

Combinatorial Synthesis of Scintillation Crystals

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Introduction

In Positron Emission Tomography (PET), scintillator crystals are employed to detect γ -rays emitted by injected tracers. The large scintillator crystals are grown via a Czochralski growth technique, which is a very time consuming process. The search for faster and more efficient scintillator crystals can be limited by the time consuming nature of the crystal growth. In this work, we will illustrate the use of combinatorial thin film synthesis process that is being used to explore new scintillator materials. The combinatorial synthesis process utilizes three individual rf magnetron sputtering sources which can be simultaneously powered to generate a wide composition space of binary or ternary systems. In this work, we have investigated cerium doped lutetium oxyorthosilicate (LSO) as it is currently the most widely used scintillator crystal in PET imaging and ideal to test the proposed approach. LSO thin films with a gradient of Cerium doping have been deposited to investigate the effects of Cerium concentration and to compare the thin film properties to those of bulk LSO crystals. Emission spectrum of the thin film material has similar characteristics compared to the bulk crystals and the integrated intensity increases with cerium concentration over the measured range. Lutetium-silicon gradients have also been grown and the measured x-ray diffraction results have been correlated to the equilibrium phase diagram. In this presentation, we will discuss the combinatorial thin film synthesis process, and will correlate the observed structural, morphological, and chemical properties of the thin films to the measured optical properties.

Procedures

The AJA International ATC 2000 R.F. magnetron sputter system employs four 2" guns spaced evenly below the substrate holder. The system uses 4" or 6" substrates. Operating parameters include R.F. or D.C. substrate bias, still or rotating substrate, heater up to 800°C, and separate substrate and target gas sources. Base pressure of the system is below 10^{-9} torr. By controlling gun power and tilt angle, substrate rotation, and system atmosphere a wide range of compositions can be generated.

Lutetium (Lu) and Silicon (Si) targets were used in an argon, oxygen atmosphere to reactively sputter Lu_2SiO_5 (LSO) onto a Si substrate. Cerium (Ce) doping was achieved by sputtering layers of cerium gradients between layers of LSO. This created a cerium concentration gradient along one axis of the deposited thin films. The concentrations in the center of the cerium gradients ranged from 0.3 at% to 5.6 at%. After sputtering, samples were annealed for 10 hours to diffuse the cerium evenly through the sample. Additionally, Lu_2O_3 to SiO_2 gradients were sputtered to validate sample composition.

Scanning electron microscope (SEM) images were taken of the deposited and annealed films using a Hitachi 4300 SEM. A Philips X'Pert x-ray machine was used to perform x-ray diffraction measurements. Excitation and emission spectrums were measured using a Hitachi F-4500 spectrophotometer. Reflectance was measured using a Filmetrics F20-UV thin film analyzer with a Hamamatsu L7893 light source. Intensity versus concentration was graphed by integrating spectrophotometer spectrums and normalizing with respect to reflectance.

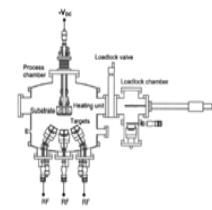


Figure 1. Diagram of sputter system



Figure 2. Picture of sputter chamber during processing. Gun targets and substrate holder are labeled.

Results

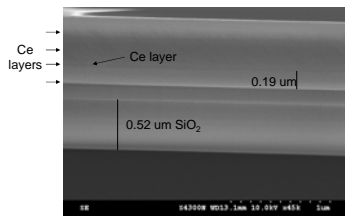


Figure 3. SEM image of as deposited structure

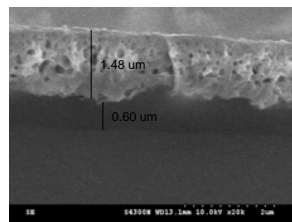


Figure 4. SEM image of annealed structure

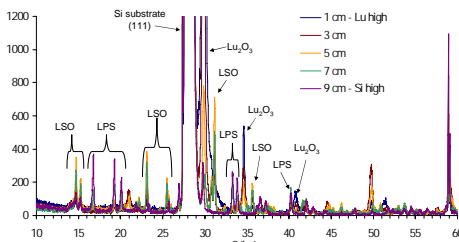


Figure 5. XRD of Lu_2O_3 to SiO_2 gradient. Measurements were taken in five spots along the 10cm long wafer. Peaks are labeled with corresponding phases.

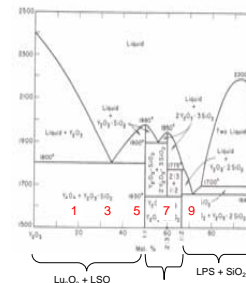


Figure 6. Phase diagram with composition along 10cm long substrate labeled in red. 5cm corresponds to the center of the wafer where target composition was LSO.

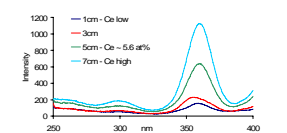


Figure 7. Excitation Spectrum. Emission at 420nm

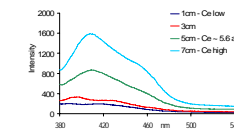


Figure 8. Emission Spectrum. Excitation at 359nm

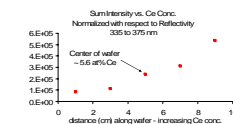


Figure 9. Integrated intensity vs. Ce concentration

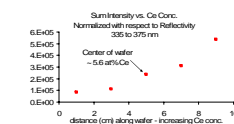


Figure 10. Integrated intensity vs. Ce concentration

Discussion

The structure of the deposited film has been confirmed with SEM imaging and the composition has been confirmed with x-ray diffraction. The Emission spectrum of the thin film material has similar characteristics compared to bulk LSO crystal. Additionally, the integrated intensity has been observed to increase with cerium concentration over the measured range.

SEM images of the annealed films show voids and other features which raise the question of substrate diffusion into the film. The possibility of substrate diffusion needs further investigation. The location of the cerium atoms in the LSO crystal structure also needs to be examined. Additionally, further work needs to be done to correlate the properties of the deposited thin films to those of bulk LSO crystal.

The use of combinatorial thin film synthesis processing to explore new scintillator materials shows promise. However, the process is still being optimized.