INVESTIGATION OF $LaBr_3$:Ce AND $LaCl_3$:Ce SCINTILLATORS FOR SPECT IMAGING

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ABSTRACT

This study aims to investigate the potential use of $LaBr_3$:Ce and $LaCl_3$:Ce materials in SPECT imaging. GATE Monte Carlo simulations of single-head gamma camera and a $^{99}Tc^{m}$ point source were performed to evaluate the energy spectra, modulation transfer function (MTF) and detection efficiency. A range of Ce concentrations (0.5, 5, 10 and 15)% of $LaBr_3$:Ce and $LaCl_3$:Ce crystals was simulated in conjunction with efficiency calculation to find optimal concentration for SPECT imaging. The MTF curves showed the excellent MTF performance of $LaCl_3$ and $LaBr_3$ particularly at low frequencies. The intrinsic efficiency results demonstrated the superiority of $LaBr_3$:Ce crystals with respect to $LaCl_3$:Ce and NaI(Tl). Results also suggest that higher Ce concentrations for both $LaBr_3$:Ce and $LaCl_3$:Ce crystals only slightly improve intrinsic efficiency. In conclusion, because $LaCl_3$:Ce and $LaBr_3$:Ce combine excellent MTF performance with increased intrinsic efficiency, they have the potential to replace NaI(Tl) as the scintillators of choice for SPECT.

Index Terms— $LaBr_3$:Ce, $LaCl_3$:Ce, SPECT, GATE

1. INTRODUCTION

There has been considerable research and development of inorganic scintillators for Single Photon Emission Computed Tomography (SPECT) imaging over the past several decades and the search for the ideal scintillator is intensifying [1]. Ideally, scintillation crystals used in SPECT should have high light output (for good energy resolution and intrinsic spatial resolution), high density (>3.5 g/cm3), an emission wavelength well matched to photomultiplier tube readout (300–500nm), short decay time (<1μs) and of course be cost-effective [2].

Cerium-doped lanthanum crystals, particularly $LaBr_3$:Ce and $LaCl_3$:Ce, have lately drawn significant interest due to their high scintillation yield and superior energy resolution which make them attractive for SPECT imaging [3]. In comparison to NaI(Tl), $LaBr_3$:Ce and $LaCl_3$:Ce have 30% and 60% higher light output respectively, and better energy resolution (6-7% vs. 9% FWHM) [3]. This higher light output would allow Anger cameras to use 76 mm PMTs to reach intrinsic spatial resolution similar to what is presently achieved with 51 mm PMTs (3.5mm FWHM), reducing the number of PMTs by 25%. The improved energy resolution would allow the Compton scatter background to be reduced from 35% to as low as 25%. $LaBr_3$:Ce has the additional benefit of shorter attenuation length, which would reduce the volume of scintillator by one quarter; hence, improving intrinsic spatial resolution [2]. Table 1 summarises the comparative scintillation properties that are most relevant for SPECT.

Table 1: Summary of comparative properties for $LaBr_3$:Ce $LaCl_3$:Ce and NaI(Tl) scintillators.

	$LaBr_3$:Ce	$LaCl_3$:Ce	NaI(Tl)
Density (gm/cm^3)	5.29	3.79	3.67
Effective atomic number of host (*Zeff*)	46.9	49.5	50.0
Energy resolution (at 140 keV)	≈ 6%	≈ 7.5%	9.5%
Light output (photons/MeV)	63,000	46,000	39,000
Wavelength (mm)	380	350	415
Decay time (ns)	20	25	240
Attenuation length @ 140keV (mm)	3.7	3.9	5.3

Unfortunately, the lanthanum halide scintillators have a few drawbacks of their own; such as, hygroscopic nature and internal radioactivity [4]. Nevertheless, the hygroscopicity needs not to be of great concern when the material seems to have very good scintillation properties and the internal radioactivity drawback is likely to be serious only for the very long times of counting in low activity measurement.

The main thrust of the present study was to investigate the potential use of $LaBr_3$:Ce and $LaCl_3$:Ce materials in SPECT compared to NaI(Tl) using GATE Monte Carlo simulation. System performance was assessed using energy spectra, energy resolution, detection efficiency, and MTF calculations.

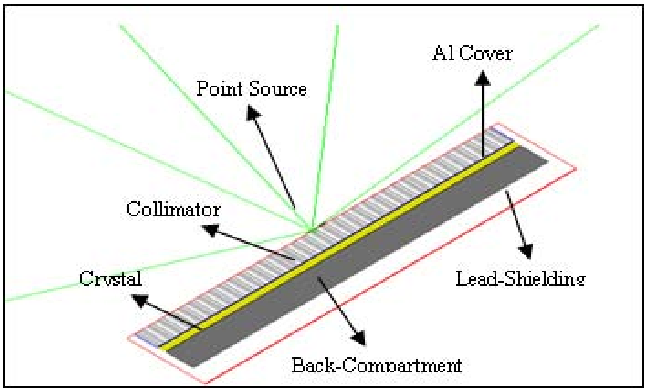

Fig. 1: Picture of single-head gamma camera as modelled by GATE.

2. METHOD

Monte Carlo simulations are increasingly used in nuclear medicine imaging to model imaging systems and to develop and evaluate tomographic reconstruction techniques and correction methods for improved image quantification. GATE (GEANT4 application for tomographic emission) is a relatively new Monte Carlo simulation package based on GEANT4 dedicated to nuclear imaging applications. GATE accurately allows for an Monte Carlo modelling of photon transport in the phantom and in the collimator, crystal, head shielding and scanning table [5]. In this study, GATE V.3.1.1 was used.

First, $LaCl_3$ and $LaBr_3$ of different Ce concentrations (0.5, 5, 10 and 20) % had to be stated in the GATE Material data base. A single-head camera was modelled as a combination of (Fig.1):

- Low-Energy High-Resolution (LEHR) collimator made of lead (hole diameter: 1.4 mm, collimator thickness: 32 mm and septal thickness: 0.156 mm);
- (560 × 560 × 9.5) mm scintillator crystal ($LaCl_3$:Ce, $LaBr_3$:Ce or NaI(Tl));
- Shielding made of lead, 35 mm thick around the camera head and 30 mm thick on the rear.

GATE allows the modelling of a so-called back-compartment to account for the photomultipliers and electronics located behind the crystal. Assie *et al* have demonstrated the vital role of back-compartment modelling in GATE, without which, large differences between simulated and experimental data were observed [6]. Hence, a back-compartment was modelled as a 50 mm Perspex layer (density 2.5 g/cm^3). For a more realistic representation, an aluminum cover of 0.1 mm thickness was simulated for each detector.

2.1. Energy spectra evaluation

A Gaussian energy blurring of FWHM= 9.5%, 7.5 % and 6.5 % at 140 keV was used in the simulation of NaI(Tl), $LaCl_3$:Ce and $LaBr_3$:Ce respectively. Our Gaussian blurring values were based on experimental energy resolution measurements of Ø 25.0 × 25.0 mm, Ø 50.8 × 50.8 mm and Ø 44.4 × 50.8 mm of $LaBr_3$:Ce, NaI(Tl) and $LaCl_3$:Ce respectively. Furthermore, the intrinsic crystal resolution for NaI(Tl) (3.4mm intrinsic crystal resolution), LaCl (3mm) and LaBr (2.4mm) was modelled based on the following respectively: (i) a prior experimental measurement[1] (ii) an estimation based on $LaCl_3$:Ce light output value compared to NaI(Tl) and $LaBr_3$:Ce (Table 1) and (iii) Ref. [7]. The energy spectra were simulated over the whole field of view (FOV) in air, with the $^{99}Tc^{m}$ point source located at the centre of the FOV, at 15 cm from the collimator. A thresholder and an upholder were used to consider only the particles detected with energies between 0 and 190 keV.

2.2. Detection efficiency

The intrinsic efficiency, defined as the number of pulses recorded divided by number of radiations striking crystal, was evaluated by simulating an intrinsic static scan of a $^{99}Tc^{m}$ point source in air, located at the centre of the FOV and at 25 cm from the surface of the NaI(Tl) crystal. An identical simulation was repeated for the $LaCl_3$:Ce(10%) and $LaBr_3$:Ce(5%) scintillator. For each simulation 500 million events were tracked. . In order to investigate the effect of Ce concentration on efficiency, the simulation was repeated for the $LaBr_3$:Ce and $LaCl_3$:Ce SPECT based crystal with Ce concentrations of 0.5, 5, 10 and 15%.

2.3. MTF Analysis

The point spread function (PSF) was simulated using a $^{99}Tc^{m}$ point source located at distance of 0 mm between the source and the collimator as shown in Fig.1. This is because of a difference between different detector materials PSF is evident only for 0 mm separation. At larger distances, the PSF is determined only by the collimator, not by the detector type (either pixellated or continuous crystal). The MTF was calculated then by taking the fast Fourier transform (FFT) of the normalised PSF [8].

3. RESULTS AND DISCUSSIONS

3.1. Energy spectra

The spectrum was analysed with ROOT (V 5.12). ROOT is based on the built-in C^{++} interpreter and provides users with the functionality needed to handle and analyse large amounts of data in a very efficient way. Fig.2 shows comparable energy spectra from the $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) crystal based systems for a $^{99}Tc^{m}$ point source in air. Cerium-doped lanthanum- based systems have

[1] The measurement was carried out using a SKYLight/Precedence gamma camera (Philips) with a 9.5 mm NaI(Tl) crystal.

significantly better energy resolution than sodium iodide-based systems.

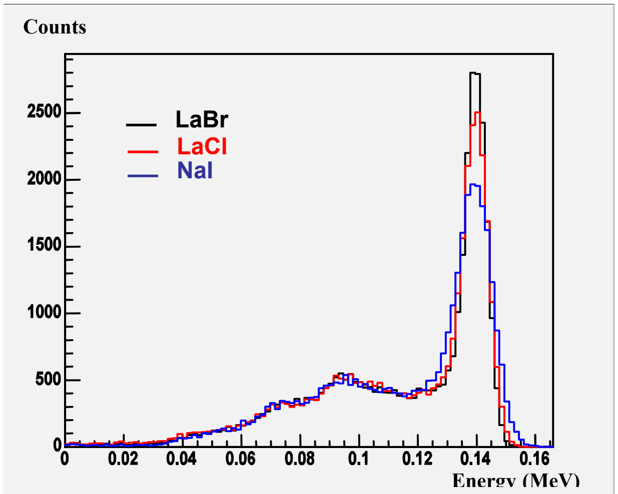

Fig.2: Comparison of energy spectra of $^{99}Tc^{m}$ obtained by $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) scintillators.

Good energy resolution is a desirable characteristic for any spectrometry system because it permits a precise identification and separation between γ-rays of very close energies, for scatter rejection. Superior energy resolution is particularly important for radionuclides with more than one photo-peak energy or for dual radioisotope imaging. The superior energy resolution of $LaBr_3$:Ce, and $LaCl_3$:Ce is due to due to a very high light output and very small non-proportionality with photon energy of the scintillator (less than 5%).

3.2. Detection Efficiency

The intrinsic efficiencies of $LaBr_3$:Ce(5%), $LaCl_3$:Ce(10%) and NaI(Tl) crystal at 140 keV were found to be (94.3±0.6)%, (91.7±0.7)% and (90.5±0.5)%, respectively. The associated relative standard deviations were calculated by repeating each simulation five times. The lanthanum bromide scintillator shows higher intrinsic efficiency than both lanthanum chloride and sodium iodide crystals. This is due to its high density as shown in Table 1.

Also, to demonstrate the superiority of detection efficiency of cerium-doped lanthanum crystals with respect to NaI(Tl) scintillator, a wide range of crystal thicknesses (1, 2, 3,…35) mm was simulated along with recording the detected photons, as shown in Fig. 3. Currently, most gamma cameras employ a large area of scintillator of ≈10 mm thickness. Fig. 3 suggests that intrinsic spatial resolution can be improved by reducing crystal thickness of $LaBr_3$:Ce and $LaCl_3$:Ce up to ≈7 mm while the detection efficiency is comparable to the NaI(Tl) crystal of 10 mm thickness.

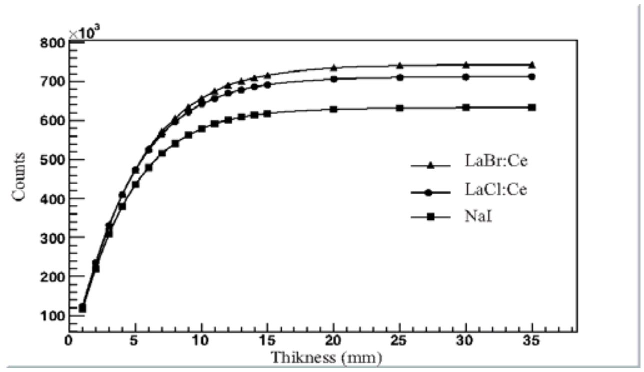

Fig. 3: Detected events versus a range of crystal thickness for $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) sintillator.

In general, Table 2 suggests that the higher Ce concentration for both $LaBr_3$:Ce and $LaCl_3$:Ce crystals only slightly improves the intrinsic efficiency. The improvement from 0.5% to 5.0% Ce concentration was more significant than the improvement from 5.0% to 15.0%. This is due to the high atomic number of Ce (Z=58) which increases Z_{eff} of the crystal detector. However, to give a more definitive judgment on the effect of Ce concentration on the overall detection performance, more investigations regarding decay time and rise time have to be performed. Furthermore, cost-effectiveness and development in crystal growth techniques have to be considered.

Table 2: Intrinsic efficiency of $LaBr_3$:Ce and $LaCl_3$:Ce with different Ce concentrations for the $^{99}Tc^{m}$ 140 keV γ-ray.

Ce Concentration (%)		0.5	5.0	10	15
Intrinsic efficiency (%)	$LaBr_3$	89.6±1.2	94.3±0.6	95.2±0.5	95.8±0.7
	$LaCl_3$	86.8±0.9	88.5±1.2	91.7±0.7	92.9±0.4

3.3. MTF Analysis

Fig.4 shows the PSF of a $^{99}Tc^{m}$ point source, located on the surface of the LEHR collimator, for the three different scintillators. The FWHM of the PSF obtained for $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) are 3.4, 3.9 and 4.1 mm respectively.

However, the FWHM of the PSF is a relatively crude expression of resolution and is also an insensitive measure of the effect of scattered radiation on resolution [7]. Therefore, a more comprehensive expression of the ability of the gamma-camera to reproduce spatial information is given by the MTF which shows directly the extent to which the information carried by each spatial frequency has been attenuated by the imaging system. Fig.5 shows the calculated MTFs for all detector scintillators. It can be clearly seen that the excellent MTF performance of $LaBr_3$:Ce and $LaCl_3$:Ce especially at low frequencies. This means that the cerium-doped lanthanum crystals, $LaBr_3$:Ce and $LaCl_3$:Ce, are better in visualising large low-contrast structures. This could be due to the fact that $LaBr_3$:Ce and

$LaCl_3$:Ce have respectively 30% and 60% higher light output than NaI(Tl).

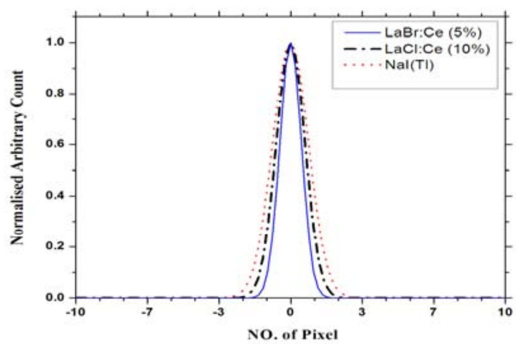

Fig.4: PSF of a ^{99m}Tc point source for the $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) scintillator.

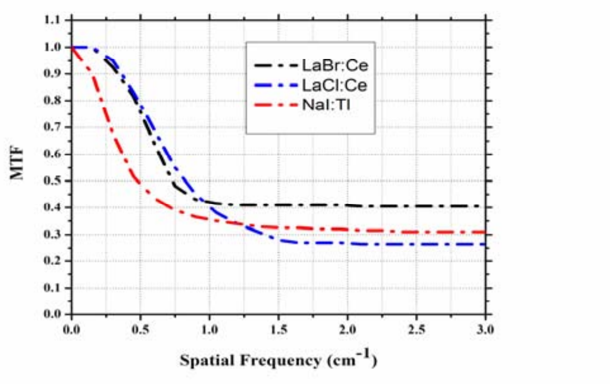

Fig.5: MTF for the $LaBr_3$:Ce, $LaCl_3$:Ce and NaI(Tl) scintillator.

4. CONCLUSSION

In this study, we have presented results of investigation for the potential use of relatively new cerium-doped lanthanum crystals, $LaBr_3$:Ce and $LaCl_3$:Ce, using GATE Monte Carlo simulation. Our research focused on evaluating energy spectra, detection efficiency, and MTF curves.

In comparison to a NaI(Tl) scintillator based SPECT system, $LaBr_3$:Ce and $LaCl_3$:Ce scintillators have excellent energy resolution, superior MTF performance and at least comparable detection efficiency. Based on intrinsic efficiency calculations, the higher Ce concentrations for both $LaBr_3$:Ce and $LaCl_3$:Ce crystals slightly improves the intrinsic efficiency. The increase in intrinsic spatial and energy resolution is leading to higher contrast resolution and improved detection of abnormalities.

In conclusion, because the relatively new cerium-doped scintillators; particularly $LaBr_3$:Ce, have excellent energy resolution and slightly higher detection efficiency notwithstanding comparison of other performance aspects, they have the potential to replace NaI(Tl) as the scintillator of choice in SPECT imaging systems. However, further investigations, such as making sure $LaCl_3$:Ce and $LaBr_3$:Ce features are maintained as the crystal volume is increased, and low cost crystal growth techniques are needed before these materials can be commonly used in preclinical and clinical SPECT systems.

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