

# Development of medical scintillator

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**Abstract** This review presents the scintillators requirements for the medical imaging modalities. The history and the development in recent years of the medical scintillators (mainly for GSO:Ce, LSO:Ce, LuAP:Ce) are expatiated in detail.

Since Rontgen discovered X-ray in 1895 and two British doctors took the first X-ray picture one year later<sup>[1]</sup>, the ray (X-ray or  $\gamma$ -ray) imaging techniques have played a vital role in diagnosing disease because they can help the doctors to observe the interior of patients bodies. A rapid development has been got in medical imaging field especially for the X-and  $\gamma$ -ray imaging techniques since the 1950s. The imaging techniques make use of the detection of rays and deal with electronic system and computers. The function of the scintillator in the imaging systems is to absorb the incident high energy photon and convert it into visible light. Medical imaging systems contain four modalities, namely planar X-ray imaging, X-ray computed tomography (X-ray CT), single photon emission computed tomography (SPECT) and positron emission tomography (PET). With the increasing requirements for medical imaging equipment, the demand for the scintillators as the detection materials in imaging systems is enormous. About 175 metric tons of scintillator are required annually and the demand for the various scintillators is shown in table 1<sup>[2]</sup>.

Table 1 Annual volume of medical scintillator

Modality	Annual production (screens)	Annual scint. volume/m <sup>3</sup>
Planar X-ray	1000000	50
X-CT	2000	0.15
SPECT	2000	6
PET	50	0.5

In general, an ideal medical scintillator should have the following properties: (1) high luminous efficiency; (2) short decay time; (3) no after glow; (4) high density; (5) short radiation length; (6) good spectral match to photodetectors; and (7) low cost<sup>[2]</sup>. The luminous efficiency must be very high in order to reduce the radiation dose to patients as safety concerns. Short decay time will improve the time-resolution of detecting high energy photon. The after glow is also a critical parameter. If the

after glow intensity exceeds specific limitations, image degradation occurs due to memory effects. The decay time and after glow are the time properties of scintillation but the decay time describes the behavior of the specific luminescence center. And the after glow is often induced by some traps from crystal defects. High density has a good stopping power and can shorten the radiation length of scintillator which can decrease the volume of scintillator and detection facility as well as the cost. The light emitted by the scintillator should have a good spectral match to photodetectors in order to ensure the high detection efficiency. However, the ideal scintillator does not exist, so each modality must compromise, selecting the most suitable combination of properties from available scintillators.

The conventional medical scintillators such as NaI (Tl) and BGO are not well satisfied with the requirements in some aspects. For example, the NaI (Tl) has a low density and a comparatively long decay time and BGO has a relatively low light yield. Opportunities for improving the scintillators used for medical imaging systems still exist in some aspects as they are somewhat far from the theoretical limits.

More and more scientists are devoting their effort to the research in medical scintillators which have become a focus in recent years. New excellent scintillators are ceaselessly proposed. More and more reports about medical scintillator appeared in recent international conferences on inorganic scintillator. The first international conference on medical scintillator was held in 2000, Russia. The development of medical scintillator will impulse the enhancement in medical diagnosis.

## 1 Scintillator requirements for medical imaging

In the field of medical imaging, the energy of the photons detected is between 15 and 600 keV. The detection system must be very efficient because there is a limit of radiation dose absorbed by patients as safety concerns. The improvement of signal-to-noise ratio cannot be obtained by increasing the ray source intensity but achieved by increasing the sensitivity of detector. In the range of 15—511 keV, photon energies used for medical imaging have the attenuation length of 2—10 cm in tissue. When the photons come into the human body, their interactions result in the Compton scattering due to the low effective atomic number of tissue and most photons undergo continually Compton scattering until they leave the body. The 10%—15%  $\gamma$  photons without the scattering can form the accurate image and other photons scattered come into being the background. In order to improve the quality of imaging, the reducing of Compton scatter photons is necessary.

Now the development of scintillators used in planar X-ray imaging, e.g. Gd<sub>2</sub>O<sub>2</sub>S:Tb and LaOBr:Tb, and used in X-ray CT, e.g. (Y,Gd)<sub>2</sub>O<sub>3</sub>:Eu,Pr and Gd<sub>2</sub>O<sub>2</sub>S:Pr,Ce,F, is relatively mature. The attention is mainly focused on the

scintillator used in SPECT and PET imaging systems.

SPECT is based on detecting individual photons emitted randomly by radioactive drug (such as  $^{99m}\text{Tc}$  with the emission of 140 keV photons) which is introduced into the body either by injection or inhalation. In practice, the only material used in SPECT is NaI:Tl. The scintillator requirements for SPECT are, in order of decreasing importance, (1) high luminous efficiency; (2) high density ( $>3.5 \text{ g/cm}^3$ ); (3) low cost; (4) good match of emission spectrum to PMT (300—500 nm); and (5) short decay time ( $<1 \mu\text{s}$ )<sup>[1]</sup>.

Similar to SPECT, the patient for PET imaging is injected with a biologically active, radioactively labeled drug that localizes at targeted sites in the body. The difference is that the drug (commonly used e.g.  $^{11}\text{C}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{18}\text{F}$ ) is positron-emission radionuclide. The emitted positron combines with a nearby electron leading to an annihilation which creates the back-to-back emission of two 511 keV  $\gamma$  photons. The scintillator requirements for PET are, in order of decreasing important, (1) short attenuation length ( $<1.2 \text{ cm}$ ) ; (2) short decay time ( $<300 \text{ ns}$ ); (3) low cost; (4) high luminous efficiency ( $>8000 \text{ photons/MeV}$ ); and (5) good match of emission spectrum to PMT (300—500 nm)<sup>[1]</sup>. In practice, the most common material used in PET is  $\text{Bi}_4\text{Ge}_3\text{O}_{12}$  (BGO).

**2 New medical scintillator**

It took one century for  $\text{CaWO}_4$  polycrystal powder to be used as scintillator<sup>[3]</sup>. Thallium doped sodium iodide, NaI (Tl), introduced by Hofstadter in 1948<sup>[4]</sup> was the most popular scintillator used in the detectors for the following tens of years because of its high light yield. However, NaI (Tl) has a low density ( $3.76 \text{ g/cm}^3$ ) and a long radiation length, which confines the energy resolution and reduces the imaging quality. This is the reason why Weber et al.<sup>[5]</sup> proposed a new heavy scintillator BGO ( $7.13 \text{ g/cm}^3$ ) in 1973, which has been extensively studied as a promising scintillator. Nowadays, the BGO has occupied more than

50% PET market<sup>[6]</sup>. But BGO has a low light yield (20%—25% of NaI (Tl)) and a long decay time ( $\sim 300 \text{ ns}$ ), which is not benefit for the improvement of time resolution. New scintillators used in PET and  $\gamma$  cameras ought to have high atomic number (high Z value), high light yield and fast decay time compared to BGO. Actually, the emissions of  $\text{Ce}^{3+}$  has fast decay time from several to several tens ns. If we could find a high density matrix and  $\text{Ce}^{3+}$  could be doped into the matrix easily, our aim for research new medical scintillators would be achieved. In fact, the recent development for medical scintillators is based on this idea. At the same time, many problems such as the crystal growth technique and the improvement for energy resolution should be solved. Nowadays, the extensively studied  $\text{Ce}^{3+}$  doped materials with high density have  $\text{Lu}_2\text{SiO}_5\text{:Ce}$ (LSO:Ce),  $\text{Gd}_2\text{SiO}_5\text{:Ce}$  (GSO:Ce) and  $\text{LuAlO}_3\text{:Ce}$ (LuAP:Ce). Some properties of the scintillators are listed in table 2.

( i ) LuAP: Ce. In 1973, Weber introduced  $\text{YAlO}_3\text{:Ce}$  (YAP:Ce)<sup>[7]</sup> and some groups researched its scintillate properties<sup>[8–11]</sup>. YAP:Ce has a fast luminescence but a low density. In order to improve its stopping power, the heaviest element lutetium in lanthanide group was used to substitute for yttrium, LuAP:Ce in some laboratory<sup>[12–18]</sup>. At room temperature, the LuAP:Ce has an emission band peaked at 355 nm which is excited by VUV (fig. 1). The decay time is 11 and 28 ns, respectively. The light yield is 20% higher than BGO. The density of LuAP:Ce is  $8.34 \text{ g/cm}^3$  which is heavier than that of YAP:Ce. So the LuAP:Ce is a promising scintillator. However, no reliable growth production processes have been obtained yet. Using the Czochralski method, only very small crystal samples could be obtained<sup>[14,19]</sup>. The LuAP:Ce samples of  $5\times 5\times 50 \text{ mm}^3$  were prepared by Petrosyan et al. using the Bridgeman method<sup>[20]</sup>. The different methods and different preparing conditions can result in the great diversities

Table 2 Properties of scintillators

Scintillator	Light yield/ photons/MeV	Density /g · cm <sup>-3</sup>	Decay /ns	Wave- length /nm	Radiation length /cm	Z <sub>eff</sub>	Refractive index	Energy resolu- tion <sup>137</sup> Cs(%)	Hygro- scopic
NaI(Tl)	38000	3.7	230	415	2.59	51	1.85	7.0	Strong
CsI(Tl)	60000	4.5	1000	560	1.85	54	1.80	9.0	Slight
BGO	8000	7.13	300	480	1.12	74	2.15	9.5	No
LSO:Ce	25000	7.35	11/36	420	1.14	66	1.82	12.0	No
GSO:Ce	8000	6.7	56/600	440	1.38	59	1.85	7.8	No
YSO:Ce	10000	4.54	37/82	420				9.0	No
YAP:Ce	16000	5.37	28	360	2.24	34	1.93	11.0	No
LuAP:Ce	9600	8.34	11/28	355					No
Lu <sub>0.3</sub> Y <sub>0.7</sub> AP:Ce	14000	6.19	25	360		53			No
Lu <sub>0.3</sub> Gd <sub>0.7</sub> AP:Ce	10800	7.93		360		63			
Gd <sub>2</sub> O <sub>2</sub> S:Tb	70000	7.3		545		64	2.1		
Y <sub>2</sub> O <sub>2</sub> S:Tb	60000	4.9		545					

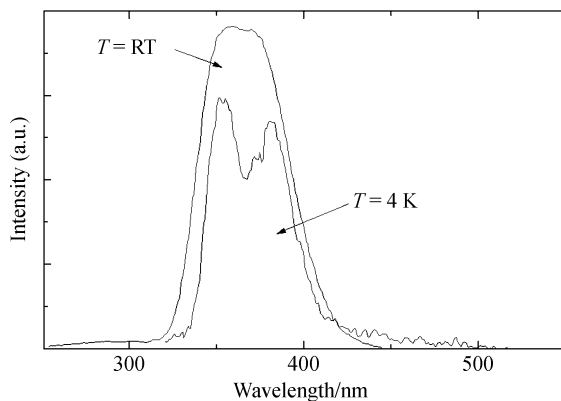


Fig. 1. Emission spectra of LuAP:Ce at RT and low temperature excited by 152 nm.

in scintillate properties<sup>[19, 21]</sup>. Other lutetium phases, especially the garnet phase, can arise in the samples. The growth of LuAP:Ce is a hard task because it is very difficult to stabilize the lutetium orthoaluminate phase. A possible way to overcome the difficulty is to grow mixed orthoaluminate crystals, especially using yttrium and gadolinium, namely  $\text{Lu}_x(\text{RE}^{3+})_{1-x}\text{AP:Ce}(\text{RE}^{3+}=\text{Y}^{3+} \text{ or } \text{Gd}^{3+})$ <sup>[22, 23]</sup>. In addition, present research shows that the light yield of  $\text{Lu}_x(\text{RE}^{3+})_{1-x}\text{AP:Ce}$  is higher than that of LuAP:Ce, which is one of the reasons of developing  $\text{Lu}_x(\text{RE}^{3+})_{1-x}\text{AP:Ce}$ . Large crystals with good quality were grown for  $\text{Lu}_x(\text{Y}^{3+})_{1-x}\text{AP:Ce}$  with  $x=0.1, 0.2$  and  $0.3$ . There is no garnet phase in crystals.  $\text{Lu}_x(\text{Gd}^{3+})_{1-x}\text{AP:Ce}$  crystals were obtained with a range between  $0.6$  and  $0.7$ <sup>[24–26]</sup>. The light yield of  $\text{Lu}_x(\text{RE}^{3+})_{1-x}\text{AP:Ce}$  is about 40%–75% higher than that of BGO. For the application of PET,  $\text{Lu}_{0.3}\text{Y}_{0.7}\text{AP:Ce}$  is a promising material.  $\text{Lu}_x(\text{Gd}^{3+})_{1-x}\text{AP:Ce}$  has higher  $Z_{\text{eff}}$  and density but lower light yield than  $\text{Lu}_x(\text{Y}^{3+})_{1-x}\text{AP:Ce}$ . In the application of PET and SPECT, high density material has a short radiation length which can decrease the thickness of scintillators and increase the spatial resolution. The LuAP:Ce is the heaviest material among all studied scintillators at present. The final aim is to find an optimal growth method and increase the light yield. The  $\text{Lu}_x(\text{RE}^{3+})_{1-x}\text{AP:Ce}$  should be a transient material from the YAP:Ce to LuAP:Ce.

There are many kinds of traps in the REAP:Ce crystals which can be reflected by thermoluminescence (TL). It is not very clear about the origin of these traps but their existence can seriously affect their scintillation properties especially for the stability of light yield and scintillation decay time. The traps in the crystals can influence the stability of light yield and the scintillation decay time<sup>[28]</sup>. The reports about TL of LuAP:Ce are very abundant<sup>[27, 29–32]</sup>. Experimental results show that the TL of LuAP:Ce peaks at 360, 500, 600, 730 K corresponding to the trap depths ranging from 0.7 to 1.9 eV<sup>[28]</sup> and the TL below the room temperature appear at 183 and 266 K<sup>[30]</sup>. The TL of  $\text{Lu}_x\text{Y}^{3+}_{1-x}\text{AP:Ce}$  appears at 50 °C, 100 °C —

130 °C, 175 °C and 255 °C<sup>[28]</sup>, and its spectral component belongs to the transition  $5d \rightarrow 4f$  of  $\text{Ce}^{3+}$  (3.45 eV). Because there is no thermal stimulated electric-conduction signal to be detected, the trapped electrons can directly enter the 5d state of  $\text{Ce}^{3+}$  without going into the conduction band. An ideal trap related to oxygen vacancies was proposed and validated by the annealing experiment. Mares reported that color centers such as  $\text{O}^-$  and  $\text{F}^+$  can be induced by  $\gamma$ -ray irradiation (3000 Gy dose)<sup>[33]</sup>. The temperature dependence, instability of light yield and the change of decay time are closely related to the defects, so it is important to enhance the defects research which will contribute to the improvement of the scintillation properties.

(ii)  $\text{RE}_2\text{SiO}_5:\text{Ce}$ . During the development of new medical scintillators, silicate doped  $\text{Ce}^{3+}$  is another focus. Such materials include YSO:Ce, GSO:Ce, LSO:Ce and LGSO:Ce. The YSO:Ce has been commercially used as phosphor<sup>[34, 35]</sup>. The GSO:Ce single crystal was introduced by Takagi et al. in 1983<sup>[36]</sup>. The GSO:Ce has faster decay time than BGO and higher density and intrinsic energy resolution than the NaI:Tl<sup>[37–39]</sup>. The LSO:Ce was first proposed by Melcher et al. in 1992<sup>[40]</sup> and got a prompt development due to its high light yield, high density and fast decay time and it becomes a very promising scintillator for PET<sup>[41–49]</sup>. The LGSO:Ce ( $\text{Lu}_{1-x}\text{Gd}_x\text{SiO}_5:\text{Ce}$ ) was firstly reported by Yamamoto<sup>[50]</sup> in 1998 and its properties were described by some groups<sup>[46, 51]</sup>. The LGSO is a new and very promising scintillator due to its lower background radiation in comparison to the LSO. But the present development stage of the LGSO shows that the LGSO has about 20% lower light output and worse energy resolution than LSO.

The emission spectrum of RESO:Ce exhibits the presence of two sets of emissions ascribed to the luminescence centers Ce1 and Ce2 respectively as shown in figs. 2 and 3. At 11 K, there are two emission bands peaked at 430 nm of Ce1 centers and at 480 nm of Ce2 centers under the excitation of 345 and 378 nm, respectively. At 296 K, the intensity decreases slightly for Ce1. However, there is a remarkable decrease for Ce2 and the emission wavelength shifts towards the short wavelength about 460 nm. Therefore, the Ce1 emission is dominant at room temperature. Shi et al. reported the emission of GSO:Ce excited by vacuum ultraviolet (VUV)<sup>[52]</sup>. It was discovered that under the excitation of 189 nm, no luminescence quenching occurred until the temperature up to 453 K and another two new band peaks, which come from Ce2 centers or some defects, appeared at 581 and 652 nm. At low temperature (6 K) a narrow excitation band peaked at 6.5 eV and a wide band peaked at 22.5 eV were observed, the former is related to the transition from the valence band to the conduction band; the latter is the excitation of a mixture state with 5p, 5d core level located in  $\text{Gd}^{3+}$ . The emission spectra of LSO:Ce excited by 188 nm

at room temperature and low temperature are shown in fig. 4. At 11 K, the excitation peaks of LSO:Ce lie in 356 nm (Ce1) and 376 nm (Ce2) and the emission peaks lie in 393, 427 nm (Ce1) and 460 nm (Ce2)<sup>[39]</sup>. Above the room temperature the emission spectrum ranges from 410 to 440 nm but it varies with the samples as well as the excitation spectrum<sup>[43]</sup> which may be caused by defects and traps from the immature growth technique.

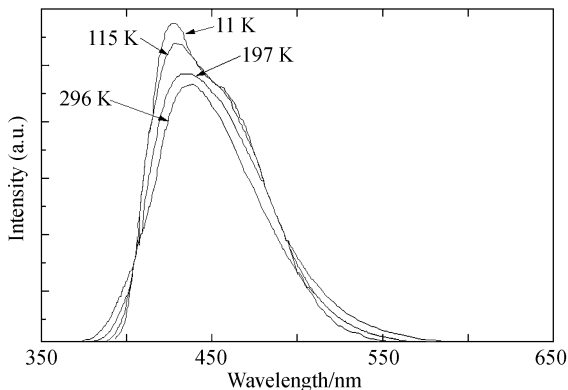


Fig. 2. Emission spectra of GSO:Ce at different temperatures excited by 345 nm (Ce1).

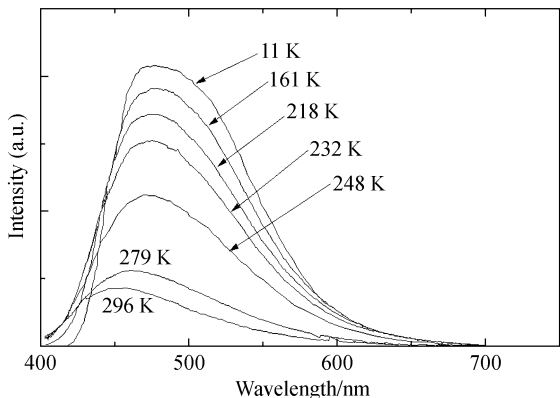


Fig. 3. Emission spectra of GSO:Ce at different temperatures excited by 378 nm (Ce2).

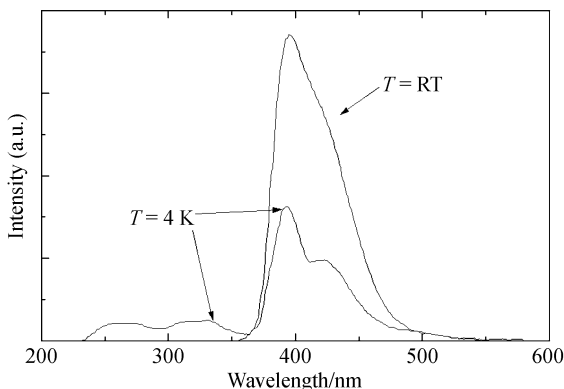


Fig. 4. Emission spectra of LSO:Ce at RT and low temperature excited by 188 nm.

In PET application, LSO:Ce is the best substitute for BGO. But it is difficult to develop LSO due to the high cost of Lu<sub>2</sub>O<sub>3</sub>. In addition, the stability of light output is rather poor for different samples, and even poor for the different parts in the same single crystal<sup>[45]</sup>. The basic opinion for the instability of LSO luminescence lies in the existence of two different centers, Ce1 and Ce2<sup>[43,47]</sup>. At room temperature the luminescence from two kinds of Ce centers induces the reduction in energy resolution of 10% with 662 keV radiation. There are two sets of Ce centers in GSO:Ce, but at room temperature the luminescence quenching occurs on Ce2<sup>[43]</sup>. So the stability of GSO:Ce luminescence is better than LSO:Ce, so is the energy resolution. Therefore, GSO:Ce is a good material to substitute for BGO in the PET system.

### 3 Light yield nonproportionality and the intrinsic energy resolution of scintillator

An ideal scintillator would produce a number of scintillation photons that is proportional to the energy deposited in the crystal. But the ideal behavior has not been observed and all scintillators are characterized by some degree of light yield nonproportionality<sup>[53]</sup>. In 1956, it was first discovered that NaI (Tl) scintillation photon production was not strictly governed by Poisson statistics and there was apparently some additional broadening which was intrinsic energy resolution<sup>[54]</sup>. At the same time, the NaI (Tl) scintillation yield nonproportionality was observed<sup>[54]</sup>. The two phenomena mentioned above had been studied independently until Iredale<sup>[56]</sup> and Zerby<sup>[57]</sup> reported in 1961 that the NaI(Tl) light yield nonproportionality significantly contributes to intrinsic energy resolution. Many reports about the scintillation yield nonproportionality and the energy resolution have been published in recent years<sup>[58–62]</sup>. All scintillators except for YAP:Ce<sup>[15]</sup> are characterized by the nonproportionality response with incident energy. For example, the LSO shows a clear sublinear increase and the light yield increases hardly when the incident energy is above 800 keV<sup>[48]</sup>. Recent results show the nonproportionality response with energy directly related to the crystal structure but not to the dopant<sup>[15]</sup>. So GSO, YSO and LSO have a similar nonproportionality response. The LuAP:Ce should also have a similar response to YAP:Ce due to the same reason. But the experimental results show a significant diversity probably caused by defects from the immature crystal growth technique<sup>[62]</sup>.

The energy resolution,  $\Delta E/E$ , where  $\Delta E$  is the full wave at half maximum (FWHM) of the full energy peak and  $E$  is the energy at the maximum measured with a scintillator coupled to a photomultiplier, can be written as<sup>[58]</sup>

$$(\Delta E/E)^2 = (\delta_{SC})^2 + (\Delta N/N)^2,$$

where  $\delta_{SC}$  is the intrinsic resolution and  $\Delta N/N$  is the photoelectron statistical contribution.

The intrinsic resolution is connected with many ef-

fects such as inhomogeneities in the crystal which causes local variations of the light yield, non-uniform reflectivity of the crystal surface, as well as the nonproportional response of the scintillator<sup>[58]</sup>. The photoelectron statistical contribution can be calculated as follows:

$$\Delta N/N = 2.36 \times (1 + \varepsilon)^{1/2} / N^{1/2},$$

where  $N$  is the number of photoelectrons and the variance of the electron multiplier gain. The typical value of  $\varepsilon$  for XP2020Q PMT is 0.1.

In the case of APD (avalanche photodiodes), the energy resolution  $\Delta E/E$  can be derived as

$$(\Delta E/E)^2 = (\delta_{SC})^2 + (\Delta N/N_{e-h})^2 + (\Delta_{noise}/N_{e-h})^2,$$

where  $\Delta N/N_{e-h}$  and  $\Delta_{noise}/N_{e-h}$  are the e-h pair statistical contribution and the noise contribution, respectively.

For using high light yield scintillator such as CsI (Tl), NaI (Tl) and LSO, the energy resolution is mainly determined by the intrinsic energy resolution of the crystals. The improvement of the intrinsic energy resolution of scintillator is the key element to enhance the energy resolution of the detector and imaging quality. The photons with fixed energy are used in PET or SPECT, but most photons with energy loss due to Compton scatter may cause some Compton scatter peaks at the low energy side of full energy peak. Therefore, the energy resolution is important for PET and SPECT.

A theoretical calculation method, which deals with the intrinsic energy resolution, induced by the energy non-proportional response of scintillators was developed by Valentine<sup>[61]</sup>. This theory can calculate the electron response and  $\gamma$  photon response, respectively and the results accord well with those of the experiments.

#### 4 Summary and prospect

Medical scintillators must have high light yield, short radiation length, fast decay time, high density, good intrinsic energy resolution and good spectral match to the PMT response and low cost. Traditional scintillators such as NaI (Tl) and BGO cannot meet all of the requirements, many researchers have carried out some searches for new scintillators. To develop new scintillators, it is an efficient way to imitate known scintillators and design future new scintillators aiming at some special requirements from the similar structure, close elements, ions radius, common chemical properties.

Luminescent efficiency is the most important factor related to the dose absorbed by patients. Normally, oxides have a higher luminescent efficiency compared with fluorides, so they will be paid more attention in our future research.

The advantage of selecting rare earth (RE) ions as a matrix is that it is easier for other RE ions to be doped into the matrix because of the close radius and common valence. Materials with high atomic number such as Lu can be used to synthesize the high density matrix, although the cost would be rather high.

The fast decay time with the order of nanosecond can be obtained from the transition from the  $5d \rightarrow 4f$  of RE.  $Ce^{3+}$  is the most widely used element because of its  $5d \rightarrow 4f$  allowed transition. Secondly, luminescence of  $Pr^{3+}$  is selected. The  $5d$  energy level of  $Pr^{3+}$  is the next higher than that of  $Ce^{3+}$ , thus they are sometimes used as dopants to create luminescence with the decay time of nanosecond order. In order to get a high effective luminescence of  $Ce^{3+}$ , the energy gap of the host must be higher than the energy difference of  $5d$  and  $4f$  of  $Ce^{3+}$ .

In current applications of CT, PET and SPECT, the scintillators are all single crystal forms. In fact, ceramic form is also chosen and being studied. Ceramic materials often show more uniformity than single crystals because ceramic materials reflect the average properties of many very small scintillator crystals. Another advantage of ceramic is the easy preparation compared with the growth of single crystal. It is noted that the ceramic materials must be transparent for the PET and SPECT application.

From the above discussion,  $Ce^{3+}$  doped scintillators such as LuAP, GSO and LuSO have become the research focus in the development of new medical scintillators. Although their light yield is still lower than that of NaI(Tl), their fast decay time and high densities show that they are promising scintillators for PET and SPECT. But many problems have not been solved technically, so the related researches are still under way.

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