Lutetium Yttrium Orthosilicate Single Crystal Scintillator Detector

7-26-2005

Bruce Chai

Yangyang Ji
Crystal Photonics, Inc.

Find similar works at: http://stars.library.ucf.edu/patents

University of Central Florida Libraries http://library.ucf.edu

Recommended Citation


This Patent is brought to you for free and open access by the Technology Transfer at STARS. It has been accepted for inclusion in UCF Patents by an authorized administrator of STARS. For more information, please contact lee.dotson@ucf.edu.
A single crystal having the general composition, $\text{Ce}_{2x}(\text{Lu}_1-y\text{Y}_y)_{2-2x}\text{SiO}_5$ where $x$=approximately 0.0001 to approximately 0.05 and $y$=approximately 0.0001 to approximately 0.9999; preferably where $x$ ranges from approximately 0.0001 to approximately 0.001 and $y$ ranges from approximately 0.3 to approximately 0.8. The crystal is useful as a scintillation detector responsive to gamma ray or similar high energy radiation. The crystal as scintillation detector has wide application for the use in the fields of physics, chemistry, medicine, geology and cosmology because of its enhanced scintillation response to gamma rays, x-rays, cosmic rays and similar high energy particle radiation.


Primary Examiner—Constantine Hannaher
Assistant Examiner—Shun Lee
(74) Attorney, Agent, or Firm—Brian S. Steinberger; Law Offices of Brian S. Steinberger, P.A.

ABSTRACT

A single crystal having the general composition, $\text{Ce}_{2x}(\text{Lu}_1-y\text{Y}_y)_{2-2x}\text{SiO}_5$ where $x$=approximately 0.0001 to approximately 0.05 and $y$=approximately 0.0001 to approximately 0.9999; preferably where $x$ ranges from approximately 0.0001 to approximately 0.001 and $y$ ranges from approximately 0.3 to approximately 0.8. The crystal is useful as a scintillation detector responsive to gamma ray or similar high energy radiation. The crystal as scintillation detector has wide application for the use in the fields of physics, chemistry, medicine, geology and cosmology because of its enhanced scintillation response to gamma rays, x-rays, cosmic rays and similar high energy particle radiation.

14 Claims, 5 Drawing Sheets
<table>
<thead>
<tr>
<th>Patent Number</th>
<th>Date</th>
<th>Inventor(s)</th>
<th>Class Code(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5,003,181</td>
<td>3/1991</td>
<td>Morotti</td>
<td>250/484.4</td>
</tr>
<tr>
<td>5,025,151</td>
<td>6/1991</td>
<td>Melcher</td>
<td>250/269.6</td>
</tr>
<tr>
<td>5,164,041</td>
<td>11/1992</td>
<td>Berkstresser et al.</td>
<td>117/19</td>
</tr>
<tr>
<td>5,500,147</td>
<td>3/1996</td>
<td>Fitzpatrick</td>
<td>252/301.6 S</td>
</tr>
<tr>
<td>5,610,967</td>
<td>3/1997</td>
<td>Moorman et al.</td>
<td>378/154</td>
</tr>
<tr>
<td>5,644,612</td>
<td>7/1997</td>
<td>Moorman et al.</td>
<td>378/98.2</td>
</tr>
<tr>
<td>5,651,047</td>
<td>7/1997</td>
<td>Moorman et al.</td>
<td>378/98.8</td>
</tr>
<tr>
<td>5,660,627</td>
<td>8/1997</td>
<td>Manente et al.</td>
<td>117/12</td>
</tr>
<tr>
<td>5,690,731</td>
<td>11/1997</td>
<td>Kurata et al.</td>
<td>117/13</td>
</tr>
<tr>
<td>5,729,584</td>
<td>3/1998</td>
<td>Moorman et al.</td>
<td>378/146</td>
</tr>
<tr>
<td>5,751,785</td>
<td>5/1998</td>
<td>Moorman et al.</td>
<td>378/146</td>
</tr>
<tr>
<td>5,859,893</td>
<td>1/1999</td>
<td>Moorman et al.</td>
<td>378/154</td>
</tr>
<tr>
<td>6,323,489</td>
<td>11/2001</td>
<td>McClellan</td>
<td>250/361 R</td>
</tr>
</tbody>
</table>

* cited by examiner
FIG. 1A (YSO)

C\text{e}^{3+}: Y_{2}SiO_{5} \text{ABSORPTION AND EMISSION SPECTRA}

Emission Cross Section (10^{-18} \text{cm}^{2})

Absorption Cross Section (10^{-18} \text{cm}^{2})

Wavelength (nm)
FIG. 1B (LSO)

$\text{Ce}^{3+} : \text{Lu}_2 \text{SiO}_5$ \text{ABSORPTION AND EMISSION SPECTRA}

EMISSION CROSS SECTION ($10^{-18}$ cm$^2$)

ABSORPTION CROSS SECTION ($10^{-18}$ cm$^2$)
FIG. 4

EFFECTIVE Z AS A FUNCTION OF Lu
IN LYSO

% OF Lu IN LYSO

40 60 80 100

20 40 60

0

EFFECTIVE Z

70 65 60 55 50 45 40 35 30
This is a Divisional of application Ser. No. 09/506,160 filed Feb. 17, 2000, now U.S. Pat. No. 6,624,420. This invention relates to a single crystal as scintillating detector for gamma ray or similar high energy radiation which single crystal is composed of Cerium doped Lutetium Ytttrium orthosilicate (LYSO) with the general composition of Ce₃₋ₓ(Lu₁₋ₓ(Y₉₋₂ₓ)ₓ)₂₋ₓSiO₅ where x=0.0001 to 0.02 and y=0.0001 to 1.9999 and claims priority based on U.S. Provisional Application Ser. No. 60/120,500 filed Feb. 18, 1999.

BACKGROUND AND PRIOR ART

There are a number of ways to detect high energy radiation. Some of the equipment can be quite bulky, such as a cloud chamber, others may not be as sensitive or quantitative. Scintillator is a very simple and also very accurate method to detect high energy radiation such as x-rays, gamma-rays, high energy particles exceeding a few kilo-electron-volts (KeV) in energy. When high energy radiation strikes on a scintillating crystal, it creates a large number of electron-hole pairs inside the crystal. Recombination of these electron-hole pairs will release energy in the range of a few eV. This energy can be emitted directly from the recombination center as light or transferred to a light emitting ion center which then emits a specific wavelength of light. This low energy emission can then be detected by a photomultiplier tube, avalanche photo diode (APD) or other detector systems with sufficient sensitivity. The higher the light emission (or light yield), the easier for the detector design.

The first scintillating crystal is calcium tungstate (CaWO₄), which was used before the turn of this century to detect x-rays. The most significant discovery of a scintillating crystal is Thallium-activated sodium iodide NaI(Tl)) in the mid-40's. Even now, it is still the most widely used scintillating crystal. This is because large size crystals are readily available and quite inexpensive. Moreover, the light yield is the highest among all the known materials and is still the benchmark standard for all other scintillator crystals even after all these years. Even though NaI(Tl)) is widely used, it is not without problems. It is hygroscopic and very soft. Moreover, the density is too low (37 g/cm³), so it has high thermal neutron capture cross-section (49,000 barns) of the gadolinium. It will interfere with the gamma rays generated by neutron irradiation source. However, since there is no neutron source involved in the PET process, gadolinium containing GSO is not a problem.

In the late 80's, the Ce doped GSO crystal was disclosed as a scintillator material. It has a density of 7.4 g/cm³ and is non-hygrosopic. The light yield is significantly better and close to 75% that of NaI(Tl)) and the decay time is even faster (42 ns). The index of refraction is also very low (n=1.82). Moreover, since LSO has a totally different crystal structure from GSO, it is fortuitous that in LSO structure, there is not any distinct cleavage plane making the material more suitable for detector block fabrication without the serious risk of fracturing. The thermal neutron capture cross-section is very low (84 barns) as compared to GSO. Lastly, it is now possible to commercially produce high quality, large size single crystals of LSO. Compared with all the other existing known scintillator crystals, Ce doped LSO seems to have the best combination of all the needed properties for PET or other high energy gamma-ray detector application.

Unfortunately, the lutetium element of the crystal contains a trace amount of a natural long decay radioactive isotope, Lu²⁷, which will provide some background count rate that can be harmful for certain highly sensitive detector applications and the crystal has very deep trap centers. This is evidenced by the very long phosphorescence after exposure to any UV light source. The light output measurement of a large number of LSO crystals shows an anti-correlation between trap-related integrated thermoluminescence output and scintillation light output over a range of several orders of magnitude. At present time, the crystal defect is the most serious issue. One can have two crystals with identical appearance with one having 100% light yield and the other failing to scintillate. Thus far, there is no understanding of how these deep traps are formed in the first place and there is also no remedy how to reduce or eliminate them.

At present, the scintillation process has been well accepted and used in many applications. The basic mechanism is also reasonably well understood. It is generally accepted that the basic scintillation process involves three steps: (1) the absorption of the incident high energy radiation and the conversion into a large number of low energy (a few multiples of the band gap energy) electrons and hole pairs;
where \( \beta \) is the conversion efficiency, \( S \) is the transfer efficiency and \( Q \) is quantum efficiency of the radiation centers. Despite the understanding of scintillating mechanism based on the known materials, there is still lack of any good model which has the capability to predict the scintillating behavior of a specific compound. The quantum efficiency of an emission center can be predicted or independently tested optically; however, neither the total number of electron-hole pairs generated by an incident gamma ray radiation nor the transfer efficiency can be predicted or independently tested. In the end, the only way to confirm the scintillating behavior of a compound is to make and then test it.

**SUMMARY OF THE INVENTION**

The first objective of the present invention is to provide a scintillation detector for use as a high-energy radiation detector.

The second objective of this invention is to provide an improved scintillation crystal for use as a gamma ray or other high energy radiation detector.

The third objective of this invention is to provide a monocrystalline scintillation crystal of improved performance for use as a gamma ray or other similar high energy radiation detector.

In the subject invention, an improved scintillation detector assembly has been realized comprising: a cerium doped lutetium yttrium orthosilicate crystal; and, a photodetector, comprising a charge-coupled device, coupled to said crystal whereby an electrical signal is generated in response to a light pulse from said crystal when exposed to a high energy gamma ray. The crystal is preferably transparent, monoclinic and of a general composition of \( Ce_5_2(Lu_1_y Y_{1_2_1_y})_3SiO_5 \) where \( x = 0.0001 \) to 0.02 and \( y = 0.0001 \) to 1.9999, with a luminescence wavelength of approximately 420 nm and a luminescence decay of approximately 35 to 45 ns, whereby the detector utilizing said crystal as the scintillator responsive to gamma and other similar high energy radiation is particularly useful in the fields of physics, crystallography, geology and cosmology.

**BRIEF DESCRIPTION OF THE FIGURES**

FIG. 1a and 1b shows absorption and emission spectra of pure YSO and LSO.

FIG. 2 is a pictoral representation of a typical cerium doped LYSO scintillating detector.

FIG. 3 shows the LYSO scintillating light yield intensity as a function of lutetium concentration.

FIG. 4 shows the change of effective Z of LYSO as a function of lutetium concentration.

**DESCRIPTION OF REPRESENTATIVE EMBODIMENT**

For illustrative purpose, a representative embodiment of the invention is described hereinafter in the context for the detection of high energy gamma rays. It will be understood that the LYSO single crystal scintillator of the invention is not limited to the detection of gamma rays but it has the general application for the detection of other types of radiation such as x-rays, cosmic and other high energy particle rays.

In the background review, it is mentioned that Ce doped LSO has the best scintillating properties among all the known materials. But is still has a few serious problems to overcome; namely, the isotopic problem and the defect (deep trap) problem. In addition to these physical issues, LSO crystals also face two tough economic issues. First is the high melting temperature for growth. The melting point of LSO is estimated around 2200° C. It is among the highest melting temperature crystals produced commercially. Special high temperature ceramics were used to build the furnace and Iridium crucibles were used to contain the melt for growth. The growth process is quite detrimental to both insulation and the crucible. High cost of frequent replacement of the hardware pushes cost too high to be bearable for practical use. Second is the high cost of raw material of lutetium oxide. It is not a common material. Moreover, the current material purity around 99.99% is not sufficient to guarantee consistent high light yield. It is highly desirable to reduce or even replace lutetium oxide as the main ingredient in new scintillator crystals.

The embodiment of this invention is to design a new crystal which can eliminate most of the problems of LSO crystal without sacrificing the scintillating properties. Our initial motivation is to reduce the growth temperature of LSO single crystals. It is a very difficult task to maintain the operation at such high temperature for long period of time greater than 1 week). Since YSO has lower melting temperature near 2070° C., we are seeking the possibility to find an intermediate composition (or LYSO composition) which may melt at lower temperature to ease the growth process. We also want to minimize the yttrium content to retain the LSO scintillating properties.

Since there is no known published phase diagram between YSO and LSO, the phase relationship of the intermediate composition is not available. We speculate the melting and crystallization behavior of the intermediate LYSO crystal composition based on two assumptions. First, since both YSO and LSO have the same crystallographic structure and the ionic size of lutetium 3+ (0.090 nm) and lutetium 3+ (0.088 nm) arc very similar, we assume that there is a 100% miscibility between the two compositions. In other words, it is possible to make any intermediate composition LYSO crystals without worry about phase separation or formation of new compounds. Second, since YSO has lower melting temperature, based on the model of ideal solid solution, it is expected that all the intermediate compositions will have lower melting temperature similar to the classic pseudo-binary phase relations even though the exact position of the solubility and liquidus lines are not known.

In order to understand the melting and crystallization behavior, four intermediate LYSO charge compositions were prepared. The compositions were: \( Ce_{0.002}(Lu_{0.7}Y_{0.3})_{1.098}SiO_5 \) designated (70% LYSO); \( Ce_{0.002}(Lu_{0.5}Y_{0.5})_{1.098}SiO_5 \) designated (50% LYSO); \( Ce_{0.002}(Lu_{0.3}Y_{0.7})_{1.098}SiO_5 \) designated (30% LYSO); and, \( Ce_{0.002}(Lu_{0.15}Y_{0.85})_{1.098}SiO_5 \) designated as (15% LYSO). The percentage refers to the fraction of the lutetium in the crystal. A pure LSO charge was also prepared to be processed in a similar way as a reference for direct comparison. To make sure that the property comparison is meaningful, all the LYSO crystal preparation procedures are identical. The same total number of moles of chemicals in each case were used so that the finished crystals are near identical in size. To minimize the
were cut from each crystal, one from the top of the crystal width at half maximum of the 511 keV gamma ray peak. The scintillating light yield of any SiO2 scintillating light from the crystal decreases systematically form the top to the bottom of melt to crystal. The resulting crystal was grown in order to convert the maximum amount of 12.5%. The energy resolution is expressed as the full width at half maximum of the 511 keV gamma ray as the incident light. The scale used for the light output measurement is arbitrary unit. In this case, the light output for a standard NaI(Tl) scintillator is set a factor of 2.

To evaluate the scintillating properties, two crystals were grown consecutively in the same crucible and melted in a 50 kilowatt maximum power radio frequency (RF) heated high temperature furnace. The slab is placed under a Na 22 radiation source which generates the 511 keV gamma ray as the incident light. The seed is in equilibrium with the melt, the seed and iridium metal crucible will last longer. Second, simply because of the top portion of each crystal has the best scintillating value since the crystalization process has been found to be a purification process. This result has many important implications. The top portion of the crystal will have the least impurity content and thus the best performance. It is interesting to note that the pure LSO crystal has produced light yield of 93% of that of NaI(Tl). This is significantly higher than the published result of 75%. This value may approach the ultimate scintillating power for LSO.

Third, the result also shows the rapid reduction of the light yield as the growth is progressing and the greater fraction of the melt is converted to crystal. This is consistent with all the published speculation that impurities are the primary cause to create the deep trap, which gives the long phosphorescence and reduces the scintillating light yield. In the case of pure LSO, the light yield drops by a factor of 2 when 80% of the melt is converted to crystal. This is the largest drop as compared to all the other LYSO crystals. In FIG. 3, it is shown that there is a linear reduction of light yield for the bottom portion of the LYSO and LSO crystals with linear increasing of lutetium content. This is the most direct evidence to show that the impurity is coming from the Lu2O3 starting material. The phosphorescence is greatly reduced with more yttrium substitution and is nearly unnotice in high temperature processes.

In addition to the advantage directly observed from the light yield measurements, LYSO also resolves other problems associated with pure LSO. First, the growth temperature of LYSO is lower than that of pure LSO by approximately 100° C. which is very significant in high temperature processes. Since the radiation heat loss is proportional to the 4th power of temperature (or Tn) the high temperature insulation and iridium crucible will last longer. Second,
substituting yttrium will reduce proportionally the trace concentration of the naturally radioactive Lu\textsuperscript{176} isotope without sacrificing the net light yield. This will, in effect, reduce the background noise of the detector. Third, both the cost and purity of the Lu\textsubscript{2}O\textsubscript{3} starting material is a serious issue. Thus, Yttrium substitution will reduce the cost and improve the uniformity of scintillating crystals to reduce the effect of total internal reflection, the substitution of yttrium reduces the already low value of the index of refraction of LSO.

Lastly, let us examine the issue of stopping power, radiation length and effective Z. For practicality, it is desirable to use material with highest Z and shortest radiation length. Interestingly enough, we find that in the case of LYSO, the effective Z increases rapidly with small substitution of lutetium. FIG. 4 illustrates such effect. As a consequence, 30% LYSO has the same effective Z as NaI (TI) and 60% LYSO has the same effective Z as GSO. In terms of the radiation length, we can save 30% lutetium with 10% increase of radiation length, save 50% and 70% lutetium with 1.5 and 2 times that of pure LSO. In other words, the reduction is not linear to the substitution. In fact, it is favor for the substitution.

FIG. 2 illustrates the structure of the scintillation device with the crystal of the invention optically connected to the photomultiplier tube, a PIN diode, and an APD (avalanche photodetector) diode.

While the invention has been described, disclosed, illustrated and shown in various terms of certain embodiments or modifications which it has presumed in practice, the scope of the invention is not intended to be, nor should it be deemed to be, limited thereby and such other modifications or embodiments as may be suggested by the teachings herein are particularly reserved especially as they fall within the breadth and scope of the claims here appended.

We claim:

1. A composition for the detection of energy radiation comprising: a cerium doped lutetium yttrium orthosilicate mono crystal wherein the crystal includes a monocristalline structure of cerium doped lutetium yttrium orthosilicate, Ce\textsubscript{2}(Lu\textsubscript{2},Y\textsubscript{3})\textsubscript{2}SiO\textsubscript{5}, wherein x is approximately 0.00001 to approximately 0.05 and y is approximately 0.0001 to approximately 0.9999.

2. The composition of claim 1 wherein x ranges from approximately 0.0001 to approximately 0.001 and y ranges from approximately 0.3 to approximately 0.8.

3. A method of making a scintillation crystal comprising the steps of:
   (a) mixing Lu\textsubscript{2}O\textsubscript{3}, Y\textsubscript{2}O\textsubscript{3}, CeO\textsubscript{2}, SiO\textsubscript{2} together to form a mixture;
   (b) heating the mixture;
   (c) interacting the heated mixture with an LSO seed crystal; and
   (d) growing an LYSO crystal from the interaction.

4. The method of claim 3 wherein Lu\textsubscript{2}O\textsubscript{3} is substantially pure.

5. The method of claim 3 wherein Y\textsubscript{2}O\textsubscript{3} is substantially pure.

6. The method of claim 3 wherein SiO\textsubscript{2} is substantially pure.

7. The method of claim 3 wherein the heating step includes: heating the mixture to a molten state.

8. The method of claim 3 wherein the growing step includes: separating said LYSO crystal from the melt and cooling said LYSO crystal.

9. A crystal scintillator comprising a parent-single crystal of cerium-activated lutetium yttrium oxyorthosilicate having the general formula Lu\textsubscript{2-x-y}Y\textsubscript{x}Ce\textsubscript{y}SiO\textsubscript{5}, wherein 0.05 \leq x \leq 1.95 and 0.001 \leq y \leq 0.02.

10. The crystal scintillator of claim 9, wherein 0.2 \leq x \leq 1.8.

11. A scintillation detector, comprising:
   (a) a crystal scintillator comprising a parent-single crystal of cerium-activated lutetium yttrium oxyorthosilicate having the general formula Lu\textsubscript{2-x-y}Y\textsubscript{x}Ce\textsubscript{y}SiO\textsubscript{5}, wherein 0.05 \leq x \leq 1.95 and 0.001 \leq y \leq 0.02; and
   (b) a photodetector optically coupled to said crystal scintillator for detecting light from said crystal scintillator.

12. The detector of claim 11, wherein said photodetector comprises a photomultiplier tube.

13. A scintillator detector, comprising:
   (a) a crystal scintillator comprising a single crystal of cerium-activated lutetium yttrium oxyorthosilicate having the general formula Lu\textsubscript{2-x-y}Y\textsubscript{x}Ce\textsubscript{y}SiO\textsubscript{5}, wherein 0.2 \leq x \leq 1.8 and 0.001 \leq y \leq 0.02; and
   (b) a photodetector optically coupled to said crystal scintillator for detecting light from said crystal scintillator.

14. The detector of claim 13, wherein said photodetector comprises a photomultiplier tube.

* * * * *