UNIVERSITY OF MANCHESTER

Development of a novel Colour X-ray Coherent Scatter Imaging system

by

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A thesis submitted in partial fulfillment for the degree of Doctor of Philosophy

in the

Faculty of Engineering and Physical Sciences
School of Materials

May 2010
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# Abbreviations

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<th>Description</th>
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<tbody>
<tr>
<td>ACRT</td>
<td>Accelerated Crucible Rotation Technique</td>
</tr>
<tr>
<td>ADD</td>
<td>Angle Dispersive Diffraction</td>
</tr>
<tr>
<td>ASIC</td>
<td>Application Specific Integrated Circuit</td>
</tr>
<tr>
<td>CAT</td>
<td>Computed Axial Tomography</td>
</tr>
<tr>
<td>CCE</td>
<td>Charge Collection Efficiency</td>
</tr>
<tr>
<td>CIE</td>
<td>Charge Induction Efficiency</td>
</tr>
<tr>
<td>CT</td>
<td>Computed Tomography</td>
</tr>
<tr>
<td>CZT</td>
<td>CdZnTe</td>
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<tr>
<td>DAQ</td>
<td>Data Acquisition</td>
</tr>
<tr>
<td>DLS</td>
<td>Diamond Light Source</td>
</tr>
<tr>
<td>EBSD</td>
<td>Electron Back Scattered Diffraction</td>
</tr>
<tr>
<td>EDD</td>
<td>Energy Dispersive Diffraction</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full Width at Half Maximum</td>
</tr>
<tr>
<td>GUI</td>
<td>Graphical User Interface</td>
</tr>
<tr>
<td>HPB</td>
<td>High Pressure Bridgman</td>
</tr>
<tr>
<td>ICDD</td>
<td>International Centre for Diffraction Data</td>
</tr>
<tr>
<td>ICSD</td>
<td>Inorganic Crystal Structure Database</td>
</tr>
<tr>
<td>KCl</td>
<td>Potassium Chloride</td>
</tr>
<tr>
<td>MVB</td>
<td>Modified Vertical Bridgman</td>
</tr>
<tr>
<td>NaCl</td>
<td>Sodium Chloride</td>
</tr>
<tr>
<td>PDF</td>
<td>Powder Diffraction File</td>
</tr>
<tr>
<td>rTEDDI</td>
<td>rapid Tomographic Energy Dispersive Diffraction Imaging</td>
</tr>
<tr>
<td>SPS</td>
<td>Spark Plasma Sintering</td>
</tr>
<tr>
<td>TEDDI</td>
<td>Tomographic Energy Dispersive Diffraction Imaging</td>
</tr>
<tr>
<td>THM</td>
<td>Traveling Heater Method</td>
</tr>
<tr>
<td>Z</td>
<td>atomic number</td>
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Abstract

The field of X-ray imaging and X-ray diffraction have been combined in a new technique called TEDDI. This diffraction imaging technique allows 3D sample images to be obtained, non-destructively, where each imaged point contains the atomic structural information associated with its diffraction pattern. The main drawback of the TEDDI technique is the long collection times needed to produce the images. In order to overcome this obstacle the rTEDDI system has been developed at the University of Manchester’s Material Science Centre. The research and development of rTEDDI has been the focus of this PhD thesis.

A proof of concept for the rTEDDI imaging technique was obtained using thin samples on station 7.6 SRS Daresbury. In this case a first generation array collimator was used in conjunction with an energy resolving Si pixelated detector. Structural information such as lattice parameters, crystal system and phase identification were obtained for metal, polymer and deer antler bone samples. The use of high Z semiconductor detector material was investigated in order to increase the potential of TEDDI for larger and more dense samples. To enable penetration of larger samples high energy X-rays needed to be utilized. In order to detect these higher energies with a good efficiency the detector media was changed from Si to CZT.

The second generation rTEDDI, using CZT as the detection media, was intended to be used under high flux/high energy synchrotron radiation conditions. Testing of the system under these conditions on station 16.3 SRS Daresbury showed an inability to produce diffraction imaging. An in depth investigation into detector and collimator array performance showed a two fold cause. The ERD2004 detector was unable to handle the high countrates experienced during high flux/high energy synchrotron radiation conditions. The MK1.2 collimator array was found to become partially transparent to X-ray energies around the absorption edge of W resulting in the swamping of the diffraction signal under high flux/high energy synchrotron radiation conditions.

A new detector ASIC design, developed by the detector division and the Rutherford Appleton Laboratory, and Data Aquisition (DAQ) system, developed by Aspect Systems, as well as a number of new collimator array designs were developed and tested. Testing of the new collimator array structures have shown positive results and the new HEXITEC detector which was designed to be able to handle high countrates, have shown an unprecedented inter pixel uniformity and energy resolution which have been attributed to the ASIC performance and the use of better quality CZT material.
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Acknowledgements

I would first of all thank my supervisor Prof. Robert J. Cernik who has served as an immense source of help and guidance during my PhD. I have appreciated all the interesting discussions we have had (sometimes they have even been on the topic of discussion!!). Without you this PhD would not have be possible nor would it have been half as much fun.

A special thanks has also got to be made to Kern Hauw Khor. I do not know how I would have got through the first year without your help.

I also wish to thank all the members of the HEXITEC collaboration. It has been truly great to be able to work with so many extremely competent and friendly researchers across different fields.

A special thank you must also be made to Matthew Wilson and Matthew Veale. You have been an amazing help and endlessly patient with what must at times have seemed to be an endless stream of questions from me. Thanks to the two of you I will also never look at a paint of strongbow the same way again!

I would also like to thank Simon Jacques, Olivier Lazzari and the rest of the UCL/Birkbeck crowd. I am grateful for all the help you have given me during, and leading up to, beamtimes. I am fairly certain I will never be witness to stranger, or more passionate, dinner discussions then I have when dining with you guys. It has been fantastic fun!

A big thank you also has to be said to my friends here in Manchester and at home. Special mention has to be made to Hans-Peter Pettersson, Karl-Fredrik Pettersson and Daniel Pedersen who is always there regardless of how long it has been. You know what you mean to me.

A special thank you also has to be made to Sarah Griffin with Family. Sparkie, you have stood by me when this work has been the hardest and the easiest. I can not put into words how much that and you mean to me.

Finally I have to thank all of my Family who has been an endless source of encouragement, comfort and support throughout my first 28-ish years. Mum and Dad, I could not have made it this far without the two of you. I am forever grateful and count myself lucky to have you as parents.

“knowledge only adds to the excitement, the mystery and the awe of a flower”

R.P.Feynman
Chapter 1

Introduction

Since the discovery of X-rays by Wilhelm Röentgen in 1896 [1] this form of electromagnetic radiation has been associated with the concept of X-ray imaging. The first X-ray image ever taken was done by Roentgen himself. The first image was of a set of weights located inside a wooden box and after this the famous image of his wife’s hand would follow (see figure 1.1 left). The form of imaging used by Roentgen is now known as radiography and is still a very important technique in medical science. Two radiographs can be seen in figure 1.1. Radiography utilizes the fact that X-rays will be absorbed in different amounts depending on the density variation, given a constant thickness, of the object they are transmitted through. By exposing a solid object to a straight through X-ray beam it is then possible to map the variation in X-ray absorption/transmission as a function of position. By interpreting the absorption/transmission variation as a

Figure 1.1: (Left) Radiograph of Bertha Roentgen’s hand (Right) Modern radiograph of a hand
variation in density of the solid object it is possible to obtain images such as the ones in figure 1.1.

X-ray transmission imaging took a massive leap forward in 1972 when Hounsfield managed to construct the first Computed Tomography (CT) scanner, also known as a Computed Axial Tomography (CAT) scanner [2]. The CT scanner records the transmission of X-rays through a sample at various angles (non-simultaneously) and uses this data to tomographically reconstruct a 2D-slice of the object by calculating absorption profiles. By reconstructing a large number of these 2D-slices a 3D image can be produced. A CT slice as well as a reconstructed 3D CT image can be seen in figure 1.2. The

![Figure 1.2: (Left) 2D CT slice of a human head [3] (Right) 3D reconstructed CT image of a foot [4].](image)

ability to use X-ray CT scanning in order to image in greater detail have improved since 1972, first with the development of X-ray micro-tomography [5, 6, 7] and subsequently X-ray nano-tomography [8].

Although the field of crystallography was well established before the discovery of X-rays it is usually attributed to have come into its modern phase when the first X-ray diffraction pattern was collected by von Laue and his colleagues [9] in 1912. What von Laue found was that sending monochromatic (single wavelength) X-rays through a crystal deflected the X-rays into specific patterns. One of these patterns, known as diffraction patterns, can be seen in figure 1.3. The deflection of the X-rays into specific patterns by crystals were correctly interpreted by Bragg and Bragg to be a consequence of the atomic structure of the crystal [10], more precisely the inter-planar spacings between the atomic planes. The ability to 'image' the crystal planes in a crystal revolutionized the way we investigate the internal structure of solids. Initially this was done by exposing the sample to monochromatic X-rays and measuring the angles at which the X-rays where deflected. Using Bragg’s law [10] the crystal lattice of the solid could then be determined. This initial setup, still greatly used and of great importance in modern
science, is known as Angle Dispersive Diffraction (ADD).

With the discovery of the semiconductor detector by Van Heerden in 1945 [12] and the subsequent improvement of this technology, mainly during the 1960’s, a second geometrical setup for diffraction was realized. This setup, known as Energy Dispersive Diffraction (EDD), was developed (for powder diffraction) independently by each other by Giessen and Gordon [13] and Buras et al [14]. EDD exposes a sample to a coloured X-ray beam (large no of wavelengths) and positions a spectroscopic detector, coupled to a collimator, at a set angle to the incoming beam and measures which wavelengths are deflected to this angle. This setup allows for faster acquisition times compared to ADD since it is not necessary to scan all angles to find the deflected X-rays, but the accuracy with which the lattice spacing can be determined is set by the energy-resolution of the detector (and collimator) instead of the ability to measure the angular difference of the deflected X-rays. Despite this technical drawback EDD has been extensively used in application where the ability to do angular scanning is limited by geometrical setup considerations, such as for high pressure diamond anvil work [15] [16], or when short collection times are needed.

Using the EDD setup and by manipulating the geometries of the X-ray beam and the collimator field of view it was realized that the diffraction from a discrete voxel within the sample could be selected out [17] allowing spatial resolution to be obtained. By step-scanning this voxel within the sample 2D and 3D sample imaging could be achieved. This form of imaging, known as TEDDI [18], does not only produce images, which can be done using both CT and radiography, but also contain for each imaged voxel the atomic structural information obtainable from a EDD diffraction pattern. In short the
technique combines the concepts of X-ray imaging and X-ray diffraction into a new, more powerful, form of imaging. TEDDI as an imaging technique has a large number of uses, some of which can be seen in figure 1.4, including (i) build up of material on reactor wall during synthesis of nanocrystalline CeO$_2$ in supercritical water environments [19]; (f) determining non-systematic change in the lattice parameter of a ferrite transformer [20] and (e) investigation of crystallite alignment and polymorphic interaction of glutamic acid [21]. Other potential uses include identifying tissue types [17], airport security and identifying strain variations in crucial engineering components.

The main drawback of the TEDDI technique is the long exposure times needed, usually 16-20 hrs using step-scanning, in order to build up a 2D image of a bulk sample. In order to overcome this problem a novel X-ray imaging system, called rTEDDI, was developed at the University of Manchester’s School of Materials [22, 23]. The rTEDDI system was realized by coupling an array collimator [24] with a pixelated, spectroscopic Si semiconductor detector [25]. This produces an array of voxels, imaging an entire 2D cross-sectional slice of the sample simultaneous, drastically reducing the collection times needed.
1.1 Thesis outline and chapter description

The work presented in this thesis was carried out from 2006-2009 at the University of Manchester, the SRS Daresbury, the detector development division at Rutherford Appleton Laboratory and at Diamond Light Source. At the start of the project a collimator array and a pixelated detector, intended as the core of the rTEDDI system, had already been developed. They had however not been incorporated into a working prototype system, nor had a proof of concept for the rTEDDI technique been obtained. The detector had not been characterized or investigated to any depth.

A few areas of interest had already been identified by the start of this project. In order to image thick samples using the rTEDDI system high energy X-rays would have to be utilized. The poor detection efficiency of Si at higher X-ray energies would therefore be an issue. It was decided that in order to try and overcome this obstacle Si would be substituted by CdZnTe (CZT) as the active detection material of the detector. A new detector Application Specific Integrated Circuit (ASIC) would also be developed with the main aims being an optimized energy resolution and a small pixel pitch while using CZT as the detector material.

The main aim of this project was therefore specified as obtaining a proof of concept for the rTEDDI technique using Si detectors; substituting Si for CZT as the active detector material and investigate any effects caused by doing so; investigate and characterize the existing detector ASIC in order to detect any weaknesses in the design with respect to imaging using the rTEDDI system and use this information to improve on the design of the new detector ASIC. Testing the rTEDDI system on high energy, high flux, synchrotron stations in order to investigate the system behavior under these conditions where also carried out during this project with the aim of improving the system for a second generation rTEDDI system.

What follows is a brief description of the chapters that follow this introduction.

- Chapter 2

Chapter 2 discusses the theory of diffraction and diffraction imaging. The chapter gives an overview of X-ray Bragg diffraction. The relationship between ADD and EDD is shown and the geometrical setup of EDD and the natural extensions TEDDI and rTEDDI leading to diffraction imaging is discussed. The chapter closes with a discussion on the use of Rietveld refinement to obtain atomic structural information from a diffraction pattern.
• Chapter 3

Chapter 3 discusses the theory of operation of semiconductor detectors, specifically CdZnTe. The chapter gives an overview of the interaction processes that occur between radiation and semiconducting material followed by a discussion of charge induction and the formation of detector signals. A discussion of different high Z semiconductor materials investigated for use as detector media is also included. The chapter closes with a discussion of the different growth techniques of CdZnTe and the effect both intrinsic and extrinsic defects have on the detector properties.

• Chapter 4

Chapter 4, the first of four results chapters, focuses on the first generation rTEDDI system leading to the proof of concept for the imaging technique. A brief discussion of the realization of the system followed by the experimental low flux, low energy synchrotron setup is given before an in-depth look and analysis of images obtained from four different sample material, Al, Cu, Nylon-6 and deer antler bone, using the technique. The chapter concludes with results obtained for the second generation rTEDDI system (using CZT as the detection media) under high flux, high energy synchrotron conditions where diffraction imaging was not possible.

• Chapter 5

Chapter 5 shows the results obtained for a TEDDI setup using a single element CZT detector. The setup was used to investigate the effects of using a high Z detector media detector when imaging using the classical TEDDI setup. Two samples, a friction stir welded Ti6246 sample and a spark sintered ceramic sample, were imaged and Rietveld refined. The concluded structural changes observed as a function of position in the samples are discussed for both samples.

• Chapter 6

Chapter 6 focuses on the development of detectors for the second generation rTEDDI system. The operation of the ERD2004 detector is discussed. Effects of substituting Si with CZT as the detector media for the ERD2004 detector is discussed with respect to response to known radiation sources; pixel and inter pixel uniformity; and bias voltage and temperature response. The effectiveness of different charge sharing correction algorithms applied to the ERD2004 ASIC read out structure was investigated and is discussed. The countrate limitations for
the ERD2004 detector with respect to both the ASIC and DAQ system was has been evaluated. The operation of the developed HEXITEC detector and initial results with both test signals and X-ray exposure is also studied. The developed data analysis and visualization software for both detectors is described in detail in Appendix A and Appendix B.

• Chapter 7

Chapter 7, which is the last of the results chapters, focuses on the development and testing of different collimator array structures. The development and testing of the initial MK-line of array collimators under low flux/low energy synchrotron conditions is followed by response under high flux/high energy synchrotron conditions evaluation of the MK1.2 array collimator. The development, manufacturing and testing under high flux/high energy synchrotron conditions of two solid collimator array designs, one using a lead glass capillary design and one using a cast lead bismuth design, is discussed. The chapter is concluded on the discussion of a third collimator array design, manufactured using chemical etching.

• Chapter 8

The project concludes with chapter 8 which gives a summary of the major findings of the project and suggestions for future works.
Chapter 2

X-ray diffraction theory

In order to understand the technical challenges that needs to be overcome in order to realize the rTEDDI system an understanding of the underlying principal and limitations of the imaging technique is needed. In this chapter diffraction and the factors limiting its ability to determine atomic structural information is discussed for different diffraction methods. This is followed by a discussion on the geometrical setup for EDD and how diffraction imaging is accomplished. The chapter is concluded by a short section on how analysis of diffraction patterns is carried using the Rietveld refinement method, due to this method of non linear least squares use to analyse raw diffraction data to extract physical parameters.

2.1 Diffraction, Bragg’s law and peak width

The discovery that X-rays where deflected into specific patterns when moving through a crystal by von Laue [9] was interpreted to be due to reflections from inter-atomic planes of atoms in the crystal by Bragg and Bragg [10]. The angle at which constructive interference occur (diffraction) is described by Bragg’s law [10] and is stated in equation 2.1

\[ n\lambda = 2dsin\theta \]  

(2.1)

where \( \lambda \) is the wavelength of the incoming X-ray, \( d \) is inter-atomic plane distance, \( \theta \) is half the angle at which diffraction occur and \( n \) is an integer number known as the order of diffraction. If a crystal is exposed to a single X-ray wavelength X-ray beam diffraction occurs at discrete angles which can be found by spatially scanning a detector. In this setup the inter-atomic plane distances of the sample is determined by the angle at which diffraction is found and is known as Angle Dispersive Diffraction (ADD). If
a crystal is exposed to a white X-ray beam (a wide band of wavelengths) any given plane of atoms will diffract discrete wavelengths which can be detected using an energy (wavelength) resolving detector. In this setup the inter-atomic plane distances of the sample is determined by the energies for which diffraction occur and is known as Energy Dispersive Diffraction (EDD) [26] [27]. When working with EDD it is often useful to re-write equation 2.1 using the expression of energy, E, found in equation 2.2

\[ E = \frac{hc}{\lambda} \]  

where h is planck’s constant and c is the speed of light. By inserting equation 2.2 into equation 2.1 the energy dispersive equation of diffraction is obtained as can be seen in equation 2.3.

\[ E_{d}sin\theta = \frac{nhc}{2} \]  

For ADD the factor limiting the resolution with which the inter-atomic plane distances can be determined is the ability to accurately determine the angle of diffraction. For EDD the resolution is mainly limited by two factors, the energy resolution of the energy resolving detector and the value of energy broadening due to beam divergence [28] [27] [29]. The peak width, \( \delta E \), due to these two effects are mathematically expressed in equation 2.4 [29]

\[ \delta E = \left[ (\delta E_{D})^2 + (\delta E_{\theta})^2 \right]^{1/2} \]  

where \( \delta E_{D} \) is the energy resolution of the detector system and \( \delta E_{\theta} \) is the energy broadening due to beam divergence. The factors effecting the energy resolution of the detector system is discussed more in detailed in the next chapter. In order to limit the beam divergence a collimator is placed in front of the detector. This results in the beam divergence being reduced to the divergence of the collimator. An expression of the energy broadening due to collimator divergence, \( \delta \theta \) is obtained by differentiating equation 2.3 and can be seen in equation 2.5 [29]. For simplicity \( C = nhc/2 \) has been used.

\[ E = \frac{C}{d_{sin}\theta} \]

\[ \delta E_{\theta} = -\frac{C\cos\theta}{d_{sin}^2\theta} \Delta \theta \]
\[ \delta E_\theta = -\frac{E \sin \theta \cos \theta}{ds \sin^2 \theta} \Delta \theta \]

\[ \delta E_\theta = -Ecot \theta \delta \theta \] (2.5)

which can subsequently be inserted into equation 2.4. It should be noted that EDD requires a sufficiently high polycrystalline nature from the sample, or sample rotation, in order to obtain all diffraction peaks of the sample material.

### 2.2 EDD geometry, TEDDI and rTEDDI

The shape and size of the volume (lozenge) being sampled in an EDD setup is set by the geometry of the X-ray beam and the field of view of the collimator at the point where the two intersect. This can be seen in expanded view in figure 2.1. By putting the sample on a xy-movement stage the lozenge can be step scanned through out the plane of the sample, allowing for a 2D map to be imaged where the size of each imaged voxel is set by the X-ray beam and collimator geometry. This form of imaging is known as TEDDI imaging [30] [31] and a 2D TEDDI map can be seen in figure 2.2. The image in figure 2.2 is of a piece of Ti that has been welded using the linear friction welding technique (for further description and details on the Ti weld sample please see Chapter 5). The imaging capability of the technique is here clearly shown as well as the

![Illustration of the experimental setup for TEDDI imaging.](image)
Figure 2.2: TEDDI image of a friction stir welded piece of Ti. Material regions with low strain (top pattern), high strain (middle pattern) and the bakelite regions (bottom pattern) are clearly visible from the image. The image was collected on station I15, Diamond Light Sources, Ltd.

The associated diffraction pattern of each imaged voxel. This allows for imaging where for each imaged voxel the associated atomic structural information contained in the EDD pattern can be obtained [19] [32] [33]. The long scanning times needed in order to create a 2D TEDDI image, usually exceeding 16-20 hours, lead to the development of the rTEDDI system at the University of Manchester’s Material Science Centre [34]. The basic concept of the rTEDDI system is illustrated in figure 2.3. By adding collimators and detectors horizontally a corresponding number of imaging voxels are created giving depth profiling in the sample. By adding collimators and detectors vertically imaging voxels are created spanning the width of the sample. When creating an array of collimators and detectors an entire plane of the sample can be imaged simultaneously [34]. Scanning the sample in the z direction, under these circumstances, allows for 3D diffraction images to be acquired with a simple one directional motion. The rTEDDI collimator-detector array offers, assuming the same detector countrate as for a classic TEDDI measurement, an increase in imaging speed compared to TEDDI imaging directly related to the number
2.3 Whole pattern fitting, Rietveld refinement and Topas

In order to extract information from the diffraction patterns obtained a non linear least square technique such as the whole pattern fitting Rietveld method is needed. Whole pattern fitting deals with the fitting of calculated diffraction patterns to measured diffraction patterns in order to determine crystal structure parameters. In 1968 Rietveld devised a structure refinement method that used peak profile intensities instead of total integrated peak intensities allowing for overlapping peaks to be resolved [35]. The Rietveld refinement method, which initially revolutionized neutron powder diffraction and later X-ray powder diffraction, uses a crystal structure model to produce a calculated diffraction pattern that is to be compared to the measured pattern. The parameters of the crystal structure model are varied in order to minimize the least squares error between the calculated and measured patterns. The term minimised is stated in equation 2.6

\[ S_i = \sum w_i (y_i(\text{obs}) - y_i(\text{calc}))^2 = \text{Minimum} \]  

(2.6)

where \( y_i(\text{obs}) \) is the measured intensity of the incremental 2θ step i, \( y_i(\text{calc}) \) is the calculated intensity of the incremental 2θ step i and \( w_i \) is the weight. The weight was originally calculated by [36]
\[ w_i^{-1} = \sigma_{ig}^2 + \sigma_{ib}^2 \quad (2.7) \]

where \( \sigma_{ig} \) is the standard deviation, usually based on counting statistics, at step i, and \( \sigma_{ib} \) is due to the background. The weight calculation stated above is now one of many schemes used \[35 \] \[37 \] \[38 \]. Parameters that can be refined include lattice parameters, atomic positions, atomic site occupancies, atomic thermal vibrational parameters, peak profiles, preferred orientation, background function, \( 2\theta \) zero correction, overall scale factor and a number of parameters accounting for instrumental conditions. Due to the non-linearity of the parameters approximate values for all parameters has to be stated as input for the first refinement cycle.

Topas \[39 \] is one of many programs developed to refine X-ray diffraction patterns using the Rietveld refinement method, and has been chosen for this project due to its high fitting stability. Topas allows for initial crystal structure parameters to be downloaded and imported from the Inorganic Crystal Structure Database (ICSD) \[40 \]. Figure 2.4 shows the rietveld refinement of a single point TEDDI measurement of a Ti6246 sample. The blue line is the measured diffraction pattern, the red line is the calculated fit and the grey line at the bottom shows the error between the two.

![Figure 2.4: Screen grab from a rietveld refinement of a Ti6246 sample done using the Topas software. The diffraction patterns are loaded in and displayed as diffraction angle vs intensity. The blue line is the measured diffraction pattern, the red line is the calculated fit and the grey line at the bottom shows the error between the two.](image)

The measured data was loaded into the Topas program on a intensity vs. \( 2\theta \)-angle format. The X-ray emission profile,
background, instrumentation parameters where specified and the crystal structure and phase parameters where loaded into the program by the use of ICSD files. Lattice parameter, relative phase relation and preferred orientation were subsequently refined. In this manner a number of physical parameters can be obtained at each voxel point and it is the choice of the experimenter to decide which to output.
Chapter 3

Detector theory

In order to understand the effects of the detector performance on the overall performance of the rTEDDI system a good understanding of how semiconductor detectors work is essential. In this chapter the interaction of ionising radiation with matter and the subsequent processes leading to, and affecting the detection of the induced signal in a detector is discussed. This is followed by a short review of the desired material properties of high atomic number (Z) semiconductors and consequentially some of the material systems that are under investigation for room temperature X-ray detection purposes at high X-ray energies. This chapter is concluded by a brief description of current growth techniques and resulting effects of defects on detector performance for the high Z semiconductor CZT.

3.1 Radiation Interactions with Matter

All direct conversion semiconductor radiation detectors work on the same basic principle. Radiation enters the active detection material where it deposits all, or most, of its energy. This deposition produces a physical response which is measured and presented in a readable form.

When radiation in the form of X-rays enter a crystal (in this case a semiconductor) it has a finite probability of interacting with the crystal atoms. As a consequence an X-ray beam trying to penetrate a piece of material will become attenuated according to equation 3.1:

\[
\frac{I}{I_0} = e^{-\mu_a \rho x}
\]  

(3.1)

where I is the resulting intensity of the X-ray beam after having traveled a distance x
through the material, $I_0$ is the incoming X-ray intensity, $\rho$ is the mass density of the material and $\mu_a$ is the attenuation coefficient of the material. X-rays can interact with matter in three principle ways that are of importance to X-ray detector physics. These are photoelectric absorption, elastic scattering and inelastic scattering. The three interaction processes are incorporated in equation 3.1 through their respective cross-sections (a measure of the probability of an event to occur) which gives rise to the value of the attenuation coefficient. The attenuation coefficient is material dependent and its expression can be seen in equation 3.2:

$$\mu_a = \sigma_{PA} + \sigma_{ES} + \sigma_{IS}$$

(3.2)

where $\sigma_{PA}$ is the photoelectric cross-section, $\sigma_{ES}$ is the elastic scattering cross-section and $\sigma_{IS}$ is the inelastic scattering cross-section. What follows is a short description of the three interaction processes.

**Photoelectric Absorption**

In photoelectric absorption [41] all the energy from a photon is transferred to an atomic electron. The electron will most likely originate from an atomic K-shell. The electron absorbing the photon energy, known as a photoelectron, is ejected from its host atom and is free to move throughout the crystal lattice. As the photoelectron does so it interacts with other bound electrons through Coulomb interactions leading to the creation of a charge cloud [42]. It is consequently the movement of this charge cloud that gives rise to the electric pulse measured in a detector. The energy obtained by the photoelectron, $E_{PE}$, is given by equation 3.3:

$$E_{PE} = E_{PH} - E_B$$

(3.3)

where $E_{PH}$ is the initial energy of the photon and $E_B$ is the binding energy of the electron shell from which the photoelectron was ejected. Although no single analytical expression exist for the photoelectric cross-section an approximation can be seen in equation 3.4 [43]:

$$\sigma_{PA} = N Z^5 \left[ \frac{m_e c^2}{E_{PH}} \right]^\frac{7}{2}$$

(3.4)

where $N$ is a constant, $Z$ is the atomic number, $E_{PH}$ is the energy of the incoming photon and $m_e c^2$ is the electron rest mass energy. It should be noted that the probability of
interaction through photoelectric absorption increases drastically with increased Z and decreases with increased photon energy. When the photoelectron is ejected the host atom will enter an exited state. Usually the atom relaxes by having a higher orbital electron drop down to the vacancy left by the photoelectron. In doing so a characteristic photon, with an energy equal to the difference between the shell states, is released. Under normal conditions the characteristic photon would be re-absorbed by the detector crystal, adding charge carriers to the charge cloud. If however the characteristic photon manages to escape the crystal without being re-absorbed, a lower number of charge carriers is registered for a specific incoming photon energy. This process leads to escape peaks in detector spectra. It should be pointed out that the characteristic photon process has a competing process in Auger electron emission.

Elastic Scattering

Both Rayleigh [44] and Thomson scattering fall under the category of elastic scattering. In elastic scattering the photon interacts with the material and is deflected with an angle, measured with respect to the incident trajectory of the photon, but without any transfer of energy from the photon to the material constituents. For Thomson scattering the photon interacts with free electrons and for Rayleigh the photon interacts with the atoms in matter. The Thomson cross section is given by [43]:

$$\sigma_{TS} = \frac{8\pi r_e^2}{3}$$ (3.5)

where $r_e$ is the classical electron radius. The Rayleigh cross section is given by [43] [45]:

$$\sigma_{RS} = \pi r_e^2 \int_{-1}^{1} |f_s(w, Z)|^2 (1 + \cos^2 \theta) d(cos\theta)$$ (3.6)

where $f_s(w, Z)$ is the atomic form factor, $w = (E/hc)\sin(\theta/2)$, $\theta$ is the scattering angle and $E$ is the photon energy. As can be seen from equation 3.5 and 3.6 the cross section for Thomson scattering is energy independent while the Rayleigh scattering cross section, being proportional to the atomic scattering form factor, is dependent on the $Z$ of the material and drops with increased energy.

Inelastic Scattering

In inelastic scattering [46], also known as Compton scattering, the photon interacts with a loosely bound electron. Part of the photon energy is transferred to the electron
and the photon is scattered at an angle, $\theta$, to the normal of the incident photon trajectory. Given that enough energy is transferred to the electron, then known as a recoil electron, it will break free from its host atom and be scattered at an angle, $\phi$, to the normal of the incident trajectory. The energy of the scattered photon, $E_{SP}$ is given by equation 3.7 [47]:

$$E_{SP} = \frac{E}{1 + \frac{E(1 - \cos \theta)}{m_e c^2}}$$

(3.7)

where $E$ is the incident photon energy, $m_e c^2$ is the rest mass energy of the electron. The energy transferred to the recoil electron is the difference in energy of the photon before and after scattering. The inelastic scattering cross-section is given by equation 3.8 [43]:

$$\sigma_{IS} = 2\pi r_e^2 \left\{ \frac{1 + \gamma}{\gamma^2} \left[ \frac{2(1 + \gamma)}{1 + 2\gamma} - \frac{1}{\gamma} \ln(1 + 2\gamma) \right] + \frac{1}{2\gamma} \ln(1 + 2\gamma) - \frac{1 + 3\gamma}{(1 + 2\gamma)^2} \right\}$$

(3.8)

where $\gamma = E/m_e c^2$. For $\gamma < 1$ ($E$ is less than approximately 500keV) equation 3.8 can be simplified as:

$$\sigma_{IS} = \sigma_{TS}(1 - 2\gamma)$$

(3.9)

The probability of Compton scattering for a specific atom depends on the number of electrons associated with that atom and will increase linearly with $Z$ [47].

Figure 3.1 shows how the attenuation coefficient, $\mu_a$, varies with photon energy for the compound semiconductor $Cd_{0.9}Zn_{0.1}Te$, as calculated by XCOM [48]. From the figure it is easy to see the relative probabilities of interaction at different energies for this material and that the total attenuation is a combination of the three interaction mechanisms. The reproducibility of a specific electrical pulse value from the detection material for each energy is a vital aspect for any spectroscopic X-ray detector. In order to enable this the same amount of energy has to be deposited into the detection material each time a specific energy photon is detected. As a consequence materials where photoelectric absorption is the strongest contributor to the total attenuation is selected on the expense of inelastic scattering. Elastic scattering is neglected in this discussion since it does not deposit any energy into the detector material, but it should be mentioned that this is the form of scattering leading to X-ray diffraction. Figure 3.2 shows the relative strength of phototelectric absorption to inelastic scattering as a function of photon energy and atomic number, $Z$, of the material causing the attenuation. Due to the energy range we are working with, marked red in figure 3.2, pair production can be neglected. As can be seen in figure 3.2 for a specific photon energy increasing the $Z$ of the absorber
Figure 3.1: Plot of calculated attenuation profile for $Cd_{0.9}Zn_{0.1}Te$ at different energies. The relative contributions to the total attenuation of the three interaction processes are clearly shown.

Figure 3.2: Cross-section for photon interactions. The boundaries where there are equal probability of photoelectric absorption to inelastic scattering and inelastic scattering to pair production are indicated [49]. The energy region of interest for material science problems that can be addressed using rTEDDI is marked in red.
will increase the likelihood of interaction taking place through photoelectric absorption on the expense of inelastic scattering. This makes higher $Z$ materials, from a purely matter-photon interaction viewpoint, a better choice of detector material. Due to the overall increase in attenuation with $Z$, as described in equation 3.4 and equation 3.8, a higher $Z$ semiconductor will also have a higher detection efficiency. Figure 3.2 shows the detection efficiency for a number of semiconductors illustrating the effect of increased detection efficiency with increasing $Z$.

![X-ray photon detection efficiency](image)

**Figure 3.3:** Plot of the detection efficiency as a function of energy for a number of different semiconductor materials. The $Z$ of the materials varies from the lowest in Si to the highest in TlBr and $HgI_2$. The detection efficiency of CdTe can be used as an approximation for CZT. The plot is used with the permission of Dr. Paul Sellin, University of Surrey

### 3.2 Leakage current and intrinsic energy resolution of semiconductor detectors

The band gap of any semiconductor is only dependent on the unit cell size of its crystalline structure [50]. For both Si and CZT the atoms are arranged in the zinc-blende structure. At absolute zero all semiconductors behave as insulator since no electrons have been thermally excited from the valence band to the conduction band. Increasing the temperature above absolute zero will result in electrons being thermally excited over the band gap, producing electron-hole pairs. If the number of thermally exited electron-hole pairs is high at the detector operation temperature the leakage current of
the detector will consequently be high. The probability of an electron-hole pair being thermally created is given by equation 3.10 [51] [52]:

\[ p(T) = C T^3 e^{-\frac{E_g(T)}{2k_bT}} \]  

(3.10)

where \( T \) is the absolute temperature, \( E_g(T) \) is the band gap energy which is in itself temperature dependent, \( k_b \) is the Boltzmann constant and \( C \) is a material constant. As can be seen in equation 3.10 the generation of leakage current, the current running through the detector under biased conditions when no X-rays are present, is strongly band gap dependent since, for a set operation temperature, an increase in bandgap results in fewer electron-hole pairs being present in the material. The band gap of the detector material is therefore an important design parameter. The thermal generation probability of electron hole pairs at different temperatures for Si, CZT and Diamond can be seen in figure 3.4. One advantage of compound semiconductors is the ability to

![Figure 3.4: The thermal generation probability of electron hole pairs at different temperatures for Si \( (E_g \approx 1.14\text{eV}, T=300\text{K}) \), CZT \( (E_g \approx 1.57\text{eV}, T=300\text{K}) \) and Diamond \( (E_g \approx 5.47\text{eV}, T=300\text{K}) \) [53].](image)

manipulate the band gap energy through band gap engineering. For CZT this is done by changing the ratio of Zn to Cd of the material. By increasing the band gap of a semiconductor the leakage current is decreased. Figure 3.5 shows the band gap energy as a function of the lattice parameter for a number of compound semiconductors. The effect of changing the Zn concentration in CZT is indicated by the vertical dashed lines.
The band gap energy as a function of lattice constant for II-VI compounds [54]. The variation in band gap energy for acCZT when varying the Cd to Zn ratio in the material is shown by the horizontal and vertical dotted lines (marked in red).

The energy it takes to create an electron-hole pair roughly corresponds to the band gap of the detector [55] and is known as the W-value, of the material. Figure 3.6 shows the relationship between band gap and W-value for a large number of semiconductors. Using the W-value the number of electron-hole pairs created, \( N \), for any specific energy...

![Figure 3.5: The band gap energy as a function of lattice constant for II-VI compounds [54]. The variation in band gap energy for acCZT when varying the Cd to Zn ratio in the material is shown by the horizontal and vertical dotted lines (marked in red).](image)

![Figure 3.6: The W-value as a function of band gap energy for a number of different semiconductors [54].](image)
deposited in the detector is given by equation 3.11

\[ N = \frac{E_{\text{dep}}}{W} \]  

(3.11)

where \( E_{\text{dep}} \) is the deposited energy and \( W \) is the W-value. Assuming the creation of electron-hole pairs follow Poisson statistics the variation in the number of electron-hole pairs created, \( \Delta E_{\text{Poisson}} \), for a specific energy can be derived from equation 3.11 and is stated in equation 3.12

\[ \Delta E_{\text{Poisson}} = \sqrt{E_{\text{dep}}W} \]  

(3.12)

The creation of all the electron-hole pairs constituting a charge cloud in semiconductors are physically related making the process differ from that described by perfect Poisson statistics. This results in a smaller \( \Delta E \) (due to a higher number of electron-hole pairs created for a specific deposited energy) than predicted in equation 3.12 [56]. The difference is described by the Fano factor [57] [58], \( F \), which is a measure of the difference between observed variance (number of observed electron hole pairs created by the deposition of a specific energy) and perfect Poisson variance (number of electron hole pairs predicted to be created for a specific energy when assuming creation according to Poisson statistics). The expression for the intrinsic energy variance for a semiconductor material is therefore given by equation 3.13

\[ \Delta E_{\text{Real}} = \sqrt{FE_{\text{dep}}W} \]  

(3.13)

This energy variance gives rise to a variation in the measured charge for the same deposited X-ray energy for repeated depositions. This energy variance manifests itself as a peak width of the measured energy peak in the detector spectra. The intrinsic Full Width at Half Maximum (FWHM) energy resolution of the spectral peak for a detector media is stated in equation 3.14

\[ E_{\text{FWHM}} = 2\sqrt{2\ln(2)}\sqrt{FE_{\text{dep}}W} \approx 2.355\sqrt{FE_{\text{dep}}W} \]  

(3.14)

Equation 3.14 assumes a Gaussian peak shape of the measured spectral peak and is media.
3.3 Charge carrier transport and the Hecht equation

On creation a electron-hole pair will in a biased detector material move on average a certain distance. This distance is known as the drift length, \( l \), and can be different for electrons and holes. The expressions for electron and hole drift lengths can be seen in equation 3.15

\[
l_e = v_e \tau_e \\
l_h = v_e \tau_h
\]  

(3.15)

where \( v_e \) and \( v_h \) are the drift velocities and \( \tau_e \) and \( \tau_h \) are the mean carrier lifetimes (average time a charge carrier remains free before being trapped). The drift velocities are given by expression 3.16.

\[
v_e = \mu_e E \\
v_h = \mu_h E
\]  

(3.16)

where \( E \) is the electric field strength used to bias the detector and \( \mu_e \) and \( \mu_h \) are the electron and hole mobilities. The mobility is a measure of how strongly a charge carriers motion is affected by an applied electric field. By inserting equation 3.16 into equation 3.15 it is possible to express the drift lengths as a function of the mobility and lifetime parameters as can be seen in equation 3.17.

\[
l_e = \mu_e \tau_e E \\
l_h = \mu_h \tau_h E
\]  

(3.17)

From equation 3.17 it can be seen that the material dependent \( \mu\tau \)-product is vitally important for the transport of charge carriers. It is often therefore stated as a measure of the quality of a detector material [59] [54] [60]. It should however be noted that while the mobility is a fundamental parameter of the material the lifetime is often a measure of the maturity of the material growth technique. The spectroscopic performance of a semiconductor material, solely from a charge transport viewpoint, is directly dependent of the ability of the charge carriers inducing the signal onto the read-out electronics to traverse the distance from interaction location to the attracting electrode. It is therefore important that the charge carrier drift length, for the operational voltage used, is equal to or surpasses the thickness of the detector. A measure of a detectors ability to collect all charges created is known as the Charge Collection Efficiency (CCE). The definition of the CCE is the ratio of the charge reaching the electrodes to charge created by the
Detector theory

An expression for the CCE as a function of interaction position for a material, using a planar electrode configuration and assuming equal trap distribution through the material, was developed in 1932 by Hecht [61]:

\[
CCE = \frac{\mu_e\tau_eE}{d} \left[ 1 - e^{\frac{-(d-x)}{\mu_e\tau_eE}} \right] + \frac{\mu_h\tau_hE}{d} \left[ 1 - e^{\frac{-x}{\mu_h\tau_hE}} \right]
\]  

(3.18)

where \( d \) is the thickness of the detector and \( x \) is the depth of interaction. It can be seen from the left hand expression of equation 3.18 that the resulting CCE is an addition of the electron CCE and hole CCE.

### 3.4 The Shockley-Ramo theorem and single polarity charge sensing

As the charge carriers, created by the ionising event, starts traversing the detector due to the bias voltage applied, the electrostatic coupling between the charge carriers and the electrodes will result in instantaneous charge induction on the electrodes. The complete induced charge on the electrodes after the charge carriers have traversed the distance from where they where created to the electrode attracting them gives rise to the electric pulse that is measured and related to the energy of the photon creating the ionising event. The classical way of calculating the time dependent charge induction on the electrode would be to calculate the electric fields for each position of the charge carriers trajectory. The complete charge induced on the electrode would be obtained by integrating the normal component of the electric field over the electrode surface for all the electric fields. In the late 1930’s a computationally much easier way of calculating the charge induced by a moving charge on an electrode was developed independently by W. Shockley [62] and S. Ramo [63]. The Shockley-Ramo theorem, which has been used extensively to calculate charge induction on complex electrode configurations, states that the induced charge, \( Q \), and induced current, \( i \), caused by a moving charge, on an electrode is given by:

\[
Q = -q\varphi_0(x)
\]

\[
i = qvE_w(x)
\]

(3.19)

where \( E_w \) and \( \varphi_0 \) is the weighting field and the weighting potential respectively. The weighting field is the electric field obtained when the electrode for which the induction is being calculated is set to unity, all other electrodes are set to zero and there are no charges present. The Shockley-Ramo theorem allows for the induced charge on an
Electrode to be calculated using a single electric field (the weighting field) without taking into account the moving charge carrier or space charge. The Shockley-Ramo theorem was initially developed for vacuum tube geometries but was later proven to hold valid for semiconductors as well \[64\]. The weighting potential as calculated for a planar detector and pixelated detector can be seen in figure 3.7. The change in variation of the weighting potential gives the strength of induction for charge carriers moving in this region of the detector. For the planar detector geometry the change of weighting potential is uniform throughout the detector. The charge induced on the electrode, for a set initial charge cloud, is therefore only dependent on the drift lengths, \(l_e\) and \(l_h\), and is an addition of the electron and hole induction. For the pixelated geometry the charge induced on a specific pixel is smaller when the charge is far away and increases rapidly when the charge is close to the anode. Physically this is an effect of the basic geometry of a pixelated device. When a charge carrier is located far away from a small pixel (anode) the pixel area covers a smaller angular field of the environment as seen from the charge carrier. The electrostatic coupling between the pixel and the charge carrier is therefore small. As the charge comes closer to the pixel the angular field covered by the pixel area as seen from the charge carrier increases resulting in an increased coupling between the charge carrier and the pixel. A charge carrier will therefore induce more charge on a pixel when moving in an area close to the anode side of the detector due to the increased electrostatic coupling. Most of the charge will be induced when the charge carrier is close to the 1-P region as seen in figure 3.7. \(P\) is the pixel to detector thickness ratio. As \(P\) becomes smaller the slope of the region from 0 to 1-P and the length from 1-P to P, known as the near field region, is reduced. This effect is known as the small pixel effect \[66\] \[67\] \[65\]. As a consequence of the small pixel effect the charge carrier type

\[\text{Figure 3.7: (Left) Weighting potential variation from cathode to anode for a (Left) planar electrode configuration (Right) pixelated (on the anode side) electrode configuration. Plots have been reproduced from \[65\]. The weighting potential for both plots ranges from 0 to 1 (unity) in accordance with the definition of the weighting field. The interaction depth is plotted from the cathode (0) to the anode (1).}\]
that is attracted to the pixelated electrode side (in this case electrons) are predominantly responsible for the induced charge, assuming a small $P$ value. It should also be noted that the detector can be operated at a higher countrate. This form of single polarity charge sensing, where the induced charge is given by the charge transport properties of the material as well as the variation of the charge induction profile resulting from the electrode geometry, is desirable for detector material where one of the two charge carrier types suffer from poor drift length, such as CZT. The improvement in energy resolution for material where one of the two charge carrier types has a low drift length by changing the electrode configuration was first noticed by Zanio et al. [68] using a spherical detector geometry. Up to date the coplanar electrode configuration [69] gives the best energy resolution but does not lend itself to imaging applications due to the non pixelated structure of its electrode configuration. For more detailed information on the coplanar electrode configuration please see [69] [70]. Improvements in energy resolution using pixelated structures was realized [67] [66] and correctly interpreted as a single charge sensing effect [66]. The Charge Induction Efficiency (CIE), the total charge induced on a electrode compared to the initial charge created by the ionising event, as a function of interaction depth can be seen for planar and pixelated electrode configurations in figure 3.8 The best spectroscopic performance is obtained with a CIE

![Figure 3.8](image)

**Figure 3.8:** The CIE as a function of interaction depth for the ionising event. (LEFT) Planar detector response with good $\mu\tau$-product for both electrons and holes (Ge response) and with typical $\mu\tau$-products obtained for spectroscopic grade CZT. (RIGHT) Pixelated detector response with typical $\mu\tau$-products obtained for spectroscopic grade CZT with different pixel sizes [59].

that does not vary with interaction depth. The left plot in figure 3.8 shows the CIE as a function of interaction depth for a planar detector with a good $\mu\tau$-product for both electrons and holes as well as for $\mu\tau$-products obtained for spectroscopic grade CZT. The right plot shows the CIE as a function of interaction depth for a planar detector, with CZT $\mu\tau$-products, using different pixel sizes. For a planar geometry and good $\mu\tau$-product (both electrons and holes can traverse the entire detector length) the CIE shows
perfect response. For planar configuration and CZT $\mu\tau$-products we see a negative linear slope with increased interaction depth. The low $\mu\tau$-product for the holes will result in very swift hole-capture leading to a charge induction that is mainly dependent on the movement of electrons. The CIE value will therefore be strongly dependent on the length for which the electrons are moving through the detector. As a result interaction close to the cathode, from where the electrons travel through the entire length of the detector, gives a higher CIE then if the interaction takes place close to the anode. The CIE response for CZT $\mu\tau$-products using pixelated electrode geometry shows the improvements when manipulating the weighting potential profile. When changing the P-value, by changing the pixel size, an decrease of CIE is seen with decreased pixel size for interactions close to the cathode. This is due to the smaller area of the pixel. As the pixel becomes smaller compared to the thickness of the detector the near-field region moves closer to the anode. For the electron cloud traversing the detector from close to the cathode an increase in trapping before reaching the near-field region will occur resulting in a lower charge being induced onto the pixel electrode compared to the same situation where a larger pixel area is used. Interactions closer to the anode show lower CIE for higher pixel sizes. This is again due to the varied size of the near field region. If the interaction site is located inside the near field region the electron cloud will not move through the initial part of the near field region and less charge will consequently be induced onto the pixel electrode. For a larger pixel area the near field region will extent further away from the anode and the effect will become more pronounced leading to a lowered induced charge with increased pixel size. The best CIE profile, 5x5mm pixel size in figure 3.8, is therefore ,for a pixelated geometry, dependent on both the ratio of detector thickness to the pixel size and the $\mu\tau$-products of the detector material [59] [71].

3.5 Charge cloud diffusion and charge sharing

As a charge cloud is created by an ionising event and starts traversing the detector due to the bias voltage it also starts to expand due to thermal energy and the charge carrier density variation. The increase of the charge cloud cross section can be approximated using a Gaussian distribution with the standard deviation, $\sigma_{CC}$ given by [56]:

$$\sigma_{CC} = \sqrt{2Dt} \quad (3.20)$$

where $t$ is the time passed since the creation of the charge cloud and $D$ is the material dependent diffusion coefficient. The diffusion coefficient is given by the product of the thermal velocity, $v_{\text{therm}}$, and the mean free length of the charge carriers $l$. Using this
definition and the theorem of equipartition of energy for one dimension the Einstein relation, relating the mobility and diffusion coefficient for a material, can be obtained:

\[ D = \left( \frac{kT}{q} \right) \mu \]  

(3.21)

where \( k \) is the Boltzmann constant and \( T \) is the absolute temperature. Inserting equation 3.22 into equation 3.20 and using the relationship \( \mu = q\tau/m_n \), where \( m_n \) is the effective mass of the charge carrier, gives:

\[ \sigma_{CC} = \sqrt{\frac{2kT x}{qE}} \]  

(3.22)

which can be used to approximate the charge cloud cross section size as a function of distance traveled, \( x \), from the point of charge cloud creation due to the applied bias. If the charge cloud cross section size becomes so large that some of the charge is induced on the electrode of the original pixel and some of the charge is induced on neighboring pixels an error is introduced in our measurement known as charge sharing. Unless correctly compensated for charge sharing leads to a degradation of the spectral response of the detector.

### 3.6 High Z semiconductors for room temperature spectroscopic radiation detectors

In 1977 Armantrout et al. published a paper ranking the most promising high Z materials for \( \gamma \)- and X-ray detection purposes [60]. The desired device characteristics were a high atomic number in order to have a good detection efficiency, good \( \mu\tau \)-products in order to obtain good collection efficiencies and special consideration was directed to the hole mobility since it is considered the most common intrinsic limitation in high Z detector materials. To initially single out material candidates a number of selection criteria was used. The selection would contain only binary compounds due to the increased stoichiometric problems of ternary and higher order compounds. Only materials with \( Z > 50 \) and materials that were not naturally radioactive would be considered. In order to enable room temperature operation a band gap range of 1.5 eV to 2.2 eV was also used. The lower band gap limit was set by leakage current consideration and the higher limit due to the decrease in mobility with increased band gap due to polar lattice scattering. These criteria resulted in 14 possibilities which is shown in table 3.1. From the list in figure 3.1 CdTe was singled out as the best choice when considering just the
Table 3.1: List of the initial 14 high Z materials selected by Armantrout et al. as best candidates for \( \gamma \)- and X-ray detection. Reproduced from [60]

<table>
<thead>
<tr>
<th>Sulfides</th>
<th>( E_g ) (eV)</th>
<th>( \mu_e ) ( (cm^2/V) )</th>
<th>( \mu_h ) ( (cm^2/V) )</th>
<th>( \tau_{min} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>HfS(_2)</td>
<td>1.96</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>HgS</td>
<td>2.21</td>
<td>(&lt; 1)</td>
<td>(&lt; 1)</td>
<td>100 µsec</td>
</tr>
<tr>
<td>InS</td>
<td>1.86</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>OsS(_2)</td>
<td>2.00</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Sb(_2)S(_3)</td>
<td>1.92</td>
<td>15</td>
<td>45</td>
<td>10 µsec</td>
</tr>
<tr>
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<td>2.21</td>
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<thead>
<tr>
<th>Selenides</th>
<th>( E_g ) (eV)</th>
<th>( \mu_e ) ( (cm^2/V) )</th>
<th>( \mu_h ) ( (cm^2/V) )</th>
<th>( \tau_{min} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>CdSe</td>
<td>1.74</td>
<td>650</td>
<td>50</td>
<td>10 µsec</td>
</tr>
<tr>
<td>La(_2)Se(_3)</td>
<td>1.97</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WSe(_2)</td>
<td>1.57</td>
<td>100</td>
<td>80</td>
<td>13 µsec</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Tellurides</th>
<th>( E_g ) (eV)</th>
<th>( \mu_e ) ( (cm^2/V) )</th>
<th>( \mu_h ) ( (cm^2/V) )</th>
<th>( \tau_{min} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>CdTe</td>
<td>1.5</td>
<td>1050</td>
<td>80</td>
<td>6 µsec</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Iodides</th>
<th>( E_g ) (eV)</th>
<th>( \mu_e ) ( (cm^2/V) )</th>
<th>( \mu_h ) ( (cm^2/V) )</th>
<th>( \tau_{min} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>BiI(_3)</td>
<td>1.73</td>
<td>680</td>
<td>20</td>
<td>5 µsec</td>
</tr>
<tr>
<td>HgI(_2)</td>
<td>2.22</td>
<td>94</td>
<td>4</td>
<td>25 µsec</td>
</tr>
<tr>
<td>SbI(_3)</td>
<td>2.22</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ReI</td>
<td>Exists?</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

mobility and HgI\(_2\) was of interest due to its very large hole trapping time. The remaining 12 choices where considered, from a crystal growth point of view, to give no advantages over CdTe. A relaxation of the atomic number criteria and the binary compound constraint yielded the final 9 candidates of potential detector materials and can be seen in table 3.2. A number of drawbacks of some of the 9 candidates where pointed out by Armantrout. WSe\(_2\), BiI\(_3\) and HgI\(_2\) all form layered structures making them prone to damage during detector packaging. Cs\(_3\)Sb has a large non stoichiometry leading to Cs and Sb vacancies [72]. For WSe\(_2\) only small single crystals, rich in Se, has managed to be grown [73] [74]. CdSe was expected to have many of the same problems as CdTe but with a lower mobility.

Of the original 9 materials only 3 are still candidates today. These are AlSb, HgI\(_2\) and CdTe [59] [54]. Another 3 materials that fall outside the criteria set up by Armantrout are also often stated as candidates. These are CdZnTe (ternary compound), PI\(_2\) (\( E_G > 2.2eV \)) and TlBr (\( E_G > 2.2eV \)). PbI\(_2\) was investigated as an alternative to HgI\(_2\) since they had similar Z but while HgI\(_2\) has a destructive phase change between the melting point and room temperature PbI\(_2\) has not [54]. Although PbI\(_2\) has shown good
<table>
<thead>
<tr>
<th>Binaries</th>
<th>Zmax</th>
<th>$E_g$ (eV)</th>
<th>$\mu_e$ ($cm^2/V/s$)</th>
<th>$\mu_h$ ($cm^2/V/s$)</th>
<th>$\tau_{min}$, $\mu$sec</th>
<th>Xtal Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>AlSb</td>
<td>51</td>
<td>1.62</td>
<td>*1200</td>
<td>*700</td>
<td>1</td>
<td>ZB</td>
</tr>
<tr>
<td>InP</td>
<td>49</td>
<td>1.35</td>
<td>4700</td>
<td>150</td>
<td>3</td>
<td>ZB</td>
</tr>
<tr>
<td>ZnTe</td>
<td>52</td>
<td>2.25</td>
<td>340</td>
<td>110</td>
<td>4</td>
<td>ZB</td>
</tr>
<tr>
<td>CdTe</td>
<td>52</td>
<td>1.50</td>
<td>1050</td>
<td>80</td>
<td>6</td>
<td>ZB</td>
</tr>
<tr>
<td>WSe$_2$</td>
<td>74</td>
<td>1.36</td>
<td>100</td>
<td>80</td>
<td>1</td>
<td>Layered</td>
</tr>
<tr>
<td>CdSe</td>
<td>48</td>
<td>1.74</td>
<td>650</td>
<td>50</td>
<td>10</td>
<td>W, ZB</td>
</tr>
<tr>
<td>BI$_2$</td>
<td>83</td>
<td>1.7</td>
<td>*680</td>
<td>*20</td>
<td>5</td>
<td>Layered</td>
</tr>
<tr>
<td>Cs$_3$Sb</td>
<td>55</td>
<td>1.6</td>
<td>500</td>
<td>10</td>
<td>40</td>
<td>Cubic (BCC)</td>
</tr>
<tr>
<td>HgI$_2$</td>
<td>80</td>
<td>2.2</td>
<td>94</td>
<td>4</td>
<td>25</td>
<td>Layered</td>
</tr>
</tbody>
</table>

Table 3.2: List of the final 9 high Z materials selected by Armantrout et al. as best candidates for $\gamma$- and X-ray detection. Reproduced from [60].

Spectral response at room temperature [75] the poor transport properties limits the detector thickness to 200$\mu$m resulting in a poor detection efficiency. AlSb was identified by Armantrout as the material with the highest potential. Calculations on the electron and hole mobilities reaching 1000$cm^2/Vs$ [76] and measurements ranging from 300-900$cm^2/Vs$ [77] [78] has been reported. Problems growing the material has however hindered the development of this material. No reliable measurements of the carrier lifetimes and no spectroscopic performance has been reported for AlSb [59] [54]. Spectroscopic performance has been obtained from TlBr [79]. Polarization effects [79] [80] [81], believed to be due to ionic conduction, and stability problems [82] are currently limiting the usability of TlBr as a detector material. HgI$_2$, CdTe and CdZnTe are currently the best high Z detector material options and are all still under development. HgI$_2$ suffers from polarization believed to be due to accumulation of space charge in the material [83] [42]. A strong improvement is seen in the spectral response for HgI$_2$ after the bias voltage has been applied for a long time [84] (on the timescale of days). CdTe is used less and less due to the higher resistivity, lower dislocation densities and lower polarization observed in CdZnTe. CdZnTe is currently being given the most research attention in the high Z material for $\gamma$- and X-ray detection area. The spectral response of HgI$_2$ and CdZnTe compared to "normal" detector materials, Si and GaAs, for two different sources can be seen in figure 3.9.
3.7 CZT growth and defects

Due to the long preparation times needed before detector operation when using HgI$_2$ [83], the problems associated with the ionic conduction properties of TlBr [83] [85] and the large research interest of CZT currently existing, CZT was chosen as the high Z material to substitute Si as the active detector crystal in the rTEDDI detector system. A number of different techniques to grow CZT has been tried over the years, all with the aim of producing large, single crystal, defect free, high resistivity material. What follows below is a description of the most common CZT growth methods and a review of the crystalline defects, and the subsequent effect on the X-ray response, found in the grown CZT.

3.7.1 The High Pressure Bridgman method

In the High Pressure Bridgman (HPB) growth process the constituent materials, i.e. Cd, Zn and Te, are placed in a crucible/ampule which is in turn placed in a high pressure chamber containing a multi zone heater. An overpressure of Argon, 10-150 atm, is
produced inside the pressure chamber. The material inside the crucible is subsequently physically moved through a specifically designed temperature profile. A schematical representation of the growth setup and temperature profile can be seen in figure 3.10. As the temperature increases in the beginning of the growth run the material constituents melts and compounds. The temperature is then lowered as the ampule moves further down the chamber and crystal. The overpressure in the chamber is used to suppress evaporation of Cd, otherwise likely due to its relatively high vapor pressure, which would lead to a gradual increase of Te in the CZT crystal along the growth axis. Despite this measure an increase of Te is present in the crystal when grown by the HPB method. The HPB

![Diagram of HPB growth chamber and temperature profile](image)

**Figure 3.10:** (a) Schematical representation of the HPB growth chamber and (b) a simulation of the temperature profile in a HPB growth chamber [47].

![Picture of a HPB grown CZT ingot](image)

**Figure 3.11:** Picture of a HPB grown CZT ingot [86].
Detector theory

method has been shown to produce good quality material with high resistivity, in the order of \(1 \times 10^{11} \Omega\text{cm}\), good \(\mu_e \tau_e\)-products ranging from \(10^{-3} - 10^{-2} \text{cm}^2\text{V}^{-1}\) [86] [87] [88] and is the favored growth method of eV PRODUCTS [89], now EI Detection & Imaging Systems. The largest problem with this growth method is the low yield of single crystalline material, making as little as 25% of the boule suitable as detector grade material [86]. Growth rates using the HPB method are around 0.1-1.0 mm hr\(^{-1}\), resulting in growth cycles on the range of 3-4 weeks.

### 3.7.2 The Modified Vertical Bridgman method

The Modified Vertical Bridgman (MVB) method is very similar to the HPB method with two main differences, the growth is done using no overpressure and the Accelerated Crucible Rotation Technique (ACRT) is introduced. The ACRT is introduced to produce convection in the melt in order to maintain stoichiometric growth leading to increased single crystal sizes and a higher uniformity of zinc concentration throughout the boule. Since no overpressure is used the system can be made lighter and technically less complicated but a loss of Cd from the melt during growth is present. To compensate for the Cd loss an excess of Cd is supplied during growth. A schematical representation of the growth setup and temperature profile can be seen in figure 3.12.

\[\text{Figure 3.12: Schematical representation of the MVB growth chamber with temperature profile [88].}\]

The MVB method has been shown to produce good quality material. Resistivities of up to \(5 \times 10^{10} \Omega\text{cm}\) has been found for doped material and \(\mu_e \tau_e\)-products of up to \(1.8 \times 10^{-2} \text{cm}^2\text{V}^{-1}\) have been
reported [90] [91]. Single crystal volumes of 50 cm$^3$ has been obtained using the MVB method and boule yields are in the excess of 50%. The MVB method was the favored growth method of Yinnel Tech. Inc. Growth rates using the MVB method are around 1.0 mm hr$^{-1}$, resulting in growth cycles on the range of 2-3 weeks.

### 3.7.3 The Traveling Heater method

In the Traveling Heater Method (THM) a polycrystalline CZT rod is used as a feed material while pure Te is used as a solvent. A schematic diagram of a THM growth system and temperature profile used to grow CdTe can be seen in figure 3.13 [92]. The solution zone, zone 3 in figure 3.13, has a temperature high enough to melt the pure Te. As the solution zone slowly moves up the polycrystalline CZT rod the liquid Te acts as a solvent, dissolving the CZT. The CZT subsequently diffuse through the solvent to a colder region where the lowered temperature decreases the CZT solubility, effectively crystal

---

**Figure 3.13:** A schematic diagram of a THM growth system used to grow CdTe [92].
3.7.4 Crystalline defects and resulting effect on the X-ray response

During material growth and detector manufacturing a number of different defects can be introduced into a CZT crystal. What follows are a discussion on some of the most important defects.

Cracks, grain- and twin-boundaries

Cracks, grain- and twin-boundaries are all produced during the growth of CZT. An axial slice of CZT showing cracks, grain- and twin boundaries can be seen in figure 3.14. Macroscopic cracks in CZT is one of the largest problems in melt grown material and is

the main reasons for the poor yield of detector grade material when using the HPB- or the MVB-method of growing. Cracking is believed to be due to the thermal and mechanical stress introduced during the growth with stress introduced at the crucible/material interface being a main contributor [88] [86]. It has been shown that using high porous crucibles and optimizing the temperature profile of the growth can reduce the degree of cracking observed [42]. The effects of cracks, grain and twin-boundaries on the detector performance can be seen in figure 3.15. Cracks gives an almost zero detector response. Large angle grain boundaries give as well an almost zero detector response. All grain boundary regions do however not show poor detector response. No correlation between the twin boundaries and detector response has been found indicating that twins do not effect the electric field or the charge collection properties to any larger extent.

Figure 3.14: Axial slice of a HPB grown CZT ingot with cracks, grain- and twin-boundaries clearly visible [88]. The average sizes of cracks are typically a few cm in length and 25µm in width [86]. The average grain size is typically 2-3cm [88].
Figure 3.15: Grain, twin and crack map (bottom) of two pieces of HPB CZT and corresponding detector response (top). The detector response represents the total number of counts (see scalebar) above the noise level for a $^{57}$Co source measured by a single channel analyzer [88]. The counts where measured by a 1mm diameter conductive rubber probe, surrounded by a guard ring, moved over the surface in 1mm steps using an X-Y stage. Cracks gives an almost zero detector response. Large angle grain boundaries give as well an almost zero detector response. All grain boundary regions do however not show poor detector response. No correlation between the twin boundaries and detector response has been found.

**Pipes**

Pipes, tubular structures running along the growth axis on the ingot, can be observed in melt grown material. They often appear in families and the pipe density decreases from the tip of the ingot to the heel. An IR image of the tip of a CZT ingot showing pipes can be seen in figure 3.16. Pipes are believed to be caused by the partial overpressure of Cd creating Cd bubbles at the growth interface. The Cd bubbles runs through the material as it is grown, crating tubular structures along the growth axis that are filled with Te [42]. The effect of pipes on the detector response can be seen in figure 3.17 [93]. It has been suggested that the reduction in alpha peak centroid, the average charge induced on the collecting electrode when exposing the material to alpha radiation, as a function of position on the sample is due to a varying amount of trapping around the pipe.
Precipitates and inclusions

Precipitates and inclusions are both accumulation of a single atomic species in a region of the grown CZT, effectively creating lumps of Te or, less commonly, Cd through out the material. They differ in size and are formed through different processes. Precipitates are usually $<1\mu$m in size and is randomly distributed throughout the material [88] [94] and consequently have little effect on the detector performance. During growth point defects are created within the crystal. As the material cools these defects form micro-cavities that become filled with Te, creating Te-precipitates. Inclusions are usually 1-100$\mu$m in size and have been shown to attract impurities due to the increased impurity solubility of Te [95]. The creation of inclusions occur at the growth interface where melt droplets are captured. An IR image of the heel of a CZT ingot showing inclusions can be seen
in figure 3.18 Te inclusions are found distributed randomly throughout the material with an increase at grain- and twin-boundaries effectively decorating them. Due to the relatively low band gap of Te a high concentration of inclusions lead to an increase in leakage current and modified electric fields within the detector. The impurities associated with the inclusions create trapping sites which lead to a variation in the charge transport properties in and around the inclusions. The effect of inclusions on the detector response can be seen in figure 3.19 It has been suggested that the reduction in alpha peak centroid is due to a varying amount of trapping, and subsequent change in charge transport properties, around the Te inclusions.
Impurities, vacancies, antisites and interstitials

The introduction of impurities as well as intrinsic crystalline point defects in grown CZT produces defect energy levels inside the band gap. Table 3.3 gives a summary of the energy levels introduced into the band gap by some common impurities in CdZnTe. Intrinsic crystalline point defects can be categorized into three groups. These are vacancies, antisites and interstitials. A vacancy is a lack of an atom in the crystal lattice. Antisites are atoms located in the wrong positions in the lattice, such as a Te atom on a Cd site \((\text{Te}_{\text{Cd}})\). Interstitials are atoms placed in between lattice sites. Table 3.4 lists some of the energy levels reportedly introduced by crystalline point defects in CdTe. As can be seen in table 3.4 there is currently no consensus within the scientific community what levels are actually introduced by these types of defects. Defect levels in the band gap are grouped into two groups, deep levels or shallow levels. Deep levels are levels close to the middle of the band gap \((E>0.4\text{eV})\) while shallow levels are close to the valence or conduction band \((E<0.4\text{eV})\). A high density of shallow levels will lead to a large number of thermally excited charge carriers reducing the resistivity of the material and increasing the leakage current. Charge carriers trapped in deep levels are usually

### Table 3.3: Some common impurities in CdTe and CdZnTe

<table>
<thead>
<tr>
<th>Element</th>
<th>Group</th>
<th>Energy (eV)</th>
<th>Type</th>
<th>Method</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na</td>
<td>I</td>
<td>0.02, 0.06</td>
<td>Acceptor</td>
<td>Theory, PL</td>
<td>[96] [97]</td>
</tr>
<tr>
<td>Cu</td>
<td>Transition</td>
<td>0.15, 0.22</td>
<td>Acceptor</td>
<td>Theory, PL</td>
<td>[97] [98] [96]</td>
</tr>
<tr>
<td>Ag</td>
<td>Transition</td>
<td>0.11, 0.12, 0.15</td>
<td>Acceptor</td>
<td>PL, Theory</td>
<td>[97] [98] [96]</td>
</tr>
<tr>
<td>Au</td>
<td>Transition</td>
<td>0.20, 0.26</td>
<td>Acceptor</td>
<td>Theory, PL</td>
<td>[96] [97]</td>
</tr>
<tr>
<td>V</td>
<td>Transition</td>
<td>0.80</td>
<td>Acceptor</td>
<td>Theory, PL</td>
<td>[99]</td>
</tr>
<tr>
<td>Al</td>
<td>III</td>
<td>0.01, 0.02</td>
<td>Donor</td>
<td>PL, Theory</td>
<td>[96] [100]</td>
</tr>
<tr>
<td>In</td>
<td>III</td>
<td>0.02, 0.04</td>
<td>Donor</td>
<td>PL, Theory</td>
<td>[100] [96] [101]</td>
</tr>
<tr>
<td>As</td>
<td>V</td>
<td>0.09, 0.10</td>
<td>Acceptor</td>
<td>PL, Theory</td>
<td>[97] [96]</td>
</tr>
<tr>
<td>Bi</td>
<td>V</td>
<td>0.30</td>
<td>Acceptor</td>
<td>Theory</td>
<td>[96]</td>
</tr>
<tr>
<td>Cl</td>
<td>VII</td>
<td>0.02</td>
<td>Donor</td>
<td>PL</td>
<td>[100]</td>
</tr>
</tbody>
</table>

Table 3.4: Native point defects in CdTe compounds.

<table>
<thead>
<tr>
<th>Defect</th>
<th>Energy (eV)</th>
<th>Type</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>(V_{Cd})</td>
<td>0.21, 0.43, 0.46, 0.47, 0.76</td>
<td>Acceptor</td>
<td>[96] [102] [103] [104] [105] [106]</td>
</tr>
<tr>
<td>(V_{Te})</td>
<td>0.71, 1.10</td>
<td>Donor</td>
<td>[96] [107] [106]</td>
</tr>
<tr>
<td>(Cd_{Te})</td>
<td>0.10</td>
<td>Donor</td>
<td>[96]</td>
</tr>
<tr>
<td>(Te_{Cd})</td>
<td>0.59, 0.74, 0.77</td>
<td>Donor</td>
<td>[96] [102] [108] [103]</td>
</tr>
<tr>
<td>(Cd_i)</td>
<td>0.45, 0.64</td>
<td>Donor</td>
<td>[96] [106]</td>
</tr>
<tr>
<td>(Te_i)</td>
<td>0.57</td>
<td>Acceptor</td>
<td>[96]</td>
</tr>
</tbody>
</table>
not able to obtain enough energy to escape, making the deep levels act as centers for carrier trapping and recombination. As a result the CIE is reduced. In large enough concentrations the deep levels are the reason for charge carrier lifetime limitations. In MVB grown material, at low temperature operation, electron trapping has been shown to produce internal electric fields resulting in degraded charge transport properties \[109\]. Under high flux operation (>60,000 counts/sec/mm\(^2\)) trapping of holes have been shown to be able to produce spatially charged regions that modifies the electric field inside a bias detector, known as polarization, resulting in dynamic lateral polarization, electric field collapse and device failure \[110\] \[111\] \[112\] \[113\]. The effect of dynamical lateral polarization in a polarizing device under varied flux irradiation conditions can be seen in figure 3.20. The build up of a spatially charged region in the detector is believed to

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3_20.png}
\caption{Counting map for a 3 mm thick, 16x16 pixel CdZnTe monolithic detector array, biased at 900 V and subjected to a (a) low, (b) intermediate, (c) high, and (d) an ultrahigh flux of x-rays through a 4 mm diameter collimator shown as a dashed white line. \[111\].}
\end{figure}

results in a modified electric potential across the detector as seen in figure 3.21. The altered potential profile produces a flux dependent lensing effect for the electron trajectories as can be seen in figure 3.22 resulting in lateral electron transport. For pixelated devices intended for imaging this effect results in detection of photons in pixels they where not absorbed, effectively destroying the imaging capability of the device.
Figure 3.21: Detector electric potential profile simulation for the detector described in figure 3.20 when exposed to high flux of X-rays [111].

Figure 3.22: Electric field lines shown as solid lines with arrows depicting electron trajectories for part of a 3 mm detector, biased at 900 V and subjected to a (a) low, (b) intermediate, (c) high, and (d) an ultrahigh flux of photons through a 4 mm diameter collimator. Equipotential surfaces are shown as dashed curves [111].
Chapter 4

Proof of concept for the rTEDDI system

At the start of the project a collimator array and a pixelated detector, intended as the core of the rTEDDI system, had already been developed. They had however not been incorporated into a working system and no proof of concept for the imaging technique had been obtained. The work done to realize the rTEDDI system and obtaining a proof of concept for the system is described in this chapter, starting with the building of the system. The proof of concept for the rTEDDI system, using a Si detector, follows with the investigation of 4 test samples. The tests conducted at a low energy/low flux synchrotron beamline, station 7.6 at SRS Daresbury. The verification of the results using International Centre for Diffraction Data (ICDD) Powder Diffraction File (PDF)’s and traditional TEDDI investigation has been obtained. Diffraction patterns from each of the 4 test samples have been Rietveld refined, by Professor R.J. Cernik, using Topas. The results from initial testing of the rTEDDI system on a high energy/high flux synchrotron beamline, station 16.3 at SRS Daresbury, is shown and observed problems discussed.

4.1 Realization of the rTEDDI system

In order to decrease the very long scan times needed to produce 2D TEDDI images the rTEDDI system was built. The underlying concept of adding collimators and energy resolving detectors in an array configuration in order to diffraction image an entire plane simultaneously can be seen in figure 2.3. The principal was realized using an existing pixelated energy resolving detector, known as the ERD detector [25] and a purposely designed collimator array [24]. One of the manufactured collimator arrays can be seen in figure 4.1. The collimator array consists of a number of tungsten plates, each with a
Figure 4.1: Photograph of one of the manufactured tungsten collimator arrays with the laser drilled holes in one of the plates blown up to be visible.

16x16 array of 50\(\mu\)m holes on a 300\(\mu\)m pitch, aligned and positioned at defined distances with respect to each other in order to prevent cross-talk between collimators. A detailed discussion on the tungsten collimator array is found in Chapter 7. The ERD detector with Si as the active detector crystal can be seen in figure 4.2. The spectroscopic detector has a 16x16 array of pixels, on a 300\(\mu\)m pixel pitch, where each pixel has its own readout channel associated with it. A detailed description of the ERD detector is found in Chapter 6. The detector and collimator array was aligned using 2 PI M-111 micro translation stages and a PI M-116 precision rotation stage, coupling each collimator to a specific detector pixel. The moving stages was controlled from a PC user interface via a PI Apollo Stepper-Motor Controller using a RS-232 communication protocol. The basic setup can be seen in figure 4.3.
4.2 First X-ray colour images using the rTEDDI system

In order to verify the rTEDDI concept the developed system was set up at station 7.6 at SRS Daresbury. The system was attached to the forward rotational stage of the station stage. A Hamamatsu S8193 Si photodiode [114], utilizing a ceramic scintillator, was placed behind the collimator array, after having verified the straight through beam hit the front end of the collimator array. The x-ray intensity was subsequently measured while step scanning the pitch and yaw angles of the collimator array with respect to the straight through beam. This was done in order to verify that the field of view of the collimator would intersect the beam, when set to a $2\theta$-angle, and increase the accuracy with which the $2\theta$-angle of the setup could be determined. The Hamamatsu detector was used in order to minimize the time the ERD Si detector would have to be exposed to the straight through beam due to radiation damage considerations. The Hamamatsu detector was subsequently substituted by the ERD Si detector which was step scanned in xy-direction and roll angle using the PI stages in order to find a good detector pixel to collimator hole alignment. Finally the rTEDDI setup was set to an angle with respect to the incoming beam, the setup $2\theta$-angle, and a sample stage was utilized to allow for the sample to be moved along the beam to find the intersection points of the collimator field of views and the beam (the imaging lozenges). The setup can be seen in figure 4.4.
When working with thin samples, such as the once used in this experiment due to the low X-ray energies required by a Si detector, an alternative imaging geometry to the vertically thin beam geometry described in figure 2.1 and 2.3 can preferentially be used. By using a large box section beam and a thin sample the plane being imaged is flipped from a horizontal direction to a vertical direction as can be seen in figure 4.5. By using a large box section beam, when working with thin samples, the entire array configuration of the rTEDDI system is utilized instead of just a single or a number of rows. For the
Proof of concept for the rTEDDI system

The four thin sample materials used to test the rTEDDI concept both imaging geometries where used. The four sample materials used where Al powder, a Cu foil, two polymer sheets with test marks cut out of them and a piece of deer antler tribecular bone. The Al powder and Cu foil was imaged using the vertically thin beam geometry while the two polymer sheets and the deer antler bone samples where imaged using the large box section beam geometry. The results from all of the four samples where verified using classical TEDDI measurements obtained at station 16.4, SRS Daresbury. The setup used at station 16.4 has been described elsewhere [115]. The diffraction peak positions found using the rTEDDI measurements for the Al powder sample and the Cu foil sample was compared to values obtained from ICDD PDF’s. Attempts at rietveld refinement was done for the diffraction patterns obtained from all 4 sample materials.

4.2.1 Al powder sample

The rTEDDI setup used when imaging the Al powder sample is illustrated in figure 4.6. The powder was created by hand-filing a piece of Al. A 2θ-angle of 22° was used. The high angle was used due to the low energy range of the white X-ray beam available at station 7.6 as can be seen in figure 4.29. The Al powder, placed in a small plastic bag, was scanned along the direction of the beam in order to find the positions of the diffraction lozenges. Figure 4.7 (a) shows the total scattering as registered for each pixel of the detector. The intensity is false coloured with lighter colour being the result of a higher number of registered events. The vertically thin beam geometry with a thin sample results in a vertical cross-section of the sample being imaged, where movement from bottom to top of the imaged sample (sample area) in figure 4.7 (a) equates to

![Illustration of the rTEDDI setup used when imaging the Al sample. A horizontally thin beam geometry was used and a 2θ-angle of 22°.](image-url)
Figure 4.7: The first rTEDDI imaged obtained using the vertically thin beam geometry. (a) A false coloured, total intensity map depicting a slice through a thin Al powder sample. Movement from top to bottom in the image equates to movement in and out of figure 4.6, while the thickness of the sample area is set by the vertical thickness of the white X-ray beam. (b) A map of the energy dispersive diffraction patterns, containing atomic structural information, recorded for each pixel. There is no scattering from the region where no material is present at the imaging lozenges.
movement into the image in figure 4.6 and the thickness of the sample area is set by the vertical dimension of the beam. The inter pixel variation in intensity is believed to mainly stem from variation in sample thickness. Figure 4.7 (b) shows the individual energy dispersive X-ray scattering pattern from each pixel. There is no scattering from the region where no material is present at the imaging lozenges. Figure 4.8 shows the peak position of the diffraction peaks for Al as a function of d-spacing, as stated in the ICDD PDF. A conversion into resulting peak position in energy is stated for a $2\theta$-angle of $22^\circ$, as calculated using equation 2.3. Three of the diffraction patterns registered, and

![Diagram showing peak positions and relative intensities](image)

**Figure 4.8:** Peak position and relative intensity of Al diffraction peaks as stated in the ICDD PDF. A conversion from d-spacing into energy for the peak positions is shown as calculated using the energy dispersive equation of diffraction and a $2\theta$-angle of $22^\circ$.

their position in the sample can be seen in figure 4.9 A good correlation between the ICDD PDF diffraction peaks and the measured peak positions, best illustrated by pixel 34 in figure 4.9, was found. The higher energy peaks in the diffraction patterns are only distinguishable in the pixels with the highest registered countrates and consequently the highest statistical quality diffraction pattern. The data was collected during 17 hours which is in the time scales stated for 2D imaging using TEDDI. The reasons for the long scanning times are discussed in section 4.2.5. Six of the highest statistic pixel spectra, plotted both vs. energy and vs. d-spacing, can be seen in figure 4.10, showing the variation in detectable peaks for the pixels. The variation is believed to mainly be due to poor statistical quality and preferred orientation effects. A direct comparison of the ICDD PDF d-spacing values against the measured d-spacing values obtained for the six pixels shown in figure 4.10 can be seen in table 4.1. A verification of the Al powder sample d-spacings was obtained by a single point scan TEDDI measurement obtained at the existing setup on station 16.4 SRS Daresbury. A plot of the resulting energy dispersive diffraction pattern can be seen in figure 4.11 both as a function of energy and d-spacing. A direct comparison of the ICDD PDF d-spacing values against the measured d-spacing values obtained for the single point TEDDI measurement on the Al powder sample is shown in table 4.2. Although a few of the spectra in figure
Figure 4.9: The registered energy dispersive diffraction patterns for three of the highest statistics pixels. Out of the three pixels only pixel 34 shows all 4 expected diffraction peaks.

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
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<td>2.386</td>
<td>2.43</td>
<td>2.336</td>
</tr>
<tr>
<td>d2</td>
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<td>2.005</td>
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<td>2.016</td>
<td>2.06</td>
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<tr>
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<td>NA</td>
<td>1.471</td>
<td>1.456</td>
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<tr>
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<td>NA</td>
<td>1.194</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
</tr>
</tbody>
</table>

Table 4.1: Comparison of resulting d-spacings for the different peaks in the pixel spectra shown in figure 4.10 compared to the d-spacing values stated in the ICDD PDF. A high degree of correlation was observed for discernible peaks.

<table>
<thead>
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<th>Database (Å)</th>
<th>Pixel 27 (Å)</th>
</tr>
</thead>
<tbody>
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<td>2.339</td>
</tr>
<tr>
<td>d2</td>
<td>2.0247</td>
<td>2.0277</td>
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<tr>
<td>d4</td>
<td>1.2209</td>
<td>1.2233</td>
</tr>
</tbody>
</table>

Table 4.2: Comparison of resulting d-spacings for the different peaks in the single TEDDI spectra compared to the d-spacing values stated in the ICDD PDF. A high degree of correlation was observed for all peaks.
Figure 4.10: Plots of the six highest statistic pixels plotted as function of both energy and d-spacing. The number of discernible peaks varies between pixels and is accredited to poor statistical quality and preferred orientation effects.
Figure 4.11: Plot of the single point TEDDI measurement plotted as function of both energy and d-spacing. The measurement used a 2θ-angle of $7.32^\circ$. The measurement was made to verify the sample material d-spacings.

Figure 4.12: An energy dispersive diffraction pattern from the indicated pixel was refined using peak (1 1 1) and (2 0 0). The refinement (Right), although suffering from low statistical quality observed data (blue), gave a good fit (red). Resulting parameter values of $\chi^2=0.94$ and $a=4.0497(38)\,\text{Å}$ was obtained.

4.10 showed capability of resolving the higher energy peaks only the two lower energy peaks, the (1 1 1) and (2 0 0) peaks where distinguishable in all pixels containing sample scattering. Using the (1 1 1) and (2 0 0) peak a lattice parameter refinement using Topas, as can be seen in figure 4.12, was possible in spite of the poor statistical quality resulting in parameter values of $\chi^2=0.94$ and $a=4.0497(38)\,\text{Å}$. The lattice parameter accuracy is found to be in the second decimal place. A higher accuracy would be needed for micro strain measurements, accuracy in the fourth decimal place, but the results show promise for tasks such as strain mapping of whole engineered components with improvement in technology and statistical quality of the diffraction patterns.

4.2.2 Cu foil sample

The rTEDDI setup was also used to diffraction image a Cu foil sample. A 2θ-angle of $22^\circ$ was used for the same reasons as stated for the Al powder sample, and a vertically
Figure 4.13: The rTEDDI image obtained for the Cu foil sample using the vertically thin beam geometry. (Left) A false coloured, total intensity map depicting a slice through the thin Cu foil sample. A bend in the material is clearly visible in the image. (Right) A map of the energy dispersive diffraction patterns recorded for each pixel. There is no scattering from the region where no material is present at the imaging lozenges.
thin beam geometry was applied. Figure 4.13 shows the rTEDDI image of the sample and the energy dispersive diffraction map. The diffraction peak positions as a function of d-spacing for Cu, as stated in the ICDD PDF, can be seen in figure 4.14. A conversion from d-spacing to energy for the peak positions are also stated. Three of the diffraction patterns registered, and their position in the sample can be seen in figure 4.15 Only two of the four diffraction peaks are visible in the registered diffraction patterns. A verification of the Cu foils sample d-spacings was obtained using a single point TEDDI measurement, obtained at station 16.4, SRS Daresbury. The energy dispersive diffraction pattern obtained from the measurement can be seen in figure 4.16, while the six highest statistic pixel spectra for the rTEDDI measurement, plotted both as a function of energy and d-spacing can be seen in figure 4.17. A direct comparison between the ICDD PDF d-spacing values and the measured values for the pixels in figure 4.17 can be seen in table 4.3. The lower d-spacing peaks, expected from the ICDD PDF value, where not present in neither the rTEDDI nor the TEDDI measurements. This verifies that preferred orientation, which posses fundamental limitation for energy dispersive

![Figure 4.14: Peak position and relative intensity of Cu diffraction peaks as stated in the ICDD PDF. A conversion from d-spacing into energy for the peak positions is shown as calculated using the energy dispersive equation of diffraction and a 2θ-angle of 22°.](image)

<table>
<thead>
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<th>Pixel 57 (Å)</th>
<th>Pixel 213 (Å)</th>
<th>Pixel 219 (Å)</th>
<th>Pixel 223 (Å)</th>
<th>Pixel 230 (Å)</th>
</tr>
</thead>
<tbody>
<tr>
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<td>2.132</td>
<td>2.058</td>
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<td>d2</td>
<td>1.8075</td>
<td>1.838</td>
<td>1.84</td>
<td>1.779</td>
<td>NA</td>
<td>1.852</td>
<td>1.821</td>
</tr>
<tr>
<td>d3</td>
<td>1.2781</td>
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<td>NA</td>
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<td>NA</td>
<td>NA</td>
<td>NA</td>
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<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
</tr>
</tbody>
</table>

Table 4.3: Comparison of resulting d-spacings for the different peaks in the pixel spectra shown in figure 4.10 compared to the d-spacing values stated in the ICDD PDF. A high degree of correlation was observed for discernible peaks.
Figure 4.15: The registered energy dispersive diffraction patterns for three of the highest statistics pixels. Only peak d1 and d2 are visible in the diffraction pattern.

Figure 4.16: Plot of single point TEDDI measurement as a function of energy and d-spacing for the Cu foil sample. A 2θ-angle of 7.324° was used. Only the higher d-spacing reflections, from planes (1 1 1) (d1) and (2 0 0) (d2), are visible. This verifies that the lack of low d-spacing reflections in the rTEDDI measurement is mainly due to preferred orientation of the sample, as would be expected.
Figure 4.17: Plots of the six highest statistic pixels plotted as function of both energy and d-spacing. Peak d1 and d2 are present in all but one diffraction pattern. The lower d-spacing peaks are not present in any diffraction pattern.
diffraction imaging, is the main reason for the lack of reflection of the low d-spacing planes.

4.2.3 Polymer sheet samples

The rTEDDI setup used when imaging the 2 polymer sample is illustrated in figure 4.18. A 2θ-angle of 13°, and the large box section beam (5x5mm) geometry was used. Both sample sheets was made from the same polymer material and had test marks in the form of an X and two circles, as can be seen in figure 4.19 (a) and (b). The test marks where positioned facing the incoming beam in order to allow for imaging of the entire test structure simultaneously using the chosen geometrical setup. The colour centres created in the polymer material by the incoming beam is clearly visible in figure 4.19 (a) and (b). Figure 4.19 (c) and (d) shows the rTEDDI images of the two polymer samples, both with the test marks clearly visible, demonstrating the imaging capability of the technique. Both images are false coloured in the same way as described for Figure 4.7 (a). Figure 4.19 (e) and (f) show the individual energy dispersive X-ray scattering pattern from each pixel for the two samples. No scattering is recorded in the area where no material is present (pink shading). One diffraction pattern for each sample and the position of origin for those diffraction patterns can be seen in figure 4.20. Two peaks are clearly distinguishable in the diffraction pattern. A positive peak verification was obtained using a single point TEDDI measurement, see figure 4.21, using the same setup parameters as those stated for the Al powder sample. Six of the highest statistics spectra obtained from the rTEDDI measurement, taken from the polymer sample with circular test marks, plotted both as a function of energy and d-spacing can be seen in figure 4.22. A direct comparison between the measured d-spacing of the six pixels are
Figure 4.19: (a) Photograph of polymer sheet sample with X shaped test mark. (b) Photograph of polymer sheet sample with circular shaped test mark. For both samples the colour centres created by the incoming X-ray beam is clearly visible. Both samples were imaged using the large box section beam geometry with the test marks facing the incoming beam. (c) and (d) False coloured, total intensity maps clearly showing the shapes of the test marks. Movement from top to bottom in the image equates to movement in and out of figure 4.18, while movement right to left equates to movement up and down. (e) and (f) Maps of the energy dispersive diffraction patterns, containing atomic structural information, recorded for each pixel, for both samples. There is no scattering from the region where the sample material has been removed to produce the test marks.
Proof of concept for the rTEDDI system

Figure 4.20: Registered energy dispersive diffraction patterns and their positions in the samples. Two peaks are clearly visible. Both sample sheets are of the same material.

Figure 4.21: Plot of single point TEDDI measurement of the polymer sample plotted as both a function of energy and d-spacing. The measurements verified the d-spacings of the sample material.

stated in table 4.4. The patterns seen are very similar but not identical. The variation is largely attributed to local variations in structure and preferred orientation. The

<table>
<thead>
<tr>
<th>Peak</th>
<th>Pixel 69 (Å)</th>
<th>Pixel 100 (Å)</th>
<th>Pixel 120 (Å)</th>
<th>Pixel 158 (Å)</th>
<th>Pixel 207 (Å)</th>
<th>Pixel 251 (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>d1</td>
<td>4.536</td>
<td>4.535</td>
<td>4.425</td>
<td>4.343</td>
<td>4.524</td>
<td>4.503</td>
</tr>
<tr>
<td>d2</td>
<td>3.678</td>
<td>3.777</td>
<td>3.693</td>
<td>3.705</td>
<td>3.687</td>
<td>3.665</td>
</tr>
</tbody>
</table>

Table 4.4: Comparison of resulting d-spacings for the different peaks in the pixel spectra shown in figure 4.22.

lattice parameter refinement of the polymer sample, seen in figure 4.23, yielded a $\chi^2$ of 1.45 with possible space groups P21, P21/m or P21/c, a=7.058(50)Å, b=10.679(36)Å, c=17.839(15)Åand $\beta$=68.4(21)$^0$, corresponding to the structure of nylon-6. This was
Figure 4.22: Plots of the six highest statistic pixels (circular test mark sample) plotted as a function of both energy and d-spacing. The patterns are similar but not identical. The variation is attributed to local variations in structure and preferred orientation.
Figure 4.23: (Left) Two typical diffraction patterns from the polymer samples. (Right) A lattice parameter refinement of a typical polymer sample diffraction pattern. A $\chi^2$ of 1.45 was achieved and lattice parameter errors were in the second decimal place.

confirmed by referring the result of a FTIR measurement of the sample to that of nylon-6 stored in the Hummel Polymer Library. Lattice parameter errors were in the second decimal place.

4.2.4 Deer antler bone sample

The rTEDDI setup used to image the deer antler trabeculae bone sample was identical to the one stated for the polymer sample, and can be seen in figure 4.24. The relevant

Figure 4.24: Illustration of the rTEDDI setup used when imaging the deer antler trabeculae bone sample. A large box section beam (5x5mm) geometry was used and a $2\theta$-angle of 13°.

d-spacing range of the bone sample was unknown at the time of the experiment and
was assumed to be in the same range as for the polymer sample, an assumption that would prove erroneous. The 2θ-angle was consequently kept at 13°. Figure 4.25 shows a photograph of the sample, the obtained rTEDDI image and the energy dispersive diffraction map. The inter pixel variation seen in the rTEDDI image is due to the structural variation of the sample material. A diffraction pattern obtained from this sample and its position in the sample can be seen in figure 4.26. No single diffraction peak is clearly discernible on first inspection of the energy dispersive diffraction pattern. A verification of the obtained energy dispersive diffraction pattern for the bone sample was obtained on station 16.4 using a single point TEDDI measurement, and can be seen in figure 4.27. For the TEDDI measurement 4 distinct peaks are visible. The measured d-spacing values are stated in table 4.5. Due to the chosen 2θ-angle, in combination with the low beam flux at energies above 20keV, the d-spacing region measured by the rTEDDI measurement shown in figure 4.26 covers roughly 2 to 5Å (peak d1 and d2). The reflections of the lower d-spacing planes are suppressed due to the low beam flux at

\[ \text{Figure 4.25: (Left) Photograph of deer antler trabeculae bone sample. The sample was imaged using the large box section beam geometry. (Centre) False coloured, total intensity map. The inter pixel variation is due to the structural variation of the sample material. (Right) Maps of the energy dispersive diffraction patterns recorded for each pixel.} \]

\[ \text{Figure 4.26: One of the registered energy dispersive diffraction patterns and its corresponding positions in the sample. No distinct peaks are directly visible. The chosen 2θ-angle and the low flux at higher energies resulted in an incomplete diffraction pattern.} \]
Figure 4.27: Plot of single point TEDDI measurement of the polymer sample plotted as both a function of energy and d-spacing. The measurement showed four distinct peaks, enabling interpretation of the rTEDDI measurements spectral response.

<table>
<thead>
<tr>
<th>Peak</th>
<th>d-spacing (Å)</th>
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<tbody>
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<td>d1</td>
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<td>d2</td>
<td>2.8031</td>
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<tr>
<td>d3</td>
<td>2.2614</td>
</tr>
<tr>
<td>d4</td>
<td>1.9493</td>
</tr>
</tbody>
</table>

Table 4.5: Resulting d-spacing values obtained for the bone sample using single point TEDDI measurement.

Higher energies resulting in an incomplete diffraction pattern being collected. Although it was the actual photon flux at higher energies that limited collection of low d-spacing diffraction peaks in this case, the same scenario would occur on a station with higher flux at higher energy, due to the decreasing stopping power of Si with increasing energy. In spite the poor statistical quality and incomplete diffraction pattern, refinement using Topas, figure 4.28, allowed for identification of the sample as one of the hydroxyapatites, but no distinction could be made regarding which type. The refinement resulted in a=9.323(9)Å, c=7.425Å in space group P6₃/m.

Figure 4.28: (a) Deer antler trabeculae bone sample. (b) Resulting rTEDDI image with pixel used for refinement clearly marked.(c) A lattice parameter refinement of a deer antler trabeculae bone sample diffraction pattern. The refinement resulted in $a=9.323(9)\text{Å}$, $c=7.425\text{Å}$ in space group P6₃/m.
4.2.5 rTEDDI at station 16.3, SRS Daresbury

The initial rTEDDI measurements, although a success in proving the concept of the technique, yielded low statistical quality diffraction patterns obtained over a long time due to the low flux, low energy beamprofile of station 7.6. Figure 4.29 shows the relative flux profiles of station 7.6, SRS Daresbury, and beamline station 16.3, SRS Daresbury. Station 16.3 is a engineering strain scanning and high resolution single crystal diffraction station allowing for operation in both monochromatic and white beam mode. The station is situated on the centre of a 6 Tesla wiggler fan giving an available energy range of 5-80keV. Station 16.3 has a beam profile closely resembling the ones normally used for TEDDI measurements. The peak flux for station 7.6 is 20 times lower than the peak flux of station 16.3 and operates in a lower energy range. The 17 hour collection time and the poor statistical quality is therefore attributed to the lower flux of station 7.6, especially at energies equivalent to the measured higher energy diffraction peaks of the samples. Detector countrate limitations, as described in Chapter 6, will also have degraded the statistical quality of the measured diffraction patterns. To test the system under high flux, high energy conditions, as initially envisioned, the rTEDDI system was set up on station 16.3, in the same way as described in section 4.2. The beamprofile for both

![Station 7.6 and 16.3 Beam Flux](image)

**Figure 4.29:** Comparison of the beam flux profiles for stations 7.6 and 16.3 at SRS Daresbury as calculated by Dr. David Laundy (station scientist for both stations). The beam profile of station 16.3 is similar to those used for TEDDI imaging. The peak flux for station 7.6 is 20 times lower and operates at a lower energy range. The low energy/low flux profile is accredited for the long collection time (17 hours) and low statistical quality of the spectra. Both of these effects would be more significant in the energy region where the higher energy diffraction peaks occurs (>20keV).
station 7.6 and 16.3 can be seen in figure 4.29. Si was substituted by CZT as the active detector material to enable effective high energy X-ray detection. The detector housing was adapted for CZT as described in chapter 6 and the detector was cooled to 268K, a bias voltage of -350V was applied and a 50kHz clocking frequency (see section 6.1 for description) was used. The spectral response of the CZT detector had been verified using an Am-241 dial source prior to the experiment. The spectral response measured on station 16.3 by the rTEDDI system for the same Al-powder sample as described in subsection 4.2.1 can be seen in figure 4.30. The large box section beam geometry was applied. No clearly visible AI diffraction peaks where observed in the spectra. Imaging of the polymer sample described in subsection 4.2.3 (cross test mark) was also tested yielding the same spectral response (see figure 4.31) as for the Al powder sample. It should also be noted that the imaging capability of the system was not existing as is apparent from figure 4.31. In order to eliminate noise as the cause of the inability to observe diffraction patterns shielding of the collimator, detector housing and DAQ system was further improved using lead sheets, a straight through beam beam stop was used and a lead pipe was inserted around the incoming beam leading up to the sample position in order to reduce air scatter. A general decrease in registered events resulted from the increased shielding but the spectral response pattern retained the same shape as seen in figure 4.30 and 4.31. A large piece of lead was finally put in front of the collimator opening, at which point the number events observed by the detector became negligible, demonstrating that the cause of the spectral response was due to X-rays traveling through the collimator and into the detector.

The lack of imaging as well as the spectral response at station 16.3 is currently believed to be due to the lack of stopping power of the tungsten collimator at energies closely

![Figure 4.30: rTEDDI imaging results obtained for the Al powder sample on station 16.3. SRS Daresbury. (Left) Spectral response registered for the Al-powder sample. No distinguishable Al diffraction is visible. This spectral plot is known as an Al-plot and is the spectral response of all the individual pixel spectral responses added together. (Right) rTEDDI image of the Al powder sample. The sample was not covering the entire imaging area of the detector.](image-url)
Figure 4.31: rTEDDI imaging results obtained for the Polymer, nylon-6, sample (circular test marks) on station 16.3. SRS Daresbury. (Left) Spectral response registered for the Polymer sample (All-plot). No distinguishable Polymer diffraction is visible. The response is almost perfectly the same as registered for the Al powder sample. (Right) rTEDDI image of the Polymer sample. The imaging capability of the rTEDDI imaging system on a high energy/ high flux station was not observed. The lack of imaging as well as the spectral response at station 16.3 is currently believed to be due to the lack of stopping power of the tungsten collimator at energies closely leading up to the absorption peak of tungsten, described in chapter 7, in combination with the ERD detectors inability to handle large countrates, described in chapter 6. These two effects explains the breakdown of the rTEDDI system when moving to a higher flux/ higher energy beamprofile synchrotron station.

4.3 Summary

In this chapter the building, and subsequent testing, leading to a proof of concept for the rTEDDI system has been described. Using a Si detector four different samples where imaged on the low flux/low energy synchrotron station 7.6 at SRS Daresbury. Two different geometrical setups where used. The imaging capability, best shown by the polymer samples, as well as the ability to extract an energy dispersive diffraction pattern for each pixel, corresponding to a spatial region of the sample, was demonstrated. Peak validation of the collected diffraction patterns where obtained using single point TEDDI measurements for all samples. Rietveld refinement using the Topas software gave values of $\chi^2=0.94$, $a=4.0497(38)$ for the Al powder sample. Refinement of the polymer sample resulted in $\chi^2=0.94$ with possible space groups P21, P21/m or P21/c, $a=7.058(50)\,\text{Å}$, $b=10.679(36)\,\text{Å}$, $c=17.839(15)\,\text{Å}$ and $\beta=68.4(21)^0$, corresponding to the structure of nylon-6. The samples were verified as Nylon6 using FTIR measurements. A lattice parameter accuracy in the second decimal point after refinement where found
for the Al powder, Cu foil and polymer sheets. Although this accuracy is not sufficient to enable micro strain scanning, where an accuracy in the fourth decimal would be needed [116] [117], it shows the promise of significantly improving the form of strain scanning described by Korsunsky et al. [117] with technological improvements. Further examples of point by point scanning TEDDI has been described by Harding et al. [17].

Partial diffraction pattern collection of the deer antler trabeculae bone sample was obtained and enabled identification as one of the hydroxyapatetites but no further distinctions could be made. Identification of fine structural changes in tissue is believed to fall outside the capability of the rTEDDI system. In vivo identification of diseased tissue, when leading to altered scattering signatures as described for certain types of cancer [118] [119], is anticipated as a main use of the technique. For tissue biopsy samples more advanced studies, such as the once described by Geraki et al. [120] [119] could be made, allowing for adipose and fibrous sample composition to be determined as well as quantification of elemental concentrations.

The energy dispersive diffraction patterns collected for each of the samples where all of poor statistical quality due to the relative low flux of the synchrotron station. Only a limited number of diffraction peaks where detected for each sample, ultimately due to the limited energy range detectable when using Si as the active detector material. The combination of poor statistical quality and low number of discernible peaks in the diffraction patterns results in a decreased lattice parameter accuracy obtainable from rietveld refinement and consequently limits the usability of the rTEDDI imaging technique. By substituting Si with CZT the available energy range of the detector would be significantly increased. This would translate into a larger number of peaks detected in a sample diffraction pattern increasing the ability to determine the lattice parameters. The larger stopping power of CZT would also increase the number of events detected increasing the statistical quality of the diffraction patterns.

The rTEDDI system was tested on a high flux/high energy station, station 16.3 SRS Daresbury, using CZT as the active detector material. The hopes where to increase the statistical quality of sample diffraction patterns as well as increasing the energy range over which the patterns were collected. A strong reduction in imaging capability and no discernible diffraction peaks in the spectral response where observed. The lack of imaging as well as the spectral response at station 16.3 is currently believed to be due to the lack of stopping power of the tungsten collimator at energies closely leading up to the absorption peak of tungsten, described in chapter 7, in combination with the ERD detectors inability to handle large countrates, described in chapter 6. As a consequence a new detector ASIC and DAQ system as well as new collimator array structures have been developed and tested. This work is described in chapter 7 and 6 respectively.
Chapter 5

TEDDI imaging using a single element CZT detector

A TEDDI setup, using a single element CZT detector, was set up at station I15, Diamond Light Source (DLS), in order to test samples of interest for the energy region envisioned for the rTEDDI system when utilizing a fully operational pixelated energy resolving CZT detector. This need is due to the vast majority of samples in the fields of security, medicine, oil+gas exploration or other applications have sample sizes in the cm range rather then the mm range which requires access to high energy X-rays resulting in a need for a detector with greater stopping power. As examples a friction stir welded Ti6246 sample as well as a spark plasma sintered ceramic sample was investigated using the TEDDI technique in the 30-80keV region. A description of the experimental setup is followed by the specifics and findings for the friction stir welded Ti6246 sample investigation and the spark plasma sintered ceramic sample investigation, and the chapter is concluded by a summary of the findings. The experimental work underlying the results in this chapter was done in co-operation with Prof. Robert. J. Cernik and Dr. Chris Martin, University of Manchester; Dr. Simon Jacques and Mr. Olivier Lazzari, University College London/Birckbeck and Prof. Alexander Korsunsky, Mr. Jonathan Belnoue and Terry Jun, University of Oxford.

5.1 The high energy TEDDI experimental setup.

A TEDDI setup, principally depicted in figure 2.1, using a single element CZT detector was constructed on beamline I15 at DLS in order to TEDDI image samples in the energy range 30-80keV. Station I15 uses multiple wigglers allowing for a user defined field value with a peak design field of 3.5T. For the durations of the measurements described in
this chapter the field was kept at 1T. The beamprofile of station I15 with a 3.5T and 1T can be seen in figure 5.1 with the energy region of interest for the this TEDDI study shown. For the energy range of interest, 30-80keV, the beamprofile can be seen to vary very smoothly over one order of magnitude making the station ideally suited for high energy TEDDI and rTEDDI imaging.

The detector used was a commercial Amptek single element CZT detector using a 3x3x1mm CZT crystal as the detector crystal. The detector was positioned behind the collimator. The collimator consisted of a 750mm long, lead covered, brass tube with a set of huber slits on each end. Each of the huber slits where set to have an opening of 200x200µm. The sample was situated approximately 315mm from the front end of the collimator resulting in an approximately 400x400µm sampling area created by the collimator field of view. The imaging voxel created by the intersection of the field of view of the collimator and the geometry of the beam was set in space while moving the sample in order to achieve TEDDI imaging.

5.2 TEDDI imaging of a linear friction welded Ti6246 sample.

Advanced Ti alloys are being widely used by the aerospace industry for high performance engine components that need to work under fail safe conditions. As a consequence the
need to be able to characterize, determine microstructural property relationships and study phase transformation that occur during processing is high in order to design microstructures that have excellent aim specific properties for this material [121].

Ti6246 is a Ti-alloy containing two crystallographic phases ($\alpha$, hexagonal and $\beta$, body-centered cubic) having a Ti-6Al-2Sn-4Zr-6Mo composition where the numbers refer to the wt% of each element with the reminder of the sample composed of Ti. Classifications with respect to the relative $\beta/\alpha$ ratios have been made sorting them into five classes $\alpha$, near $\alpha$, $\alpha+\beta$, near $\beta$ and $\beta$ alloys [122]. A quantification of the classes has however not been done. Recently alloys with greater $\beta$-phase fraction has been introduced in aero-engine components due to the observed improvements in creep strength, heat treatment response and higher operation temperatures [121] [123] [124] making it necessary to quantify the strain contribution of the $\beta$-phase. Earlier investigations using electron microscopy and X-ray diffraction has shown a 25:75 $\beta/\alpha$ ratio for the Ti6246 alloy [121].

The Ti alloy plates used in this TEDDI investigation where received as machined blocks with an oxygen content between 700-1200 ppm. This alloy is normally forged in the $\beta$-phase at 1213±15 K [122], followed by annealing at the $\alpha+\beta$ phase field between 1175-1195K for 1-2hrs, and then again at approximately 868K for 8 hours. This treatment is developed to attempt to create a microstructure with coarse (primary) and fine (secondary) $\alpha$ [121]. The sample used for this TEDDI investigation, supplied by Dr. Michael Presuss, consisted of two Ti6246 plates that had been linear friction welded together. The sample had been cut from a larger plate. In linear friction welding one part, in this case one of the Ti plates, is held stationary while the other is rubbed against it repeatedly in a linear fashion resulting in heating of the interface leading to softening of the material [124]. A significant axial pressure load is applied during the process leading to material (referred to as the flash) to be expelled from the weld region resulting in self-cleaning and removal of surface contaminants. Linear friction welding has been shown to introduce significant changes in microstructure and stress variation across the weld region in Ti [124]. The Ti6246 sample used in this study can be seen in figure 5.2 (Right) with the weld clearly visible in the middle and has the dimensions 1.8cmx1.3cmx2mm.

The large box section beam geometry setup, described for the polymer sample in section 4.2.3, but with a smaller incoming beam and a $\theta$-angle was used. Imaging was achieved by step scanning the sample in xy-direction using 400$\mu$m steps. Data from each voxel was collected for 10 seconds and the entire scan took approximately 13hrs.

The resulting TEDDI image can be seen in figure 5.2 (Left) and is plotted as a false coloured representation of the total number of counts collected at each voxel. The sample shape is faithfully represented in the TEDDI image and a correlation between the
external coloration of the optical micrograph and the TEDDI intensity map was observed. The sample had been mounted in Bakelite for preparation purposes, which was still clearly visible in the regions surrounding the weld flash. The Bakelite regions supporting the weld flash can be seen in yellow in the TEDDI image. A trend towards high countrate diffraction patterns, depicted as red pixels in the TEDDI intensity image, as well as high countrate region distributed throughout the sample.

Three types of diffraction patterns were observed for the voxels making up the TEDDI image and can be seen in figure 5.3. The diffraction patterns in figure 5.3 a)-c) originate from the designated voxels indicated by the arrows. The diffraction pattern from group a) contains 8 fluorescence/escape peaks up to a reconstructed 2θ-angle of 20° which are also present in the low angle spectra of the other two diffraction pattern groups. The 8 fluorescence/escape peaks correspond to Pb Lβ; Zr Kα; Mo Kα; Zr Kβ; Mo Kβ and two Cd escape peaks from the two main intensity diffraction peaks. For diffraction pattern a) this is followed by a series of strong narrow diffraction peaks corresponding to the α and β phases of Ti6246. The diffraction patterns from group c) have much broader peaks compared to that observed for diffraction patterns in group a), with a very broad main peak encompassing the α(010),(002),(011) and β(011) reflections. The c) diffraction pattern group is typical of the diffraction patterns observed in the weld region of the sample. Diffraction pattern b) was identified as diffraction from the Bakelite.

The high countrate voxels mentioned earlier, shown as red voxels in the TEDDI image, typical of voxels on the weld line and distributed throughout the sample, are all of the broad peak diffraction pattern type b). Prior knowledge of the material system would
Figure 5.3: Expanded view of the TEDDI image of the sample. The areas used for closer investigations in figure 5.6 and figure 5.7 are indicated by white boxes. On the right hand side the three distinctive diffraction types observed (a,b and c) are expanded with an arrow showing their original location. The 4 pink circles show the locations of the pixels used to plot the 3 spectra in figure 5.5.
suggest the increased peak width is caused by localized strain in the material with regions of built in strain being indicative to Ti6246. A direct comparison between the diffraction peak width increase and the observed peak width increase of the fluorescence peaks show a 100% correlation and can be seen by the two overlaid Rietveld refined pixels in figure 5.4. The increase in fluorescence line peak width for voxels located in the weld line, high countrate and matrix region can be seen in figure 5.5 and for each pattern the change in fluorescence line width was matched with an similar change in diffraction pattern peak width. The increase in peak width of the fluorescence lines would suggest pressure broadening of spectral lines due to built in lattice strain [125] [126] [127]. This would further suggest the increased diffraction peak width was caused by strain in material.

The EDD pattern from every voxel were refined using the known starting structures of for the Ti6426 α and β phase. The range and statistical quality of the diffraction patterns where not of high enough quality to refine atomic coordinates or microstructural properties. Cell parameters, preferred orientation directions and trends in particle sizes could be determined for all cases.

Figure 5.6 shows ten refined diffraction patterns, five originating from column 24, row a-e, and five from column 20, row a-e. The positions of the of the corresponding voxels in the TEDDI image, all around the weld line, is shown in figure 5.3. The variation seen between spectra labeled a,b,d or e are mainly attributed to preferred orientation of
Figure 5.5: The fluorescence peaks at different parts of the sample showing how fluorescence peaks broaden with increasing strain. The locations of the pixels plotted can be seen in figure 5.3 indicated by pink circles where the highly strained response is furthest to the left and the matrix response furthest to the right.

the sample. The spectra labeled c, on the weld line, were different from the other observed spectra having broader peak widths. The data was insufficient to separate out a particle size or strain broadening component however investigations of the sample using Electron Back Scattered Diffraction (EBSD) indicated particle sizes between 500nm and 1\(\mu\)m around the weld line and larger, 10-100\(\mu\)m, elsewhere. For all spectra both the \(\alpha\) and \(\beta\) phase was observed in the diffraction pattern due to a larger voxel than particle size. Both these results would suggest that particle size broadening would not be a dominant effect. The values outputted from the rietveld refinement for the 10 voxels seen in figure 5.6 are shown in figure 5.7 (Top), column 20, and figure 5.7 (Bottom), column 24, respectively. A significant and non-systematic variations in lattice parameter and volume of both phases across the sample was observed. The goodness of fit was acceptable in all cases. With the exception of the weld line the ratio of \(\alpha\) to \(\beta\) was 25\%:75\% to within experimental error which is consistent with previous studies [121]. The variation of the lattice parameters in between voxels in the same column can be seen in figure 5.8 for five almost complete columns. The colour coding of the plotted lines correspond to the columns seen in the TEDDI image, figure 5.2, where red correspond to column 12; black column 16; blue column 20; purple 24 and green column 29. The weld region is indicated in figure 5.8 by the gray shaded area. A significant change in lattice parameter is seen at the weld line for all columns and is attributed to strain broadening. In addition to the expected shifts at the weld line random, non-correlated regions of shifts in lattice parameters are observed throughout the bulk of the material,
Figure 5.6: Five Rietveld refinement output scans from column 20 and column 24 in the region of the weld. Differences between 20a and 20b are due to preferred orientation. Similarly between 20d and 20e and 24a, 24b, 24d and 24e. The assumed strain effects observed across the weld region in 20c and 24c manifest themselves in the form of peak broadening. The large peak visible is an amalgamation of the (010), (002), (011) and (011) reflections. Preferred orientation has been modified by spherical harmonics.
TEDDI imaging using a high Z detector

Figure 5.7: (Top) Parameters obtained from Rietveld refinement on voxels found in column 20, row a-e in figure 5.3. (Bottom) Parameters obtained from Rietveld refinement on voxels found in column 24, row a-e in figure 5.3.

The investigations using TEDDI imaging of the Ti6246 linear friction welded sample has faithfully imaged the sample; shown preferred orientation directions and rough trends in particle size. It is from the findings believed that the observed variation in diffraction peak width, along the weld line and regionally and non systematically throughout the bulk of the material, is due to regions of strain in the material which is also shown in the refinement output.
Figure 5.8: The lattice parameter variations between pixels along five complete lines. The red line correspond to column 12; black column 16; blue column 20; purple 24 and green column 29. There is a large change at the weld line as expected but there are equally large localized variations which are non-systematic.

5.3 TEDDI imaging of a spark sintered Al$_2$O$_3$ ceramic sample.

Sintering is a technique that goes back thousands of years with ancient Mesopotamian civilizations utilizing it as far back as 6000 BC [128] to create bricks from clay. One way of activating the sintering process in a material is by the use of electrical current. This method of activating sintering goes back to the early 1900’s, with patents issued as early as 1933. Although the use of electric current to activate sintering has a long history recent times have seen a renewed interest due to the commercially available devices that has come on to the market. One of the techniques that has been commercialized is the Spark Plasma Sintering (SPS) technique [129]. A schematical setup of the SPS
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Figure 5.9: Schematical representation of the SPS process. A high pulsed DC current in combination with uniaxial pressure is used to consolidate powders through the creation of ‘necks’ between particle surfaces [130].

The SPS technique can be seen in figure 5.9. The SPS technique utilizes pulsed high DC current in combination with uniaxial pressure in order to consolidate powders. To a large extent the physical processes active in SPS sintering is not completely understood and the creation of a plasma state in the material during sintering has never been verified [130]. A first description of the sintering process when using the SPS technique was done by M. Tokita [129]. As a spark discharge appears between the particles of a material, a local high temperature state is momentarily generated. This causes evaporation and melting of the powder surfaces and ‘necks’ are formed between particles binding them together. A high degree of success has been achieved using the SPS technique including cleaner grain boundaries in sintered ceramic materials, a remarkable increase in superplasticity of ceramics, higher permittivity in ferroelectrics, improved magnetic properties, improved bonding quality, improved thermoelectric properties, and reduced impurity segregation at grain boundaries. For a fuller description of the material advantages obtained when using the SPS technique and full referencing please see [130]. One of the materials that have shown promise when manufactured by the SPS technique is Al₂O₃, a ceramic that crystallises in a rhombohedral form and is used as a armor material [131].
Two SPS manufactured pieces of Al$_2$O$_3$, referred to as sample#7820 and sample3, was investigated using the TEDDI setup in the 30-80keV range. The top surface of the ceramic samples were set facing the incoming beam and the beamsize was set to 200x200µm. The samples were step scanned along the beam using 200µm steps, creating a depth profiling of the sample, starting at the top surface and ending at the bottom surface of the SPS samples.

Rietveld refinement of the measured data for two lines of sample#7820 and one line from sample3 can be seen in figure 5.10. In the Rietveld refinement plots of the TEDDI imaged lines, figure 5.10 a)-c), the observed and fitted diffraction pattern for all of the voxels comprising the line can be seen (top). The gray line in the middle of the image shows the error between the calculated and observed diffraction patterns. At the bottom the observed reconstituted angle for all the reflections are shown for all of the measured voxels, effectively showing the d-spacing variation as a function of sample depth. For all of the Rietveld refinement plots a number of the diffraction patterns are seen to have peaks of substantially lower intensity than average. During the scan the imaging voxel was initially placed outside of the sample. The imaging voxel was subsequently scanned through the sample and the scan was stopped with the voxel positioned outside of the sample on the other end. The lower intensity diffraction patterns where observed when moving in and out of the sample, when the imaging voxel was only partially inside the sample. For the reconstituted d-spacing angles for the different voxels a clear trend with higher observed angles at the edge of the sample is seen for all three lines, in both samples. This effect is also seen in the plot of the refined unit cell size a as a function of position in the sample, figure 5.10 d), f) and h), where a decrease in a parameter is seen at the edged for all scans. The a parameter varied across the sample for sample#7820 line 1 from 5.100-5.130Å. The observed variation for sample#7820 line 2 and sample3 line 1 was 5.102-5.135Å, and 5.098-5.113Å, respectively. The lower statistics of the edge voxels, described earlier, was considered as a possible reason for the shift of the refined a-parameter at these locations. The statistical quality was however believed to be sufficiently high to not be causing a problem even at the edges. The consistent trend towards higher constituted angles at the edge voxels for all measured lines would also lend support to the observed effect being caused by an actual a parameter shift. No clearly observable variation of the rhombohedral alpha angle as a function of sample depth, figure 5.10 e), g) and i), could be observed.

The TEDDI investigations of two SPS Al$_2$O$_3$ ceramic samples showed a variation in the unit cell size with lowered a parameter value at the edges of the sample, the position where the electrodes were placed during the sintering process. An interpretation of the cause of the observed effect could be regionally higher temperature or current densities during the SPS process, as has been observed for non-conducting Cu and conducting Al samples previously [130].
Figure 5.10: Rietveld refinement plots (a-c) (as described in chapter 2) fitted for the angular range of 22° to 74°; visualization of the refined rhombohedral unit cell size parameter $a$ (d, f and h) as a function of sample depth (200µm stepsize) and visualization of the refined rhombohedral alpha angle parameter (e, g and i) as a function of sample depth (200µm stepsize) for three scans in two SPS manufactured Al$_2$O$_3$ ceramic samples. The data was collected using the TEDDI technique. A clear reduction in unit cell size was observed at the top and bottom of the samples for all 3 line scans. No clear trend was observed for the rhombohedral angle variation as a function of position in sample.
5.4 Summary

A TEDDI setup using a single element CZT detector was constructed at beamstation I15, DLS, in order to investigate two samples in the energy region 30-80keV. This high energy region, which is envisioned to be used for the rTEDDI system, enables diffraction imaging investigations of high density/thickness samples which would be of strong interest for industry by allowing non-destructive evaluation of, for example, whole engineered components and friction stir welds. Two sample materials where investigated under these high energy conditions, a friction stir welded Ti6246 sample and two SPS manufactured Al$_2$O$_3$ ceramic sample.

The friction stir welded Ti6246 sample was imaged using the large box section beam geometry. The shape of the sample was faithfully reproduced by the TEDDI image and a correlation between the external coloration of the optical micrograph and the TEDDI total intensity map was observed. A trend of high total intensity countrate voxels along the weld line was seen as well as random regions of high intensity voxels. Three distinct classifications of diffraction patterns where seen for the voxels imaged. The high countrate voxels corresponded to diffraction patterns indicative of high diffraction peak peak widths. Prior knowledge of the sample would indicate that the increased peak width was in fact due to localized strain in the material. An increase in the peak width of the low energy fluorescence peaks, 100% correlated to the increased diffraction peak peak width, would support this suggestion [125] [126] [127]. The α and β phase of the material was present in all voxels, indicating a particle size smaller then the imaging voxel dimension. Measurements done using EBSD showed particle sizes in the range of 10 to 100µm in the bulk material. Both these results would suggest the observed peak width increase was not caused by particle size broadening but was in fact a strain effect. The two other classes of diffraction patterns observed for the voxels imaged differed from the high peak width patterns by, one having lower peak width diffraction peaks, and the other having the characteristic spectra corresponding to diffraction from Bakelite (used during the friction stir welding process). For the low peak width class of diffraction patterns 8 low reconstituted 2θ-angle fluorescence/escape peaks where observed followed by a series of strong diffraction peaks corresponding to the α and β phases of the material. The diffraction patterns for all of the imaged voxels where Rietveld refined. Cell parameters, preferred orientation directions and trends in particle sizes could be determined for all cases when using the TEDDI technique while the statistical quality and range of the diffraction patterns where to low to allow refinement of atomic coordinates and microstructural properties. The refined data showed β:α ratios of 25%:75%, within experimental error and weld region neglected, throughout the material which is in agreement with previous studies [121]. Variation in refined lattice parameters showed significant change along the weld line and in non systematic regions of the material in
agreement with the high countrate voxels observed in the TEDDI image.

The SPS manufactured Al$_2$O$_3$ ceramic samples where TEDDI imaged in lines going through the sample, creating a sample depth profiling, by using a 200x200µm beam and step scanning the sample along the direction of the incoming beam. 3 lines where imaged, one in sample3 and two in sample#7820. Rietveld refinements of the measured data showed a systematic decrease in unit cell size at the top and bottom of sample for all three measured lines and both samples. No alpha angle variation as a function of position in sample was readily visible. Possible speculation to the cause of the observed effect could be regionally higher temperature or current densities during the SPS process, as has been observed for non-conducting (Al) and conducting (Cu) samples previously [130].
Chapter 6

Development of high Z detectors for rTEDDI imaging

At the start of this project the ERD2004 detector had been developed and tested for spectroscopic performance but had not been characterized or investigated to any depth. The ERD2004 detector using Si as the active detector material was successfully used to obtain the proof of concept for the rTEDDI imaging technique. This has been thoroughly described in chapter 4. In order to image thicker samples, such as jet engine turbine blades or friction stir welds, using the rTEDDI system high energy X-rays would have to be utilized. A wider detectable X-ray energy range was also desirable in order to increase the accuracy with which lattice parameters could be determined. The poor stopping power and quantum efficiency of Si at higher X-ray energies would prevent any useful observations. To overcome this obstacle Si was substituted by CZT as the active detector material of the detector. A new ASIC specifically designed for the rTEDDI imaging system was also developed within the framework of this project. The ASIC, known as the HEXITEC ASIC, was developed with the main aims being optimized energy resolution and small pixel pitch while using CZT as the active detector material. This chapter describes the effects found when substituting Si with CZT and work on optimization and characterization of the ERD2004 detector. The results found for the ERD2004 detector were used to improve the design of the HEXITEC detector. Initial findings on the first iteration of the HEXITEC detector are also described.

6.1 The ERD2004 detector.

The ERD2004 detector was developed by the detector development group at Rutherford Appleton Laboratory (RAL) [25]. Although spectroscopic performance using Si as the
active detector material was determined for the detector, no extensive characterization was performed on the detector. In order to increase the usability of the rTEDDI technique Si was substituted with CZT and limitations of the ASIC design were investigated. The results obtained were used to improve the design of the HEXITEC detector system. This sub-chapter describes the structure and read-out scheme for the ERD2004 detector; the complete detector system when using both Si and CZT as the active detector material and the developed software used to analyze the detected X-ray events. This is followed by results obtained for initial measurements of the ERD2004 detector response when substituting Si for CZT. This includes response to known radiation sources, inter pixel uniformity investigation and effects of varying parameter values. Investigations into effects resulting from the ASIC design and detector geometry, including countrate limitations and charge sharing correction, has also been done and is shown. The problems need to be solved in order make the ERD2004 detector useful for the rTEDDI technique.

6.1.1 The ERD2004 detector system

The ERD2004 detector is a pixelated energy resolving X-ray detector developed by the Rutherford Appleton Laboratory [25]. The active detector crystal is bonded to 16x16 array of pre-amplifiers, on a 300µm pixel pitch, known as the MAC04. The MAC04 circuit layout and a single amplifier schematic diagram can be seen in figure 6.1. The pre-amplifier is a single ended cascode configuration with switched reset. The pre-amplifier can be used in two gain setting modes, high gain and low gain. The high gain mode is designed for low leakage detector crystals and uses a 25fF feedback capacitance. The low gain mode can be used for high leakage detectors and uses a 250fF feedback capacitance. The MAC04 is in turn wire bonded to two 1x128 arrays of shaping, peakhold and comparator circuits, known as the SHC04. The SHC04 circuit layout and a
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Figure 6.2: (Left) SHC04 integrated circuit and (Right) schematic diagram of the SHC04 readout channel. The channel contains a shaper, peak hold and comparator circuit. The shaper has a shaping time of 4µs shaping time and a gain of 10. The channel is triggered for readout if the induced charge exceeds a user defined voltage in the comparator.

Figure 6.3: Photograph of the ERD2004 ASIC. The MAC04 is seen in the middle. The MAC04 is wire bonded to two SHC04, one on each side. Each pixel is read out by a single MAC04-SHC04 channel.

single SCH04 channel schematic diagram can be seen in figure 6.2. The SHC04 channel contain a CR-RC shaper, with a 4µs shaping time and a gain of 10, a peak-hold and comparator circuit. Each pixel has a corresponding MAC04-SCH04 channel dedicated to its event detection and readout. The MAC04-SHC04 ASIC can be seen in figure 6.3. A channel is triggered for readout if the charge induced from a pixel exceeds a user defined voltage in the comparator circuit. This flags a register which activates a multiplexer which reads out the event and activates the local reset. Two reset schemes are implemented in the ERD2004 ASIC, the global reset and the local reset. The global reset is used to reset the MAC04 in order to prevent the pre-amplifiers to be saturated due to leakage current. The local reset is used to reset single channels of the SHC04
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after an event has been read out. The active period, $T$, of the global reset, during which events can be registered is given by:

$$T = \frac{2000}{\text{freq}} \times (\text{number of reads per frame})$$

(6.1)

where $\text{freq}$ is the clocking frequency of the detector system. Both $\text{freq}$ and (number of reads per frame) are user defined variables set in the detector Graphical User Interface (GUI).

The active detector material, i.e a 4.8mm x 4.8mm x 300$\mu$m Si crystal or a 4.8 x 4.8 x 2mm CZT crystal, is bump bonded to the ERD2004 ASIC. Bump bonding is realized by the deposition of gold balls, using a gold ball bonder, one on each bond pad of the MAC04. A conductive silver epoxy dot is deposited on each of the pixels of the pixelated metalization of the active detector crystal, as can be seen in figure 6.4. The

![Figure 6.4: Image showing the deposited silver epoxy dots, used in the bump bonding process, on a the pixelated side of a CZT crystal.](image)

MAC04 and the crystal is carefully aligned and the gold studs are mechanically pushed into the silver epoxy dots, creating the bonds. The process is illustrated in figure 6.5. Currently all bump bonding is done at the Rutherford Appleton Laboratory. Finished

![Figure 6.5: Illustration of the bump bonding process. The aligned gold balls (MAC04) and the silver epoxy dots (detector crystal) are mechanically pushed together, thereby creating the bonds.](image)
ERD2004 detectors bump bonded with Si and CZT crystals can be seen in figure 6.6. The data from the ERD2004 detector is read out using a DASH-E readout logic card to which it is connected via a 40 pin connector as can be seen in figure 6.7. The ERD2004 detector can be connected to the DASH-E card directly using the 40 pin socket or via a connection cable as shown in figure 6.8. The DASH-E readout logic card is in turn read and controlled from a LabView [132] detector GUI running on a PC to which it is connected. The detector power and bias voltage sources are supplied via the DASH-E card. A schematic layout of the power supply connections needed to run the EDR2004 detector and the DASH-E card is shown in figure 6.9. Bias for the Si detector is achieved by connecting a number of batteries in series (36V). Bias for the CZT detector is achieved using a Keithley 248 High voltage supply [133] running in reversed bias mode ( -200V to -400V).

When running the detector system using Si as the active detector material the ERD2004 detector is simply secured in a bracket as can be seen figure 6.10 and connected to

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**Figure 6.6:** Photographs of ERD2004 detectors. The top image shows the ERD2004 bonded to a Si crystal, ERD2004 Si detector, and the bottom image shows bonding to a CZT crystal, ERD2004 acCZT detector.

**Figure 6.7:** Photograph of a ERD2004 ASIC with the 40 pin connection used to connect the detector to the DASH-E readout logic card clearly marked.
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Figure 6.8: Photograph of the DASH-E readout logic card. The card acts as an interface between the ERD2004 detector and the control PC where the LabView detector GUI is running. Biasing of the detector and powering of the detector and the DASH-E card is supplied through connections made on the DASH-E card.

Figure 6.9: Schematic diagram of the power supply connections for the DASH-E card. When biasing a ERD2004 Si detector a number of serially connected batteries are used. When biasing the ERD2004 CZT detector the biasing is done using a Keithley 248 High voltage supply.

the DASH-E card using a connection cable. When using CZT as the active detector material adaptations to the system in order to enable cooling and high voltage biasing was needed to ensure optimized performance of the detector. An Aluminium box with
a mylar window was constructed to house the ERD2004 detector. A pair of pyramid stacked peltier coolers where attached to one of the inside walls of the box to enable cooling of the internal environment. The peltier coolers where in turn cooled using a PC processor liquid cooling system in order to allow for further cooling of the system. This configuration allowed for cooling down to approximately -10 to -15 degrees Celsius dependent on the ambient room temperature. A copper cold finger, glued using a thermal glue to the back of the ERD2004 detector, was secured to a copper fixture on the Peltier coolers, enabling cooling and securing of the detector inside the aluminium box. The internal setup of the aluminium box can be seen in figure 6.11. A temperature sensor

![Figure 6.10: Detector bracket holding the ERD2004 Si detector. The ERD2004 detector is connected to the DASH-E card using a connection cable and a Pb cover is used to shield the detector from background radiation noise.](image)

![Figure 6.11: Internal structure of the Al box used to house the ERD2004 CZT detector. The design allows for controlled cooling in a dry air environment while applying a high voltage bias.](image)
was attached to the copper fixture and connected to an external temperature controller. The temperature controller monitored the temperature of the internal sensor and varied the current to the peltier coolers accordingly to obtain the specified temperature. A dry air environment was created with a bambi dry air compressor \cite{134}, before cooling to prevent the build up of condensation when reaching temperatures below 0°C. The results of cooling in a non dry air environment can be seen figure 6.12. The introduction of an

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure6.12.png}
\caption{Results of cooling the ERD2004 CZT detector below 0°C without a dry air environment. Supplying a dry air environment completely removed the condensation problem.}
\end{figure}

dry air environment completely removed the build up of condensation. An external view of the detector box and the setup used when using CZT as the active detector material can be seen in figure 6.13.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure6.13.png}
\caption{Picture of the setup used when using CZT as the active detector material.}
\end{figure}
6.1.2 Initial dial source measurements of the ERD2004 CZT detector

To test the effect of substituting Si with CZT as the active detector material a 4.8x4.8x2mm piece of eV PRODUCTS [135], now eV Microelectronics, CZT was bump bonded to a ERD2004 ASIC and tested in the Al housing described in subsection 6.1.1 using an Am-241 dial source. The setup can be seen in figure 6.14. The Am-241 dial source, seen in figure 6.14, is a sealed radiation source containing an active Am-241 alpha particle emitter. The alpha particles is irradiated onto selectable thin metal films which in turn radiate characteristic radiation specific for that metal. The selectable metal films and the resulting X-ray radiation energy can be seen in the table 6.1. During these measure-

![Experimental setup for X-ray flood field illumination using an Am-241 dial source. The detector is placed inside the Al housing and cooled to desired temperature in a dry air environment.](image)

<table>
<thead>
<tr>
<th>Metall</th>
<th>$K_\alpha$ (keV)</th>
<th>$K_\beta$ (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>8.04</td>
<td>8.91</td>
</tr>
<tr>
<td>Rb</td>
<td>13.37</td>
<td>14.97</td>
</tr>
<tr>
<td>Mo</td>
<td>17.44</td>
<td>19.63</td>
</tr>
<tr>
<td>Ag</td>
<td>22.10</td>
<td>24.99</td>
</tr>
<tr>
<td>Ba</td>
<td>32.06</td>
<td>36.55</td>
</tr>
<tr>
<td>Tb</td>
<td>44.23</td>
<td>50.65</td>
</tr>
</tbody>
</table>

Table 6.1: The $K_\alpha$ and $K_\beta$ fluorescence energies for the available thin metal films of the Am-241 dialsource.

ments the Cu source was never registered due to the relatively high low energy cut-off of the ERD2004 detector system. In order to investigate the effects of using CZT as the active detector material, and optimize its performance, the detector was exposed to the characteristic radiation from the different metal films while varying the detector temperature and bias voltage. The measurement scheme used can be seen in figure 6.15. The varied measurement times where used to compensate for the varied photon flux for the different metal films. Four main avenues of investigation was pursued. There where
spectral response to known radiation sources (Am-241 dial source); detector response variation with varied temperature; detector response variation with varied bias voltage and inter pixel uniformity.

**Spectral response to known radiation sources**

To test the spectral response of the ERD2004 CZT detector fluorescence spectra where collected for five of the metal films and compared to the response obtained for the same sources using a ERD2004 Si detector. For all these measurements the ERD2004 CZT detector was cooled to -50°C, a clocking frequency of 50kHz and a bias voltage of -350V was used. The spectral response obtained when exposing the ERD2004 Si detector (All plot, see AppendixA) and ERD2004 CZT detector (All plot and single pixel plot, AppendixA) to the Rb fluorescence radiation can be seen in figure 6.16. The All plots used in this section are plotted as a function of intensity vs. voltage while the single pixel spectra is plotted as intensity vs. energy. The pixel pedestal values have been removed from all plots unless stated otherwise. The Rb Kα and Kβ fluorescence lines are clearly visible in the ERD2004 Si All plot spectra. A comparison between the ERD2004 CZT All plot spectra and the single pixel spectra verifies the peaks visible in the All plot spectra as spectral peaks and not artifacts of the single pixel spectra addition. The Rb Kα peak is visible, (1), in the ERD2004 CZT detector All plot spectra. No Kβ peak is directly visible in the All plot. Three high energy peaks, (2)-(3), are present that are not present in the Si spectra. Approximate energy values for the observed peaks are stated in table 6.2, verifying peak (1) as the Rb Kα fluorescence line.

The spectral responses obtained when exposing the detector to Mo fluorescence can be seen in figure 6.17. The Si detector All plot response clearly shows the Mo Kα and Kβ fluorescence lines. The Kα ,(1), but not the Kβ peak is distinguishable in the CZT
Figure 6.16: (Left) All plot of the spectral response for the ERD2004 Si detector using the Rb target on the Am-241 dial source. (Centre) All plot of the spectral response for the ERD2004 CZT detector using the Rb target on the Am-241 dial source. (Left) Single pixel plot for the spectral response of the ERD2004 Si detector using the Rb target on the Am-241 dial source. The correlation between peaks in the CZT detector All plot and single pixel spectra shows that the All plot spectral peaks are not an effect of adding the single pixel spectra. The $K_{\alpha}$ peak and three higher energy peaks are visible in the ERD2004 CZT detector spectra.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>13</td>
</tr>
<tr>
<td>2</td>
<td>22</td>
</tr>
<tr>
<td>3</td>
<td>50</td>
</tr>
<tr>
<td>4</td>
<td>63</td>
</tr>
</tbody>
</table>

Table 6.2: Approximate energy values of the spectral peaks observed when exposing the ERD2004 CZT detector to Rb fluorescence. The peak numbers corresponds to those stated in the central plot of figure 6.16.

Figure 6.17: (Left) All plot of the spectral response for the ERD2004 Si detector using the Mo target on the Am-241 dial source. (Centre) All plot of the spectral response for the ERD2004 CZT detector using the Mo target on the Am-241 dial source. (Left) Single pixel plot for the spectral response of the ERD2004 Si detector using the Mo target on the Am-241 dial source. The $K_{\alpha}$ peak and two higher energy peaks are visible in the ERD2004 CZT detector spectra. Both $K_{\alpha}$ and $K_{\beta}$ are visible in the CZT detector single pixel plot. Two high energy peaks, (2) and (3), are visible in the CZT detector spectra that can not
be seen in the Si detector spectra. The approximate energy value of the observed energy peaks for the CZT detector can be seen in table 6.3.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>17</td>
</tr>
<tr>
<td>2</td>
<td>49</td>
</tr>
<tr>
<td>3</td>
<td>62</td>
</tr>
</tbody>
</table>

Table 6.3: Approximate energy values of the spectral peaks observed when exposing the ERD2004 CZT detector to Mo fluorescence. The peak numbers corresponds to those stated in the central plot of figure 6.17.

The spectral responses obtained when exposing the detector to Ag fluorescence can be seen in figure 6.18. The Si detector All plot response clearly shows the Ag K\(\alpha\) and K\(\beta\) fluorescence lines. Only the K\(\alpha\) peak, (2), is clearly visible in the CZT detector All plot while both K\(\alpha\) and K\(\beta\) are visible in the single pixel plot. The two high energy peaks, (3) and (4), are still present in the spectra. At this energy a low energy peak (1) is also starting to emerge in the CZT detector spectra. The approximate energy values of the observed peaks are stated in table 6.4.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>13</td>
</tr>
<tr>
<td>2</td>
<td>22</td>
</tr>
<tr>
<td>3</td>
<td>49</td>
</tr>
<tr>
<td>4</td>
<td>60</td>
</tr>
</tbody>
</table>

Table 6.4: Approximate energy values of the spectral peaks observed when exposing the ERD2004 CZT detector to Ag fluorescence. The peak numbers corresponds to those stated in the central plot of figure 6.18.
The spectral responses obtained when exposing the detector to Ba fluorescence can be seen in figure 6.19. The Si detector All plot response show both the Ba K\(\alpha\) and K\(\beta\) fluorescence lines. The CZT detector All plot clearly show the K\(\alpha\) peak, (3), and an indication of the K\(\beta\) peak, (4), is starting to emerge. Both K\(\alpha\) and K\(\beta\) are clearly visible in the single pixel plot. The two high energy peaks, (5) and (6), are still present. At the lower energy end of the spectra the peak two peaks, (1) and (2), are present. Peak (1) correspond to the peak (1) seen in the Ag fluorescence spectra and peak (2) correspond to peak (2) in the Rb fluorescence spectra. The approximate energy values of the observed peaks are stated in table 6.5.

<table>
<thead>
<tr>
<th>Peak number</th>
<th>Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>11</td>
</tr>
<tr>
<td>2</td>
<td>23</td>
</tr>
<tr>
<td>3</td>
<td>32</td>
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<tr>
<td>4</td>
<td>37</td>
</tr>
<tr>
<td>5</td>
<td>48</td>
</tr>
<tr>
<td>6</td>
<td>62</td>
</tr>
</tbody>
</table>

Table 6.5: Approximate energy values of the spectral peaks observed when exposing the ERD2004 CZT detector to Ba fluorescence. The peak numbers corresponds to those stated in the central plot of figure 6.19.

The spectral responses obtained when exposing the detector to Tb fluorescence can be seen in figure 6.20. The Tb K\(\alpha\) and K\(\beta\) fluorescence lines are visible in both the Si and CZT ((3) and (4)) All plots. The observed CZT K\(\beta\) fluorescence line is believed to be a combination of the of the actual K\(\beta\) line and the observed line occurring around 50keV.
The high energy peak observed for all the measurements where present present for the Tb fluorescence as well, (5). The approximate energy values of the observed peaks are stated in table 6.6. The presence of high energy peaks for the CZT detector not visible for the Si detector is believed to be due to the superior detection efficiency of CZT at these energies. The spectral response obtained from the five different fluorescence sources using the ERD2004 CZT detector can be explained by the respective K\(\alpha\) and K\(\beta\) fluorescence lines in combination with 4 additional spectral lines occurring at approximately 13, 22, 50 and 60keV. Possible sources of the additional spectral lines where considered. The observed K\(\alpha\) and K\(\beta\) fluorescence lines for the different illuminated metals where accounted for and their values are stated in table 6.1. The radioactive material of the dial source is a piece of Am-241. The electromagnetic spectrum for Am-241 has been investigated \[136\] and the spectral lines, in keV, are stated in table 6.7. The
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<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>13.5 (keV)</td>
<td>17.3 (keV)</td>
<td>20.9 (keV)</td>
<td>26.3 (keV)</td>
<td>59.7 (keV)</td>
</tr>
</tbody>
</table>

Table 6.7: X-ray and γ-ray energies emitted by an Am-241 source for the energy range 0-60keV. The lower energy peaks (Np Lα X-ray to Am-241 γ-ray 2) are a result of emission from Np-237, the daughter product of Am-241.

lower energy peaks (Np Lα X-ray to Am-241 γ-ray 2) are usually referred to as the Np daughters, since most of them are an effect of Am-241 decaying into the daughter product Np-237. The Kα and Kβ fluorescence lines for the detector material constituents (Cd, Zn and Te) are stated in table 6.8 and the resulting escape peaks registered when illuminating the CZT detector with radiation from the different metal films can be seen in table 6.9.

<table>
<thead>
<tr>
<th>Cd</th>
<th>Zn</th>
<th>Te</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kα  (keV)</td>
<td>23.17</td>
<td>8.64</td>
</tr>
<tr>
<td>Kβ  (keV)</td>
<td>26.09</td>
<td>9.57</td>
</tr>
</tbody>
</table>

Table 6.8: Kα and Kβ fluorescence energies for Cd, Zn and Te. Observed escape peaks in the spectral response will correspond to these values.

<table>
<thead>
<tr>
<th>All values in keV</th>
<th>Rb</th>
<th>Mo</th>
<th>Ag</th>
<th>Ba</th>
<th>Tb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>8.89</td>
<td>21.06</td>
</tr>
<tr>
<td>Zn</td>
<td>4.73</td>
<td>8.8</td>
<td>13.46</td>
<td>23.42</td>
<td>35.59</td>
</tr>
<tr>
<td>Te</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>4.66</td>
<td>16.83</td>
</tr>
</tbody>
</table>

Table 6.9: Energy values where detector escape peaks would occur for the different fluorescence targets available from the Am-241 dial source.

The resulting possible explanations for the observed peaks when exposing the CZT detector to different Am-241 dial source metal fluorescence can be seen in figure 6.21. The four additional spectral lines are consequently accredited to effects stemming from the higher detection efficiencies and atomic species of the constituent atoms in the detector material (Cd, Zn and Te). The lowest of the four peaks, occurring roughly at 13 keV is accredited to a combination of low energy tailing of the spectra (due to the increased leakage current of CZT) and Np L-α radiation originating from the Am-241 alpha emitter inside the dial source. The peak centered at approximately 22keV is accredited to a combination of Np daughters (Np L-β, Np L-γ, Np γ-ray 2) and escape peaks (Cd and Te) originating from the unknown peak at approximately 50keV. The origin of the peak at 50 keV is currently unknown but it has been suggested that backscattered columb radiation from the source could be an explanation. The fourth peak is accredited to the main Am-241 γ-ray (γ-ray 1) having an energy of 59.7keV. To verify that no overseen processes where the actual cause of any of the metal flouresence peaks a spectra was collected using a pure Am-241 source at Rutherford Appleton Laboratory. The resulting spectra and a comparison with an Am-241 reference spectra obtained from
Figure 6.21: (Left) All plot spectral response for the different fluorescence energies of the Am-241 dial source. Right approximate energy of the observed peaks and possible explanation. The discrepancy between the Si and CZT spectral response can be explained (disregarding the 50keV peak) when taking into account the higher detection efficiency and the energy levels of the constituent atomic species of the CZT detector.
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Figure 6.22: (Left) All plot spectral response of the ERD2004 CZT detector to a pure Am-241 source. (Right) Reference Am-241 spectra obtained from Amptek’s web page [137]. The reference spectra was collected using a single element CdTe detector. A high degree of correlation between the ERD2004 CZT detector spectra and the reference spectra was found.

Amptek [137] can be seen in figure 6.22. The collected spectra has been corrected for pedestal variation and charge sharing (discrimination, see section 6.1.4). The main Am-241 peak, the Cd and Te escape peaks as well as the Np-daughters are all clearly visible.

Detector response with varied temperature and bias voltage

The effects of varying the operational temperature of the ERD2004 CZT detector was tested by illuminating the detector using fluorescence from the Am-241 dial source (Rb, Mo, Ag, Ba and Tb) while varying the temperature. A clocking frequency of 50kHz and a bias voltage of -350V was applied. The spectral response variation with varied temperature while illuminating the detector with Tb fluorescence can be seen in figure 6.23. An improvement in both countrate and spectral response was observed when cooling the detector from 19°C to -5°C. The limited countrate at higher temperature was attributed to leakage current saturation of the pre-amplifiers during part of the collection.
time. Any improvement in response when cooling from -5°C to -10°C was unclear. The Al detector housing did not allow for cooling below -10°C and quantification of spectral performance of the pixelated CZT detector was severely hindered due to inter-pixel non-uniformities as discussed in the next sub-section. Investigations into planar detector charge transport properties of eV PRODUCTS CZT was carried out by Mathew Veale [109], University of Surrey/Rutherford Appleton Laboratory, to temperatures of -163°C indicating internal electric field build up at temperatures of -10°C and below adversely effecting the detector performance. It is therefore believed it is advantageous to stay above this temperature and allow cooling to 0 to -5°C.

The effect of varying the bias voltage of the CZT detector was tested by illuminating the detector using fluorescence from the Am-241 dial source while varying the bias voltage. A clocking frequency of 50kHz and a temperature of -5°C. was applied. The resulting spectral and spatial response can be seen in figure 6.24 when exposing the detector to Rb fluorescence at different temperatures. An increased spectral response was observed

![Image of figure 6.24]

Figure 6.24: ((Bottom) All plot of spectral response for the ERD2004 CZT detector when exposed to Rb fluorescence at different bias voltages. An increased spectral response was observed at higher bias voltages indicating a higher bias would be preferable. (Top) Pixel total intensity map at different bias voltages. At higher voltages non-responsive pixels appeared in the centre of the detector, an effect termed black hole effect. The countrate fell as a function of increased number of non-responsive pixels.

with increased bias voltage, as expected due to the resulting increase in charge carrier drift length, \( l \). The spatial response was found to suffer with increased bias voltage with a significant number of pixels becoming non responsive when changing the bias voltage from -350V to -450V. The effect was termed the black hole effect Spectrally the black
hole effect resulted in a lower peak count rate in the All plot. The temperature dependence of the black hole effect was investigated and the result is shown in figure 6.25. A decreased black hole effect was observed with increased temperature indicating that

![Figure 6.25: The black hole effect as a function of voltage at two different temperatures. An decrease in the severity of the black hole effect with applied bias voltage was observed at higher temperatures, indicating that the effect is not a result of increased leakage current.](image)

the effect was not caused by leakage current swamping the pre-amplifiers of the MAC04 circuit. The pixel pedestal values (operational offset from base value for the readout electronics) was investigated as a function of bias voltage and the result can be seen in figure 6.26. An increase in the number of high pedestal value pixels for the -450V biased detector was observed when comparing it to the -450V biased detector. A correlation between the high pedestal value pixels and the non-responsive pixels where found at both bias voltages. The high pedestal value leading to non-responsive pixels indicated a bias voltage dependent effect driving the readout channels of the effected pixels high for all stages of the readout cycle. An ERD2004 CZT detector from the same batch as the one shown above was sent to KSI for investigation of bump bonding quality using acoustic microscopy. The result can be seen in figure 6.27. Delamination and missing bump bonds where found to be present. The bump bonding quality found is believed to be a possible reason for the observed black hole effect. Improvement in detector manufacturing (i.e. in house bump bonding at Rutherford Appleton Laboratory) eradicated the black hole problem in later detector batches. This is shown in figure 6.28 where the spatial response at -450V for a detector from a later batch is compared to the detector displaying the black hole effect.
Figure 6.26: A correlation between high pedestal value and the black hole effect was found. An increase in the number of pixels having high pedestal values increased with increased bias voltage. The high pedestal pixels where the same as the non responsive pixels.

Figure 6.27: Acoustic microscopy image of a ERD2004 CZT detector from the same batch as the detector exhibiting the black hole effect. Poor bonding in the form of delamination and missing bump bonds were observed. The poor bonding quality is believed to be the cause of the black hole effect.
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Figure 6.28: Total intensity maps of two ERD2004 CZT detector, one from the ‘black hole effect batch’ (Cold16) and one from a batch bonded after improvement to the bonding process had been implemented (CA2). The black hole effect was no longer present at -450V bias voltages.

Inter pixel spectral variation

The spectral uniformity between pixels was investigated using Am-241 dial source flood field irradiation. The spectral response for the different pixels as found when illumination the detector using the Ba target can be seen in figure 6.29. A high degree of inter pixel non-uniformity was observed. In order to try and quantize the inter-pixel uniformity response as a function of energy the detector was energy calibrated using the five fluorescence sources of the Am-241 dial source and the teddi_v1 program (see Appendix A). A voltage to energy linearity plot for each pixel was subsequently produced as can be seen for the top half of the detector in figure 6.30. A pixel quality definition was subsequently done, defined a pixel as good or bad, using the deviation of the measured fluorescence points from linearity. From the pixels labeled as ‘bad pixels’ using the described definition, three sub-groups of ‘bad pixels’ where found on closer inspection. Typical observe linearity responses for each of the three sub-groups can be seen as the the bottom right corner plot in figure 6.31, figure 6.32 and figure 6.32 respectively. The response at different energies for a group one pixel can be seen in figure 6.31. The group one pixel had an non responsive behavior regardless of energy and was identified by a close to vertically linear calibration linearity. The response at different energies for a group two pixel can be seen in figure 6.32. A group two bad pixel was characterized by an energy response that was present but poor, resulting in a high linear least squares fit error. One of the measured calibration points could for some of the group two pixels be
Figure 6.29: Spectral response pixel map as obtained when exposing the ERD2004 CZT detector to Tb fluorescence from the Am-dial source. A high degree of inter-pixel variation is clearly visible.

Figure 6.30: Calibration linearity pixel map for the top SHC04 of the ERD2004 CZT detector as created by the teddi_v1 program in Matlab. The voltage value specified as the Kα peak for each pixel and fluorescence source corresponds to the the highest intensity value found in the single pixel spectral plot.
Figure 6.31: Spectral response for the five fluorescence sources used and the resulting calibration linearity for a typical group one pixel. The group one pixel had a non-spectral response regardless of energy.

Figure 6.32: Spectral response for the five fluorescence sources used and the resulting calibration linearity for a typical group two pixel. A group two pixels has a poor spectral response at all energies.

strongly offset from the linear fit. The response at different energies for a group three pixel can be seen in figure 6.33. The most notable feature of the group three pixels is the
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Figure 6.33: Spectral response for the five fluorescence sources used and the resulting calibration linearity for a typical group three pixel. A group three pixels is the non responsiveness at low energies with an increased responsiveness for higher energies non responsiveness at low energies (i.e. Rb fluorescence) with an increased responsiveness for higher energies. This resulted in a good calibration linearity at higher energy while having a strongly offset Rb measurement point. The behavior of group three pixels has a possible explanation in the existence of a Te precipitate located close to the cathode electrode. The destructive influence of Te inclusion on detector performance has been well documented [88] [93]. A Te inclusion close to the cathode electrode would affect the response of Rb due to the shallow interaction depth of low energy X-rays. At higher X-ray energies the mean interaction depth would be located further into the bulk of CZT, resulting in a bypassing of the influence of an surface located inclusion. The poor spectral response for the ’bad pixels’ is currently not understood within the scientific community. Possible explanations include the effect of material imperfection such as Te inclusions [88] [93], cracks and twin boundaries [88]; poor bonding quality and/or low inter-pixel resistance (poor passivation) [139]. Using the stated definition of ’bad pixels’, 30% of the pixels for the ERD2004 CZT (eV PRODUCTS CZT) detector was defined as belonging to one of the three groups.
6.1.3 Further investigation of inter pixel non-uniformity

In order to further investigate spectral performance and inter pixel non-uniformity of the ERD2004 eV PRODUCTS CZT detector experiments where carried out using synchrotron radiation at DLS and using sealed radiation sources. The work was done in co-operation with Mr. Matthew Wilson (Rutherford Appleton Laboratory) and Dr. Julien Marchal (DLS). The synchrotron data visualization and analysis and the sealed source measurements where done by Matthew Wilson.

Spectral response of the ERD2004 CZT detector eV2.

The ERD2004 CZT detector, known as eV2, was initially tested for spectral performance using a sealed Am-241 source. The detector was cooled to 50°C and biased to -400V. The obtained spectral All plot can be seen in figure 6.34. A FWHM of 1.93keV and a peak to valley ratio of 14.9 was obtained. The main Am-241 peak, the Cd and Te escape peaks and the Np-daughters are clearly visible in the All plot.

![Am Spectrum -400V-278K](image)

**Figure 6.34:** Am-241 energy spectrum obtained for the ERD2004 CZT detector eV2 at 50°C and -400V. Charge sharing discrimination and pedestal removal has been done. A FWHM for the main Am peak of 1.93keV and a peak to valley ratio of 14.9 was obtained [140].

and a peak to valley ratio of 14.9 was obtained. The main Am-241 peak, the Cd and Te escape peaks and the Np-daughters are clearly visible in the All plot.

ERD2004 CZT detector uniformity.

In order to test the detector uniformity it was uniformly exposed to monochromatic X-ray beams at 15 and 20keV at station B16 DLS. The detector was biased to -350V and cooled to 50°C. The uniformity of the detector was measured as the total no events registered for each pixel. This has been visualized, in 2- and 3-D, for the 20keV measurement in figure 6.35. A region of with a low number of registered events was observed,
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Figure 6.35: (Left) 2D and (Right) 3D pixel intensity map for the ERD2004 CZT detector eV2, as found when exposing the detector to a uniform X-ray beam of 20keV. All the the 3D plots in this section, except for figure 6.36, has been normalized against the highest total number of events for any one pixel registered for each measurement. This has been done to ease the comparison of response between pixels. Lighter colour indicates a higher number of registered events. A region of low number of registered events, indicated by the white arrow is shown in both plots [140].

indicated by the arrows in figure 6.35, and two regions of high number of registered events centered around pixel 101 and 45 respectively. The uniformity of the detector was further investigated using the data obtained from the uniform exposure of a 15keV beam by analysis of the uncalibrated spectra of each pixel. Figure 6.36 shows a pixel map plot of the position of spectral peak (V). A lower voltage value for the peak is interpreted as a reduced charge collection efficiency for the pixel in question. A lower peak position is observed for the pixels in the low number of registered event region of figure 6.35.

A sealed radiation source (Am-241 dial source) uniformly irradiating the detector was
used to investigate the uniformity for the detector eV2 at different temperatures (i.e. 22°C and -5°C) and different X-ray energies (Ag=22.1keV and Tb=44.23keV). A bias voltage of -350V was used as in the DLS experiments. The no of registered events for all the pixels, while being uniformly irradiated using the Ag fluorescence, can be seen for two different temperatures, 22°C and -5°C, in figure 6.37 The region of low number of registered events are present at both temperatures. A higher uniformity was found at 22°C compared to -5°C. Four regions with pixels having a high number of registered events was observed at -5°C. The region of low number of registered events has shown a reduced peak position voltage indicating a lower charge collection efficiency. The improvement of uniformity with increased temperature could suggest that the non uniformity is caused, at least to some extent, by trapping sites in the detector crystal. This would be consistent with the work done by Soldner and Szeles [110] [141] [113] showing long ranging non-uniformities resulting from Te inclusions decorating a twin boundary. The detector was also uniformly irradiated by Tb fluorescence while biased -350V for a temperature of 22°C. A comparison of the uniformity when exposed to Ag fluorescence and Tb fluorescence can be seen figure 6.38. An increased uniformity was observed at higher energies. The region of low number of registered events was still present. The total intensity pixel map for a ERD2004 Si detector is also shown in figure 6.38 as a material reference. The Si detector was irradiated by the same Ag source as for the CZT detector while at room temperature and biased at +50V. The much higher degree of uniformity of the ERD2004 Si detector strongly indicates that the non-uniformity observed for the eV2 detector is material related. One explanation to the increased uniformity could be the decreased trapping caused by a shorter transport distance for events taking place further into the bulk crystal.

**Figure 6.37:** (Left) Normalized pixel intensity map of the eV2 detector for a uniform irradiation of Ag fluorescence. The detector was run at a temperature of 22°C and biased to -350V. (Right) Normalized pixel intensity map of the eV2 detector for a uniform irradiation of Ag fluorescence. The detector was run at a temperature of -5°C and biased to -350V [140].
Synchrotron raster scan investigation of the eV2 detector.

To investigate the sub-pixel spatial uniformity of the eV2 detector the 15keV beam was collimated using a pair of Huber slits to a size of 40x40\textmu m. The shaped beam was step scanned over a section of the detector. The scanning was done in lines. The distance between two measurement points in the same line was 50\textmu m and each point was illuminated for 2 seconds. The distance between subsequent, parallel, lines was 50\textmu m. The initial line was scanned from pixel 182 to pixel 73 (see figure 6.35 for reference on pixel numbering). The final line was scanned between pixel 238 to pixel 17. Part of the area scanned covered a section containing pixels having demonstrated a low no of registered events under uniform illumination. The detector was cooled to 5\textdegree C and biased at -350V. The number of events registered for each pixel for the entire scan can be seen in figure 6.39. The individual pixel spectral response for three lines are shown in figure 6.40. The first line, plotted in blue, is the registered response of the scan from pixel 182 to pixel 73. The length of the scanned line was longer then the distance between pixel 182-73 resulting in significant spectral response on pixel 181 and pixel 74. Irradiation of the inter-pixel area of pixels 182/181 and pixels 73/74, leading to a strong charge sharing effect is believed to be a main contributor. The same effect is also observed in the other inspected lines. The second line, plotted in red, is the registered response from pixel 198 to pixel 57. The most notable feature of this scan was a significant charge collection on pixel 64 when pixel 56 was irradiated. The third line, plotted in green, is the response obtained when scanning from pixel 214 to pixel 41. When irradiating pixel 40 a significant charge was collected on pixel 32. When irradiating pixel 41 the majority of charge was collected on pixel 33. When correlating these findings with figure 6.35, the
Figure 6.39: Normalized total intensity map for the entire scan [140]. The scan was conducted using a 40x40μm beam of a monochromatic, 15keV, X-ray beam. The detector was biased to -350V and cooled to 5°C.

Figure 6.40: The uncalibrated pixel spectra for three lines obtained from the raster scan experiment. The three lines go from pixel 182 to pixel 83 (blue), pixel 198 to pixel 57 (red) and pixel 214 to pixel 41 (green) [140].
number of registered events for uniform illumination, shows pixel 41 having a low number of registered events while the neighboring pixel 33 has a high number of registered events. The same is true for the relationship between pixel 56 (low) and pixel 64 (high). This is consistent with the result found by Szeles et al. [113], where pixels with high registered events is neighbored by pixels with low number of events. The channeling of charge from one pixel to its neighbor is possibly caused by lateral electric fields in the detector. The existence of lateral electric fields is believed to be caused by spatially charged regions [142] existing due to charge carrier trapping. Another possible reason could be poor pixelation leading to a changed weighting potential.

6.1.4 Investigations on charge sharing correction

When substituting a 300\(\mu\)m thick Si crystal with a 2mm thick CZT crystal it was realized that charge sharing would be present and lead to a degraded spectral response. Charge sharing is due to the expansion of the charge cloud when traversing the detector material, leading to the charge being collected on two or multiple pixels. In order to test the effects of charge sharing in the ERD2004 CZT detector, and the best way of correction for it, two charge sharing correction scripts, charge sharing addition and charge sharing discrimination, has been developed in the matlab environment and tested. The basis of the two programs are the same. For each registered event the program looks to see if any event took place on one of the neighboring pixels at the same time. Due to the sparse logic of the ERD2004 detector system (a pixel is only read out if an event is registered) this is done by comparing subsequent rows of data as it has been saved. The basic structure of the charge sharing correction programs can be seen in figure 6.41. If an event is registered at the same time for a neighboring pixel the event is labeled as a charge sharing event. For the charge sharing addition script pixels being labeled as sharing charge is added together and the full registered charge is added to the pixel with the highest initial value. The rest of the pixels are set to zero. For the charge sharing discrimination script all pixels that have been labeled as charge sharing are set to zero. The ERD2004 CZT detector was uniformly illuminated using a Am-241 radiation source while cooled to -50\(^\circ\)C and biased at -300V. The collected raw data was then corrected using both charge sharing addition and discrimination. Figure 6.42 shows the resulting All plot when applying charge sharing addition, charge sharing discrimination and no charge sharing correction (pedestals removed only) to the collected Am-241 radiation data. The charge sharing discrimination correction was found to yield the best spectroscopic performance. Charge sharing addition show a strong tailing of the main peak. This has been attributed to the user defined comparator trigger level controlling the read-out of pixels. If the part of the charge that has diffused into one
or several neighboring pixels is too small to trigger a readout it goes unregistered. The result is an event that has a lower value leading to a low energy tailing of peaks. This effect is also in present when using charge sharing discrimination but for discrimination only one pixel has to be labeled as charge shared for the error to be rectified, drastically reducing the effect. For the charge sharing discrimination spectra a drop in counts compared to the uncorrected spectra is seen at approximately 50 keV. This again due to comparator trigger value. The trigger level for the collected data was approximately 10 keV which is the difference between the main peak and the value of the drop. This shows a fundamental limit on the ability to correct for charge sharing using discrimination set by the user defined voltage value of the comparator with lower trigger voltage resulting in a better correction.
6.1.5 Investigations into the ERD2004 detector system countrate capability

In order to swiftly collect a rTEDDI image the countrate capability of the detector is of vital importance. A countrate for the pixelated device of between 1.000-10.000 hits*pixel^{-1}*sec^{-1} is currently the aim of the rTEDDI detector system [144].

Countrate limitations due to the global reset scheme.

In order to investigate the countrate capability and limitations of the ERD2004 CZT detector the detector was uniformly exposed to Tb fluorescence from an Am-241 dial source. The number of registered events observed for the entire detector during two minutes at different clocking frequencies and temperatures where collected. The detector was biased to -300V during all experiments explained in this section. The resulting plot can be seen in figure 6.43. The ideal detector behavior is illustrated by the black dot-dash line in the plot. After reaching a sufficiently high enough clocking frequency all the events emitted by the radiation source is detected, resulting in a constant countrate with increased clocking frequency. A strongly temperature dependent peaking of the countrate with increased clocking frequency was observed. A subsequent decrease in
The number of registered events observed for the entire detector during two minutes at different clocking frequencies and temperatures when exposed to Tb fluorescence. For each temperature a peak countrate was observed followed by a region where the countrate decreased when increasing the clocking frequency of the detector system. The variation of the maximum countrate with temperature was due to saturation of the pre-amplifiers during part of the active period at higher temperatures.

countrate with increased clocking frequency was observed following the peaking for all temperatures. The increased maximum countrate observed for decreased temperature was found to be due to saturation of the pre-amplifiers during part of the active period of the global reset period, $T$. The saturation resulted in an effective shorter measurement time at higher temperature. The active part of the global reset period, $T$, which dictates the time length during which events can be registered is dependent on the user defined clocking frequency and ‘number of reads per frame’ as stated in equation 6.1. The decrease of $T$ with increased frequency was investigated as a possible cause of the decrease in countrate with increased clocking frequency following the maximum countrate peaking. The initial measurement was repeated but as the frequency was increased the ‘number of reads per frame’ was varied in order to keep the length of $T$ constant. Measurements where taken for two $T$ values, $T=50$ and $T=150$, and the result can be seen in figure 6.44. The detector was cooled to $-50^\circ$C. The decrease in countrate with increased clocking frequency was slightly improved when keeping $T$ constant. The decrease was however still present. An overall decrease in countrate with increased length of $T$ was observed. An increased $T$ would yield a larger number of events (more data) for each frame. A decreased countrate therefore suggested a problem of the DAQ-system to handle larger amounts of data. A measurement was done by Matthew Wilson at the Rutherford Appleton Laboratory investigating the relative time of the active part of the global reset period ($T$) to the inactive part of the of the global reset period (the detector dead time) as a function of clocking frequency for a set $T$ value, $T=150$. The result can be seen in figure 6.45. The increased dead time with increased clocking frequency, seen in figure 6.45, inversely mirrors the decrease in countrate with increased clocking frequency.
The number of registered events observed for the entire detector during two minutes at different clocking frequencies while keeping T constant, for T=50 and T=150, when exposed to Tb fluorescence. The decrease in T with increased clocking frequency, as stated in equation 6.1, did not explain the decrease in countrate with increased clocking frequency. An overall countrate loss was observed with increased T between T=50 and T=150, indicating problems with the DAQ-systems ability to handle larger amounts of data [143].

Plot of dead time as a function of clocking frequency for the ERD2004 CZT detector. The increase in dead time with increased clocking frequency explains the decrease in countrate with increased clocking frequency [143].

An overall decreased countrate for increased T values where observed as can be seen in figure 6.44. In order to investigate the T length dependence of the countrate the detector was exposed to Tb fluorescence. The number of events registered during 2 minutes at different clocking frequencies for a number of set T values where collected. The result can be seen in figure 6.46. The countrate for different T as a function of frequency are rotated clockwise and the peak value is transported towards lower frequencies when going towards higher T values. Plotting the same data as a function of ‘number of reads per frame’ instead of frequencies can be seen in figure 6.47. An optimum countrate was seen at approximately the same ‘number of reads per frame’ regardless of the T value.
and is due to an optimization of the active part of T compared to the dead time. The drop in overall countrate seen in figure 6.46 is a consequence of operating the detector in a less favorable 'number of reads per frame' region shifting the countrate response in accordance to the observed pattern seen with increased T as seen in figure 6.47. An increase of the optimal countrate as a function of 'number of reads per frame', as seen in figure 6.47, for increasing T, up to T=50, is accredited to a lack of detector oversampling with respect to the photon X-ray flux. As T increases the clocking frequency for a specific 'number of reads per frame' will be less likely to have reads where no photon detection has been registered. Optimized performance for the detector at high count rates will therefore be realized by setting the 'number of reads per frame' to the specified optimal value while decreasing the T value resulting in an effective increase of detection frequency.
Countrate limitations due to the local reset scheme.

The ERD2004 CZT detector was tested using a $\theta/2\theta$ measurement, as seen in figure 6.48 on station 16.3 at the SRS Daresbury, initially to test the energy resolution of the detector at different energies with a cleaner spectral source then found using the Am-241 dial source. The setup utilized a white square X-ray beam incident on a single crystal Si sample set at an angle ($\theta$). The detector housing was placed at an angle ($2\theta$) behind the Si crystal resulting in the 20keV Laue spot (and harmonics) from the $(2 2 0)$ plane of the Si crystal uniformly irradiating the detector. The detector housing was well shielded using a Pb/Cu/Al sandwich structured housing to ensure reduced noise from the surrounding environment. A vacuum tube and a lead shield was used to reduce the noise due to forward scattering of the incoming beam. When illuminating the entire area of the detector when attenuating the incoming beam with only 1mm of Al to reduce the flux, the detector was non responsive, registering no events at all. When using a 2mm thick Pb attenuation the detector registered events but the expected spectral peaks at 20, 40, 60 and 80keV where not distinguishable. When using a 3mm thick Pb attenuation a single high energy peak at approximately 86keV. The spectral response of the detector when attenuating the incoming beam with 2mm and 3mm Pb can be seen in figure 6.49 If more then two events, is registered for the same pixel, takes place within the same local reset period the readout chain of the pixel freezes and becomes non responsive [145] explaining the non responsiveness when using only 1mm of Al to attenuate the incoming beam. If two events, is registered for the same pixel, takes
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Figure 6.49: The spectral response of the detector when exposed to 20keV radiation over all pixels using the $\theta/2\theta$ setup for different attenuation of the incoming X-ray beam. (Left) All plot of the spectral response when attenuating the incoming beam using 1mm Pb. (Centre) Single pixel plot of the spectral response when attenuating the incoming beam using 1mm Pb. (Right) Single plot of the spectral response when attenuating the incoming beam using 3mm Pb. When using 1mm of Al to attenuate the beam freezing of the pixel readout chains due to multiple events occurring in the same local reset period [145] was observed. As the attenuation was increased to 1mm Pb the detector became responsive but the spectral response was non-sensible. Further increase of the attenuation to 3mm Pb resulted in spectral response place within the same local reset period the value of the resulting measurement will take a non determinable value [145] resulting in a non interpretable observed spectra, explaining the response as seen when using 2mm Pb to attenuate the incoming beam. The similarities between the 2mm Pb All-plot spectra shown in figure 6.49 and the one shown in figure 4.31 would suggest that the inability of the rTEDDI system to produce any diffraction images on station 16.3 was due to the detectors inability to handle the necessary X-ray flux. The normalized theoretical station 16.3 beam flux profile when attenuating with 3mm Pb can be seen in figure 6.50 The correlation between the 3mm Pb attenuation theoretical beam profile and observed detector spectra indicated that in order to reduce the flux enough to achieve spectral performance all lower energy X-rays

Figure 6.50: Theoretical beam profile for station 16.3 SRS Daresbury when attenuated using 3mm of Pb. The shape of the beam profile explained the observed spectral response of the ERD2004 CZT detector when the incoming beam was attenuated using 3mm of Pb.
where absorbed by the attenuator, resulting in no discernible 20, 40 or 60keV peaks. The 80 keV peak is observable in the spectra. The theoretical beam flux profile with 3mm Pb attenuation and without attenuation can be seen in figure 6.51. The heavily reduced beam flux needed in order to get spectral performance from the detector would translate to very long scanning times needed for the rTEDDI system making the ERD2004 CZT detector undesirable as part of the system. When reducing the beam size to one pixel spectral response was observed, as can be seen in figure 6.52, when attenuating using 2mm Pb. This is attributed to the increased readout speed of this pixel when not needing to compete with with other pixels.

**Figure 6.51:** Relative beam profiles for station 16.3 SRS Daresbury without attenuation (blue) and with 3mm Pb attenuation (red). The high reduction of photon flux needed to obtain spectral response for the ERD2004 CZT detector would severely hinder the speed and hence the usability of the rTEDDI system.

**Figure 6.52:** (Left) Total intensity pixel map of the ERD2004 CZT detector when exposing a single pixel to 20 keV radiation using the $\theta/2\theta$ setup. The incoming beam was attenuated using 2 mm of Pb. (Right) The resulting single pixel spectral response. The sensible spectral response observed at a lower attenuation is attributed to the higher readout rate of the single pixel when not competing for readout with other pixels.
6.1.6 Summary ERD2004 detector

At the start of this project the ERD2004 detector had already been developed [25]. The detector, using a Si crystal, had been tested for spectroscopic performance but no further performance characterization had been done. The detector was initially identified as a viable candidate for the rTEDDI imaging system, and was in fact used in the proof of concept for the technique described in chapter 4. The active detector material was substituted from Si to CZT to increase the detection efficiency and allow the detection of a wider range of X-ray energies. The increased detection efficiency would allow the rTEDDI system to image thicker/denser samples such as jet engine turbine blades or friction stir welds. The increase in detectable energy range would allow for a higher accuracy in determining atomic structural information obtainable using the rTEDDI system. An in depth characterization of the ERD2004 CZT detector was carried out and has been described.

Early EDR2004 CZT detectors were found to be suffering from a bias dependent malfunctioning of pixels, referred to as the black hole effect, which was corrected for in later ERD2004 CZT detectors by improvements in the device packaging process.

When exposing the ERD2004 CZT detector to radiation from known radiation sources (Am-241 dial source, Am-241 source) variation in spectral response compared to measurements done using Si as the active detector material was observed. The changes in number of observed peaks and intensity of the observed peaks were explained by the increased detection efficiency and atomic constituents of the detection material when substituting to CZT crystals.

A high degree of inter pixel non-uniformity was observed for the ERD2004 detector when using CZT as the active detector material that was not observed when Si was used. The degree of non-uniformity observed would render the detector unusable in any quantiUsing the linearity plot obtained for each pixel during detector energy calibration it was found that the spectral response did not only vary in between pixels at a specific energy but also varied within a single pixel when changing the energies of the X-ray detected. Pixels exhibiting spectral response variation with changing energy was classed into three groups of 'bad pixels' with the aid of the energy calibration linearity plots. Out of all the pixels for the ERD2004 CZT detector, using eVPRODUCTS grown CZT, approximately 30% was suffering from 'bad pixel' behaviour.

Spectral improvement by charge sharing correction of the ERD2004 CZT detector was found to be severely limited by the user defined voltage of the comparator driven readout scheme inherent to the detector. The user defined voltage effectively set a hardware low energy cut-off of the detector system preventing charge sharing between pixels below the trigger level from being detected introducing an error into any correction done. Charge sharing discrimination was found to be a superior correction algorithm compared to
The countrate capability of the ERD2004 detector system was found to be severely limited by the DAQ ability to handle large data amounts. The inability of the DAQ system to handle large data amounts resulted in very high detector deadtimes in between global reset periods of the detector. The optimum countrate capability was consequently achieved by setting the 'number of read per frame' parameter constant (approx 800) and adapting the clocking frequency of the system to the X-ray flux. At higher X-ray fluxes optimum countrate is achieved by increasing the clocking frequency, effectively minimizing the active global reset period. If two events would take place, for the readout channels of the ERD2004 detector, during the same local reset period the value on the peak hold circuit would take a random nature [145]. If more than two events occurred during the same local reset period the readout chain would freeze, becoming non responsive [145]. Investigations using synchrotron X-ray diffraction showed that the incoming beam had to be attenuated (3mm Pb attenuation) to a small fraction of the normal beam profile in order for the ERD2004 detector local reset speed to be sufficient to produce accurate spectroscopic performance, making the detector unsuitable for rTEDDI imaging at higher flux stations. At higher X-ray fluxes a nonsensical spectral response was observed. The spectral response obtained showed strong similarities to the spectral response found during the unsuccessful rTEDDI imaging test on station 16.3 SRS Daresbury, described in section 4.2.5, suggesting this was the cause of the unsuccessful imaging trial. A further increase of the X-ray flux rendered the ERD2004 CZT detector completely non responsive.

The limiting effects found during the investigation of the ERD2004 CZT detector was used to further improve the design of the HEXITEC ASIC with respect to imaging using the rTEDDI system. The HEXITEC detector is a detector designed purposely for spectroscopic X-ray imaging using CZT as the detector medium, with rTEDDI being the initially envisioned application, and is the subject of the next sub chapter. The limitation of the ERD2004 detector caused by the user defined comparator voltage, used to trigger read out of channels, when using charge sharing correction to optimize energy resolution was overcome in the HEXITEC ASIC design by the use of a rolling shutter read out scheme. The rolling shutter read out scheme remove the hardware low energy limit seen in the ERD2004 detector and will allow for a much improved charge sharing correction. The observed countrate limitations of the ERD2004 detector allowed for an improvement of the HEXITEC DAQ system specifications and a re-design of the HEXITEC ASIC to allow for a higher detector countrate. The observed inter pixel
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non-uniformity when using CZT as the active detector material is a major problem destroying the capability to perform spectroscopic imaging. In an attempt to overcome this obstacle CZT material from a number of different manufacturers have been tested on the HEXITEC ASIC and is described in the next sub chapter.

6.2 The HEXITEC detector

The HEXITEC ASIC was developed at the Rutherford Appleton Laboratory [146] as part of the HEXITEC collaboration [147]. The detector was developed with the main aim being optimized energy resolution and small pixel pitch while utilizing CZT as the active detector material. The limitations found while investigating the ERD2004 CZT detector performance with respect to imaging using the rTEDDI system was used to improve the design of the HEXITEC detector system. The problems of using a comparator triggered readout scheme with respect to energy resolution was addressed in the design and countrate limitations imposed by the ERD2004 detector DAQ resulted in improved specifications for the HEXITEC detector DAQ. This sub-chapter described the structure and readout scheme of the HEXITEC detector and the complete detector system. This is followed by a description of initial testing done on the HEXITEC ASIC and detector system using test signals. Initial testing of the HEXITEC detector using CZT as the active detector material to detect X-ray radiation was done by Chris Youd (University of Manchester) under the supervision of the author and Matthew Wilson (Rutherford Appleton Laboratory) and is described here. Investigations into inter pixel uniformity was done by Chris Youd (University of Manchester), Matthew Wilson (Rutherford Appleton Laboratory) and Mathew Veale (Rutherford Appleton Laboratory), using synchrotron radiation and active radiation sources, and is also shown. The developed software programs used to analyze the data from the HEXITEC detector is described in detail in AppendixB.

6.2.1 The HEXITEC detector system

The HEXITEC ASIC was developed by the Rutherford Appleton Laboratory [146] and is a 20x20 pixel array energy resolving detector with a 250µm pixel pitch. The ASIC was designed to have a 2mm thick CZT crystal bump bonded to it; be able to detect X-rays in the energy range of 1-150 keV (optionally 10keV to 1.5MeV) and have a spectral resolution of 200eV (FWHM) when having a 300fF detector capacitance and 5pA leakage current. Each pixel in the 20x20 pixel array has a readout chain associated with it consisting of a pre-amplifier; shaper and filter; peak hold and three sample hold circuits as can be seen in figure 6.53. The pre-amplifier is a charge amplifier with a selectable
Figure 6.53: Block diagram of the HEXITEC ASIC pixel readout chain. Each pixel has a readout chain dedicated to its readout consisting of a pre-amplifier; shaper and filter; peak hold and three sample hold circuits. The lack of comparator circuit triggered readout will increase the energy resolution obtainable using charge sharing correction but considerations has to be taken to preferential high energy sensing as well as incorrect charge sharing addition, the severity of which both will increase with increased X-ray flux. The three sample hold circuits are used in order to enable ‘end of frame correction’.

The optional ranges are set by the value of the feedback capacitance and will take either a value of 15fF, corresponding to a detection range of 1-150keV, or 15+135fF, corresponding to a detection range of 10keV-1.5MeV. The amplifier also has a feedback circuit enabling compensation for leakage currents up to 50pA and a power down option, accessible from the user GUI. The output of the amplifier is shaped and filtered using a CR-RC type configuration bandpass filter and a 2nd order low pass filter, resulting in a shaping time of $2\mu$s. The 2nd order low pass filter was incorporated in order to reach the low specified noise levels in spite the use of the long shaping times ($up to 1\mu$s) needed when using CZT. The peak hold circuit follows the voltage of the filter output and maintains the voltage of the peak of the shaped signal until it can be read out. Unlike the ERD2004 ASIC pixel readout for the HEXITEC ASIC is not triggered by a comparator but set by a specific time interval from the last readout. This allows better charge sharing correction capabilities since no hardware low energy cut-off is present. It does also introduce a number of effects that has to be considered. If a second, higher energy event takes place for a single channel (pixel) before the first event has been read out the second event, having a higher pulse height, is held and read out. The detector will as a result preferentially detect high energy events. This effect will become more significant under high flux conditions. If the lower energy event would result in charge sharing any charge sharing addition would result in the higher energy event and the
shared charge from the low energy event being added and an error being introduced by the correction. At the end of each channel three sample holds, known as S1, SPH and S2, have been incorporated and are sampled sequentially. S1 and S2 samples the shaper output while SPH samples the peak hold circuit. The SPH sample contains the value corresponding to the energy of the detected X-ray photon while S1 and S2 are used to veto incorrect peak hold data due to partially developed signals at the time of readout, known as ‘end of frame correction’. ‘End of frame correction’ becomes more important at higher readout rates, such as envisioned when using the detector in the rTEDDI system, due to the relative time ratios of the shaper rise and reset time to time between read outs. Three programmable registers, known as the Read Enable Register, Power Enable Register and Calibrate Enable Register, as can be seen in figure 6.54, can be used to define and select smaller regions of the detector 20x20 array to be read out. These registers can be used to create regions of interest to be read out. These smaller regions of interest can be read out at a faster rate then is possible when reading all the pixels. The HEXITEC ASIC pixel array is read out using a rolling shutter read out scheme. This means that one row is output onto the column outputs and the peak hold circuits in the specific row reset. The next row is subsequently read out and the process continuous to the last row. When the last row is reset the first row is next in line and the

**Figure 6.54:** Block diagram of the HEXITEC ASIC. The active area being read out by the detector can be set using the Power, Read and Calibrate Enable Registers. The selection of a smaller active region allows for region of interest investigations. A smaller active region also allows for a faster readout rate. The readout is done using a rolling shutter readout scheme.
readout continue in this manner in a rolling manner. The values on the column output are continuously read out. The time it takes to read on row is given by equation 6.2:

\[ t_{\text{row}} = (50\text{ns} \times (n_{\text{pixels}} + 1)) + 2\mu\text{s} \quad (6.2) \]

where \( n_{\text{pixels}} \) is the number of pixels in the row set by the active area chosen and the extra 2\( \mu \)s is due to the readout of S1 and S2 needed to enable 'end of frame correction'. The time it consequently takes to read out one frame (first row to last row once) is given by equation 6.3:

\[ t_{\text{frame}} = (n_{\text{rows}} + 1) \times t_{\text{row}} \quad (6.3) \]

where \( n_{\text{rows}} \) is set by the chosen active area of the detector. When reading out all 20x20 pixels the time it takes to read one frame is \( 6.405 \times 10^{-5} \)s resulting in a frame readout rate of 15.6kHz. Assuming a 10 percent occupancy rate, in order to prevent for preferential high energy sensing, a maximum number of 1.560 photons*pixel\(^{-1}\)second\(^{-1}\) is possible, which is in the desired range, 1.000-10.000 photons*pixel\(^{-1}\)second\(^{-1}\), for the TEDDI system [144]. The data coming out from the HEXITEC ASIC is handled by the HEXITEC detector DAQ system developed by aSpect Systems [148]. Each frame of data is converted to a greyscale image, where each pixel have a greyscale value corresponding to the voltage value read out of the pixel peak hold circuit, and is transported via a camera link system and saved to a PC disc memory. The HEXITEC detector ASIC and DAQ system is controlled from a user GUI running on the PC.

6.2.2 Initial testing of the HEXITEC ASIC and detector system using test signals.

The HEXITEC detector system was initially evaluated on assembly, without having a CZT crystal bump bonded to the ASIC, using test signals in the 3-frame mode (described in Appendix B) in order to validate the performance of the system. Test signals are also referred to as calibration signals and work on the premises of inducing a user defined voltage pulse onto the VCAL input, as seen in figure 6.53, of each channel and thereby triggering the entire readout chain. A number of problems where discovered and addressed through this investigation.

The ability to select regions of interests to be read out using the Power, Read and Calibrate Enable Registers was investigated using the settings seen in figure 6.55 as a starting point. The Power Enable Register defines the region of pixels that are powered
Figure 6.55: Register base settings for investigations of the region of interest collection for the HEXITEC detector. Using these register settings the pixels in grey should be read out and contain test signal values. The pixels marked in light blue should be read out but not contain any test signal. The dark blue pixels are powered up but are not read out. Pixels not being read out will reduce the size of the greyscale image produce as data output, but the are not read can not be reconstructed without external knowledge of the register settings.

up. The Calibrate Enable Register defines the region of pixels that are connected to the calibrate input (activating the test signal). The Read Enable Register defines the region of pixels that are to be read out. If the Read Enable for a row is set to 1 the row is read out and result in a row in the greyscale image. If the Calibrate Enable for a row is set to 1 a signal is induced on that row and a event is registered giving a darker value in the greyscale image. When setting the column values to select the region as seen in figure 6.55 but keeping all row registers active the greyscale image seen in figure 6.56 was obtained. 4 rows have the Read Enable set to 0 resulting in only 16 rows being read out and displayed in the greyscale image. The setting of the Calibrate Enable for the rows will induce a test signal in two 4 pixel wide regions with 4 pixels in between having no signal induced. The resulting image shows the region containing the ’real’ SPH data, obtained in a 3-frame mode run from the centre of the image, having two vertical dark stripes with a vertical light stripe in between. The dark pixels in the SPH data is due to activation of both the Calibrate and Read Enable Registers being active resulting in
signals being induced onto the channels and the values of those signals being read out. The light stripe is due to the pixels being read out but no signal being induced on the channel. It is also seen that vertically the SPH region contains less than 20 pixels while horizontally there are 20 pixels. The vertical decrease in size of the SPH region is due to the 8 pixels having their Read Enable Registers in an inactive mode. The values are consequently not read out and the image is reduced. It should be noted that the actual position of the columns that have been deactivated cannot be obtained from the data.

Any spatial reconstruction of the data obtained using the HEXITEC detector therefore depends on the user noting down which areas of the detector are active and which are not. When setting the column and row register values as seen in figure 6.55 the image seen in figure 6.57 was obtained. The image obtained was very much different from what was expected. Signals induced where seen in both of the two reset S1/SPH/S2 regions and the image was distorted. An investigation into the cause of the inability to select a region of interest was carried out by Matthew Wilson at Rutherford Appleton Laboratory, and an error was found in the ASIC design. The packages used in the ASIC design of filler cells had been corrupted and resulted in the electronics shorting itself out when trying to activate region of interest settings on the row registers. As a consequence only 20x20 pixel measurement was done on this generation of the HEXITEC ASIC. The problem has been corrected in the design of the second generation of the 20x20 pixel HEXITEC ASIC (not discussed in this thesis) allowing for region of interest settings.

Test signals where used in order to test the countrate of the HEXITEC detector system when running in the 3-frame mode. The number of collected frames (greyscale images) was varied while monitoring the time taken to finish a run and the size of the data stored in the memory. The result is shown in table 6.10. An increase in run time disproportional to the increase in number of frames was observed. Calculations on the 20,000 frames
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Figure 6.57: Resulting greyscale image produced when all row and column registers are set as seen in figure 6.55. The resulting greyscale image is distorted and signals read where there should be none. The reason for the inability to freely choose regions of interest to collect data from was found to be an error in the filler cell design of the ASIC creating short circuits when selecting regions of interest settings for the row registers. The filler cell problem was corrected in the design of the second iteration of HEXITEC ASIC’s.

<table>
<thead>
<tr>
<th>Number of Frames</th>
<th>Run time</th>
<th>Data size</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,000</td>
<td>A few seconds</td>
<td>7.81MB</td>
</tr>
<tr>
<td>20,000</td>
<td>11 minutes</td>
<td>171MB</td>
</tr>
<tr>
<td>50,000</td>
<td>50 minutes</td>
<td>390MB</td>
</tr>
</tbody>
</table>

Table 6.10: Collection time and data size measurements for varied number of collected frames.

run showed a countrate of 36.6 events*second\(^{-1}\)*pixel\(^{-1}\), far below expected values for the HEXITEC ASIC. Investigation into the deadtime of the detector system showed a deadtime of approximately 90%. This was found to be due to the operation of the Aspect DAQ system. The DAQ system was designed so that it operated by collecting events, stopping collection of events, allocating memory space, writing to memory and the start the cycle over again. The system was consequently inactive the majority of the time. The low number of events*second\(^{-1}\)*pixel\(^{-1}\) was a result of the inactive period and not the sped of the HEXITEC ASIC. Modifications of the Aspect DAQ system resulted in a deadtime close to zero [149].

The effects of the large data amounts collected during runs on the performance of Matlab was investigated by measuring the time needed to produce an All-plot using the Conny_All_plot.m program (see Appendix B) for different number of frames collected. The binning region of interest for which the program was run was 6000-8000 LSB. The plotted result can be seen in figure 6.58. Producing one All plot using matlab using 50,000 frames of data took approximately 20 minutes. Investigation into the increase in time needed to produce an All plot over a larger LSB region of interest showed that the time needed was proportional to the size of the region of interest. The time needed to produce a single All plot using 50,000 frames of data if the region of interest was
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Figure 6.58: Plot of time needed to produce an All plot using the Conny_All_plot.m program (binning region of interest 6,000-8,000 LSB) as a function of number of frames collected. Increasing the binning region of interest increased the time to produce a All plot in proportion. The long times needed to do basic data analysis and visualization of the HEXITEC detector data using Matlab suggested another solution was needed. Extrapolation from the trend shown in figure 6.58 for 150,000 frames of data and setting the region of interest to 0-10,000 LSB would yield a run time for the Matlab program of 4.6 hours in order to produce a single All plot. The excessive times needed to produce basic data analysis and visualization of HEXITEC detector data has been attributed to Matlab’s problem handling large matrix operations. Basic data handling has been handled in the short term by adapting the EasyEDD program, developed by Taha Sohi at University College London as part of the HEXITEC collaboration, to be able to read and analyze the HEXITEC data format. A long term solution with respect to analysis of the HEXITEC detector data for rTEDDI applications has been proposed and system specification requirements been worked out by the author in co-operation with DsoFT Solutions Inc [150]. For the document stating the requirement specifications of the proposed system please see Appendix D.

The All plot and single pixel spectral resolution of the HEXITEC ASIC was investigated in the 3-frame mode using 0.2V test signals. The resulting All plot and single pixel plot, both after pedestal subtraction, can be seen in figure 6.59. A FWHM of 215 LSB was found for the All plot while a FWHM of 35 LSB was found for the single pixel plot. The large variation between the All plot and single pixel plot in combination with the peak structure found for the All plot was thought to be due to the varied maximum peak position observed between single pixel plots. To verify this a program was developed in the Matlab environment that found the LSB value corresponding to the maximum
spectral intensity and subsequently shifted all single pixel spectra to make their maximum spectral intensity coincide with the one found for the All plot. The output of the shift program was a 20x20 array of values where each value corresponded to the LSB shift of the maximum intensity between that pixel and the All plot value. The resulting spectral response before and after the shift correction can be seen in figure 6.60. An All plot response close to the single pixel response was found after the shift correction.

The spectral response for two more test signal voltages, 0.15V and 0.1V, was collected and the shift correction file created at 0.2V was used to shift correct at these voltages in order to see if the correction was translatable with signal voltage. The result can be seen in figure 6.61. The All plot FWHM before shift correction are approximately 200 LSB regardless of the test signal voltage. The spectral peak response of the All plot before shift correction also show a high degree of similarities. The single pixel response regardless of test signal voltage is approximately 30 LSB. When observing the All plot spectral response after shift correction, using the shift correction file produced for the 0.2V data set, for the different test signal voltage plots a variation can be seen. At 0.2V the spectra is well corrected and the peak has a highly Gaussian shape. For the 0.15V

Figure 6.59: All plot and single pixel plot obtained for test signal data collected in the 3-frame mode. A All plot FWHM of 215 LSB and a single pixel plot FWHM of 33 LSB suggested a variation between pixels degraded the All plot spectra.

Figure 6.60: All plot before and after the use of shift correction. An increased spectral resolution was observed after shift correction.
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Only minor differences with voltage was observed for single pixel responses and All plot response before shift correction. The variation in improvement using shift correction as a function of voltage suggested a gain variation factor between pixels.

spectra a low energy tailing is starting to appear, degrading the energy resolution. At 0.1V the energy resolution of the peak has been reduced to 72LSB FWHM. Crating a shift correction file at 0.1V and applying it to the other voltages has the same effect but the best spectral response is obtained at 0.1V and the worst spectral response at 0.2V. This effect has been attributed to gain factor variations between pixel channels. The degrading spectral effect of a gain factor variation between pixels could be effectively removed by energy calibrating each pixel separately.

6.2.3 Initial testing of the HEXITEC CZT/CdTe detector.

Detector crystals from a number of manufacturers, Acrorad (CdTe) [151]; eVPROUDUCTS (CZT) [89]; Yinnel Tech (CZT) and Redlen Technologies (CZT) [152] was gold stud bump bonded to the HEXITEC ASIC and exposed to a number of different X-ray radiation sources in order to enable initial testing of the detector system when exposed to ionising radiation. Initial testing of the HEXITEC detector using CZT as the active detector material to detect X-ray radiation was done by Chris Youd (University of Manchester) under the supervision of the author and Matthew Wilson (Rutherford Appleton Laboratory) and is described here. Investigations into inter pixel uniformity was done by Chris Youd (University of Manchester), Matthew Wilson (Rutherford Appleton Laboratory) and Mathew Veale (Rutherford Appleton Laboratory), using synchrotron
radiation and active radiation sources, and is also shown.

**Initial spectroscopic limitations of the HEXITEC detector.**

Initially no spectroscopic performance was obtainable using the HEXITEC detector with a CZT crystal as the active detector material. Investigation into the cause of the non responsiveness, by monitoring the outputs from single pixel channels, showed the induction of a large noise pulse during readout. The observed noise pulse can be seen in figure 6.62. The noise pulse observed went low at S1 and high at S2. The shape of the noise pulse corresponds to a square wave pulse after having gone through the shaper and filter circuits as can be seen in figure 6.63. The time at which the pulse was induced (S1) and the length of the pulse (S1 to S2) resulted in the ENO signal being the most likely candidate to be causing the problem. The ENO signal is a clock signal dealing with the readout of the collected data. The ENO signal switches the current of the row output lines to drive the data to the readout multiplexers. A total current of 4mA for the entire detector (200µA for each channel) is switched on and off for each ENO clock
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This was found to be causing a temporary potential increase of the ground effectively causing a ground bounce. A 1pF capacitor in the leakage current compensation circuit of the pre-amplifier was coupled to the ground. As the ground bounce occurred the noise signal was consequently induced in the pre-amplifier and reversed through the readout channel. The amplitude of the noise pulse drove the peak hold circuit to a value corresponding to 46keV. Any X-ray event having an energy below this value was therefore not registered by the detector, resulting in a lack of spectral response below 46keV. The permanent solution of the problem has been achieved for the second generation of the HEXITEC 20x20 ASIC. A temporary solution using a compensation signal was developed in order to test the first generation ASIC as extensively as possible in spite the ground bounce problem. The compensation signal solution used a square pulse of opposite polarity of the pulse created by the ground bounce to cancel out the noise signal and restore spectral response of the detector.

Spectroscopic performance when using compensation signal correction.

Due to the observed noise pulse, resulting from the ground bounce, the HEXITEC DAQ system was modified to enable cancellation of the noise pulse through the induction of a pulse of opposite polarity. The cancellation pulse, also known as the compensation signal, is induced onto each channel through the VCAL input, originally used to induce test signals. The compensation signals takes three inputs, the start time with respect to the last reset, the stop time with respect to the last reset and the magnitude of the pulse. These parameters are user defined in the GUI. The difference between the start and the stop time gives the length of the square pulsed test signal and the magnitude the voltage of the square pulse. Due to the nature of the noise pulse the compensation pulse starts at same time as S1 is activated and stops at the same time as S2 is activated. The optimum magnitude of the compensation signal had to be experimentally determined by varying the magnitude and monitor the value of the base level of the peak hold circuit at the end of a readout when no ‘real’ signal had been sent in. Figure 6.64 shows the effect of the compensation on the base level as seen for a HEXITEC detector using eVPRODUCTS CZT as the active detector material. The HEXITEC detector uses negative signals (electrons inducing the pulse). A observed higher greyscale level (LSB value) consequently means a lower voltage stored on the peak hold circuit and a lower signal having been induced on the readout chain. Testing the effect of the compensation showed an increased in the measured LAB value up to a peak maximum (at approx. 0.09V). This is attributed to the compensation signal partially canceling out the noise pulse with increased magnitude. At 0.09V optimum compensation is achieved. As we increase the compensation signal magnitude further a noise pulse is induced not
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Figure 6.64: Plot of the mean effect on the base level value of the Peak hold circuit when correcting using compensation signal. The channels of the HEXITEC detector are designed to work for negative (electron induced) signals. A increase in LSB value therefore correspond to a lower voltage value measured at the SPH (peak hold circuit) and S1/S2 (output of the filter circuit) positions. An initial increase of the base level was seen when increasing the amplitude of the compensation signal. The curve peaked at the optimum correction (approx. 0.09V-0.01V for the eVPRODUCTS detector). A subsequent decrease in LSB value followed due to over compensation using the compensation signal.

due to the ground bounce but due to a over compensation with the compensation signal. It should be noted that the peak hold circuit is falling edge triggered. For the ground bounce noise signal, being a negative signal, the peak hold circuit is triggered as the signal comes in. For the over compensation noise signal, being a positive signal, the peak hold circuit is triggered as the signal leaves. The detector bias voltage dependence of the compensation signal was tested for detectors bonded to eVPRODUCTS CZT and Acrorad CdTe, by repeating the measurement at different bias voltages. The result can be seen in figure 6.65. The different y-axis windows used for the two plots in figure 6.65

Figure 6.65: (Left) Plot of the SPH base level value as a function of compensation signal amplitude for the HEXITEC detector bump bonded to eVPRODUCTS CZT at different bias voltages. (Right) Plot of the SPH base level value as a function of compensation signal amplitude for the HEXITEC detector bump bonded to Acrorad CdTe at different bias voltages. The difference in the y-axis between the two plots should be noted. For the eVPRODUCTS detector no bias dependence was found. For the Acrorad detector a shift towards lower LSB values with increased bias voltage up to -350V was observed.
should be noted. The eVPRODUCTS detector showed close to no bias dependence and had a optimum compensation signal amplitude of 0.09-0.10V. The Acrorad detector showed good correspondence for a decrease in LSB value with increased bias voltage and had a optimum compensation signal amplitude of 0.08-0.10V. A direct comparison of the compensation signal response for both the eVPRODUCTS detector and the Acrorad detector with a HEXITEC ASIC without any crystal bonded to it can be seen in figure 6.66. A comparison between the eVPRODUCTS detector and Acrorad detector showed a stronger influence of the compensation signal for the eVPRODUCTS detector. A comparison between the eVPRODUCTS detector and just the ASIC showed overall higher LSB values for the ASIC on its own and an optimum compensation signal amplitude shifted down towards 0.04V. Both the eVPRODUCTS and the Acrorad detector was subsequently exposed to a number of X-ray fluorescence energies from the Am-241 dial source. The resulting spectral response for both detectors can be seen in figure 6.67. The Acrorad detector showed no spectral response at any X-ray energy even at higher bias voltages. This is accredited to the lack of correction when injecting the compensation signal as observed in figure 6.66. For the eVPRODUCTS detector spectral response was obtained for the two highest energies, Ba (44keV) and Tb (32keV), of the Am-241 dial source enabling distinction between the Kα and Kβ peaks. At the lower fluorescence energies of the dial source, starting with Ag (22keV), no spectral response was obtainable for the eVPRODUCTS detector. By energy calibrating the eVPRODUCTS detector using the Tb and Ba Kα and Kβ peaks, as can be seen in figure 6.68, the y-axis (LSB axis) zero point occurred around 22 keV. The detector cutting out at this

![Figure 6.66: Direct comparison of the SPH base level value as a function of the compensation signal amplitude for the eVPRODUCTS detector, Acrorad detector and the HEXITEC ASIC without any crystal bump bonded to it. A stronger variation of the SPH base level value of the compensation signal was found for the eVPRODUCTS detector compared to the Acrorad detector. The optimum compensation signal amplitude was lower for the ASIC on its own compared to when bonded to eVPRODUCTS CZT.](image-url)
Figure 6.67: Spectral response of the eVPRODUCTS detector and Acrorad detector, using compensation signal correction, when exposed to the X-ray fluorescence of a number of Am-241 dial source targets. No spectral response was observed for the Acrorad detector even at higher bias voltages. The eVPRODUCTS detector showed spectral performance down to 22keV.

point would explain the lack of spectroscopic performance at lower X-ray energies. The difference in base level LSB value of the S1 and SPH circuit as a function of amplitude of the compensation signal can be seen in figure 6.69. At the optimum correction (0.09V-0.10V) there was a difference in the S1 value, measured at the filter output, and the SPH, measured at the peak hold output, corresponding to vertical shift seen in the energy calibration. The difference in S1 and SPH values are not completely understood but would suggest an incomplete correction of the ground bounce noise signal by the compensation signal. A variation in the ability of the compensation signal to be able to correct for the ground bounce noise seem to be material dependent with hardly no correction taking place for Acrorad CdTe and correction leading to spectral response down to X-ray energies of 22keV for the eVPRODUCTS CZT. Tests using Redlen CZT allowed for spectral response down to and possibly below 8keV when correcting using
Figure 6.68: Energy calibration of the eVPRODUCTS detector using the Tb and Ba Kα and Kβ peaks. The y-axis (LSB axis) zero point occurred around 22keV indicating spectral response would not be possible at lower energies, as had indeed been observed. The observed variation between pixels are believed to be due to inter pixel gain variations (see section 6.2.2).

Figure 6.69: Difference between the base level value of S1 and SPH as a function of compensation signal amplitude for the eVPRODUCTS detector. The difference in LSB value at optimum compensation signal amplitude (0.09V-0.1V) corresponded to the shift seen in figure 6.68 suggesting a incomplete correction of the ground bounce noise signal using the compensation signal resulted in a lack of spectral response below 22keV.
the compensation signal as can be seen in the next section. The eVPRODUCTS detector linearity was tested, in high gain mode, using test signals with increasing amplitude. The result for 4 of the detector pixels, plotted as Energy vs. LSB value, can be seen in figure 6.70. A high degree of linearity was found up to approximately 200keV, meaning

![Image 6.70: Linearity of the eVPRODUCTS detector as a function of energy as obtained using test signals. A good linearity was found up to approximately 200keV and saturation occurred at approximately 260keV.](image)

the detector exceeded the specifications set on it. Saturation occurred at approximately 260keV.

**Spectroscopic performance and inter pixel uniformity at different X-ray energies for the HEXITEC detector using Redlen CZT.**

The spectroscopic performance and inter pixel uniformity at different X-ray energies was investigated for two HEXITEC detectors, known as the Redlen A and Redlen B detector, which had been bonded to Redlen CZT crystals. Compensation signal correction was used on both detectors to enable spectroscopic performance. X-ray energies from 8keV to 140keV was used. Energies from 8keV to 67.2keV was produced by fluorescence of different metal foils (Cu, Mo, Ce and W) produced when exposed to monochromatic synchrotron light at station I15 DLS. Energies above this up to 140keV was produced by radiation sources (Co-57 and Tc-99m) used at the Royal Marsden Hospital.

Figure 6.71 shows the spectral pixel map of the Redlen B detector when exposed to fluorescence from a Cu foil. The characteristic radiation energies, Kα and Kβ, for Cu is 8.05keV and 8.91keV respectively. The Kα peak is clearly visible showing an improved effectiveness of the compensation signal correction when using Redlen CZT compared to eVPRODUCTS CZT and Acrorad CdTe, allowing for low energy X-ray detection. A significantly higher degree of uniformity is observed then seen for the ERD2004 detector using eVPRODUCTS CZT seen in figure 6.29. The Redlen CZT crystals can
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Figure 6.71: Spectral pixel map of the Redlen B detector when exposed to fluorescence from a Cu foil. The effectiveness of the compensation signal correction when using Redlen CZT allowed for detection of X-ray radiation down to at least 8keV. A high degree of spectral uniformity was observed as compared to the spectral response of the ERD2004 detector when using eVPRODUCTS CZT. A improved spectral uniformity at Mo and Ce energies compared to Cu energies was observed suggesting the detector suffered from low energy effects degrading the spectral response.

only be bought with passivation and electrode deposition already done. The reason for improvement in uniformity observed for the Redlen material can therefore not be easily investigated to determine if it is caused by better bulk material properties or surface treatment and electrode manufacturing. The spectral uniformity observed when exposing the detectors to Mo and Ce fluorescence, as can be seen in figure 6.72 and figure 6.73 respectively, is higher then observed when exposing the detector to Cu fluorescence. A possible reason for this could be the short mean interaction depth of X-rays at Cu fluorescence energies, forcing the electrons to traverse the entire thickness of the detector, increasing the electron trapping probability. The Cu fluorescence Kα peak is clearly visible in the spectra but not the Kβ peak. This is accredited to the small energy difference between the two, ΔE=0.86keV, not allowing them to be resolved due to the spectral resolution of the detector. Figure 6.72 shows the spectral pixel map, as well as the 3D Kα peak position and peak height maps, of the Redlen A detector when exposed to fluorescence from a Mo foil. A higher degree of uniformity is observed between pixels
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Figure 6.72: (Left) Spectral pixel map, (Right Lower) 3D Kα peak position pixel map and (Right Upper) 3D Kα peak height pixel map as observed for the Redlen A detector when exposed to Mo X-ray fluorescence. An improved inter pixel uniformity compared to the one found for Cu fluorescence was observed. During these measurements a cable fell down covering the lower left corner of the detector causing an attenuation of the X-ray flux in this region visible in both the spectral and peak height maps. A lower quality spectral response, unrelated to the cable attenuation, was observed for the left side of the Redlen A detector. The Kα and Kβ peaks where both visible in the spectra.

when exposing the detector to Mo fluorescence compared to Cu fluorescence. During the experiment a cable for the electronics fell down in front of the Redlen A detector on the left hand side causing attenuation of the incident X-rays. The effect can clearly be seen in figure 6.72 both in the 2D spectral pixel map as well as in the 3D Kα peak height pixel map. A poorer spectral response was found on the left side of the Redlen A detector, unrelated to the cable attenuation, throughout the experiments. The reason for this reduced spectral response for the Redlen A detectors left side is unknown but is not present for the Redlen B detector. Both the Mo fluorescence Kα (17.5keV) and Kβ (19.6keV), ΔE=2.1keV, peaks are visible in the spectra on the right hand side while for the poor spectral response region of the left side only the Kα peak is discernable.

Figure 6.71 shows the spectral pixel map, as well as the 3D Kα peak position and peak height maps, of both the Redlen B and Redlen A detector when exposed to fluorescence from a Ce foil. The left row of Redlen detector B was non-functional due to bonding problems. The wire in the lower left corner of the Redlen detector A is still present. A high degree of uniformity was seen for both detectors. The Redlen A detector had a lower spectral response on the left side, as stated before, but the Ce fluorescence Kα (34.7keV) and Kβ (39.3keV), ΔE=4.6keV, was visible in the pixels spectra of both detectors. A higher degree of uniformity for both detectors was observed for the Kα peak position compared to the Kα peak height. For the rTEDDI imaging technique peak
Figure 6.73: Spectral pixel map (a and b), as well as the 3D Kα peak position (d and f) and peak height maps (c and e), of the Redlen A and Redlen B detector respectively when exposed to fluorescence from a Ce foil. The left column of pixels for the Redlen B detector where non functional due to bonding issues. Both Kα (34.7keV) and Kβ (39.3keV) where visible in both detectors. A higher inter pixel uniformity was observed for the Kα peak position compared to the Kα peak height.
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positions is currently the main parameter of interest. The response uniformity with respect to the peak shape and height might be estimated more accurately by taking the integrated peak intensity of the Kα peak instead of the Kα peak height since the latter is more easily noise influenced. Figure 6.74 shows the spectral pixel map, as well as

![Spectral pixel map (a and b), as well as the 3D Kα peak position (d and f) and peak height maps (c and e), of the Redlen A and Redlen B detector respectively when exposed to fluorescence from a W foil. A Mo wire was placed in front of the Redlen B detector in order to verify the imaging capability of the detector. The resulting X-ray attenuation is clearly visible in both the spectral pixel map as well as the Kα peak position pixel map. Good uniformity was observed for both detectors, especially with regards to peak position uniformity. A better peak height uniformity was observed for the Redlen B detector compared to the Redlen A detector. The degradation in peak height uniformity observed for both W and Co-57 (Redlen A detector) suggest it was a high energy effect. For both detectors the Kα and Kβ peaks are visible.](image)
the 3D $\mathrm{K}\alpha$ peak position and peak height maps, of both the Redlen B and Redlen A detector when exposed to fluorescence from a W foil. A Mo wire was positioned in front of the Redlen B detector to test for imaging of the detector. The reduced flux caused by the wire can be clearly seen as a reduction of the number of total events in the spectra for the effected pixels in the spectral pixel map and as a strong reduction of the $\mathrm{K}\alpha$ peak height of the 3D $\mathrm{K}\alpha$ peak height map. The $\mathrm{K}\alpha$ peak position map is however not effected by the attenuation showing a uniform response across all pixels. An overall good spectral uniformity was found for both detectors, especially with respect to $\mathrm{K}\alpha$ peak position, at this energy. A relatively poorer peak height uniformity was observed for the Redlen A detector compared to the Redlen B detector. The increase in $\mathrm{K}\alpha$ peak height non uniformity between pixels observed for the Redlen A detector seemed to be true for Co-57 as well, seen in figure 6.75, suggesting it might be a high X-ray energy effect.

Both the $\mathrm{K}\alpha$ (59.3keV) and $\mathrm{K}\beta$ (67.2keV) peaks, $\Delta E=7.9$keV, of the Ce fluorescence is visible in the spectral response. A FWHM of the $\mathrm{K}\alpha$ peak (59.3keV) was found to be approximately 4keV. Note should however be taken to the fact that these results where obtained without any correction algorithms (charge sharing correction or 'end of frame' correction) having been applied, nor was the detector cooled as is normally done to approximately -5 to -30°C when using CZT detectors. Figure 6.75 shows the spectral pixel map, as well as the 3D $\mathrm{K}\alpha$ peak position and peak height maps, of the Redlen A detector when exposed to fluorescence from a Co-57 foil. Figure 6.76 shows the spectral

![Figure 6.75: (Left) Spectral pixel map, (Right Lower) 3D $\mathrm{K}\alpha$ peak position pixel map and (Right Upper) 3D $\mathrm{K}\alpha$ peak height pixel map as observed for the Redlen A detector when exposed to radiation from a Co-57 source. A spectral peak at 122keV is clearly visible in the spectral plots. A decrease in the spectral uniformity is visible in the right side of the detector.](image)
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Figure 6.76: (Left) Spectral pixel map, (Right Lower) 3D Kα peak position pixel map and (Right Upper) 3D Kα peak height pixel map as observed for the Redlen A detector when exposed to radiation from a Tc-99m source. Spectral peaks are visible at 140keV and 75keV (Pb fluorescence). Just as for the Co-57 radiation response a decrease in spectral uniformity is seen on the right side of the detector suggesting high energy effects are degrading the spectral response of the detector.

pixel map, as well as the 3D Kα peak position and peak height maps, of the Redlen A when exposed to fluorescence from a Tc-99m foil. A good Kα peak position uniformity was observed for both detectors. A peak at 122keV was observed for the Co-57 spectra measurement and two peaks, 140kev and 75keV (Pb fluorescence), was observed for the Tc-99m measurement. A decreased uniformity of Kα peak height was observed at both these two higher energies although not as strongly for the Tc-99m measurement. A more non uniform spectral response was also observed in the right hand side of the spectral pixel map for the Redlen A detector at these energies, suggesting both these observations are high energy phenomena. A possible explanation could stem from the high mean interaction depth obtained at these energies. The long transport distances needed for the holes under these circumstances and the poor hole $\mu\tau$-product in CZT [153] would result in hole trapping in the bulk material, resulting in spatially charged regions reducing the detector performance. This would be consistent with the cause of polarization in CZT observed under high X-ray flux conditions [110] [111] [112] [113] and in other material systems prone to polarization build up (i.e. HgI$_2$) [83].
6.3 Summary

In this chapter the work done on effects of substituting CZT for Si as the detector crystal for the ERD2004 detector has been described. The change of detector material was done to improve detection efficiency, facilitate detection of high energy X-rays and enable a wider detectable X-ray range to ensure essential improvements in scanning times needed, wider range of samples available for investigation and higher accuracy of refined atomic structural data respectively for the rTEDDI imaging technique. An in depth characterization of the ERD2004 detector resulting in specification changes to the HEXITEC detector system and subsequent initial testing of the HEXITEC detector has also been described. The characterization was done in order to produce a pixelated energy resolving detector capable of serving as one of the core modules of the rTEDDI system. As described in section 6.1.2 early EDR2004 CZT detectors where found to be suffering from a bias dependent malfunctioning of pixels, referred to as the black hole effect, which was corrected for in later ERD2004 CZT detectors by improvements in the device packaging process.

When exposing the ERD2004 CZT detector to radiation from known radiation sources (Am-241 dial source, Am-241 source) variation in spectral response compared to measurements done using Si as the active detector material was observed. This has been described in section 6.1.2. The changes in number of observed peaks and intensity of the observed peaks where explained by the increased detection efficiency and atomic constituents of the detection material when substituting to CZT crystals.

A high degree of inter pixel non uniformity was observed for the ERD2004 detector when using CZT as the active detector material that was not observed when Si was used ad has been described in section 6.1.2 and section 6.1.3. The degree of non uniformity observed would render the detector unusable in any quanti Using the linearity plot obtained for each pixel during detector energy calibration it was found that the spectral response did not only vary in between pixels at a specific energy but also varied within a single pixel when changing the energies of the X-ray detected. This was described in section 6.1.2. Pixels exhibiting spectral response variation with changing energy was classed into three groups of 'bad pixels' with the aid of the energy calibration linearity plots. Out of all the pixels for the ERD2004 CZT detector, using eVPRODUCTS grown CZT, approximately 30% was suffering from 'bad pixel' behaviour. The complete cause for inter pixel non uniformity in CZT detectors is currently not understood within the scientific community. Possible explanations include the effect of material imperfection such as Te inclusions [88] [93], cracks and twin boundaries [88] ; poor bonding quality and/or low inter-pixel resistance (poor passivation) [139].

Spectral improvement by charge sharing correction of the ERD2004 CZT detector, described in section 6.1.4, was found to be severely limited by the user defined voltage
Development of high Z detectors for rTEDDI imaging

of the comparator driven readout scheme inherent to the detector. The user defined voltage effectively set a hardware low energy cut-off of the detector system preventing charge sharing between pixels below the trigger level from being detected introducing an error into any correction done. Charge sharing discrimination was found to be a superior correction algorithm compared to charge sharing addition. The addition algorithm was found to introduce an error into the correction due to charge sharing events on neighboring pixels not being detected that was significantly higher than for the discrimination script, although the discrimination script was also affected. The limitation to charge sharing correction set by the comparator circuit in the ERD2004 detector was circumvented in the HEXITEC detector design by using a rolling shutter readout scheme. No low energy hardware cut-off is consequently present in the HEXITEC ASIC enabling better charge sharing correction.

As described in section 6.1.5 the countrate capability of the ERD2004 detector system was found to be severely limited by the DAQ ability to handle large data amounts. The inability of the DAQ system to handle large data amounts resulted in very high detector deadtimes in between global reset periods of the detector. The optimum countrate capability was consequently achieved by setting the 'number of read per frame' parameter constant (approx 800) and adapting the clocking frequency of the system to the X-ray flux. At higher X-ray fluxes optimum countrate is achieved by increasing the clocking frequency, effectively minimizing the active global reset period. If two events would take place, for the readout channels of the ERD2004 detector, during the same local reset period the value on the peak hold circuit would take a random nature \[145\]. If more than two events occurred during the same local reset period the readout chain would freeze, becoming non responsive \[145\]. Investigations using synchrotron X-ray diffraction showed that the incoming beam had to be attenuated (3mm Pb attenuation) to a small fraction of the normal beamprofile in order for the ERD2004 detector local reset speed to be sufficient to produce accurate spectroscopic performance, making the detector unsuitable for rTErrD imaging at higher flux stations. At higher X-ray fluxes a nonsensical spectral response was observed. The spectral response obtained showed strong similarities to the spectral response found during the unsuccessful rTEDDI imaging test on station 16.3 SRS Daresbury, described in section 4.2.5, suggesting this was the cause of the unsuccessful imaging trial. A further increase of the X-ray flux rendered the ERD2004 CZT detector completely non responsive. The HEXITEC detector was consequently designed to be able to handle countrates up to 1.560 counts*sec\(^{-1}\)*pixel\(^{-1}\). The countrate was assumed for a 10% occupancy rate making the probability of two events occurring for a single pixel within the same reset period highly unlikely. If two or multiple events occurring during the same readout the HEXITEC ASIC is designed so the highest energy X-ray is detected. The design of the HEXITEC ASIC is however susceptible to problems with preferential high energy sensing during high flux conditions.
and partially developed signals during high clocking frequencies.

Initial testing of the HEXITEC detector system described in section 6.2.2 showed an inability to select region of interest scans, a DAQ deadtime of approximately 90% and a lack of spectroscopic performance. The inability to perform region of interest scans was found to be due to a ASIC design error of the filler cells resulting in short circuiting when applying region of interest setting for the pixel rows. This error was consequently corrected in the second iteration of the HEXITEC ASIC design (not discussed in this thesis).

The 90% deadtime of the DAQ system was due to the structure of the data transfer from the ASIC to the control PC resulting from a miscommunication during system specifications stage. The error was consequently corrected by upgrading the DAQ system leading to a close to 0% readout deadtime.

The lack of spectroscopic performance was found to be caused by a periodic large current pulse temporarily increasing the potential of the ground, creating a ground bounce, resulting in the induction of a large noise signal on the readout channels. A temporary solution was devised, in order to allow testing of spectral performance, by sending in a pulse of opposite polarity, known as the compensation signal, to cancel out the noise pulse. The compensation signal correction was tested in order to find optimal correction settings with respect to start, cancellation and signal magnitude. The effectiveness of the compensation signal correction was found to be material dependent with no spectroscopic performance available in spite of correction when using Acrorad CdTe, spectroscopic performance for X-ray energies above 22keV when using eVPRODUCTS CZT and no observed limitation in spectral response due to the ground bounce noise signal when using Redlen CZT. The reason for the material dependence in the effectiveness seen when using compensation signal correction is not currently understood. Possible causes could be differences in leakage current or a change in the input impedance of the pre-amplifiers when using different materials. The problem of the ground bounce induced noise signal has been corrected for the second iteration of the HEXITEC 20x20 pixel ASIC (not discussed in this thesis), eliminating the need for compensation signal correction.

Measurements on the inter pixel uniformity of the HEXITEC detector bump bonded to a piece of Redlen CZT (see section 6.2.3) showed a inter pixel spectral uniformity unprecedented in the literature. If the improved inter pixel uniformity seen for the Redlen CZT is a cause of bulk material properties or processing is currently unknown. The varied spectral response within a single pixel when changing the energies of the X-ray detected seen for the ERD2004 eVPRODUCT CZT detector was not observed for the HEXITEC Redlen CZT detector. An indication of low (around 8keV) and high (around 122keV) energy effects leading to degraded inter pixel spectral uniformity where observed for the
HEXITEC CZT detector and has a possible cause in the trapping of electrons and holes respectively due to the mean interaction depths of X-rays in these energy regions. Problems in the long times needed to analyze and visualize the HEXITEC detector data using purposely written Matlab m-scripts where found, and has been shown in section 6.2.2, with simple All plots of 150,000 frames and full histogramming range taking 4.6 hours to produce. For practical considerations visualization of the data collected using any detector being part of rTEDDI system has to be able to be viewed 5-10 minutes after finishing a scan. A short term solution was devised by adapting the EasyEDD program (based on C++ coding) to be able to handle basic visualization of the HEXITEC detector data. A long term solution to data visualization and analysis for the rTEDDI system, using the HEXITEC detector, has been specified (see Appendix D) and submitted as part of a grant application.
Chapter 7

Manufacturing and evaluation of array collimators

At the start of this project an array collimator, known as the MK1.2 collimator, had already been developed and tested under low energy/low flux synchrotron radiation conditions with positive results. This array collimator was successfully used in order to obtain the proof of concept for the rTEDDI imaging technique, described in section 4.2. The inability to produce rTEDDI images under high flux/high energy synchrotron conditions, described in section 4.2.5, resulted in a re-evaluation of the collimator design under these circumstances.

This chapter describes the development, initial (low flux/low energy) evaluation and re-evaluation (high flux/high energy) of the MK-line of collimators. The results of the investigations led to the development of other array collimator designs of which the development and initial testing is also described. Re-evaluation of the MK1.2 collimator, development and testing of the alternative collimator structures where done in collaboration with Mr. Olivier Lazzari, Dr. Simon Jacques and Mr. Taha Sohi from University College London/Birkbeck College. This chapter has been co-written with Mr. Olivier Lazzari and sections of this chapter will occur in Mr. Lazzari’s PhD thesis as well as here. Their contributions are made clear in the

7.1 The MK line of array collimators

At the start of this project a line, known as the MK-line, of tungsten plate array collimators had been developed. Collimators from the MK line are referred to as the W (for tungsten) collimators. This section deal with the testing and evaluation of these collimators under low and high energy synchrotron X-ray radiation.
7.1.1 Manufacturing of the MK-line of array collimators

A number of aspects were considered in the specification of the W collimators. The collimators had to be in an array structure with a 300\(\mu\)m pitch in order to allow it to be coupled to the ERD2004 detector, enabling rTEDDI imaging. The bulk material that was used had to have a high mass attenuation coefficient in order to allow a high degree of attenuation in the solid part of the collimators. No cross talk between collimators could be allowed. The aspect ratio for each collimator (collimator length to diameter ratio) had to be sufficient, approximately 6000:1, to limit the beam divergence enough to obtain an energy resolution matching the ERD2004 detector (approximately 250eV when using Si). A number of different manufacturing methods where considered [24] including photochemical and microforming processes (LiGa) [154]; chemical machining; micro electron-discharge-machining and laser based machining [155]. Non of these techniques allowed for holes (individual collimators in the collimator array) being drilled with an aspect ratio higher then 500:1. A layered collimator array structure was therefore decided upon where a number of thin, 100\(\mu\)m, W plates, each having an array of holes drilled into it, where aligned at specific intervals to make sure no cross talk occurred. The collimators where manufactured using a new manufacturing principle, patent pending, where femto second pulse duration laser drilling was used to simultaneously drill and align the W plates. The laser drilling technique was used instead of the other mentioned techniques due to the higher drilling speed possible. Three W collimators, MK1.1, MK1.2 and MK1.3 were manufactured.

<table>
<thead>
<tr>
<th>Collimator</th>
<th>No of W plates</th>
<th>hole size ((\mu)m)</th>
<th>Total collimator length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MK1.1</td>
<td>11</td>
<td>50</td>
<td>330</td>
</tr>
<tr>
<td>MK1.2</td>
<td>6</td>
<td>50</td>
<td>80</td>
</tr>
<tr>
<td>MK1.3</td>
<td>11</td>
<td>75</td>
<td>400</td>
</tr>
</tbody>
</table>

Table 7.1: The variation in number of W plates used, the hole size (collimator diameter) and total length for the three W collimators MK1.1, MK1.2, and MK1.3.

Figure 7.1: Photographs of the three W collimators (Left) MK1.1, (Centre) MK1.2 with the collimator array region of a W plate blown up and (Right) MK1.3. The MK1.2 collimator was used to obtain the proof of concept for the rTEDDI imaging technique described in section 4.2.

MK1.2 and MK1.3 where manufactured. The variation in number of W plates used, the
hole size (collimator diameter) and total length for the three collimators can be seen in table 7.1. The three collimators can be seen in figure 7.1.

### 7.1.2 Low energy X-ray testing of the MK-line collimators

The W collimators was initially performance tested by Dr. Kern Hauw Khor on the low flux/low energy synchrotron station 7.6 SRS Daresbury to evaluate their performance. The experimental setup can be seen in figure 7.2. The collimator transmission angle was tested, for each of the three W collimators, by illuminating the front of the collimator using a 6mmx6mm white box beam and monitor the transmitted signal detected at the end of the collimators as a function of both pitch and yaw angle. The beam size was subsequently set to 25µmx25µm while at the optimum transmission angle and the collimator was step scanned in x-y direction in order to test the spatial transmission profile. Collimator MK1.1 showed both cross fire between collimators and satellite transmission paths attributed to non circular holes and plate misalignment and was consequently discarded. The observed transmission angle and spatial transmission profile for the collimator MK1.2 can be seen in figure 7.3. A transmission angle FWHM of 0.05° was
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Figure 7.3: (Left) Spatial transmission function, (Right Top) 3D plot of the transmission angular dependence and (Right Bottom) 2D plot of the transmission angular dependence for the MK1.2 collimator array. An angular acceptance angle of 0.05° FWHM was observed with a majority of the collimator holes showing good hole forming.

observed, exceeding the expected FWHM of 0.06°. The higher observed angular resolution was attributed to overlapping effects in the alignments of holes between W plates. The spatial transmission profile showed a majority of holes being well formed. The minority of holes displaying lower X-ray intensities are believed to have been caused by W debris from the drilling partially blocking the holes. The observed transmission angle and spatial transmission profile for the collimator MK1.3 can be seen in figure 7.4. A transmission angle FWHM of 0.01° was observed with an expected FWHM of 0.02°. The spatial transmission profile showed two rows of collimators in the collimator array missing. The cause of this was found to be an error in the programming of laser drill process. The collimators observed in the spatial transmission profile exhibited good hole formation with an increased intensity compared to MK1.2 as would be expected due to the increase in hole diameter of the collimators. Collimator MK1.3 was later found not working as a collimator. The reason for this is believed to be due to misalignment of the final W plate caused by a blow to the cross beam holding the plates occurring during transport after these initial tests [156]. The one working collimator, W collimator MK1.2, was used for the successful proof of concept test for the rTEDDI imaging technique on the low flux/low energy synchrotron station 7.6 SRS Daresbury, described in section 4.2. However, the inability to produce rTEDDI images under high flux/high energy synchrotron conditions, described in section 4.2.5, resulted in a re-evaluation of the collimator design under these circumstances.
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7.1.3 High energy X-ray testing of the MK-line collimators

The spectral collimation ability of the MK1.2 collimator was tested on the high flux/high energy station 16.4 SRS Daresbury in order to test the ability to collimate under these conditions. The experimental setup can be seen in figure 7.5. A white X-ray beam having a square cross section penetrates a Potassium Chloride (KCl) sample. The horizontal and vertical beam sizes of the incoming beam are referred to as Bh and Bv respectively. The resulting diffraction X-ray beam consequently irradiates the front end of the collimator array. For an ideal collimator array only X-rays traversing through the collimator holes emerge from the back end. An energy resolving detector situated a distance away from a slit is positioned behind the back end of the collimator array selecting a area that is monitored. The diameter of the slit, referred to as Ds, typically measures 50m for the final setup. A 2D, spectroscopically resolved, scan of the of the transmission function at the back end of the collimator is realized by moving the collimator in steps equal to Ds along the Y and Z axes. The detector used was an Amptek XR-100T-CdTe detector [157] having a 3mmx3mmx1mm piece of CdTe as the active detector crystal. For these experiments, the detector was shielded with 2 thick layers of copper and lead. The heavy lead plating first stops most X-rays, while the copper layer then absorbs the fluorescence photons emitted by the lead. A number of alignments has to be successfully...
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Figure 7.5: Experimental EDD setup used to re-evaluate the performance of the MK1.2 W collimator. A cross sectional side view (Left Top) and top view (Left Bottom) of the setup is shown as envisioned for a solid (non layered) collimator array. The incoming white synchrotron X-ray beam is shown in light green; the sample is pink colored; the diffracted X-ray beam is shown in blue and the beam entering through the slit to the detector is shown in dark green. The view from the back end of the collimator (Right) shows the effect of the different geometrical relationships of the setup. The height of the diffracted beam is mainly set by the thickness (in the direction of the incoming beam) of the sample while the width is set by Bh. The dark green spot shows the monitored area of the back end of the array collimator.

carried out for this setup. The collimator field of view has to be aligned the diffraction lozenge created by the interaction of the sample with the incoming beam. The acceptance angles created by the detector crystal surface area and the slit has got to be aligned with acceptance angles of the collimators in the collimator array. When correct alignment has been found a diffraction pattern should be observed when monitoring a collimator hole. The spectral response observed for the MK1.2 detector can be seen in figure 7.6 when using a KCl. The acceptance angle of the slit was increased, resulting in the monitoring of multiple collimator holes, to ensure the detection of a diffraction signal if one came out at the back end of the collimator. No diffraction peaks were visible in the spectral response observed showing the collimator array does not collimate at these energies. A single spectral peak was observed having a maximum value at approximately 67keV. An explanation to the spectral response can be obtained from the X-ray transmission through 600µm W as a function of energy as can be observed in figure 7.7. The increase in transmission leading up to the absorption edge for 600µm W is attributed to be the observed spectral response. The peak shape correlation between the observed spectra and the transmission spectra is in agreement. The difference in peak value is believed to be due to imperfect energy calibration of the measured spectra and the reduced beam flux with increased X-ray energy in this region. The observed spectral response is
Figure 7.6: The spectral response observed for the MK1.2 W collimator when using a KCl sample. The acceptance angle of the detector-slit configuration was increased, making the area monitored span several collimator holes, in order to ensure detection of any diffraction emerging from the back end of the collimator array. The change did not alter the spectral response observed. The observed spectra did not correlate to the expected diffraction spectra of KCl.

Figure 7.7: Calculated I/I₀ as a function of X-ray energy for a 600µm W plate. An increase in transmission leading up to the absorption edge of W is observed. The response provides an explanation to the observed spectra seen in figure 7.6. The inability of the MK1.2 W collimator to collimate under high flux/high energy conditions is attributed to the increase in transmission leading up to the absorption edge of W in combination with the geometrical layout of the W plates with respect to the collimator hole vs. W surface area. For the W collimator array design an increase in the thickness of the W plates or increase in the number of plates used would rectify an error of this nature.

believed to be due to the relatively high transmission signal (I/I₀=0.05 of the incoming X-rays at 69.5keV); the wide spectral range of the transmission signal and the area of
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holes vs. W in the W plates (2% to 98% respectively for just in the collimator array region seen blown up in the central image of figure 7.1). An increase in the attenuation of the array bulk material would correct for this problem and could be done with the W collimator design by either increasing the thickness of the W plates or increasing the number of plates used. In the current form the MK1.2 W collimator does however not collimate under high energy X-ray conditions.

7.2 Alternative collimator structures

Although the MK1.2 W collimator failed to perform under high flux/high energy synchrotron conditions the layered W design is believed to be valid if an increased attenuation is achieved. The design does however suffer from a low production yield (33%) and the manufactured collimators are very sensitive to physical strain making them hard to transport and handle. Solid collimator array designs have therefore been investigated. A Pb glass collimator matrix has been manufactured by Collimated Holes, Inc. [158] on behalf of the advanced characterisation techniques group at the University of Manchester’s Material Science Centre. A Pb-Bi cast collimator array and the method of manufacturing it has been developed by Mr. Olivier Lazzari at the University College London/Birkbeck College.

7.2.1 Manufacturing of the lead glass array collimator

The Pb glass collimator is manufactured by stacking 16x16 glass capillaries, referred to as the glass matrix, which is then heated and drawn into the desired shaped and enclosed in a glass casing used to enhance the structural integrity of the collimators. The front view of one of the manufactured glass collimators can be seen in figure 7.8 [159]. The matrix glass formula can be seen in table 7.2. The capillary hole diameter is 60µm and

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Wt %</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>45.5</td>
</tr>
<tr>
<td>PbO</td>
<td>44.7</td>
</tr>
<tr>
<td>BaO</td>
<td>0.18</td>
</tr>
<tr>
<td>NaO</td>
<td>3.02</td>
</tr>
<tr>
<td>K₂O</td>
<td>6.43</td>
</tr>
<tr>
<td>As₂O₃</td>
<td>0.28</td>
</tr>
</tbody>
</table>

Table 7.2: Matrix glass formula for the Pb glass collimators.

the matrix has a 300µm pitch allowing it to be coupled to the ERD2004 detector. A long collimator was drawn and subsequently broken of at desired intervals to produce the
collimator lengths wanted. Three primary collimator lengths, 10, 15 and 20cm, where produced. A photograph of the 20cm Pb glass collimator can be seen in figure 7.9.

7.2.2 Manufacturing of the lead bismuth array collimator

In addition to the glass collimators the decision was made to construct a second prototype collimator which was done by Mr Olivier Lazzari. Its conception is different from that of the previous collimators, as this one is cast. A mold is formed by passing steel wires, whose diameter corresponds to that of the future holes (100µm), through two
laser-drilled plates similar to those used for the W collimator. The wires are coated with silicon oil. Here the plates, distant of each other by 15cm, are used as guides that ensure the wires are perfectly parallel. About one gram of finely ground Sodium Chloride (NaCl) is added onto the bottom laser-drilled plate to ensure it is leak tight. The external walls of the mold are assembled around the plates and wires. A weight of approximately 150g is suspended to each wire to induce a tension and make sure it follows a straight line. The mold is then placed vertically in an oven and heated along with the low-melting metal alloy that will constitute the future casting. The melted metal is poured into the mold, where it solidifies within a minute as the temperature of the oven decreases. Finally, the cast is separated from the mold and the steel wires removed carefully, thus forming the desired collimator. For the making of the mold, a 100µm diameter stainless steel 302 wires, ordered from the California Fine Wire company [160], was used. 1kg of 58/42 Pb-Bi alloy bars, whose melting point is 168°C, was purchased from Alec Tiranti Tools Inc [161]. Two tungsten plates were laser-drilled by Kun Li, a PhD student in Dr Bill O’Neill’s research group at the Institute for Manufacturing, Cambridge University. These plates contain a grid of 16x16 holes whose diameter is about 120µm, which is slightly bigger than that of the steel wires. The thickness of the tungsten plates is 500µm and the spacing between the centers of two consecutive holes is 300µm. SEM pictures of one such plate are shown in figure 7.11. The external walls of the mold and the weights, made of brass, were machined by Paul Stukas from the Workshop at Birkbeck College. Two collimators were produced with this method: the first one with just one hole, and the second one with 4 holes forming a square with a side of 600µm. They both measure about 13cm in length. Only the first collimator was imaged due to time restraints during synchrotron ‘beamtime’.

Figure 7.10: (Left) Schematics of the mold of a 2x2 holes collimator. (Right) The equipment used to make the collimator. 1) 100µm steel wire spool. 2) Bar of lead-bismuth alloy. 3) Laser-drilled plate. 4) Body of the mold, made of brass. 5) Brass weights.
7.2.3 High energy X-ray testing of the lead glass and lead bismuth collimators

The Pb glass collimator was initially tested alongside the MK1.2 W collimator during the experiment shown in figure 7.5. A large acceptance angle of the detector slit geometry was used to find proof of diffraction on the expense of imaging. The resulting spectral response for the 10cm Pb glass collimator, using a KCl powder sample, can be seen in figure 7.12. The peak pattern was recognized as KCl diffraction [162]. No discernible diffraction patterns were observable when using the 15 and 20cm Pb glass collimators. The experimental setup seen in figure 7.5 was reproduced at station I15 DLS in order to
evaluate the Pb-Bi collimator and further investigate the 10cm Pb glass collimator. A portion of the cross-section of the 10cm Pb glass collimator was scanned. The horizontal beam size \(B_h\) was 200\(\mu m\). The vertical beam size \(B_v\) was 600\(\mu m\). The slit size \(D_s\) was 50\(\mu m\). The vertical and horizontal step sizes were 30\(\mu m\). The 2D scan is made of 33 lines of 47 pixels each. The total scanned area measures about 1x1.4mm and comprises 20 holes out of 256. The collection time per pixel was 10 seconds, for a total collection time of about 4h30mn. A 3D total intensity map of the scan can be seen in figure 7.13. An array structured increase in total intensity can be clearly seen suggesting an increase in the number of detected X-rays when the detector is situated over a glass capillary. The majority of intensity peaks are situated 9-10 steps (approximately 270-300\(\mu m\)) from their neighbors suggesting a close to 300\(\mu m\) collimator pitch. A variation in peak intensity position between peaks in the same row had a maximum value, within the region scanned, of 3 steps (approximately 90\(\mu m\)) while a maximum difference in columns where found to be 2 steps (approximately 60\(\mu m\)). A 'beam crash' occurred during the scan resulting in the drop of base intensity seen at row 15. The drop in intensity gives and indication of the background noise present despite the precautions taken to eliminate it. A 2D scan of the Pb-Bi collimator (single hole) was done as well. For this 2D scan over the Y and Z axes, the detector slit diameter, the vertical and the horizontal beam sizes were equal to 200\(\mu m\). The sample was again KCl powder. The vertical and horizontal step sizes were 20\(\mu m\). The 2D scan is made of 19 lines of 21 pixels each. The collection time for one pixel was 15 seconds. The total collection time was about 1h

Figure 7.13: Total intensity map of a section of the back end of the 10cm Pb glass collimator. A 300\(\mu m\) pitch between maximum intensities where observed with maximum spatial variation of peak intensities within rows being 90\(\mu m\) and columns 60\(\mu m\). the drop in intensities with increased row number is believed to be due to beam decay. The drop in observed minimum intensity seen at row 15 was due to a 'beam crash'.
45mn. The spatial total intensity map as well as selected spectral responses observed for the 10cm Pb glass collimator and the Pb-Bi collimator can be seen in figure 7.14 and figure 7.15 respectively. The shape of the hole observed in figure 7.15 is regular and its size corresponds to expectations. For both collimators a clear set of spectral peaks starts to evolve when moving into the assumed collimator holes that are not present outside of the holes. The spectral response is exactly the same for both collimators as would be expected since the same KCl powder sample was used. Using the d-spacings of the crystal planes in KCl the expected energies for which diffraction peaks would occur was calculated using a 2θ-angle of 6° and can be seen in table 7.3.

<table>
<thead>
<tr>
<th>Crystal Plane</th>
<th>d-spacing (Å)</th>
<th>Diffraction peak energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(111)</td>
<td>3.632</td>
<td>32.613</td>
</tr>
<tr>
<td>(200)</td>
<td>3.145</td>
<td>37.663</td>
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<tr>
<td>(220)</td>
<td>2.224</td>
<td>53.260</td>
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<tr>
<td>(311)</td>
<td>1.897</td>
<td>62.441</td>
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<tr>
<td>(222)</td>
<td>1.816</td>
<td>65.226</td>
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<tr>
<td>(400)</td>
<td>1.573</td>
<td>75.302</td>
</tr>
<tr>
<td>(331)</td>
<td>1.443</td>
<td>82.086</td>
</tr>
<tr>
<td>(420)</td>
<td>1.406</td>
<td>84.246</td>
</tr>
<tr>
<td>(422)</td>
<td>1.284</td>
<td>92.251</td>
</tr>
</tbody>
</table>

Table 7.3: The d-spacing and resulting diffraction peak energy of the different crystal planes of KCl as expected for a 2θ-angle of 6°.
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Figure 7.15: Result of the 2D scan of the lead-bismuth collimator hole and detail of the patterns of 3 pixels. The background observed at high energy is most likely due to insufficient shielding of the detector. The patterns are displayed in counts as a function of the channel number (1024 channels with the Amptek XR-100T-CdTe detector), and the 2D scan in total count number as a function of the step number along the Y and Z axes. A KCl powder sample was used. The spectral response observed for both the Pb glass and Pb-Bi collimator are the same.

Figure 7.16: Spectral response observed for the holes in the collimator array as a function of energy. Three main peaks where observed at 47.35keV, 2.77keV and 16.6keV. These peaks to not correspond to the ones expected from diffraction of the crystal planes of a KCl powder sample. The reason for the unknown observed spectral peaks are currently not understood and need further investigation.

...
for this indiscrepancy and the origins of the observed spectral peaks are currently not understood and need further investigations.

7.2.4 Manufacturing of the chemically etched molybdenum array collimator

A third alternative collimator array design has just started to be investigated. The method intends to stack chemically etched Mo plates on top of each other in order to build a solid array collimator. The Manchester group has contacted Photofabrication Limited [163] to manufacture the collimator. Initial designs of the mask to be used to etch the Mo plates can be seen in figure 7.17. 4 different masks are under consideration with different, 75, 100, 150 and 200µm, collimator hole diameter. The reasoning behind the different diameters are the desire to enable testing of minimum diameter that enables the stacking process to produce acceptably uniform collimator holes. At the higher diameters possible cross talk between collimators due to poor inter collimator attenuation is considered a possible concern.

![Etching masks designed by Photofabrication Limited on behalf of the Manchester group. The development of a third alternative collimator design has been started in order to try and find a working concept to get a high attenuation solid collimator array. This design envisions stacking of a large number of chemically etched Mo plates together in order to get a collimator array. Four masks with different collimator hole diameter have been designed to enable testing of the design concept.](image-url)
7.3 Summary

In this chapter the testing of the W collimator design under low flux/low energy as well as high flux/high energy synchrotron conditions has been described. Manufacturing and testing of 2 alternative collimator array design has also been shown and the plans of a third solid collimator array design discussed.

Three W collimators, MK1.1, MK1.2 and MK1.3, where initially investigated under low flux/low energy synchrotron conditions. The MK1.1 W collimator where shown to be suffering from cross fire between collimators and satellite transmission paths attributed to non circular holes and plate misalignment. The MK1.2 collimator, used to obtain the proof of concept for the rTEDDI imaging concept described in section 4.2, showed a transmission angle FWHM of $0.05^0$ while exhibiting good collimator hole formation on the majority of holes. The MK1.3 collimator initially showed a transmission angle FWHM of $0.01^0$ and good collimator hole formation on the majority of holes. Collimator MK1.3 was tested again later and did no longer work. The reason for this has been attributed to misalignment of the last W plate due to physical shock to the device.

When MK1.2 collimator showed an inability to work as a collimator under high flux/high energy conditions which is believed to be a result of the decrease in attenuation at energies leading up to the absorption edge of W and the geometrical ratios of hole area to total area of the W plates used in the design. this effect is believed to have attributed to the inability of the rTEDDI system to image under high flux/high energy conditions as described in section 4.2.5. It is believed that an increase in the number or thickness of W plates would remedy this problem making the W collimator array design successful under high flux/high energy conditions as well as low flux/low energy ones.

The layered design of W collimator array design was however found to be suffering from both low production yield (33%) and shock sensitivity, making it both economically and practically problematic. Two initial solid collimator designs was consequently designed, manufactured and tested. The shorter, 10cm, Pb glass collimator, manufactured by drawn Pb glass capillaries, showed an ability to collimate under the same conditions at station 16.3 SRS Daresbury under which the MK1.2 collimator failed, while the two longer versions, 15 and 20cm showed no collimation. The Pb glass collimator where found to have an array structured increase in total intensity with a pitch of 300µm showing a working attenuation profile as expected. A variation in peak intensity position between peaks in the same row of the matrix had a maximum value, within the region scanned, of approximately 90µm while a maximum difference in columns where found to be approximately 60µm. The shape of the hole observed for the Pb-Bi collimator, manufactured by casting, showed a regular size corresponding to expectations.

The setup used to initially verify collimation taking place using the 10cm Pb glass collimator was replicated at DLS in order to test both the Pb glass and and Pb-Bi collimator.
The spectral response obtained for both collimators where found to be the same but did not correspond to the expected diffraction pattern of the sample (KCl powder sample). Nor could the spectral response be explained by fluorescence from any of the constituent atoms of the collimators or Cd and Te escape peaks from the detector. The spectral response observed is currently not understood.

The spectral response corresponding to KCl diffraction observed for the 10cm Pb glass collimator at SRS Daresbury showed that collimation is taking place. The spatial response found for both of the alternative collimator designs would suggest the same. The spectral response observed for the collimators at DLS is however not understood. Further investigation is therefore needed in order to verify if the collimator designs work as collimator array devices. Investigation of a third design method, using chemical etching, has also started.
Chapter 8

Conclusions and Further Work

It has been shown that the fields of X-ray imaging and X-ray diffraction, both important fields in their own right, can be coupled together to create a powerful X-ray diffraction imaging technique in the form of TEDDI. A major drawback of TEDDI imaging is the long time spans needed to produce 2D and 3D images. In order to overcome this obstacle the rTEDDI imaging system has been developed as a focus of this PhD project.

The rTEDDI system was realized by coupling an array collimator (MK1.2, as described in chapter 7) with a pixelated energy resolving detector (the ERD2004 detector described in section 6.1.1). The coupling of the collimator and detector in combination with shaping of an incoming X-ray beam allowed for an entire plane of a sample to be diffraction imaged simultaneously allowing for images to be produced a lot faster. A proof of concept was successfully obtained for the rTEDDI imaging system, using Si as the detector detection media, on a low energy/low flux synchrotron station (Station 7.6 SRS Daresbury). Metal (Al and Cu), Polymer (Nylon-6) and tribecular bone samples where all diffraction imaged using the rTEDDI system. Only thin samples where imaged.

It is highly desirable to be able to image thicker samples, such as jet engine turbine blades and friction stir welds, and would be of great interest to industry. In order to be able to image thicker samples higher energy X-ray need to be utilized. The detection efficiency for Si is however very poor for X-ray energies exceeding 20-25keV. In order to overcome this problem the detection media of the ERD2004 detector was changed from Si to CZT. CZT, being a high Z semiconductor material, has a higher detection efficiency for X-rays and a wider range of X-rays for which the detection efficiency is high. This would allow for thick samples to be imaged using the rTEDDI system. A wider range of detected X-rays would also result in a higher accuracy of refined sample lattice parameters assuming the same detector energy resolution for the two media.
rTEDDI imaging using the ERD2004 CZT detector failed to produce diffraction images on a high energy/high flux synchrotron beamstation (station 16.3 SRS Daresbury). During investigations into the ERD2004 CZT detector and MK1.2 array collimator performances the lack of diffraction imaging was found to have a two fold cause. The ERD2004 detector’s inability to handle high enough countrates, described in section 6.1.5, resulted in erroneous energy values or non responsive channel behavior during high flux synchrotron radiation exposure. At energies approaching the absorption edge of W (69.5keV) the attenuation caused by the W plates of the MK1.2 array collimator was substantially decreased. For a X-ray beam energy profile where a high degree of X-rays were at these energies, such as for station 16.3 SRS Daresbury, a strong signal could be transmitted through the collimator, effectively swamping the diffraction signal (Thereby destroying the collimator function).

As a solution to the problem the HEXITEC detector was developed (described in section 6.2). This detector has a read out that can handle a much higher countrate.

A number of limitation investigations and correction schemes for the HEXITEC detector have not yet been developed and need further work. Due to the read out scheme of the HEXITEC detector preferential high energy sensing, as described in section 6.2.1, would be an issue under to high flux conditions. There is no way to detect this effect other then a shift towards higher energy in the spectra. Investigations are consequently necessary in order to test for maximum allowable X-ray fluxes of the incoming X-ray beam under rTEDDI imaging conditions. 'End of frame'- (see section 6.2.1) and 'charge sharing correction'-algorithms has still got to be developed and tested for the HEXITEC detector.

The very significant problem of inter pixel non uniformity and variation in pixel response with changed energy observed for the ERD2004 CZT detector when using eVPRODUCTS grown CZT seems to have been solved to a large extent by using the HEXITEC ASIC bonded to Redlen grown CZT. If a higher degree of inter pixel uniformity should be needed a correction (i.e. multiplication) factor for each energy of each pixel, using a specific pixel response as a reference, could be tried. Possible low and high X-ray energy effects where observed for the HEXITEC CZT detector and should be further investigated.

As has been seen in the literature CZT grown by different manufacturers have shown different bias voltage and temperature settings yielding optimum detector performance. For each detector to be used in the rTEDDI system the optimum bias voltage and temperature has to be systematically determined.

At the moment no temperature control system exist for the HEXITEC detector system. One is currently being developed by NASA’s Marshall Space Flight Center, who are
interested in using the HEXITEC detector system in conjunction with their X-ray high-
resolution grazing-incidence optics [164]. Hopefully this temperature control system will be compatible with the rTEDDI system, otherwise another temperature control system will have to be developed independently.

An increased number of tungsten plates in the MK1.2 collimator array design is believed to be able to solve the high energy exposure collimator problem due to the resulting increase in X-ray attenuation. The fragile nature of the current collimator array design does however make a solid collimator array design much more desirable.

A number of solid array collimator designs have been developed and tested during this PhD with encouraging although not conclusive results (see chapter 7). Further investigations are needed into the response of the collimator.

With the observed initial results of the HEXITEC detector and promising findings for a number of solid collimator array designs it is the authors belief that the completion of a high energy/high flux capable rTEDDI system is technically close to completion pending the proposed further work aspects described earlier in this section.

Investigations into both collimator designs and detector performance could be expanded in the future at Manchester and at RAL and Diamond light source. A number of computer controlled xyz- and rotation-stages have been purchased in order to produce an automated control system (the rTEDDI control system). This system could initially be developed to be set up at the newly opened, 2.5M EPSRC funded, Henry Moseley X-ray Imaging Facility at the University of Manchester’s Material Science Centre. The number of stages should allow for a system that can be used for advanced in-house collimator testing and detector testing (spatial and flood field measurements at varied flux rates). It would also be able to work as a stand alone control system for the second generation rTEDDI imaging system, allowing for the system to be transported between locations.

A few issues with respect to DAQ, visualization and refinement and the rTEDDI control system for the future rTEDDI system should be considered. Practically any rTEDDI system situated on a synchrotron beamline will have to be able to visualize the data within 5-10 minutes after the end of the experiment. This in order for the experimenter to be able to interpret what is happening with the sample and logically proceed with the next experiment depending what has been discovered without spending hours visualizing the image, losing valuable experimental time. The TEDDI Data analysis requirements specifications (see Appendix D) has therefore been developed (with the HEXITEC detector in mind) by the author in collaboration with D-soft solutions. The proposed system will allow for swift data handling and visualization. A data rate of 320Mb/sec
is required to be handled and visualized. The specification of the Data analysis system calls for a 3rd party program or a specifically designed program to Rietveld refine the visualized EDD data, ideally as an instantaneous step in the data visualization, and allowing for a second visualization of the refined parameters.

One refinement issue that has been problematic for both the rTEDDI data, described in chapter 4, and the TEDDI data, described in chapter 5, is the selection of peak shape for fitting. In order to achieve a good Rietveld fit a correctly defined peak shape (Gaussian, Lorentzian, Pseudo Voight etc.) has to be specified. Due to the current restraints in CZT material parameters and its effect on the CIE uniformity, described in chapter 3, peak tailing will occur for spectra collected using this detector media. The development of a new peak shape function specifically tailored for the HEXITEC CZT detector would therefore be advisable. One possible route to take would be to fit the lower part of the peak using one specified function and the upper half using a different function. This issue has been briefly discussed with Dr. Simon Jacques (UCL/Birkbeck).

The TEDDI data analysis system, the ASPECT DAQ (used as the HEITEC detector DAQ system) and the rTEDDI control system will have to be controlled together from a single user interface in order to make the rTEDDI system easily usable. This would imply that a talker/listener (master/slave) relationship would have to be implemented between the three, currently unrelated, systems. This issue has been raised by the author in discussions with D-soft solutions and the TEDDI Data analysis requirements specifications (Appendix D) has taken this into account as a long term objective. A collaboration between D-soft solutions, Aspect and the person developing the rTEDDI control system will most likely have to be set up.

A number of different samples might be of interest for initial testing of the second generation rTEDDI system. One area that is believed to of interest for the rTEDDI imaging technique is medicine. As described in section 4.3, 'identification of fine structural changes in tissue is believed to fall outside the capability of the rTEDDI system. In vivo identification of diseased tissue, when leading to altered scattering signatures as described for certain types of cancer [118] [119], is anticipated as a main use of the technique. For tissue biopsy samples more advanced studies, such as the once described by Geraki et al. [120] [119] could be made, allowing for adipose and fibrous sample composition to be determined as well as quantification of elemental concentrations'. A literature review of scattering signatures and changes of these in diseased tissues could prove advantageous. For identification of elicit material in security applications using the rTEDDI system a somewhat different analysis chain then the one outlined by the TEDDI Data analysis requirements specifications (Appendix D) would likely have to be considered. One possible analysis method would be the use of libraries containing spectral look up tables of the EDD scattering profiles of illicit materials. A literature review,
or systematic collection, of scattering signatures containing to material of interest for the security industry would likely be required.

The second generation rTEDDI system has a number of very exciting applications. Using the system for rTEDDI imaging and 3D fluorescence tomography will allow for fast analysis of structure, strain and chemical contamination within whole engineered components such as fail safe components used for aerospace applications. The addition of a rTEDDI module to existing security scanners would allow for a faster and more reliable identification of elicit materials. The sample chemical information obtainable from the rTEDDI technique in combination with the already existing absorptive information in a security scanner will also allow for a much reduced false alarm rate.

The use of rTEDDI imaging in tissue biopsy would allow for an improved identification of cancerous tissue. This in turn will allow for optimized surgical procedures and steer post operative radiological treatment. A drop in the number of false positives will save patients from non needed, painful and expensive treatment.

In the field of oil and gas exploration a real time analysis of core samples will allow for rapid identification of mineral and rock species, saving rig time and deliver data for geo-steering and formation evaluation.
Appendix A

The ERD2004 detector data analysis program
No data analysis software previously existed for the data obtained using the ERD2004 detector. A basic analysis program was as a result developed within the advanced characterization techniques group at the University of Manchester. The main core of the program, called teddi\_v1 was developed by Dr. Kern Hauw Khor, scripted using Matlab .m files [165], but has since then been adapted and extended. Figure A.1 shows a section of data obtained using the ERD2004 detector when exposed to Ag fluorescence X-ray radiation. The ERD2004 detector uses a sparsitic logic in collection of data. Each row contains the data obtained for a single photon event. Subsequent rows therefore correspond to events occurring in chronological order. The first column contains the voltage value, corresponding to an X-ray photon energy, of the registered event. The second column states the channel number of the pixel for which the event took place. Special care has to be taken when dealing with the pixel number value. Due to operations done to the data by the Labview detector GUI the stored pixel number, referred to as the data file channel number, differs from the physical address number of the pixels, referred to as the real channel number. The correspondence between the data file channel number and the real channel number can be seen in figure A.2. Data column three for the data obtained using the ERD2004 detector contains a voltage value used by the hardware as a reference to identify the voltage value of column 1. Column four states during

<table>
<thead>
<tr>
<th>Induced Voltage</th>
<th>Channel number of registered event</th>
<th>Reference voltage</th>
<th>Frame number for event</th>
<th>Data Valid</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.732</td>
<td>19</td>
<td>0.496</td>
<td>37717</td>
<td>1</td>
</tr>
<tr>
<td>0.291</td>
<td>118</td>
<td>0.496</td>
<td>37717</td>
<td>1</td>
</tr>
<tr>
<td>0.273</td>
<td>255</td>
<td>0.503</td>
<td>37717</td>
<td>1</td>
</tr>
<tr>
<td>0.305</td>
<td>238</td>
<td>0.505</td>
<td>37717</td>
<td>1</td>
</tr>
<tr>
<td>0.322</td>
<td>211</td>
<td>0.503</td>
<td>37717</td>
<td>1</td>
</tr>
<tr>
<td>0.337</td>
<td>204</td>
<td>0.503</td>
<td>37718</td>
<td>1</td>
</tr>
<tr>
<td>0.215</td>
<td>83</td>
<td>0.496</td>
<td>37718</td>
<td>1</td>
</tr>
<tr>
<td>0.278</td>
<td>91</td>
<td>0.496</td>
<td>37718</td>
<td>1</td>
</tr>
<tr>
<td>0.251</td>
<td>129</td>
<td>0.503</td>
<td>37718</td>
<td>1</td>
</tr>
<tr>
<td>0.31</td>
<td>195</td>
<td>0.503</td>
<td>37718</td>
<td>1</td>
</tr>
<tr>
<td>0.33</td>
<td>0</td>
<td>0.496</td>
<td>37718</td>
<td>1</td>
</tr>
</tbody>
</table>

**Figure A.1**: Data structure of the data obtained from the ERD2004 detector system. Each row contains the data registered for a single event. Column 1 contains the registered voltage value of the detected event. Column 2 states in which pixel the event took place. The third column states the value of a reference voltage used to determine the voltage in Column 1. Column 4 states during which frame number (which global reset period) the event took place. The final column contains a data valid/invalid check number. Due to the sparsitic logic (comparator triggered activation of read out) of the ERD2004 detector consecutive rows, and therefore events, are in chronological order.
Figure A.2: Conversion chart for channel no between stored data file channel number and real channel number. The data is stored using file channel number while the data is displayed in the GUI as real channel number.

which frame the event was registered and column five is a data valid check (1=valid, 0=invalid). The root menu options available when running the teddi_v1 program can be seen in figure A.3. Option 1 loads in the specified data file, in the format seen in Table

```
** TEDDI Analysis Tools **

   (Overall spectra plot and pixel map)

   --- ENERGY CALIBRATION ---

[2] Create k-alpha peak file. (1)

   --- PIXEL PLOT ---

[6] Load fitting constants file. (1)
[7] Full SHAMROC plot. (1, 6)
[8] Multi/Single pixel plot. (1, 6)

   --- PEDESTAL ---


[0] QUIT
```

Figure A.3: The root menu options available when running the teddi_v1 program. Typing teddi_v1 in the Matlab main command window will produce this text and allow you to choose one of the options available in the program.

2, from the computer memory into matlab work memory. For the purpose of describing the developed data analysis software we will be assuming we are working on a data file called Data1.txt. Option 1 will ask if you which to remove the pedestal noise, which will
be described later when discussing option 9. It will then remove all data lines in the data files where the data valid number is not 1 (removes corrupted data). All data channel numbers are changed from the software data channel number to their physical address in order to create consistency in the labeling and identification of pixels. Option 1 then outputs a histogram spectral plot of the detector response and a pixel relative intensity map as can be seen in figure A.4. The spectral response seen in figure A.4 (Left) is an addition of the spectral response of all the individual pixels when exposed to a Mo X-ray source. This is referred to as an All Plot. As can be seen the spectral response is plotted as voltage vs. intensity. At this stage in the program no energy calibration has been done for the detector and as a result the spectral response is plotted as voltage vs. intensity as opposed to energy vs. intensity as is the end goal.

Option 2 uses the data loaded in Option 1 (Data1.txt) and finds the highest intensity peak in the spectral response for each pixel which is assumed to be the peak corresponding to the incident photon energy. The output from this Option is a file with the name extension Data1_kalphamax.txt. This file will consist of a single column with 256 lines. Each line corresponds to a single pixel and contains the highest intensity peak value for that pixel.

The input to Option 3 is 4_kalphamax.txt files created by irradiating the detector with different energy X-ray sources and running the resulting data files through Option 2. Which these _kalphamax files are is defined by the user. The aim of this option is to find the linear conversion between the induced voltage of the detector and the incoming photon energy throughout a specific photon energy range. The output from this option is the creation of 4 files. These files will have the name extensions: The first file consists of 4 columns each having 256 rows. Each row corresponds to a specific pixel and each of the columns in a specific row contains the peak voltage value from the spectral response.
Table A.1: Extension name of the 4 files created as an output to Option 3.

<table>
<thead>
<tr>
<th></th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>_kalphamax_4S_kalphamax.txt</td>
</tr>
<tr>
<td>2.</td>
<td>_kalphamax_4S_LFconstant.txt</td>
</tr>
<tr>
<td>3.</td>
<td>_kalphamax_4S_mean_kalphamax.txt</td>
</tr>
<tr>
<td>4.</td>
<td>_kalphamax_4S_mean_LFconstant.txt</td>
</tr>
</tbody>
</table>

Table A.1: Extension name of the 4 files created as an output to Option 3.

for the radiation used to produce the 4 input _kalphamax files (i.e. column 1 = Rb source = 13.37keV, column 2 = Mo source = 17.44keV, column 3 = Ag source = 22.10keV, column 4 = Ba source = 32.06keV). The second file consists of 2 columns having 256 rows. Each row corresponds to a single pixel and the columns of each row contain the gradient and intersection point of the linearity between photon energy and induced detector voltage that can be derived from the values in the _kalphamax_4S_kalphamax.txt file. In short this file contains the conversion ratio between energy and voltage for each pixel that is the energy calibration for the detector. The third file has 4 columns and 1 row. Each column contains the mean value of the 256 lines of their counterpart in the _kalphamax_4S_kalphamax.txt file, thereby stating the mean linearity and intersection point of the detector. In the same way the values in the fourth file contains the mean gradient and intersection point of the detector calculated using the values found in the _kalphamax_4S_LFconstant.txt file. When running Option 3 graphs showing the energy to voltage linearity of each pixel is plotted. An example of these plots can be seen in figure A.5.

Option 4 does the exact same thing as Option 3 but uses 5 sources to find the energy to voltage linearity instead of 4. The first 4 sources for Option 4 is the same as those stated for Option 3. The fifth source is a Tb source that emits radiation with energy of 44.23 keV.

Option 5 enables the user to pick out a single pixel and plot the energy to voltage linearity for that specific pixel.

Option 6 allow you to decide if you want to load the gradient and intersection point values for the different pixels that has been stored in the _kalphamax_4S_LFconstant.txt into the matlab work memory or not. If the values are not loaded up into the memory the spectral response plots created by running Option 7 are plotted as voltage vs. intensity, if they are loaded into the memory the plots will be plotted as energy vs. intensity. Option 7 plots the spectral response of the detector for each pixel. An example of this kind of plot can be seen in figure A.6. The spectral response of the pixels can be plotted either as voltage vs. intensity or energy vs. intensity.

Option 7 allows the user to single out one pixel or one row of pixels from one SHC04 and plot the spectral response accordingly.

In an ideal case no charge at all would be detected by the detector under dark conditions. This is however not the case. Every single pixel shows a small voltage, which fluctuates
slightly with time, under dark conditions. This voltage offset is known as the pedestal values. In order to correct for this source of error the voltage value for each pixel is collected when no source of radiation is presence during a few read out cycles. Option 9 calculates the mean offset for each pixel using the measurement data and outputs a mean pedestal file that can be used in Option 1 to remove this source of noise. The mean pedestal file is given the name extension `avgped.txt` by the program.

An alternative analysis program has been developed by Matthew Wilson, Rutherford Appleton Laboratory. A copy of both the `teddi_v1` script and the alternative analysis software can be found in Appendix A.
Figure A.6: Example of pixel spectral response plot as created by Option 3 in the developed data analysis software. A Rb source was used to illuminate the detector.
Appendix B

The HEXITEC detector data analysis programs
No data analysis software existed for the data obtained using the HEXITEC detector. A number of programs was consequently developed in order to enable basic data analysis with HEXITEC detector data. The programs where scripted using Matlab .m files [165]. There are two basic data format available form the HEXITEC detector known as 1-frame mode and 3-frame mode. The 1-frame mode is used when collection actual data using an X-ray source while the 3-frame mode is used to detect events when using test signals to analyze the performance of the ASIC. Both formats are visible in figure 6.53. For both formats the data is saved as greyscale images. For the 1-frame mode the image is a 20x60 pixel greyscale image where each pixel greyscale value represents a voltage read out from the detector. The pixels in area 20x(1-20) are the S1 values of the 20x20 pixel channels from one data frame. The pixels in area 20x(21-40) are the SPH value(peak hold value containing the actual photon energy measurement) of the 20x20 pixel channels from one data frame. The pixels in area 20x(41-60) are the S2 values of the 20x20 pixel channels from one data frame. The 3-frame mode contains three sets of S1, SPH and S2. For the central set of S1, SPH and S2, pixels (21-40)x60, the values contains the same information as for the 1-frame mode. The first and third sets, pixels (1-20)x60 and (41-60)x60 respectively, are resets of the S1, SPH and S2 circuits before and after the injection of a test signal voltage directly onto the pixel channel. A complete run of the detector, regardless of run mode, contains a large number of read out frames and the resulting data is saved in a folder containing a large number of greyscale images. 7 programs where initially scripted to allow basic data analysis of the 3-frame mode but was programmed for easy conversion to be able to use the 1-frame mode of data collection. The developed programs and the data analysis flow is shown in figure B.2. The Create_datafile.m program is used in order to create a single data file from all the
The HEXITEC detector data analysis programs

Figure B.2: Illustration of the data analysis program flow for basic HEXITEC data analysis. The data collected (number of greyscale images) is saved as a single data file by program 1. Creation and removal of pedestal values, program 2-3, are optional before data visualization and analysis. Four visualization and analysis programs, program 4-7, allow the user to produce single pixel plots, All-plots and total intensity pixel maps.

greyscale images containing to one detector run. The user have to specify in what folder the greyscale images are saved and the number of greyscale images in the folder as input parameters to the program. The program selects the SPH data from the greyscale image, pixels 20x(21-40) in 1-frame mode and pixels (21-40)x(21-40) in the 3-frame mode. Subsequent SPH value greyscale images are then added together creating a 20x20x'no of frames’ matrix where the x-y position gives the value for a specific detector pixel in the frame you are in and the z value states which frame the data comes from. The 3D array containing the SPH data from all the greyscale images obtained during one detector run is saved to the user specified filepath as the output of the program. The datafile produced by program 1 can then be used to visualize and analyze the data, using program 4-7, with our without subtracting the pedestal data, program 2-3.

To enable pedestal subtraction (remove variation in base value voltage level for the pixel channels) a set of data, ideally containing at few thousand frames, has to be collected while not illuminating the detector with X-rays (1-frame mode) or while not injecting any voltage pulse onto the channels (3-frame mode). The inputs to the Create pedestal file.m specified by the user are the filepath to the greyscale images as well as the number of greyscale images contained in the folder. A 3D matrix is created just as described for the Create_datafile.m. For each pixel (x-y value) the the average of all the frames is calculated (adding together all the z values and dividing the value with the number of frames) and the program outputs an 20x20 array of pedestal values, each value corresponding to a specific pixel channel, the the user defined filepath. The Remove_pedestals.m takes the raw datafile created by the Create_datafile.m program and the pedestal file created by the Create_pedestal_file.m program and subtracts the pedestal values from the raw
data. The output of the program is a new data file on the same format as the raw data, but with the pedestals removed, which is saved to the user defined filepath. Visualization and analysis of the data, program 4-7, can be done in any order the user wishes. The input to program 4-7 is a dataset on the format as created as an output by Create_datafile.m or Remove_pedestals.m program. The All_plot.m program histograms, using the Matlab hist() function, the entire specified datafile without taking into account which pixel any event belongs to. The binning size used for the histogramming is specified by the user. The output of the program is a spectral response plot for the detector, where the intensity is plotted as a function of greyscale (LSB) value, as can be seen in figure B.3. The All_plot.m could only handle data containing less then approximately 40,000 greyscale images. This was found to be due to the inability of the Matlab hist() function to handle large arrays leading to the function crashing. To overcome this problem the Conny_All_plot.m program was scripted. The Conny_All_plot.m program has the same inputs and outputs as the All_plot.m program but has a does not utilize the Matlab hist() function to do the histogramming. The histogramming is instead done by a specifically developed code, developed by the author, that uses a 3D mask for each binning region to find the intensity values, reducing the amount of virtual memory needed for the operation by Matlab. The Conny_All_plot.m allowed for All plot histogramming of data exceeding 150,000 frames. The Single_pixel_plot.m program plots the spectral response of a single pixel, specified by the user, as intensity versus greyscale (LSB) value. A single pixel plot can be seen in figure B.4. The Matlab hist() function is used in this program due to the 400 times decreased data amount posing no problem with respect to virtual memory allocation. The Pixel_intensity_map.m program

\[ \text{Figure B.3: Resulting All-plot as produced using the All_plot.m program. The data was collected using a 0.2V test signal.} \]
The HEXITEC detector data analysis programs

Figure B.4: Resulting single pixel plot as produced using the Single_pixel_plot.m program. The data was collected using a 0.2V test signal.

produced a pixel map containing the number of events registered for each pixel within a user defined region of interest. A resulting total intensity map can be seen in figure B.5. A copy of the 7 program scripts can be found in Appendix A.

Figure B.5: Resulting total intensity map as produced using the Pixel_intensity_map.m program.
Please see accompanied CD for programming scripts.
Appendix D

TEDDI Data Analysis
Requirements Specification
TEDDI Data Analysis
Requirements
Specification
A TEDDI detector system has been under development for a number of years by a consortium of STFC, UK Universities and commercial companies. The requirement under specification here is to produce a data visualization and analysis system for data collected from the TEDDI detector. The detector is currently a 20x20 pixelated Cadmium Zinc Telluride detector and readout ASIC. An 80x80 version is under development but in the foreseeable future this is still likely to be readout in the 20x20 mode.

To construct an image a sample is scanned by an X-Ray beam, using a combination of repeated positioning the sample in the beam, the alignment of very fine collimator assemblies and detector readout. The intersection of the beam and the alignment of collimator assemblies allow the detector pixels to 'see' separate volumes within the sample. The detector pixels measure photons scattered from small individual volumes of the sample. There are no requirements to control the sample/collimator motions. As the sample is scanned a 2D/3D array of data is collected from the sample.

The detectors count electron-hole pairs, created by incident photons in the detector elements. The number of electron hole pairs is related to the energy of the incident photon. The detector is interfaced via a number of specifically designed integrated circuits, called the stack, developed by Aspect. The number of electron hole pairs is read out as a voltage. Once started the interfacing stack constantly reads the detector elements, clearing and waiting for new photons as it progresses. Reading data from the stack causes its memory to be cleared. A software system is available, developed by Aspect, to interface to and control data acquisition from the detector. The process of readout by the Aspect software converts each voltage into a greyscale, proportional to the readout voltage. The width of these greyscales is set such that no data resolution is lost. The Aspect software does not have to read out the whole area of the detector, a smaller area can be specified.

As the detector readout progresses the detector provides a constant stream of data. Each data point consists of 3 greyscales per pixel, S1 (shaper 1), SPH (peak hold) and S2 (shaper 2). S1 and S2 provide checks on the validity of the real signal SPH. The Aspect software allows the correction and saving of data in different manners and formats.

A TEDDI data analysis system, TDA, is required to allow the following, either running as data is collected or offline with previously collected data. TDA will not do any data corrections; these are only done by the Aspect software.

- Binning and histogramming of data.
- Energy calibration of histogrammed data.
- Saving histogrammed data.
- Visualization of data. Image data as 2D/3D plots. Histogrammed data as 1D spectrum.
- Refinement of crystal structure to fit theoretical and experimental data using TOPAS.
- Save refined crystal parameters.
- Visualize refined crystal parameters.

If the Aspect software is reading out a sub-section of the detector TDA needs to perform its operations on just that area. Determining whether a sub-section is being used cannot be determined from the data files. This will either need to be passed from the Aspect software to TDA or entered by the user manually.

TDA will have a GUI and operate either at the same time as data acquisition or standalone. When operating with data acquisition there will be a requirement to operate in synchronization with that data acquisition. This may require direct communication with the Aspect software.

The data analysis system has to be capable of processing data at the same rate as it is collected by the data acquisition system. Data analysis should interfere with data acquisition as little as possible, ideally not
slowing the maximum speed of acquisition. A computer system architecture is to be specified as part of the grant application. This should take into account future developments such as the following:

- The inclusion of control of the sample and TEDDI moveable stages within the overall system, to achieve 3D sample viewing.
- The inclusion of some element of intelligence into the system, for example taking decisions based on the output of data analysis.

Many commercial applications are foreseen on top of the research at SR facilities e.g. drug detection inside containers, cancer scanning. Ideally a standalone data acquisition system needs to be developed.

**Requirements**

1. **Data histogramming and calibration**

Detector data is read out by the Aspect software continuously once started. When a photon hits a detector pixel this is known as an event at that pixel. The detector is read fast enough so that each time the detector is read a pixel should have either one or no event in it. When readout each event will result in a greyscale reading proportional to the energy of the incident photon. Data saved by the Aspect software can currently have two different formats, grey level png files or ASCII dat files.

In the first case there will be a separate png file for each detector image in the sequence. The greyscale level at each pixel in an image corresponds to the readout at each pixel. The data in this case is completely uncorrected. Three images are saved in each png file, S1, S2 and SPH data.

In the second case data a number of detector images are stored in individual dat files. The entire data collection will result in a number of such files dependent upon its duration and the number of raw images in each file is specified by the following.

\[ \text{Number of raw images} = \text{Number of images per buffer} \times \text{Number of buffers per file} \]

Three modes of operation are possible for the Aspect software here, *Event Detection Hardware*, *Event Detection Software* and *Event Detection Reduced*. Only the SPH data is saved. The following corrections are or will be done by the Aspect software in this case:

- Pedestal, or dark noise, removal.
- Charge sharing correction.
- End of frame correction.

The dat file data is saved as ASCII in sequence through the dat file, one sequence of 400 pixel data points per image assuming a 20 by 20 detector. The current data format is defined in an Aspect document, *Hexitec Data Format for saved raw data*. The data formats are likely to change. The TDA system should be created so that it is easy to add other input data formats.

The basic flow required for initial binning and calibration of data saved by the Aspect software is represented in Figure 1.
The 2D images contain the greyscales of events that have occurred at each pixel on the detector. These greyscales first have to be arranged into an array of greyscales at each pixel. These are then counted into the number of events in greyscale ranges at each pixel. The output of this stage is a number of events in each greyscale bin at each pixel i.e. histogrammed data. The size of the greyscale bins must be selectable by the user.

In order to convert these histogrammed greyscale data into energy data a calibration histogram needs to have been collected. There are two ways this can be done:

- Collection of calibration spectra via the use of a standard sample, producing known energy diffraction peaks, in a Synchrotron beam. One calibration file is produced containing the greyscales of events for a number of peaks with energies dependent upon the standard sample.
- The use of a dial source when collecting from an X-Ray generator. This source produces X-Rays of known energy dependent upon the dialled up energy. A number of individual calibration files are produced each containing the greyscales of events for a single energy.

Each pixel will have an individual calibration spectrum. The user will have to input the names of the calibration files. Separate calibration is required for different detector settings. Usually during an experiment there will be one set of calibration data. These calibration spectra allow a conversion between greyscale and energy. If the files do not internally define the energy of peaks they contain then the user will have to enter this. TDA will determine the positions of peaks in the input calibration files. TDA will have the options of applying either a linear or polynomial calibration. When calibration is performed data giving the number of events versus both energy and greyscale will be saved. If not the data will be saved as the number of events versus greyscale value.

2. Visualisation of collected data

The Aspect software has no facility for the visualisation of data collected from the detector. There are no plans for this to be added to the Aspect software. A 2D false colour representation and a 3D surface representation of the events/pixel are required. Associated with each pixel will be a 1D energy dispersive spectrum. It must be possible to display these easily selectable from the 2D image.
2D Image Display

Each pixel in the 2D array of data should see a number of events as data collection proceeds. The TDA GUI will support the visualization of 2D detector data as a false colour image. For this purpose the events at each pixel need to be converted into a single number and there are multiple possibilities for achieving this.

- Sum all events to produce a total number of events.
- Region of Interest (ROI) definition and combination. It will be possible to define and combine events from a number of ROIs. These ROIs will be defined as the lower and upper bounds of greyscale or energy regions, depending on whether calibration has been performed. Having defined the ROIs events in each will be summed. It will then be possible to combine these in a number of ways to produce a single number.
  - Sum all events within one or more ROIs.
  - Produce the ratio between the summed ROIs.
  - Simple mathematical combinations of summed ROIs.
- Full Width Half Maximum (FWHM). First defining a window where a peak is expected to appear it will then be possible to curve fit any peak that occurs in that window and to calculate its FWHM.

3D Image display

A surface representation of the detector events will be possible. This will be possible either as a 3D bar chart or a surface plot. The viewing position will be selectable.

Display of Spectra

The TDA GUI will support the display of event spectra at individual detector pixels, in a separate area of the visualization GUI. As the mouse is moved over the 2D display two modes of operation will be available to refresh the spectrum display area to show the event spectrum for a pixel.

- Clicking on a pixel to refresh the spectrum.
- Positioning over a pixel to refresh the spectrum.

In 3D image display mode this will not operate, but it will be possible to select a spectrum by entering image co-ordinates if required.

There will be an option to display all histograms for the detector as one histogram, spectral all-plot. This will add corresponding histogram elements for all pixels to produce a single histogram, and display it in the spectrum display area.

3. Data Analysis

The TOPAS suite of software, originally developed by Bruker, now maintained and developed by Alan Coelho will be used to perform Rietveld analysis and refinement of the powder diffraction spectra. TOPAS runs on Windows XP or later. TOPAS requires data to define a theoretical crystal model for the sample. This will be obtained prior to analysis by the user from the crystal database. It will be stored in a file and input to TOPAS via TDA. TDA will need to allow the user to specify graphically from the crystal parameters which are to be refined. TOPAS performs a refinement by adjusting crystal structure parameters, repeatedly calculating the theoretical diffraction and matching this to the experimental diffraction.
The TDA system needs to allow the input of data required to initiate data analysis and the creation of scripts to run TOPAS. The data analysis will then be farmed out as a batch job. Once complete the program’s output will be parsed, saved to a crystal structure file and the latter used to inform follow on visualization of the results by TDA.

4. Visualization of refinement results

Once the TOPAS refinement is complete it should be possible to visualize the results in a similar manner to that described in section 2. However it will also be possible to display the output of the TOPAS parameter refinements used to define the false colour at each pixel. Some simple mathematical combination of these parameters will be required to define the pixel value.

5. Computer System

The Aspect system runs on a Windows based PC. The system in Manchester is based on an Intel Core 2 Quad CPU with a Gigabit Ethernet connection (Marvell Yukon 88E8056 PCI-E). The detector is interfaced via a dedicated card. The data is stored to a local disk. Data processing of data stored on this local disk would have an impact on the maximum data acquisition rates. A system is required where any impact of data analysis on data acquisition rates is minimised. Any significant impact would preclude the possibility of simultaneous analysis and acquisition.

One possible system is a collection of discrete CPUs communicating via a dedicated Gigabit network. A central switch will isolate the system from other local and wide area network traffic, and vice versa. As well as the data acquisition CPU a dedicated file server and data analysis CPU would be attached. The file server’s performance needs to match that of the current internal disk and be capable of accepting requests from both the acquisition and analysis CPUs without degrading the performance of either.

In order to ensure data analysis can keep pace with data acquisition it may be necessary to have multiple CPUs collaborate on the task, a cluster. To take advantage of such a cluster would require either parallelisation of TOPAS or some scheme whereby a bigger job could be split into multiple concurrent runs of TOPAS and the outputs combined. The use of a cluster may well have implications for the overall hardware design.
Appendix E

Published Papers
X-ray colour imaging

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A prototype X-ray colour imaging system has been assembled using the principle of tomographic energy-dispersive diffraction imaging (TEDDI). The new system has been tested using samples of nylon-6, aluminium powder and deer antler bone. Non-destructive three-dimensional images of the test objects have been reconstructed on a 300 μm scale with an associated diffraction pattern at each voxel. In addition, the lattice parameters of the polycrystalline material present in the sampled voxels have been determined using full pattern refinement methods. The use of multiple diffracted parallel X-ray beams has allowed simultaneous spatially resolved data collection across a plane of the sample. This has simplified the sample scan motion and has improved data collection times by a factor scaling with the number of detector pixels. The TEDDI method is currently limited to thin samples (approx. 1–2 mm) with light atoms owing to the very low detection efficiency of the silicon detector at X-ray energies above 25 keV. We describe how these difficulties can be removed by using semiconductor detectors made from heavier atomic material.

Keywords: imaging; synchrotron; energy dispersive diffraction

1. INTRODUCTION

X-rays currently provide three-dimensional density contrast images for a wide range of applications most commonly encountered in the fields of medicine and security in the form of spiral CT or baggage scanners. However, the information carried by different wavelength (or colour) X-rays is rarely used in these modalities. This information is potentially very useful because it can be used to identify tissue types (Harding et al. 1990), strain distributions in whole engineered components or the presence of illegal substances in luggage (McGann et al. 1993). A full X-ray colour imaging modality needs spectroscopy-grade pixellated solid-state detectors and corresponding high aspect ratio collimators which have hitherto not been available. We show for the first time how the necessary energy-sensitive detector and collimator arrays can produce genuine three-dimensional X-ray ‘colour’ images with associated energy-dispersive diffraction (EDD) patterns at each voxel for a range of samples across scientific disciplines.

The underlying principle of tomographic energy-dispersive diffraction imaging (TEDDI) has been described before using a single Ge solid-state detector scanning the sample in three dimensions on a 20 μm scale (Hall et al. 1998; Barnes et al. 2000). This is very time consuming, often requiring 20 hours or more to complete. The developments we describe here have enabled us to obtain much faster X-ray colour images with one simple scan motion while delivering a lower dose of X-rays to the subject under investigation.

The imaging geometry of the TEDDI X-ray colour imaging system is illustrated in figure 1a. A polychromatic X-ray beam from a synchrotron or X-ray tube is incident upon the study object. The X-rays are scattered into a set of highly parallel collimator apertures each with an aspect ratio of 4000 : 1 (Tunna et al. 2006) or higher. The X-ray beams that pass through the collimator apertures are recorded by an energy-sensitive pixellated detector (Seller et al. 1998). The tomographic element of the scattering process is enabled because a specific gauge volume (shown shaded in figure 1) is sampled by the intersection of the incident and diffracted beams. At the low 2θ angles necessary for observing diffraction patterns at high energy, the spatial resolution along the beam direction may be 10 times lower than the collimator aperture. An alternative scattering geometry is shown in figure 1b, but this is possible only with thin samples and a box beam illuminating the sample. The gauge volume is dictated by the sample thickness and the collimator geometry gives vertical rather than depth spatial separation. Despite this spatial limitation the method provides very simple and informative images.

2. RESULTS

We present here the first results in X-ray colour imaging using our prototype collimator and detector array system. The detector consists of a 16×16 array of energy-sensitive pixel elements (300×300 μm) and a corresponding 16×16 array of 50 μm diameter collimators. Each pixel records the EDD pattern from the intersection volume in the sample. This can be used to identify the material present at that point in the sample. The total scattering count gives an image of the whole sample which is proportional to the density contrast. In addition, the scattered spectra contain X-ray fluorescence (XRF) information that can also be used for material characterization. Figure 2a shows the experimental system on beamline 7.6 of the SRS at Daresbury Laboratory. The Si-pixellated detector was carefully aligned to our very fine tolerance laser-drilled X-ray collimators. A thin polymer sheet with a cross test mark (figure 2b) was placed in a 5×5 mm X-ray beam as shown in figure 1b. The total scattering from each pixel is shown in figure 2c, the intensity is false coloured, the lighter the colour the greater the count rate. Figure 2d shows the individual energy-dispersive X-ray scattering pattern from each pixel. There is no scattering where the polymer material has been
removed to form the test cross mark. The remainder of the pixels clearly display an energy-dispersive X-ray diffraction pattern. This pattern can be used in a number of ways; for example, to identify the material at the gauge volume position in the sample against a set of stored spectra, refine lattice parameters to determine strain distributions or identify materials based on characteristic fluorescence. This information is present vertically small X-ray beam large box section X-ray beam scattering gauge volume or intersection lozenge collimator apertures (**a**) (**b**) Figure 1. (**a**) A vertically thin polychromatic beam incident on a thick sample. Two of the collimator apertures are shown. The scattering is measured only where the incident beam intersects with the projected collimator aperture (shaded), thus giving spatial resolution to the sample. (**b**) A similar effect but with a large box beam and a very thin sample. Note the direction of special resolution is now vertical.

**Figure 2.** (**a**) Thin polymer sample mounted in the X-ray beam with the collimator and detector array inclined at an angle of 22° to the horizontal. (**b**) Polymer sample with a test cross cut out, the beam discolouration can be seen. (**c**) The TEDDI image from the polymer sample (lighter colour indicates higher photon count). (**d**) The energy-dispersive diffraction patterns recorded at each pixel. Note the absence of scattering where the material has been removed (shaded).
Figure 3. (a) Two typical diffraction patterns from different pixels obtained from a thin polymer sample later identified as nylon-6. (b) A lattice parameter refinement of the nylon-6 sample where the observed data points ($I_{obs}$) are fitted with the smooth red line ($I_{calc}$). The $I_{obs}$-$I_{calc}$ curve is shown in grey together with tick marks at the expected Bragg peak positions (blue). The refinement converged to a $\chi^2$ of 1.45 with lattice parameter errors in the second decimal place.

Figure 4. (a) A TEDDI image of a slice through a thin aluminium pressed powder sample. (b) The energy-dispersive spectra from a typical pixel have been refined. Although the observed data (blue) are of poor statistical quality, a good fit (red) was obtained with $\chi^2=0.94$ and $\alpha=4.0497(38)$ Å.

Figure 5. (a) A 1 mm thick section of deer antler bone together with the 5 mm$^2$ area investigated using rapid TEDDI. (b) The corresponding rapid TEDDI image. (c) A diffraction pattern from one of the pixels together with output of a refinement of the known hydroxyapatite lattice parameters.
in each pixel, it is the choice of the experimenter to decide which to analyse.

Figure 3a shows the EDD patterns from two pixels in the detector array. The patterns are similar but not identical. Local variations in structure and preferred orientation are largely responsible for these differences. A lattice parameter refinement was carried out using TOPAS (Kern & Coelho 1998) and despite the limited statistical quality in each pixel due to the low count rate a \( \chi^2 \) of 1.45 was achieved with the possible space groups \( P 2 1, P 2 1/m \) or \( P 2 1/c \), \( a = 7.058(50), b = 10.679(36), c = 17.83(15) \) Å and \( \beta = 68.4(21)^{\circ} \) corresponding to the structure of nylon-6. This was later confirmed by collecting an FTIR spectrum and referring the result to that of nylon-6 stored in the Hummel Polymer Library (A. Wilkinson et al. 2007, private communication). The accuracy of the lattice parameters from the TOPAS refinement is lower than that expected from traditional powder measurements, occurring in this case to the second decimal place; however, detector efficiency improvements will alleviate this problem. The intensity variations due to local structural differences can be treated qualitatively to give information on grain clustering (Cernik et al. 2007). However, the integrated intensities are not sufficiently reliable to enable structure determination or full Rietveld refinement.

There is a great deal of interest within engineering communities in the non-destructive determination of residual stresses in manufactured components especially in critical areas such as aircraft wings and engine casings. The TEDDI system can be used for strain scanning whole fabricated components in the automotive or aerospace industries, however, even though we are currently limited to light alloys. The TEDDI system generates new imaging possibilities in the physical sciences including the analysis of structural changes in collagen associated with cancerous tissue. These features were measured with small angle scattering over a size range of 75–1390 Å. Structural features of these dimensions cannot be identified by biological sample in 180 s.

We anticipate that one of the main uses of the TEDDI method in the life sciences will be in distinguishing normal from abnormal tissue types. Geraki et al. (2002, 2004) have developed a synchrotron-based method combining XRF and EDD to examine biopsies of healthy and cancerous breast tissue. The XRF was used to examine the differential accumulation of iron, copper, zinc and potassium between the abnormal and diseased tissue while the EDD data were used to evaluate the quantities of adipose and fibrous tissue present. The shape and distribution of the EDD spectra from the healthy cells are quite different from those of the tumour cells and could be easily separated out by automatic sampling methods. Their XRF analysis revealed that all four elements are found in elevated levels in the tumour specimens. The rapid TEDDI method described in this paper gives simultaneous fluorescence and diffraction measurements which will greatly facilitate this type of biopsy sampling. Lewis et al. (2000) have described the structural changes in collagen associated with cancerous tissue. These features were measured with small angle scattering over a size range of 75–1390 Å. Structural features of these dimensions cannot be measured with TEDDI since the diffraction angles are practically too small. However, it is quite possible with low detector angles and energies in the region of 20 keV to probe structures on a 30 Å scale. The usefulness of this was described by Castro et al. (2005) with regard to the analysis of uterine, kidney and breast duct carcinoma biopsies. The observed structural changes in the first case were too subtle for the TEDDI method to distinguish without improvements to the energy resolution. However, the structural changes measured for the last two cases are of the correct order or magnitude to be observed by the rapid TEDDI system. The TEDDI system generates new imaging possibilities in the physical sciences including the analysis of strain distributions in fabricated components for the

3. CONCLUSIONS

The prototype TEDDI system has significant potential as a multidisciplinary imaging tool. However, our samples were limited in physical size and counting statistics by the low quantum counting efficiency of silicon detectors at high X-ray energies. In our case this was less than 1% above 25 keV. This single factor was responsible for the undesirably long data collection times. These were significantly shorter than the 20 hours point by point scans previously described (Barnes et al. 2000) but were typically 2–3 hours. This would allow the study of a thin alloy sample for use in aerospace engineering applications, but would not be suitable for studying a biopsy sample owing to radiation damage problems. We are currently developing new, high purity, high atomic weight semiconductor detector materials \(^1\) that will remove this difficulty. Cadmium zinc telluride area detectors will be used to replace silicon, these have been shown to be 60% efficient at 100 keV for 500 μm thick material. We have designed 2 mm thick active sensors and expect data collection times to drop by two orders of magnitude giving a 300 μm resolution reconstruction of a 2 cm\(^3\) biological volume.

\(^1\)HEXITEC project funded by EPSRC http://www.hexitec.co.uk/.
aerospace industry; the identification of oil- and gas-bearing seams in geological core samples and for security scanning at airports. In these cases the radiation dosages are less critical. However, there is still a need to use more efficient detector material and higher energy X-rays owing to the need to study fabricated or machined components with large volumes. The result has exciting implications for non-destructive strain scanning of assembled light alloy components because an image, structure variation and strain distribution can be obtained simultaneously. This development will significantly enhance the type of strain scanning studies with whole fabricated components such as those described by Korsunsky et al. (2002). Further examples of the point by point scanning TEDDI method were described by Harding & Schreiber (1999) with reference to the security industry and the need to identify specific substances inside large containers. Energy-dispersive spectra from a variety of samples were demonstrated to be quite clearly distinct and identifiable despite rather poor counting statistics. The range of samples unambiguously identified from their diffraction patterns included smetex, cocaine and heroin.

It would be highly desirable to develop laboratory-based TEDDI systems for medical, security and process control applications, but it is probable that the higher energy applications for engineering or earth sciences will need synchrotron radiation. However, Honemaki et al. (1990) made a comparison of the X-ray flux available for sealed tube, rotating anode and synchrotron sources, demonstrating that if a large opening angle can be accepted (as in the case of TEDDI) the fluxes at the samples can be very similar. We therefore conclude that the X-ray flux available from a tungsten sealed source tube with a short source to sample distance and a wide opening angle will be very similar to the limited photon flux of beamline 7.6 at Daresbury Laboratory. With better detector efficiency we could expect the TEDDI images to be produced from a laboratory-based source which is very important if biopsy screening is to become viable.

We have demonstrated for the first time that our TEDDI imaging system can deliver spatially resolved images with one very simple scanning motion. These images fully use the multiwavelength, or colour, information present in the incident X-ray beam. This gives three-dimensional density contrast images and structural information at each voxel point. The rapid TEDDI system forms the basis of a powerful new modality in X-ray imaging.

We would like to thank the EPSRC and CCLRC for research support and access to the research facilities.

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Performance limitations of the pixelated ERD detector with respect to imaging using the rapid Tomographic Energy Dispersive Diffraction Imaging system

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ARTICLE INFO

Available online 12 February 2009

Keywords:
Semiconductor thin film neutron detector
2D imaging array
Beamport monitor

ABSTRACT

A prototype X-ray imaging system, using the principle of Tomographic Energy Dispersive Diffraction Imaging (TEDDI) has been developed at the University of Manchester’s School of Materials. The non-destructive 3D imaging system makes use of a state of the art collimator array and a pixellated Si energy resolving detector. The new rapid TEDDI system is limited to thin, low density materials due to the low stopping power of Si at higher X-ray energies. In this paper the results of substituting Si for CdZnTe as the active detection element and limitations to the key parameters of energy resolution and count rate, for the detectors is presented. The findings have led to the design of a new ASIC which is discussed.

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1. Introduction

Tomographic Energy Dispersive Diffraction Imaging (TEDDI) has been shown to be able to produce three-dimensional (3D) images of bulk objects, with the associated diffraction pattern for each imaged voxel, on a μm scale [1,2]. This makes the technique potentially very useful for medicine, biology and the physical sciences as well as for security applications [1,3].

The imaging speed of the technique has been limited by current detector and collimator technology, often requiring 20 h or more to step scan a two-dimensional (2D) image [2,4]. In order to overcome this drawback the rapid TEDDI (rTEDDI) technique has been assembled in the School of Materials, University of Manchester, and a proof of concept has been obtained at SRS Daresbury Laboratory, station 7.6 [5]. The rTEDDI technique uses a state of the art, femtosecond laser drilled, collimator array [6] where each one of the 16 × 16 collimators is coupled to a pixel on a corresponding 16 × 16, 300 μm pitch, pixellated Si energy resolving detector (ERD) [7]. This setup allows for voxels constituting a 2D plane to be imaged on a 300 μm scale, simultaneously.

Use of the rTEDDI system is currently restricted to thin samples (1–2 mm) due to the low stopping power of Si for photon energies above 25 keV. In order to overcome this problem and further increase the speed of the system, enabling its use at a higher energy/higher flux, the 300-μm-thick Si crystal has been replaced by a 2-mm-thick CdZnTe (CZT) crystal.

The functionality of the ERD with CZT as the active detection material was investigated using flood field irradiation from an Am-241 source. In order to investigate the suitability of the ERD at high incident flux, count rate measurements were conducted—changing the clocking frequency of the detector system while exposed to flood field irradiation in the form of Tb fluorescence of an Am-241 dial source. Due to the detector dimensions charge sharing was foreseen to be a major source of energy resolution degradation. As a consequence charge-sharing compensation scripts were developed in the form of charge-sharing addition and charge-sharing discrimination. These two methods were tested on Am-241 spectra taken by the CZT ERD detector. A comparison between the two charge-sharing correction methods has been made.

2. Structure and resetting scheme of the ERD application specific integrated circuit (ASIC)

The detector currently used in the rTEDDI system, known as ERD, is a pixellated X-ray detector developed by the Rutherford Appleton Laboratory [7]. The active detector element is bump bonded to a 16 × 16 array of pre-amplifiers on a 300 μm pixel pitch-MAC04 in Fig. 1, which is in turn wire bonded to two 128 × 1 arrays of shaping, peak-hold and comparator circuits, -SCH04. Each pixel has a MAC04-SCH04 channel dedicated to its operation. Two reset schemes are implemented in the system. The global reset is used to reset the MAC04 in order to prevent the pre-amplifiers becoming saturated by leakage current while the local reset is used to reset individual channels of the SCH04 after an...
event is read-out. The active part of the global reset period, \( T \), during which events can be registered, is given by Eq. (1), where \( f \) is the clocking frequency of the detector system. Both these variables are user definable parameters and the local reset is only active during \( T \). A channel is triggered for read-out when an event exceeds a user set voltage in the comparator. This flags a register activating a multiplexer which then reads out the event and activates the local reset. The dead time of the pixel read-out channel following a local reset is 25 clocks. The ASIC is read-out using a data acquisition system (DAQ) consisting of a National Instruments card controlled by software developed in the LabVIEW environment:

\[
T = \frac{2000}{f} \times \text{(number reads per frame)} \tag{1}
\]

3. Flood-field irradiation setup

During the flood-field irradiation measurements the ERD was attached to a cold finger inside an Al box and connected to the read-out and power electronics.

The inside of the box was filled with dry air in order to prevent condensation on the detector during cooling. The detector was cooled to approximately \(-5^\circ\text{C}\) in order to reduce the relatively high leakage current inherent to CZT detectors. The cooling was implemented using two pyramid stacked peltier coolers. The Am-241 dial source was placed on top of a Mylar window in the Al box, with the detector crystal situated on the other side of the window, irradiating the ERD. This setup, shown in Fig. 2, has the advantage of being very simple but the disadvantage that any movement of the dial source will change the effective flux hitting the surface of the detector. Consequently all related measurements were done during the same experimental session.

4. The charge-sharing correction programs

Charge sharing in pixellated detectors takes place when the photon-induced charge carriers diffuse to the extent that part of the charge cloud is registered by the pixel where the interaction took place and part of the charge cloud is registered in one or several of the neighbouring pixels. The ERD uses sparse logic where an event is read-out after having been triggered due to an interaction. This structure of read-out ensures that the timing sequence of events is saved in the order in which events occur. A post-processing charge-sharing correction script was then developed by checking if two or more events took place in physically neighbouring pixels. This structure of charge sharing has the drawback that if two real events take place, one after the other and next to each other, they would fall under the programs definition of charge sharing and is erroneously corrected. At low levels of irradiation this may be considered to be a statistically acceptable drawback. Using the charge-sharing correction structure explained above, two scripts were developed in the Matlab environment, charge-sharing addition and charge-sharing discrimination. For the charge-sharing addition script, the events that
were considered to be charge sharing were compared to see which pixel had the highest collected charge. The pixel with the highest charge was awarded the charge for itself and the sharing pixels. The sharing pixels were set to zero. In the charge-sharing discrimination script all pixels involved in a charge-sharing event were set to zero.

5. Results and discussion

5.1. ERD response using CZT as the detector crystal and count rate measurements

In order to determine whether or not the ERD worked with CZT as the active detection material the detector was exposed to an Am-241 radiation source. The spectral result of adding together the spectra of all 256 pixels can be seen in Fig. 3. The spectra clearly show the main peak of the Am-241; the Cd and the Te escape peaks as well as the Np daughters. To be able to use the rTEDDI system under high-flux conditions the detector used has to be able to operate with a very-high clocking frequency. In order to find the frequency response of the ERD detector it was exposed to Tb fluorescence in the flood-field irradiation setup as previously described.

The clocking frequency was varied while keeping the global reset time $T$ constant by compensating using the $f_{reads}$ parameter and measuring the number of counts. Two $T$ values were investigated, $T = 50$ and $150$. Each data point was measured for 2 min. The results of the measurements can be seen in Fig. 4. The ideal detector behaviour for this measurement is also shown in Fig. 4. There is an increase of the number of counts detected when working at the lowest frequencies due to the fact that the system is under-clocked, and not counting all the photons coming in.

Once the system is not under-clocked, all the photons should be counted and there should be no variation in the number of counts with increased clocking frequency. Measurements did however show a drop in number of counts with increased frequency. It was also observed that the number of counts registered were lower when the system was working under $T = 150$ than at $T = 50$. This suggested that the data acquisition system could not handle large amounts of data. The time intervals between activation of the global reset was measured and compared to the active part of the global reset period, $T$ as described by Eq. (1), in order to determine the deadtime of the system. The result can be seen in Fig. 5.

The deadtime of the system due to the DAQ's inability to handle large amounts of data increases with clocking frequency and this explains the drop in number of counts with increased clocking frequency observed. The need for high counting capabilities of the rTEDDI system at high-flux beam stations makes the ERD unsuitable for this application.

5.2. Comparison between charge-sharing addition and charge-sharing discrimination

In order to determine the most effective method for charge-sharing correction both the charge-sharing addition and charge-sharing discrimination analyses were performed on the same set of ERD data taken with the Am-241 radiation source. The result can be seen in Fig. 6.

Using the non-corrected data as a reference, charge-sharing discrimination was found to be a superior correction method to charge-sharing addition when optimising energy resolution. Charge-sharing addition shows a strong tailing of the main Am-241 peak. This tailing has been attributed to the user defined voltage of the comparator which determines the triggering level for read-out. When the part of the charge cloud that has diffused...
into neighbouring pixels is too small to trigger the comparator it
goes unregistered. The event is therefore not registered as charge
shared and considered good. The result is an event registered as
having a smaller value than it had leading to tailing on the low
end side of peaks. This phenomenon is less significant when using
charge sharing discrimination due to the fact that only one
neighbouring pixel has to be registered as sharing for the error to
be corrected.

6. Conclusions

The ERD detector currently used in the rTEDDI system has been
shown to work satisfactorily when CZT is used as the detection
material at low-count illumination. The large dead time of the
ERD DAQ system when operating at high frequencies has
demonstrated that the detector will not be able to cope with the
high count rates needed to move the rTEDDI system to a higher-
flux synchrotron beam station. The sparse logic inherent to the
 ERD design has shown that charge-sharing discrimination is the
optimal method of charge-sharing correction. This however
comes at a loss of efficiency for the rTEDDI system since it works
on the basis of erasing shared events resulting in longer imaging
times. As a result of these findings a new ASIC, known as
the HEXITEC ASIC, has been designed and is currently under
manufacture. The new ASIC using CZT as a detector will be read-
out using a rolling shutter scheme enabling good response to
charge sharing addition correction and is designed to work with a
5 MHz clocking frequency.

Acknowledgements

We would like to thank EPSRC and STFC for research support
and access to research facilities. This work is motivated and
funded by the HEXITEC collaboration (www.hexitec.co.uk).

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X-Ray Performance of Pixilated CdZnTe Detectors

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Abstract—The X-ray performance of CdZnTe detectors with 300µm pixels was investigated. 2mm thick CdZnTe from eV Products Inc. was bump bonded to ERD2004 detector modules. Preliminary experiments with an eV Products detector at room temperature and -400V bias gave a FWHM of 1.93keV at 59.9keV and a peak to valley ratio of 14.9. The performance of individual pixels varied significantly across the devices due to the non-uniformities in the CdZnTe. Experiments at the Diamond Light Source were conducted to investigate the spatial non-uniformity of the detector. A monochromatic X-ray beam was used to measure the energy resolution and peak position to quantify the spatial charge collection efficiency of the detector. A collimated X-ray beam was used to scan the detector. The scanning experiment was used to investigate charge sharing and lateral electric fields within the detector.

I. INTRODUCTION

CADMIUM ZINC TELLURIDE (CdZnTe) is used as a high energy X-ray detector as it has the desirable properties of high stopping power and can operate at room temperature. CdZnTe could be used as a detector for many applications including medical imaging [1], radiation protection [2],[3], astrophysics [4] and material science [5].

In this work the performance of eV Products CdZnTe on ERD2004 was investigated by experiments at the Diamond Light Source (DLS) and with sealed radiation sources. The CdZnTe was 2mm thick and had a 16x16 array of pixels on a 300µm pitch with a guard ring. The ERD2004 detectors have two types of ASIC, a MAC04 and a SHC04 [6]. The MAC04 has a 16x16 array of 256 channels on a 300µm pitch, each with a charge sensitive preamplifier. The SHC04 has 128 channels, each with a shaper, peak hold and comparator circuit. The pixilated CdZnTe was bump bonded to the MAC04. The MAC04 was wire bonded to two SHC04 ASICs. The SHC04 ASICs were wire bonded to a PCB which fits into a 40-way zero insertion force socket on the external readout electronics, as shown in Fig.1. The readout electronics were connected to a PC via National Instruments data acquisition cards. All of the bonding was conducted at Rutherford Appleton Laboratory.

Fig.1. An ERD2004 detector with 2mm thick eV Products CdZnTe. The CdZnTe was bump bonded to the MAC04 ASIC (centre of the detector). The MAC04 is wire bonded to two SHC04 ASICs, one either side of the MAC04.

II. SPECTRAL PERFORMANCE

The detector was tested in the laboratory with sealed radiation sources. The detector was biased to -400V and cooled to 278K in a vacuum cryostat. Each pixel was calibrated separately. The pixels were calibrated using the coefficients of a linear fit to a plot of the uncalibrated spectral peak positions (V) against the energies of the radiation sources (13.4keV to 59.9keV) used to capture the spectra.

The ERD2004 ASICs uses a comparator circuit to identify an X-ray event on a pixel. The low energy threshold for the comparator was around 10keV with CdZnTe as the detector material. Therefore any event below 10keV was not recorded. This limited the ability of charge sharing corrections in the ERD2004 detectors. Any events that were identified as charge sharing events were discriminated against as it was highly likely that the entire energy of the events was not recorded. The comparator threshold of the ERD2004 detector can be lowered when silicon is used as the detector material. The ERD2004 detector has measured X-rays below 5keV when silicon was used as the detector material.

Fig.2 shows a calibrated energy spectrum from an Am-241 gamma ray source. The full width half maximum (FWHM) of the main energy peak at 59.9keV was 1.93keV. There was a low energy shoulder to the main energy peak due to charge sharing events where a pixel sharing the event had an energy value below the comparator threshold. The peak to valley ratio for the main energy peak was 14.9. The valley was taken to be the average value between 38 and 45keV. The peaks from Np, the daughter product of Am-241, were between 10 and 30keV [7]. The escape peaks of Te and Cd were at 33keV and 37keV respectively.
III. DETECTOR UNIFORMITY

Pixilated CdZnTe detectors need to have a uniform performance over all pixels to be viable option for imaging applications. The uniformity of the detector was tested at the DLS on station B16 [8]. A monochromatic beam at 15 and 20keV was used to uniformly expose the detector. The detector was cooled to 278K and biased to -350V.

The uniformity of the detector was measured by the number of events captured on each pixel, Fig. 3. The lighter colored pixels indicate a greater number of events recorded on that pixel. The 2D pixel intensity map (top) shows the pixel number (0 to 255). The numbering of the pixels is a consequence of the orientation of the two separate SHC04 ASICs in the ERD2004 detector. The most striking feature of the detector non-uniformity is the region of pixels with a low number of events, running from pixel 224 (x=8mm and y=1.2mm) to pixel 79 (x=4.8mm and y=3.3mm). The feature is more evident in the 3D pixel intensity map (bottom). The pixels with the low number of events run from the back to the front of the image. The overall uniformity of the pixels outside of this region is generally poor. The difference in the number of events per pixel was so great that a histogram of the data gave no statistical information. Another notable feature was the region around pixel 101 (x=4.2mm and y=3.8mm). This pixel and the nearest neighbors had a comparatively large number of events. A similar feature was seen in pixel 45 (x=4.2mm and y=1.8mm).

The uniformity of the detector performance was investigated further by examining the uncalibrated spectra on each pixel. The detector was uniformly exposed to 15keV X-rays when cooled to 278K and biased to -350V. Fig. 4 shows the position of the spectral peak (V) from the 15keV X-rays. The lower the 15keV peak position in a pixels spectrum, the lower the charge collection efficiency on that pixel. The pixels with a spectrum with a lower peak position are shown with lower peaks and blue colors. The region of pixels with a low number of events in Fig. 3 has a lower peak position relative to the other pixels.

![Normalized pixel intensity maps for a uniform exposure of 20 keV X-rays. The plots show the number of events captured per pixel in 2D (top) and 3D (bottom). The lighter colors indicate a larger number of recorded events. The 3D pixel map has been rotated to show the region of pixels with a low number of events.](image-url)
The energy resolution of the pixels was measured by taking the FWHM of the main energy peak of the uncalibrated spectrum from a uniform exposure of 15keV X-rays. The pixels with better energy resolution are those with lower FWHM. Fig.5 suggests that the region of pixels with a lower number of events (see Fig.3) have a better energy resolution than those around them. This result does not represent the true spatial variation in the detector energy resolution. It is a consequence of low statistical numbers on some pixels and a poor fitting algorithm. The data was collected with X-ray energy close to the low energy cut-off of the detector which may also contribute to the unusual results seen here. This experiment should be repeated at higher energy with a greater number of events on each pixel to get an accurate measurement of the spatial variation in energy resolution of the detector.

The CdZnTe detector was tested in the laboratory with a fluorescence foil X-ray source. The detector was uniformly irradiated by an Ag fluorescence X-ray source (22keV). The detector was biased to -350V as in the experiments at DLS. The temperature of the detector was set to 295K and 268K. The number of events per pixel was used to determine the detector uniformity. At 295K (Fig.6) the detector was generally more uniform than at 268K (Fig.7). The region of pixels with a low number of events present at all temperatures. At 268K there were four regions where the number of events per pixel was high, and all other pixels had a comparatively low number of events.

The CdZnTe detector was tested with a Tb fluorescence X-ray source (44keV). The detector was biased to -350V and was at 295K. The number of events per pixel was plotted in Fig.8. The pixels were generally more uniform than with the lower energy X-rays. The region of pixels with the low number of events was still present. To the left hand side of the pixels with low number of events (as viewed in Fig.8) the pixels have a relatively high number of events.
These experiments show that there are extreme non-uniformities in CdZnTe detectors. The experiments identified a region of pixels that had a comparatively low number of events from a uniform exposure. The same region of the detector had pixels with a lower spectral peak position, indicating lower charge collection efficiency. The non-uniformity of the detector improved slightly with increased temperature. This evidence suggests that the non-uniformities are caused by trapping sites in the detector crystal. This is consistent with the extensive work of Soldner, Szeles et al. [9]-[11], who have reported that high Te inclusion densities along twin defects can cause long ranging non-uniformities in CdZnTe detectors. The detector is more uniform when exposed to high energy X-rays (Fig.8). A simple explanation could be that on average the electrons from low energy events have to travel further through the crystal and are therefore more susceptible to the trapping sites. However, the distribution and size of the trapping sites [12], [13], the electric field distribution [14] and contact properties could also contribute to this effect.

IV. LATERAL ELECTRIC FIELDS

A monochromatic 15keV X-ray beam at B16 in DLS was collimated to a size of 40x40μm (FWHM). The detector was biased to -350V and cooled to 278K. The collimated beam was initially positioned over pixel 182 (x=2.1mm and y=3mm). The beam was moved in the x-direction to pixel 73 (x=3mm and y=3mm) with 50μm increments. The scan in the x-direction was repeated at y-axis positions spaced by 50μm, up to the row of pixels, pixel 238 to pixel 17 (x=2.1mm to x=3mm at y=0.9mm). (Please refer to Fig.3 for pixel numbering). The exposure time at each position was 2 seconds.
The number of events per pixel for the entire scan is shown in Fig. 10. The region of pixels with a low number of events was present in the central region of the scanned area. There were a low number of events recorded on the pixels outside of the scanning area due to charge sharing at the edges of the scan.

Fig.10. Normalized pixel intensity map for the complete scanning experiment. The detector was biased to -350V and cooled to 278K.

The scanning experiment was analyzed by viewing the spectrum on each pixel for each scan step across the detectors x-direction (top to bottom in Fig. 11). Three separate scan steps across the x-direction are shown in Fig. 11. The first scan was from pixel 182 to pixel 73. The spectra are shown in blue on the left hand side of Fig. 11. There was significant charge sharing at the start and end of the scan. This charge sharing was caused by irradiating the inter-pixel region between pixel 181 and 182 at the start of the scan, and irradiating the inter-pixel region of pixel 73 and 74 at the end of the scan. The second scan shown in the middle of Fig. 11 with red spectra was from pixel 198 to pixel 57. The notable feature of this scan was the significant charge collected on pixel 64 when pixel 56 was irradiated. The third scan shown on the right hand side of Fig. 11 with green spectra was from pixel 214 to pixel 41. There was significant charge collected on pixel 32 when pixel 40 was irradiated. When pixel 41 was irradiated the majority of the charge was collected on pixel 33.

These spectra can be correlated with the data in Fig. 3. Pixel 41 has a very low number of events compared to pixel 33. Likewise, pixel 56 has fewer events than pixel 64. This is consistent with observation of pixels with low number of events neighboring pixels with a high number of events, by Szeles et al. [11]. The movement of the charge carriers from one pixel to a neighboring pixel must be caused by lateral electric fields in the detector crystal. The origin of the lateral electric fields is believed to be caused by the charge that is trapped in the detector [15]. Further investigations to correlate the trap distribution (and therefore trapped charge distribution) with the lateral movement of charge carriers are required.

V. FUTURE WORK

CdZnTe from eV Products was supplied with pixilation and passivation. The properties of the detector crystal cannot be studied without polishing off the contacts and passivation layer. CdZnTe material from YINnel Tech. Inc. was supplied as unprocessed crystals. Using YINnel material would allow the crystals to be studied before they are contacted, passivated and bonded to ASICs. A transmission infrared (TIR) imaging system was integrated with a stereomicroscope at Rutherford Appleton Laboratory to investigate CdZnTe. Three YINnel crystals were polished within the HEXITEC [16] collaboration. The crystals were TIR imaged on the modified stereomicroscope. The software of the stereomicroscope automatically imaged the crystals at multiple x-y positions and focal planes (z-axis) through the detector. The software can combine the multiple images in the x-y plane at any one focal plane to form an image across the entire crystal x-y area. Additionally, images taken at multiple focal planes through the detector can be combined and projected onto a single image. By combining the two techniques the entire detector volume can be imaged and visualized in a single image. Fig. 12 shows a TIR image of a YINnel crystal. The image combines multiple images in the x-y axis and at different focal planes separated by 25µm. As the depth of focus of the stereomicroscope is 30µm the image shows the entire crystal volume in focus. The automatic measurement software of the stereomicroscope can be used to measure the spatial density of the Te inclusions and give a distribution of the sizes of the Te inclusions. Although these 2D images remove the information of the depth of the Te inclusions, which is significant [13], they offer a simple way of visualizing the distribution of Te inclusions in the entire crystal. It is noted that the automatic measurement software could be applied to the images at
individual focal planes to give the 3D spatial distribution of Te inclusions. A lens with greater magnification has been purchased to investigate smaller defects in the crystal. A future investigation will take TIR images of crystal samples to map the Te inclusion distribution. The CdZnTe crystals will be processed into detectors within the HEXITEC collaboration. The detectors will have the spatial performance analyzed and compared to the Te inclusion distribution.

The performance of ERD2004 detectors is limited by the low energy cut-off of the comparators [17]. The HEXITEC collaboration is developing a new ASIC for CdZnTe detectors [18]. There will be two ASICs, a first iteration has an array of 20x20 pixels and the second iteration will have a larger area with 80x80 pixels. The pixel pitch is 250μm for both ASICs. The ASICs have leakage current compensation circuits and a rolling shutter readout scheme. The ASICs have two gain modes to operate in the energy range of 1-150keV and 1-1.5MeV. The HEXITEC20x20 ASIC has been manufactured and is currently being combined with a custom designed data acquisition system.

VI. SUMMARY

The performance of an eV Products CdZnTe detector on ERD2004 ASICs was tested in B16 at DLS and with sealed radiation sources in the laboratory. The detector had an energy resolution (FWHM) of 1.93keV at 59.9keV and a peak to valley ratio of 14.9. There was a region of pixels with poor performance, believed to be caused by Te inclusions along a twin boundary. A scanning beam experiment showed that lateral electric fields in the detector caused charge carriers generated above a pixel to be collected on neighboring pixels. The non-uniformities and the lateral electric fields could limit the detectors performance as an energy resolving imaging detector. A transmission infrared imaging system has been developed to investigate the correlation of Te inclusions and the non-uniformities and lateral electric fields in CdZnTe detectors. The HEXITEC collaboration has a new ASIC for CdZnTe detectors which will aid future investigations.

ACKNOWLEDGMENT

We thank the beam line scientists and technicians at DLS for the support and advice during the beam time at B16.

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