Comparative Investigation of Ce\textsuperscript{3+} Doped Scintillators in a Wide Range of Photon Energies Covering X-ray CT, Nuclear Medicine and Megavoltage Radiation Therapy Portal Imaging Applications

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Abstract—The aim of the present work is to study the performance of scintillators currently used in PET and animal PET systems, under conditions met in radiation therapy and PET/CT imaging. The results of this study will be useful in applications where both CT and PET photons as well as megavoltage cone beam CT (MV CBCT) photons could be detected using a common detector unit. To this aim crystal samples of GSO, LSO, LYSO, LuYAP and YAP scintillators, doped with cerium (Ce\textsuperscript{3+}) were examined under a wide energy range of photon energies. Evaluation was performed by determining the absolute luminescence efficiency (emitted light flux over incident X-ray exposure) in the energy range employed in X-ray CT, in Nuclear Medicine (70 keV up to 662 keV) and in radiotherapy 6 MV (approx. 2.0 MeV mean energy)—18 MV (approx. 4.5 MeV mean energy). Measurements were performed using an experimental set-up based on a photomultiplier coupled to a light integration sphere. The emission spectrum under X-ray excitation was measured, using an optical grating monochromator, to determine the spectral compatibility to optical photon detectors incorporated in medical imaging systems. Maximum absolute luminescence efficiency values were observed at 70 keV for YAP:Ce and LuYAP:Ce and at 140 keV for LSO:Ce, LYSO:Ce and GSO:Ce. Highest absolute efficiency between the scintillators examined was observed for LSO:Ce, followed by LYSO:Ce and GSO:Ce. The detector optical gain (DOG) exhibited a significant variation with the increase of energy between 70 keV to 2.0 MeV. All scintillators exhibited low compatibility when combined with GaAsP (G5645) photodetector.

Index Terms—Biomedical applications of radiation, cerium compounds, radioluminescence, scintillation detectors.

I. INTRODUCTION

CERIUM doped (Ce\textsuperscript{3+}) inorganic single crystal scintillators such as GSO, LSO, LYSO, LuYAP and YAP have already been used or suggested for use in PET, animal PET and other nuclear medicine modalities [1], [2]. Their high light yield and high density make them interesting for applications involving imaging at megavoltage energy X-rays, such as radiotherapy treatment beams [3].

Current trends in multimodality imaging detectors provide indications that the exploitation of the above scintillators in a wider range of energies, covering CT/PET and Portal Imaging, [3], [4] could offer additional advantages. A PET/CT imaging system relying on the same detectors and electronics would eliminate the need to align images, would reduce the space needed to house the whole apparatus, and would reduce the overall cost of the system [5]. CT detectors require small thicknesses of scintillation crystals. However in order to construct a PET/CT system based on the same detector, crystal thickness should be increased at the expense of CT resolution. The short decay time of the Ce\textsuperscript{3+} doped single crystal scintillators favors their use in combined PET/CT detectors [6]. Megavoltage cone-beam computed tomography (MV CBCT) using portal imaging detectors, is a highly promising technique for providing volumetric patient position information in the radiation treatment room [7]. If the quantum efficiency of the X-ray detector was considerably improved, it is reasonable to expect that soft tissues could be visualized at considerably lower, clinically acceptable doses using megavoltage cone-beam (or fan-beam) CT. For this reason, high efficiency X-ray detectors have been widely investigated [8].

The purpose of the present study was to investigate the variation of the luminescence emission efficiency of LSO:Ce, LYSO:Ce, GSO:Ce, LuYAP:Ce and YAP:Ce single crystal scintillators in a wide range of photon energies, from diagnostic X-ray CT up to Portal Imaging applications used in megavoltage Radiation Therapy, i.e., between 70 keV (corresponding to X-ray CT imaging) and 18 MV photons (approx. 4.5 MeV mean energy). Results would be of value in the design
of a combined PET/CT and/or Portal Imaging system. For comparison, the luminescence emission efficiency of BGO crystal, which has been successfully employed in many PET and occasionally in some X-ray CT systems, was also examined. To our knowledge the aforementioned scintillating crystals have never been systematically studied and inter-compared in the above energy range for imaging applications.

II. MATERIALS AND METHODS

Crystal samples of LSO:Ce, LYSO:Ce, GSO:Ce, LuYAP:Ce and YAP:Ce, with thicknesses between 8 mm to 20 mm were irradiated with photons in a wide imaging energy range from 130 keV X-ray tube voltage (corresponding to 70 keV mean energy) through 140, 364 and 662 keV gamma ray energy, to 18 MV photons used in radiotherapy (corresponding to a mean energy of 4.5 MeV ± 0.5 MeV). In particular, the samples of GSO:Ce, LSO:Ce and LYSO:Ce crystals had dimensions of 10 mm × 10 mm × 10 mm. The sample of LuYAP:Ce crystal had dimensions of 2 mm × 2 mm × 8 mm while the YAP:Ce crystal had dimensions of 10 mm × 10 mm × 20 mm. All crystals were doped with Ce³⁺ and all their surfaces were polished.

Evaluation was performed by determining:

A) The absolute luminescence efficiency (AE) [13], i.e., the ratio of the light energy fluence $\Psi(\lambda)$ (light energy per unit of area) emitted by the irradiated scintillator over the air KERMA, $K_{air}$, corresponding to the X-ray or gamma ray beam, incident on the scintillator surface. AE expresses the ability of a scintillator to convert incident X-ray or gamma ray exposure rate into emitted light energy. It may be used to describe the radiation detection sensitivity of energy integrating detectors, i.e., detectors producing a signal directly related to the total energy absorbed within the scintillator’s mass. The latter can be determined through ionization chamber measurements in the energy range employed in X-ray CT, in Nuclear Medicine (70 keV up to 662 keV) and in radiotherapy (6 MV and 18 MV). Absolute efficiency is thus expressed in units of $\mu W \cdot m^{-2} / mGy \cdot s^{-1}$ [or AE units]. The use of high AE scintillator materials may reduce patient radiation dose in many medical imaging examinations.

The crystal samples were irradiated: a) in a Philips Optimus general radiography X-ray unit with a tungsten (W) target anode and 2 mm Al filter (W/Al X-ray spectrum); operated at 130 kVp (tube voltage) to produce X-rays, corresponding to a mean energy of approximately 70 keV, which is within the energy range used in general purpose computed tomography and possible breast CT applications, b) at 140 keV (Tc-99 m), 364 keV (I-131) and 662 keV (Cs-137) $\gamma$-rays covering the range of energies used in nuclear medicine imaging and c) in a Siemens Primus High (Siemens Medical Systems, Concord, CA, USA) radiation therapy unit using 6 MV and 18 MV radiotherapy megavoltage treatment beams (corresponding to approximately 2.0 MeV ± 0.2 MeV and 4.5 MeV ± 0.5 MeV mean energies) used in MV-CBCT applications. Light energy fluence measurements were performed using an experimental apparatus and methods that have been previously described by Valais et al., [14], [15]. In the X-ray diagnostic energy range the mean photon energy was determined as described in [16]. In the megavoltage energy range, the mean energy of the 6 MV photon beam was taken from Fix et al., [17] while for the 18 MV beam it was calculated through linear fitting of the mean energy values calculated for 6 MV and 10 MV beams as described in [18].

The X-ray and $\gamma$-ray Air Kerma rate, was determined by ionization chamber (UNIDOS, PTW) measurements.

B) The spectral matching factor (SMF Or $\alpha_S$), which expresses the spectral compatibility of the excited scintillator emitted light to the spectral sensitivity of the optical photon detectors used and can be expressed by the ratio [19]

$$SMF = \alpha_S = \frac{\int S_P(\lambda)S_D(\lambda) d\lambda}{\int S_P(\lambda) d\lambda}$$  \hspace{1cm} (1)

where $S_P$ is the spectrum of the light emitted by the scintillator, $S_D$ is the spectral sensitivity of the optical photon detector and $\lambda$ denotes the wavelength of the emitted light. SMF was estimated from the emitted spectrum ($S_P(\lambda)$) of the examined scintillator crystals, which was measured using a grating optical spectrometer (Ocean Optics Inc., HR2000) and from the spectral sensitivity ($S_D(\lambda)$) data, which were obtained from corresponding manufacturer’s (Hamamatsu, EMI, etc.) datasheets. Optical spectrometry measurements were performed under X-ray excitation previously described by Valais et al., [16].

Six optical photon detectors commonly used in a wide range of medical imaging modalities (digital and conventional radiography, computed tomography, nuclear medicine, portal imaging, etc) and their spectral matching factor with the examined scintillator spectra were assessed (Table II). These optical detectors were the following: (i) XP2020 Photonis photocathode, (ii) a-Si:H/108H Hamamatsu amorphous silicon photodiode corresponding to intrinsic layer thickness of 800 nm (108 H), (iii) H8500/9500 Hamamatsu photocathode and (iv) S8550 Hamamatsu avalanche photodiode (APD), (v) S10420–1006 Hamamatsu CCD and vi) G5645 GasAsP photodiode.

C) The detector optical gain (DOG) Which was defined as the number of light photons emitted per incident X-ray or gamma ray photon, given by the ratio [19]

$$DOG = \frac{\Phi_X}{\Phi_X}$$  \hspace{1cm} (2)

where $\Phi_X$ is the emitted light photon flux and is the X-ray or gamma ray photon flux incident on the scintillator. The latter was calculated by converting the air KERMA data
Fig. 1. Absolute efficiency of LSO:Ce, LYSO:Ce, GSO:Ce, LuYAP:Ce and YAP:Ce crystals in the energy range between 70 keV–4.5 MeV.

The light photon flux was determined as follows

$$\Phi_\lambda = \frac{\Psi_\lambda}{\hbar \nu} \quad (3)$$

where $\nu$ is the mean value of the light frequency which was determined from optical spectrometry data. DOG is used to express the net amplification effect (in terms of number of quanta) of an X-ray imaging scintillator as a unitless quantity [20]. The knowledge of DOG is useful in calculations related to X-ray image quality parameters such as the quantum noise power spectrum and modulation transfer function [17], [21].

III. RESULTS AND DISCUSSION

The variation of the absolute luminescence efficiency of GSO:Ce, LSO:Ce, LYSO:Ce, LuYAP:Ce and YAP:Ce scintillators in the energy range examined in this study (70 keV–5.4 MeV) is shown in Fig. 1. For comparison reasons the absolute efficiency of BGO crystal (10 mm × 10 mm × 10 mm), measured under the same conditions, is also shown. In the energy range from 70 keV to 140 keV, the absolute efficiency of LSO:Ce, LYSO:Ce, LuYAP:Ce and GSO:Ce crystals was found to increase with photon energy (Fig. 1). For energies higher than 140 keV the absolute efficiency of these crystals decreased significantly. However, an additional slight increase was observed at 2.0 MeV–4.5 MeV, for LSO:Ce, LYSO:Ce, GSO:Ce and at 662 keV–4.5 MeV, for LuYAP:Ce and YAP:Ce crystals. The similarities in the AE variation between the GSO:Ce and the Lu-based scintillators maybe attributed to their similar absorption properties due to: (i) the presence of the corresponding heavy elements, with close atomic number values (64 for Gd and 71 for Lu), (ii) the high density of these materials (see Table I). On the other hand the absolute luminescence efficiency of the YAP:Ce scintillator, with lower atomic number and lower density was found with significantly different variation, decreasing continuously between 70–662 keV (Fig. 1).

Maximum difference in the AE between LSO:Ce and LYSO:Ce scintillators was observed up to 140 keV, whereas at higher energies this difference decreases. Between 2.0 MeV and 4.5 MeV these two scintillators exhibited almost equal AE values. Similarly, the difference in AE between LuYAP:Ce and YAP:Ce was maximized between 70 to 140 keV.

All the Ce-doped scintillators, exhibited higher AE values compared to BGO for the whole energy range examined in the present work.

Although below 140 keV LuYAP:Ce and YAP:Ce exhibited higher AE values than GSO:Ce, as the energy increases, this finding is reversed up at approximately 364 keV. At higher energies, the AE of GSO:Ce decreases continuously to values lower than the corresponding of YAP:Ce in the energy range between 662 keV–4.5 MeV. These findings indicate that the luminescence efficiency of the examined scintillators may vary significantly with photon energy. In cases where comparison between scintillators is required, such efficiency variations should be taken into account and efficiency should be examined within the required energy range.

Fig. 2 shows the variation of energy absorption coefficients (proportional to cross section) of the examined scintillation crystals with energy (obtained from Hubbell and Seltzer, 1995). The Lu-based scintillators have similar energy absorption coefficients with a very narrow spread of their K-characteristic. The presence of Yttrium in YAP decreases absorption in this
Fig. 2. The variation of energy absorption coefficients of the examined scintillation crystals with energy (obtained from Hubbell and Seltzer, 1995). Above 300 keV energy absorption coefficient values are almost constant and close to zero.

Fig. 3. Detector Optical Gain (DOG) of LSO:Ce, LYSO:Ce, GSO:Ce, LuYAP:Ce and YAP:Ce crystals in the energy range between 70 keV–4.5 MeV. On the other hand, the presence of Gd in GSO results in higher energy absorption coefficient values, than those of Lu-based crystals, in the energy range between 60–90 keV. However as the energy increases, the energy absorption coefficients of GSO tend to decrease to values lower than those corresponding to the Lu-based crystals.

As it can be observed Lu-based crystals have higher absorption coefficient values, which obviously affect the increased light flux of these materials. However this is not the case for GSO:Ce crystal which although has high cross section values, it showed lower light flux. This behavior could be attributed to the low intrinsic light yield of this material as shown in Table I.

The calculated detector optical gain (DOG) for the examined scintillators is shown in Fig. 3. The value of DOG which can be a measure of light yield, varies as photon energy increases with maximum value at 140 keV for Lu-based and Gd-based scintillators.

The shape of the DOG curves can be explained by considering that, in the range of lower X-ray energies (50–140 keV), since X-ray photons are totally absorbed in the crystal and since their energy increases gradually, the light created per photon is larger and therefore DOG increases. At higher photon energies more and more photons are transmitted through the crystal without any interaction. Thus DOG shows a tendency to decrease.

DOG reflects more accurately the detector intrinsic properties, since it is not affected by the air-ionization chamber properties used to determine AE [15]. The latter depends on the X-ray energy fluence to exposure conversion factor, which exhibits a highly non-linear variation with X-ray energy [20]. This may explain the differences in the curves between AE and DOG. As it can be observed from Fig. 3 Lu and Gd based crystals show their highest DOG values in the range of 100–140 keV employed in CT imaging. At PET energies DOG is significantly lower. However, since this is also the case for presently used scintillators, it is not expected to degrade detector performance of a common scintillator system [6].

The calculated spectral matching factor (SMF) values are shown in Table II. All scintillators, except LuYAP:Ce and YAP:Ce, exhibited high compatibility (SMF values higher than 0.5) with the optical photon detectors examined in this study. LuYAP:Ce and YAP:Ce scintillators had the lowest compatibility when combined with amorphous silicon photon detectors (a-Si:H).

GaAsP G5645 photodiode exhibited low compatibility with all scintillators examined and therefore under our experimental condition it may not be considered as a photodetector of choice.
The effective efficiency of all the scintillators investigated in our study. GaAsP G5645 photodetector exhibited low compatibility when combined with amorphous silicon (a-Si:H) and when combined with avalanche photodiode (APD) is shown in Figs. 4 and 5 respectively. The effective efficiency of YAP:Ce and LuYAP:Ce scintillators remain almost constant to the lowest values, exhibiting the maximum deviation from their luminescence efficiency values when combined with a-Si photodiode (Fig. 5, Table II).

IV. CONCLUSIONS

The absolute luminescence efficiency of all the scintillators examined showed a decrease as the energy of the radiation increases above 140 keV. LSO:Ce was found to have the best luminescence efficiency of all the scintillators investigated in our study. GaAsP G5645 photodetector exhibited low compatibility with all scintillators. YAP:Ce and LuYAP:Ce scintillators show very low compatibility with amorphous silicon photodiode and this result put into consideration their use in portal imaging.

REFERENCES