ORTEC 935 CFD. Time spectrum was measured with an ORTEC 566 TAC and recorded by the PC-based MCA.

Results and discussion

Light Yield and Energy Resolution. Fig. 1 presents a comparison of the pulse height spectra of 662 keV γ-rays from a ^{137}Cs source measured with LuAG:Ce and $LaBr_3$:Ce crystals. The energy resolution of 3.5% obtained with $LaBr_3$:Ce is superior compared to the value of 6.7% obtained with LuAG:Ce. Note a higher photofraction in the spectrum measured with LuAG:Ce, as would be expected due to a higher effective atomic number and density of the LuAG:Ce material.

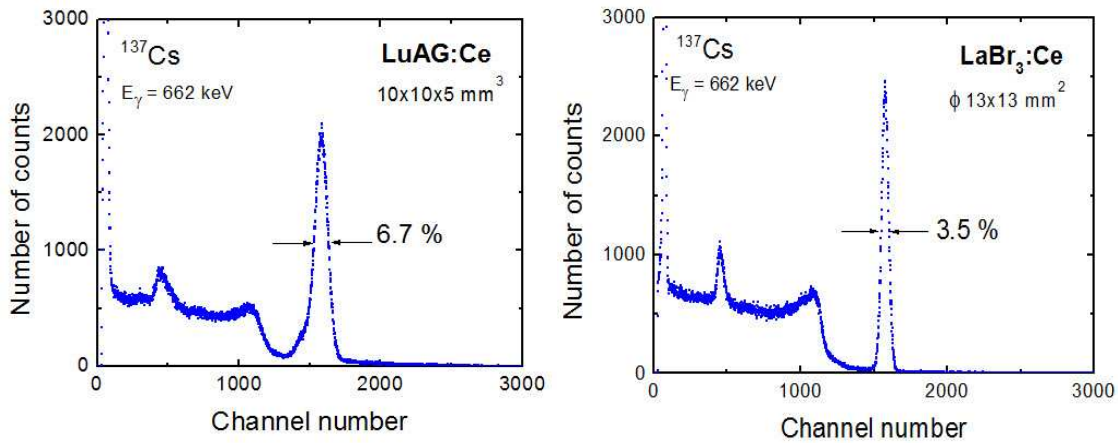

Fig.1 Pulse height spectra of 662 keV γ - rays from a ^{137}Cs source measured with LuAG:Ce and $LaBr_3$:Ce crystals.

Photoelectron yield was determined using 662 keV γ-rays from a ^{137}Cs source. $LaBr_3$:Ce exhibits the photoelectron yield of about 12,320 phe/MeV, which is much larger than the value of about 3,730 phe/MeV for LuAG:Ce. The number of photoelectrons measured for both crystals was recalculated to the number of photons assuming the quantum efficiency of 29% and 18% for the XP5500B PMT, respectively, at the peak emission of $LaBr_3$:Ce (370 nm) and LuAG:Ce (525 nm). The light yield of about 42,500 ph/MeV and 20,700 ph/MeV was obtained for $LaBr_3$:Ce and LuAG:Ce, respectively.

The energy resolution (ΔE/E) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [17]

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2, \quad (1)$$

where δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution and δ_{st} is the statistical contribution of PMT to the resolution.

The statistical uncertainty of the signal from the PMT can be described as

$$\delta_{st} = 2.355 \times 1/N^{1/2} \times (1+\varepsilon)^{1/2}, \quad (2)$$

where N is the number of the photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for an XP5500B PMT.

The transfer component depends on the quality of optical coupling of the crystal and PMT, homogeneity of quantum efficiency of the photocathode and efficiency of photoelectron collection at the first dynode. The transfer component is negligible compared to the other components of the energy resolution, particularly in the dedicated experiments [17].

Overall energy resolution and PMT resolution can be determined experimentally. If δ_p is negligible, intrinsic resolution δ_{sc} of a crystal can be written as follows

$$(\delta_{sc})^2 = (\Delta E/E)^2 - (\delta_{st})^2. \quad (3)$$

The results summarizing the photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals are presented in Table 1.

Table 1 Photoelectron yield, light yield, and energy resolution for LuAG:Ce and $LaBr_3$:Ce.

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	Energy resolution [%]		
			ΔE/E	δ_{st}	δ_{sc}
LuAG:Ce	3,730 ± 200	20,700 ± 2,000	6.7	5.0	4.5
$LaBr_3$:Ce	12,320 ± 600	42,500 ± 4,000	3.5	2.7	2.2

Fig. 2 presents a comparison of the intrinsic resolution for the studied crystals. The intrinsic resolution of $LaBr_3$:Ce is almost a factor of two lower than that of LuAG:Ce. This result should be reflected in better proportionality of light yield vs γ-ray energy for $LaBr_3$:Ce, as the non-proportionality of light yield is a fundamental limitation to δ_{sc} of the scintillators [17,18].

Non-Proportionality of Light Yield. The non-proportionality is defined as the ratio of photoelectron yield measured at specific γ-ray energies relative to the photoelectron yield at the 662 keV γ-peak. Fig.3 presents the non-proportionality curves of $LaBr_3$:Ce and LuAG:Ce crystals. $LaBr_3$:Ce is clearly superior to LuAG:Ce in terms of light yield proportionality. It is proportional down to 80 keV and exhibits only 7% deviation at 16.6 keV, while LuAG:Ce shows larger non-proportionality (22% deviation at 16.6 keV). The high proportionality of $LaBr_3$:Ce is one of the important reasons behind its high energy resolution. The high proportionality characteristics of $LaBr_3$:Ce is reflected in its low intrinsic resolution(see Fig.2).

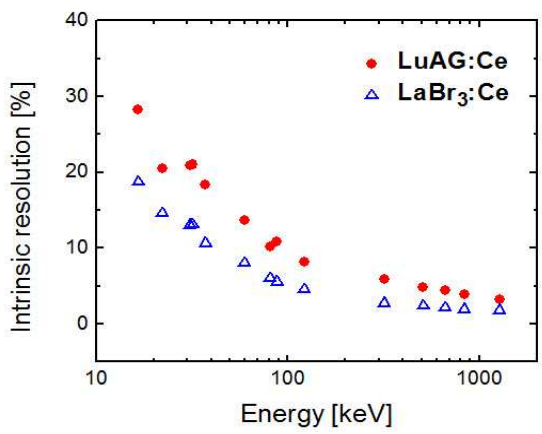

Fig. 2 Intrinsic resolution of LuAG:Ce and $LaBr_3$:Ce crystals versus energy of γ-rays.

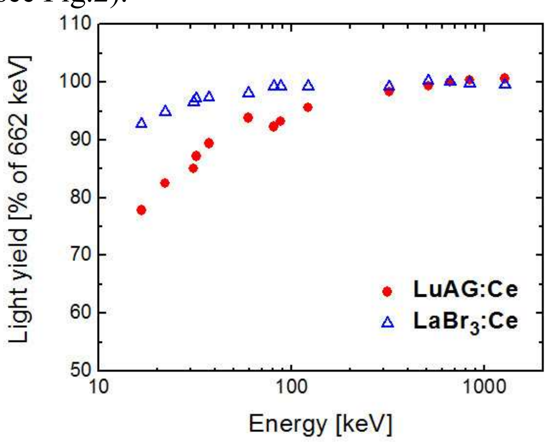

Fig. 3 Non-proportionality of the light yield as a function of γ-ray energy, measured with LuAG:Ce and $LaBr_3$:Ce crystals.

Photofraction. The photofraction is defined here as the ratio of counts under the photopeak to the total counts of the spectrum as measured at a specific γ-ray energy. The photofraction for LuAG:Ce and $LaBr_3$:Ce at 320, 662 and 835 keV γ-rays is collected in Table 2. For a comparison, the cross-section ratio (σ-ratio) defined here as the ratio of the photon cross-section for the photoelectric effect to the total one calculated using the WinXCom program [19] is given too. The LuAG:Ce exhibits much higher photofraction than $LaBr_3$:Ce in a similar trend as the σ-ratio obtained from the WinXCom program. The reason is due to higher effective atomic number (58.9 vs 46.9) and density (6.67 vs 5.29 g/cm^3) of the LuAG:Ce crystal. However, the σ-ratio is closer to the measured photofraction for LuAG:Ce than for $LaBr_3$:Ce. It may be due to a smaller size (a factor of 2.5) of LuAG:Ce sample.

Table 2 Photofraction for $LaBr_3$:Ce and LuAG:Ce crystals.

γ energy (keV)	320	662	835	
Source	^{51}Cr	^{137}Cs	^{54}Mn	
Photof. (%)	44.7	16.2	12.7	$LaBr_3$:Ce
σ- ratio (%)	31.3	8.7	5.9	
Photof. (%)	64.8	28.0	22.3	LuAG:Ce
σ- ratio (%)	52.8	19.9	14.2	

Coincidence Timing Resolution. Fig. 4 presents the coincidence timing spectra measured for LuAG:Ce and $LaBr_3$:Ce detectors in combination with a BaF_2 detector. The timing resolution was measured to be 583 ps and 204 ps, respectively, for LuAG:Ce and $LaBr_3$:Ce detectors. The results of the measurements are collected in Table 3. The measured timing resolution, presented in the second column, is corrected for the contribution of the reference BaF_2 detector (128 ps) and shown in the third column. The last column shows the number of photoelectrons corresponding to the 511 keV peak. Note an excellent timing resolution of 159 ps for $LaBr_3$:Ce detector. No doubt that it is the effect of a large number of photoelectrons and a very fast principal decay time constant of about 15 ns for $LaBr_3$:5%Ce [20]. In contrast, the scintillation decay of LuAG:Ce exhibits a fast component of 61 ns contributing with low intensity of 47% in the scintillation pulse as measured within 1 μs range [21]. This is the reason for much worse timing resolution of LuAG:Ce as compared to $LaBr_3$:Ce.

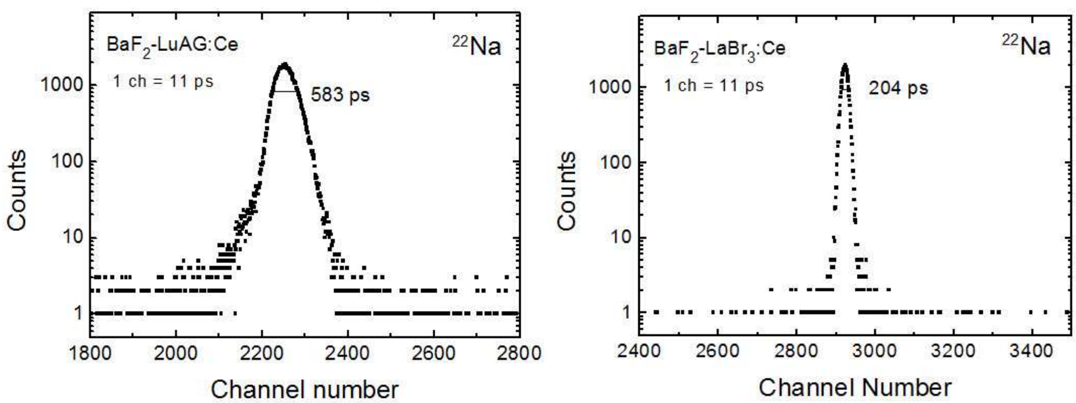

Fig. 4 Timing resolution spectra measured for LuAG:Ce and $LaBr_3$:Ce detectors in coincidence with a BaF_2 detector.

Table 3 Coincidence timing resolution measured with $LaBr_3$:Ce and LuAG:Ce detectors.

Crystal	Timing resolution, δ_t (ps)		N (phe) at 511 keV
	Measured	Tested detector	
$LaBr_3$:Ce	204 ± 5	159 ± 5	6220 ± 400
LuAG:Ce	583 ± 18	568 ± 18	1290 ± 80

Summary

The scintillation properties of LuAG:Ce and $LaBr_3$:Ce scintillators were studied and compared for γ-ray detection. The high energy resolution of 3.5 % for 662 keV γ-rays obtained with $LaBr_3$:Ce is much better than that of 6.7 % for LuAG:Ce. The high light yield and very good proportionality of $LaBr_3$:Ce are the important reasons behind its high energy resolution. It has the potential to replace NaI:Tl as the scintillator of choice for SPECT camera and γ-ray spectroscopy. An advantage of LuAG:Ce is its high density and effective atomic number, which results in a higher detection efficiency for γ-rays. A drawback of LuAG:Ce is its very intense slow component in the scintillation pulse [22,23], which is due to retrapping of charge carriers at shallow traps and appearance of the delayed radiative recombination at the Ce^{3+}-emission centers. It points to a chance to enhance its scintillation intensity of fast component determining both the energy and time resolutions, if related shallow traps could be suppressed. This fact together with the considerably fast scintillation decay and high detection efficiency for γ-rays, would make LuAG:Ce the material of choice for γ-ray spectrometry and PET imaging.

Acknowledgments

This work was supported by King Mongkut's University of Technology Thonburi under the National Research University Project of Thailand's Office of the Higher Education Commission.

References

[1] C.W.E.van Eijk: Nucl. Instrum. Methods Phys. Res.A, Vol.460 (2001), p. 1

[2] K.W. Kramer, P. Dorenbos, H.U. Gudel, and C.W.E.van Eijk: J. Mater. Chem., Vol.16 (2006), p. 2773

[3] M. Nikl: Meas. Sci. Technol. Vol.17 (2006), p. R37

[4] P. Lecoq, A. Annenkov, A. Gektin, M. Korzhik, and C. Pedrini: *Inorganic Scintillators for Detector Systems*, the Netherlands, Springer (2006)

[5] R. Autrata, P. Schauer, J. Kvapil, and J. Kvapil: J. Phys. E, Sci. Instrum., Vol.11(1978), p. 707

[6] M. Moszynski, T. Ludziewski, D. Wolski, W. Klamra, and L.O. Norlin: Nucl. Instrum. Methods Phys. Res. A., Vol.345 (1994), p. 461

[7] A. Lempicki, M.H. Randles, D. Wisniewski, M. Balcerzyk, C. Brecher, and A.J. Wojtowitz: IEEE Trans. Nucl. Sci., Vol.42 (1995), p. 280

[8] M. Nikl, E. Mihokova, J.A. Mares, A. Vedda, M. Martini, K. Nejezchleb, and K. Blazek: Phys. Status Solidi B, Vol.181 (2000), p. R10

[9] J.A. Mares, A. Beitlerova, M. Nikl, N. Solovieva, C. D'Ambrosio, K. Blazek, P. Maly, K. Nejezchleb, and F. de Notaristefani: Rad. Meas., Vol.38 (2004), p. 353

[10] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk K. Kramer, and H.U. Gudel: Appl. Phys. Lett. Vol.77 (2000), p. 1467

[11] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K. Kramer, and H.U. Gudel: Appl. Phys. Lett. Vol.79 (2001), p. 1573

[12] P. Dorenbos, J.T.M. de Haas, and C.W.E. van Eijk: IEEE Trans. Nucl. Sci., Vol.51 (2004), p. 1289

[13] Z. Guzik, S. Borsuk, K. Traczyk, and M. Plominski: IEEE Trans. Nucl. Sci., Vol.53 (2006), p. 231

[14] M. Bertolaccini, S. Cova and C. Bussolatti: *A technique for absolute measurement of the effective photoelectron per keV yield in scintillation counters*, in Proc. Nuclear Electronics Symp., Versailles, France (1968)

[15] M. Moszynski, M. Kapusta, M. Mayhugh, D. Wolski and S.O. Flyckt: IEEE Trans. Nucl. Sci.,Vol.44 (1997), p.1052

[16] M. Moszynski, M. Kapusta, D. Wolski, M. Balcerzyk, S.O. Flyckt, P. Lavoute, C. Marmonie, and H. Mach: IEEE Trans. Nucl. Sci., Vol.51 (2004), p.1701

[17] M. Moszynski, J. Zalipska, M. Balcerzyk, M. Kapusta, W. Mengeshe, and J.D. Valentine: Nucl. Instrum. Methods Phys. Res.A,Vol.484 (2002), p. 259

[18] P. Dorenbos, J.T.M. de Haas, and C.W.E. van Eijk: IEEE Trans. Nucl. Sci.,Vol.42(1995), p.2190

[19] L.Gerward, N. Guilbert, K.B. Jensen, and H. Levring: Rad. Phys. and Chem.,Vol.71(2004), p.653

[20] K.S. Shah, J. Glodo, M. Klugerman, W.W. Moses, S.E. Derenzo, and M.J. Weber: IEEE Trans. Nucl. Sci.,Vol.50 (2003), p. 2410

[21] W. Chewpraditkul, L. Swiderski, M. Moszynski, T. Szczesniak, A. Syntfeld-Kazuch, C. Wanarak and P. Limsuwan: IEEE Trans. Nucl. Sci.,Vol.56 (2009), p. 3800

[22] W. Chewpraditkul, L. Swiderski, M. Moszynski, T. Szczesniak, A. Syntfeld-Kazuch, C. Wanarak and P. Limsuwan: Phys. Status Solidi A, Vol.206 (2009), p.2599

[23] M. Nikl: Phys. Status Solidi A, Vol.202 (2005), p.201

Materials Processing Technology, ICAMMP2011
10.4028/www.scientific.net/AMR.418-420

Comparison of $Lu_3Al_5O_{12}$:Ce and $LaBr_3$:Ce Scintillators in Gamma-Ray Spectrometry
10.4028/www.scientific.net/AMR.418-420.922

DOI References

[1] C.W.E. van Eijk: Nucl. Instrum. Methods Phys. Res. A, Vol. 460 (2001), p.1.
http://dx.doi.org/10.1016/S0168-9002(00)01088-3
[2] K.W. Kramer, P. Dorenbos, H.U. Gudel, and C.W.E. van Eijk: J. Mater. Chem., Vol. 16 (2006), p.2773.
http://dx.doi.org/10.1039/b602762h
[5] R. Autrata, P. Schauer, J. Kvapil, and J. Kvapil: J. Phys. E, Sci. Instrum., Vol. 11(1978), p.707.
http://dx.doi.org/10.1088/0022-3735/11/7/028
[6] M. Moszynski, T. Ludziewski, D. Wolski, W. Klamra, and L.O. Norlin: Nucl. Instrum. Methods Phys. Res. A., Vol. 345 (1994), p.461.
http://dx.doi.org/10.1016/0168-9002(94)90500-2
[7] A. Lempicki, M.H. Randles, D. Wisniewski, M. Balcerzyk, C. Brecher, and A.J. Wojtowitz: IEEE Trans. Nucl. Sci., Vol. 42 (1995), p.280.
http://dx.doi.org/10.1109/23.467836
[9] J.A. Mares, A. Beitlerova, M. Nikl, N. Solovieva, C. D'Ambrosio, K. Blazek, P. Maly, K. Nejezchleb, and F. de Notaristefani: Rad. Meas., Vol. 38 (2004), p.353.
http://dx.doi.org/10.1016/j.radmeas.2004.04.004
[10] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk K. Kramer, and H.U. Gudel: Appl. Phys. Lett. Vol. 77 (2000), p.1467.
http://dx.doi.org/10.1063/1.1308053
[11] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K. Kramer, and H.U. Gudel: Appl. Phys. Lett. Vol. 79 (2001), p.1573.
http://dx.doi.org/10.1063/1.1385342
[12] P. Dorenbos, J.T.M. de Haas, and C.W.E. van Eijk: IEEE Trans. Nucl. Sci., Vol. 51 (2004), p.1289.
http://dx.doi.org/10.1109/TNS.2004.829375
[13] Z. Guzik, S. Borsuk, K. Traczyk, and M. Plominski: IEEE Trans. Nucl. Sci., Vol. 53 (2006), p.231.
http://dx.doi.org/10.1109/TNS.2006.869819
[15] M. Moszynski, M. Kapusta, M. Mayhugh, D. Wolski and S.O. Flyckt: IEEE Trans. Nucl. Sci., Vol. 44 (1997), p.1052.
http://dx.doi.org/10.1109/23.603803
[16] M. Moszynski, M. Kapusta, D. Wolski, M. Balcerzyk, S.O. Flyckt, P. Lavoute, C. Marmonie, and H. Mach: IEEE Trans. Nucl. Sci., Vol. 51 (2004), p.1701.
http://dx.doi.org/10.1109/TNS.2004.832322
[17] M. Moszynski, J. Zalipska, M. Balcerzyk, M. Kapusta, W. Mengeshe, and J.D. Valentine: Nucl. Instrum. Methods Phys. Res. A, Vol. 484 (2002), p.259.
http://dx.doi.org/10.1016/S0168-9002(01)01964-7
[18] P. Dorenbos, J.T.M. de Haas, and C.W.E. van Eijk: IEEE Trans. Nucl. Sci., Vol. 42(1995), p.2190.
http://dx.doi.org/10.1109/23.489415
[19] L. Gerward, N. Guilbert, K.B. Jensen, and H. Levring: Rad. Phys. and Chem., Vol. 71(2004), p.653.
http://dx.doi.org/10.1016/j.radphyschem.2004.04.040

[20] K.S. Shah, J. Glodo, M. Klugerman, W.W. Moses, S.E. Derenzo, and M.J. Weber: IEEE Trans. Nucl. Sci., Vol. 50 (2003), p.2410.
http://dx.doi.org/10.1109/TNS.2003.820614
[21] W. Chewpraditkul, L. Swiderski, M. Moszynski, T. Szczesniak, A. Syntfeld-Kazuch, C. Wanarak and P. Limsuwan: IEEE Trans. Nucl. Sci., Vol. 56 (2009), p.3800.
http://dx.doi.org/10.1109/TNS.2009.2033994
[22] W. Chewpraditkul, L. Swiderski, M. Moszynski, T. Szczesniak, A. Syntfeld-Kazuch, C. Wanarak and P. Limsuwan: Phys. Status Solidi A, Vol. 206 (2009), p.2599.
http://dx.doi.org/10.1002/pssa.200925161
[23] M. Nikl: Phys. Status Solidi A, Vol. 202 (2005), p.201.
http://dx.doi.org/10.1002/pssa.200460107