Structured CsI(Tl) Scintillators for X-ray Imaging Applications

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Abstract

We are developing large-area, thick, structured CsI(Tl) imaging sensors for a wide variety of X-ray imaging applications. Recently we have fabricated structured CsI(Tl) scintillators ranging from 30 μm (16 mg/cm^2) to 2000 μm (900 mg/cm^2) in thickness and up to 15 x 15 cm^2 in area. Even 2000-μm-thick film showed well-controlled columnar growth throughout the film. Material characterization confirmed that the film is crystalline in nature and that the stoichiometry is preserved. To improve the spatial resolution of thick films, post-deposition treatments were performed. The effect of these treatments on film characteristics was quantitatively evaluated by measuring signal output, modulation transfer function [MTF(f)], noise power spectrum [NPS(f)], and detective quantum efficiency [DQE(f)]. The data show that by proper film treatments, the film DQE(f) can be improved.

I. Introduction

A. Overview

Imaging X-ray and gamma-ray detectors with large area, high detection efficiency, and excellent spatial resolution over a broad X-ray energy range have applications in non-destructive testing (NDT), astronomy, medical imaging, macromolecular crystallography, and basic research. Film radiography has long been used as a principal imaging method for the above applications. Although it provides superior spatial resolution, this method is inefficient, extremely time consuming, labor intensive, and unsuitable for real-time applications. Modern and more sophisticated digital X-ray imaging systems are based on combinations of a series of scintillating phosphor screens coupled to the new state-of-the-art charge-coupled device (CCD) or amorphous silicon detector arrays (a-Si:H). This combination offers the potential for very high spatial resolution, dynamic range, and a wide range of system formats that can be easily modified to meet specific application requirements. However, if these optical detectors are used with conventional phosphor screens, the compromise between X-ray stopping power and spatial resolution limits the performance of the detector. To address this limitation, we have been developing structured CsI(Tl) scintillator films for a wide variety of applications. A partial list of these applications along with the structured scintillator requirements is given in Table 1.

B. Structured X-ray Imaging Scintillators

At RMD, we are conducting research to develop a cost-effective method of producing large-area (up to 20 x 25 cm^2), micro-structured CsI(Tl) scintillators that provide a superior combination of stopping power, spatial resolution, light output, and fast scintillation decay time compared to the currently available X-ray converter screens. The CsI(Tl) micro-columnar structures are high-density fibers of CsI(Tl) scintillator with a structure resulting from growth on a specially designed substrate [1]. This scintillating material is grown in preferential microstructured columns, which reduces the width of the point response function, resulting in superior spatial resolution compared to bulk or polycrystalline scintillators. The CsI(Tl) scintillator converts incident X-rays into visible light with very high conversion efficiency of 64,000 optical photons/MeV [2]. The micro-columnar structure (controllable to diameters as small as 5 μm) suppresses lateral spreading of the scintillation light even when the film is made very thick (150-2000 μm). This allows high spatial resolution, on the order of 15 lp/mm, along with higher detection efficiency of 97% at 30 kV X-rays as used in medical imaging (150-μm-thick film) and 50% at 400 kV X-rays as used in NDT applications (>1000-μm-thick film).

C. Structured Scintillator Fabrication

Our fabrication methods use an inherently inexpensive, modified vapor deposition system to produce large-area, very thick X-ray converter screens. Initial developments of thin CsI(Tl) screens at RMD have been reported for both low-energy [3] and high-energy NDT applications [4]. Our recent research has resulted in extending these thin film deposition techniques and process controls to fabricate substantially thick, over 2000 μm (~900 mg/cm^2), structured films. Through this research we have developed a process that allows well-controlled thick columnar growth while maintaining the crystallographic orientation and stoichiometry throughout the film. Additionally, these films exhibit better than 7% aerial thickness uniformity over 5 cm x 5 cm area, low defect densities, and essentially full bulk density packing fraction.

1) Surface Morphology

The surface morphology of 2000-μm-thick films was performed using an Environmental Scanning Electron Microscope (ESEM). Figures 1(a) and 1(b) show ESEMs of a center portion of the 2000 μm film (after approximately 1000 μm growth) and the film top. The film consists of well-defined columns separated by dense grain boundaries. Each grain is formed by a dendritic process and can be interpreted on the basis of Structure Zone Model (SZM) [5]. Between the columns, deposited films preserve voids that are free from any deposited material. Contrary to initial observations by others [6], annealing at 450°C decreased the gaps between the grain boundaries.

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Table 1
Structured CsI(Tl) Film Requirements For Various X-Ray Imaging Applications.

Application	X-ray Energy (keV)	Film Thickness (µm)	Area (cm^2)	Spatial Resolution (lp/mm)	Response Time (ms)
Crystallography	8 – 20	30 – 50	30 x 30	10	< 0.5
Mammography	20 – 30	100 – 150	20 x 25	15 – 20	< 0.1
Dental Imaging	50 – 70	70 – 120	2.5 x 3.5	7-10	NA
NDT	30 – 400	70 - >1000	>10 x 10	5 – 10	< 0.1
Astronomy	30 – 600	70 - >2000	30 x 30	4 – 5	<0.05

2) Crystallographic Orientation

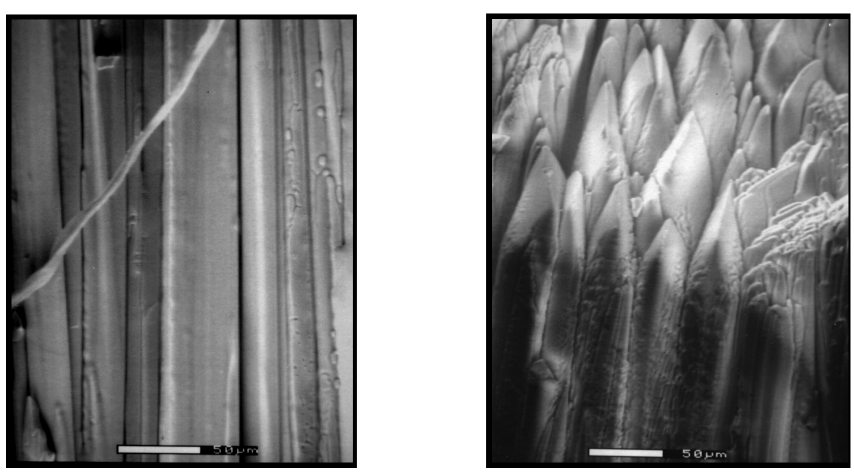
While columnar growth is necessary for preserving film spatial resolution, controlled stoichiometry and crystallographic orientation are essential for preserving the high specific light output of the CsI(Tl) scintillator and the excellent optical transmission characteristics of the resulting structure. To confirm the crystalline nature of thick films, X-ray diffraction studies were performed using a Rigaku X-ray diffractometer, model #RU300. The lattice parameters, both parallel and perpendicular to the interface, were obtained from 2θ scans with Cu K_α line. As shown in Figure 2, films exhibit an absence of any amorphous structure and grow in a preferred direction. The preferential growth for a 2000 µm film is along <310> compared to <110> for a pure CsI crystal from Harshaw chemical company. The probable cause for this is the lattice stress developed due the presence of orthorhombic TlI inside the cubic CsI. This stress is higher with thinner films which show preferred orientation of <200>.

3) Film Stoichiometry

It is well known that the dopant thallium (Tl) atoms in alkali halide scintillators work as activators which play an important role in increasing the scintillating efficiency and producing longer wavelength (540 nm) emission spectra of CsI(Tl). A homogeneous concentration of Tl is necessary to optimize the performance of the CsI(Tl) scintillator. From our experimental measurements and those performed by others [7], it has been found that a Tl concentration between 0.04 and 0.06 mole percent in the CsI matrix is necessary for optimal scintillating efficiency. Thick CsI(Tl) samples were analyzed using a Perkin Elmer atomic absorption spectrometer model #3300. The analysis of different samples revealed that the concentration of Tl in the CsI(Tl) matrix was approximately 0.04-0.07 mole percent.

II. Experimental

While the thick film morphology shows excellent columnar growth, it was found that film resolution degrades with increasing thickness [4]. For example, a 100-µm-thick CsI(Tl) film shows >60% modulation at 3 lp/mm compared to <40% modulation obtained by 140-µm-thick film at the same spatial frequency. Computer modeling of the film performance revealed that one of the major causes of this inverse relationship in columnar films is the optical scattering that takes place at the film top surface. To verify this hypothesis and to improve the resolution of thick CsI(Tl) films, post-deposition film treatments were carried out. These treatments included 1) surface finishing with absorptive or



(a) (b)

Figure 1. ESEM micrograph of 2000 µm CsI(Tl) film. Photo (a) central portion of film; (b) film surface.

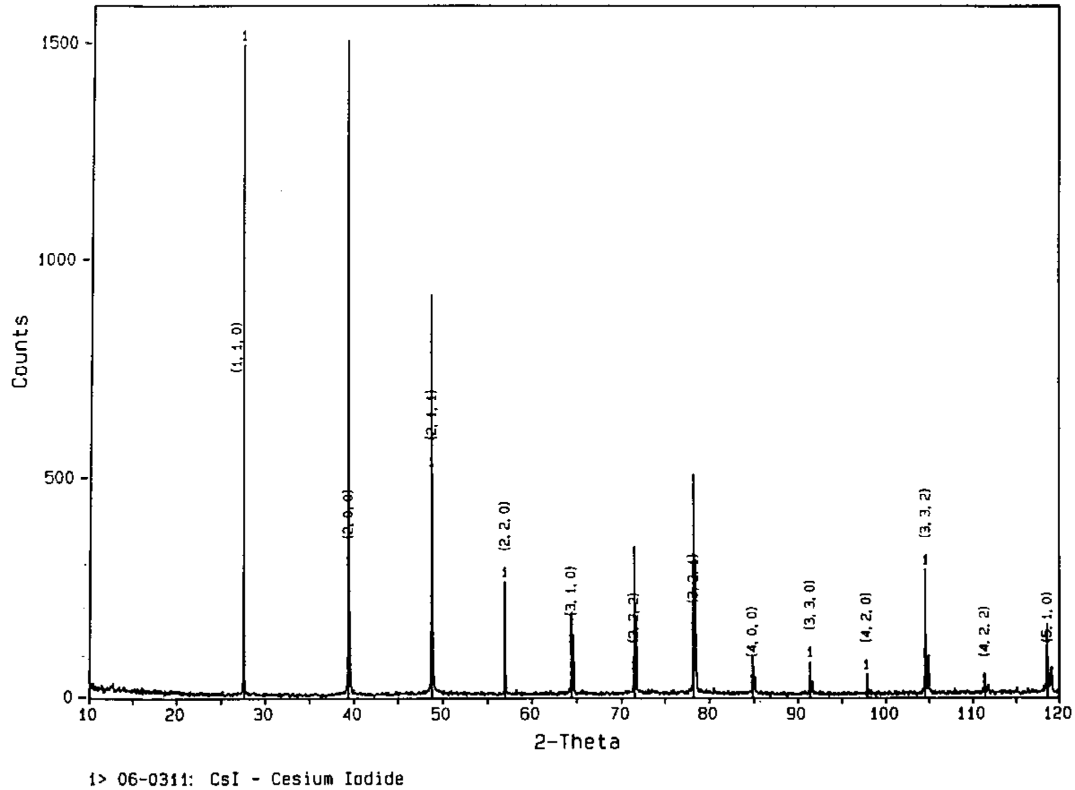


Figure 2. 2θ diffraction scans for CsI(Tl) film grown by vacuum deposition.

reflective coatings, and 2) high pressure (30,000 psi) compression to flatten the film surface as well as to force some of the coating material in between inter-columnar gaps to minimize cross-talk. A detailed characterization of changes in signal output, MTF(f), NPS(f), and overall DQE(f) before and after each processing step was carried out.

For experimental evaluation, structured CsI(Tl) scintillating screens having thicknesses in the range of 150 µm (68 mg/cm^2) to 2000 µm (900 mg/cm^2) were integrated into the Photometrics XR-200 cooled CCD camera specifically designed for direct imaging of phosphor screens. The camera consists of a 3:1 demagnification ratio fiberoptic taper directly bonded to a Thomson TH7896M scientific grade CCD resulting in an effective imaging area of 5.8 x 5.8 cm^2. With the CCD pixel size of 19 µm and 3:1 demagnification, the effective pixel size is 57 µm. Consequently the intrinsic Nyquist-limiting resolution is 8.6 lp/mm. To minimize the noise, the CCD is thermo-electrically cooled to -20°C. This results in a dark noise of 0.6 e$^-$/pixel/sec at -20°C and readout noise of 10 e$^-$ rms at 5.6 Mpixels/sec readout rate. The CCD efficiently captures 1024 x 1024 pixel high-resolution X-ray images with 14 bit digitization.

A tungsten anode X-ray generator generated the beam incident on the detector with continuously variable settings of 30 kV to 100 kV. The detector was positioned ≈ 45 cm from the X-ray source in order to illuminate the converters uniformly.

III. Results

A. Light output efficiency testing

The light conversion efficiency and uniformity of CsI(Tl) screens as deposited, and after surface coating and compression treatments, were evaluated. The scintillators were exposed to X-ray flood fields for a fixed period of 1.25 seconds and the digitized images were analyzed. These data are shown in Figure 3. As expected, screens coated with an optically absorptive coating showed 40% to 50% lower light output than screens before surface treatment, while those

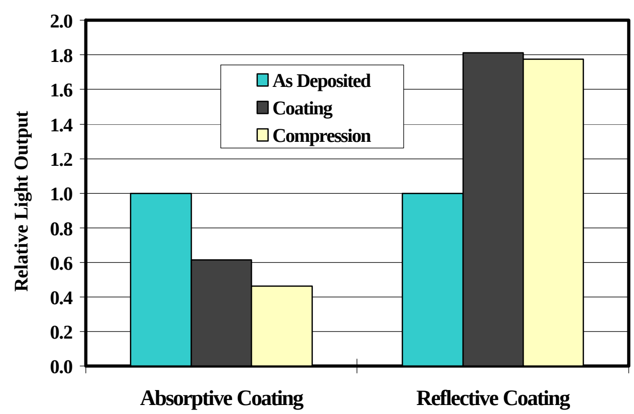


Figure 3. Light output conversion efficiency of a CsI(Tl) screen after absorptive and reflective coatings.

coated with reflective coating showed an 80% increase. The X-ray flood field non-uniformity over the 2.5 cm diameter area was measured to <3% for all the screens.

B. Modulation transfer function

The modulation transfer function, MTF(f), for spatial frequencies in the range of 0 to 12 lp/mm for various screens was calculated from the Fast Fourier Transform of the line spread function (LSF) data [8]. A 10-µm-wide tantalum slit was placed in front of the scintillator at a 0.9° angle with respect to the CCD pixel columns. This resulted in a highly sampled LSF with a sampling frequency of 0.85 µm which eliminated the aliasing artifacts in the MTF calculation. The detector was exposed to a flood field of 30 kV W X-rays. Figure 4 compares the MTF(f) for a 150-µm-thick CsI(Tl) converter before and after treatment with absorptive coating and compression.

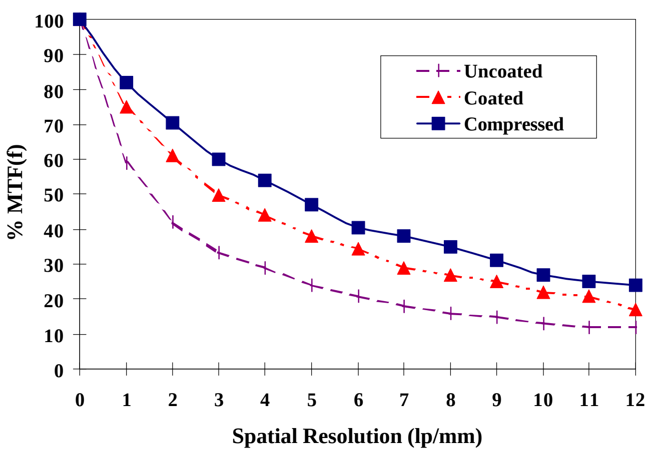


Figure 4. Modulation transfer function [MTF(f)] for a 150-µm-thick CsI(Tl) converter before and after treatment with absorptive coating and compression.

The data show dramatic improvements in screen MTF(f) after surface treatments. This is attributed to the fact that the absorptive coating effectively removes light from the film surface that is otherwise scattered by the surface roughness back into the film. High pressure compression (30 Kpsi) helps reduce scatter by surface flattening and by forcing some of the absorptive coating into the CsI(Tl) inter-columnar gaps. The effective removal of scattering substantially reduces the screen glare (or background noise) resulting in superior resolution performance. Spatial resolution of screens coated with a reflective coating did not change significantly (data not shown here).

C. Noise power

The noise power as a function of frequency, NPS(f), for spatial frequencies in the range of 0 to 12 lp/mm for various screens was evaluated by Fourier analysis of a flat field image [9]. One-pixel-wide and 256-pixel-long regions of interest (ROI) were used. A 256-point FFT was performed on the pixel values of the ROI. Several such FFTs of different ROIs were averaged. The resulting curve was further smoothed by four-point averaging. Figure 5 compares the NPS(f) for a 150- µm-thick CsI(Tl) converter before and after treatment with absorptive coating and compression.

The data show improvements in the screen NPS(f) after absorptive coating over the entire range of spatial frequencies. The NPS(f) is further improved by film compression. For films treated with reflective coating the NPS(f) remained unchanged up to the spatial frequency of 5 lp/mm and showed higher noise power after 5 lp/mm.

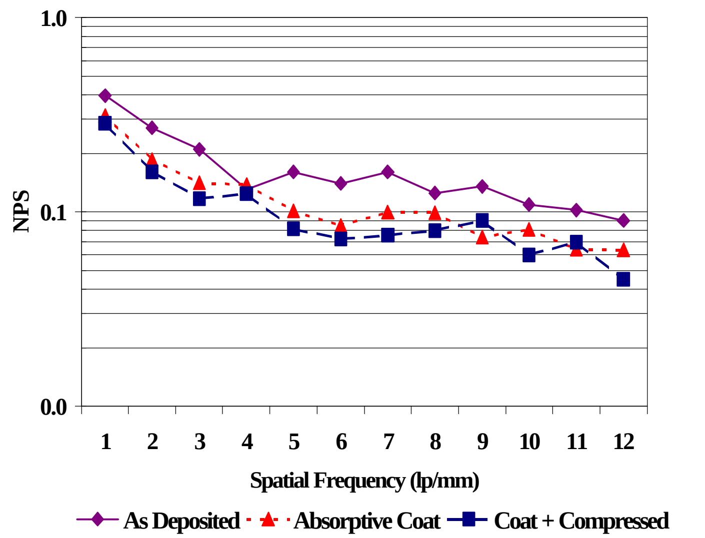


Figure 5. Noise power spectrum [NPS(f)] for a 150 µm CsI(Tl) converter before and after treatment with absorptive coating and compression.

D. Detective quantum efficiency

The detective quantum efficiency, DQE(f), for 30 kV X-rays and 150 µm CsI(Tl) screens was calculated from the measurement data described above using a formula given by Hillen et. al [10] and Roehrig et. al [11]. Accordingly the DQE(f) is given by (1) where Signal(ϕ) = Mean signal in units of ADU at X-ray fluence ϕ.

$$DQE(f,\phi) = [Signal^2(\phi)]*[MTF^2(f)]/ NPS(f)*(\phi) \qquad (1)$$

The incident X-ray fluence (ϕ) was kept constant throughout the Signal, MTF(f), and NPS(f) measurements and therefore was not entered in the calculation of relative DQE(f) of films before and after the treatment.

Figure 6 shows the effect of absorptive coating and compression on DEQ(f). Although the signal output of CsI(Tl) screens decreases with absorptive coating, the overall DQE is improved over the frequency range of 1 to 12 lp/mm. This is attributed to significant improvements in film MTF(f) (note that DQE(f) is proportional to the square of MTF(f)) and overall NPS(f). On the other hand, CsI(Tl) films treated with reflective coating and compression showed somewhat reduced DQE(f) due to lower MTF(f) values, especially at high spatial frequencies of 5 lp/mm and above. Thus, the performance of X-ray screens cannot be judged by signal strength or spatial resolution alone.

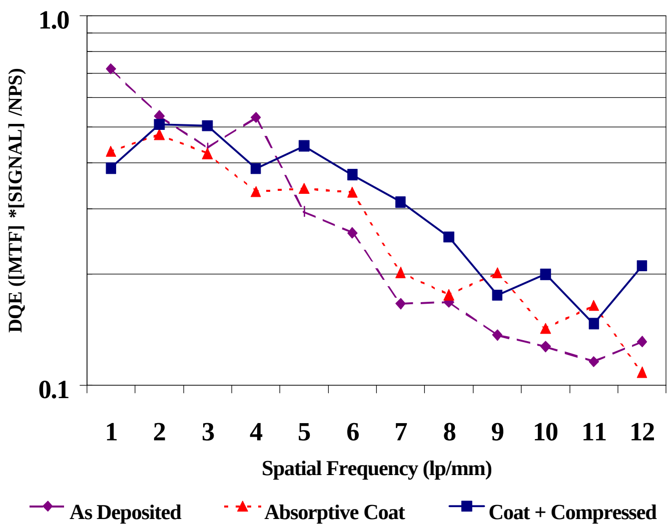


Figure 6. Detective quantum efficiency [DQE(f)] of a 150-µm-thick CsI(Tl) converter before and after treatment with absorptive coating and compression.

IV. Summary and Conclusions

We have developed structured CsI(Tl) scintillators for a wide variety of X-ray imaging applications. Using existing facilities at RMD, Inc., we are capable of producing very large area, thick scintillators on a wide variety of substrates such as commercial fiberoptics or optical sensors including CCDs or a-Si:H pixel arrays. Recently, >2000-µm-thick film structures have been successfully deposited, which is a very significant accomplishment, especially for the high-energy imaging applications. Even when such thick films are fabricated, the stoichiometry and columnar crystallographic orientation are maintained throughout the film. This allows for better scintillation efficiency and excellent optical transmission properties of thick film.

In an attempt to improve the thick film's spatial resolution, we have carried out experimental studies of the effect of post-deposition surface treatments on the film properties. Films treated with absorptive or reflective coating were further subjected to high pressure compression. Detailed characterization of changes in signal output, MTF(f), NPS(f), and overall DQE(f) before and after each processing step have been carried out. The results show that in spite of the fact that the films treated with absorptive coating produce a significantly lower signal, the overall DQE(f) is improved due to the corresponding improvements in MTF(f) and the NPS(f). Thus, by controlling the optical density or the light absorption properties of the coating, films may be tailored to suit a given application.

V. References

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