Scintillation Crystals for PET*

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In PET, inorganic scintillator crystals are used to record γ-rays produced by the annihilation of positrons emitted by injected tracers. The ultimate performance of the camera is strongly tied to both the physical and scintillation properties of the crystals. For this reason, researchers have investigated virtually all known scintillator crystals for possible use in PET. Despite this massive research effort, only a few different scintillators have been found that have a suitable combination of characteristics, and only 2 (thallium-doped sodium iodide and bismuth germanate) have found widespread use. A recently developed scintillator crystal, cerium-doped lutetium oxyorthosilicate, appears to surpass all previously used materials in most respects and promises to be the basis for the next generation of PET cameras.

Key Words: PET; scintillation crystals; lutetium oxyorthosilicate

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A scintillator is a material with the ability to absorb ionizing radiation, such as x- or γ-rays, and to convert a fraction of the absorbed energy into visible or ultraviolet photons. The conversion process typically takes place on a time scale of nanoseconds to microseconds, thus producing a brief pulse of photons corresponding to each γ- or x-ray that interacts with the scintillator material. The light pulse, the intensity of which is usually proportional to the energy deposited in the scintillator, is sensed by a photodetector and converted into an electrical signal.

Scintillators may be liquid or solid, organic or inorganic, and crystalline or noncrystalline. Organic liquid and plastic scintillators often are used for detection of β particles and fast neutrons. For the detection of x- and γ-rays, such as the 511 keV γ-rays used in PET, inorganic single-crystal scintillators are used, because of their generally higher density and atomic number, which lead to better detection efficiency.

A typical scintillator is a transparent single crystal in which valence and conduction bands are separated by a band gap of 5 eV or more. In a perfect crystal, free of defects or impurities, there would be no electronic energy levels in this gap. However, most scintillators are doped with an activator ion that provides energy levels in the otherwise forbidden band gap. After absorption of γ-ray energy by the bulk crystal, a fraction of the energy localizes on the activator ions. Relaxation of the activator ions results in the emission of scintillation photons, typically around 4 eV, corresponding to visible blue light.

In the early years of PET, detectors were made of single crystals of thallium-doped sodium iodide (NaI[Tl]), individually coupled to photomultiplier tubes (PMTs). With the discovery of bismuth germanate ($Bi_4Ge_3O_{12}$ or BGO), most detector designers converted to this material because of its much greater efficiency for detecting γ-rays. A block detector became the most widely used design, in which a BGO block is segmented into as many as 64 elements and coupled to 4 PMTs (*1*). Other scintillators have included barium fluoride (BaF_2 [*2*]), yttrium aluminate ($YAlO_3$[Ce] or YAP) (*3*), and cerium-doped gadolinium oxyorthosilicate (Gd_2SiO_5[Ce] or GSO) (*4*). In recent years, a promising new material, cerium-doped lutetium oxyorthosilicate (Lu_2SiO_5[Ce] or LSO) (*5*), has emerged and is likely to be used widely in future generation PET scanners.

THE SCINTILLATION PROCESS

The process whereby a scintillator converts the energy deposited in it by a γ-ray into a pulse of visible (or ultraviolet [UV]) photons includes 3 main steps. According to Lempicki et al. (*6*), the overall efficiency (η) of the conversion process may be characterized as the product of 3 factors:

$$\eta = \beta SQ,$$

where β is the conversion efficiency of the γ-ray energy to electron hole pairs, S is the transfer efficiency of the energy held by the electron-hole pairs to the activator ions or other luminescence centers, and Q is the quantum efficiency of the luminescence centers themselves.

Robbins (*7*) demonstrated that β can be calculated from the fundamental physical properties of the scintillator crystal, including the electronic band gap, the high-frequency and static dielectric constants, and the optic longitudinal photon energy. For many scintillator materials, these parameters are well known; consequently, the conversion efficiency can be calculated with reasonable confidence. For some newer scintillators, however, some of the parameters are not known precisely, thus resulting in a significant uncertainty in the calculated conversion efficiency.

Despite the obvious importance of S, no model exists to calculate it reliably. In fact, perhaps the primary challenge of current scintillator research is to develop an accurate model of this crucial process.

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Q is measured by directly exciting the centers with UV light, the energy of which matches the excitation energy of the center. In this way, the electron-hole creation step and the energy transfer step are bypassed, and the efficiency of the luminescence center itself can be observed directly.

CHARACTERISTICS OF THE IDEAL SCINTILLATOR

The ideal scintillator would have a combination of several physical and scintillation properties (Table 1). A high detection efficiency for the γ-rays of interest requires both high atomic number for a large photoelectron cross section and high density for a large Compton-scattering cross section. These are the 2 main interactions through which 511 keV γ-rays interact with the scintillator crystals. For good coincidence timing and high count-rate capability, a short decay constant is required. In other words, the pulse of scintillation photons must be as brief as possible. A high light output allows a large number of crystal elements to be coupled to a single photodetector, and good energy resolution allows a clear identification of full energy events. The transmission of the scintillation light pulses into the photodetector is best when the refractive index of the scintillator material is similar to that of the entrance window and coupling material, usually near 1.5. In some materials, color centers may be easily produced by ionizing radiation, thus impeding the transmission of the scintillation light through the scintillator itself. Therefore, a resistance to this effect, known as radiation hardness, is desirable. Some scintillators are hygroscopic, i.e., they readily absorb water from the atmosphere and therefore require special packaging to hermetically seal them. Nonhygroscopic materials have an advantage, in that simpler packaging may be used. Mechanical ruggedness is desirable, because it makes fabrication of small crystals easier. Because a PET scanner may use 5,000–10,000 cc of scintillator crystals, the growth of large volumes of crystals at a reasonable cost must be feasible.

TABLE 1
Properties of the Ideal Scintillation Crystal for PET

Crystal property	Purpose
High density	High γ-ray detection efficiency
High atomic number	High γ-ray detection efficiency
Short decay time	Good coincidence timing
High light output	Allows large number of crystal elements per photodetector
Good energy resolution	Clear identification of full energy events
Emission wavelength near 400 nm	Good match to photomultiplier tube response
Transparent at emission wavelength	Allows light to travel unimpeded to photomultiplier tube
Index of refraction near 1.5	Good transmission of light from crystal to photomultiplier tube
Radiation hard	Stable crystal performance
Nonhygroscopic	Simplifies packaging
Rugged	Allows fabrication of smaller crystal elements
Economic growth process	Reasonable cost

TABLE 2
Physical Properties of Some Common Scintillator Crystals

Crystal	Density (g/cm³)	Effective atomic number	Hygroscopic	Rugged
$CdWO_4$	7.90	64	No	No (cleaves easily)
$Lu_2SiO_5(Ce)$ (LSO)	7.40	65	No	Yes
$Bi_4Ge_3O_{12}$ (BGO)	7.13	75	No	Yes
$Gd_2SiO_5(Ce)$ (GSO)	6.71	59	No	No (cleaves easily)
BaF_2	4.88	53	No	Yes
CsF	4.64	53	Very	No
CsI(Na)	4.51	54	Yes	Yes
CsI(Tl)	4.51	54	Slightly	Yes
NaI(Tl)	3.67	51	Yes	No
$CaF_2(Eu)$	3.18	17	No	No

PROPERTIES OF COMMERCIAL SCINTILLATORS

Because the ideal scintillator does not actually exist, one must look at the characteristics of materials that do exist and choose the one best suited to the application. Table 2 shows the physical properties of some commonly available scintillator materials, listed in order of decreasing density. Both BGO and LSO have excellent physical properties. They have high density and atomic number that result in efficient detection of γ-rays and are also rugged and nonhygroscopic, which allows relatively simple detector fabrication. Cadmium tungstate and GSO are also good candidates, except that both cleave easily, which makes detector fabrication more difficult.

Table 3 shows the scintillation and optical properties of some common scintillators, listed in order of increasing decay constant. BaF_2 has the shortest decay constant by far: 0.8 ns. Unfortunately, the emission is weak and located in the far UV at 220 nm, which requires PMTs with more expensive quartz windows. It also has a long secondary component of 600 ns. Cesium fluoride (CsF) has a very short decay constant of 4 ns, but its intensity is so weak that this scintillator is seldom used. LSO has the best combination of a short decay constant, 40 ns, and high emission intensity. In addition, it has no secondary decay component.

SCINTILLATORS USED IN PET

NaI(Tl) was discovered in 1948 by Hofstadter (*8*). It quickly became the scintillator of choice for radiation detection because of its high light output, i.e., efficient conversion of deposited γ-ray energy to scintillation photons. The large light pulses are easily processed by conventional pulse-shaping electronics. The main disadvantage of NaI(Tl) is its low detection efficiency for γ-rays above 200 keV, as a result of low density and moderately low atomic

TABLE 3
Scintillation and Optical Properties of Some Common Scintillator Crystals

Crystal	Primary decay constant (ns)	Secondary decay constant (ns)	Relative emission intensity	Emission wavelength (nm)	Index of refraction
BaF_2	0.8	600	12	220 and 310	1.49
CsF	4		5	390	1.48
$Lu_2SiO_5(Ce)$ (LSO)	40		75	420	1.82
$Gd_2SiO_5(Ce)$ (GSO)	60	600	30	430	1.85
NaI(Tl)	230	~10,000	100	410	1.85
$Bi_4Ge_3O_{12}$ (BGO)	300		15	480	2.15
CsI(Na)	630		75	420	1.84
$CaF_2(Eu)$	900		40	435	1.44
CsI(Tl)	1000		45	565	1.80
$CdWO_4$	5000	~20,000	20	480	2.20

number. At the energies typically used in SPECT (140 keV), the detection efficiency of NaI(Tl) is satisfactory, and it is used almost exclusively in that application. However, for higher energy applications, such as PET (511 keV), NaI(Tl) has been replaced, for the most part, by materials with higher density and atomic number. An additional disadvantage of NaI(Tl) is that it is highly hygroscopic. As a result, a great deal of effort has gone into the development of hermetic packaging to protect the material from moisture in the atmosphere.

BGO emerged in the early 1970s, with initial studies reported by Weber and Monchamp (*9*). Although the light output of BGO is only about 15% of that of NaI(Tl), its dramatically higher detection efficiency, as a result of density almost twice that of NaI as well as a much higher atomic number, has made it a very popular choice for the detection of radiation above a few hundred keV. PET is the major ongoing application of BGO crystals today, despite the fact that their relatively long decay constant of 300 ns limits coincidence timing resolution.

Scintillators with extremely short decay constants offer the possibility of time-of-flight PET, in which opposing detectors measure the difference in the arrival times of a pair of γ rays. In this way, the location of the positron event can be localized along the line connecting the 2 detectors. Two possibilities that appear in Table 3 are CsF and BaF_2. CsF has very low light output and is very hygroscopic and, consequently, has seen little use despite its short decay constant of 4 ns. BaF_2 has an even faster decay of less than 1 ns, greater light output, and is nonhygroscopic. Therefore, in the early 1980s it was used in several PET scanners (*10*). However, because of its relatively low density and atomic number, it eventually gave way to BGO.

One way to increase the spatial resolution of a tomograph is to couple multiple scintillator crystals with different decay constants to a single photodetector. Pulse-shape discrimination is used to identify the crystal element of interaction. GSO has been used in conjunction with BGO in this way for a high-resolution tomograph (*4,11*). A tomographic design using GSO exclusively has also been reported (*12*). Fabrication of GSO detectors requires great care, because the crystals cleave easily. Thus, special techniques are needed to avoid cracking the crystal elements during cutting.

LSO offers the best combination of properties for PET of any scintillator known today (*13*). It has high density and high atomic number for good γ-ray detection efficiency, a short decay constant for good coincidence timing, and high light output that allows the use of many small elements per PMT. In addition, it is mechanically rugged and nonhygroscopic, thus allowing relatively simple fabrication of detectors. LSO has a low level of natural radioactivity as a result of the presence of ^{176}Lu, but the counting rate from this isotope is a small fraction of the typical counting rates from the injected tracers; thus, it is not a significant problem for PET. LSO has been used in a high-resolution brain tomograph (*14*), high-resolution animal tomographs (*15*), combined PET/MRI detectors (*16*), and combined PET/SPECT cameras (*17*). Large-scale commercial production of LSO has been realized during 1999 (*18*), and widespread use of this scintillator is expected in the future.

PROPERTIES OF SCINTILLATORS FOR PET

Despite the investigation of virtually every known scintillator for possible use in PET, only 2 have seen widespread use so far, NaI(Tl) and BGO. A third, LSO(Ce), is expected to see widespread use in the future. In this section, the properties of these 3 important scintillators are compared in more detail.

One of the most important properties of a scintillator for PET is γ-ray detection efficiency. Because of the desire to shorten scan times and maintain low tracer activity, the crystals must detect as many of the γ-rays emitted as possible. This is the primary reason for the popularity of BGO. γ-rays with energy of 511 keV interact with solid matter primarily through 2 phenomena, the photoelectric effect and the Compton effect. In the photoelectric effect, the γ-ray is absorbed by an atom that ejects an electron (photoelectron) and also produces either characteristic x-rays or Auger electrons. The end result is that the full energy of the γ-ray is absorbed. In Compton scattering, the γ-ray loses a fraction of its energy to an electron through scattering. The energy of the electron is likely to be absorbed

in the crystal, whereas the scattered γ-ray may or may not be absorbed. The distribution of energy between the electron and the γ-ray is determined by the scattering angle.

The detection efficiency of a detector may be characterized by the fraction of incident γ-rays that are partially or fully absorbed by it. For a detector of thickness x, exposed to a mono-energetic beam of γ-rays, the initial γ-ray intensity, I_o, is attenuated according to:

$$I(E) = I_o(E) \exp(-\mu x),$$

where I is the intensity of γ-rays passing through the detector without interacting at all, and μ is the linear attenuation coefficient. The γ-rays that do interact in the detector by depositing either their full or partial energy are given by:

$$A = 1 - \exp(-\mu x).$$

Thus, it is clear that the fraction of incident γ-rays that are partially or fully absorbed is determined by the linear attenuation coefficient (for an idealized geometry). Figure 1 compares the linear attenuation coefficients for NaI, BGO, and LSO. From these data, the advantages of BGO and LSO over NaI are clear. At 511 keV, $\mu = 0.96$ cm^{-1} for BGO and $\mu = 0.87$ for LSO, whereas for NaI, μ is only 0.35 cm^{-1}. Consequently, to achieve similar efficiency, NaI detectors must be more than twice as thick compared with BGO and LSO detectors.

In most PET scanners, the emission of the scintillation crystals is converted to electrical signals by PMTs. To produce the largest signal, the scintillation emission should be as intense as possible, and the wavelength of the emission should match the wavelength of maximum photomultiplier sensitivity. Because bi-alkali photomultipliers with glass entrance windows, the most commonly used type, have a maximum sensitivity near 400 nm, it is advantageous for the scintillator to have its emission maximum near this wavelength. Both NaI and LSO have intense emissions that peak near that wavelength, whereas BGO has a much weaker and longer wavelength emission (480 nm) (Fig. 2). The intensity of the scintillation emission strongly affects the number of crystal elements that can be coupled to a single PMT or, stated another way, the ratio of scintillation elements to electronic channels. With BGO, block detectors today use up to 16 crystal elements per PMT, whereas LSO detectors use up to 144 crystal elements per PMT. Thus, LSO makes possible significant cost savings as a result of the reduced number of photomultipliers.

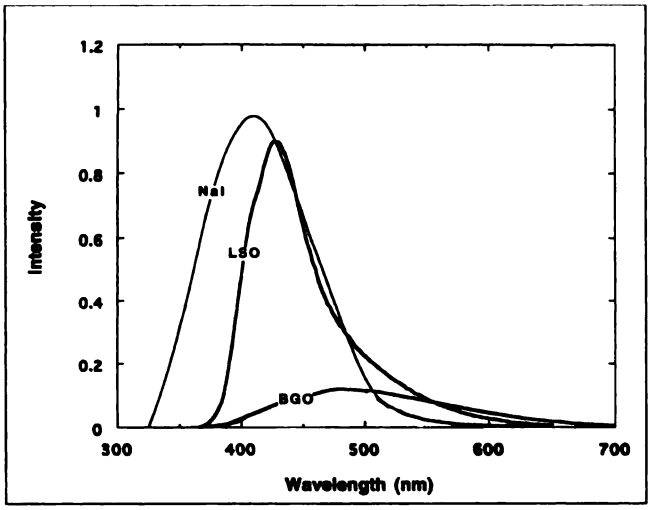

FIGURE 2. Scintillation emission spectra of NaI(Tl), BGO, and LSO(Ce), under γ-ray excitation.

In PET, the decay constant of the scintillation emission is very important, because singles count rates are typically very high, and coincidence resolving time should be as small as possible to reject unwanted random events. A short scintillation decay constant is a benefit in both instances. Figure 3 compares the scintillation decay of NaI(Tl), BGO, and LSO. BGO has the longest decay, 300 ns. The primary decay constant of NaI(Tl) is somewhat shorter, 230 ns, but

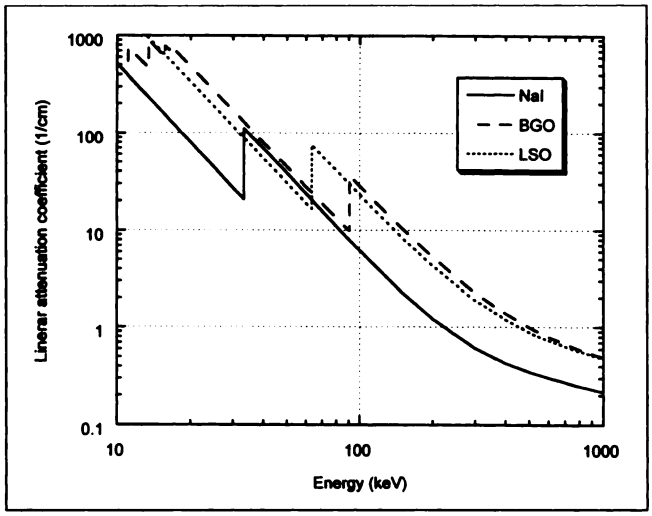

FIGURE 1. Total linear attenuation coefficients of NaI, BGO, and LSO.

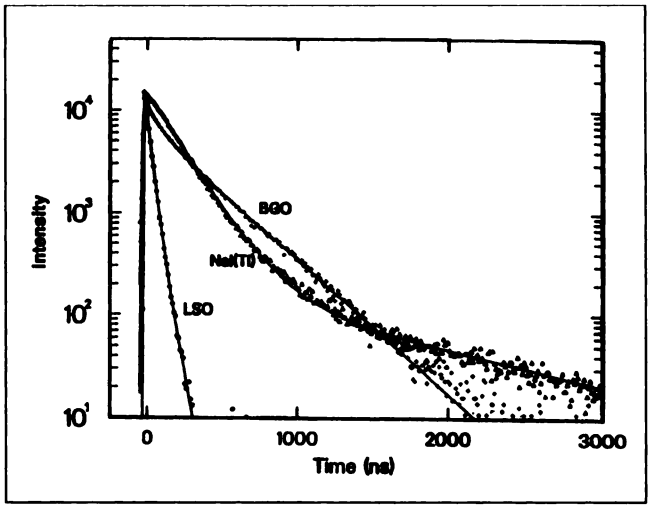

FIGURE 3. Decay of the scintillation emission of NaI(Tl), BGO, and LSO(Ce), after excitation by γ-rays.

an additional secondary decay of several microseconds is also present. The decay constant of LSO is several times shorter, 40 ns, and no secondary component is present.

Because the quality of a PET image is strongly dependent on the coincidence resolving time of the detectors, a figure of merit that has been used for PET scintillators is the number of photons emitted per nanosecond. In this way, both the overall intensity of the emission as well as the duration of the pulse are accounted for in 1 parameter. Figure 4 compares the photons emitted per nanosecond for NaI, BGO, and LSO. LSO has a large advantage over BGO in this respect, because LSO's emission is both intense and fast, whereas BGO's emission is much weaker and slower. LSO is also better than NaI in this respect, because although NaI's emission is stronger than LSO, the decay constant is more than 5 times longer.

FUTURE RESEARCH DIRECTIONS

Researchers continue to actively investigate potential new scintillator materials, because a tomograph's performance depends so strongly on the characteristics of the detectors. Most of the effort has focused on high-density and high-atomic-number materials as well as materials with potentially very short decay constants. Surveys of large numbers of candidates (*19*) have failed to identify any practical scintillators for PET. Recent research, therefore, has shifted toward computational efforts to model potential materials (*20,21*). Hopefully, an improved understanding of the underlying physics of scintillation processes will lead to the ability to predict the characteristics of new scintillator materials without actually synthesizing them.

In addition to searching for new scintillator materials, investigators have combined known scintillators in novel ways, such as phoswich detectors, which combine 2 dissimilar scintillators in the same detector. For instance, layers of LSO and NaI(Tl) have been combined to make a camera that provides both PET and SPECT capability (*18*). The LSO layer is used for PET imaging and the NaI(Tl) layer is used for SPECT imaging. Similarly, researchers have constructed prototype detectors of LSO and YSO for the same purpose (*22*).

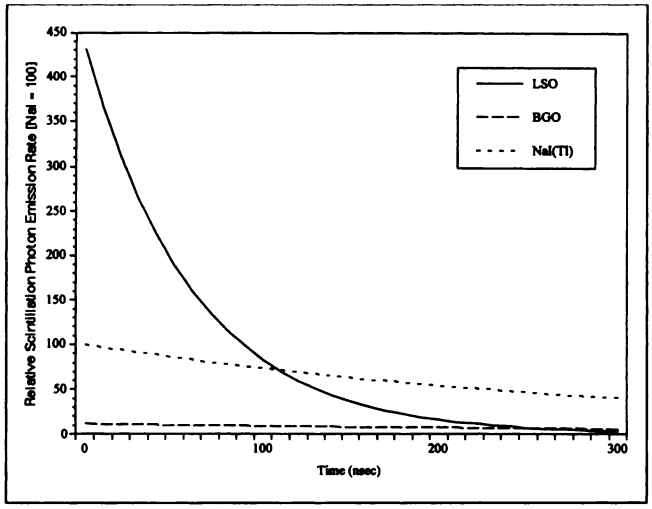

FIGURE 4. Comparison of rate of scintillation emission (photons emitted per nanosecond after excitation) for NaI(Tl), BGO, and LSO(Ce).

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