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Growth and scintillation properties of ternary eutectic scintillators for radiation imaging applications.

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Abstract

Tl:CsI/CsCl/NaCl eutectic scintillators were grown by the Bridgman method. When sample is pulled down at 0.2 mm/min, eutectic the phases were uniformly distributed in the transverse direction and slightly aligned along the growth direction. The expected emission peak observed at 550nm was ascribed to Tl^{+} s^2-sp transition from Tl:CsI under X-ray excitation. The emission image map was observed by the confocal laser microscope. Only Tl:CsI scintillator fiber phase showed the luminescence.

1. Introduction

Radiation detectors composed of scintillators and photodetectors such CCD are widely used in X-ray imaging applications. In the X-ray imaging applications, radiation imaging sensors were composed of photodetector arrays and indirect flat panel detector (FPD) coupled with a scintillator plate such as Tl:CsI. However, grown CsI:Tl columnar reduces the spatial resolution due to the light scattering in the micro-meter size Tl:CsI fiber crystals. Light diffusion through scintillatior materials on photodetector degrades resolution of radiation imaging sensors and limits the sensitivity. Earlier, pixilated photodetector arrays have been improved to achieve a micrometer scale special resolution. However, pixel size of scintillator arrays and the light diffusion limit the spatial resolution. Up to now, phase-separated scintillator fibers (PSSFs) in eutectic materials were reported by our group for high resolution radiation imaging such GAP/α-Al_2O_3[1], HfO_2/α-Al_2O_3[2],and LiF/CaF_2/$LiBaF_3$[3]. These eutectic consist of higher refractive index scintillator fibers of several μm in diameter surrounded by lower refractive index matrix material. Such eutectic structure works as an optical waveguide. As a result, high resolution X-ray imaging can be achieved[4].

CsI:Tl is one of the most common scintillators with good scintillation properties such 55,000 photon/MeV of light yield, 1050ns of decay time and 550 nm of emission peak. This emission peak is suitable for wavelength sensitivity of silicon photodiode based photodetectors such CCD, CMOS, Si-APD etc. CsI show refractive index of 1.80@550nm and is expected as promising higher refractive index scintillator fiber phase. In this work, we investigate Tl:CsI/CsCl/NaCl eutectic fibers scintillator for radiation imaging[5]. Here, refractive index of CsI, CsCl and NaCl are 1.80, 1.64 and 1.54 @ 550nm, respectively. Therefore, CsI:Tl can be work as PSSFs and optical waveguide. Fabrication technology of the eutectic with PSSFs structure and evaluation of scintillation properties were performed.

2. Experimental produce

a) crystal growth

The starting materials were prepared using CsI, CsCl, NaCl, and TlI powders (4N). Tl and non-doped CsI/CsCl/NaCl eutectics were grown at the eutectic composition of 0.78 mol.%:TlI, 31.5mol.%:CsI, 45.2mol.%:CsCl, 22.5mol.% NaCl and 31.8mol.%:CsI, 45.7mol.%:CsCl, 22.5mol.%: NaCl, respectively. The details of CsI/CsCl/NaCl eutectic point were described in Ref.[6] . The starting powders were enclosed in a quartz tube with 3.8 mm inner diameter under high vacuum (~10-4 Pa). Crystal growth was performed by the vertical Bridgman-Stockbarger (BS) method. The pulling rates were 0.01, 0.1, 0.2, 0.5, 1 mm/min.

b) Phase identification of the eutectics

Circular samples with 1 mm thickness were obtained from the grown crystal. The cut surface was optically polished and the eutectic phase structure was observed by back scattered electron image (BEI) using Hitachi S3400N. The eutectic structures on transverse and vertical cross-section were observed. The obtained phases in the eutectics were investigated by powder X - ray diffraction (XRD) using RIGAKU RINT-2000.

c) Measurements of luminescence properties

Photoluminescence properties were investigated by a spectrometer (Edinburgh FLS920) using optical source (Edinburgh Xe 900). At the SR-163 spectrometer (ANDOR TECHNOLOGY) equipped with the CCD detector DU920P (ANDOR TECHNOLOGY) the radioluminescence spectra were measured under X-ray irradiation (40mV, 30mA, 60s). The emission image map was observed by the confocal laser microscope (ZEISS, LMS 710) at 405nm excitation.

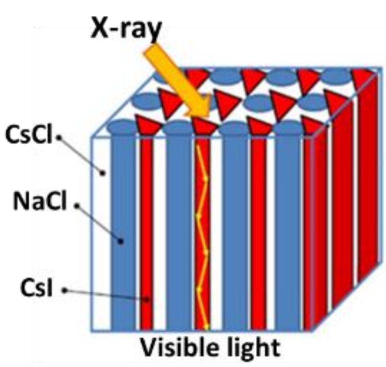

Fig. 1. Schematic view of PSSFs for light-guide system.

3. Results

The Tl doped CsI/CsCl/NaCl and non-doped CsI/CsCl/NaCl eutectics were grown by the BS method. Orange rods with 3.8mm in diameter and 30 mm in length were obtained. Samples with 3.8mm diameter and 1 mm thickness were cut from the grown crystals and mechanically polished. The BEI image of transverse cross-section of the eutectic is shown in Fig.3. The three phases seem to be uniformly dispersed. The size of each phase become smaller as the pulling rate becomes higher. The BEI of vertical cross-section is shown in Fig.4. The phases were well aligned along the growth direction in the case of 0.01, 0.1, 0.2 mm/min samples, random solidification. These phases were determined as follows White:CsI, Gray:CsCl and Black:NaCl by the EDX mapping and powder XRD pattern. CsI (cubic, Fm-3m, 225), CsCl (cubic, Pm-3m, 221) and NaCl (cubic, Fm-3m, 225) phases are observed in the powder XRD pattern. Radioluminescence spectrum measured under x-ray excitation is shown in Fig.5. In the Tl doped sample, the expected emission peak was observed at 550nm and ascribed to Tl^{+} s_2-sp transition from Tl:CsI under X-ray excitation. For the Tl-doped samples, Image of the luminescence of CsI fibers is shown in Fig.6. Tl^{+} s_2-sp emission under 405nm excitation are observed as 3~5μm diameter dots on the surface of the sample. The shape and position of the dots are well corresponding to the CsI phase in the BEI.

4. Conclusions

We have fabricated the Tl and non-dope CsI/CsCl/NaCl eutectic scintillator by directional solidification. In the eutectic each phase was uniformly dispersed in transverse direction and slightly aligned along the growth direction. Especially in Tl-doped eutectic scintillator, the expected emission peaking at 550nm was observed. This wavelength is suitable to detector with pixilated photodetector arrays like CCD, CMOS. The obtained emission image map by the confocal laser microscope evidenced emission from only the CsI phase.

In our presentation, relationship between chemical composition of starting materials, growth rate, and eutectic structure will be discussed. Details of scintillation properties, emission image map and radiation imaging using the grown eutectic plates will be also reported.

References

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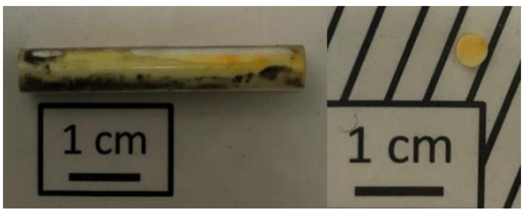

Fig. 2 Example photographs of the grown eutectic as grown. (left) and polished wafer (right).

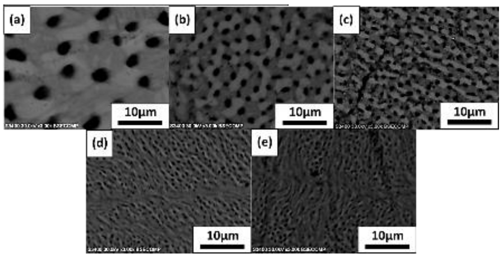

Fig. 3. BEI of CsI/CsCl/NaCl eutectic on the transverse cross-section. The growth rate: a)0.01 mm/min, b)0.1 mm/min, c)0.2 mm/min, d)0.5 mm/min, e)1 mm/min

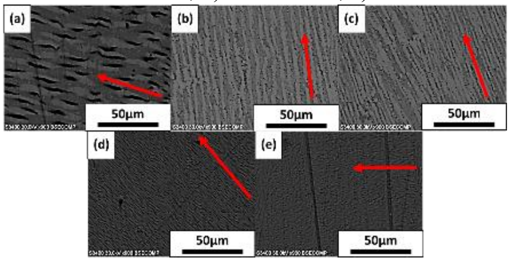

Fig. 4. BEI of CsI/CsCl/NaCl eutectic on the vertical cross-section. The growth rate: a)0.01 mm/min, b)0.1 mm/min, c)0.2 mm/min, d)0.5 mm/min, e)1 mm/min. The red arrow indicate pulling direction.

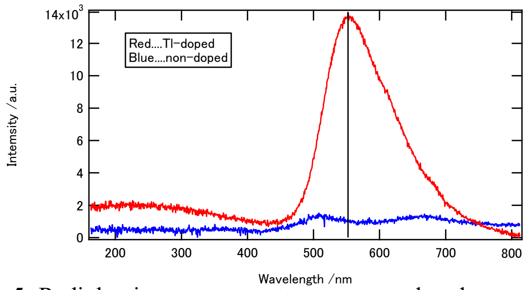

Fig. 5. Radioluminescence spectrum measured under x-ray excitation

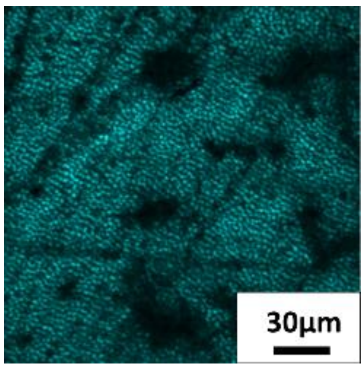

Fig. 6. Image of the luminescence of CsI fibers