

Cranfield University

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**Diffraction Enhanced Kinetic Depth Effect
X-ray Imaging**

Department of Engineering and Applied Science

PhD Thesis

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Abstract

An increasing number of fields would benefit from a single analytical probe that can characterise bulk objects that vary in morphology and/or material composition. These fields include security screening, medicine and material science. In this study the X-ray region is shown to be an effective probe for the characterisation of materials. The most prominent analytical techniques that utilise X-radiation are reviewed. The study then focuses on methods of amalgamating the three dimensional power of kinetic depth effect X-ray (KDEX) imaging with the materials discrimination of angular dispersive X-ray diffraction (ADXRD), thus providing KDEX with a much needed material specific counterpart. A knowledge of the sample position is essential for the correct interpretation of diffraction signatures. Two differing sensor geometries (i.e. circumferential and linear) that are able to collect and interpret multiple unknown material diffraction patterns and attribute them to their respective loci within an inspection volume are investigated. The circumferential and linear detector geometries are hypothesised, simulated and then tested in an experimental setting with the later demonstrating a greater ability at discerning between mixed diffraction patterns produced by differing materials. Factors known to confound the linear diffraction method such as sample thickness and radiation energy have been explored and quantified with a possible means of mitigation being identified (i.e. via

increasing the sample to detector distance). A series of diffraction patterns (following the linear diffraction approach) were obtained from a single phantom object that was simultaneously interrogated via KDEX imaging. Areas containing diffraction signatures matched from a threat library have been highlighted in the KDEX imagery via colour encoding and match index is inferred by intensity. This union is the first example of its kind and is called diffraction enhanced KDEX imagery. Finally an additional source of information obtained from object disparity is explored as an alternate means of calculating sample loci. This offers a greater level of integration between these two complimentary techniques as object disparity could be used to reinforce the results produced by the linear diffraction geometry.

*To my nan
gone but not forgotten*

Acknowledgments

I would like to take this opportunity to speak plainly and express my gratitude to a number of individuals, without which I would not have been able to complete this work. Firstly I would like to acknowledge my PhD supervisors. Professor Keith Rogers has acted as a mentor to me, providing guidance and encouragement on countless occasions and Professor Paul Evans, who's insight and support has been vital. As well as my supervisors there are a number of individuals from both Cranfield University and Nottingham Trent University that I would like to express my gratitude towards. Dr. Jer Wang Chan has been invaluable, his technical expertise is second only to his support as a friend. A special thank you should also be given to Mr Joseph Rogers and Mr Adrian Mustey for their technical advice.

Finally I would like to thank my family and close friends. I consider myself very fortunate to be surrounded by so many loved ones. In my times of need they have always found time to support and encourage me, giving me reason to stay the course.

Preface

This PhD thesis is the result of a collaborative, wider study conducted between Cranfield University and Nottingham Trent University to investigate novel configurations that exploit 3D transmission X-ray imaging modalities augmented with X-ray scatter. The goal, to provide compelling 3D X-ray images with spatially resolved materials discrimination. The entire project was funded by EPSRC (under grant numbers EP/F017804/1 and EP/F017596/1).

The novelty of this work is the generation of the first kinetic depth effect X-ray (KDEX) image sequences combined with angular dispersive X-ray diffraction. This information has been embedded via colour encoding. Novel yet practicable scatter sensor geometries have been investigated that facilitate information fusion with KDEX. The overall aim of this work is to develop methods of augmenting KDEX with a much needed material specific counterpart.

Contributions to Knowledge

The main contributions to knowledge of this thesis are:

- Novel X-ray image sequences derived from the fusion of kinetic depth effect X-ray transmission imagery augmented with X-ray diffraction obtained in the laboratory (video 3, section 4.4)
- A method of representing the imaging modality. Material identification is highlighted through colour encoding and match index inferred by intensity (section 4.4).
- Simulating a circumferential detector geometry able to provide materials identification of multiple unknown polycrystalline materials at unknown loci (sections 3.3 and 4.2).
- Simulating a linear detector geometry able to provide materials identification of multiple unknown polycrystalline materials at unknown loci (sections 3.4 and 4.3).
- Experimental proof of principle of the circumferential and linear detector configurations (illustrated in sections 4.2 and 4.3.4, respectively).
- Quantitative evaluation of the linear detector configuration by computer simulation when factors such as sample thickness (section 4.3.1), detector distance (4.3.2) and radiation energy (section 4.3.3) are considered.

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4. *Video file illustrating a kinetic depth effect X-ray image sequence. The locus of objects is calculated by tracking object disparity. Tracked objects are encoded in green. Concatenated images of this video are illustrated in figure 4.38.*

Thesis Outline

Chapter 1. Introduction

Within this chapter imaging technology is introduced. A description of the fundamental requirements are given, as well as a brief synopsis of the main applications. The chapter then summaries the current state of art in imaging technology typically, although not exclusively associated with security screening whilst highlighting the current gaps in knowledge.

Chapter 2. X-rays

This chapter starts by describing X-ray radiation and its interactions with matter. A comprehensive review of its use as both an imaging tool and materials characterisation tool is provided. Each of the differing modalities are identified, explored and critiqued. The chapter concludes with the thesis aims and objectives.

Chapter 3. Method

The principles of X-ray diffraction have already been discussed in chapter 2. This chapter introduces the reader to obtaining diffraction patterns in practice. The complications associated with collecting and interpreting diffraction patterns from volumes is explained. Different scatter sensor geometries and methods are hypothesised. Some permutations are simulated and tested. A method of obtaining transmission X-ray imagery is presented. Finally a rationale for colour encoding transmission imagery with diffraction data is proposed.

Chapter 4. Results

This chapter has two distinct themes. The first explores the hypothesised scatter sensor geometries outlined in chapter 3. The differing methods are validated by a combination of computer simulation and experiments in the laboratory. Factors such as elongated samples and radiation energy are explored. The second theme demonstrates dynamic transmission X-ray imagery with material specific colour encoding. A back checking method using image disparity is also demonstrated.

Chapter 5. Discussion

The chapter begins with a review of the current gaps in knowledge highlighted in chapters 1 and 2. The thesis aims and objectives are re-iterated. The performance and feasibility of the differing scattering sensor geometries presented in chapters 3 and 4 are discussed. Confounding factors highlighted through computer simulation in the results chapter are addressed and a possible means of mitigation is proposed. The impact of dynamic transmission X-ray imagery with material specific colour encoding is discussed.

Chapter 6. Conclusion

This chapter summaries the main findings of the project and details the contributions to knowledge. A list of the contributions to knowledge has been included in the Preface.

Chapter 7. Recommendations for Further Work

This project has yielded substantial findings. However given the nature of this research there is still significant blue sky thinking that may prove valuable. A comprehensive list of recommendations for future work is given.

Chapter 1

Introduction

Within this chapter imaging technology is introduced. A description of the fundamental requirements are given, as well as a brief synopsis of the main applications. The chapter then summaries the current state of art in imaging technology typically, although not exclusively associated with security screening whilst highlighting the current gaps in knowledge.

1.1 Motivations and Background

Imaging is the process of providing a visual representation of an item and/or area of interest. However visual representation does not apply exclusively to morphology (i.e shape). Accurate representation of morphology is a requirement of many appli-

cations (examples of which are described in section 1.2). However various imaging modalities have also been developed that describe the distribution of other features (i.e via a map). The features that can be visually represented are diverse, examples of which include temperature [9], stress [10] and humidity [11]. This study is focused on the characterisation and inspection of objects via radiation, therefore the imaging modalities of interest (other than morphology) include elemental composition [12], electron density [13], refractive index [14] and phase / material identification [15].

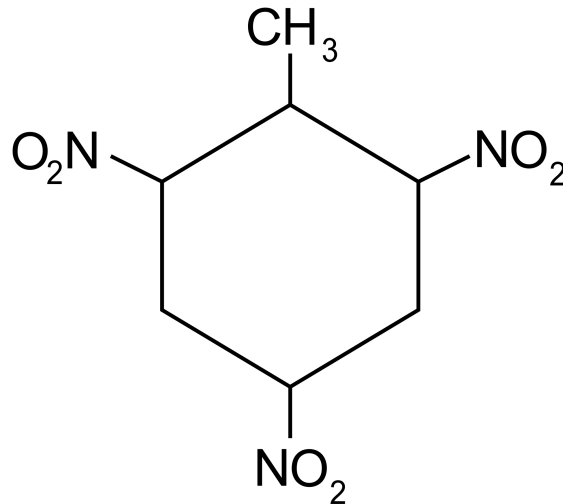
Imaging technologies are designed to address the needs (or answer the questions) dictated by specific applications, although some demands are cross-disciplinary. These same applications also typically impose their own requirements. These act as a framework of rules that the imaging technology must adhere to (e.g. speed, cost etc). Imaging modalities have been designed that utilise differing proportions of the electromagnetic spectrum [16] and although they are often capable of characterising individual materials, some of the more complex applications encountered in real life require an imaging modality that can characterise many different types of material (e.g security screening). A brief synopsis of some prevalent applications (section 1.2) and their associated requirements (section 1.3) are provided in the following sections. Technologies are reviewed with regard to their effectiveness in the following chapter. Areas requiring improvement are also highlighted.

1.2 Applications

Radiation based imaging technologies have been developed to address a diverse spectrum of applications. However, arguably the largest areas relevant to this study include security screening, medicine and material science.

Security screening is conducted to identify and subsequently prevent threat items from entering prohibited areas [17]. The items requiring identification are typically those that pose an immediate safety risk such as knives, guns and explosives and/or illegal items such as controlled substances [18, 19] that although have no immediate safety concerns do have a detrimental socioeconomic impact [20, 21]. Items such as knives and guns are most distinctly recognisable based on morphology [22]. Conversely explosives, and in particular improvised explosives devices (IED's) [23] can be manipulated into any number of benign shapes [18] (e.g. such as sheet explosives,[24]) and so morphological recognition is limited. High explosives such as trinitrotoluene (i.e TNT illustrated in figure 1.1) contain varying proportions of C, H, N and O [25], are generally polycrystalline and have a slightly higher density than typical organic mediums [26](e.g. $> 1.6 \text{ g cm}^{-3}$ [1]). Differing imaging technologies have been developed to aid in identifying these prohibited items with varying degrees of success. However the medium that is examined (e.g. baggage or personnel) will dictate differing operational requirements (section 1.3).

Figure 1.1: *Molecular structure of trinitrotoluene (TNT) illustrating the varying proportions of C, H, N and O (after Akhavan [1])*



Medicine encompasses a wide variety of differing applications from examining anatomy and physiology to disease diagnosis. Accurate morphological representation is often important [27], perhaps in three dimensions. However the types of material to be examined vary dramatically. Bones and teeth have much higher densities than soft tissues such as organs (e.g. the heart and blood vessels). Specimens can also exhibit indicative material characteristics such as the collagen structure for tumor diagnosis [28, 29] or characterisation of urinary calculi [30] for applications in veterinary medicine. Again the medium in which the examination is conducted is prevalent. Some technologies must be suitable for *in-vivo* [27] studies as well as functional imaging that requires speed of data acquisition (described in section 1.3).

Material science is more commonly associated with the quantitative examination of materials in terms of structure. This may include detecting manufacturing defects, an area of importance in forensic science [31] by way of morphological analysis at a macroscopic scale or alternately the quality control of industrial processes (e.g. metal [32], concrete [33] or catalyst [34]) which may warrant characterisation by other means. Within the quality control sector alone there are numerous applications. For instance detecting the presence of impurities (e.g. metal manufacture [32]) or specific phases during the production process (e.g. semi-conductor technology [35, 36]). Alternate means of identification (derived from the use of radiation based imaging) are often employed to aid in characterisation whereby an impetus is placed on high fidelity.

1.3 Requirements

Radiation based inspection systems are employed to fulfill needs set by applications (a selection of which have been described previously, section 1.2). However as well as providing the necessary information (e.g. is an explosive present) other constraints may be imposed. It is important that these requirements be addressed so that the limitations offered by current technology (and thus the gaps in knowledge) that are discussed later may be appreciated.

Speed is often a constraint imposed on imaging technology. In the USA alone over two million passengers and their baggage must be checked for threat items at airports in a single day [37]. The general consensus is that airport screeners must declare an article of baggage is safe in approximately 3 seconds [38, 39]. Speed is also a necessity for many medical applications, particularly where procedures are conducted *in – vivo* and thus areas of interest may not be static (e.g. a beating heart) [2] or for functional imaging.

Cost is a crucial factor for the commercialisation of technologies. Lower cost (in both initial purchase and maintenance) is more desirable. However should the technology reduce the personnel requirements (e.g. less security screening personnel required per shift) then a higher initial cost may be tolerated.

Practicality Some technologies are capable of producing high caliber results but are not feasible for widespread implementation. A good example is synchrotron sources (discussed in section 2.1.4) where their size makes them unfeasible for many applications.

Radiation energy must be suitable for purpose. The energy will dictate the penetrating capability of the radiation. Penetrating through objects with high absorption coefficients (described later) and densities will require high energy incident radiation. However radiation can cause photo-ionization in soft tissues [40] and so is generally avoided where possible. High radiation energies (such as those generated by neutron radiation) will require the use of shielding for safety.

Performance provides an indication of the quality of result returned by the technology. The types of materials in need of evaluation (described in section 1.2) can be characterised in terms of morphology, refractive index, density, elemental composition and phase. These characteristics may be common or unique to a material thus offering differing degrees of fidelity. A common method of measuring performance in medicine is through the use of a receiver operator characteristic (ROC) curves [41] (ROC curves are described in more detail in section 3.5.3).

1.4 Summary

Imaging modalities are often required to characterise materials that vary in morphology, density, elemental composition and material identity. In recent years developments have been made that can achieve unprecedented differentiation in each of these characteristics. However a growing number of applications would benefit from a single imaging modality that can be multifunctional, capable of detecting and representing multiple characteristics simultaneously. The technique should also achieve this whilst conforming to the wealth of additional requirements needed to make it application ready. Taking the security screening example further (although the needs described here are largely cross-disciplinary), no one of the a fore mentioned characteristics is capable of identifying the multitude of prohibited items that may pass through a luggage screening system. Morphology is arguably the most dis-

tinctive characteristic lending itself to the identification of knives and guns, yet is not sufficient to identify improvised explosive devices (IED's) that can conceivably have a benign shape. Similarly the most obvious manner by which to identify explosives and controlled substances is based on chemical composition, yet knives and guns are not chemically distinct (i.e the presence of metal within baggage would not raise suspicion). The area of security has been populated with several imaging and material specific techniques. For a comprehensive review the reader is directed to Yinon [16] and Singh [42]. A more specific review of the technologies currently being developed for detecting concealed weaponry (in particular explosives) can be found in section 8.1. This review is by no means complete but gives a fair representation of the most major areas including neutron technologies, nuclear quadrupole resonance (NQR) and terahertz. These technologies have differing modes of operation providing varying degrees of materials characterisation. However this is often at the expense of cost, speed and / or practicality. A major shortcoming is also their inability to visualise morphology. This means that it would not be prudent to replace traditional X-ray based methods outright. Including these technologies as a supplementary screening stage is conceivable although this would add to both an already exhaustive waiting time and cost.

X-ray detection methods are the most common means to inspect luggage at airports [37] because they provide sufficient performance across a range of applications to make them widespread [43]. This means that X-ray related equipment often benefits from economies of scale. X-ray technology is also mature yet the current research

has also shown its capacity to provide high materials fidelity. From this point on the thesis will concentrate on the use of high energy photons. X-ray interactions are discussed in the following chapter as well as a review of the technologies exhibiting the most promise.

Chapter 2

X-ray Characteristics

This chapter starts by describing X-ray radiation and their interactions with matter. A comprehensive review of their use as an imaging tool is provided. The chapter goes on to describe their use for materials characterisation. Each of the differing modalities are identified, explored and critiqued. The chapter concludes with the thesis aims and objectives.

To appreciate X-rays ability to provide imagery as well as elemental and material discrimination it is important that some fundamental concepts regarding their interactions with matter are understood. However, firstly the manner with which X-rays are generated in a typical X-ray tube will be explained for completeness. There are overlapping concepts and nomenclature regarding the interactions of high energy electrons and the interactions of high energy photons with matter. The explana-

tions of which may facilitate a better understanding. For a more comprehensive overview of X-ray generation and their interactions with matter the reader should review Graham et al. [2].

2.1 Generation of X-rays

Generating X-rays in a typical X-ray tube involves the thermionic emission of electrons from a filament that are accelerated toward a metal target (e.g. copper or tungsten). The kinetic energy with which the electrons strike the target is equivalent to the voltage placed across the filament (e.g. 40 kV). The spectrum of X-ray energies and heat produced by an X-ray source can be attributed to the manner with which these accelerating electrons lose their kinetic energy. The electrons can have **elastic** or **inelastic** collisions with the target. Elastic collisions of electrons conserve kinetic energy. Inelastic collisions involve the loss of kinetic energy and in the majority of circumstances involves the release of electromagnetic radiation. Please note that the difference between elastic and inelastic collisions of electrons described here and elastic and inelastic scattering of photons (discussed later) although the energy relationship still holds true.

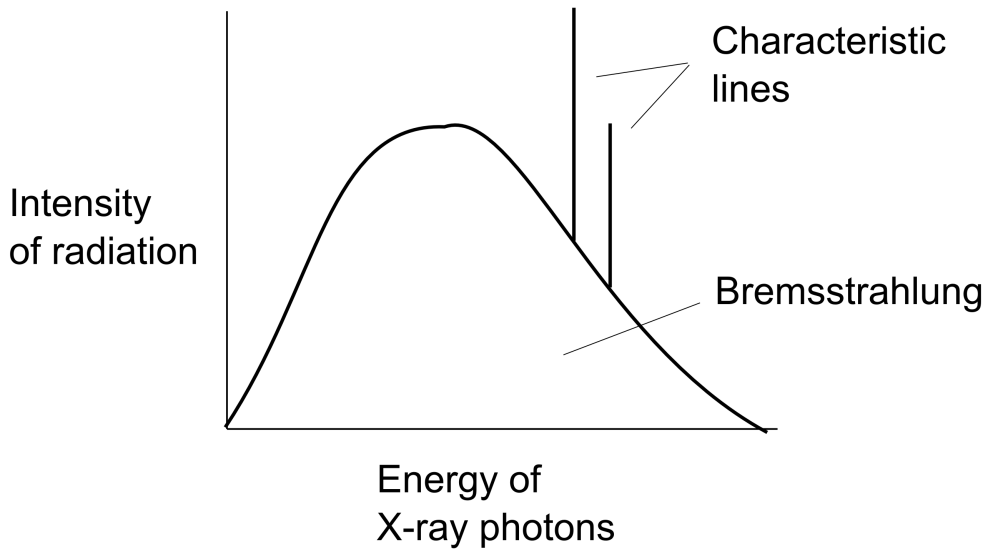
2.1.1 Heat Production

The filament electrons interact with the outer shell electrons, inner shell electrons and the nucleus of the metal target. The majority of interactions are small deflections of the filament electrons with the outer shell electrons of the target. These result in the emission of electromagnetic radiation in the infrared range and thus the generation of heat.

2.1.2 Bremsstrahlung Radiation

Filament electrons can lose kinetic energy by interacting with the nucleus of the target. The kinetic energy lost is emitted as electromagnetic radiation (which may be in the X-ray region). These interactions are inelastic, therefore the amount of kinetic energy lost and energy of X-ray radiation emitted will vary over a continuous spectrum. This continuous spectrum is denoted “Bremsstrahlung”. The maximum X-ray energy emitted is equivalent to the accelerating voltage of the electrons (e.g. 40 kV yields a maximum photon energy of 40 keV) where the electron gives up all of its energy at once. The Bremsstrahlung contribution is indicated in figure 2.1 which illustrates a typical X-ray spectrum.

Figure 2.1: A schematic illustrating a typical X-ray spectrum containing both *Bremsstrahlung* and characteristic lines (after Graham et al. [2]).

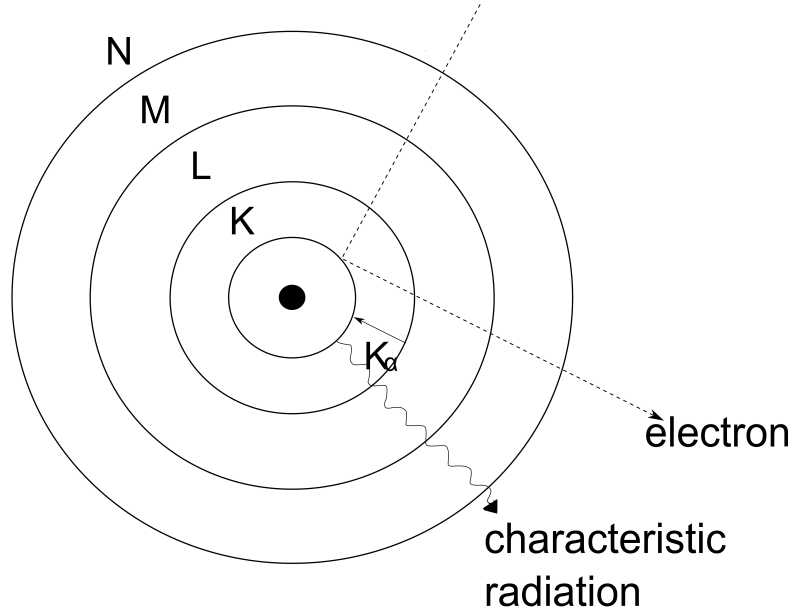


2.1.3 Characteristic Radiation

Characteristic radiation is generated by the inelastic collisions of the filament electrons with the inner shell electrons of the target. In each case kinetic energy is transferred to the inner electron. This may result in excitation or ionisation of the target atom leaving the inner shell with a vacancy. This vacancy is filled via a quantum jump from an electron in an outershell. Any residual energy from this transition is emitted as radiation. The transition depicted in figure 2.2 is an electron moving from the L shell to the K shell. This is known as a $K\alpha$ transition. Other permutations (not shown) can also occur. These transitions result in the production of characteristic radiation energies indicative of the target material (illustrated in figure 2.1). This feature is an important tool utilised to identify the chemical

composition of samples (described later in this chapter).

Figure 2.2: *Depiction of the ionisation of an inner shell electron within a metal target resulting in the release of characteristic $K\alpha$ radiation (after Graham et al. [2]).*



2.1.4 Synchrotron Radiation

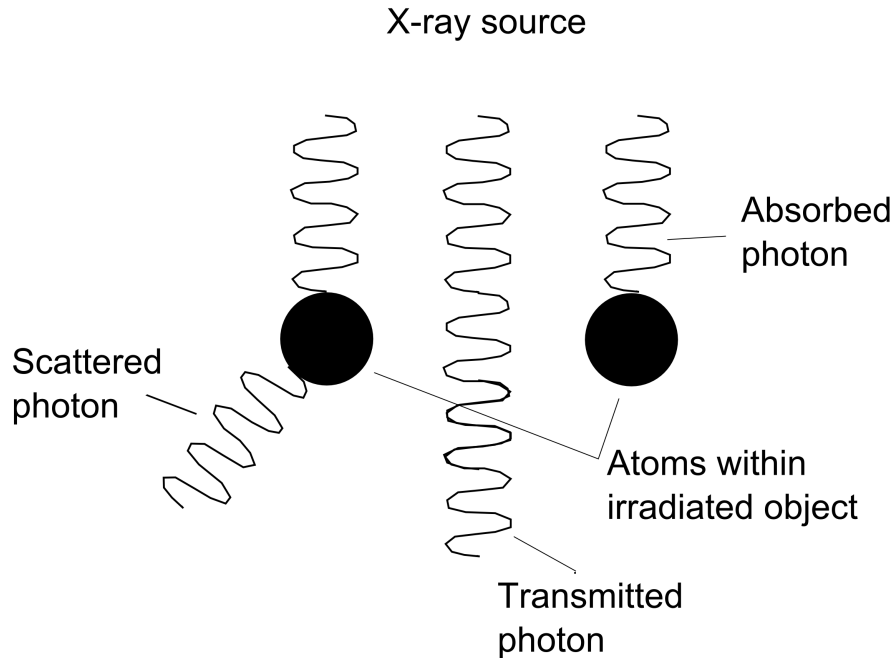
A synchrotron is an example of a particle accelerator in which charged particles (i.e. electrons) are accelerated to a high speed traveling on a circular orbit [44]. The circular orbit is maintained through the use of magnets and the acceleration is induced by an electric field. In the case of electrons, the acceleration and the change in direction (i.e deflection) of the electrons instigates the release of high intensity electromagnetic radiation [45, 46]. Typical characteristics that are anticipated from synchrotron X-radiation included high energies, flux densities and monochromaticity.

The interested reader should review Wiedemann [45]. The maximum curvature of the electrons is governed by the magnetic field strength. This imposes a minimum radius on the size of the synchrotron. Size constraints as well as cost make this type of X-ray source impractical for many applications (section 1.3). This study will concentrate on the use of traditional X-ray tubes.

2.2 X-ray Interactions with Matter

When X-ray photons are incident on matter the result is one of three possible outcomes. These are illustrated diagrammatically in figure 2.3. X-rays can be transmitted through the material (i.e. no loss in energy), absorbed by the material (i.e. total loss in energy) or scattered. The scattering mechanisms can be either **elastic** or **inelastic** resulting in the energy being conserved or transferred by the photon, respectively. These scattering mechanisms are described in greater detail later. The name for the collective absorption and scattering of X-rays by a material is called the attenuation. The four mechanisms by which X-rays may become attenuated are presented in the following sections.

Figure 2.3: *An illustration of the differing X-ray interaction mechanisms that occur with matter (i.e. transmission, absorption and scatter) (after Graham et al. [2]).*



2.2.1 Photoelectric Effect

The photoelectric effect (τ) describes the mechanism whereby energy from an incoming X-ray photon is absorbed by the sample [48]. This mechanism is inelastic and typically the energy of the incoming photon is equal to or slightly above the binding energy of the electron. The photon energy is transferred to the electron and it becomes disassociated from the atom. Any residual photon energy is transferred to the electron as kinetic energy and it is referred to as a photo-electron. Vacancies in the samples inner shell(s) will be filled by quantum jumps resulting in the production of characteristic radiation (similarly to that described previously, section 2.1.3). In radiology the sample is typically organic and thus has low atomic num-

bers (Z). This results in the production of the characteristic radiation in the infrared region (i.e heat) and so is largely ignored. For samples of higher Z the characteristic radiation produced may well be in the X-ray region. X-ray fluorescence (described later) makes use of this interaction. The photoelectric effect is most dominant with lower Z materials [43] because the ionization energies are typically less. Dual energy X-ray imaging (discussed later in section 2.5.1) makes use of this relationship to provide qualitative information regarding atomic number. This is described further in section 2.2.5.

2.2.2 Compton Scattering

Compton scattering (σ) is also an inelastic scattering mechanism. This interaction occurs when the energy of the incoming photon greatly exceeds the binding energy of the sample's electrons. In this circumstance the electron is considered to be a free electron. A proportion of the energy from the incoming photon is partially absorbed when it scatters off the electron. The energy of the incoming photon is reduced and the residual energy is transferred to the electron. However, the total energy of the interaction is conserved [2]. For low energy X-rays (up 100 keV) the photons are scattered isotropically. This form of radiation is useful because it is related to the local electron density of the target [49] and so it is predominately used to provide relatively high resolution density images [49]. Factors affecting the Compton attenuation coefficient are described later in section 2.2.5.

2.2.3 Thompson Scattering

Thompson scattering (σ_{coh}) (i.e elastic or coherent scattering) occurs when the energy of the incoming photon is less than the binding energies of a sample electron. Rather than ionize or excite the electron (i.e photoelectric effect) or completely disassociate the orbital electron (i.e Compton scattering) it raises the electron's energy slightly. When the electron returns to its previous energy state it emits an X-ray photon of equivalent energy as the incident photon. This scattered photon is distributed isotropically from the atom (i.e the X-ray suffers no change in energy merely in direction [50]).

The elastic scatter field is regarded as the most material specific of all the X-ray techniques, competing with neutron based techniques in specificity [43]. Coherent scatter is described later for use with X-ray diffraction.

2.2.4 Pair Production

Pair production (π) is the highest energy X-ray interaction. This effect is caused when a high energy X-ray photon strikes a charged object such as a nucleus with an energy exceeding 1.02MeV [51]. The energy is sufficient to produce an electron and positron pair. If the electron and positron pair remain in close proximity to each other after their production then they will annihilate (this process effectively absorbs energy). The energy of the initial photon after this process is equal to its original

energy minus 1.02MeV. This interaction is not prevalent to the method discussed later in this document as the X-ray energies used here are widely considered to be too low to bring about this interaction.

2.2.5 Attenuation Coefficients

The degree to which an X-ray beam is attenuated by a sample is calculated by the linear attenuation coefficient (μ) or more commonly the mass attenuation coefficient (μ/ρ), where ρ is the material density. It is the difference in attenuation coefficient between sample materials that produces contrast in a radiograph, allowing identification based on morphology. However the total mass attenuation coefficient is the combined effect of the four attenuation mechanisms described previously (i.e photoelectric effect, Compton scattering, Thompson scattering and Pair production). This is expressed in equation 2.1 where τ , σ , σ_{coh} and π represent the photoelectric effect, Compton scattering, Thompson scattering and Pair production contributions respectively.

$$\frac{\mu}{\rho} = \frac{\tau}{\rho} + \frac{\sigma}{\rho} + \frac{\sigma_{coh}}{\rho} + \frac{\pi}{\rho} \quad (2.1)$$

These attenuation mechanisms are affected differently by atomic number (Z) and X-ray photon energy (E). This has facilitated the development of more sophisticated techniques (i.e dual energy imaging, section 2.5.1) that provide insight into the

material makeup of an interrogated sample. For the X-ray energies discussed in this study (i.e. $\ll 1.02$ MeV) the attenuation coefficients owed to pair production (π) can be ignored. However the attenuation coefficients for the photoelectric effect and Compton scattering are of importance. These are illustrated in equations 2.2 and 2.3, respectively.

$$\frac{\tau}{\rho} \propto \frac{Z^3}{E^3} \quad (2.2)$$

$$\frac{\sigma}{\rho} \propto \frac{\text{electron density}}{E} \quad (2.3)$$

Equation 2.2 is an approximation for X-ray energies (E) up to 200 keV. The photoelectric coefficient is reliant upon X-ray energy (E) and the atomic number (Z) where as the Compton scattering is reliant on the X-ray energy (E) and electron density. The electron density for most materials is approximately the same (i.e. $\sim 2.5 \times 10^{23}$ electrons per gram with the exception of hydrogen because of the absence of a neutron [2]). Therefore the attenuation due to Compton scattering occurs largely independent of the sample material. This relationship is utilised later in section 2.5.1 (dual-energy X-ray imaging) to derive effective atomic number. Please note that here it is the photons absence due to scatter that is used. Other techniques discussed later harvest the scattered X-rays (both coherent and incoherent) to provide additional sample information.

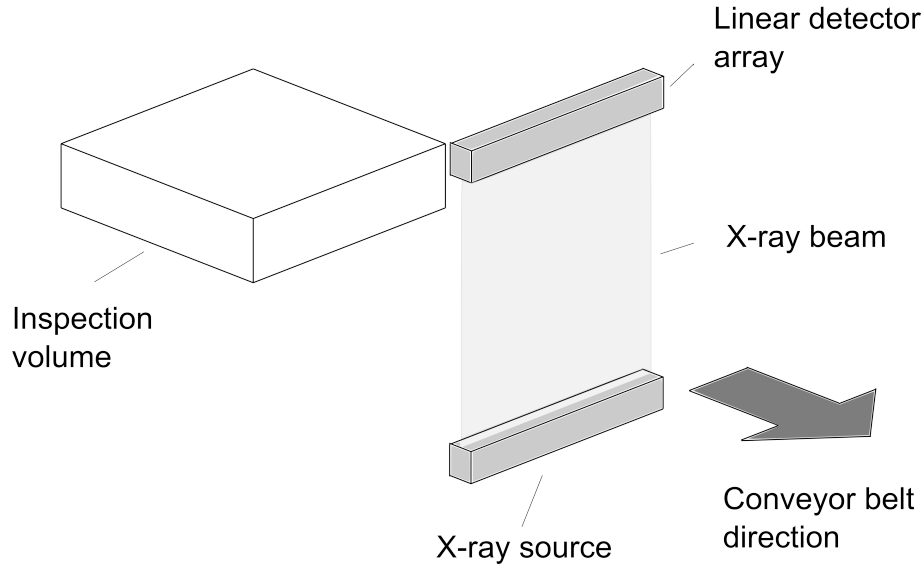
2.3 X-ray Imaging Techniques

In this section the most prominent X-ray imaging technologies are discussed. Particular interest is given to the manner in which they address the requirements (discussed in section 1.3) of an imaging system from an application perspective.

2.3.1 Transmission X-ray Imaging

Transmission X-ray imaging (i.e. radiography) is perhaps the simplest and most mature X-ray imaging technique discussed in this section. In principle a broad spectrum of X-rays (similar to that illustrated in figure 2.1) is used to irradiate a sample (i.e a person or baggage). A proportion of the X-rays from this spectrum are attenuated by the sample before being captured by a detector [2]. Different areas within the sample attenuate the X-ray beam by differing amounts. This is due to the differences in mass attenuation coefficient (μ/ρ) between differing materials, discussed in the previous section (i.e section 2.2.5). These differences produce contrast in the resulting X-ray image permitting objects to be identified based on morphology. This imaging technique can operate at speed (e.g. 0.2 m/s [4]). Transmission X-ray screening systems (commonly found at airports) are adapted to work on a conveyor belt. Luggage is translated on a conveyor belt at constant speed so that it intersects a fan beam arranged normal to the conveyor belt direction. A typical geometry is illustrated in figure 2.4. Individual lines through the inspection volume (i.e baggage)

Figure 2.4: *A depiction of a typical transmission X-ray screening geometry such as those found at airports where an inspection volume is passed through an X-ray beam at constant speed to build a transmission X-ray image.*

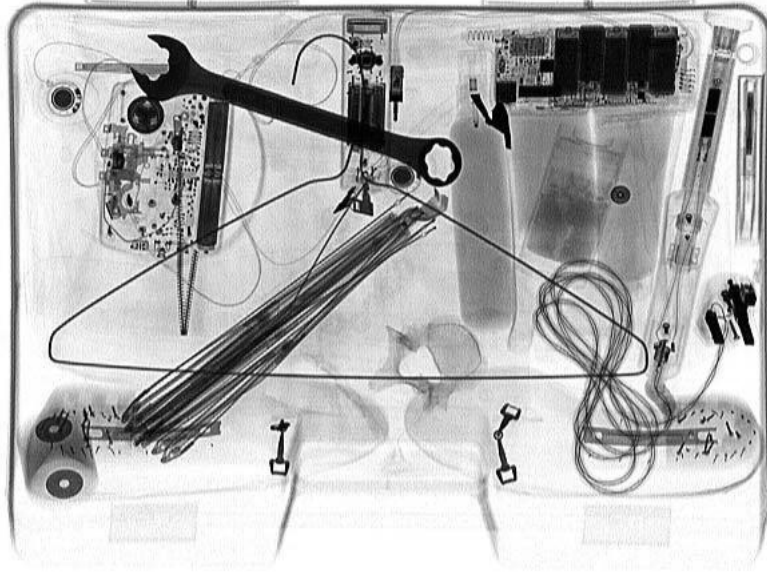


are imaged in quick succession (i.e line scan, [43]) to build a 2D image, an example of which is illustrated in figure 2.5.

High Z materials such as metals stand out prominently due to their high attenuation coefficients. This is advantageous from a security perspective as typical threats (including knives and guns) that are made of metal are readily recognisable based on morphology. This technology is also typically cheap which makes it the work horse of airport security screening [37].

One of the main shortcomings of this technology is that it cannot differentiate between a thin sheet of a high attenuating material and a thick sheet of a low atten-

Figure 2.5: *An example of a transmission X-ray image illustrating the contents of a suitcase.*



uating material [37] (i.e $\mu x = \mu x$ though the linear attenuation coefficient (μ) and path length through the material (x) are different) . Also this technique represents a 3 dimensional object within a 2D image producing super positioning. These effects, when combined can result in the transmission imagery appearing cluttered (i.e numerous objects separated in depth overlap in the image) making interpretation problematic. Objects can also become occluded (i.e hidden) by high attenuating materials. These problem could be alleviated if more than one perspective were considered (a common theme adopted by the forthcoming techniques).

2.3.2 Computerised Tomography

Computerised tomography (CT) was largely developed for medical imaging [52, 53] but has also been applied to other fields including security screening [37] and mate-

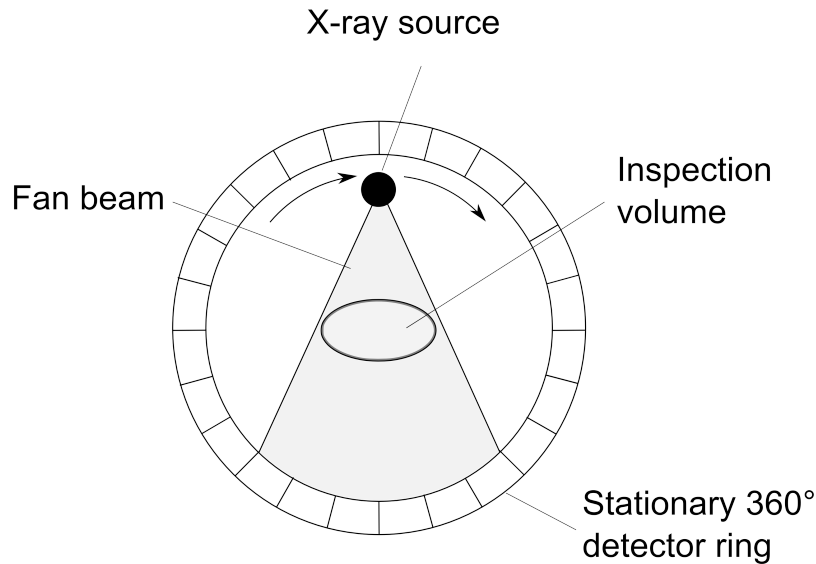
rial science [54, 55] (which have been summarised in section 1.2). This technology involves capturing of a series of X-ray line scan images (referred to as views) around 360° that all intersect one plane through an object of interest[2]. These views are used to compute an image of a cross sectional plane through the inspection volume via image summation (referred to as a tomogram or a slice). The object under inspection is then traditionally translated and the process repeated so that differing planes through the volume are intersected by the interrogating beam. These axial “slices” can then be collated to produce a 3D reconstruction of the objects contents [37]. This approach provides an improvement over traditional X-ray imaging technology in that super positioning (described previously) is removed. Also objects that may have been occluded by high attenuating materials may be revealed by the computation of multiplanar cross sectional images. Differing CT geometries have been developed. Figure 2.6 illustrates a fourth-generation geometry where the cross section of an object under inspection is being irradiated to obtain such a view. In this configuration the inspection volume and detector ring remain stationary and the X-ray source rotates around a central axis.

Arguably the main developmental drive for CT scanners has been for faster data acquisition (described previously in section 1.3) [3]. In the field of medicine this is of particular importance as some areas in need of examination do not remain static for a significant time period (e.g. lungs of a patient) [3]. A brief synopsis of the scan times and mode of operations of differing generations of CT scanner is illustrated in table 2.1.

Table 2.1: *A table illustrating a synopsis of the major developments in CT scanners. This includes a summary of their mode of operation and the scan times of differing generations (after Delchar [3]).*

Type	Mode	Scan time (per slice)	Comments
First-generation	Pencil beam, 160 lateral steps followed by 1° rotation for each perspective image	4-5 min	Source and detector move together on a gantry
Second-generation	Fan beam subtending 10° angle, reduced number of lateral steps followed by 10° rotation for each perspective image.	~ 20 s	As above but with the inclusion of multiple detectors. Data collected for several sets of parallel rays, total number of rays increased.
Third-generation	Wide angle fan beam encompasses inspection volume, no lateral steps, source now pulsed and rotates without stopping.	$\sim 4-5$ s	Detector balance and calibration problem; several hundred detectors required. Several hundred readings obtained for each pulse.
Fourth-generation	Wide angle fan beam (non-pulsed) source rotates without stopping within ring of stationary detectors.	$\sim 4-5$ s	Real time calibration of detectors possible.
Fifth-generation	Scanning X-ray source, no mechanical moving parts.	~ 50 ms	Essentially simultaneous multiple slice images possible.

Figure 2.6: A depiction of a fourth-generation CT geometry whereby an X-ray source is rotated through 360° around an inspection volume to obtain a cross sectional image. (after Delchar [3])



The latest generation of CT scanners (i.e sixth and seventh) often referred to as “spiral” CT scanners continually move along the object obtaining images whilst rotating, in effect moving on a helical path (rather than the traditional approach of capturing all of the necessary views to construct one cross sectional plane before moving to inspect the next plane) [56]. This approach presents obvious advantages in speed of examination but is computationally demanding as data is continually streamed to the computer rather than the a fore mentioned approach where data is sent in discrete packets [2].

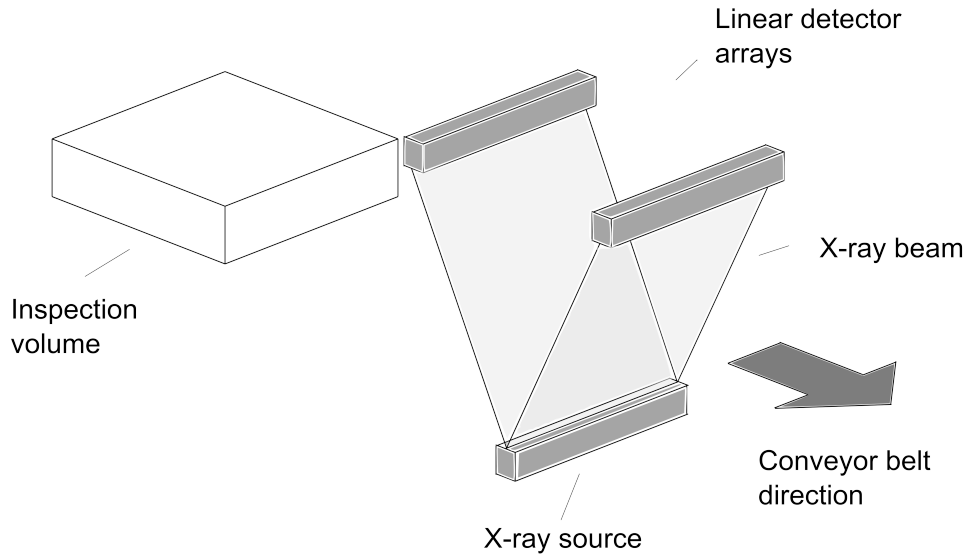
The main limitations of CT scanners are that they require very high signal/noise ratios to produce viable images for reconstruction. This means that typically X-ray

sources with high energies and currents are required to fully penetrate objects from many different radial positions [5]. From a luggage inspection perspective this creates radiation protection issues and the further requirement for shielding [5]. There is also a high associated system cost which is largely due to the complexity of the mechanical equipment and difficulty in collimation alignment [37]. This complexity also has a significant contribution to the mass of the unit with a typical CT scanner mounted on a gantry weighing in the order of a hundred kilo's or more [5]. The 3D aspect that this technique can provide is of great importance. However it has been argued that CT scanners are particularly data 'hungry' [37, 57] requiring many images to create accurate image reconstruction [58] although the speed of their operation has vastly improved.

2.3.3 Stereoscopic X-ray Imaging

Stereoscopic X-ray imaging is an imaging technique that only requires two transmission X-ray images [57]. These images are obtained at differing perspectives to the inspection volume. A typical geometry applying this technology to security screening is illustrated in figure 2.7 although it should be noted that other applications including medicine [59] make use of this technique. The perspective images (i.e right and left) are displayed separately to the viewers eyes (via polarising glasses or alternate medium) to instigate binocular parallax [60]. Binocular parallax provides the viewer with depth perception, thus addressing the superposition issues that have

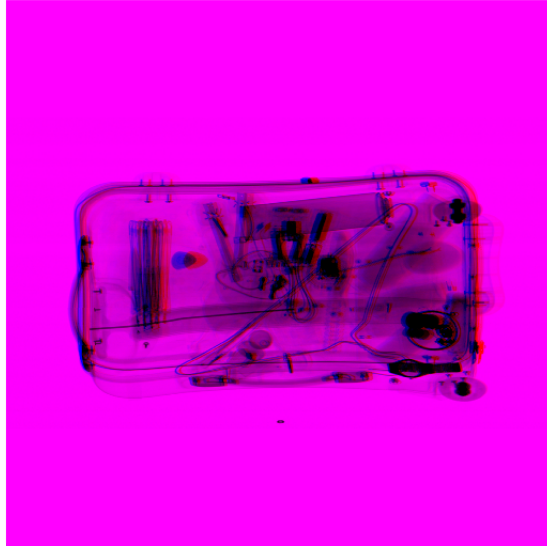
Figure 2.7: *A depiction of a typical stereoscopic X-ray imaging geometry such as those found at airports where an inspection volume is passed through two X-ray beams at constant speed to build a left and right perspective X-ray image (after [4]).*



been discussed previously (section 2.3.1). An example image depicting a suitcase has been included in figure 2.8. The right and left perspective images have been captured with an angular separation of 3° . Experiencing binocular parallax from this image requires the use of red/blue filtered '3D' glasses (providing that the image is in colour). Please note that this image has been included for completeness and its viewing is strictly optional.

Increasing the angular separation between the perspective images increases the observed disparity. This in turn increases the depth of field [60] enabling objects that have a closer proximity in depth to be more readily separated. However it has been

Figure 2.8: *An example of a stereoscopic transmission X-ray image illustrating the contents of a suitcase. Left and right perspective images have been encoded in red and blue. Binocular parallax can be realised with the use of red and blue filtered glasses.*



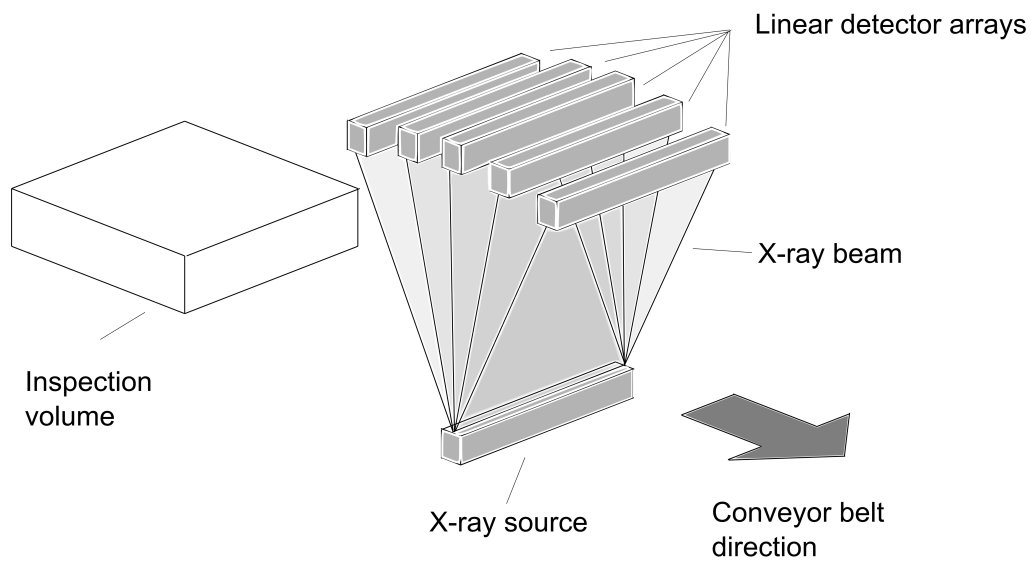
demonstrated that there is an upper limit in angular range that can be accommodated. Choosing too high an angular separation between the perspective images may prevent viewers from successfully “fusing” the images into a single coherent image. An angular separation of $\pm 2^\circ$ has been found to be optimum by Darambara et al. [59]. This technique can operate at speed (i.e typically 5 seconds per bag [60]) and is anticipated to be somewhat cheaper than that offered by CT systems. However it cannot reasonably compete with the depth perception afforded by CT systems owed to the limited angular separation requirement imposed by the screener.

2.3.4 Kinetic Depth Effect X-ray Imaging

Kinetic depth effect X-ray imaging (KDEX) utilises multiple (>2) transmission images obtained with an incremental angular separation between each “perspective” image [5]. A typical geometry applying this technology to security screening is illustrated in figure 2.9. A “line scan” image is obtained from each of the linear detector arrays when the inspection volume is translated through the X-ray fan beam at constant speed. Conceivably these detectors could be arranged in such a manner as to not impede the conveyor belt time [5]. The images (each representing a slightly different perspective) are then displayed in sequence to the viewer so that the inspection volume appears to rotate. This is conceptually equivalent to fixing the linear detector array and X-ray beam orientation and incrementally rotating the inspection volume. An example of a KDEX image sequence has been included (video 1) which illustrates a suitcase imaged from $\pm 15^\circ$ with 1° angular separation between each perspective image.

Video 1 induces a parallax effect to the operator brought about by the differences in angular velocity of items within the inspection volume. In the example provided the items nearer the top of the bag modify their position (i.e demonstrate greater disparity) between each perspective image then those at the bottom of the bag. The relative rate of modification therefore inferring a position in depth. This affords depth perception via motion parallax [5]. The motion parallax offered by KDEX is more descriptive than the binocular parallax effect described previously for stereo-

Figure 2.9: *A depiction of a kinetic depth effect X-ray imaging geometry where an inspection volume is passed through a series of X-ray beams at constant speed to build a transmission X-ray images at differing orientations to the conveyor belt (after Evans et al. [5]).*



stoscopic imaging and Evans [39] suggests that the depth perception is perhaps more compelling. This technology addresses the super positioning and occlusion issues encountered by 2D transmission imagery (section 2.3.1) similarly to computerised tomography (section 2.3.2) and stereoscopic X-ray imagery (section 2.3.3). However arguably it presents some significant advantages over both. The depth perception offered by stereoscopic X-ray imaging is limited by the angular separation of the perspective images that can reasonably be “fused” (i.e approximately $\pm 2^\circ$) by the screener. KDEX has no such limitation and typically operates over $\pm 15^\circ$. This makes the depth perception more compelling Evans [39]. Once more this can theoretically be achieved at an equivalent speed (providing the inclusion of enough linear arrays similarly to those illustrated in figure 2.9) without the need for moving parts. The latest spiral CT scanners (discussed in section 2.3.2) can also operate at a similar speed and the 3D imaging ability is arguably better. However this typically comes at great cost and requires mechanical movements which is undesirable. In fact KDEX technology is anticipated to cost at least an order of magnitude less than the purchase and running costs of a CT scanner [5] and so for financially sensitive markets KDEX may offer a viable alternative.

2.4 Elemental and Phase Specific X-ray Techniques

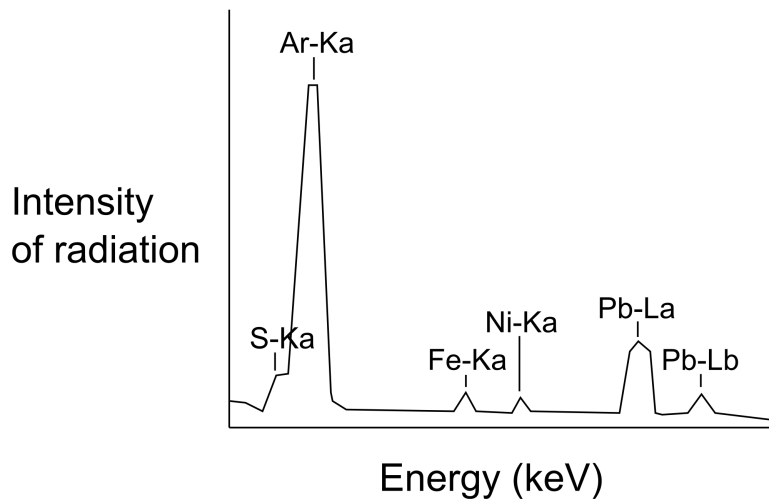
X-rays are a versatile analytical probe and are often employed to evaluate materials based on other characteristics rather than morphology (i.e dictated by mass attenuation coefficient, μ/ρ). Some of the main areas are described and discussed in the following sections.

2.4.1 X-ray Fluorescence

X-ray fluorescence (XRF) can be used to identify the elemental composition of a sample [61]. The mechanism's involved are conceptually similar to the manner with which characteristic radiation is produced (described earlier in section 2.1.3). Figure 2.2 illustrates that should electrons have sufficient energy they can be used to ionise or excite inner shell electrons of a material, creating a vacancy. This vacancy is subsequently filled with an electron from an outer shell (relaxation) [62], and may result in the emission of an X-ray quanta. The energy of the emitted quanta is element specific governed by the differing energy states of the orbitals. In XRF, the high energy particle instigating the excitation / ionisation of the inner shell electron (in the sample material) can be a photon rather than an electron (similarly to that described in the photoelectric effect, section 2.2.1, although other high energy particles can be used in technologies such as energy-dispersive X-ray

(EDX) [63, 64, 65] and particle induced X-ray emission (PIXIE) [66, 67, 68]). The instigating photons are referred to as primary X-rays and the characteristic X-rays emitted by the sample are referred to as secondary X-rays. The secondary X-rays are energy resolved to provide the elemental composition [49] (an example of which is illustrated in figure 2.10). The elemental composition can be measured for differing loci to produce a map [69].

Figure 2.10: *Schematic example of an X-ray fluorescence spectrum where peaks at differing energies can be attributed to the elemental composition of the sample (after [6]).*



There are some aspects of X-ray fluorescence that are undesirable. The emitted radiation from lighter elements is prone to interaction with the surrounding sample medium and atmosphere. This typically puts a lower limit on the elements that can be detected (i.e $Z < 19$) [74], which can lead to materials being identified based on the absence of elements rather than their presence [75]. Therefore there is an argument to suggest that this technique is ill-equipped to deal with the problems faced by the

applications stated in section 1.2. Typical materials of interest (e.g. biological or explosive materials) are comprised of low atomic numbers (e.g. C, H, N and O) [2, 25] and so may not yield detectable fluorescence signatures. Even if these particular elements were detected, their presence is not indicative of a material. Instead it is the molecular arrangement of the atoms that govern the chemical properties of the materials not just the atomic constituents [76]. This last point is a common theme and discussed later for dual energy X-ray imaging (section 2.5.1)

2.4.2 X-ray Diffraction

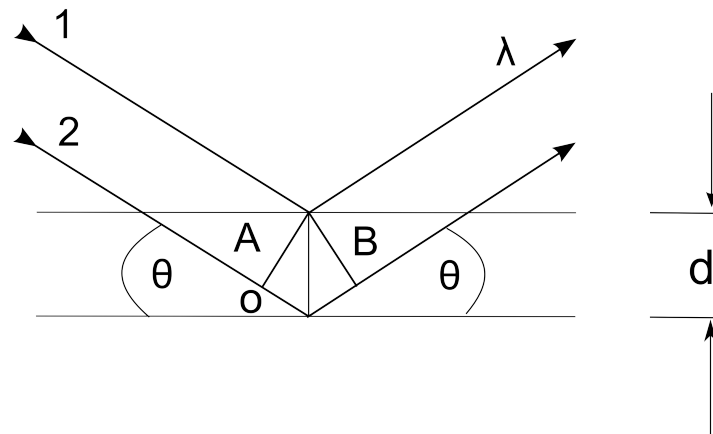
X-ray diffraction is routinely used in material science and has recently been applied to security screening because of its almost unequivocal ability to provide material discrimination.

When a material is exposed to radiation a proportion of the incident rays are elastically scattered from the electrons surrounding the atoms (discussed in section 2.2.3). This elastic scatter is distributed isotropically [77]. Each atom therefore acts as a source of an ever increasing sphere of elastically scattered radiation. Should two atoms lie within close proximity then their respective spheres of elastic scatter will interact. Radiation can be represented by a sinusoidal wave (similarly to that illustrated in figure 2.3). Therefore the scattered rays from the adjacent atoms may interact by interfering constructively or destructively depending on the angle of propagation, wavelength of radiation (λ) and importantly the distance between the

atoms. Constructive interference will occur if the rays are in phase with one another (i.e. aligned in peak and trough), resulting in a ray of the same wavelength but increased amplitude. Conversely should the rays be out of phase (e.g. by $\frac{n}{2}\lambda$) then a reduction in amplitude will be observed. It is the spatial relationship between these atoms that is of importance, altering the distance between the atoms will invoke a change in the angle(s) giving rise to constructively interfered rays.

Crystalline materials have a high degree of periodicity. The spacings between atoms are highly regimented. These are governed by the Z number of the atoms and their bonds [78]. These spacings are therefore indicative of the material identity. The spacings between the atoms are typically in the order of angstroms and therefore in the X-ray region of the electromagnetic spectrum.

Figure 2.11: A diagram that illustrates the derivation of Bragg's law (i.e. $\lambda = 2d \sin \theta$) (after Bowen and Tanner [7]).



When monochromatic X-rays strike a crystalline material, constructive and destructive interference effects result in the elastically scattered X-rays being emitted at characteristic angles. These angles (θ) are indicative of the spacing (d) between the

planes of the crystal structure and are governed by Bragg's law [48]. This can be visualised with the use of figure 2.11.

In figure 2.11 rays 1 and 2 illustrate the paths of monochromatic radiation that before interacting with the sample are in phase. Both rays are reflected isotropically from the planes within the sample although only one direction of reflection has been illustrated. Ray 2 suffers phase retardation [79] by traveling the additional path difference AOB. For the rays to still be in phase with each other on exiting the material and thus constructively interfere (i.e diffract) then the path difference (AOB) must be equal to $n\lambda$ where n is an integral number of wavelengths [7]. This relationship is governed by Bragg's Law:

$$n\lambda = 2d \sin \theta \quad (2.4)$$

Where λ is the wavelength of the X-rays used, d is the inter-planar spacing within the material and θ is the angle between the X-ray beam and the atomic plane [62].

However it should be noted that not all d-spacings will yield diffracted rays at a characteristic angle (for a given wavelength). Diffraction can occur at the same characteristic angle simultaneously from more than one d-spacing within a unit cell for the same wavelength of radiation (where the path difference for one set of diffracting rays may be λ and another path difference may be 2λ). These diffracting rays (from differing d-spacings) are independent. They may constructively inter-

ferre (further increasing peak amplitude at the characteristic angle) or they could destructively interfere and entirely extinguish the scattered rays emerging from that particular d-spacing (i.e systematic extinctions) [79]. In this respect the absence of diffracted rays (i.e Bragg maxima) is in effect as telling as the presence of diffracted rays. The amplitude of scatter arising from parallel planes of atoms is therefore related to scattering from each atom (j) in the unit cell and is given by the structure factor (F_{hkl}) illustrated in equation 2.5 after Jenkins and Snyder [79].

$$F_{hkl} = \sum_{j=1}^m f_j \exp[2\pi i(hx_j + ky_j + lz_j)] \quad (2.5)$$

where x , y and z are the fractional coordinates of the atom within the unit cell, h , k and l are the scattering vectors and m is the total number of atoms within the unit cell. To identify the “phase” of a material (i.e material identity) typically all of the d-spacings are measured and compared to a material database.

Illuminating a single crystal from one orientation will not yield diffraction signatures that represent all of the d-spacings within the material. It is more likely that no planes within the material will fulfill the Bragg geometry. The single crystal is typically rotated in the interrogating beam so that all planes fulfill the Bragg geometry at a given orientation. An alternate method is to investigate a powder (comprised of much smaller crystals) of the same material (i.e powder diffraction). It can then be assumed that the sample under consideration has a random arrangement of crystals

so that the angles over which diffraction occurs are representative of all the differing d-spacings. Typically the wavelength (λ) of radiation is kept constant and the angle over which diffraction occurs is measured in reflection. However an alternate technique, energy-dispersive X-ray diffraction (EDXRD) is currently receiving much interest. Angular-dispersive X-ray diffraction (ADXRD) and EDXRD are discussed in the following section.

Powder X-ray diffraction can be used for more than phase identification. Examples of other features that can be characterised include:

- Residual stress
- Residual strain
- Crystallite size
- Preferred orientation

These features are not particularly relevant to this study. However the interested reader should review Cullity [48].

Comparison of ADXRD and EDXRD

This section serves to compare the use of angular and energy dispersive X-ray diffraction (ADXRD and EDXRD, respectively) for powder diffraction models. ADXRD

utilises a monochromatic interrogating X-ray beam (e.g. typically one of the characteristic lines described previously in section 2.1.3) and the coherently scattered radiation is measured over an angular range (typically denoted 2θ) [86, 78, 48]. Measurements can be collected by a point detector that is moved sequentially with constant radius to the sample. This incurs a low cost for the detector but increased time overhead for data acquisition. Alternatively the entire diffraction pattern can be collected simultaneously on a 2D area detector. This offers a relative gain in speed (because no mechanical movement is required) but has a higher associated cost for the detector. ADXRD has been applied in security screening studies [76], medicine [28] and material science [87, 88, 89] with authors such as Jupp et al. [76] demonstrating that explosives can be unequivocally identified in 5 seconds.

Most commercially available ADXRD systems (with the exception of those discussed in section 2.5.4 as well as Roncallo et al. [35, 36]) are not designed for simultaneous measurements of spatially separated materials (i.e. mapping). Additional time overheads owed to sample movement are often required (or alternatively the angle of incidence for depth profiling [90]) to acquire spatially separated measurements. An alternate method has been brought about with the development of EDXRD which is arguably the best technique currently available (indicated by the number of publications cited later). This technique utilises a polychromatic X-ray source (similar to that illustrated in figure 2.1) which boasts a comparatively higher flux (because it utilises all of the generated Bremsstrahlung wavelengths) than the monochromatic beam used for ADXRD. The coherently scattered X-rays are then collected and en-

ergy (λ) resolved by the detector after diffracting through a fixed angle determined by a diffraction cell. This instrumentation requires no moving parts as the angle of diffraction is fixed. Luggar et al. [91] reports the production of real time measurements of explosives showing characteristic profiles measured in 0.1 seconds. This is a common theme as authors report similar measurements acquired at speed when identifying explosives [92, 93] as well as other applications including medicine [94] and material science [33, 32].

However EDXRD has limitations. The increase in scattering flux obtained from using a polychromatic source (over a monochromatic source) is seldom realised because the majority of the scatter beam flux ($\sim 99\%$) is lost due to collimation [95]. This collimation defines the angle of intersection between the incident and diffracting rays. There is some debate as to the optimum angle of intersection. Coherent X-ray scatter dominates over incoherent scatter in the forward direction [37]. Lazzari et al. [96] suggests the use of low 2θ angles in the region of $1-5^\circ$. Greater sensitivity can be obtained by selecting a lower angle closer to the path of the incident beam (e.g. 3°). This facilitates the analysis of higher attenuating materials because greater penetration is achieved. However at higher angles it is possible to achieve better energy resolution [91]. This choice of angle also presents a problem in that the diffraction pattern obtained from one angle may not be as descriptive as the pattern obtained from an alternate angle [97]. Sun et al. [20] reports a difference in the optimum diffraction angle to detect trinitrotoluene (TNT) and heroin (i.e. 10° and 12° , respectively). Another confounding factor is that of energy resolution. Solid state

energy dispersive detectors cannot offer an equivalent resolution to that of ADXRD [93]. EDXRD typically employs silicon, germanium or cadmium zinc telluride (CZT) detectors. Silicon detectors demonstrate the best energy resolution (~ 150 eV) for soft X-rays but are inappropriate for high energies (> 20 keV) [98]. Germanium and CZT demonstrate better absorbance at increasing energies [99, 100] and offer energy resolutions of 400 eV [101] and 2 keV [102] at ~ 60 keV (i.e tungsten $K\alpha$), respectively. Although the latest research suggests that CZT can now operate at 1.09 keV (with a standard deviation of 0.46 keV) [103]. Assuming a fixed diffraction cell of 12° [20] (which is on the larger side of angles that are reported) then the equivalent angular resolutions can be calculated at the appropriate wavelengths for each detector. Silicon equates to $\sim 0.23^\circ$ angular resolution (at ~ 8 keV, Cu $K\alpha$). Germanium would yield $\sim 0.08^\circ$ angular resolution at ~ 60 keV. CZT would provide an equivalent angular resolution of $\sim 0.22^\circ$ at ~ 60 keV. Typical ADXRD systems achieve angular resolutions of 0.02° , a 4-10 fold gain in equivalent resolution. Another issue is beam hardening. Attenuation through the sample produces preferentially absorption of lower energy X-rays [101], which can make identifying materials based on the energies of the scattered X-rays problematic. With this in mind it may be worth revisiting ADXRD as a means of providing phase identification.

2.5 Elemental / Phase Specific X-ray Imaging techniques

Section 1.4 states that some complicated applications requiring the inspection of a multitude of materials may benefit from a single imaging modality capable of collecting and interpreting multiple material characteristics simultaneously. The manner with which X-rays have been employed to identify material's individual characteristics (i.e morphology, elemental composition and phase) has been discussed in sections 2.3 and 2.4. The following section provides an overview of the techniques that have been founded on the basis of combining one or more of these modalities. This section concludes with a summary as to their success, the gaps in knowledge and the thesis aims and objectives.

2.5.1 Dual Energy X-ray Imaging

Dual energy X-ray imaging provides a method of representing effective atomic number (i.e elemental composition) and morphology simultaneously. An example image is illustrated in figure 2.12. This technique has also been combined with kinetic depth effect X-ray imaging [104] (section 2.3.4) to provide a much needed 3D imaging counterpart. This technique can be implemented in a similar manner to the transmission geometry illustrated earlier (figure 2.4) except, importantly, two images are obtained simultaneously from different energy (E) X-ray beams. These

A collage artwork titled "The Machine" by Robert Rauschenberg. The composition features a variety of mechanical and electronic components layered over a textured, orange-toned background. A large, dark wrench is positioned diagonally across the upper left. To its right is a green circuit board with several black integrated circuits. Below the wrench, a bundle of blue wires is coiled. In the lower left, there are two green circular components, possibly capacitors or small speakers. The overall style is abstract and layered, with a focus on industrial and technological motifs. The background has a mottled, painterly quality with shades of orange and yellow.

However instead of the atomic number, the effective atomic number is reported. The “effective atomic number is an estimate of the hypothetical single crystal element that will give the same X-ray attenuation as the substance being evaluated” [37]. In actuality if >1 materials were overlaid with contrasting atomic numbers (i.e. high

and low Z) along the interrogating beam path, then the resulting effective atomic number would be somewhere in between (and therefore not representative of either material). For this reason dual energy imaging qualitatively categorizes materials into one of three categories (namely organic, inorganic and metallic depicted in orange, green and blue, respectively). Organic indicates the sample is comprised of $Z_{ave} \leq 10$, inorganic is $10 < Z < 20$ and metallic is $Z \geq 20$ [104]. For this reason dual energy imaging is ill equipped to provide material specific information (arguably worse than X-ray fluorescence as it qualitatively groups adjacent atomic numbers, described in section 2.4.1) as it is the molecular arrangement of the atoms and not the individual elements atomic number (Z) that is characteristic of the material [43].

2.5.2 Diffraction Enhanced Imaging

Diffraction enhanced imaging (DEI) is somewhat of a misnomer. This is because the image enhancement is brought about via the removal of both the incoherent and coherent (described in sections 2.2.2 and 2.2.3, respectively) attenuation contributions. The intensity of X-rays that are transmitted through an object can be described by equation 2.6 following Chapman et al. [14] where the total recorded intensity (I_N) is the sum of the refracted intensity (I_R), diffracted intensity (I_D), coherently scattered intensity (I_C) and the incoherently scattered intensity (I_I) [14].

$$I_N = I_R + I_D + I_C + I_I \quad (2.6)$$

Although X-ray scattering has been shown to be useful (described further in section 2.4) these scattering contributions (i.e. I_D , I_C and I_I) are detrimental to the transmission imagery as they result in a loss of spatial resolution and contrast. DEI utilises a highly monochromised X-ray source (typically generated by a synchrotron, section 2.1.4) that is incident on the sample. An analyser crystal with a narrow acceptance angle (i.e. micro radians) is then used to selectively diffract X-rays onto a detector. Only X-rays that conform to the acceptance angle of the analyser crystal will contribute to the resulting image (i.e. those that deviate very little from their original path). X-rays that have deviated from their original path by milliradians owed to scattering are rejected [14].

The contrast in the resulting image is the result of refraction and extinction [105]. The benefit is that greater contrast can be obtained between objects of a similar mass attenuation coefficient (μ/ρ) that cannot be readily differentiated by conventional imaging systems [106] (including all those discussed in section 2.3) and has found favor in medicine [107, 108]. Recent studies have also involved DEI being combined with CT [109].

2.5.3 Phase Contrast X-ray Imaging

Phase contrast X-ray imaging offers a method of displaying supplementary information regarding a samples refractive index within transmission X-ray imagery negating the problem of poor image contrast from materials with weak absorption contrasts

[110, 111]. This technique is therefore similar to DEI (described in section 2.5.2) but is achieved using quite different mechanisms. Phase contrast X-ray imaging involves the use of a highly coherent X-ray source (which can be either monochromatic or polychromatic). When the X-ray beam wavefront passes through objects of differing refractive indices, some of these coherent waves can suffer a small degree of phase retardation. If a detector is placed close to the object (to collect a transmission image) this small degree of phase retardation is unnoticed. However moving the detector further from the object accentuates this effect and the waves interfere with one another. In practice these interference effects appear in the image at the edge of objects, where there is a discrepancy between the refractive indices. Similarly to DEI this results in contrast between objects (of similar mass attenuation coefficient (μ/ρ)) [112] that cannot be identified by conventional imaging modalities (e.g. CT). Recent developments have seen phase contrast imaging combined with dual energy X-ray imaging [113, 114], stereoscopy [115] as well as CT and fluorescence [116] (discussed in sections 2.4.1 and 2.3.2).

2.5.4 X-ray Diffraction Imaging

Recently techniques have been developed to provide X-ray diffraction imaging of samples whereby characteristic patterns are obtained and attributed to spatially separated loci. This has been performed in two and three dimensions using angular and energy-dispersive approaches (the later of which have already been compared in

this document, section 2.4.2). The angular and energy dispersive imaging approaches are discussed separately in the following sections.

Energy-Dispersive

The manner with which energy dispersive X-ray diffraction (EDXRD) patterns are obtained is discussed in section 2.4.2. However this technique has also been applied to imaging. Energy dispersive X-ray diffraction tomography [82, 117] permits scatter signatures to be attributed to a voxel (locus) within a sample. The locus within the sample is defined by the intersection of a highly collimated set of incident and diffraction rays. Developments in diffraction collimation have permitted several loci in depth to be evaluated simultaneously [49]. The hypothesised geometrical configuration for obtaining some 10^4 voxels (each of which contain a characteristic spectra for a locus within the sample) has also been proposed by Harding [85] but has yet to be realised. The most advanced EDXRD imaging system currently in operation is perhaps the rapid tomographic energy dispersive diffraction imaging (rTEDDI) system [118]. This system (a development of the TEDDI system [33]) permits simultaneous (i.e spatially separated) measurements to be collected. The sample is illuminated by white radiation and a laser cut array of collimator's (each with a fixed angle) ensure that individual pixels on a 2D array are exposed to scattered radiation from a discrete sampling volume [96, 15]. The scattered radiation is then independently energy resolved at each pixel producing an array of diffraction

patterns obtained from differing sample loci . This technology serves as a benchmark for future developments and has lead to the production of compelling imagery colour encoded with material's identification [96].

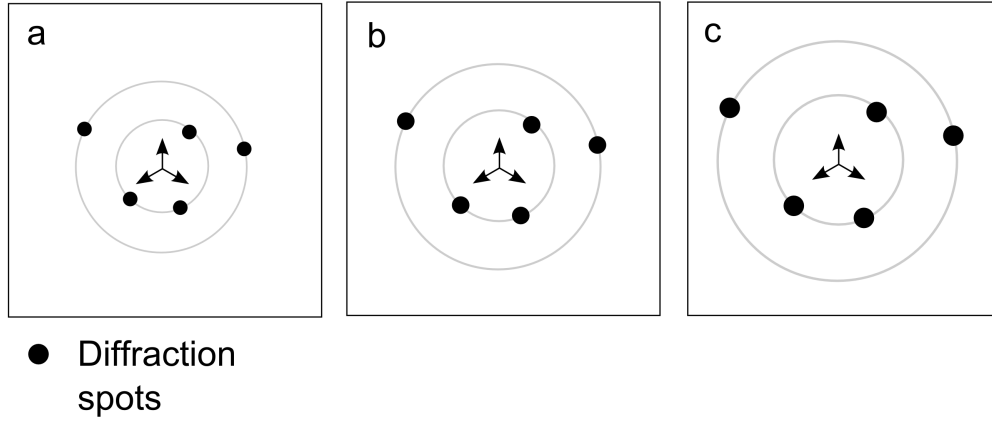
Angular-Dispersive

Perhaps the most novel imaging technique utilising an angular-dispersive X-ray diffraction approach (although there are others [119, 120]) is the 3DXRD microscope [8] which is typically used to map grain boundaries statically [121] or dynamically [122, 123, 124].

This approach involves illuminating a thin line (e.g. $0.5\text{ }\mu\text{m}$ by 0.8 mm) through a small sample (e.g. cylindrical specimen 3 mm in height and radius of 0.4 mm [121]) with a highly monochromised beam. Should any of the grains within the sample be at the correct orientation to fulfill the Bragg condition then it will results in a spot of radiation being diffracted onto a 2D detector. Rotating the sample ensures that all grains fulfill the Bragg condition at one orientation. For each sample orientation the detector is also translated to increasing sample to detector distances along the primary beam path. This has been illustrated in figure 2.13 where detectors a, b and c represent three diffraction images obtained at increasing sample to detector distances.

Any diffraction spots present in the first image (a) will move radially outwards

Figure 2.13: *Diagrammatic representation of diffraction spots resulting from the irradiation of individual crystals collected at increasing sample to detector distances (a, b and c) similarly to that employed for 3DXRD (after Nielsen et al. [8]).*



when the detector distance is increased. This direction of radial movement has been indicated in figure 2.13 by the three arrows. The amount that the diffraction spots move per frame and the detector positions are linearly related and are used to calculate both 2θ and the locus of the grain within the sample. This permits highly detailed 3D imagery to be created such as those presented in Ludwig et al. [125].

2.5.5 Summary

Some applications (described in section 1.2) require the ability to interrogate and examine sample volumes that contain complicated items varying in structural morphology and material composition. The X-ray region has been shown to be an effective probe for evaluating these characteristics and has given rise to a host of analytical techniques.

X-ray imagery (afforded because of the differing attenuation coefficients in matter) has progressed significantly (section 2.3) from the traditional 2D static transmission imagery (section 2.3.1) into exciting 3D systems that negate super positioning. These systems provide better morphological visualisation and recognition but come at cost. Kinetic depth effect X-ray (KDEX) imaging (2.3.4) has been shown to be extremely valuable because it offers a compromise between stereoscopy (section 2.3.3) and computerised tomography (section 2.3.2) in terms of 3D effect and hypothesised cost.

This technique alone offers very limited information regarding material composition. Many promising techniques including dual-energy, X-ray fluorescence, diffraction enhanced imaging and phase contrast imaging have been combined with X-ray imaging to provide a better description of sample materials. For the most part the accompaniment of these techniques have led to the development of important tools and compelling imagery, each of which have their own niche. However the X-ray diffraction field is arguably the most descriptive technique reviewed in this document. This is because refractive index and elemental composition (offered by the aforementioned techniques) cannot be used to unequivocally identify materials based on their molecular structure (i.e. phase).

X-ray diffraction tomography (section 2.5.4) is prolific and differing modalities utilising energy and angular dispersive approaches have been discussed. The energy-dispersive approach (favored by many authors) offers speed of data acquisition

yet there are numerous reasons (described in section 2.4.2) that suggest angular-dispersive X-ray diffraction (ADXRD) may be worth revisiting. Current tomographic ADXRD systems such as the 3DXRD system (section 2.5.4) demonstrate great promise but require the use of synchrotron radiation (section 2.1.4) which is not practical for all applications. It also only evaluates small sample volumes (e.g. 1 mm^3) that give rise to diffraction spots rather than Debye cones that may be anticipated from truly polycrystalline materials.

2.6 Aims and Objectives

The aim of this study is to combine kinetic depth effect X-ray (KDEX) imagery with angular dispersive X-ray diffraction (ADXRD). This technique fusion should yield synergy, facilitating the simultaneous interrogation of materials that are morphologically and/or molecularly distinct. Attention should be paid to the speed of data acquisition as this is one of the main shortcomings of ADXRD. Ideally the technique should also be realised via the use of a traditional X-ray tube as it increases practicality. The individual thesis objectives are listed below:

- Devise a method or methods of obtaining and interpreting angular dispersive X-ray diffraction patterns from volumes.
- Simulate the methods response to factors such as sample thickness and appropriately critique the result.

- Demonstrate proof of principle within the laboratory.
- Simultaneously interrogate a sample via KDEX and angular-dispersive X-ray diffraction.
- Produce image sequences that demonstrate combined X-ray diffraction and kinetic depth effect imaging.

Chapter 3

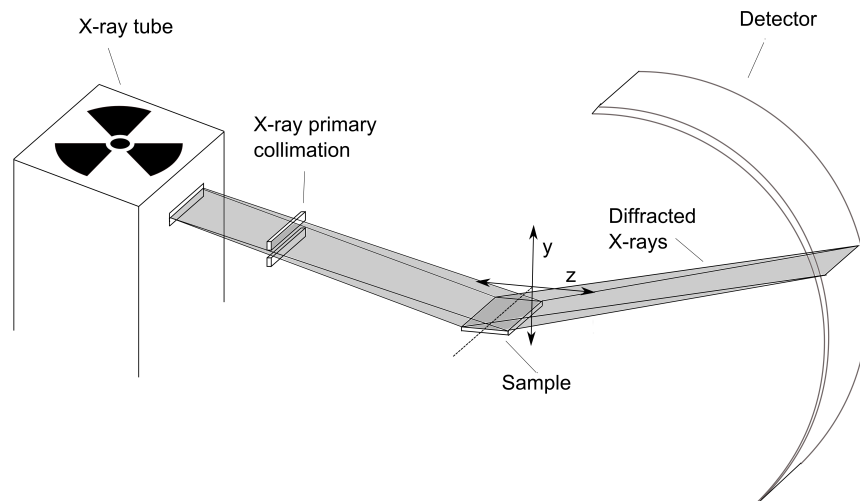
Method

The principles of X-ray diffraction have already been discussed in chapter 2. This chapter introduces the reader to obtaining diffraction patterns. The complications associated with collecting and interpreting diffraction patterns from volumes is explained. Different scatter sensor geometries and methods are hypothesised. Some permutations are simulated and tested. A method of obtaining transmission X-ray imagery is presented. Finally a rationale for colour encoding transmission imagery with diffraction data is proposed.

3.1 Geometry of the Conventional Powder Diffractometer

The principles governing powder X-ray diffraction have been discussed in section 2.4.2. This section discusses how angular dispersive X-ray diffraction (ADXRD) patterns are obtained in practice. Figure 3.1 depicts the conventional Bragg-Brantano ADXRD geometry. Primary X-rays irradiate the sample (held in reflection) and the coherently scattered X-rays are emitted at characteristic angles. A typical powder X-ray diffractometer illustrating this same geometry is illustrated in figure 3.2.

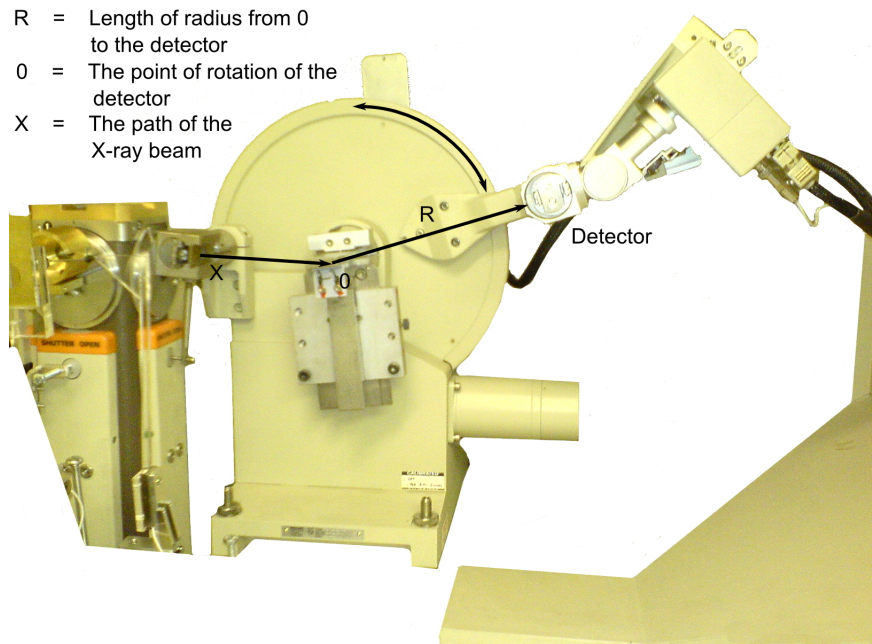
Figure 3.1: *A depiction of an angular dispersive X-ray diffraction geometry. A highly collimated X-ray beam is incident on a polycrystalline sample. Diffracted rays are then collected by a point detector which moves at constant radius to the sample. The detector illustrated in this figure is a superposition of such a detector.*



The monochromatic X-rays are emitted from the X-ray housing (X) that are collimated so they strike the sample situated at θ . A scintillator detector attached to

a mechanical arm is moved with constant radius (R) to the sample collecting the scattered radiation at differing angles. These characteristic angles and the wavelength of the monochromatic radiation can be used to calculate the d-spacings of the material by rearranging Bragg's law (2.4). These d-spacings are compared to a database of standards to provide a phase (material) identification.

Figure 3.2: *An image of a typical powder X-ray diffractometer. A highly collimated X-ray beam (X) is incident on a polycrystalline sample situated at 0 . Diffracted rays are then collected by a point detector which moves at constant radius (R) to the sample.*



3.2 Sample Position

In the geometry presented in section 3.1 (figure 3.1) the correct positioning of the sample is paramount in identifying the sample. If the sample is displaced from the rotation axis y then a subsequent change in diffraction angle and hence d-spacing

will be observed. This instance provides problems that are two fold:

- The unknown material cannot be unequivocally identified as the observed d-spacings are incorrect.
- The locus of the scattering material is unknown.

In practice it is not possible to position the sample exactly on this axis because the sample has a finite thickness. However if the sample is displaced from the rotation center on the y-axis by an amount s , then the subsequent change in scattering angle can be estimated from the well known displacement error equation 3.1 following Jenkins and Snyder [79]

$$\Delta 2\theta = -\frac{2s \cos \theta}{R} \quad (3.1)$$

The geometries presented thus far show reflection geometries. If the sample were irradiated in transmission then an alternate equation would be necessary to account for sample displacement in the z-axis.

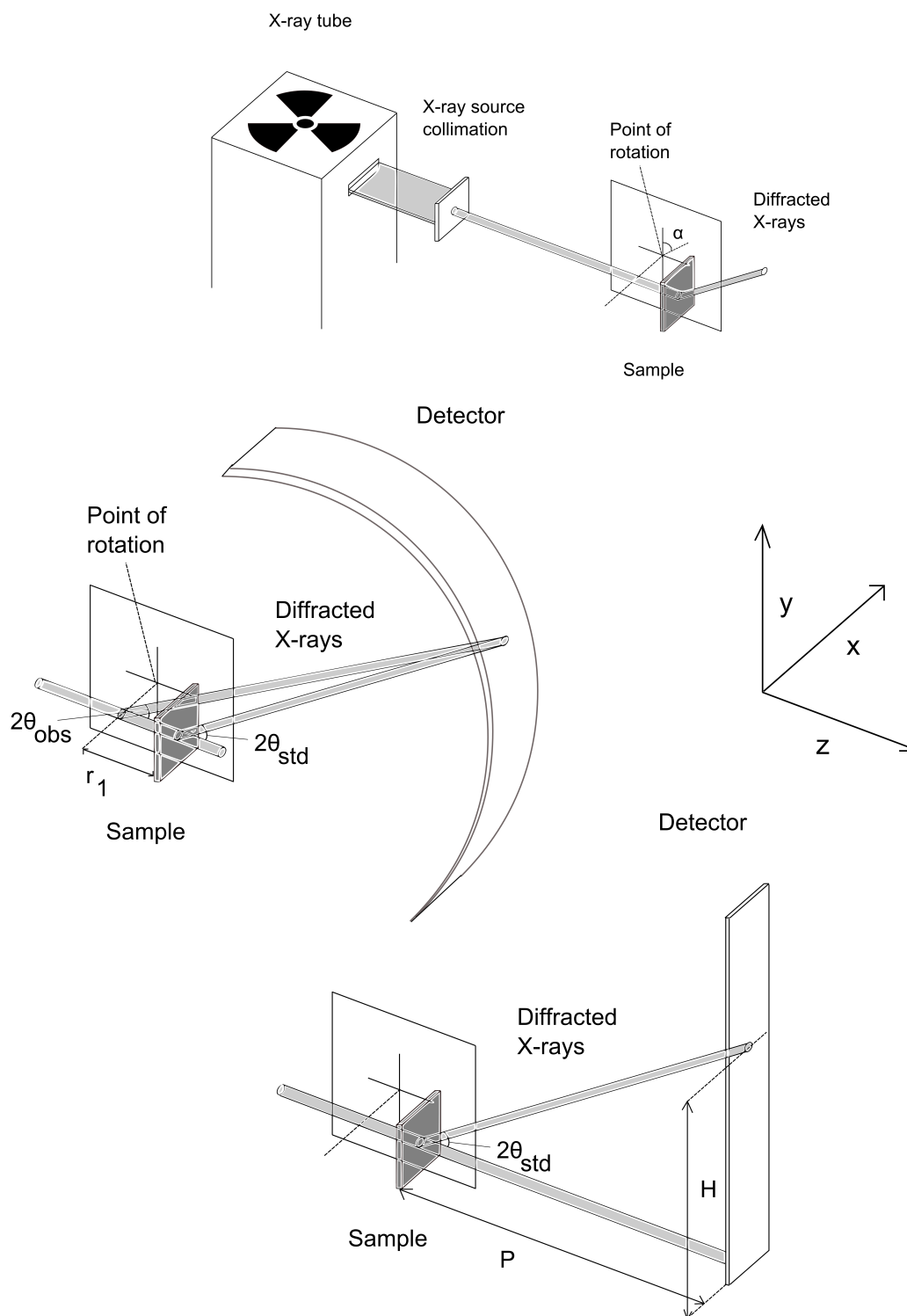
The purpose of this research is to utilise the material discriminating capabilities of X-ray diffraction to augment kinetic depth effect X-ray (KDEX). KDEX is designed to provide transmission X-ray images of inspection volumes (e.g. baggage, described in section 2.3.4). It is likely that the sample locus of an unknown diffraction material (within the inspection volume on the z-axis) will not be known. Therefore,

before KDEX and angular dispersive X-ray diffraction was combined this issue was investigated in transmission mode. The following problems were investigated:

- Identifying an unknown material from diffraction patterns originating from an unknown locus along the primary X-ray beam path.
- Identifying the subsequent locus of the now known diffracting material.
- Resolving patterns originating from multiple unknown materials at unknown locations along the primary X-ray beam path.

The following sections explore the use of differing detector geometries to resolve these issues. Circumferential detector configurations (section 3.3) akin to traditional diffraction geometries as well as linear detector configurations (section 3.4) more likened to transmission imaging systems are explored. All of the proposed configurations consider the use of “open collimation”. The primary incident beam is tightly collimated to a pencil beam and no collimation is placed in front of the scatter sensors. This permits diffraction signals from any locus on the primary beam path (z-axis) to reach the detector. The use of “open collimation” is validated in section 4.1. A master diagram (figure 3.3) has also been included at this point to illustrate the axes and nomenclature for various geometries used in the following sections.

Figure 3.3: A master image illustrating the geometry and nomenclature of various configurations utilised in this study. These include the circumferential geometry (section 3.3), linear geometry (section 3.4) and kinetic depth effect X-ray geometry (3.6).



3.3 Circumferential Detector Configuration

Circumferential detector configurations are traditionally used in powder diffraction experiments. For this reason circumferential detector configurations were explored to resolve the sample position issues (highlighted in section 3.2) as it required little equipment engineering. The method outlined in this section is repeated elsewhere [126].

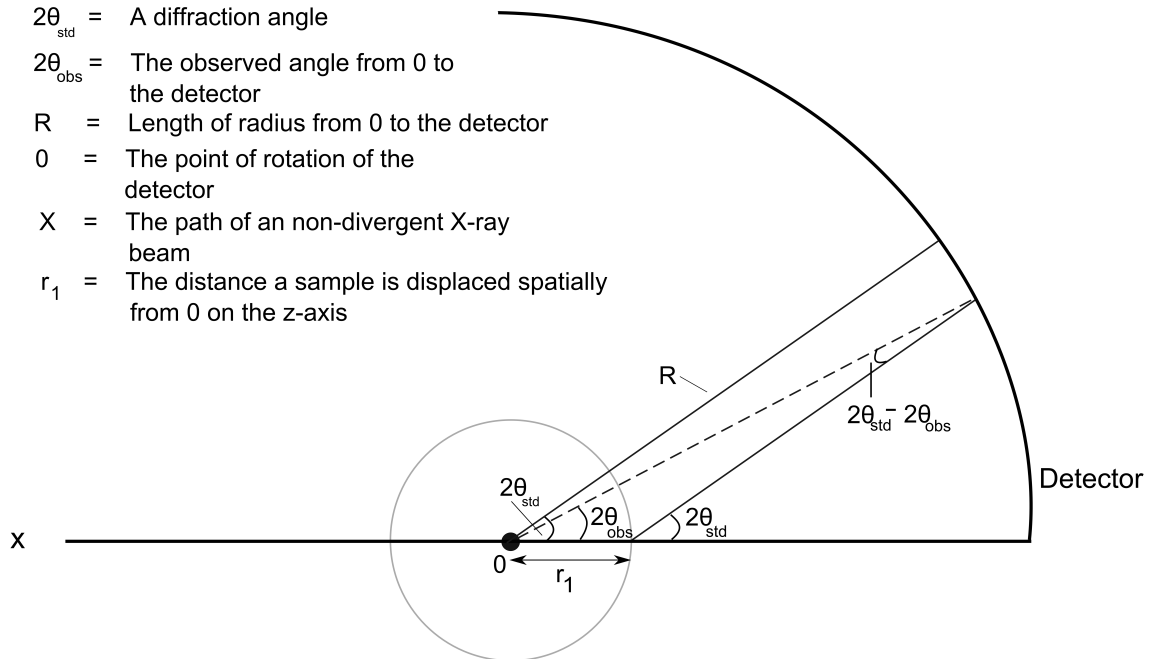
3.3.1 Identifying Displaced Patterns

To identify an unknown diffracting material at an unknown locus with a circumferential detector configuration (i.e similarly to that illustrated in figure 3.2) the impact that moving (or displacing) the material from the z-axis has on the resulting diffraction pattern was first investigated. Figure 3.4 illustrates the relationship between the material diffraction angle ($2\theta_{std}$), the measured or observed angle ($2\theta_{obs}$) and the displacement (r_1). Equation 3.2 was derived to transform diffraction patterns to how they would appear if they were displaced by distance r_1 along the z-axis. A systematic approach that used this equation to identify the material was investigated. A library of known diffraction patterns (illustrated in table 3.1) were systematically transformed to how they would appear at a series of calculated loci (r_1). These were individually compared to the unknown pattern. If any of these transformed diffraction patterns matched the unknown (i.e. have concordant 2θ values) then the

material and its calculated location were reported. In this study a relatively small number (~ 15) of diffractograms were used. It should be noted that the number of known diffraction patterns that need to be transformed will impinge on the analysis time. This is discussed further in section 5.

$$2\theta_{obs} = 2\theta_{std} - \sin^{-1} \left[\frac{r_1 \sin(180 - 2\theta_{std})}{R} \right] \quad (3.2)$$

Figure 3.4: *The geometry of the X-ray diffractometer when a sample is displaced from the rotation axis by distance r_1 . Displacing the sample from the rotation axis toward the detector results in a downward shift in the observed angle of diffraction making materials identification problematic.*



3.3.2 Resolving Multiple Patterns

One of the assumptions made in section 3.3.1 when identifying the displaced patterns is that all of the diffracted signals share the same locus on the primary beam path. In practice this is unlikely. The unknown pattern may have a contribution of Bragg maxima from multiple unknown materials that are spatially separated on the primary beam path. For this reason resolving multiple unknown material's diffraction patterns was considered.

Consider a diffractogram that contains Bragg maxima from ≥ 2 materials that are situated at different loci on the primary beam path. In order to identify these material's, their respective Bragg maxima need to be separated according to their loci. Each set of maxima (from differing loci) can then be individually subject to the systematic approach outlined in section 3.3.1. This differentiation cannot be achieved from a single pattern because there is no inherent position information. To resolve this issue a new variable was introduced that involved rotating the inspection volume. Providing the primary beam path and the detector remain stationary, then altering the orientation of the inspection volume through a rotation (by an amount α) provides an alternate "perspective". This is illustrated in figure 3.5. The locus of the material (i.e r_1) on the primary beam path (i.e. z-axis) is altered for different perspectives. This alteration is linked to the original location. For this reason materials at different loci are altered by different amounts when the inspection volume is rotated. This relationship is governed by equation 3.3 whereby

r_1 is the original displacement, r_2 is the new displacement and α is the angular separation between each of the perspectives diffractograms.

$$r_2 = \frac{r_1}{\cos \alpha} \quad (3.3)$$

Providing multiple diffraction patterns are obtained from a series of rotations (α) of the inspection volume then the resulting diffraction patterns will be comprised of Bragg maxima that modify their positions ($2\theta_{obs}$) by different amounts. The Bragg maxima modify their position because of two factors.

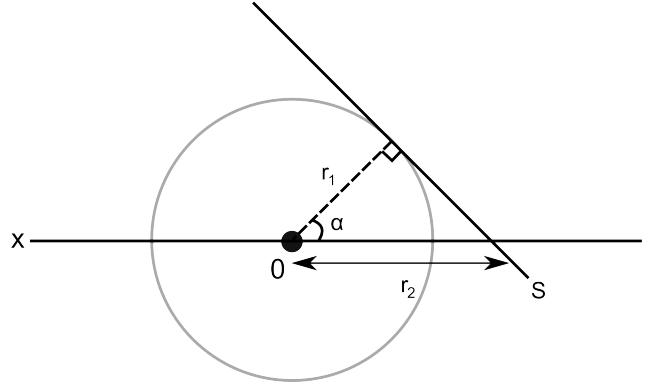
- The change in location (r_1 being modified to r_2) of each material, largely dependent on the original location (r_1)
- The diffraction angle ($2\theta_{std}$) of each Bragg maxima.

If the change from $2\theta_{std}$ can be determined then any additional Bragg maxima position modification can be attributed to the diffracting materials locus on the primary beam path. For an unknown material, $2\theta_{std}$ is not known. Therefore an approximation method was derived which does not require $2\theta_{std}$. For the purposes of this explanation it was assumed that the same Bragg maxima can be identified (or tracked) through differing perspective diffractograms (α). $2\theta_{obs2}$ represents the same observed Bragg maxima as $2\theta_{obs}$ accept its locus was modified by a rotation

of α . Providing $2\theta_{std}$ is small (i.e less than 30°), the small angle approximation for sine can be used to derive equation 3.4. The Bragg maxima position modification with α is no longer dependent on $2\theta_{std}$ and instead was used to infer the origin of each Bragg maxima (r_1). A full derivation for equation 3.4 can be found in section 8.2. Equation 3.4 can be rearranged further to determine the sample position (r_1) analytical, illustrated by 3.5 (a full derivation of which is given in section 8.2). Both an empirical and analytical approach were investigated to resolve multiple pattern diffractograms obtained with this configuration. These are described further in section 3.5 and demonstrated in section 4.2.

Figure 3.5: *The geometry of a sample arranged normal to the X-ray beam path after being subject to a rotation of an amount α . If both r_1 and α are positive such as the example depicted then sample is effectively displaced toward the detector resulting in a downward shift in the observed angle of diffraction.*

- S = An infinitely thin sample rotated by α about 0
- 0 = The point of rotation of the detector
- X = The path of a non-divergent X-ray beam
- r_1 = The distance a sample is displaced spatially from 0 on the z-axis
- r_2 = The new displacement position on the z-axis after rotation α
- α = Degree of rotation about the point of rotation



$$\frac{2\theta_{obs2}}{2\theta_{obs}} = \frac{R - \frac{r_1}{\cos \alpha}}{R - r_1} \quad (3.4)$$

$$r_1 = \frac{R - R \frac{2\theta_{obs2}}{2\theta_{obs}}}{\frac{1}{\cos \alpha} - \frac{2\theta_{obs2}}{2\theta_{obs}}} \quad (3.5)$$

3.4 Linear Detector Configuration

Transmission X-ray imaging systems conventionally employ linear or planar detectors. A similar configuration was investigated that could collect and interpret scatter signatures. Please note that for the practical application of this method diffraction patterns were collected on a 2D area detector and radially integrated to produce a conventional 1D pattern (described in greater detail in section 3.6) . However, importantly the same result can be achieved with a linear detector. Further information regarding the linear detector configuration can be found in Dicken et al. [126].

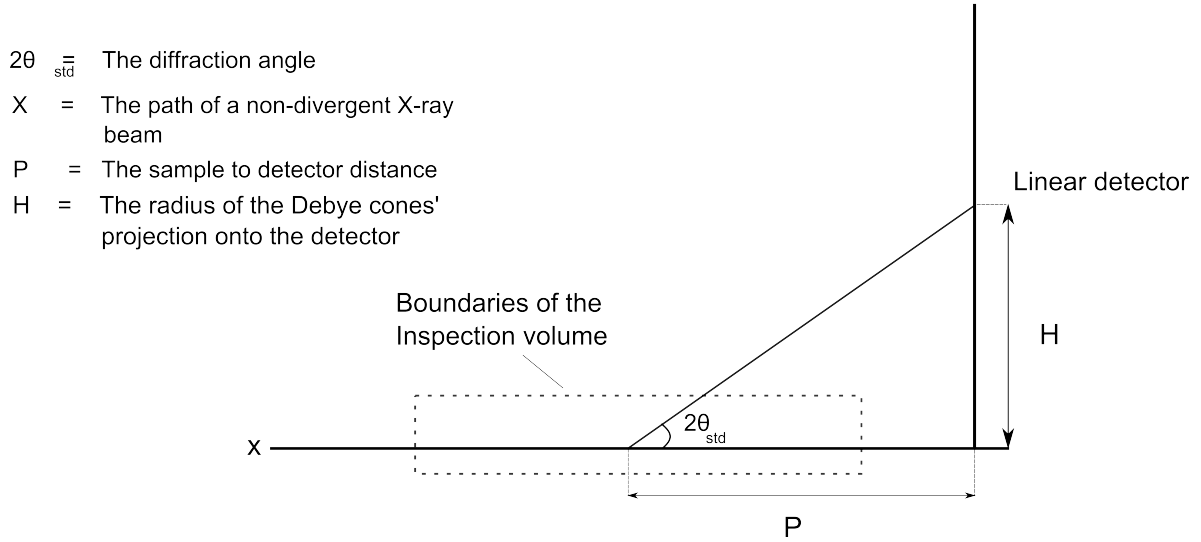
The sample locus on the primary beam path is critical for providing materials identification with a planar detector (illustrated in figure 3.6). If the sample to detector distance (P) is known then $2\theta_{std} = \tan^{-1}(H/P)$ where H is the radius of the Debye cones' projection onto the detector. However because the sample locus cannot be determined from a single diffraction pattern similarly to section 3.3.2 a new (yet different) variable was introduced. Consider the diffraction pattern resulting from the geometry illustrated in figure 3.6 whereby the planar detector is arranged normal to the incident beam. Translating the detector along the primary beam path (z-axis)

away from the source and sample (and hence increasing P) will result in the Debye cones' projection striking higher on the detector (resulting in a subsequent increase in H). The angle of scatter ($2\theta_{std}$) can then be calculated by equation 3.6.

$$\frac{dH}{dP} = \tan 2\theta_{std} \quad (3.6)$$

The sample to detector distance was calculated by $P = H/\tan(2\theta_{std})$. The sample locus was expressed as a distance from the source rather than the detector (as the source is static). This principle enables both the sample locus and $2\theta_{std}$ to be solved simultaneously. Providing the same Bragg maxima's can be identified or tracked through differing diffractograms (collected on detectors at differing distances) then in principle only two diffractograms are necessary to solve the problems outlined in section 3.2. However due to unreliable coincidence information between Bragg maxima in differing diffractograms (discussed in more detail in section 5) this differentiation is not possible. To resolve this issue a method of "peak tracking" was developed which is discussed in the following section.

Figure 3.6: A diagram illustrating the linear detector configuration. The radius of the Debye a Debye cones' projection (H) onto the linear detector is directly a function of the sample to detector distance (P) as well as the diffraction angle ($2\theta_{std}$).



3.4.1 Peak Tracking

Peak tracking is a method that was developed to identify common Bragg maxima (i.e the same Bragg maxima) on differing detectors that are spatially separated along the primary beam path. Traditional features such as peak amplitude cannot be used to achieve this differentiation (discussed in section 5) so a new approach was developed. If multiple Bragg maxima (n) are measured at two detector positions (separated by ΔP) all possible $2\theta_{std}$ permutations and their loci can be calculated by cross-referencing every Bragg maxima position (H) on the first detector with every Bragg maxima position on a second detector. Boundary conditions such as $2\theta_{std} > 0$ restrict the number of permutations to range from $(n(n+1)/2)$ to n^2 . If the

number of Bragg maxima (n) is large the proportion of correct permutations (and hence represent the correct $2\theta_{std}$ values) to false will reduce.

To remove incorrect permutations, diffraction patterns were measured from additional (>2) detector positions. If a $2\theta_{std}$ permutation was correct then the location that its Debye cone intercepts additional detectors (H values) at known detector positions (P) can be anticipated. Only $2\theta_{std}$ permutations with a full compliment of H values at the correct locations at each detector position were accepted. Other boundary conditions were used to inspect the calculated $2\theta_{std}$ permutations. These are listed below for clarity:

- $2\theta_{std} \geq 0$
- A full compliment of H values (one per detector) at each of the detector positions, concordant with the angle of diffraction ($2\theta_{std}$) and point of intersection (sample locus on the primary beam path)
- The point of intersection (sample locus) must fall within the boundaries of the inspection volume
- The $2\theta_{std}$ value must not exceed 40° .

Four detector positions were used in this study, the rationale for which is demonstrated in section 4.3 and discussed in section 5. After the $2\theta_{std}$ permutations were refined the remaining values were grouped according to their locus on the primary

beam path. Bragg maxima calculated to a common locus were deemed to be from the same sample (although this sample could be mixed phase). The maxima were cross referenced against a library of diffraction patterns to provide materials identification. Some anomalous $2\theta_{std}$ permutations (and their loci) evaded the boundary conditions. A further inspection step involving object disparity is introduced later in section 3.8 and discussed in section 5.

3.5 Diffraction Simulations

The rationale for two different methods that detect and interpret diffraction patterns of unknown locus have been discussed in sections 3.3 and 3.4. To demonstrate these methods simulations were conducted in Matlab®. These simulations were used to assess the performance of each method, evaluate confounding factors such as sample elongation and aid in selecting configurations prior to experimental tests. Each simulation can be separated into three distinct phases. The generation of diffraction patterns, interpreting the diffraction patterns and finally assessing the accuracy of interpretation. These phases are described in the following sections.

3.5.1 Generating Diffraction Patterns

The first stage in the simulation procedure was to generate diffraction patterns that would be expected given a predefined experimental arrangement. The primary data was obtained from the International Centre for Diffraction Data (ICDD) cards (a list of which is given in table 3.1) that list the d-spacings and intensity of a number of materials. FWHM² were set at 0.5° . This primary data was placed within a material library (illustrated in figures 4.18, 4.19 and 4.20 as they would appear at molybdenum $K\alpha$). A selection of these materials were randomly assigned a locus on the primary X-ray beam path within a boundary representing an imaginary inspection volume. The boundaries of the inspection volume were different for the circumferential and linear detector simulations. These were imposed because of known equipment limitations. The diffraction patterns were then projected onto the detectors and linearly summed. Diffractograms were created for differing rotations in α (0 - 45° in 1° increments) for the circumferential simulations and differing detector distances (P) typically anywhere from 210-280 mm from the X-ray source for the linear simulations. It should be noted that an idealised detector was simulated with a resolution of $10\mu m$ ($\sim$ the same detector resolution as the PIXIS camera described later in section 3.6). Each projected pattern contributed equally to the resulting patterns at each α rotation /detector position. To achieve this each pattern was normalised (with respect to the highest intensity peak) and then linearly interpolated onto the $10\mu m$ resolution detector. No allowances for the material's scattering cross sections, re-absorption effects or preferred orientation were made as

these factors only affect Bragg maxima amplitude. Material identifications in this study were based entirely on d-spacing, the justifications for which are presented in section 5. The minimum detector length in the y-axis (illustrated in figure 3.3) was considered (which inevitably varied for simulations with different radiation energies), to optimize the simulation time. However in each case it was ensured that the detector was of a suitable size to detect all diffracted X-rays.

The impact that variables such as sample elongation (i.e. samples with thickness along the primary beam path) and radiation energy on Bragg maxima position in the resulting diffraction patterns were also examined. To simulate a material with thickness in the z-axis the material was assumed to be at a range of loci (e.g. 150 mm from the X-ray source on the z-axis, ± 1.5 mm). A projected pattern was created for the same material in 0.1 mm increments over the elongated distance. These patterns were then linearly interpolated on the $10\mu\text{m}$ resolution detector and the intensity values were normalised with respect to the highest intensity Bragg maxima. To simulate different radiation energies the primary data was converted into d-spacing using a rearrangement of Bragg's law (equation 2.4, section 2.4.2) and then angle by substituting the wavelength of radiation that was being simulated.

3.5.2 Interpreting Diffraction Patterns

After the patterns were generated two algorithms were designed to utilise the concepts outlined in sections 3.3 and 3.4 for the circumferential and linear detector

configurations, respectively. A Savitzky-Golay filter was applied to each of the 1D patterns (to maintain continuity with experimental patterns obtained later). The patterns were then normalised with respect to the highest intensity Bragg maxima and a modified first differential was applied to provide a robust peak finder in the presence of noise. A peak finder was written in Matlab®. The criteria required to satisfy a peak are represented by equations presented in section 8.3. Circumferential and linear detector algorithms were then applied to the patterns. Their method of operation were quite different and so will be described separately.

Circumferential Algorithm

Both an empirical and analytical approach were explored to resolve multiple material diffraction patterns. In each case the peak finding algorithm was used to identify the position of Bragg maxima ($2\theta_{obs}$) in the diffractograms. Firstly the Bragg maxima were separated according to their locus on the primary beam path. For the empirical approach, Bragg maxima positions ($2\theta_{obs}$) from $1 - 45^\circ$ in α were divided by their respective positions in the 0° α diffractogram (the rationale for which is demonstrated in Equation 3.4). This can be thought of as normalising the peak position modification with respect to $2\theta_{std}$. These normalised peak positions were then plotted against α (illustrated in figure 4.8) for each Bragg maxima. Bragg maxima that have the same locus adhere to the same characteristic curve (approximated by equation 3.4). To separate the Bragg maxima quantitatively each line was fit with

$y = ax^2 + bx + c$ rather than a cosine function. The Taylor series expansion of $\cos(x)$ (illustrated by equation 8.5, section 8.4) suggests that a 2nd order polynomial will provide a suitable linear fit.

A plot of the coefficients results in Bragg maxima with a common origin occupying the same region (illustrated in figure 4.9). The locus on the primary beam path (r_1) of each Bragg maxima was then predicted empirically from the second order coefficient of the $y = ax^2 + bx + c$ fit. An alternate r_1 prediction was derived analytically using only the extreme orientations (0 and $45^\circ\alpha$) based primarily on equation 3.5. A comparison of these approaches is illustrated in section 4.2 and discussed in section 5.

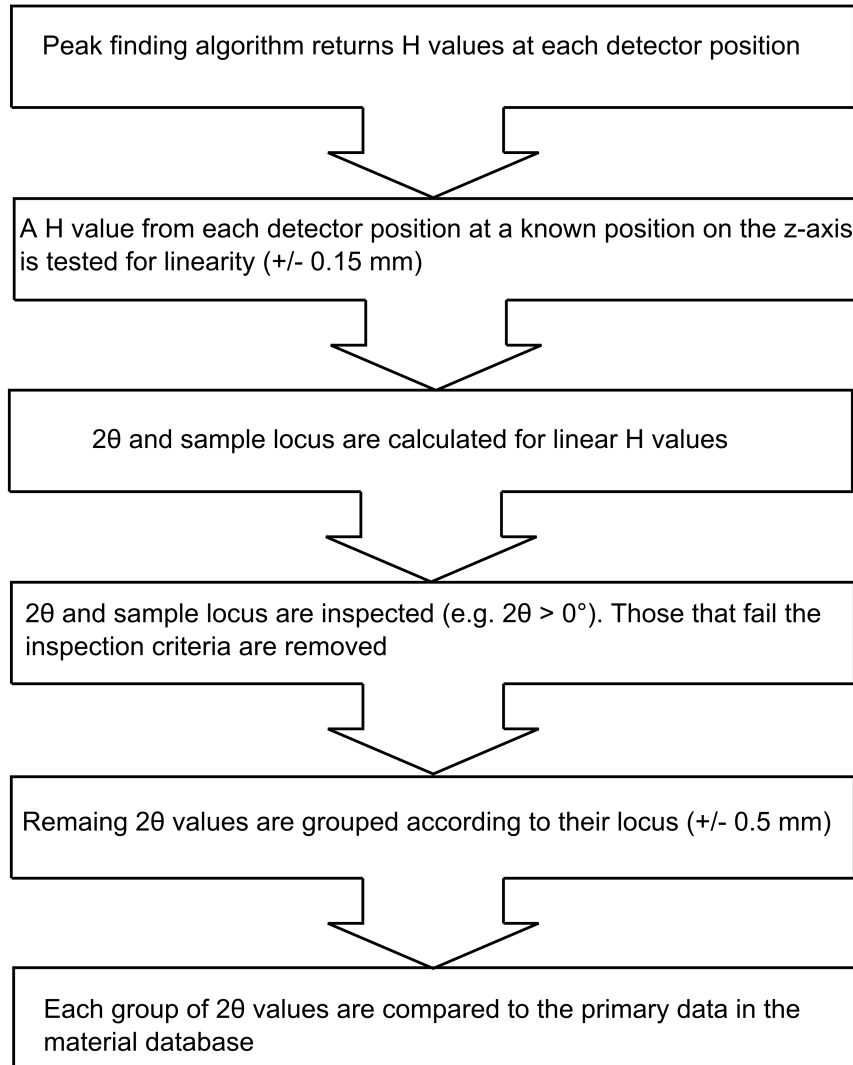
The next step was to identify the unknown materials. Each diffraction pattern in the material library was transformed using equation 3.2 to how it would appear at the predicted r_1 positions and cross referenced against the unknown sets of Bragg maxima (searching for concordant $2\theta_{obs}$ values). Material matches and their respective positions were reported. Match index's were also generated based on the amount of matching Bragg maxima.

Linear Algorithm

The peak finding algorithm was used to identify Bragg maxima positions (in this instance referred to as H values) at each detector position. One Bragg maxima

position (H value in mm) was chosen from each detector position and these were tested for linearity. A tolerance of ± 0.15 mm was applied to account for discrepancies in Bragg maxima position. This was repeated so that all permutations were considered. When straight lines were found (drawn between a single Bragg maxima position on each detector) the tangent of the gradient ($2\theta_{std}$) and the sample locus (calculated from the intercept) were reported in a match table. The match table was then inspected to remove permutations that do not match the boundary conditions listed in “Peak Tracking” (section 3.4.1). Increasing the number of detector positions decreases the number of permutations. This was evaluated empirically (illustrated in section 4.3) by tracking Bragg maxima permutations through differing numbers of detector positions (e.g. 2-6) and cross referencing them with the number with Bragg maxima (n) in the primary data. Four detector positions were then used in both the simulation and experimental aspects of the investigation.

Figure 3.7: A flow diagram that illustrates the manner with which the linear diffraction configuration is implemented to find coincident Bragg maxima from diffraction patterns obtained from increasing sample to detector distances, ultimately resulting in the identification of an unknown sample(s) and an associated position within an inspection volume .



To identify the material, the $2\theta_{std}$ values from the match table were grouped according to their sample locus (± 0.5 mm). These were cross referenced against the

primary data in the material library directly (the benefits over the circumferential method are discussed in section 5). The number of matching Bragg maxima were used to generate a match index (described in section 3.7). This was used in assessing the algorithm’s performance at a later stage. These steps have been summarised and are illustrated via a flow diagram in figure 3.7

3.5.3 Assessing Algorithm Performance

Each configuration was assessed by comparing the known data inputs required to generate the patterns (i.e. primary data and locus) with the $2\theta_{std}$ and loci matches generated by each simulation.

The linear method was subject to a more thorough examination than the circumferential method (the reasons for which are discussed in section 5). Taking the security screening example further, a simulation was created to assess the peak tracking method’s ability to identifying “threat” (illicit) diffraction patterns (indicative of “threat” or illicit materials) in the presence of benign diffraction patterns that could conceivably be found in a suitcase. It should be noted that all the materials considered were crystalline. Approximately 1000 multiple material diffraction patterns were created by randomly combining two materials’ diffraction patterns from the material library. This library contained both illicit (e.g. trinitrotoluene / TNT and pentaerythrite tetranitrate / PETN) and benign (e.g. nylon , sucrose) materials, the locations of which were randomly assigned in the inspection volume by random

number generator between 70 - 170 mm from the X-ray source on the primary beam path (z-axis). Table 3.1 illustrates the material identity of the diffraction patterns that were used (illustrated in figures 4.18, 4.19 and 4.20 at molybdenum $K\alpha$) as well as their empirical formulae and International Centre for Diffraction Data (ICDD) cards numbers .

Table 3.1: *Material identity, empirical formulae and International Centre for Diffraction Data (ICDD) card numbers for the diffraction patterns used in the simulations outlined in section 3.5.*

Material identity	Empirical formulae	ICDD card number
Calcite	CaCa_3	5-586
Sucrose	$\text{C}_{12}\text{H}_{22}\text{O}_{11}$	24-1977
Polystyrene	$(\text{C}_8\text{H}_8)_n(\text{C}_{23}\text{H}_{28}\text{O}_4)_n$	53-1879
Cobalt	Co	15-806
Iron	Fe	6-696
Nylon II	$(\text{C}_{11}\text{H}_{21}\text{NO})_n$	48-2146
Potassium Chlorate	KClO_3	44-1619
Pentaerythrite Tetranitrate (PETN)	$\text{C}_5\text{H}_8\text{NO}_3$	42-1771
Cyclotrimethylene- trinitramine (RDX)	$\text{C}_3\text{H}_6\text{N}_6\text{O}_6$	44-1619
Ammonium Nitrate	NH_4NO_3	47-866
L-Cocaine Hydrochloride	$\text{C}_{17}\text{H}_{21}\text{NO}_4 \text{ HCl}$	30-1629
2,4,6-Trinitrotoluene	$\text{C}_7\text{H}_5\text{N}_3\text{O}_6$	28-1945
Talc-2M (Talcum Powder)	$\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$	13-553
Native Cellulose	$(\text{C}_6\text{H}_{12}\text{O}_6)_x$	3-289

A binary recording system was used to record whether the primary data included an illicit material (1) or if both diffractograms were benign (0). Tracked Bragg maxima were then grouped according to their calculated locus (± 0.5 mm) and compared to a smaller database that only contained the illicit materials. The number of concordant ($\pm 0.05^\circ$) Bragg maxima (between the tracked Bragg maxima and a material in the

illicit material database) was used as the discriminating threshold. Another binary recording system then recorded either 1 if the number of concordant Bragg maxima positions between an illicit material diffractogram $\geq$ the discriminating threshold indicating the presence of a “threat” material or 0 if the number of concordant Bragg maxima positions $<$ the discriminating threshold indicating there was no threat.

This procedure was repeated ~ 1000 times for different mixtures of materials at different loci. The combination of these binary systems was used to ascertain a true positive (1:1), true negative (0:0), false positive (0:1) or a false negative (1:0). Sensitivity (true positive rate) and 1-specificity (false positive rate) were then calculated for a range of discriminating thresholds and plotted against each other to produce a receiver operator characteristic (ROC) curve [41]. The closer the area under the ROC was to 1 the better the system performance. If the area under the ROC curve is equal to 0.5 (out of 1) then the system is able to detect the presence of a threat material half the time (the no-discrimination line).

The number of matching Bragg maxima was an appropriate variable to use as a discriminating threshold. For instance when only 1 concordant Bragg maxima was sufficient to indicate the presence of a “threat”, the sensitivity of the system was high (e.g. 1) (i.e all simulations that included an illicit material in its primary data were identified as containing a threat material). However the specificity was very low (i.e 0.1) because it was likely that a benign diffraction patterns would share a single concordant Bragg maxima ($\pm 0.05^\circ$) with one of the illicit material diffractograms.

Alternatively when a larger number of concordant Bragg maxima was required (i.e 30) to indicate the presence of a “threat” material the sensitivity was lower yet the specificity and false alarm rate were also lower.

ROC simulations were also conducted for different detector distances (section 4.3.2) and different radiation energies (section 4.3.3) at differing degrees of elongation / sample thickness (described separately in section 4.3.1) to produce ROC surfaces. To quantitatively compare these variables both the areas under the ROC curves, (table 4.2) as well as the volumes of the surfaces have been evaluated. These may be found in section 4.3.3 and discussed in section 5.

3.6 Experimental Validation

It is important to verify the simulation results with empirical evidence obtained in the laboratory. The experimental parameters for the differing configurations are described in the following sections. In each scenario sample materials were placed at known locations so that the results could be verified later. The experimental parameters for the KDEX configuration are also given. A description of how KDEX and X-ray diffraction were combined is given later in section 3.7.

Circumferential Configuration

The diffraction patterns for the circumferential configuration were made on a Phillips PW1820 X-ray diffractometer (illustrated in figure 3.1) incorporating a sealed, fine focus X-ray tube with a molybdenum target. The accelerating voltage and current were 40 kV and 30 mA, respectively. The scattered photons were detected by a scintillator detector located on an arm that rotated about a central axis. Diffraction patterns were collected from $5 - 35^\circ$ in 0.02° increments and 5 second integration time. A second stage was used to rotate the sample about the same central axis independently (enabling differing perspectives to be obtained). The degree of rotation of this second stage was denoted α in this study. Different collimation configurations were explored (demonstrated in section 4.1 and discussed in section 5) after which the majority of diffraction patterns were collected with $1/6^\circ$ primary collimation and no detector collimation. This produced an X-ray footprint of ~ 0.4 mm in height on the y-axis (illustrated in figure 3.3) when the sample is located at 0.

Linear Configuration

X-rays were produced by the same generator (Philips PW1830) as the circumferential configuration and operated under equivalent conditions. A 4 mm thick brass plate with a 0.66 mm aperture was used to collimate the interrogating primary beam to a pencil beam. Instead of using a scintillator detector, an X-ray camera was placed in front of a different window. The X-ray camera was a PIXIS 1024x1024 16bit cooled

(-40°C) CCD with a $\text{Gd}_2\text{O}_2\text{S:Tb}$ phosphorus screen. The camera pixel size was $13\text{ }\mu\text{m}^2$ giving a total CCD area of $13312\text{ }\mu\text{m}^2$. A Thorlab stage was used to translate the camera vertically (or raster scan) to collect the scattered photons (on the y axis normal to the primary beam path). 5 images were collected from 13 mm to 65 mm from the primary beam path (on the y-axis) with a step size of 13 mm. Each image was exposed for 3 minutes. These 5 images were stitched together (1000 pixels from each image), an example of which is illustrated in figure 3.8a.

The Debye-Scherrer rings incident on the 2D detector were integrated into a conventional 1D diffraction pattern (illustrated in figure 3.8c) by an algorithm written in Matlab®. Pixel intensities were integrated across the perimeters of a series of circular arcs. These circular arcs had a continuum of radii ranging from the bottom to the top of the detector. A superposition of these circular arcs is illustrated in yellow in figure 3.8b. Please note that some portions of the Debye rings were missed during this integration process (toward the bottom of the detector) due to the finite opening angle of the circular arcs. However this did not detrimentally affect the final intensities of the afflicted Debye rings as these tend to be from the lower angle diffraction peaks that benefit from greater flux densities. Alternate software is available to integrate Debye rings from 2D detectors such as FIT2D [127] which also has the capacity to apply appropriate tangent geometry intensity corrections. However this requires a knowledge of the sample position and is therefore not appropriate for this study.

Once radially integrated a Savitzky-Golay filter was then applied to the 1D pattern to reduce noise. Additional stages were used to translate the camera along the primary beam path (typically from 210 mm to 240 mm from the X-ray source, although the actual detector distances have been stated in the necessary sections) so that diffraction patterns could be collected at increasing sample to detector distances (P). A background subtraction was applied to some diffraction patterns (such as those illustrated in figure 4.34) to remove the characteristic sloped background nearer the primary beam. This was achieved by acquiring raster scan images and a subsequent 1D diffraction pattern (similarly to figure 3.8) from a control area (where no sample was intersected). This “background” pattern was normalised (with respect to the highest intensity value) and then subtracted from the diffraction patterns.

KDEX configuration

X-rays were produced by the Philips PW1830 generator similarly to the aforementioned configurations except the accelerating voltage and current were 20 kV and 10 mA, respectively. A 0.95 mm aluminium plate was used to attenuate the primary beam further to limit the number of photons striking the X-ray camera when placed in transmission. This was a necessary precaution given the sensitivity of the camera. The same PIXIS camera for the linear configuration was used for this configuration. The camera was translated in x and y axes in 13 mm steps to obtain 4 images which were later stitched together (1000 pixels per image) to obtain a 26 mm^2 transmission

Figure 3.8: *Diagram illustrating the raster scanned area (a), a superposition of the region that was radially integrated (b) and the resulting diffraction pattern from the radially integrated area (c). Please note that materials demonstrating preferred orientation (i.e incomplete Debye rings) can result in Bragg maxima with diminished intensity by adopting this approach.*

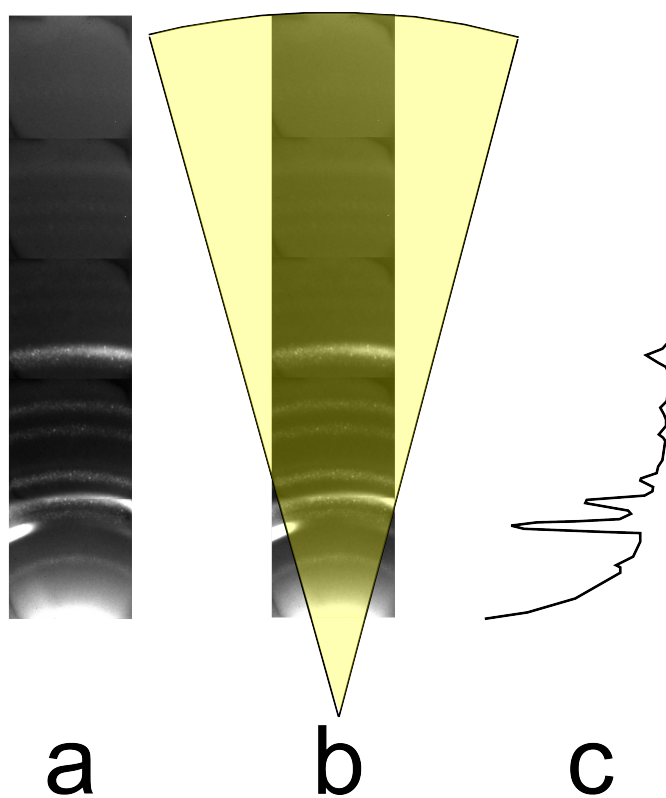
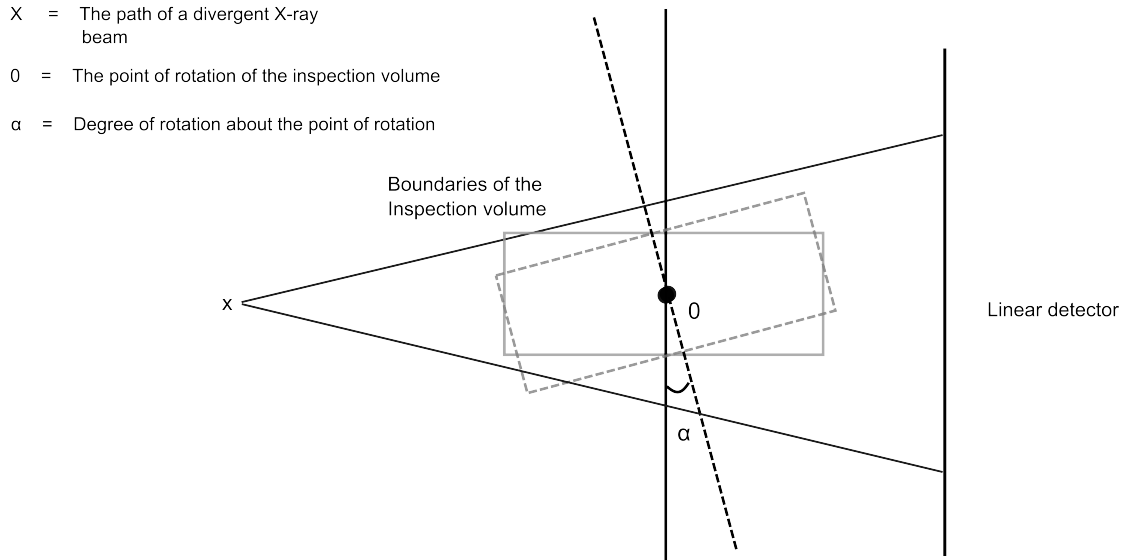


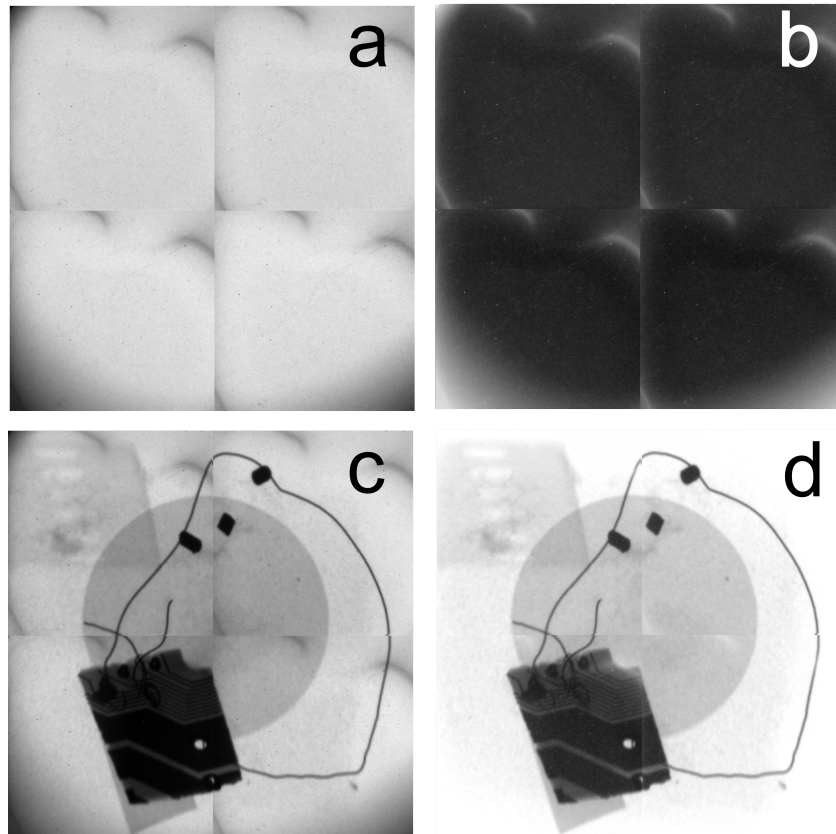
image (equating to a 2000x2000 pixel array, an example of which is illustrated in figure 3.9). Each of the 4 images were exposed for 5 seconds. A bright-field background subtraction was also applied to the rastered transmission image. This was performed by obtaining an equivalent rastered transmission image (i.e equivalent grid size and exposure time) with no sample in the primary beam path (illustrated in figure 3.9a). This background image was inverted (illustrated in figure 3.9b) and linearly added to the rastered transmission image. The result is illustrated in figure 3.9d.

Figure 3.10: *Schematic illustrating the kinetic depth effect X-ray (KDEX) configuration used in this study. Altering the orientation of inspection volume by an amount α results in a change in the transmission X-ray image seen at the linear detector.*



Multiple transmission images were obtained using this method after the inspection volume (or sample) was rotated through 1° increments from $+15^\circ$ to -15° in α whilst the X-ray source and detector remained stationary (illustrated in figure 3.10).

Figure 3.9: *Illustration of the necessary background correction steps that were applied to the transmission images as part of the KDEX configuration. Image a illustrates a flat field image with no sample present (non-uniformity in the intensity distribution can be attributed to the X-ray source and imperfections in the phosphorus). Image b illustrates the inverse of the flat field image. Image c illustrates the sample present within the X-ray beam. Image d illustrates a corrected image whereby image b and c have been linearly summed.*



This is equivalent to the traditional KDEX geometry where the inspection volume remains stationary and the source and detector are rotated to different perspectives (discussed more in section 2.3.4). Concatenated images of such an image sequence are illustrated in figure 4.32.

Materials

The relatively low energy X-rays (~ 17 keV) limited the test materials to those of low mass attenuation coefficients (μ/ρ). Early experiments utilised thin sheets of aluminium ($\simeq 0.2$ mm). This was an ideal candidate material because it has a low linear absorption coefficient (~ 13.1 cm $^{-1}$ at molybdenum $K\alpha$) and density (~ 2.69 g cm $^{-3}$). It is also easily extruded (which mitigated against complications expected from elongated/thick samples). It is self-supporting and its high crystal symmetry means its diffraction pattern gives relatively few Bragg maxima positions. Sintered aluminium oxide (Al₂O₃) plates ($\simeq 0.1$ mm) were also evaluated. Aluminium and aluminum oxide are difficult to differentiate by elemental techniques (as the emitted fluorescence radiation from light elements such as oxygen are prone to interaction by the surrounding atmosphere discussed in section 2.4.1) but their crystal structures and hence diffraction pattern are quite different. Later experiments where diffraction information was combined with transmission imagery (discussed in section 3.7) involved constructing phantoms (illustrated in figure 4.30) and obtaining diffraction patterns from cyclotetramethylenetetranitramine (HMX, a military grade high

explosive) in the presence of surrounding paraphernalia. This material is directly relevant to an airport screening environment.

3.7 Encoding Material Information within KDEX Imagery

The linear diffraction configuration (section 3.4) was chosen as the most appropriate method of incorporating material information into the KDEX imagery (the reasons for which are discussed in section 5). The interrogating pencil beam was produced by a 0.66 mm aperture through a 4 mm thick brass plate. This pencil beam was able to interrogate the entire sample in depth (i.e along the primary beam path on the z-axis) but its footprint in the x and y axes was limited ($\sim 0.34 \text{ mm}^2 + \text{beam divergence}$) in comparison to the size of the inspection volume. To comprehensively interrogate the sample translation stages were used to systematically move the inspection volume in the x and y dimensions (i.e. raster scan). The linear diffraction method was repeated for each interrogation site. These sites formed a grid and are illustrated in figure 3.11. The size of the grid and the frequency of interrogation was varied depending on the experiment (an example of which is illustrated in figure 4.33). This was conducted when the inspection volume was held at 0° in α (where α is the same variable illustrated in figure 3.10).

The tracked Bragg maxima for each interrogated site were compared to the material

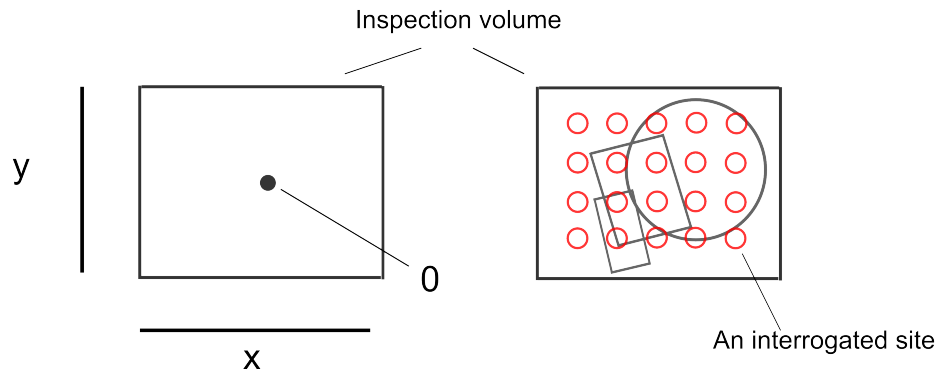
database discussed in section 3.5.3. The number of Bragg maxima (from a common locus on the primary beam path, ± 1.5 mm in this particular example) matching that of a material in the database was used as “an index of match certainty”. The interrogation sites giving rise to diffraction patterns from the material database were then colour encoded. The method of colour encoding is discussed later in this section. When the inspection volume was imaged at 0° in α the colour encoded area and the interrogation site shared the same coordinates. However when multiple images of the inspection volume are obtained for different rotations (typically $\pm 15^\circ$ in α for the KDEX configuration) the colour encoded areas (for these other images) need to be re-mapped in y . The position of the matching Bragg maxima on the z -axis was calculated by the peak tracking algorithm. This was given as a distance relative to the X-ray source but for the purposes of the re-mapping procedure was converted to a distance from the rotation axis (i.e 0 illustrated in figure 3.10) of the inspection volume. The variable r_1 was used similarly to section 3.3 as it denotes the same distance. y can be altered by Δy using equation 3.7

$$\Delta y = \cos \alpha (y - r_1 \tan \alpha) \quad (3.7)$$

where α represents the degree of rotation of the inspection volume, y represents the vertical position of the interrogating beam in the $0^\circ \alpha$ KDEX image and r_1 represents the location of the diffracting material along the primary beam path relative to the rotation axis. Relative distances within the image sequence (arbitrary) pixel values

Figure 3.11: *A diagram illustrating an example grid of pencil beam diffraction interrogation sites applied to an inspection volume*

- 0 = The point of rotation of the inspection volume
- x = Distance on the x axis relative to 0
- y = Distance on the y axis relative to 0



were calibrated using an internal standard (the ruler apparent in the top left hand corner of figure 4.30 and 4.33a-c).

To colour encode areas within the image sequence, the 16bit .tiff files were split into their RGB components. The pixel values in the red (R) channel denoted (rv) were then multiplied by a weighting factor based on the match index. This weighting factor was $rv + (rv * (\text{number of matching Bragg maxima} / 10))$ (i.e The original red pixel value plus the original red pixel value multiplied by the number of matching Bragg maxima for that region divided by 10). In doing so the intensity of red reflects the index of match. The result of this method is demonstrated in section 4.4.

3.8 Measuring Object Disparity

Although $2\theta_{std}$ permutations produced by the linear diffraction configuration were heavily inspected (outlined in section 3.6) by the boundary conditions it was still possible to get anomalous values and corresponding loci by chance (illustrated in figure 4.13). These anomalous values could produce false positives/alarms. KDEX image sequences contain positional information. This can be calculated by the rate of movement (disparity) of individual objects in the inspection volume as it is rotated. A more detailed description of disparity may be found in Tzovaras et al. [128] and Wei et al. [129]. The automatic tracking of objects in image sequences involves resolving the “correspondence problem”. This is a large field of research and is beyond the scope of this study, therefore objects were tracked manually (or by eye) rather than automatically in a well described phantom. The disparity of each object was then used to calculate their respective loci in depth on the primary X-ray beam path relative to the rotation axis.

This phantom (illustrated in figures 4.30 and 4.37) had numerous objects at known loci (r_1) within the volume. The change in position (Δy) was tracked through the KDEX image sequence of the same phantom and used to calculate r_1 in pixels. This value was converted to a distance in mm from the X-ray source using the internal standard (ruler apparent in the top right hand corner of figures 4.32). A comparison of the known loci and the calculated loci is demonstrated in section 4.5. A discussion as to its suitability for informing the diffraction information produced by the linear configuration is presented in section 5.

Chapter 4

Results

This chapter has two distinct sections. The first explores the hypothesised scatter sensor geometries outlined in chapter 3. The differing methods are validated by a combination of computer simulation and experiments in the laboratory. Factors such as elongated samples and radiation energy are explored. The second section demonstrates dynamic transmission X-ray imagery with material specific colour encoding. A back checking method using image disparity is also demonstrated.

4.1 Open Collimation

The circumferential and linear diffraction configurations are reliant on the use of “open collimation” (described in section 3.2), where diffraction signals can strike the

detector originating from any loci on the primary beam path. The source and detector collimator's were independently varied on the Phillips PW1820 X-ray diffractometer (illustrated in figure 3.1 and described in section 3.6) to provide justifications for such a geometry.

Source Collimation

The diffraction patterns obtained from a 0.2 mm thick sheet of aluminium obtained in transmission are illustrated in figure 4.1 where the largest aperture detector collimator (1.8°) which allowed for the greatest acceptance angle of scattered radiation was used but the source collimation was varied. Decreasing the divergence angle of the source collimator (i.e from 1° to $1/6^\circ$) and thus reducing the size of the interrogating primary beam resulted in Bragg maxima with decreased full width half maximum (FWHM) (i.e. narrower Bragg maxima) and hence a greater resolution diffraction pattern. Normalising the patterns (again illustrated in figure 4.1) helps to illustrate that a better signal to noise ratio was also achieved by decreasing the source collimator aperture size.

Detector Collimation

The diffraction patterns obtained from a 0.2 mm thick sheet of aluminium obtained in transmission are illustrated in figure 4.2 when the smallest aperture source col-

limator ($1/6^\circ$, identified as producing the best resolution diffraction pattern previously) was used but the detector collimation was varied. Conversely, increasing the acceptance angle of the detector collimator (from 0.75 to 1.8°) did not affect the resolution of diffraction pattern or the signal to noise ratio.

This implies that an open source collimation geometry can be used (where there is no defined acceptance angle for diffracted X-rays to strike the detector) providing suitable source collimation is used to provide a narrow interrogating primary beam (discussed further in section 5). Thick / elongated samples (along the primary beam path) do impinge on the diffraction pattern resolution when an open collimation geometry is employed. This is investigated further in section 4.3.1 and discussed in section 5.

4.2 Circumferential Configuration Approach

The rationale for interpreting diffraction patterns emerging from an unknown locus utilising a circumferential detector has been discussed in section 3.3. An example of its use to resolve and identify multiple sheets of aluminium is presented here. This particular example is also illustrated in Dicken et al. [126]. An empirical and analytical approach to predict the locus (r_1) of the Bragg maxima is also demonstrated. These are compared and then discussed later in section 5.

Figure 4.1: *Normalised diffractogram of sheet aluminium obtained in transmission with 1.8° detector collimation and specified degrees of source collimation.*

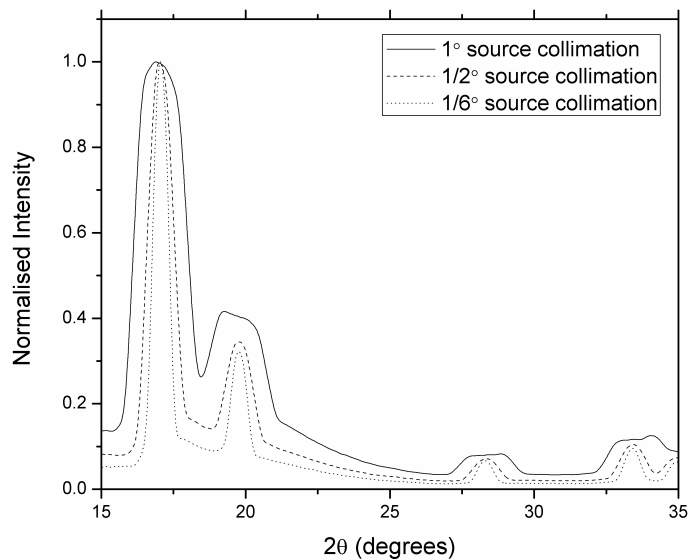
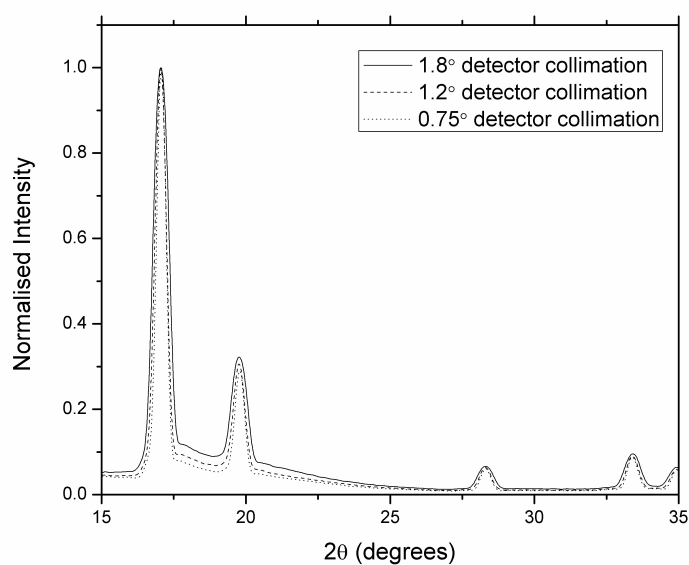


Figure 4.2: *Normalised diffractogram of sheet aluminium obtained in transmission with $1/6^\circ$ source collimation at specified degrees of detector collimation.*



4.2.1 Displaced Patterns

In order to test the validity of the method outlined in section 3.3.1 a sheet of aluminium $\simeq 0.2$ mm thick was evaluated in transmission using the experimental parameters detailed in section 3.6. Figure 4.3 illustrates the experimentally collected diffraction pattern of aluminium when it is moved / displaced (from the rotation axis by 10 mm along the primary beam path toward the detector) as well as the diffraction pattern of aluminum when it is located on the rotation axis (i.e no displacement) to demonstrate the movement incurred. Displacing the sample toward the detector results in a shift in the observed Bragg maxima ($2\theta_{obs}$) as was anticipated by the transmission displacement error equation (3.2 in section 3.3.1).

With no a priori knowledge it is not possible to unequivocally attribute the displaced pattern to aluminium. A systematic approach was applied to this displaced pattern to provide an identification. Diffraction patterns (with no displacement) were modified using equation 3.2 (illustrated in section 3.3.1) to how they would appear at a multitude of r_1 positions. Each calculated pattern was then systematically compared to the unknown. The calculated pattern that had concordant $2\theta_{obs}$ Bragg maxima positions to that of the unknown is illustrated in figure 4.4. The calculated pattern is aluminium as it would appear when displaced by 10 mm from the rotation axis toward the detector (i.e the same as the unknown).

Figure 4.3: *Two normalised diffraction patterns of sheet aluminum measured in transmission. One is located on the rotation axis (as is the convention). The second is displaced by 10 mm from the rotation axis along the primary X-ray beam path toward the detector.*

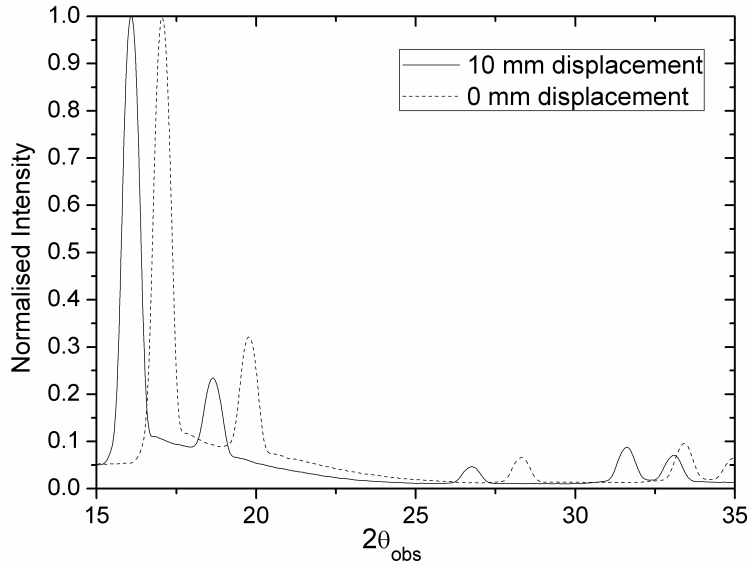
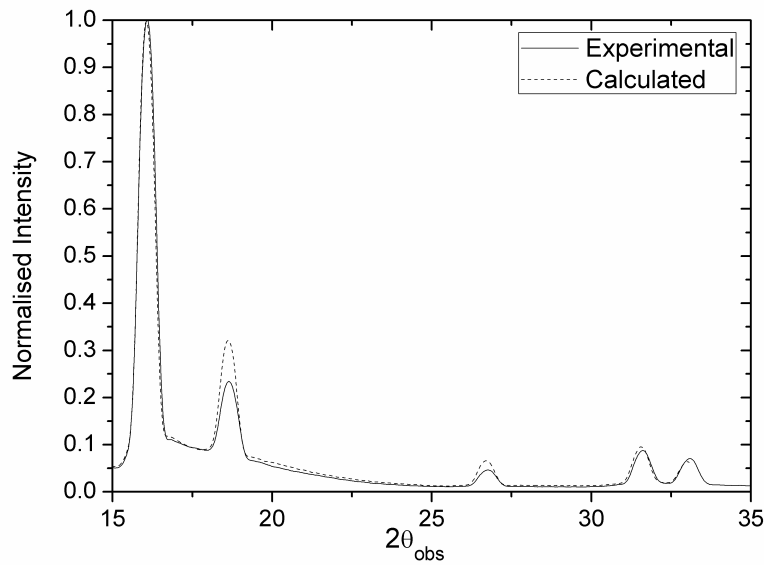


Figure 4.4: *Two diffraction patterns are illustrated. An experimentally measured diffraction pattern of sheet aluminium measured in transmission displaced by 10 mm similarly to that illustrated in figure 4.3. Also a calculated pattern of the same material at the same hypothesised locus.*



4.2.2 Multiple Patterns

To resolve multiple material diffraction patterns and thus test the theory described in section 3.3.2, diffraction patterns containing two spatially separated sheets of aluminum were created in Matlab® for differing orientations (α) of the inspection volume. Similarly to that described in section 3.5.1 the diffraction patterns of aluminium at differing displacements (2 and 12 mm from the rotation axis toward the detector) were transformed to how they would appear from 1 - 45° in α (in 1° increments). The two patterns for each α increment were then linearly summed. Four of these summed patterns (0, 15, 30 and 45° in α respectively) are illustrated in figure 4.5. Simulating this data set was a necessity as obtaining such a set of diffraction patterns at the same resolution experimentally would have taken ~ 83 hours. To ensure the validity of the synthesised data set two of the combined diffraction patterns (0 and 45° in α) were obtained experimentally and compared to the simulated patterns. These comparisons are illustrated in figures 4.6 and 4.7, respectively. The $2\theta_{obs}$ values between the experimental and simulated patterns are concordant. A discrepancy in Bragg maxima amplitude is observed but this is seldom used in crystallography (and not used at all in this study) because of the unknown amount of preferred orientation in the sample. Bragg maxima amplitude (intensity) is discussed in section 5.

The manner with which Bragg maxima modify their position ($2\theta_{obs}$) with α is governed by the Bragg maxima locus on the primary beam path (i.e sample position or

Figure 4.5: A series of simulated combined diffraction patterns (containing sheet aluminium displaced by 2 and 12 mm from the rotation axis along the primary X-ray beam path toward the detector) as they would appear if the inspection volume were rotated through 0, 15, 30 and 45° in α . Please note each pattern is normalised to 1 and a positive offset has been included for clarity.

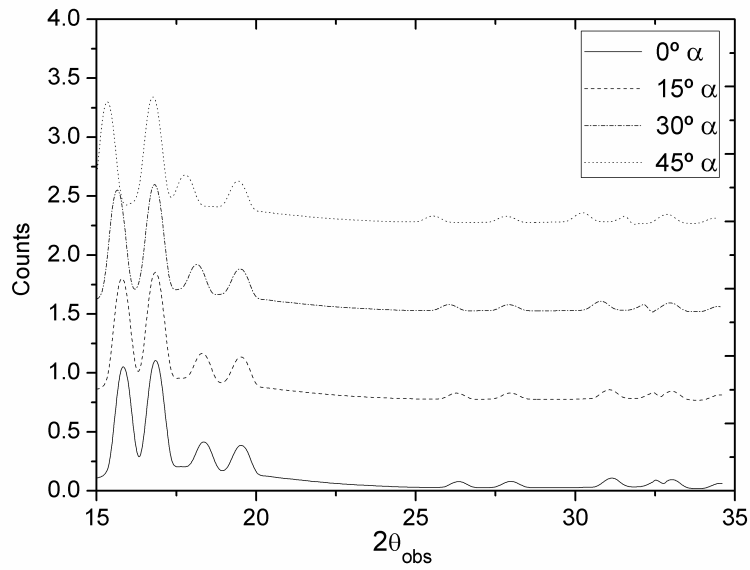


Figure 4.6: *Experimentally collected combined diffraction pattern (containing sheet aluminium displaced by 2 and 12 mm from the rotation axis along the primary beam path toward the detector) at 0° in α overlaid with the simulated data of the same scenario.*

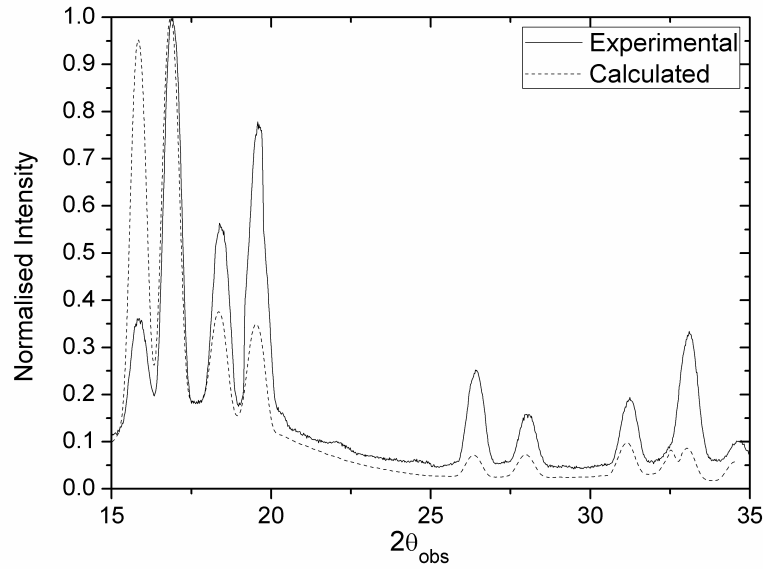
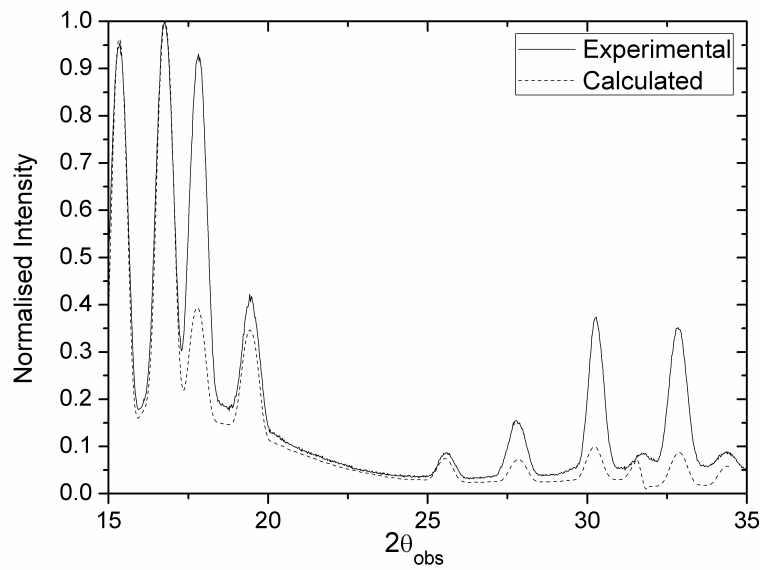


Figure 4.7: *Experimentally collected combined diffraction pattern (containing sheet aluminium displaced by 2 and 12 mm from the point of rotation along the primary beam path toward the detector) at 45° in α overlaid with the simulated data of the same scenario.*



r_1) and $2\theta_{std}$. These Bragg maxima were first separated according to their loci before an identification was made. The position modification contributed by the unknown $2\theta_{std}$ must be removed. The empirical approach (described earlier in section 3.3.2) demonstrates that dividing the $2\theta_{obs}$ values for rotations 1 - 45° in α by their respective $2\theta_{obs}$ value in the 0° diffraction pattern will remove the position modification contributed from $2\theta_{std}$ (and thus peak modification can be attributed to equation 3.4, section 3.3.2 for small angles of $2\theta_{std}$) . This can be thought of as normalising the Bragg position modification with respect to $2\theta_{std}$. The result is illustrated in figure 4.8. Each of the lines (representing individual peak positions) adhere to one of two characteristic curves. This enables the Bragg maxima to be separated qualitatively as each curve is characteristic of a sample loci altered with α . To separate the Bragg maxima quantitatively each line was fit to $y = ax^2 + bx + c$. The Taylor series expansion of $\cos(x)$ is illustrated by equation 8.5 (section 8.4). This illustrates that the second order term is the most influential factor within the cosine expansion, therefore a second order polynomial was deemed appropriate. Plotting the coefficients of the fitted polynomials resulted in Bragg maxima from the same locus falling in the same region of the graph. This is illustrated in figure 4.9.

Figure 4.8: Illustration of the $2\theta_{obs}$ position modification with α for the Bragg maxima positions illustrated in figure 4.5 after the variation from $2\theta_{std}$ has been removed.

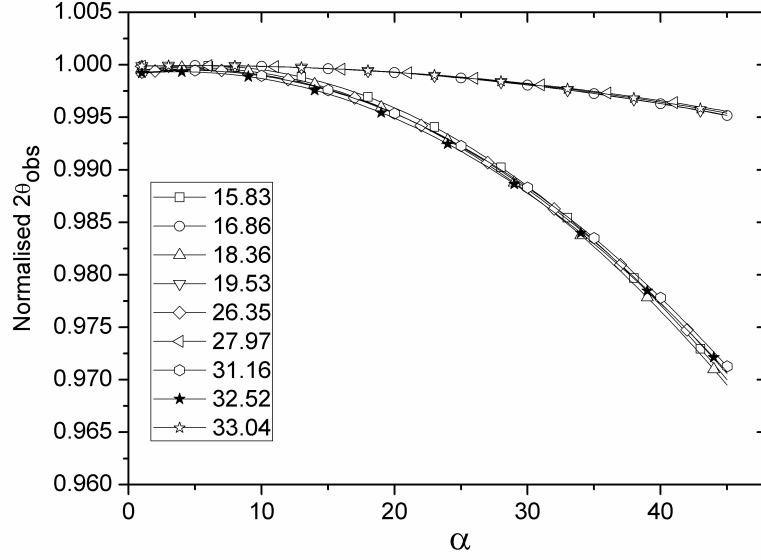


Figure 4.9: Plot of the first and second order polynomial coefficients for a fitted curve that represents $2\theta_{obs}$ position modification with α after being normalised with respect to $2\theta_{std}$. Ovals that circle Bragg maxima originating from the same loci have been included for clarity.

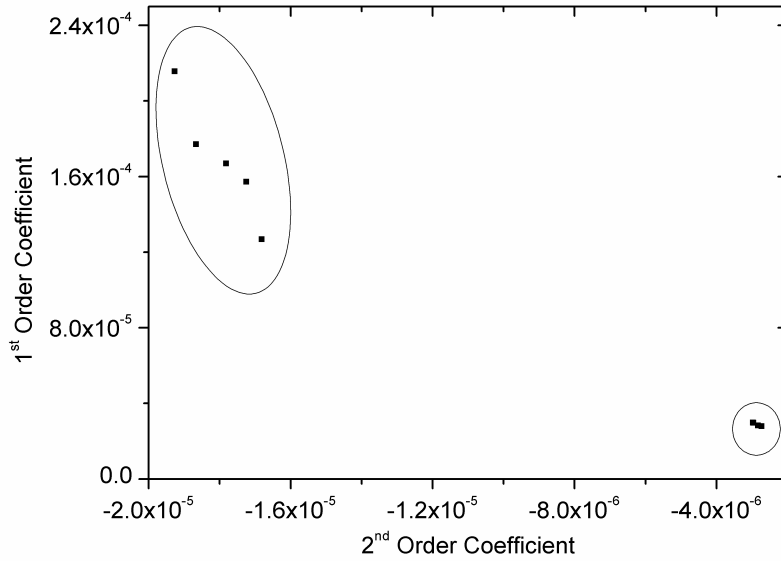
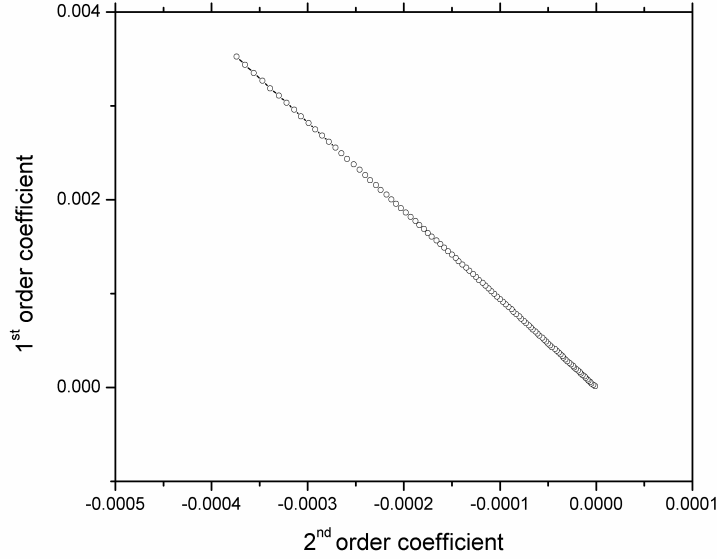


Figure 4.11: Graph illustrating the linear relationship between the 1st and 2nd order coefficients of the polynomial fits that were applied to the normalised sample position movement illustrated in figure 4.10. Coefficients are illustrated for values of r_1 between 1 to 100 mm from the rotation axis along the primary beam path toward the detector.



To ascertain the locus of the Bragg maxima, equation 3.4 (section 3.3.2) was evaluated graphically for known r_1 positions (a selection of which are illustrated in figure 4.10). The position modification illustrated in figure 4.8 is an approximation of the position modification illustrated in figure 4.10. Each of the r_1 curves in figure 4.10 were similarly fit to $y = ax^2 + bx + c$. A plot of a against b for all of the fitted r_1 loci (between 1 and 100 mm from the rotation axis along the primary beam toward the detector) is illustrated in figure 4.11. This illustrates that the relationship between the first (a) and second (b) order coefficients are linear. As the first and second order coefficients are not independent only a single coefficient was deemed necessary to predict r_1 . A plot of the second order polynomial coefficients against r_1 is illustrated

in figure 4.12. This was fit to $y = ax^4 + bx^3 + cx^2 + dx + e$ (a fourth order polynomial, equation 4.1). This equation is demonstrated below:

$$r_1 = -3.9E^5x^4 - 4.4E^{12}x^3 - 2.1E^9x^2 - 6.5E^5x + 0.26 \quad (4.1)$$

where x is the 2nd order coefficient. An alternate, analytical approach for predicting r_1 that only requires $2\theta_{obs}$ from two diffractograms (based on equation 3.5, section 3.3.2) was also explored. To compare these approaches table 4.1 illustrates the predicted r_1 values for the Bragg maxima positions illustrated in figure 4.5 as predicted by the empirical and analytical approach. Material identity for each set of Bragg maxima were systematically identified as aluminium. A more detailed discussion of the result of this method and in particular a comparison of the empirical and analytical approach to identify r_1 (i.e the sample locus) is offered in section 5.

Table 4.1: *Comparison of the known sample locus of the Bragg maxima positions illustrated in 4.5 with those predicted from an empirical and analytical approach.*

$2\theta_{obs}$ in the $0^\circ\alpha$ diffractogram ($^\circ$)	Known sample position (r_1) in mm	Empirically predicted sample position (r_1) in mm	Analytically predicted sample position (r_1) in mm
15.83	12.0	12.1	11.5
16.86	2.0	2.2	1.9
18.36	12.0	11.7	11.8
19.53	2.0	2.2	1.9
26.35	12.0	11.2	11.2
27.97	2.0	2.0	1.8
31.16	12.0	10.9	11.2
32.53	12.0	10.6	11.2
33.04	2.0	2.1	1.8

Figure 4.10: Graph illustrating normalised sample position movement with α for specified values of r_1 .

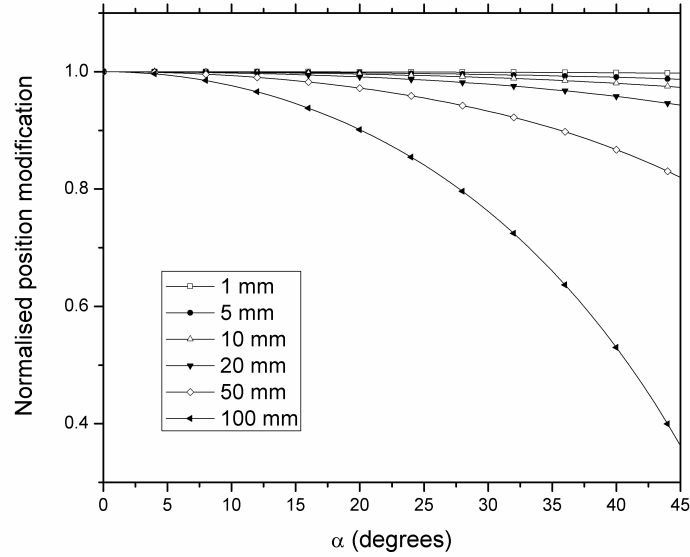
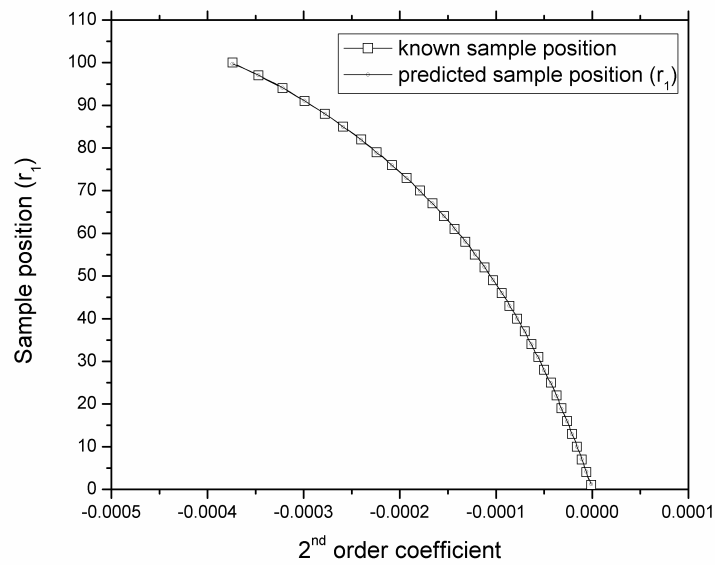


Figure 4.12: Graph illustrating the relationship between the 2nd order coefficients illustrated in figure 4.11 and the known r_1 positions. As well as the empirically calculated r_1 predictions from equation 4.1 (section 4.2).



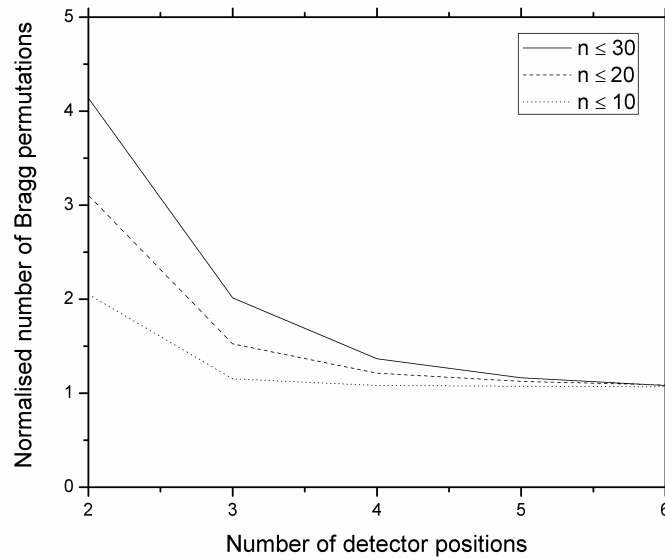
4.3 Linear Configuration Approach

The linear configuration (described in section 3.4) was subject to a more thorough examination than the circumferential configuration, justifications for which are given in the discussion (section 5). This section starts by demonstrating the premise of this technique by computer simulation and justifying the use of four detector positions. Confounding factors such as sample thickness / elongation, detector distance and radiation energy are simulated and explored. Finally the configuration is put into practice in the laboratory. Some of the examples demonstrated here are presented in Dicken et al. [130].

A minimum of two detector positions are required to calculate the Bragg maxima position ($2\theta_{std}$) and the corresponding sample locus. However with no coincidence information cross referencing every Bragg maxima position (n) from detector 1, to detector to 2 results in many ($(n(n+1)/2)$ to n^2) possibilities/permutations. The inclusion of boundary conditions (illustrated in section 3.4.1) removes some false permutations. However a significant additional reduction can be obtained by including more detector positions and only accepting Bragg maxima that propagate linearly through all detectors. The relationship between the number of Bragg maxima permutations and the number of detector positions was investigated empirically (illustrated in figure 4.13). The permutations that are evaluated here are from the same ~ 1000 simulations used to generate the ROC surfaces described in section 3.5.3 and demonstrated in sections 4.3.1 - 4.3.3. The number of permutations are

represented as a proportion of the number of Bragg maxima positions (n) in the primary data. Increasing the number of detector positions reduces the number of permutations, with the most significant proportion being removed at three detector positions and less so for more detector positions. Simulations involving a greater number of Bragg maxima positions (n) in the primary data (i.e $n \leq 30$) result in more permutations than simulations involving fewer Bragg maxima positions (i.e $n \leq 10$) as would be anticipated. Four detector positions were used for the simulated experiments as well as those obtained in the laboratory to act as a compromise between accuracy and efficiency. This is discussed further in section 5.

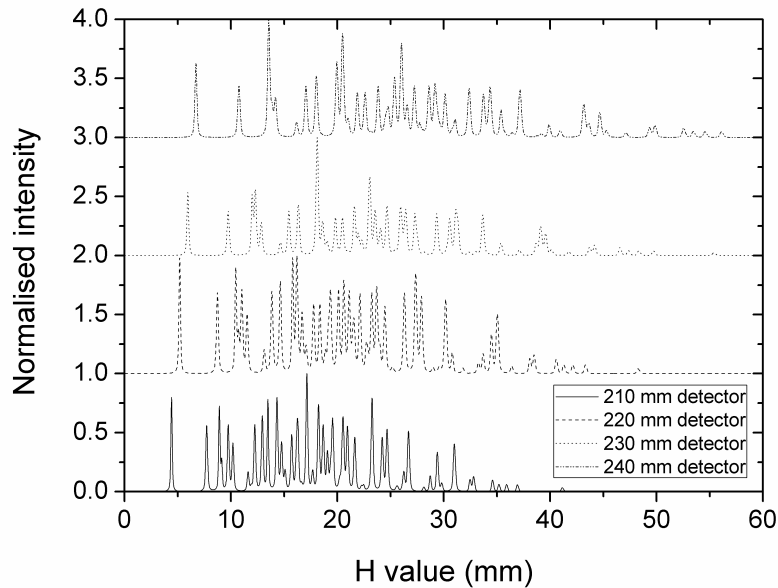
Figure 4.13: *Graph illustrating the number of Bragg maxima positions tracked through differing numbers of detector positions for differing numbers of Bragg maxima (n) in the primary data. The permutations have been normalised with respect to the number of Bragg maxima (n) in the primary data to give an idea of the proportion of correct permutations.*



To demonstrate the premise of the linear configuration figure 4.14 illustrates simu-

lated diffraction patterns at four detector positions created by a mixture of spatially separated trinitrotoluene (TNT) and talcum powder ($\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$). These samples were simulated to have 0.1 mm thickness along the primary beam path. These two materials were chosen at random from the material library described in section 3.5.1 as well as randomly being assigned a position within the inspection volume. Please note that the Bragg maxima position's (H values) on the detectors for this geometry are given in mm. This should not to be confused with Bragg maxima position's given in angle ($2\theta_{obs}$) used by the circumferential configuration. For the linear configuration the change in Bragg maxima position (ΔH) is a function of $2\theta_{std}$ and the sample position along the primary X-ray beam path (as illustrated in figure 3.6).

Figure 4.14: *Simulation of the diffraction patterns observed on a series of linear arrays arranged normal to the primary beam path at 210, 220, 230 and 240 mm from X-ray the source. The diffracting materials are spatially separated TNT and Talcum powder ($\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$). Please note each pattern is normalised to 1 and a positive offset has been included for clarity.*



TNT and talcum powder collectively give rise to 41 Bragg maxima positions (n). Cross referencing all of these Bragg maxima results in anywhere from 861 - 1681 positive $2\theta_{std}$ and corresponding locus permutations. Applying the peak tracking method described in section 3.4.1 (i.e the need for the Bragg maxima to propagate linearly through four detector positions) removed the majority of these false permutations. Boundary conditions (also discussed in section 3.4.1) for additional refinement were then applied. Removing 2θ values exceeding 40° reduced the permutations to 242. Removing permutations that lie outside the boundaries of the inspection volume (for this example, 70 - 170 mm from the source) reduced the number further to 120. Permutations sharing a similar locus (~ 1 mm) were then grouped together and compared systematically against the material library database. Two of these groups of Bragg maxima are illustrated in figures 4.15 and 4.16. Each set of Bragg maxima had concordant 2θ values to that of TNT and talcum powder respectively. They also proved to be the most likely candidate materials as determined by a conventional diffraction matching algorithm (Crystallographic Search-Match®) that searched $\sim 200,000$ diffraction patterns.

Figure 4.15: Plot of a grouping of Bragg maxima tracked through the patterns illustrated in figure 4.14 believed to have a common locus (~ 1 mm). The pattern corresponds well with Trinitrotoluene (TNT). Please note each pattern is normalised to 1 and a positive offset has been included for clarity.

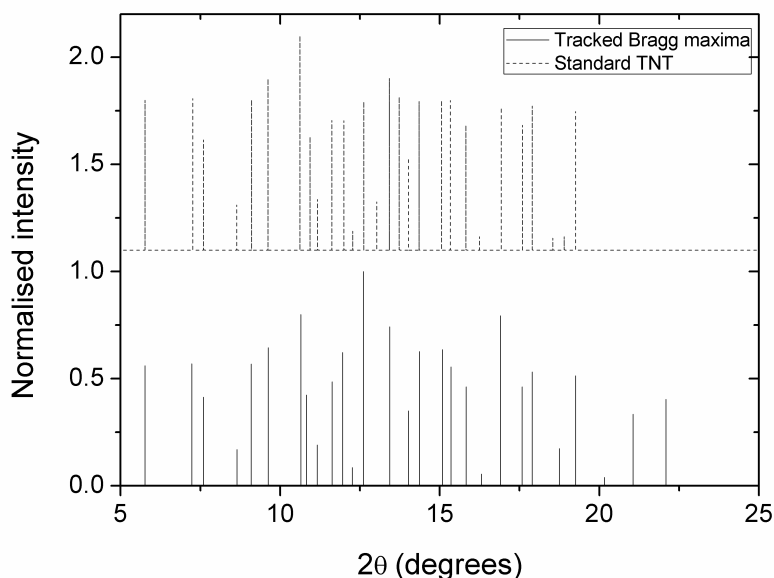
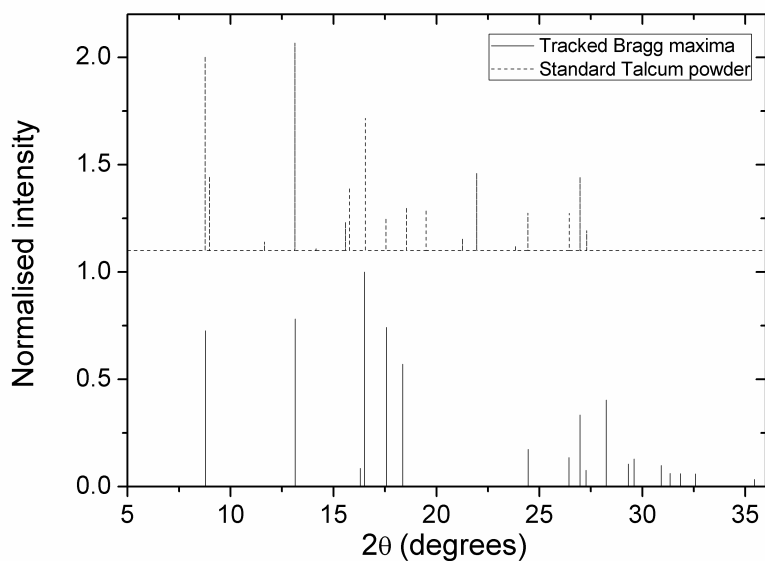


Figure 4.16: Plot of a grouping of Bragg maxima tracked through the patterns illustrated in 4.14 believed to have a common locus (~ 1 mm). The pattern corresponds well with talcum powder ($H_2Mg_3(SiO_3)_4$). Please note each pattern is normalised to 1 and a positive offset has been included for clarity.



4.3.1 Simulating Sample Elongation

When an open collimation system (described in section 4.1) is considered, the extension of the sample along the primary beam path results in broadening of the diffraction maxima (when obtained in transmission). An example of this broadening is illustrated in figure 4.17. It was hypothesised that Bragg maxima broadening will detrimentally impinge on the linear diffraction configuration (and the circumferential configuration) performance because it causes ambiguity in Bragg maxima position.

Figure 4.17: *Simulation illustrating the width of the $1.0321d$ Bragg maxima produced by iron situated at 150 mm from the source and detected at 210 mm at differing degrees of elongation. This d -spacing is diffracted at 20.24° at molybdenum $K\alpha$.*

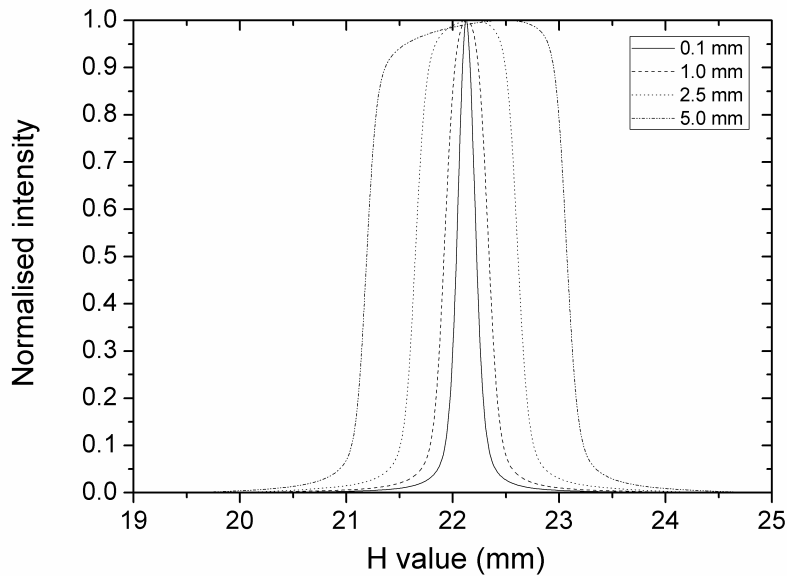


Figure 4.18: *Material diffractograms for calcite, sucrose, polystyrene, cobalt, iron and nylon simulated at molybdenum $K\alpha$, used to assess linear configuration performance. ICDD card numbers and empirical formulae are illustrated in table 3.1.*

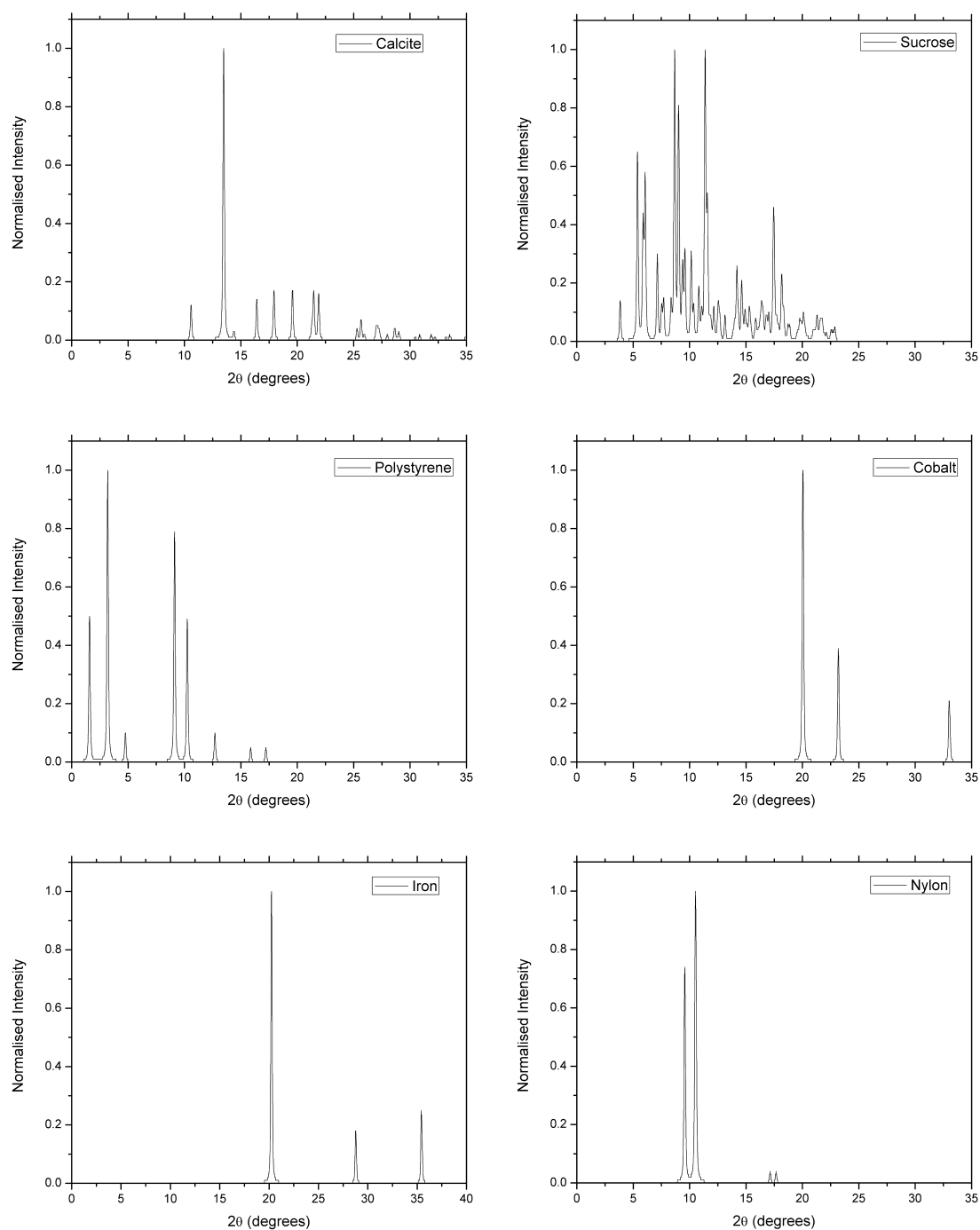


Figure 4.19: Material diffractograms for potassium chlorate, PETN (pentaerythrite tetranitrate), RDX (cyclotrimethylenetrinitramine), ammonium nitrate, cocaine (cocaine hydrochloride) and TNT (trinitrotoluene) simulated at molybdenum $K\alpha$, used to assess linear configuration performance. ICDD card numbers and empirical formulae are illustrated in table 3.1.

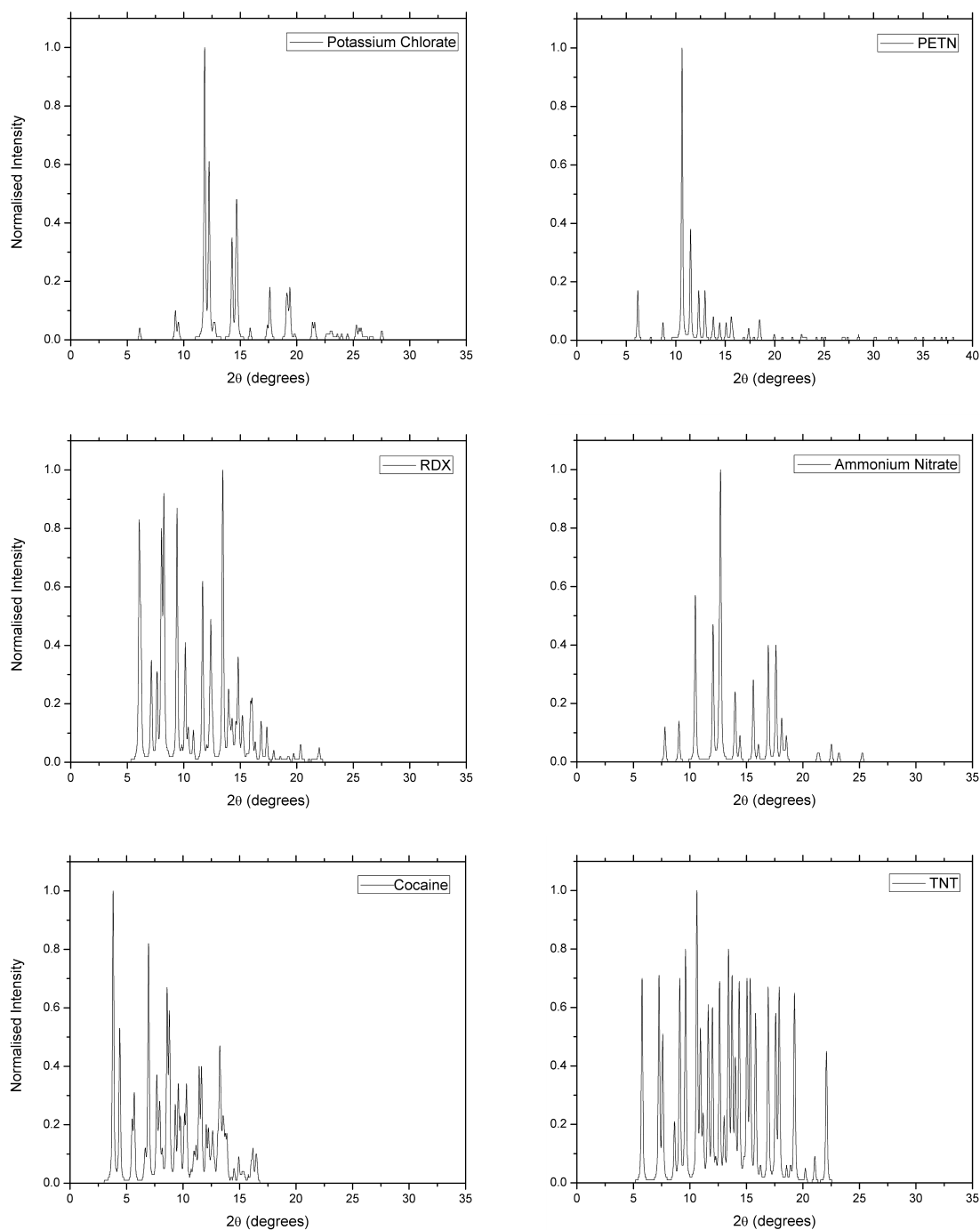


Figure 4.20: Material diffractograms for talcum powder ($Mg_3Si_4O_{10}(OH)_2$) and cellulose simulated at molybdenum $K\alpha$, used to assess linear configuration performance. ICDD card numbers and empirical formulae are illustrated in table 3.1.

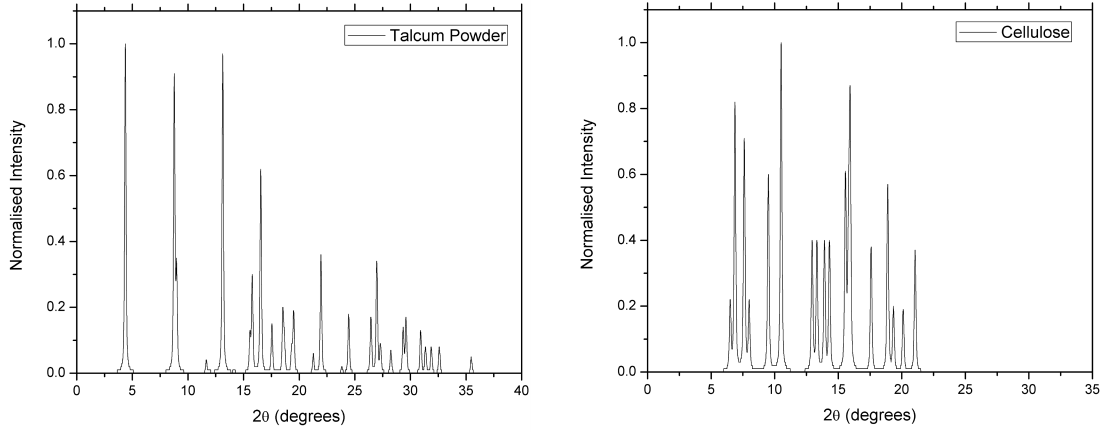
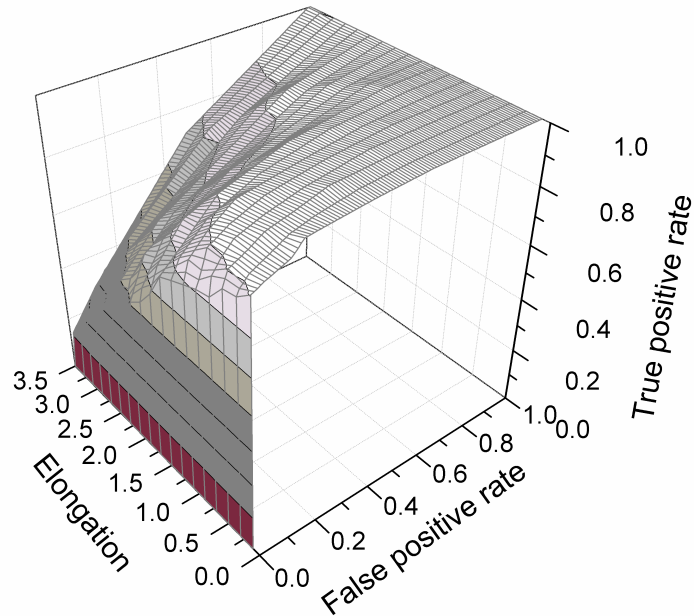


Figure 4.21: Surface illustrating the change in shape of a receiver operator characteristic (ROC) curve with elongation of the sample materials. Simulated detectors were placed at 210, 220, 230 and 240 mm from the X-ray source with molybdenum $K\alpha$ interrogating radiation.



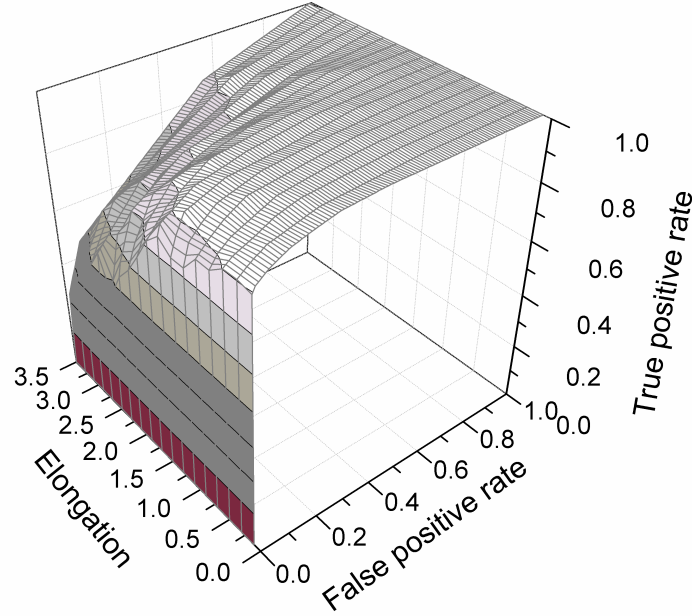
To explore this effect receiver operator characteristic (ROC) curves were created to assess the performance of the linear diffraction configuration and peak tracking method at identifying illicit material diffraction patterns at differing degrees of sample elongation (thickness). A mixture of benign and illicit material diffractograms (illustrated in figures 4.18, 4.19 and 4.20) were randomly combined (using the rationale described in section 3.5.3). The combined patterns were then interpreted using the peak tracking method and the results were compared against the primary data. The result is illustrated in figure 4.21 when detectors are placed at 210, 220, 230 and 240 mm from the X-ray source and the diffracting materials were randomly assigned a locus from 100 to 170 mm representing the boundaries of the inspection volume. When thinner (i.e 0.1 mm) elongated samples were simulated the area under the ROC curve was notably greater than when thicker (i.e 3.5 mm) elongated samples were considered. A more thorough break down of the areas under this surface (per elongation) is illustrated in table 4.2. The results from this simulation are discussed in more detail later in section 5.

4.3.2 Simulating Detector Distances

It was hypothesised that increasing the sample to detector distance (P) would improve the angular resolution of the linear configuration. This is because the concomitant increase in relative diffraction peak width is less than the corresponding positional change. This performance impact was explored by conducting the same

simulation described in section 3.5.3 and demonstrated in section 4.3.1 (i.e ROC curves for detecting illicit materials in the presence of benign materials) at differing detector positions. The detector positions as well as the spread between the detectors were increased from 210, 220, 230 and 240 mm to 220, 240, 260 and 280 mm from the X-ray source. The result is illustrated in figure 4.22. The area under the ROC curve appears to diminish with increased sample thickness (similarly to figure 4.21), but at a lower rate. The individual areas under the ROC curves (per sample thickness / elongation) are provided in table 4.2, section 4.3.3. ROC curves are typically compared using their respective areas. However since a third variable is compared here (sample thickness / elongation) it is appropriate to compare the volumes bounded by the ROC surfaces for these different simulations. These volumes are illustrated in table 4.2. The ROC surface volumes and inferences about the affect that these variables have on the system performance are discussed in section 5.

Figure 4.22: Surface illustrating the change in shape of a receiver operator characteristic (ROC) curve with elongation of the sample materials. Simulated detectors were placed at 220, 240, 260 and 280 mm from the X-ray source with molybdenum $K\alpha$ interrogating radiation.



4.3.3 Simulating Radiation Energy

The energy (wavelength) of the principle primary beam has a known affect on the radiations penetrating capability. However one of the more prevalent factors to this study is the manner with which its alters the angular resolution of the diffraction patterns. By evaluating Bragg's law (equation 2.4, section 2.4.2) decreasing the wavelength of radiation (and hence increasing the radiation energy) reduces 2θ . Figure 4.23 illustrates the same diffraction pattern from iron as it would appear if interrogated by molybdenum (~ 17 keV) and tungsten (~ 59 keV). Increasing the radiation energy reduces the angular range of the diffraction pattern from ~ 0 - 38° to

$\sim 0-12^\circ$. A scaled up version of the technology presented here will warrant an increase in radiation energy (i.e from molybdenum primarily used in this study, to tungsten used for the majority of medical and security screening applications). Therefore the manner with which radiation energy affects the performance of the linear diffraction configuration was investigated. To assess performance, ROC surfaces were created using the method outlined in section 3.5.3 with tungsten radiation. Detectors were placed at 210, 220, 230 and 240 mm (illustrated in figure 4.24) and 220, 240, 260 and 280 mm (illustrated in figure 4.25) from the X-ray source so that direct comparisons could be made with the previous surfaces simulated for molybdenum radiation (illustrated in figure 4.21 section 4.3.1 and figure 4.22 section 4.3.2, respectively). The volumes bounded by these surfaces give an indication of their performance at identifying illicit materials from benign materials at a range of sample thickness's (elongation's). These volumes are presented and discussed in section 5.

Figure 4.23: *Normalised diffraction patterns of iron, simulated for molybdenum (~ 17 keV) and tungsten (~ 59 keV) radiation.*

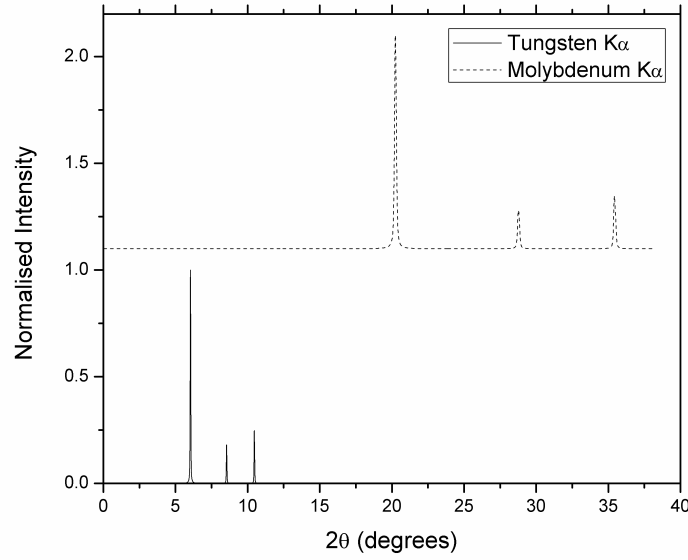


Figure 4.24: *Surface illustrating the change in shape of a receiver operator characteristic (ROC) curve with elongation of the sample materials. Simulated detectors were placed at 210, 220, 230 and 240 mm from the X-ray source with Tungsten $K\alpha$ interrogating radiation.*

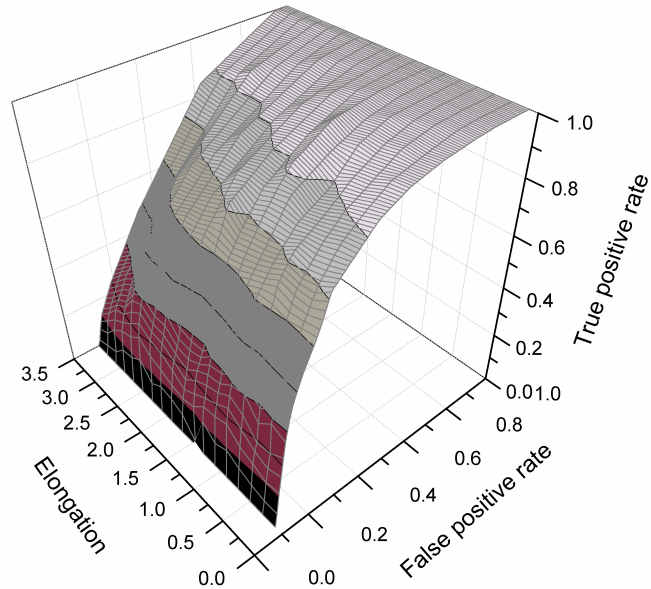


Figure 4.25: Surface illustrating the change in shape of a receiver operator characteristic (ROC) curve with elongation of the sample materials. Simulated detectors were placed at 220, 240, 260 and 280 mm from the X-ray source with Tungsten $K\alpha$ interrogating radiation.

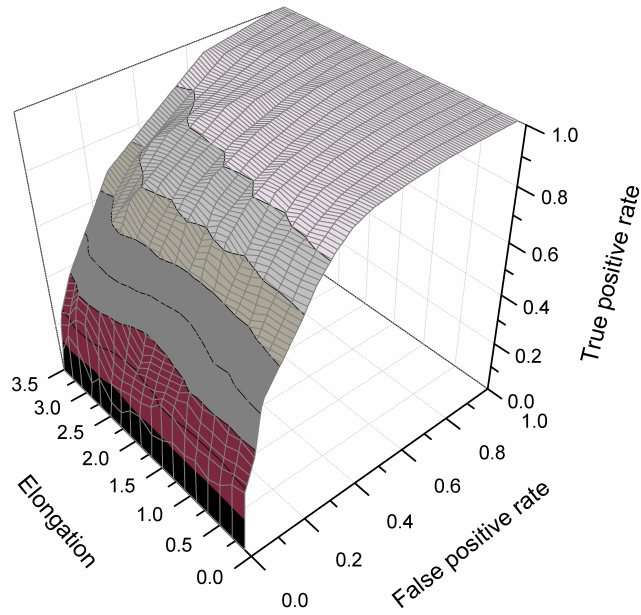
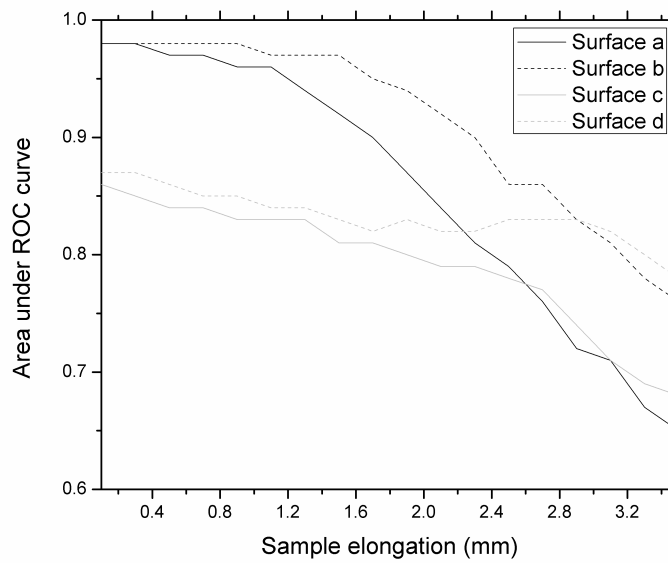


Figure 4.26: Plot of the area under the receiver operator characteristic (ROC) curve's for various sample thickness's (elongations) for differing radiation energies and detector distances. The data presented here is also illustrated in table 4.2.



The area's under each ROC curve (one for each sample thickness / elongation) for each of the surfaces described previously provide interesting trends that cannot be appreciated by evaluating each of their volumes alone. Table 4.2 illustrates a break down of the individual ROC areas (per sample thickness) for the ROC surfaces illustrated in figures 4.21, 4.22, 4.24 and 4.25 (in sections 4.3.1, 4.3.2 and 4.3.3, respectively). An area of 1 represents perfect discrimination (i.e all illicit materials identified as illicit materials and all benign materials identified as benign). An area of 0.5 represents no discrimination. The closer the area to 1 the better the system performance. Increasing the sample to detector distances (from 210, 220, 230 and 240 mm to 220, 240, 260, 280 mm) with tungsten radiation (surface c to surface d) results in better system performance similarly to the simulations performed with molybdenum radiation (surface a to surface b), explored in section 4.3.2. Increasing the sample thickness (elongation) along the primary beam is detrimental to system performance (mentioned previously in section 4.3.1) for all simulations. However, interestingly tungsten radiation is affected to a lesser extent than molybdenum radiation. Inferences from these observations are discussed in section 5.

Table 4.2: *Illustration of the areas under the receiver operator characteristic (ROC) curves for differing degrees of elongation when (a) detectors are at 210, 220, 230 and 240 mm from the source with molybdenum $K\alpha$ radiation (illustrated in figure 4.21 section 4.3.1), (b) detectors at 220, 240, 260 and 280 mm from the source with molybdenum $K\alpha$ radiation (illustrated in figure 4.22 section 4.3.2), (c) detectors are at 210, 220, 230 and 240 mm from the source with tungsten $K\alpha$ radiation (illustrated in figure 4.24 section 4.3.3) and (d) detectors at 220, 240, 260 and 280 mm from the source with tungsten $K\alpha$ radiation (illustrated in figure 4.25 section 4.3.3).*

Sample thickness (elongation)	Surface a	Surface b	Surface c	Surface d
0.1	0.98	0.98	0.86	0.87
0.3	0.98	0.98	0.85	0.87
0.5	0.97	0.98	0.84	0.86
0.7	0.97	0.98	0.84	0.85
0.9	0.96	0.98	0.83	0.85
1.1	0.96	0.97	0.83	0.84
1.3	0.94	0.97	0.83	0.84
1.5	0.92	0.97	0.81	0.83
1.7	0.90	0.95	0.81	0.82
1.9	0.87	0.94	0.80	0.83
2.1	0.84	0.92	0.79	0.82
2.3	0.81	0.90	0.79	0.82
2.5	0.79	0.86	0.78	0.83
2.7	0.76	0.86	0.77	0.83
2.9	0.72	0.83	0.74	0.82
3.1	0.71	0.81	0.71	0.80
3.3	0.67	0.78	0.69	0.78
3.5	0.65	0.76	0.68	0.78
Volume (%)	86%	91%	79%	83%

4.3.4 Experimental Proof of Principle

To evaluate the linear diffraction configuration empirically sheets of aluminium ($\simeq 0.2$ mm elongation) and aluminium oxide ($\simeq 0.1$ mm elongation) were placed on the primary beam path in known locations within the inspection volume. Four

diffraction patterns were obtained by rastering the camera normal to the primary beam path at 210, 220, 230 and 240 mm from the X-ray source (similarly to that illustrated in figure 3.6). These patterns were radially integrated to produce the 1D scattering patterns illustrated in figure 4.27.

The peak tracking algorithm was applied to these four patterns and the resulting 2θ permutations were grouped according to their locus (i.e. within 1 mm of each other). Two of these groupings are illustrated in figures 4.28 and 4.29. These tracked Bragg maxima are concordant with aluminum and aluminium oxide. Some Bragg maxima are missing. This implies that either the peaking finding algorithm failed to identify the Bragg maxima in all four diffraction patterns or alternatively the Bragg maxima failed the linearity test imposed by the peak tracking method (section 3.4.1). A thorough examination of these results is offered later in the Discussion (section 5).

Figure 4.27: Radially integrated diffraction patterns of spatially separated aluminum and aluminium oxide measured normal to the primary beam at specified distances from the X-ray source. Please note each pattern is normalised to 1 and a positive offset has been included for clarity.

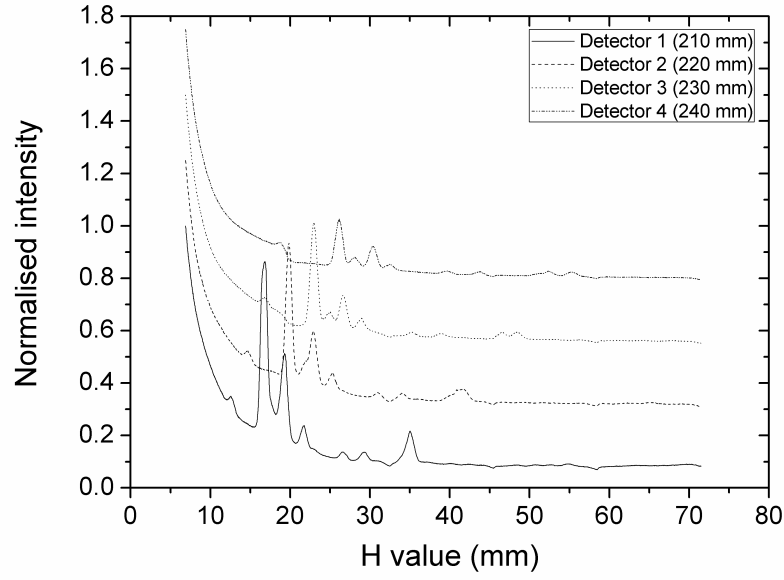


Figure 4.28: Plot of a group of Bragg maxima tracked through the diffraction patterns illustrated in figure 4.27 that share a common (± 0.5 mm) location on the primary beam path. The most likely candidate material (Al) is also illustrated. Please note each pattern is normalised to 1 and a positive offset has been included for clarity.

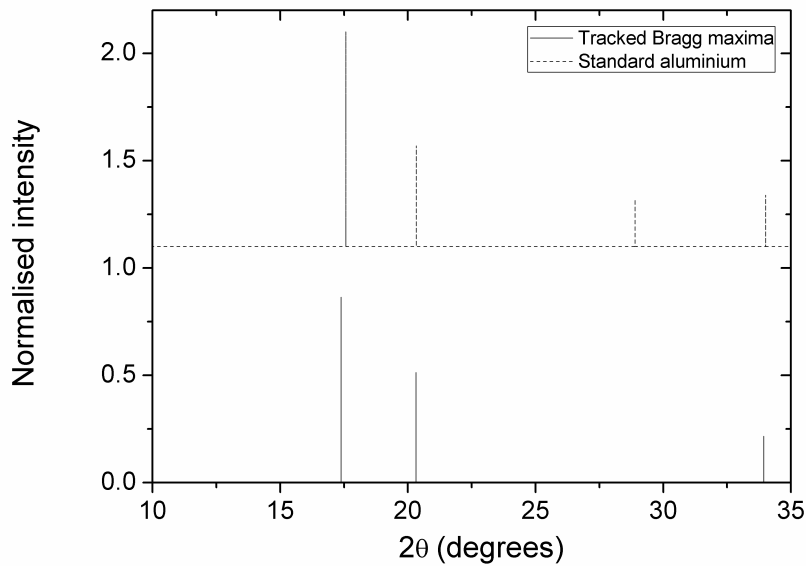
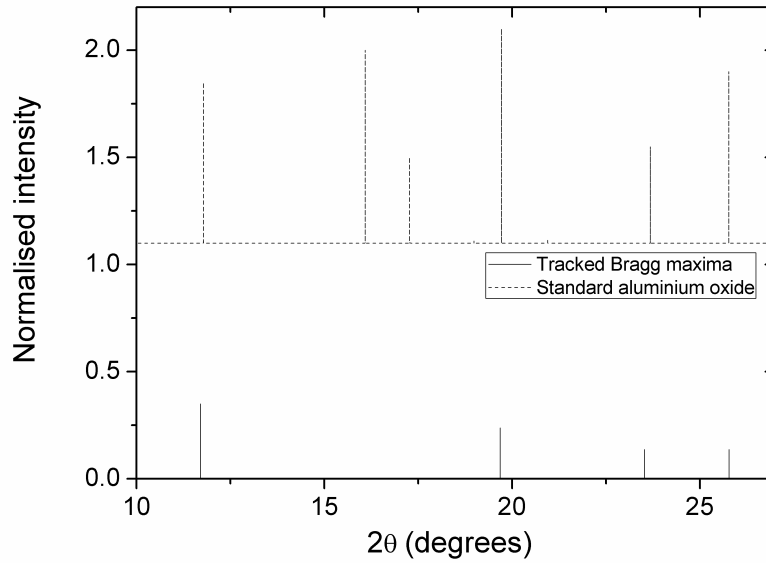


Figure 4.29: *Plot of a group of Bragg maxima tracked through the diffraction patterns illustrated in figure 4.27 that share a common (± 0.5 mm) location on the primary beam path. The most likely candidate material (Al_2O_3) is also illustrated. Please note each pattern is normalised to 1 and a positive offset has been included for clarity.*



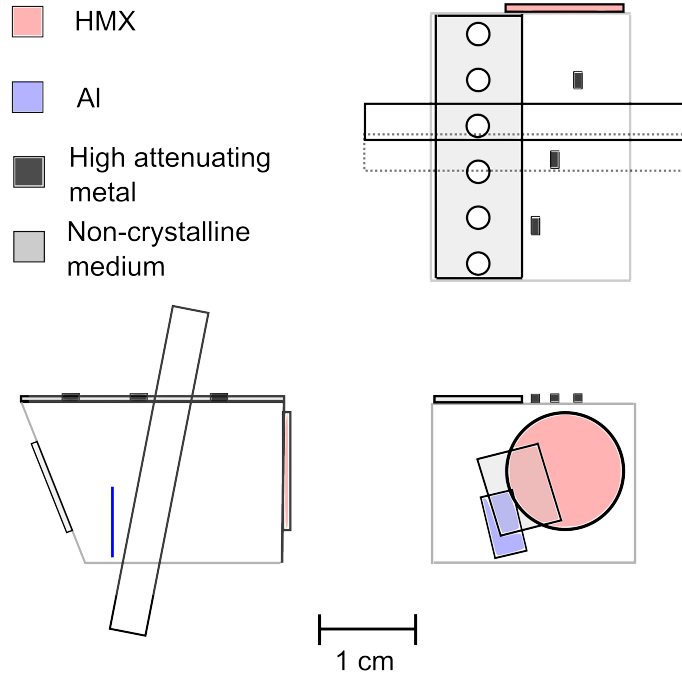
4.4 Combining KDEX and X-ray Diffraction

The principles of combining KDEX with material specific X-ray diffraction described in this section have been presented elsewhere Dicken et al. [131]. In the example presented here HMX (cyclotetraminemethylenetetranitramine, a military grade explosive) is “targeted” rather than Al_2O_3 . To combine KDEX and X-ray diffraction a small scale phantom was constructed that resembled an improvised explosive device (IED, discussed previously in section 1.2). This phantom contained a disc of HMX as well as other scattering and attenuating objects. A scaled schematic of the

phantom is illustrated in figure 4.30 as well as a 3D perspective drawing in figure 4.31. Material compositions of items within the phantom are highlighted via colour encoding. A KDEX image sequence of this phantom was obtained from $\pm 15^\circ$ with 1° increments similarly to the procedure described in section 3.6. Depth perception is afforded by parallax (described in section 2.3.4), this is best illustrated in the accompanying video (video 2). These image frames have also been concatenated and included in figure 4.32. The linear diffraction configuration was chosen as the most appropriate method to identify the “target” material. A pencil beam was used to interrogate the phantom when held at $0^\circ \alpha$. Please note that the orientation of the scattering materials within the phantom were idealised for this experiment. The “target” was placed in its thinnest orientation along the primary beam path yet occupied a large area in the other two dimensions (x and y) to improve the number of interrogation sites. 25 locations (x and y) were interrogated (illustrated in figure 4.33a). Similarly to the procedure described in section 4.3.4 four diffraction patterns were collected for each site and subject to the peak tracking method. An example of these patterns is illustrated in figure 4.34, this demonstrates the four diffraction patterns obtained from interrogation site A3 (illustrated in figure 4.33). After the linearly propagating Bragg maxima positions were tracked through the diffraction patterns they were grouped according to their position (on the primary beam path) within the inspection volume. Bragg maxima with a locus within 3 mm (rather than 1 mm previously used) of each other were grouped and identified. A larger tolerance ($\pm 0.2^\circ$) was required for this particular sample because there was greater

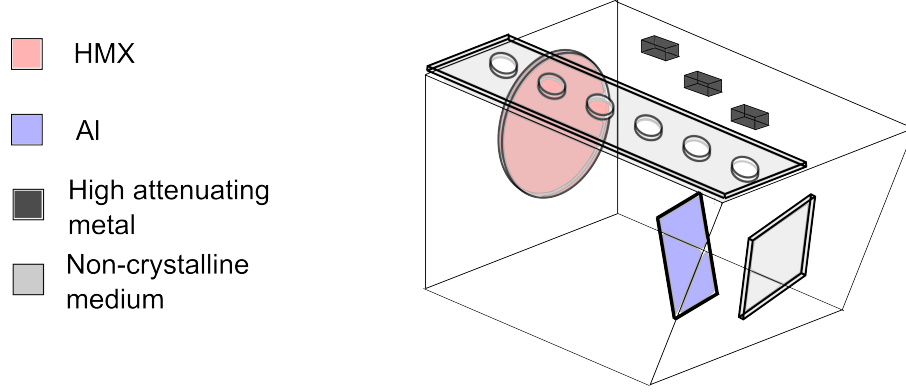
ambiguity in the Bragg maxima positions and hence the subsequently calculated locus position. This was probably due to the sample having a large grain size and is discussed later in section 5. A comparison between HMX and the tracked Bragg maxima for interrogation site A3 is demonstrated in figure 4.35.

Figure 4.30: *Scaled schematic drawing illustrating the locations and material composition of objects placed within a phantom object.*



In total 6 tracked Bragg maxima for interrogation site A3 shared concordant 2θ positions ($\pm 0.2^\circ$) with the standard HMX diffraction pattern. A map of the number of concordant 2θ positions for the other interrogation sites is illustrated in figure 4.33b. To colour encode these “tagged” areas the pixel values in the red (R) channel were multiplied by a weighting factor based on number of concordant 2θ positions (or “match index”). The greater the match index, the greater the weighting factor

Figure 4.31: *3D perspective drawing illustrating the locations and material composition of objects within a phantom object.*



applied to the red channel and hence intensity of red. The subsequent colour encoding based on the map illustrated in figure 4.33b is shown adjacently in figure 4.33c. Each of the concordant Bragg maxima also had a calculated locus on the primary beam path. Table 4.3 illustrates the interrogation site, the number of concordant Bragg maxima (with HMX) and an average of the sample position calculated from the X-ray source.

The “tagged” regions illustrated in figure 4.33c were recalculated for the other KDEX transmission images ($\pm 15^\circ$) illustrated in figure 4.32. A ruler (apparent in the top right hand corner of the transmission images) was used to convert the depth of the tagged locations from mm into their equivalent location in pixels (used by the program in Matlab®). The images within the Matlab® program were kept in pixels rather than mm. Objects within the phantom from differing locations (in depth on the primary beam path) were subject to differing amounts of magnification and thus a mm scale was not appropriate. This ruler has a known position on the primary beam path (spanning 146.5 - 173.5 mm from the X-ray source) which equates to

Figure 4.32: *KDEX* image sequence of an improvised explosive device (IED) phantom from $\pm 15^\circ$ rotation (α) with 1° increments.

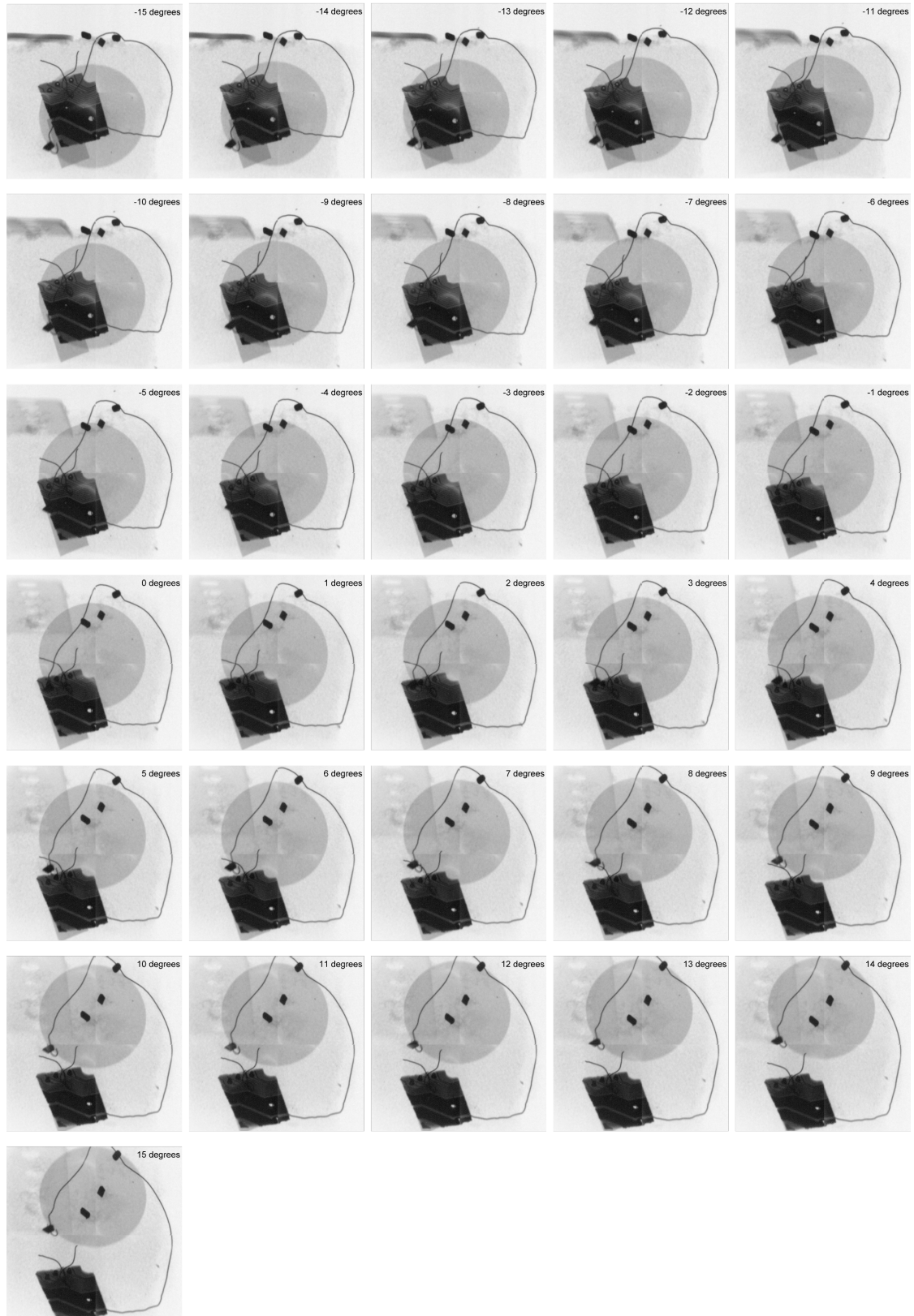


Figure 4.33: *Diagram illustrating the pencil beam interrogation sites (a), the number of matching Bragg maxima (b) and the subsequent colour encoding (c) of the phantom.*

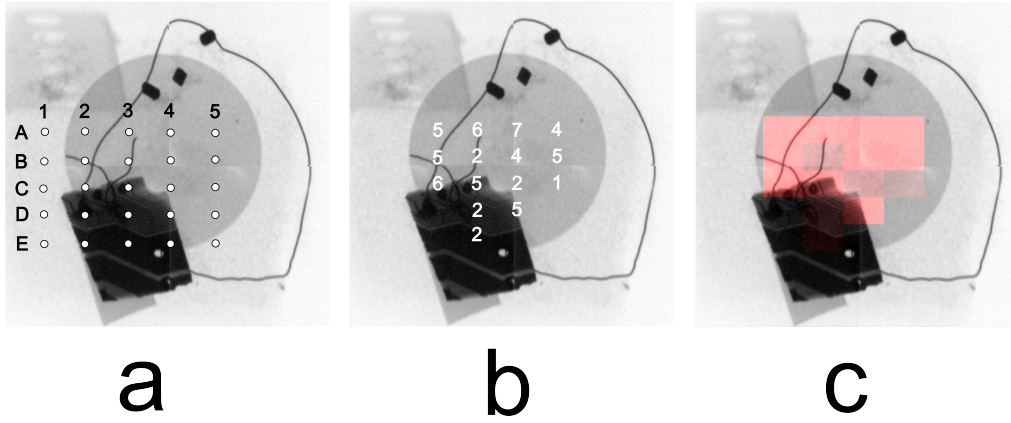


Figure 4.34: *Diffraction patterns obtained from interrogation site A3, also illustrated in figure 4.33 at specified distances along the primary beam path. A additional background subtraction has been applied to remove the high background signature at low H values. Please note each pattern is normalised to 1 and a positive offset has been included for clarity.*

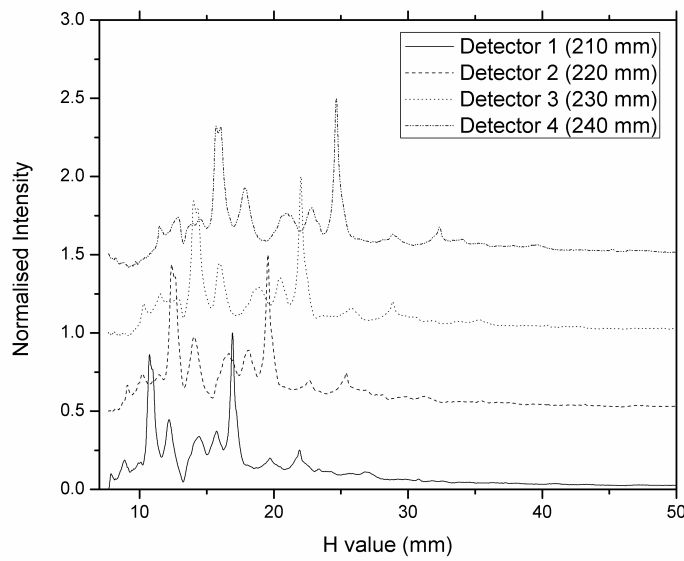


Figure 4.35: Plot illustrating a group of Bragg maxima tracked through the diffraction patterns illustrated in figure 4.34 from position A3 that share a common (± 1.5 mm) location on the primary beam path. The most likely candidate material (HMX) is also illustrated. Please note each pattern is normalised to 1 and a positive offset has been included for clarity.

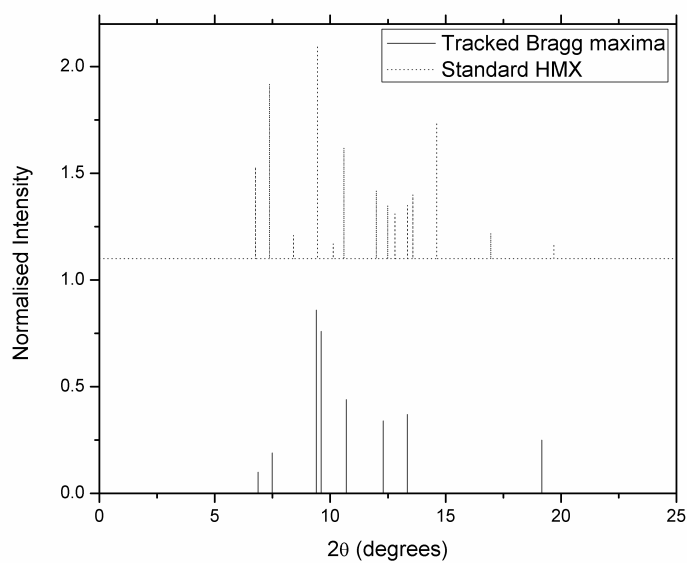


Table 4.3: *Comparison of the interrogation sites illustrated in figure 4.33 with the number of matching Bragg maxima for HMX as well as an average of their calculated locus position from the X-ray source.*

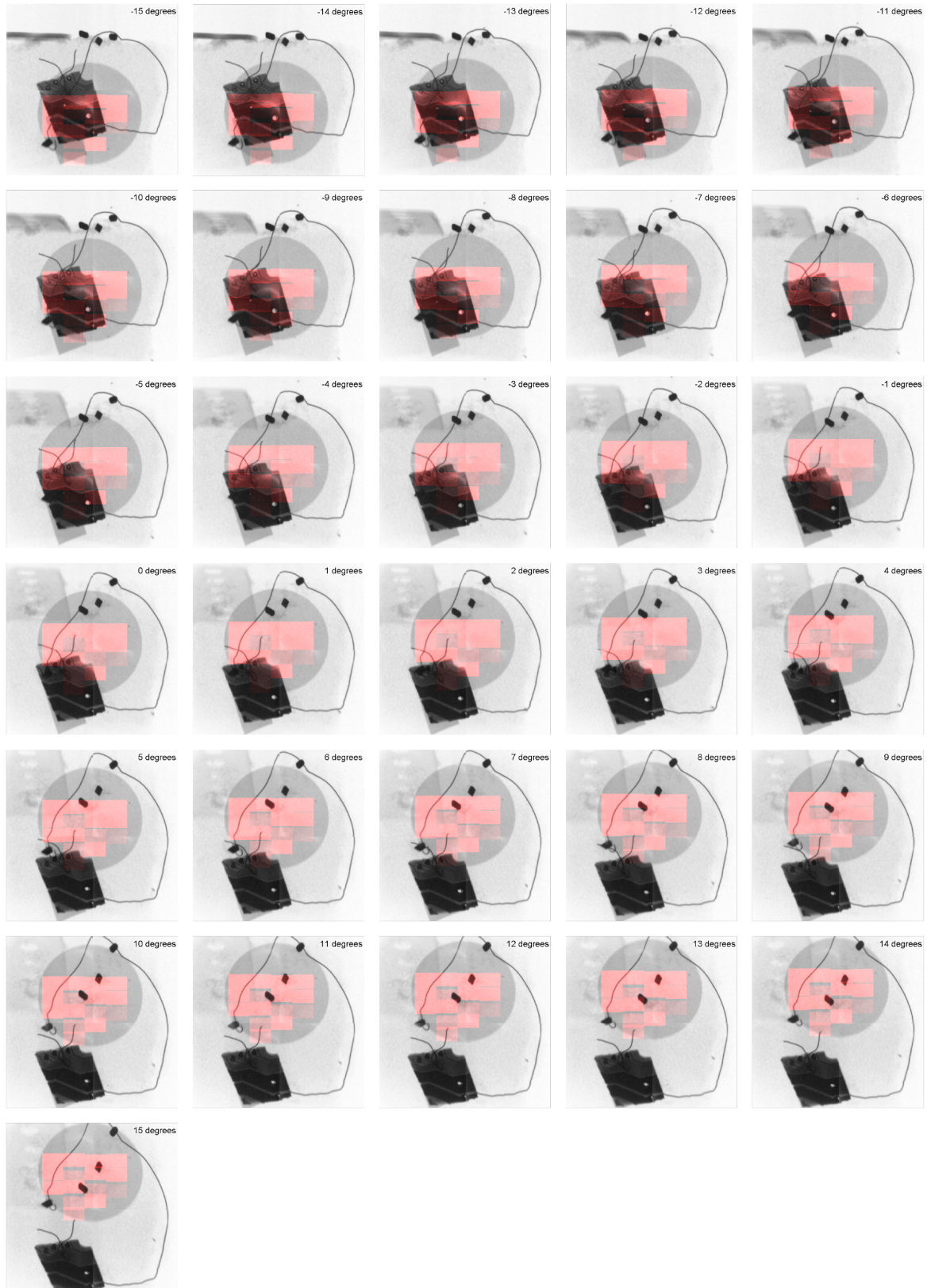
Interrogation site coordinate	Number of matching Bragg maxima position's with HMX	Average location of the tracked Bragg maxima (mm from the X-ray source)
A2	5	144.20
A3	6	144.2
A4	7	144.8
A5	4	144.44
B2	5	144.1
B3	2	146.2
B4	4	143.9
B5	5	144.7
C2	6	144.3
C3	5	143.7
C4	2	145.7
C5	1	145.9
D3	2	145.1
D4	5	144.0
E3	2	143.4

Table 4.4: *Comparison of the location of “tagged” regions listed by interrogation site, given in both mm from the X-ray source and pixels.*

Interrogation site coordinate	location in mm from the X-ray source	Location in pixels
A2	144.20	-323
A3	144.19	-337
A4	144.80	-328
A5	144.44	-326
B2	144.09	-332
B3	146.16	-352
B4	143.92	-326
B5	144.69	-322
C2	144.31	-318
C3	143.67	-318
C4	145.71	-344
C5	145.89	-346
D3	145.14	-315
D4	143.97	-326
E3	143.44	-355

± 300 pixels in the Matlab® program. Table 4.4 illustrates the positions of the tagged regions in depth in both mm from the X-ray source and pixels. Equation 3.7 (described in section 3.7) was used to remap the tagged regions (the variable r_1 being the value in pixels). The result is the tagged regions appear to follow the HMX disc through the image sequence. This is best seen in a video format (illustrated in accompanying video 3). However the individual images have been concatenated and are illustrated in figure 4.36. This result is discussed more thoroughly later in section 5.

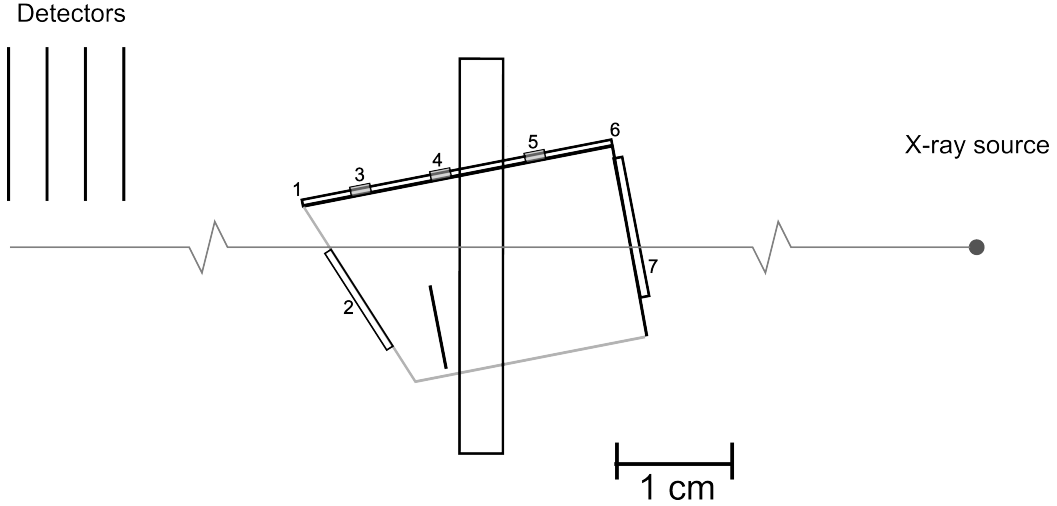
Figure 4.36: *KDEX* image sequence of an improvised explosive device (IED) phantom from $\pm 15^\circ$ rotation (α) with 1° increments. Areas giving rise to materials from a match library have been encoded visually in red.



4.5 Contribution of Object Disparity

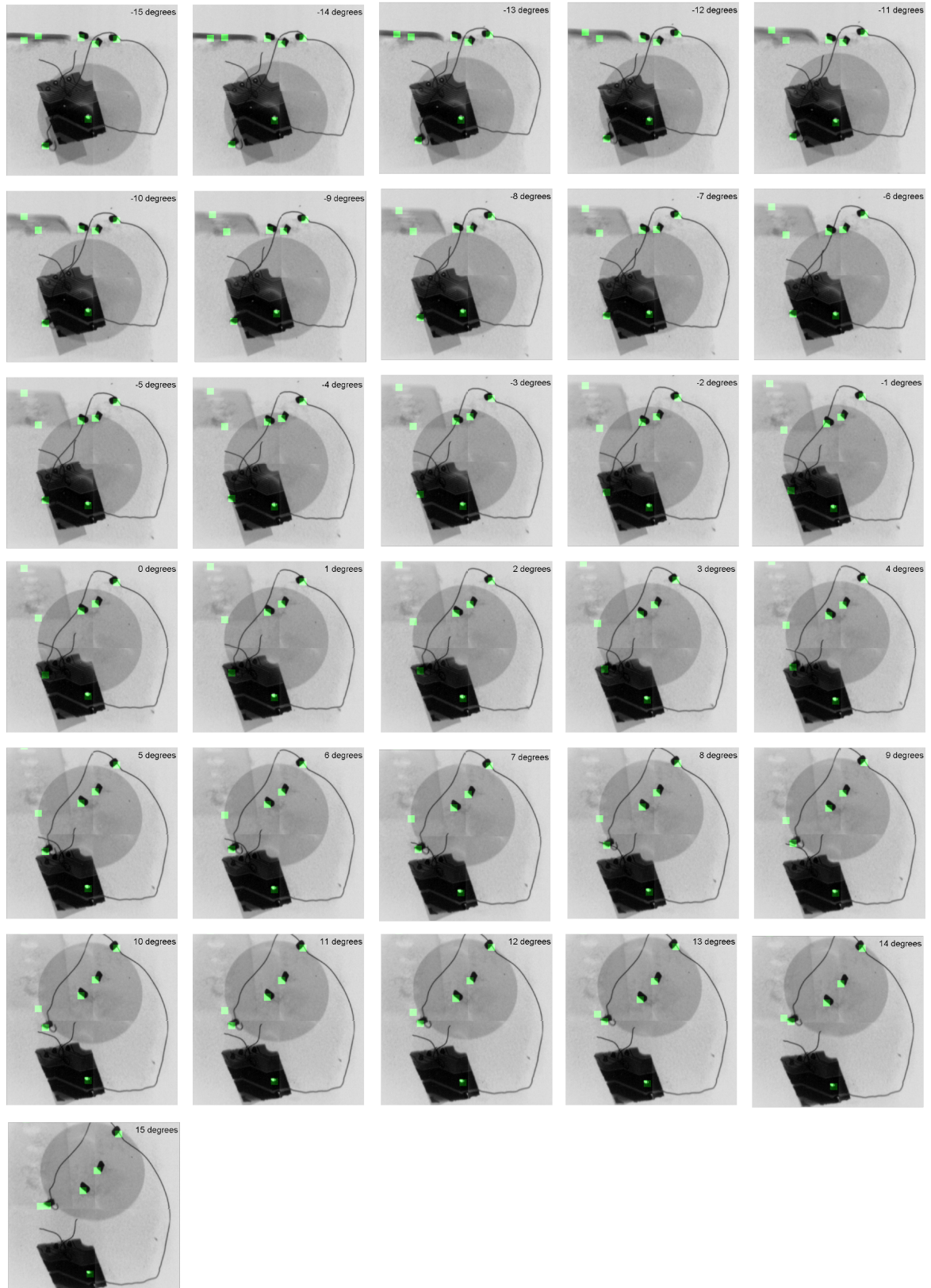
Object disparity (discussed in section 3.8) has the potential ability to inform the $2\theta_{std}$ and sample position permutations produced by the peak tracking method. This is because calculating the disparity of objects within the KDEX image sequence has inherent position information. Figure 4.37 illustrates a schematic of the side view of the HMX phantom (also illustrated in figures 4.30 and 4.32).

Figure 4.37: *Scaled schematic illustrating objects within a phantom at differing positions in depth. Objects are only to scale within the discontinuation boundaries.*



Each of the positions highlighted in figure 4.37 were tracked manually in the KDEX image sequence. Differing r_1 values (in pixels) were evaluated until the colour encoding remapped at the same rate of the objects in depth. These tracked objects and the corresponding colour encoding (in green) are indicated in video 4. The same images that comprise this video have been concatenated and are illustrated in figure 4.38.

Figure 4.38: *Objects within a phantom manually tracked through a KDEX image sequence. Tracked regions are colour encoded in green.*



The r_1 values (in pixels) were used to calculate the locus of objects on the primary beam path relative to the X-ray source using the ruler (apparent in the top right hand corner of the images in figure 4.32). This in effect is the complementary conversion used in section 4.4. The loci of each object (1 - 7) from the X-ray source (illustrated in figure 4.37) are known. A comparison between the calculated loci (based on object disparity in the KDEX image sequence) and the known loci is illustrated in table 4.5. Objects 1 and 6 represent the two ends of the ruler (on which this calculation was based) so there is no discrepancy between the calculated and known loci. The calculated loci and the known loci for the other objects are within 1 mm. The suitability of object disparity to inform the 2θ / location permutations produced by the peak tracking method is discussed in section 5.

Table 4.5: *Comparison between the calculated loci of objects in depth based on object disparity with the known loci. Object positions are illustrated in figure 4.37.*

Object number	r_1 (pixels)	Calculated loci (mm from X-ray source)	Known loci (mm from X-ray source)
1	300	173.5	173.5
2	210	169.4	168.4
3	200	169.0	168.6
4	50	162.2	161.6
5	-130	154.1	153.5
6	-300	146.5	146.5
7	-330	145.1	144.3

Chapter 5

Discussion

The chapter begins with a review of the current gaps in knowledge highlighted in chapters 1 and 2. The thesis aims and objectives are re-iterated. The performance and feasibility of the differing scattering sensor geometries presented in chapters 3 and 4 are discussed. Confounding factors highlighted through computer simulation in the results section are addressed and a possible means of mitigation is proposed. The impact of dynamic transmission X-ray imagery with material specific colour encoding is then discussed.

Recently 3 dimensional imaging modalities that can provide rapid morphological assessment of objects *in-situ* have been subject to significant development. This is perhaps due to the increasing number of applications (including medicine, security screening and material science). However, most applications also wish to establish

the presence (or distribution) of chemically specific “targets” within interrogated volumes. Developments (although arguably to a lesser extent) have also been made in the chemically specific counterparts [16]. However no one technique currently dominates the field. The majority of these chemically specific counterparts identify elemental signatures (e.g. X-ray fluorescence [6, 71, 94], dual energy imaging [37, 43], neutron technology[132, 133, 134]), which make them fundamentally ill equipped for identifying the majority of the materials of interest (e.g. cancerous tissues, explosives, illicit drugs) as they often share similar elemental compositions and densities with benign materials [134, 26].

X-ray diffraction makes its identification based on the “targets” molecular structure. This makes it more appealing as it provides significantly greater specificity [43]. Highly sophisticated angular dispersive diffraction imaging systems are already in operation as a research tool that have successfully combined X-ray diffraction with imaging[119, 120, 8, 121, 122, 123, 124]. However these typically interrogate volumes that are an order of magnitude smaller than the ones investigated here and also employ synchrotron radiation (section 2.1.4) which may have limited wide-scale application.

Tomographic energy dispersive diffraction imaging (TEDDI) [15, 118] is arguably the most advanced imaging system in this field. However, although it benefits from relatively fast data acquisition at high energies it currently lacks comprehensive material specificity. This is perhaps due to the cadmium zinc telluride detectors

currently being employed that afford good detection efficiency at high energies but at the expense of energy resolution[103] although this is certain to improve.

We have investigated augmenting the (more) material specific angular dispersive X-ray diffraction [93, 20, 97] with kinetic depth effect X-ray (KDEX) imaging (an efficient 3 dimensional transmission X-ray imaging modality). This prospect is conceptually straightforward as the X-ray interactions that underpin these techniques occur simultaneously. However in actuality numerous considerations have had to be made. The most pressing issue investigated in this thesis is that of sample position. Powder diffraction systems typically employ two sets of intersecting collimators (i.e. source and detector) whereby the sample resides within the intersecting volume. The angle of scatter between the interrogating beam and the sample (2θ) enables the d-spacing to be calculated (and subsequent material identification). KDEX provides volumetric information. Repeating the diffraction procedure for a volume (by adjusting the intersecting volume of the collimator's to inspect a new location) is not practicable. In contrast we have evaluated an entire dimension illuminated by a pencil beam simultaneously. To achieve this “open collimation” has been used.

5.1 Open Collimation

Open collimation involves the use of source collimation (to achieve an interrogating pencil beam) but no detector collimation. This permits diffracted radiation originat-

ing from any locus on the primary beam path to strike the detector. The use of such a system has been explored in section 4.1. Providing suitable source collimation is used to produce a narrow interrogating primary beam, diffraction patterns can be obtained with no detector collimation that have the same resolution (i.e. FWHM) and signal to noise ratio as those with detector collimation. The important point to note when using an “open collimation” system is the source of diffraction may be positioned on the primary beam path (i.e. z-axis). If the relationship between the interrogating beam direction, sample position and detector position is unknown then the correct angles of diffraction cannot be calculated.

5.2 Circumferential Detector Configuration

A circumferential detector configuration was explored. Circumferential detector configurations (illustrated in figure 3.2) are widespread and routinely used for traditional powder diffraction experiments. This choice was prudent as it required little equipment engineering. A rationale for identifying patterns that have been moved (or displaced) to an unknown locus on the primary beam path has been hypothesised (in section 3.3) and demonstrated (in section 4.2). This was achieved by transforming (or re-calculating) diffraction patterns from a material library to all possible loci on the primary beam path. Each of these transformed patterns were then systematically compared to the unknown material pattern (with unknown locus) to find a match. The criterion for a match was based on concordant d-spacings

(i.e. Bragg maxima positions, 2θ) and not intensity. This single criterion is frequently used throughout this document. Relative intensities of Bragg maxima are conventionally used to aid in a phase identification. However conventional diffraction patterns are obtained from a well defined system. Preferred orientation effects or large grain sizes can be mitigated against by rotating the sample (thus increasing particle statistics). Intensity corrections can also be put in place for the differences in sample to detector distance for differing diffraction angles providing the location of the sample is known. The same corrections cannot be achieved for the *in-situ* analysis studied here. An additional complication that affects Bragg maxima intensity can be attributed to overlapping maxima from differing samples (overlapped on the primary beam path). This additional complication is discussed later.

The success of the circumferential detector configuration is best illustrated in figure 4.4 where both the material and locus of an unknown sample has been systematically identified. This approach was developed further to resolve diffraction patterns produced by multiple unknown materials at differing loci overlapped in depth on the primary beam path (section 4.2.2). Multiple diffraction patterns were simulated (and experimentally verified, illustrated in figures 4.6 and 4.7) after the inspection volume had been rotated (by an amount α). The manner with which the observed Bragg maxima positions ($2\theta_{obs}$) become modified with rotation was used to differentiate Bragg maxima positions originating from differing loci on the primary beam path. Two methods, an empirical and analytical approach were investigated (their method of operation are described in sections 3.3.2 and 4.2.2).

Both the empirical and analytical approach provide reasonable r_1 predictions (i.e within 10%). However there are subtle variations in their predictions. The analytical approach generally underestimates r_1 in all cases (i.e for both differing amounts of displacement and 2θ). The predictions also degrade for increasing $2\theta_{std}$, this can be attributed to the small angle approximation for sine (discussed later) which cannot reasonably be mitigated against (as $2\theta_{std}$ is unknown). The empirical approach generally shares the same trends as the analytical approach, but additional variation is offered by the polynomial fit (equation 4.1, section 4.2.2). The accuracy of the polynomial fit varies with the sample location (r_1) (illustrated in figure 4.12, section 4.2.2) and consequently predictions are improved for samples located closer to the rotation axis (i.e 2 mm rather than 12 mm). In the examples demonstrated here both approaches arguably achieve the goal of separating the Bragg maxima into those emerging from differing loci (to ~ 1 mm). However because their modes of operation are quite different they both present practical limitations.

The analytical approach in principle only requires two diffraction patterns (0 and $45^\circ \alpha$ in the examples shown here) as opposed to 46 diffraction patterns (0- $45^\circ \alpha$) for the empirical approach. The empirical approach therefore is inherently inefficient requiring considerably more information to produce a similar, (and arguably) more ambiguous result. The manner with which 45 diffraction patterns (from presumably 45 scatter sensors) may be obtained simultaneously (e.g. on a conveyor belt for security screening) also presents obvious practical considerations. From a physical point of view (i.e. physically arranging 45 scatter sensors in an appropriate configuration

around an item of interest within the confines of a radiation proof housing) as well as the associated equipment costs. The analytical approach therefore would seem more practicable, but this approach does little to account for the correspondence problem, referred to as “peak tracking” within this document. This limitation is one of the main shortcomings of this technique (although there are others, discussed later). To calculate r_1 via the analytical approach (and the empirical approach) the same Bragg maxima must be identified in each perspective diffractogram (0 and $45^\circ\alpha$). This has not been resolved for the circumferential configuration. However the empirical approach is arguably closer as the angular separation between the perspective diffractograms is smaller (1° rather than 45°). As a result the position modification ($2\theta_{obs}$ with α) is also smaller and so the Bragg maxima position modification (theoretically) be easier to track.

This method has been experimentally validated in section 4.2 and the premise has been subject to peer review [126], although there are some fundamental limitations. The manner with which the Bragg maxima positions ($2\theta_{obs}$) from a mixed spectrum are differentiated and attributed to differing loci (described in section 4.2.2) makes three assumptions. The first limitation is of the correspondence problem. This limitation has been discussed previously and a potential solution for which is discussed later via the linear configuration. The second limitation can be attributed to the small angle approximation for sine. For small angles $\sin(x)$ tends to x . For its application here the angle (x) is $2\theta_{std}$. When applied practically this approximation begins to deteriorate when $2\theta_{std}$ exceeds 30° ultimately impacting on the success of

the multiple pattern separation method. The small angle approximation has not impeded the performance of the experiments demonstrated here. This is largely because the experiments that have been demonstrated were obtained with molybdenum $K\alpha$ radiation. This resulted in the majority of materials producing Bragg maxima positions between ~ 5 - 30° . This approximation would be more problematic if the principle radiation was copper $K\alpha$.

The equivalent molybdenum d-spacing diffracted by copper radiation would be ~ 10 - 80° . At these angles the small angle approximation would deteriorate. However, the applications that have been envisaged for this technique (highlighted in section 1.2) are likely to require higher energy X-rays (e.g. tungsten $K\alpha$ used for security screening and/or medicine). Higher energy X-rays produce lower angle Bragg maxima for the same d-spacings. The use of higher energy X-rays have been explored and are discussed later. Another point to note is that the materials relevant to these same applications (e.g. explosives and biological materials) typically have larger d-spacings anyway (and hence produce comparatively low angle Bragg maxima). This means that the small angle approximation (although a fundamental flaw) does not reveal itself practically in its use here. The third limitation is the requirement for the samples to be planar. For equation 3.3 (section 3.3.2), the basis for many equations in this configuration to be true the samples (located at differing loci along the primary beam path) must be initially normal to the primary beam and planar (similarly to that depicted in figure 3.5). This is a significant oversight of this method as the majority of materials (with the exception of perhaps sheet explosives) will not

conform to this shape. Even if the sample materials were linear this problem is not completely resolved. This is because the equivalent points on each material are not represented in each perspective diffractogram (i.e the interrogating beam intersects differing points on each material for each perspective diffractogram). If preferred orientation is present within the (linear) sample materials then this will exhibit itself when the sample is rotated. This is illustrated by comparing the experimental data illustrated in figures 4.6 and 4.7 for the extruded aluminum samples.

5.3 Linear Detector Configuration

The flaws identified for the circumferential configuration have been resolved (at least in part) by the linear configuration. This rationale has been described in section 3.4, the results have also been verified in section 4.2. The rate of divergence of the H values is a linear relationship and in principle provides the sample locus and $2\theta_{std}$ simultaneously without the use of any approximations such as those utilised by the circumferential configuration. This is advantageous because the calculated $2\theta_{std}$ values can be compared against the material library directly, without the need to systematically transform the entire material library (cross-referencing with the unknown) to subsequently obtain a phase identification. The empirical and analytical determination of the sample locus (discussed previously as part of the circumferential configuration) has helped mitigate against this issue, because they both reduce the distance over which the material library has to be systematically transformed.

However, the number of diffraction patterns in the material library should be taken into account before this (computational step) can be considered negligible. The vast majority of the examples shown here utilise a material library containing ~ 15 diffraction patterns. Systematically altering such a small number of patterns is not particularly computationally demanding. However if an extreme example is taken, it would not be practicable to repeat such a step for all the diffraction patterns in something like Crystallographic Search-Match® (which contains $\sim 200,000$ diffraction patterns). We can therefore infer that removing the necessity to systematically transform the material library is preferred. The linear diffraction configuration also provides the sample locus simultaneously. This resolves the issue of separating multiple material diffraction patterns. Importantly this differentiation is achieved through trigonometry and does not require approximations (used by the circumferential configuration) such as the small angle approximation for sine or the Taylor series expansion of $\cos(x)$. In practice this means that providing the Bragg maxima positions (at each detector position) can be identified (which is not always the case but discussed later) then there is no requirement for $2\theta_{std}$ to be below 30° (i.e. lower angles being more accurately approximated). Therefore this approach could conceivably be implemented with copper incident radiation.

Another significant advantage is that the sample materials do not necessarily have to be arranged normal and planar to the interrogating pencil beam. The linear configuration actually compares equivalent points on any sample materials that are irradiated along the primary beam path (i.e the same locations on any irradiated

materials contribute to the diffractograms obtained at differing detector distances). This means that there is no size or orientation restriction placed on the sample materials. Also this means that should the sample(s) have preferred orientation, the phenomenon will not result in a noticeable relative change in Bragg maxima amplitude (between the diffractograms) as the equivalent portions of the Debye cones (from each sample) strike the detectors. A change in amplitude will be noticed because of the inverse square law and increased air absorption (given the detectors are located at increasing distances from the source) but this can be removed by normalising the patterns with respect to the highest intensity Bragg maxima.

For the linear configuration to correctly determine both 2θ and the sample locus the same Bragg maxima must be identified in each diffractogram. Unfortunately there is no coincidence information between the same Bragg maxima in each diffractogram. At first it may seem prudent to use intensity to identify corresponding Bragg maxima, particularly given the way that preferred orientation does not manifest itself in the diffraction patterns using this configuration. However intensity is unreliable given the likelihood of overlapping peaks. The open collimation system permits scatter signatures from multiple materials to strike the detector simultaneously. This overlapping peak phenomenon is discussed later. For this reason intensity is an unreliable method of identifying corresponding Bragg maxima. Instead, a method of “peak tracking” was developed (described in section 3.4.1) that only uses Bragg maxima position. Bragg maxima are obtained from multiple detector positions (in the examples shown here four). A single Bragg maxima position from each detector

is tested for linearity. This is repeated for all permutations to find all Bragg maxima positions that appear to propagate linearly within a tolerance (~ 0.15 mm). This tolerance was applied to account for any ambiguity in Bragg maxima position offered by calibration issues, noise and change in position offered by the peak overlapping phenomenon.

Justifying the number of diffraction patterns that are required (and hence the number of detector positions) for the linear configuration is difficult to rationalise. In principle only two detector positions are necessary to calculate 2θ and the sample locus. However with no coincidence information cross-referencing every Bragg maxima (n) from the two detector positions results in anywhere from $(n(n+1)/2)$ to n^2 Bragg maxima / sample loci permutations. The “peak tracking” method is based on testing for linearity. Three detector positions should be sufficient to uniquely identify corresponding Bragg maxima positions based on linearity. In fact three detector positions are used by the 3DXRD system [8] (section 2.5.4). However it was found when a tolerance is applied this is not necessarily the case. Figure 4.13 illustrates the number of Bragg maxima positions / sample loci permutations (expressed as a proportion of the Bragg maxima positions (n) in the primary data). Increasing the number of detector positions reduces the number of permutations. The introduction of the third detector position results in the largest reduction in these permutations and is therefore perhaps the most efficient number of detectors to use. We have chosen to include a fourth detector position throughout these experiments. This additional detector position removes anomalous permutations that both evade the

boundary conditions (described in section 3.4.1) and pass the linearity test due to the tolerance that was put in place. Four detector positions is not necessarily the “optimum” number to use for all experiments. Increasing the number of detector positions reduces the likelihood that false Bragg maxima / sample loci permutations will evade the boundary conditions and the linearity test. This reduces false positives / false alarms. However, this is a trade-off as including additional detector positions also increases the likelihood that peaks will become overlapped (at one of the detector positions). When two peaks overlap (such that they cannot be resolved) they may become combined into a single peak that does not truly reflect either of the constituent Bragg maxima positions. This ambiguity (at any one of the detector positions) can cause correct Bragg maxima permutations to fail their linearity test (i.e producing a false negative). Decreasing the tolerance applied to the linearity test will mitigate against this issue but at the risk of increasing false positives. This peak overlapping phenomenon is discussed later for simulations that concern variables such as sample thickness / elongation, detector distances and radiation energy. In summary the introduction of the third detector position contributes to the largest reduction in false Bragg maxima position / sample loci permutations. Including a fourth detector position can be seen as additional refinement. In actuality the “correct” number to use may depend on other constraints such as the type of material(s) that are being evaluated, time / cost constraints and the certainty of the match that is required.

If analysis is being performed on many (>5) materials with high crystal symmetry

or fewer materials with low crystal symmetry then the number of Bragg maxima positions (n) and thus the number of permutations produced will typically be high. This is illustrated in figure 4.13 where the number of permutations from simulations where $n \leq 30$ diminishes at a lower rate with the number of detector positions than where $n \leq 10$. Evaluating scenarios with higher n may require additional detector positions to refine the tracked Bragg maxima / sample loci. Conversely in scenarios concerning lower numbers of Bragg maxima or perhaps where a lower linearity tolerance can be implemented (e.g. for well calibrated, bespoke equipment) the minimum number (i.e 3) of detector positions could be used. Additional detector positions will incur either a higher system cost (owed to the inclusion of additional detectors) or increased analysis time (if a single detector is translated to differing sample to detector distances). The degree of “match certainty” (section 3.7) for the application should also be a consideration. If a high degree of match certainty is required (perhaps owed to a full complement of tracked and concordant Bragg maxima positions) then having a higher number (i.e 5 or 6) of detector positions may prove detrimental. Four detector positions have been used consistently in this study by both computer simulation (discussed next) and experimentally in the laboratory so that fair comparisons can be drawn. However the comprehensive evaluation of differing numbers of detector positions has been highlighted for future work (section 7).

5.4 Sample Thickness, Radiation Energy and Detector Distance

Other factors such as sample thickness / elongation and incident radiation energy have been explored as they have an effect on the angular resolution of the diffraction patterns. The extension of the sample(s) (i.e. sample thickness) along the primary beam path results in broadening of the Bragg maxima (described in section 4.3.1 and illustrated in figure 4.17) when obtained in transmission mode with an open collimation system. This broadening generates ambiguity in the Bragg maxima position and increases the overlapping likelihood. However it should be noted that sample thickness is also detrimental to some energy dispersive systems [84]. Some Bragg maxima broadening can be tolerated providing individual Bragg maxima positions can be resolved. The degree of broadening is dependent on the sample locus to detector distance and the angle of scatter (2θ). This factor was evaluated empirically (described in detail in section 3.5.3) to assess the capability of the linear configuration through the production of ROC curves. The ROC curves evaluated the configuration's ability to discern between illicit and benign materials at differing degrees of elongation. In the first instance two materials (from the material library illustrated in table 3.1) were randomly assigned a position on the primary beam path (by random number generator) and projected onto detectors (at specified distances).

Figure 4.17 (and table 4.2) illustrate that good (i.e. 0.98 out of 1) performance can

be achieved for thin (i.e. 0.1 mm thick) samples. However when the samples are extended along the primary beam path the performance deteriorates (i.e. 0.65 out of 1 for 3.5 mm thick samples) nearing the no discrimination line (i.e. 0.5). The peak width can be expressed by equation 5.1 if it is assumed that beam divergence is negligible.

$$width = elongation(\tan 2\theta) \quad (5.1)$$

Increasing the elongation increases the peak width thus creating greater ambiguity in the Bragg maxima positions and increasing the likelihood of the peak overlapping phenomenon. These collectively impede the success of the peak tracking algorithm. Increasing the sample to detector distance(s) was explored as a possible means of improving the peak tracking algorithm for increased sample thickness (section 4.3.2). To give some perspective the peak width described by equation 5.1 should be expressed relative to its footprint at the detector. The minimum length of the detector (s) is nominally described by equation 5.2

$$s = P(\tan \phi) \quad (5.2)$$

where P is sample to detector distance (P) and ϕ is the greatest scattering angle that can be anticipated. Increasing the sample to detector distance (P) will warrant

an increase s . However the extremes of the elongated diffracted radiation (from the front and back surfaces of the sample) are nominally parallel, thus increasing the sample to detector distance effectively increases the angular resolution of the system. Elongated samples therefore, impede the peak tracking algorithm to a lesser extent at increased sample to detector distances (illustrated in figure 4.22). This can be expressed quantitatively by comparing the volumes (out of 100%) bounded by the surfaces for 210, 220, 230 and 240 mm with those from 220, 240, 260 and 280 mm from the X-ray source illustrated in figures 4.21 and 4.22. These volumes are 86% and 91%, respectively. However, it should be noted that the benefits of increasing the sample to detector distance are not without cost. Increasing the sample to detector distance will result in a lower scattering intensity measured at the detector due to the inverse square law and increased air absorption. The detector(s) will need to be either more sensitive or exposed for a longer time period. The requirement for the active area to be larger will also require additional cost.

The effect of increasing radiation energy (from molybdenum used in the laboratory to tungsten) was also explored. The simulations (described in section 3.5.3) for molybdenum were repeated for tungsten radiation and are illustrated in figures 4.24 and 4.25. The volumes bounded by their respective surfaces were 79% and 83%. Similarly to the aforementioned simulations the configuration with increased sample to detector distances perform better by relatively the same margin. Section 4.3.3 illustrates that increasing the radiation energy decreases the angular range over which a diffraction pattern propagates. The decrease in performance (79 and 83%

compared to 86 and 91%) can be attributed to this decrease in angular resolution given that the same detector resolution ($10\mu m$ pixel size) was used for all simulations. By evaluating these percentages alone it could be inferred that increasing the radiation energy is detrimental to the success of the linear configuration and although this may be the case by large there are other trends that are of interest. Evaluating the individual areas under the ROC curves (per elongation, illustrated in table 4.2) can provide much more information about these factors that is not necessarily apparent by evaluating their respective volumes. The same data illustrated in table 4.2 has also been illustrated graphically in figure 4.26. Previously highlighted trends such as better performance for increased sample to detector distances and generally poorer performance for greater elongations can still be appreciated. However the gradient of the lines particularly between the molybdenum and tungsten (surfaces a and b, and surfaces c and d respectively) are of interest. Although tungsten demonstrates poorer performance (than molybdenum) at thin (0.1 mm) sample elongations (which is perhaps owed to the decrease in angular resolution yet fixed detector resolution), it is affected by a much lesser extent by increasing sample elongation (i.e. the gradient of the molybdenum lines are steeper than the tungsten lines). Peaks originating from the same sample material (or single locus) will become affected by peak overlapping (discussed previously) by proportionally the same extent whether it is radiated by tungsten or molybdenum radiation when the sample is elongated (i.e Tungsten d-spacings dispersed over a proportionally smaller range, yet also exhibit a proportionally small increase in peak width). How-

ever, the simulations involve multiple sample materials separated on the primary beam path. D-spacings from molybdenum radiation propagate over a greater angular range than tungsten, it can therefore be anticipated that Bragg maxima from differing loci are more likely to interleave at the detector. When these interleaved Bragg maxima (from differing sample loci) are simulated at increased elongations then peak overlapping will become prevalent. This could offer an explanation as to why the performance of the molybdenum simulations at increased elongations deteriorates more readily than those with tungsten.

5.5 Colour Encoding KDEX Imagery

The linear diffraction configuration was chosen as the most appropriate method of incorporating X-ray diffraction information into the KDEX image sequences (the reasoning for which has been discussed previously). The result is the production of dynamic 3D transmission imagery with material specific colour encoding. This is illustrated in section 4.4 and peer review [131]. To demonstrate the benefits of fusing these techniques, the phantom (illustrated in figures 4.30 and 4.31) was carefully considered. The depth perception afforded by KDEX is most prominent when objects are overlapped (with a large separation) on the primary beam path. For this reason sample materials had to be reliably fixed in differing locations in depth (discussed later). Significant restrictions were placed on these (overlapping) sample materials due to equipment limitations. The low intensity X-rays (molybdenum

$K\alpha$) limited the types of materials that could be evaluated (i.e low μ/ρ). The size of the phantom was also restricted by maximum divergence of the X-ray beam. It was ensured that all of the phantom could be held within this X-ray footprint. Although the sample materials had a finite size, the transmission imagery that could be achieved was of a relatively high resolution (owed to a relatively small pixel size, $13 \mu m^2$). To produce high quality image sequences objects that were small, yet detailed (e.g. circuit boards) were incorporated. The sample materials were “fixed” in a suspension medium of low density foam rubber to ensure that that no movement was exhibited when the phantom was imaged at differing orientations (rotations). This foam rubber was chosen because it was largely radio transparent at Mo $K\alpha$ (i.e. the foam is hardly visible in the transmission imagery, figure 4.32) and therefore was not prominent in the KDEX imagery. A control region of this foam rubber was also interrogated with a pencil beam to ensure that it did not provide a scatter signature. Some complications were introduced by using low density foam rubber. Although the material is radio transparent it is light opaque. It also had very little strength / rigidity. This combination of traits made measuring the locations of the sample materials problematic. The locations of the sample materials had to be re-measured when the phantom was attached to the sample holder because it under went compression when very little force was applied (altering the locations of the sample materials). Re-measuring the objects *in-situ* was rather cumbersome because the foam was light opaque. The suspension medium was also highly porous, this meant that the sample materials (crystalline or not) needed to be self support-

ing (i.e. no powders or liquids could be evaluated). These phantom restrictions (i.e. low attenuation materials and small sample size) discussed previously, as well as the need for a suspension medium would not be required if a phantom was evaluated at higher energies. However, should similar experiments need to be conducted with Mo $K\alpha$ then it may be prevalent to find an alternate suspension medium. Scattering, “target materials” (e.g. HMX, Al_2O_3) were placed in the suspension material with favorable orientations (i.e. normal) to the interrogating beam. This ensured that a large area was available for diffraction interrogation whilst sample thickness (known to be problematic) was minimized. Unfavorable orientations (i.e. where the sample is orientated parallel to the primary beam path) may confound the linear diffraction configuration as it is demonstrated here, the reasons for which are twofold. Firstly the sample will be elongated along the primary beam path which has been shown to be detrimental to the peak tracking algorithm (discussed previously) and secondly, it is less likely that the sample will be intersected by an interrogating beam given the inherently poorer spatial resolution of the diffraction grid.

There is a discrepancy between the spatial resolution of the transmission imagery and the diffraction pattern counterpart demonstrated here (section 4.4). Each diffraction image required a 3 minute exposure time. 5 diffraction images were obtained per detector position and 4 detector positions were collected. This resulted in approximately a one hour overhead per interrogation site. The area of the phantom that was interrogated was approximately equal to the footprint of the pencil beam ($\sim 0.34 \text{ mm}^2$, section 3.7). Conversely each KDEX transmission image interrogated

the entire volume. These images required the camera to be raster scanned to 4 positions and each image was exposed for 5 seconds. Therefore each KDEX transmission image had a time overhead of approximately 20 seconds. Obtaining the diffraction patterns can effectively be described as point scanning. Given the time overhead associated with each of these scanned points (i.e interrogation sites) it was not practicable to obtain a diffraction grid at the same spatial resolution as that achieved in the transmission image. Therefore the area that was raster scanned and the frequency of the spacing between the interrogation sites was a trade-off between sensitivity (i.e the likelihood that one of the interrogating pencil beams would intersect a target material) and acquisition time. A bespoke system would potentially mitigate against this trade-off.

For the experiments demonstrated here both the transmitted and diffracted X-rays were collected on the same PIXIS detector (described earlier in section 3.6). The size of the camera's active area as well as its dynamic range meant that it was not practicable to obtain both the transmission and diffracted signals simultaneously, which is arguably more time efficient. Instead each aspect was obtained consecutively with differing exposure times. An idealised system would use detectors with an appropriately size active area. This would remove the requirement to raster scan the camera (to 4 positions for the transmission imagery and 5 positions for the diffraction images used in this study). It would also be prudent to use differing specification detectors, robust sensors (to detect transmitted X-rays) and sensitive sensors (to detect the diffracted X-rays). This may make the exposure times more

comparable. Appropriate collimation (enabling simultaneously measurements to be collected) rather than the need to point scan may also offer increased spatial resolution of the diffraction grid as well as speed of acquisition. However the spatial resolution of the diffraction grid is unlikely to match that of the transmission imagery. Possible collimation configurations that enable measurements to be made in tandem have been illustrated and highlighted for potential future work.

Areas within the phantom giving rise to diffraction patterns of interest were colour encoded in differing intensities of red. Red was chosen because it seemed like an appropriate colour normally associated with a “threat” material (and has been employed by other authors [135, 136]). However in practice any colour could have been chosen. For instance dual-energy imaging systems typically colour encode in blue, orange and green which have little association with the materials they represent (i.e. metallic, organic and inorganic materials respectively). A different colour may well be more appropriate from a human factors perspective, this is beyond the scope of this study and has been highlighted for future work. To colour encode highlighted regions, pixel values in the red channel were then skewed based on the match index whilst the green and blue pixel values were maintained from the original transmission image (and therefore demonstrated absorption). Absorption features in the highlighted regions were still visible but the intensity of red reflected the certainty of match.

Demonstrating the certainty of match (match index) via intensity of red was con-

ducted to provide an example of how additional information from the linear diffraction configuration could be relayed via the KDEX imagery (i.e an example of technique fusion). However, the mathematics behind the match index may require further refinement. Currently each red pixel value in the highlighted regions are multiplied by their original intensity plus a tenth of the number of matching Bragg maxima (i.e if 5 Bragg maxima are attributed to a region the red pixel values are multiplied by 1.5). This particular calculation was able to exploit the dynamic range of the image without saturation. However, it may not be appropriate for all data sets. The maximum pixel value illustrated in figure 4.36 was $\sim 38,000$ and the maximum number of matching Bragg maxima was 7 (illustrated in figure 4.33). Therefore the maximum red intensity that could be achieved with this calculation was $\sim 64,600$ (i.e $38,000 * 1.7$) which is below the saturation value ($\sim 65,500$). Therefore this calculation ensured that the maximum number of matching Bragg maxima exhibited the greatest intensity in red (without saturating the camera), also the dynamic range in intensity (i.e from 38,000 - 65,000) was divided equally among the number of matching Bragg maxima. This ensured that a change in red intensity for differing match index's (e.g. attributed to 5 matching Bragg maxima and 6 matching Bragg maxima) was noticeable in the image. Data sets with a different maximum pixel value or with different numbers of matching Bragg maxima may warrant a more sophisticated calculation to alter the red intensity.

Another point to note is the relationship between the likelihood of match and the match index. It is fair to presume that the greater the number of matching Bragg

maxima the greater the likelihood of match. However the way in which the number of matching Bragg maxima impacts the match index (and therefore intensity) should be considered. In the example demonstrated here the relationship is linear. However, differing materials give rise to differing numbers of Bragg maxima (due to their crystal symmetry). For instance iron has 4 diffraction maxima where as TNT has ~ 30 . If both materials were targeted it may not be prudent to designate the same match index (and therefore increase in red intensity) to two areas, whereby one area has 4 matching Bragg maxima with iron and another has 4 matching Bragg maxima with TNT. This is because arguably the area giving rise to the iron Bragg maxima is more likely to be produced by iron than the area giving rise to Bragg maxima matching TNT. This issue has not presented itself in this study as the Bragg maxima (illustrated in figure 4.33) were all matched to a single material (HMX). For studies with multiple target materials it may be prudent to consider the number of matching Bragg maxima as a proportion of those in the target material before assigning a match index.

Each of the matching Bragg maxima also had an associated sample locus within the inspection volume calculated by the linear diffraction configuration. To successfully remap the highlighted regions for alternate rotations (α) an internal standard was used to relate the sample locus in mm from the source to their equivalent locus in pixels (described in section 3.7 and illustrated in section 4.4). The two ends of an internal standard (a plastic ruler, illustrated in figure 4.31) were used to achieve this. These ends were measured to find their respective loci. Differing pixel values were

used to remap highlighted regions in software until they remapped at the same rate as each end of the internal standard. This relationship permitted sample locus values (expressed as mm from the source) to be converted into pixels (allowing highlighted regions to be remapped) and vice versa (i.e. to infer the sample locus of materials based on their object disparity, discussed later). The important point to note is the crucial role of the internal standard. The internal standard was a plastic ruler with a low attenuation coefficient (and thus was largely transparent in the transmission imagery). This was chosen purposefully so that it could be seen but did not detract from the parallax of other objects in the volume. However, at some orientations the super positioning of the ends of the ruler made identifying their exact location in the image problematic. This ambiguity (although slight) when calibrating the image sequence (from mm to pixels) impinged on the rate at which the highlighted areas (that indicate a threat) remapped. Fortunately this ambiguity has not detrimentally affected the encoded image sequence illustrated in this study (video 3). If aspects of the experiment demonstrated here should be repeated it may be appropriate to use an alternate internal standard that's position(s) can be unequivocally identified in each perspective transmission image.

5.6 Object Disparity

An area of interest that was not comprehensively explored is that of object disparity (described in section 3.8). Up to this point the diffraction information has been

used to inform the transmission information (with regard to threat materials and their locus within the interrogated volume). Object disparity was investigated to, in effect close the loop and have the transmission information inform the diffraction information. Anomalous 2θ /sample locus permutations can be produced by the linear diffraction configuration which can affect the system specificity (discussed previously). A knowledge of the objects locus provided by an alternate means (i.e. KDEX imagery) was investigated to potentially remove anomalous results. Manually tracking known points on objects through the image sequence and calculating their locus is illustrated in section 4.5. Objects were calculated to ± 1 mm of their “known” position via object disparity. This was surprisingly accurate given the same ambiguity offered by the super positioning of the internal standard (discussed previously) had an equivalent complementary affect on calculating the locus of objects based on pixel modification. Another point to note is that measuring the “known” locus of the objects within the inspection volume had an associated error given the light opaque and compression qualities of the suspension medium discussed previously. Should the automatic tracking of objects be perfected then it could be used to complement the diffraction information. This would bring another level of integration between the two techniques. This has been highlighted for future work.

Chapter 6

Conclusion

A new diagnostic technique able to simultaneously provide 3D morphology and spatially resolved material discrimination has been systematically investigated and demonstrated. This system is based on the amalgamation of two established, yet complementary techniques that have previously never been combined. Kinetic depth effect X-ray imaging (KDEX) is a proven and efficient technique able to provide increased morphological recognition (in the presence of complicated volumes) acquired from transmitted X-ray images. Conversely, X-ray diffraction makes no distinction on morphology. Instead it provides chemico-specific phase identification from X-rays that scatter from (crystalline or semi-crystalline) materials intersected by the transmitted beam. The fusion of these techniques demonstrates synergy and the possible applications are widespread (e.g. security screening, medicine, material science). Taking security screening as an example, KDEX provides a much needed imaging modality permitting prohibited yet non-chemically distinct items (e.g. knives

and scissors) to be identified, whilst X-ray diffraction provides the near unequivocal identification of items such as explosives that may have a benign appearance (morphology).

The techniques, although generated from the same source are fundamentally different. X-ray diffraction typically provides information regarding materials placed at a known locus. KDEX typically provides information of entire volumes. As well as these intrinsic geometric differences there is a discrepancy between their respective acquisition times (given the difference in transmitted and scattered X-ray fluxes). Uniting these two techniques has been a significant undertaking.

In attempt to address the differences in geometry and (to a lesser extent) acquisition time, differing scatter sensor geometries utilising open collimation have been evaluated. Open collimation permits scattered radiation emerging from any locus (illuminated by a primary pencil beam) to strike the detector simultaneously without the need for detector collimation (which is typically used to provide the sample locus at the expense of the scattered flux). This has been shown to be a viable alternative to point scanning (i.e with intersecting collimators) although arguably still requires development. Instead of using detector collimation, differing detector geometries (circumferential and linear) have been evaluated that provide the sample locus and angle of scatter (2θ). These configurations have demonstrated differing degrees of success.

The linear configuration can be more readily augmented to KDEX than the cir-

cumferential configuration. The circumferential configuration is reliant upon approximations (e.g. small angle approximation) to resolve multiple (spatially separated) material diffractograms. This introduces varying degrees of ambiguity in the calculated sample locus / scattering angle and places an upper restriction on the scattering angle(s) (and thus radiation energies) that can be evaluated. There is no such limitation associated with the linear configuration. The correspondence problem of identifying common Bragg maxima in differing perspective diffractograms has also been addressed by large by the peak tracking method. Bragg maxima positions propagating linearly with respect to the detector positions is used to identify coincident peaks. The ambiguity associated with calculating the aforementioned scattering angle(s) makes applying a similar method of peak tracking to the circumferential configuration inappropriate. Four detector positions have been used throughout this study and there is sufficient justification provided in the discussion to validate this method. However, four detector positions may not be optimum. In actuality the “correct” number to use may depend on constraints such as the type of material(s) that are being evaluated, time / cost constraints and the certainty of the match that is required. The linear configuration has no restrictions on sample morphology (i.e the necessity for the sample materials to be both linear and normal to the primary beam path for the circumferential configuration) with the exception of perhaps samples with pronounced elongation / thickness along the primary beam path. A side effect of the open collimation is that thick samples (interrogated in transmission) produce broadening in the diffraction maxima. This broadening has

been shown to be detrimental to the peak tracking method. A possible method of mitigating against this broadening, by increasing the sample to detector distance, has been explored. However, this solution would require larger and more sensitive detectors to capture the increased angular spread of the propagating Debye cones which will diminish in intensity owed to increased air absorption and the inverse square law. Physical limitations that are application specific should be considered before deciding if this concept has sufficient merit. Increasing the radiation energy decreases the angular divergence of the diffraction patterns. This is also detrimental to the peak tracking method as it increases the likelihood that Bragg maxima will overlap. The resolution of the detector may also become a limiting factor even if the sample materials are suitably thin.

Material identity inferred from X-ray diffraction has been represented visually within the KDEX transmission imagery via colour encoding. These highlighted region(s) have an associated locus within the inspection volume calculated from the peak tracking method. Threat item(s) in the KDEX imagery can be readily discriminated from benign items overlapped in depth. Once more the certainty of match (match index) is also represented through intensity of red. This is the first time that these two techniques have been fused. Object disparity (i.e depth of locations inferred by evaluating the KDEX imagery) has shown the potential to inform the diffraction information. However, as yet the automatic tracking of objects is in its infancy. Therefore the contribution of object disparity should be investigated more thoroughly in future work.

Chapter 7

Recommendations for Future Work

This project has yielded substantial findings (summarised in the preface). However given the nature of this research there is still significant blue sky thinking that may prove valuable. A comprehensive list of recommendations for future work is given.

7.1 Investigating Intensity

In the simulations conducted in this study detectors with perfect efficiency were hypothesised. This negated the need to evaluate factors that affect intensity. Examples of these factors include preferred orientation, scattering cross sections of the material(s) and re-absorption effects. It may be prudent to conduct simulations that more realistically model existing detectors as well as exploring the impact that these

factors have on peak amplitude. A particular area of interest would be in modeling the intensity response when the detector positions are altered. The proportion of X-rays that are scattered compared to transmitted is notoriously low and so it may also be worth investigating novel configurations that promise to improve diffraction efficiency such as those presented by Evans et al. [138] and Rogers et al. [139].

7.2 Number of Detector Positions

Four detector positions have been used throughout this study to find coincident Bragg maxima that propagate linearly through the detector positions. However, four detector positions may not be optimum. In actuality the “correct” number to use may depend on constraints such as the type of material(s) that are being evaluated, time / cost constraints and the certainty of the match that is required. This relationship should be comprehensively explored in future work.

7.3 Object Disparity

X-ray diffraction has been predominately used to inform the KDEX imagery with regard to material identity and corresponding locus within the inspection volume. This information flow is largely one directional. To make the technique fusion more compelling, KDEX should also inform the X-ray diffraction information. There is

inherent depth information within the KDEX imagery. This has been superficially investigated within this study. Correctly and automatically calculating the object's locus (via its disparity) should be explored as a way of either re-enforcing or refining the information (e.g. sample position) produced by the peak tracking algorithm.

7.4 Human Factors

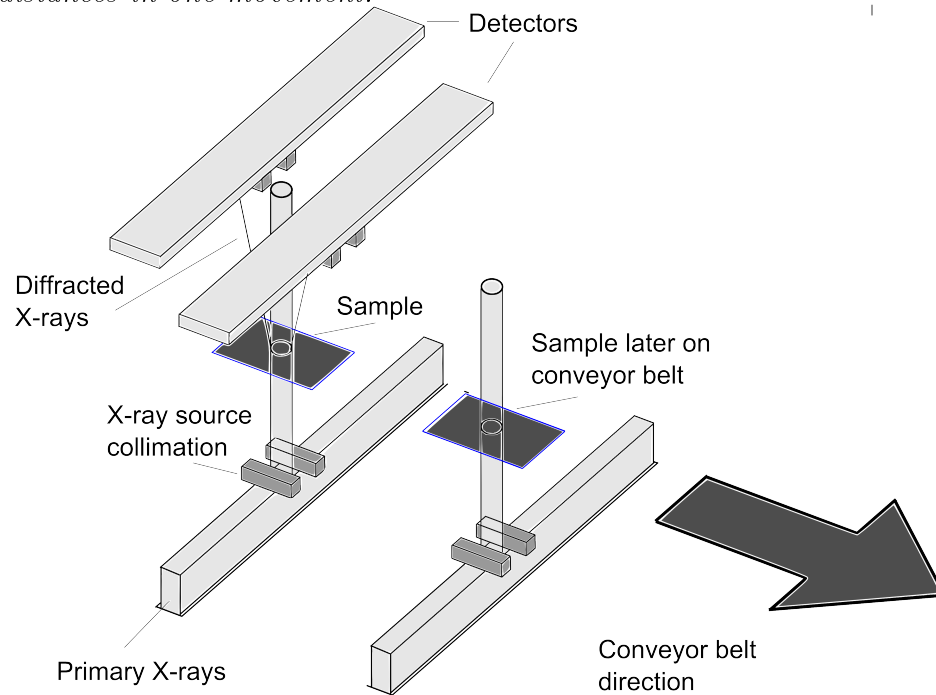
In this study material identity (acquired from X-ray diffraction) has been encoded visually within the KDEX imagery by skewing the red channel of the highlighted region(s). The intensity of the highlighted region(s) also reflects the match index. This is a simplistic method of representing both pieces of information and is not necessarily the best from a human factors perspective. Differing ways of representing the materials ID within the KDEX imagery should be investigated.

7.5 Scale up of Technology

Scaling up the technology illustrated in this study will probably require an increase in radiation energy. Simulations have been conducted here with tungsten radiation (typically used for security screening and medicine). Future work should obtain diffraction patterns with tungsten radiation in the laboratory. The practical limitations of applying the geometries to an application should also be considered.

Figure 7.1 demonstrates a hypothesised configuration utilising the linear approach. The widths of collimators and the interrogating pencil beam has been accentuated to make the illustration more clear. Two perspective diffractograms (from a single depth dimension) can be obtained on the two detectors (situated at differing distances from the sample) by inspecting opposite portions of the propagating Debye cones. Another two perspective diffractograms (thus totaling four) could be acquired by employing a second generator further down the conveyor belt. Two more detectors (not shown) would need to be placed at differing distances from the sample. The speed of the conveyor belt would be known and therefore could be used to calculate when the sample would be equivalently intersecting the second interrogating pencil beam (from the second generator). Four diffraction patterns can thus be acquired from a single depth dimension through the inspection volume and subject to the peak tracking method.

Figure 7.1: A diagram illustrating a hypothesised conveyor belt X-ray screening configuration designed to capture four diffraction patterns at increasing sample to detector distances in one movement.



Chapter 8

Appendix

8.1 Security Screening

8.1.1 Neutron Based Techniques

Neutron based technology has been shown to demonstrate highly desirable characteristics that make it suitable for the detection of unknown materials (e.g. explosives). These include “the ability to penetrate through various materials to large depths” [134], high detection sensitivity [132] and high elemental specificity [134]. In principle this technique involves bombarding the nuclei of the target material with neutrons of a particular energy [25]. This can result in numerous nuclear reactions, the most pertinent being the production of gamma rays [134, 140]. The elemental

components of explosives (typically H, C, N and O) differ strongly in their reactions with neutrons producing characteristic gamma rays [25]. The energies of the emitted gamma rays as well as their relative proportions are used to identify the material. Several different techniques exist that use this principle. Typically the neutron energy used is varied (lower, higher or both) to influence the type of nuclear reaction achieved. This helps to tailor the elemental specificity of the technique. Another point to note is that neutrons are electrically neutral. This means unlike other techniques (e.g. X-rays) they do not interact with electron shells. Some authors perceive this to be beneficial as more interaction is seen with lower atomic numbered materials (e.g. H, C, N and O) rather than high atomic numbered materials (e.g. metals). It is suggested that this lends itself well to identification of materials that are concealed [141].

Neutron based techniques have been shown to be a powerful tool in the identification of explosives which can rival traditional X-ray based methods in specificity and sensitivity. Typical receiver operator characteristic (ROC) curves occupy the upper left quadrant, characterized by a high figure of merit ($FOM > 2.5$) [132]. However this technology is still immature and faces many challenges. Practical constraints of constructing the collimation, cost (for the system and given the significant shielding required), operational safety (given the ionizing effect of neutrons exceeds that of X-rays) and perhaps the most influential, public acceptance limit this technique's appeal. Another point to note is that although many authors including Eberhardt et al. [141] champion the technique's transparency to metals to be a positive point

it is important to note that although explosives themselves are not comprised of metals, many of the components of bombs (including wiring, detonators and timers) do contain metal. Other traditional threats such as knives and guns would be invisible to this technique and so it would not be prudent to replace traditional X-ray methods outright with neutron technology. If the current limitations with these techniques could be resolved then perhaps it would be fit to combine them with traditional methods [142] as an advanced screening stage which would add to both an already exhaustive waiting time and cost.

8.1.2 Terahertz Time Domain Spectroscopy

Terahertz time domain spectroscopy is another radiation based inspection system utilising the $10\text{-}300\text{cm}^{-1}$ range of the electromagnetic spectrum [143]. This region of radiation has proven to be particularly useful because its ionizing capability is considerably lower than current X-ray based techniques (“one million times weaker than X-ray photons”) [40]. It has penetrative capabilities which are “able to propagate through barrier materials such as clothing and common packaging materials” [143] and also perhaps most importantly explosives and other contraband materials have “spectral fingerprints” in the terahertz range [40]. This technique utilises terahertz waves to spectroscopically detect explosives by evaluating their characteristic spectra [16]. Huang et al. [144] has shown that RDX (a high explosive) demonstrates absorption features in the terahertz region in transmission. Arguably the

main limitation of terahertz spectroscopy in transmission mode is that real world application measurements cannot be collected. THz radiation is able to penetrate some non-polar and nonmetallic materials [144]. However its penetrative capability is somewhat inferior to that of higher energy sources such as X-rays and neutrons. If a bulky target were examined the THz radiation is likely to be attenuated completely [40]. To mitigate against this issue efforts have been made to measure THz radiation in reflection (from the outer surface of the object) instead. This does not increase the penetrative capabilities of the technique but merely allow some signal to be measured if a high attenuating material resides within the inspection volume. There are different types of reflection techniques namely Specular Fresnel Reflection [143] and Diffuse Fresnel Reflection [40] each of which have differing positives but which share similar flaws. Terahertz is sensitive to the materials effect on both amplitude and phase. A detailed discussion regarding these can be found in Yinon [16]. The important thing to note is that both are required to correctly identify a material. Obtaining the phase information in reflection has been shown to be problematic. Typically the detector must be placed in a specific location depending on the location of the source and orientation of the sample surface [40]. In a real world environment the orientation of the sample surface will not be known. An accurate phase measurement also requires a reference reflector in the exact same position as the target and to add further complication it should share a similar surface morphology [40]. Another inherent problem of THz is its affinity for water. If water vapour is present during the analysis of RDX then majority of the characteristic peaks are

lost. This problem means that most systems are flushed with nitrogen which itself has practical considerations [40].

Terahertz time domain spectroscopy demonstrates a lot of promise for the detection of explosives in a screening environment. It has inherent benefits that distinguish it from traditional methods none more so than the fact that the “low photon energies that it uses will not cause harmful photon-ionization in biological tissues” [40]. The use of ionizing radiations in current screening techniques make them unpopular. This appears to be a trade off for penetrating capability. It has been made clear that THz in transmission is not sufficient for screening. This area is now reliant on the development of reflection modes but this is still in its infancy. Liu et al. [40] shows that identifying RDX in reflection with common barrier materials is problematic. Rough surfaces and inhomogeneous thickness’s cause distorted reflection spectra making the identification of the explosive near impossible. For this reason it makes the implementation of this technique in an airport screening environment difficult to imagine. The high absorption coefficient of water in this region also raises extra cause for concern as dealing with this problem in the laboratory (by flushing the system with nitrogen) is relatively rudimentary but a lot more costly and bothersome if it has to be done online.

8.1.3 Nuclear Quadrupole Resonance

Nuclear Quadrupole Resonance (NQR) is a promising technology because of its unique and discriminating probing capabilities of chemical matter (e.g. explosives) [145, 146]. It is relatively inexpensive when compared with other techniques (particularly neutron technology) and it provides a non-destructive and viable alternative [147]. The radiation that is used is deemed to be non-ionizing which means that there are no additional safety concerns. In principle the technique pulses a gradient of radio frequencies to excite the target (in this circumstance illicit materials) [16]. When the target falls from this excited state the same energy radio frequency is emitted. The energy detected is distinctive [148]. This energy reflects the local arrangement of electronic charge distributions around the nucleus under study which in turn depends not only on the nature of the nucleus (atomic number) but also its chemical environment (molecular structure) [147]. An inherent benefit of NQR that has been noted by a number of authors is its sensitivity to nitrogen. Nitrogen is a key elemental constituent of high explosives and nitrogen¹⁴ one of the more common isotopes exhibits a relatively large internal electric-field gradient due to its non-spherical charge distribution. This in turn lends itself well to identification by NQR [147]. Also because this technique is sensitive to the chemical environment that the species is present in it is also able to detect dangerous explosive compounds from innocuous materials which is a weakness of Dual energy X-ray systems (discussed in section 2.5.1) .

The main problem with this technique is that the discriminative radiation that it detects is of a very low frequency which is comparable to thermal noise. This consequently means that a considerable effort has to be made to extract a reliable signal from background noise [147]. Radio frequency (RF) interference from transmitters or local electronic devices can also produce electromagnetic noise which cloud the signal [147]. This means that extensive shielding may again need to be employed but in this circumstance it is not to protect the public from radiation (e.g. such as neutron based detection systems described in section 8.1.1) but to protect the NQR sensors from environmental electromagnetic noise. Interference artifacts can also be created intrinsically in the form of “magneto-acoustic ringing” [147]. This phenomenon is a result of “electrical reverberations on the probe coil” from the initial RF pulse (of a relative high power compared to the emission pulse) used to excite the sample [147]. These reverberations only occur for a short amount of time before they decay to negligible levels. However some materials (e.g. TNT) also share this short decay time [147]. The a fore mentioned issues coupled with the fact the general requirement for the samples to contain many grams or tens of grams of the compound of interest (e.g. explosive) before it will be detected limit the scope of this techniques appeal.

8.2 Removing Variation Caused by $2\theta_{std}$

To resolve diffraction patterns originating from multiple unknown spatially separated materials such as those described in section 3.3.2 it is important that the contribution in peak modification caused by $2\theta_{std}$ is isolated and removed. The first step is to rearrange equation 3.2 in terms of r_1 :

$$2\theta_{obs} = 2\theta_{std} - \sin^{-1} \left[\frac{r_1 \sin(\pi - 2\theta_{std})}{R} \right]$$

$$2\theta_{obs} - 2\theta_{std} = -\sin^{-1} \left[\frac{r_1 \sin(\pi - 2\theta_{std})}{R} \right]$$

$$2\theta_{std} - 2\theta_{obs} = \sin^{-1} \left[\frac{r_1 \sin(\pi - 2\theta_{std})}{R} \right]$$

$$\sin(2\theta_{std} - 2\theta_{obs}) = \frac{r_1 \sin(\pi - 2\theta_{std})}{R}$$

$$R \sin (2\theta_{std} - 2\theta_{obs}) = r_1 \sin (\pi - 2\theta_{std})$$

$$r_1 = \frac{R \sin (2\theta_{std} - 2\theta_{obs})}{\sin (\pi - 2\theta_{std})}$$

If the angles of θ (in radians) is said to be small the small angle approximation for sine can be used. Let $\sin x = x$ and $\sin \pi = 0$.

Therefore:

$$r_1 = \frac{R(2\theta_{std} - 2\theta_{obs})}{2\theta_{std}}$$

$$r_1 = \frac{R2\theta_{std}}{2\theta_{std}} - \frac{R2\theta_{obs}}{2\theta_{std}}$$

$$r_1 = R - \frac{R2\theta_{obs}}{2\theta_{std}}$$

This can be rearranged to give:

$$\frac{2\theta_{obs}}{2\theta_{std}} = \frac{R - r_1}{R} \quad (8.1)$$

When the sample has been subject to a global rotation of α equation 3.2 is altered.

Here $2\theta_{obs2}$ represents the same observed peak as $2\theta_{obs}$ accept its position has been modified by a global rotation of α This can also be rearranged in terms r_1 :

$$2\theta_{obs2} = 2\theta_{std} - \sin^{-1} \left[\frac{r_1 \sin(\pi - 2\theta_{std})}{R \cos \alpha} \right]$$

$$2\theta_{obs2} - 2\theta_{std} = -\sin^{-1} \left[\frac{r_1 \sin(\pi - 2\theta_{std})}{R \cos \alpha} \right]$$

$$2\theta_{std} - 2\theta_{obs2} = \sin^{-1} \left[\frac{r_1 \sin(\pi - 2\theta_{std})}{R \cos \alpha} \right]$$

$$\sin (2\theta_{std} - 2\theta_{obs2}) = \frac{r_1 \sin (\pi - 2\theta_{std})}{R \cos \alpha}$$

$$R \cos \alpha \sin (2\theta_{std} - 2\theta_{obs2}) = r_1 \sin (\pi - 2\theta_{std})$$

$$r_1 = \frac{R \cos \alpha \sin (2\theta_{std} - 2\theta_{obs2})}{\sin (\pi - 2\theta_{std})}$$

If the angles of θ (in radians) is said to be small the small angle approximation for sine can be used. Let $\sin x = x$ and $\sin \pi = 0$.

Therefore:

$$r_1 = \frac{R \cos \alpha (2\theta_{std} - 2\theta_{obs2})}{2\theta_{std}}$$

$$r_1 = \frac{R \cos \alpha 2\theta_{std}}{2\theta_{std}} - \frac{R \cos \alpha 2\theta_{obs2}}{2\theta_{std}}$$

$$r_1 = R \cos \alpha - \frac{R \cos \alpha 2\theta_{obs2}}{2\theta_{std}}$$

This can be rearranged to give:

$$\frac{2\theta_{obs2}}{2\theta_{std}} = \frac{R \cos \alpha - r_1}{R \cos \alpha} \quad (8.2)$$

Equation 8.1 and 8.2 represent rearrangements of the analytical forms of $2\theta_{obs}$ and $2\theta_{obs2}$ respectively. Dividing these analytical forms results in the $2\theta_{std}$ term to cancel. This is demonstrated in equation 8.3.

$$\frac{2\theta_{obs2}}{2\theta_{obs}} = \frac{2\theta_{obs2}}{2\theta_{std}} / \frac{2\theta_{obs}}{2\theta_{std}}$$

$$= \frac{2\theta_{obs2}}{2\theta_{std}} \cdot \frac{2\theta_{std}}{2\theta_{obs}}$$

$$= \frac{2\theta_{obs2}}{2\theta_{obs}}$$

Therefore:

$$\frac{2\theta_{obs2}}{2\theta_{obs}} = \frac{R \cos \alpha - r_1}{R \cos \alpha} / \frac{R - r_1}{R}$$

$$\frac{2\theta_{obs2}}{2\theta_{obs}} = \frac{R \cos \alpha - r_1}{R \cos \alpha} \cdot \frac{R}{R - r_1}$$

$$\frac{2\theta_{obs2}}{2\theta_{obs}} = \frac{R - \frac{r_1}{\cos \alpha}}{R - r_1} \quad (8.3)$$

Rearrange equation 8.3 to solve for r_1

$$\frac{2\theta_{obs2}}{2\theta_{obs}} = \frac{R}{R - r_1} - \frac{\frac{r_1}{\cos \alpha}}{R - r_1}$$

$$\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right)(R - r_1) = R - \frac{r_1}{\cos \alpha}$$

$$R\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right) - r_1\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right) = R - \frac{r_1}{\cos \alpha}$$

$$-r_1\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right) = R - \frac{r_1}{\cos \alpha} - R\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right)$$

$$\frac{r_1}{\cos \alpha} - r_1\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right) = R - R\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right)$$

$$r_1\left(\frac{1}{\cos} - \frac{2\theta_{obs2}}{2\theta_{obs}}\right) = R - R\left(\frac{2\theta_{obs2}}{2\theta_{obs}}\right)$$

$$r_1 = \frac{R - R\frac{2\theta_{obs2}}{2\theta_{obs}}}{\frac{1}{\cos \alpha} - \frac{2\theta_{obs2}}{2\theta_{obs}}} \quad (8.4)$$

8.3 Peak Finding Algorithm

For x to be considered a peak at position n then the following criteria must be met:

$$\sum_{n=-10}^{n=-1} (x_n - x_{n+1}) > \sum_{n=-10}^{n=-2} (x_n - x_{n+1})$$

$$\sum_{n=0}^{n=10} (x_n - x_{n+1}) > \sum_{n=1}^{n=10} (x_n - x_{n+1})$$

$$x_n - x_{n+i} \geq 0$$

for values of i between -10 to + 10

$$\sum_{n=-10}^{n=-1} (x_n - x_{n+1}) \geq \text{Threshold}$$

8.4 Taylor Series Expansion of Cos(x)

$$\cos(x) = 1 - \frac{x^2}{2!} + \frac{x^4}{4!} - \frac{x^6}{6!} \cdots \quad (8.5)$$

8.5 Publications

Title	The separation of X-ray diffraction patterns for threat detection
Journal	Applied Radiation and Isotopes
Received	17th July 2009
Revised	20th October 2009
Accepted	25th November 2009

Title	Position determination of scatter signatures
Journal	The International Journal of Pure and Applied Analytical Chemistry
Received	7th July 2010
Revised	6th September 2010
Accepted	25th September 2010

Title	Combined X-ray diffraction and kinetic depth effect imaging
Journal	Optics Express
Received	21st January 2011
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Accepted	21st March 2011



The separation of X-ray diffraction patterns for threat detection

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ABSTRACT

We introduce a novel method for identifying materials using a series of X-ray diffractograms collected in transmission. A multiple perspective approach is used to identify the diffractograms produced by materials located at different positions along the primary X-ray beam. This technique promises to enhance materials identification performance in cluttered environments such as those prevalent in aviation security screening.

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1. Introduction

The security screening industry is faced with the problem of developing technology that can provide high quality X-ray images of sufficient specificity to inspect items prior to their placement on board an aircraft. Concealed explosives in hold or carry-on luggage are considered by many authors as the most severe threat to airport security (Yinon, 2007). They are not readily recognisable by shape (Singh and Singh, 2003), only a small amount is required to bring down an aircraft (Speller, 2001) and they appear identical to benign materials on dual-energy X-ray scanners (Jupp et al., 2000; Chan et al., 2004; Wee and Evans, 2003). Harding (2009) and Speller (2001) have demonstrated the capacity of X-ray diffraction for the detection of concealed explosives. In each case energy dispersive X-ray diffraction (EDXRD) is used to obtain characteristic spectra from individual locations within the inspection volume. In contrast, we have considered the application of angular dispersive X-ray diffraction (ADXRD) to this problem. This is not intended to replace EDXRD but instead provides an alternate viewpoint, looking at this issue from geometrical considerations. We have been pursuing ADXRD because of its inherently superior materials specificity.

2. Experimental system and methods

2.1. Experimental parameters

The measurements were made on a Philips PW1820 X-ray diffractometer incorporating a sealed, fine-focus X-ray tube with a

molybdenum target. The accelerating voltage and current were 40 kV and 30 mA, respectively. The scattered photons were detected by a scintillator detector located on an arm that rotates about a central point as illustrated in Fig. 1. Finally, primary X-ray collimation of $1/6^\circ$ was used; this gave an approximate spot height of 0.4 mm at the sample when located at the point of rotation.

2.2. Geometry of ADXRD system

The majority of suspect materials within a suitcase will be deemed to be polycrystalline. Thus a powder diffraction method will be considered. If X-rays strike a target exhibiting long range order then diffraction occurs. Constructive and destructive interference effects result in X-rays only being emitted at characteristic angles (Cullity, 1956). This relationship is well known and governed by Bragg's law

$$\lambda = 2d \sin \theta \quad (1)$$

where λ is the wavelength of the radiation used, d the interplanar spacing within the material and 2θ the angle through which the radiation is scattered. Typical X-ray diffractometers irradiate the sample under investigation with a well collimated beam of monoenergetic X-rays. A detector located on a moving arm moves around the sample with constant radius collecting scattered radiation. The angles at which diffraction occur are then converted to d spacings using Eq. (1). A more detailed discussion of this process is present in Cullity (1956). To identify the material the d spacings are compared to a library of diffraction profiles to find the best match. In this scenario the location of the sample is crucial. The sample must be located on the axis of rotation of the

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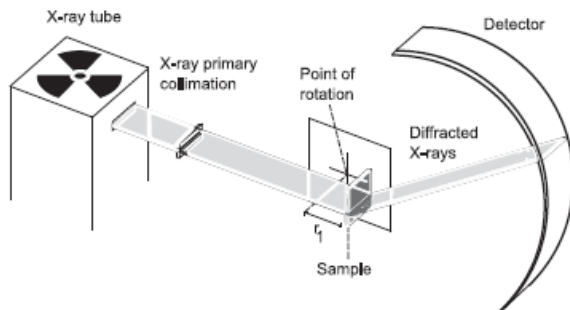


Fig. 1. The geometry of the X-ray diffractometer when the sample is displaced from the point of rotation by distance r_1 .

detector for the correct angles and subsequent d spacings to be derived. In practice this is not possible because the sample is not infinitely thin. However, if the sample is displaced from the rotation centre by an amount s , then the subsequent change in scattering angle can be estimated from the well known displacement error Eq. (2)

$$\Delta 2\theta = -2s \cos \theta / R \quad (2)$$

where R is the distance from the sample to the detector (Jenkins and Snyder, 1996). An important point to note is that angular dispersive diffraction patterns are typically detected in reflection. Therefore, Eq. (2) only corrects for this displacement issue in diffraction patterns collected in reflection. A more detailed explanation can be found in Jenkins and Snyder (1996).

2.3. Materials used

Sample materials were chosen because of their ease of use not necessary their relevancy to an airport screening environment. In the examples shown sheet aluminium was used in transmission (approximately 0.2 mm thick). This material was chosen because it is readily available, self supporting, easily manipulated and most importantly its diffraction pattern has relatively few peaks. This makes the demonstration of this method more straightforward.

2.4. Identification of displaced patterns

If an airport screening system is considered then it is conceivable that the diffraction producing material may be located at any point within the inspection volume. If diffraction signals (collected in transmission) can be isolated within a single dimension illuminated by the X-ray beam then Eq. (3) can be derived. This relationship can be used to transform ideal experimental data (where the sample is situated at the point of rotation) to how they would appear at a designated displacement position (referred to as r_1). To achieve this it is assumed that the angles of diffraction ($2\theta_{std}$) for the standard pattern are known

$$2\theta_{obs} = 2\theta_{std} - \sin^{-1}[r_1 \sin(180 - 2\theta_{std})/R] \quad (3)$$

$2\theta_{obs}$ represents the angle of diffraction measured from the point of rotation and R is the distance from the point of rotation to the detector. This method lends itself to transforming threat diffraction pattern libraries (of which there are finite amounts) to how they would appear at a multitude of locations in the illuminated dimension. The hypothesised diffraction patterns (from differing r_1 positions) can be systematically compared against the suspect pattern. The threat material that demonstrates the strongest

resemblance to the suspect pattern and its hypothesised location can then be reported to the operator. An assumption that is made to facilitate this method is that no collimation is used on the detector. This will ensure that scattered X-rays can strike the detector from any position along the inspection volume that is illuminated by the X-ray beam. Preliminary experiments have demonstrated that this does not affect resolution providing a suitably thin incident X-ray beam is used.

2.5. Separation of multiple patterns

One of the assumptions made in Section 2.4 for identifying displaced samples is that the diffraction information and thus any associated diffraction peaks come from a single material occupying the same point in space. In practice this is unlikely. Considerations should be made for resolving multiple unknown material (or combined material) diffraction patterns. Consider a diffractogram that contains a contribution of peaks from multiple materials. In order to identify a material and identify its location the peaks that comprise the diffractogram need to be grouped according to the material they came from. It is impossible to achieve this differentiation from a single diffractogram. Thus a new variable must be introduced. The consideration of other perspectives is one such variable. Consider an inspection volume with two different materials at differing locations from each other. Introducing a global rotation (hereby referred to as α) of the inspection volume (whilst the X-ray source and detector remain stationary) will change the apparent separation distance that is measured between the samples. This causes the distance between the materials in the inspection volume to effectively change. This will modify the measured peak positions within the diffraction pattern.

If numerous diffraction patterns are obtained from a series of rotations of the inspection volume then the resulting diffraction patterns will be comprised of peaks that modify their positions at differing rates. The rate at which the peaks modify their position is related to the sample location within the inspection volume. Fig. 2 illustrates this principle. Eq. (4) depicts the relationship between the original displacement position (r_1), the new displacement position (r_2) and the rotation of the inspection volume (α)

$$r_2 = r_1 / \cos \alpha \quad (4)$$

Analysing the rates at which the individual peaks shift position when the inspection volume is rotated will elucidate which peaks have a common origin.

3. Measured diffraction profiles

3.1. Experimental and predicted diffraction patterns

In order to test the validity of the method suggested in Section 2.4 a series of patterns were collected from a known material at a range of displacement positions. Fig. 3 shows the pattern obtained from aluminium in a displaced location (10 mm from the point of rotation) and the standard pattern (no displacement) to illustrate the movement incurred.

A standard pattern collected from the point of rotation was transformed using Eq. (3) to how it would appear at the same location as the suspect. This is demonstrated in Fig. 4.

3.2. Multiple pattern separation

To test the hypothesis for resolving combined diffractograms a new dataset was synthesised. Two diffraction patterns of aluminium collected in transmission from differing displacement positions (2 and 12 mm from the point of rotation) were added. This superposition represented what would be expected for the 0°

- S = An infinitely thin sample rotated by 45 degrees in α about O
- O = The point of rotation of the arc detector
- X = The path of an non-divergent X-ray beam
- r_1 = The distance a sample is displaced spatially from O normal to the X-ray beam
- r_2 = The new displacement position along the X-ray beam once the sample has been rotated by α
- α = Degree of rotation about the point of rotation

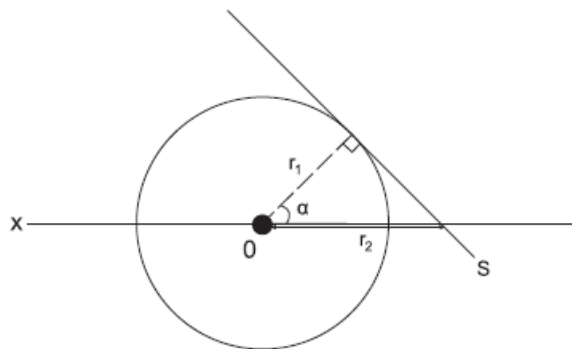


Fig. 2. Geometry illustrating the change in displacement position achieved along the X-ray beam when the sample is rotated about the point of rotation by α .

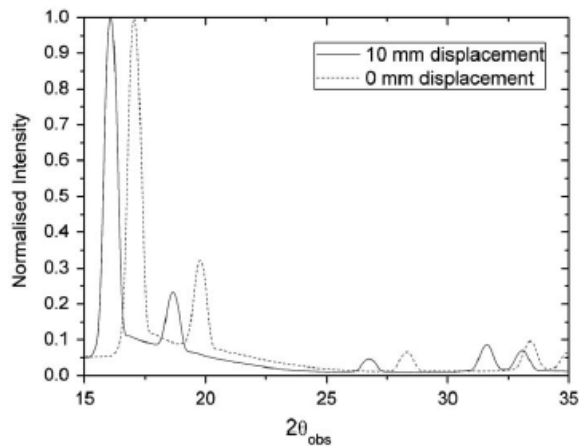


Fig. 3. Two normalised diffraction patterns of sheet aluminium measured in transmission. One is located at the point of rotation (standard). The other is displaced by 10mm from the point of rotation along the X-ray path toward the detector.

α rotation diffractogram. Each displaced diffractogram was also altered to simulate rotation positions 1–45° in α . The two resulting diffraction patterns for each rotation position were then added to comprise the rest of the dataset. The differing rates that peaks modify their position when the inspection volume is rotated are best demonstrated in a movie format. Fig. 5 illustrates snapshots of such a movie showing the diffraction patterns at a select number of rotation positions.

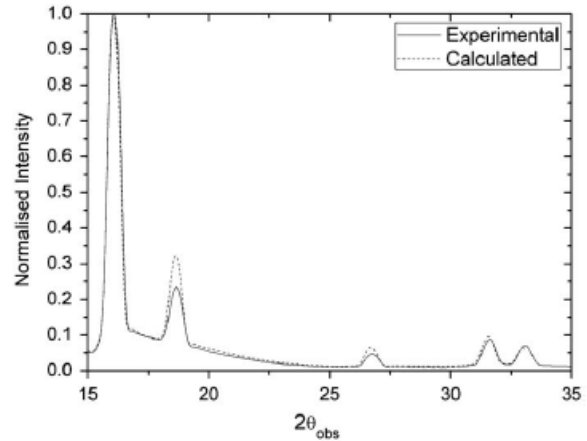


Fig. 4. Two diffraction patterns are demonstrated. An experimentally measured diffraction pattern of sheet aluminium measured in transmission displaced by 10 mm from the point of rotation along the X-ray path toward the detector. Also a predicted (calculated) pattern of the same material at the same location.

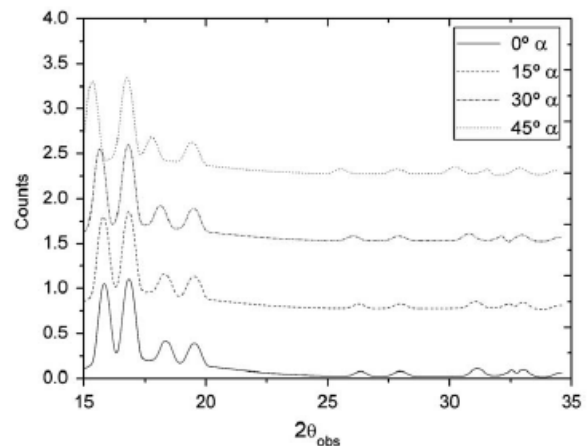


Fig. 5. A series of simulated combined diffraction patterns (containing sheet aluminium displaced by 2 and 12 mm from the point of rotation along the X-ray path toward the detector) as they would appear if a global movement of the inspection volume through 0°, 15°, 30° and 45° in α around the point of rotation were incurred.

To ensure the validity of the synthesised dataset two rotation positions were collected experimentally (0° and 45° in α) and cross referenced with the simulated data. These are demonstrated in Figs. 6 and 7 respectively. The peak positions between the experimentally obtained dataset and the synthesised dataset are compatible. A discrepancy in peak amplitude can be seen, however peak amplitude is seldom used because of the unknown amounts of preferred orientation within the sample.

The rate at which peaks modify their position that is observed within the diffraction patterns can be attributed to two factors. The first factor is the discrepancy in displacement position induced by the global rotation of the inspection volume by α . The second factor is caused by the peak angle (which we have referred to as $2\theta_{std}$). If the contribution from $2\theta_{std}$ can be isolated and removed then any remaining peak movement can be attributed to sample movement. It is proposed that the $2\theta_{obs}$ positions for each peak in the first α position (0°) serve as a

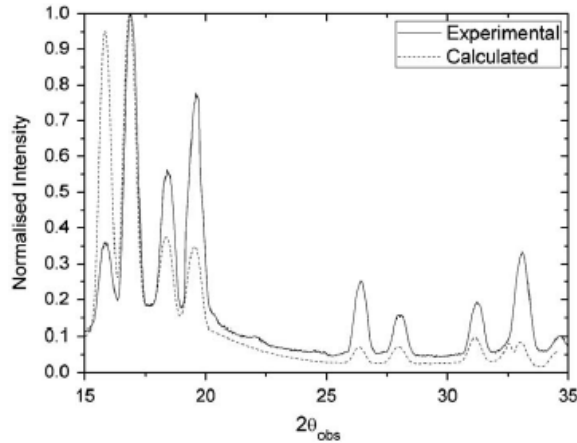


Fig. 6. Experimentally collected combined diffraction pattern (containing sheet aluminium displaced by 2 and 12 mm from the point of rotation along the X-ray path toward the detector) at 0° α to the point of the rotation overlaid with the simulated data of the same environment.

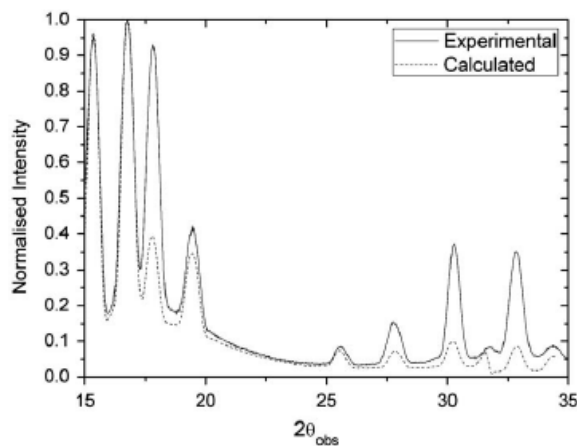


Fig. 7. Experimentally collected combined diffraction pattern (containing sheet aluminium displaced by 2 and 12 mm from the point of rotation along the X-ray path toward the detector) at 45° α to the point of the rotation overlaid with the simulated data of the same environment.

reasonable approximation to $2\theta_{std}$. Every peak position for all rotation positions (α increments from 0° to 45°) were divided by their respective approximations to $2\theta_{std}$. This in effect can be thought of as normalising the peak positions with respect to $2\theta_{std}$. The result is demonstrated in Fig. 8. Each of the lines (representing individual peak positions) adheres to one of two characteristic curves. This helps to separate the peaks qualitatively as each curve is characteristic of a single material. To separate the peaks quantitatively each line was fitted with a second order polynomial. Plotting the first and second order coefficients of the fitted polynomials results in the peaks that originate from the same material falling in the same region of the graph. This is illustrated in Fig. 9. For clarity gray circles have been used to indicate peaks that belong to the same material. The example shown demonstrates an amalgamation of two diffraction patterns of the same material (aluminium). This is merely for the sake of clarity. It should be noted that patterns comprised of peaks from differing materials can be separated in the same manner.

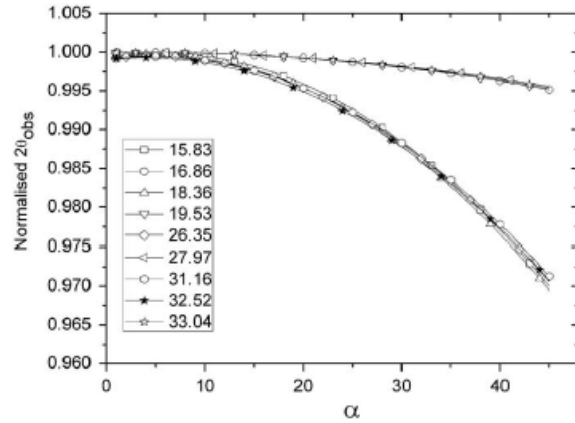


Fig. 8. Illustration of the change in $2\theta_{obs}$ for each peak when the inspection volume is rotated about the point of rotation for α increments of 0 – 45° after being normalised with respect to an approximation to $2\theta_{std}$.

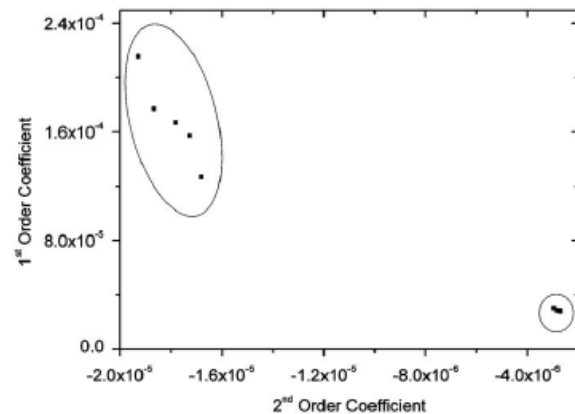


Fig. 9. Plot of the first and second order polynomial coefficients for each peak curve. The curves were derived by reviewing the change in $2\theta_{obs}$ for each peak when the inspection volume is rotated about the point of rotation for α increments of 0 – 45° after being normalised with respect to an approximation to $2\theta_{std}$.

3.3. Sample thickness

The extension of a sample along the principal X-ray path results in a broadening of the diffraction maxima. This inevitably impacts upon the angular resolution of the system and ultimately, the success of any pattern separation method. If molybdenum radiation is used then ~ 2.1 mm of aluminium would be sufficient to absorb 99% of the primary beam flux. The expected increase in diffraction peak width at this penetration depth (calculated for the first peak in aluminium when the sample is situated at the point of rotation) would be around 21% of its original width. Our experience of materials identification using diffraction suggests that such an increase in peak width would not significantly reduce our ability to discriminate materials. However, this issue would be exacerbated if a shorter wavelength radiation source were considered. Tungsten radiation will penetrate around 6 cm of aluminium before the primary beam flux is reduced to 1% of its original intensity thus giving a significant increase in diffraction peak broadening. This would be detrimental to the angular resolution of the system.

However it should be noted that this is an extreme example. In reality samples of particular interest are likely to be relatively thin because such structures are difficult to detect with 'standard' dual-energy X-ray screening techniques. For example, Luggar et al. (1997) concentrate on a typical sheet explosive thickness of 5 mm with similar values being used by Malden and Speller (2000). A sample thickness of this magnitude would increase the width of the first diffraction peak of RDX by approximately 21% (W radiation). Changes in the experimental geometry may also help to combat this loss of resolution.

4. Conclusions

A new method for identifying materials located in a displaced position has been proposed and shown to be viable providing the samples are suitably thin along the X-ray path axis. A suspect diffractogram can be compared with a library of threat materials but with one key difference. The diffraction pattern for each threat material must be transformed to how they would appear at a multitude of locations first and then cross referenced with the suspect pattern. Altering the number of transformations considered allows control over the system sensitivity. Specificity can be tailored by applying a minimum threshold distance to determine how close peaks need to be before they are considered a match. The problem of overlapping spectra caused by differing materials located at different displacements has also been investigated. A potential solution has been found by employing a multiple view approach that utilises a global rotation of the inspection volume. The rates at which peaks modify their position as the inspection volume is rotated are used to differentiate between peaks from one origin with those from a different origin. Multiple materials located in the same location (mixed phase) can still be identified, however the improvement made to the identification of unknown samples located in differing positions in depth is significant.

5. Future work

The parallels that this technique has with that of kinetic depth effect X-ray imaging (KDEX) developed by Evans et al. (2006) cannot be ignored. Both techniques utilise differing perspectives to obtain more information about the inspection volume. KDEX employs sequential parallax to realise the 3D visualisation of a

structure under inspection. Whereas this paper introduces a technique in which samples at different depths (or position along the X-ray beam) produce differential sequential shifts in the angular distribution of diffracted X-rays, which in turn reveal materials discrimination information. By hypothesis, the complementary sources of information realised by each technique maybe combined to realise enhanced threat detection performance. Time will be spent investigating the potential amalgamation of these two approaches. During the course of this investigation differing sample and detector positions will be hypothesised. The incorporation of an optical lever (i.e. increasing the distance from sample to detector) may combat any loss in resolution introduced by extended samples. This ongoing programme of work is funded by the EPSRC and is in collaboration with the UK Home Office Scientific Development Branch (HOSDB) and the US Dept of Homeland Security (DHS).

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Position determination of scatter signatures – A novel sensor geometry

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ABSTRACT

A novel diffraction sensor geometry able to provide the diffraction pattern of a suspect material without prior knowledge of the samples location is introduced. The sensor geometry can also resolve diffraction patterns originating from multiple unknown materials overlapped along the primary X-ray beam path. This is achieved through tracking Bragg peak maxima that linearly propagate from the inspection volume at a series of X-ray detector positions. A series of simulations and experiments have been performed to verify this technique and provide an insight into its characteristics. Such a technique could have widespread appeal in the security industry. Areas of most relevance include the materials characterisation of volumes such as those prevalent in an airport screening environment or equally the rapid screening for illicit drugs trafficked through the postal system.

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1. Introduction

A single approach with high sensitivity and specificity that can effectively and non-destructively screen volumes for illicit materials is yet to be adopted by the security industry. Materials that need to be identified include concealed explosives and controlled substances. Explosives are considered by many authors to be the most severe threat to airport security [1], and their perceived presence has placed an unprecedented moral and economic burden on the aviation industry. In the long term the illegal trafficking of controlled substances through the postal system (both domestically and internationally) can be considered equally as detrimental to society.

X-ray diffraction has been shown to be an effective probe for detecting both concealed explosives [2–5] and controlled substances [6–9]. These materials are generally of a crystalline nature and therefore produce characteristic diffraction maxima likened to “fingerprints” by some authors [6]. The majority of these proposed screening systems use an energy dispersive method whereby characteristic X-ray spectra are obtained from individual locations within the inspection volume that is illuminated with a broadband of radiation. Generally a sophisticated set of collimators which restrict the direction of the incident and diffracted rays is required to ensure that the origin of the scattered radiation (sample position) is well defined. This collimation impinges on the system sensitivity

and can result in as much as a 99% loss of the scattered beam flux [4].

We have been investigating a new approach which uses an angular dispersive method [10] due to its superior resolution [7]. The majority of angular dispersive systems also rely on a known sample position to correctly interpret the diffraction pattern. We have been investigating a new technique which determines the sample location without the use of collimation. This has the capacity to speed up screening times as a whole depth dimension can be evaluated simultaneously. Efforts have also been made to consider the impact of sample thickness, which is known to be problematic in transmission mode [8,10]. This preliminary study demonstrates a small scale system that utilises molybdenum radiation. The penetrating ability of this radiation is at the lower limit for letter or parcel screening and certainly too low to penetrate a full suitcase. Typically for higher penetration imaging, airports may employ a tungsten target with a characteristic line at approximately 59 keV. This radiation comes with a penalty of increased Bremsstrahlung. However the use of appropriate K edge filtration would mitigate against this effect and so theoretically the techniques presented here could be used with tungsten radiation in a scaled up version. K edge filtration would also come with the additional benefit that it will improve the image quality of transmission based imaging systems.

1.1. Principles of X-ray diffraction

The majority of illicit materials such as explosives and drugs are polycrystalline and so a powder diffraction model has been

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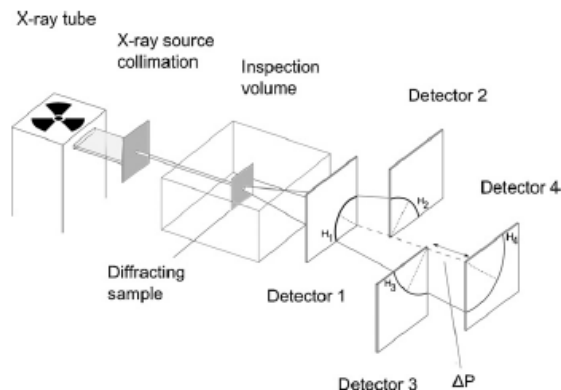


Fig. 1. The geometry of the X-ray diffractometer with four detectors superimposed at different positions along and normal to the primary X-ray beam path.

adopted. Diffraction maxima occur when a mono-energetic X-ray beam strikes a crystalline material (or any material exhibiting long range order) at a specific angle of incidence. Constructive and destructive interference effects result in radiation being scattered at characteristic angles [11]. These are directly related to the differing interplanar spacings within the material. This relationship is governed by Bragg's law,

$$\lambda = 2d \sin \theta \quad (1)$$

where λ is the wavelength of the radiation used, d is the interplanar spacing within the material and 2θ is the angle through which the radiation is scattered relative to the incident beam direction. Thus if monochromatic radiation is used and the angle of scatter for any Bragg peak measured, then the interplanar spacings (characteristic of material) can be determined. Clearly the sample position relative to the detector is a critical factor for calculating the angle of scatter in most experimental systems. We have been developing a new approach which does not require a prior knowledge of sample position.

1.2. Rationale

Consider a diffraction pattern consisting of multiple diffraction maxima collected on a planar detector arranged normal to the incident beam (see Fig. 1). Translating the detector along the primary beam, will result in the Bragg maxima striking the detector at different locations i.e. the radius of the Debye cones' projection onto the detector (H) will change. The angle of scatter for each Bragg peak can thus be calculated from,

$$\frac{dH}{dP} = \tan 2\theta \quad (2)$$

where P is the sample to detector distance. Therefore, by measuring the rate at which the radius changes as the sample to detector distance is changed, it is possible to determine both the scatter angle and the sample position (relative to the detector) simultaneously.

In principle then a minimum of two diffraction patterns need be measured in order to determine both 2θ and the sample position. However in practice this is not possible, particularly in the case of measured diffraction patterns consisting of multiple Bragg peaks as there is no reliable coincidence information that can be used to identify the common maxima between each pattern. We have developed a method for correctly identifying such corresponding Bragg peaks ("peak tracking") and this is discussed below. It should be noted that peak amplitude is an unreliable characteristic to exploit in this context due to multiple factors affecting peak heights (e.g. overlapping maxima).

1.3. Peak tracking

With no coincidence information, and assuming that all diffraction maxima (n) measured at one detector position are also observed at a second position, the number of possible Bragg peaks that could have created peaks in the observed detector positions ranges from $(n(n+1)/2)$ to n^2 . Only a proportion of these Bragg peak possibilities will be correct. It is evident that for scenarios where n is large the proportion of correct Bragg peak possibilities to false will diminish. Our approach to identify corresponding diffraction maxima uses Eq. (2) applied to diffraction data collected from multiple (>2) detector positions. The positions of any maxima are linearly related through the detector positions. Thus determining those peak positions which possess a fixed gradient when plotted against detector position enables coincidence grouping of the peaks. A correct d -spacing and source position may then be determined.

For non-ideal conditions (e.g. where diffraction peaks have a finite width and thus peak overlapping is more frequent), a greater number of detector positions may be required. The inclusion of additional detector positions will increase the specificity of the system, however it will also warrant either a higher associated system cost (for the additional detectors) or an extra time constraint (where the detector is moved to different positions). We have found that four detector positions will eliminate enough false Bragg peaks to differentiate simple volumes containing few materials. For complex volumes that contain >5 materials with low symmetry the technique may require more than four detector positions to achieve high levels of sensitivity (true positive rate) and 1-specificity (false positive rate).

2. Method

2.1. Simulation

A simulation was created in Matlab® to demonstrate the premise of the peak tracking method and to aid in selecting possible configurations prior to experimental tests. A number of materials of a chosen thickness (elongation) were randomly assigned a position along the X-ray beam path within a boundary representing an imaginary inspection volume. The diffraction patterns for each material were then projected onto detectors at specified distances and linearly summed. It should be noted that the projected pattern from each material contributed equally to the resulting patterns at each detector position and no allowances for the scattering cross sections of the materials, re-absorption effects or preferred orientation were made. However, these factors only affect peak amplitude not position. Each of the 1D patterns were smoothed using a Savitzky-Golay filter, normalised and then a modified first differential was applied to provide a robust peak finder in the presence of noise. Finally, each pattern was evaluated to find potential correct 2θ solutions and their associated position on the primary X-ray beam path, the result of which is reported in Section 3.1.1.

To test the robustness of the system a mixture of illicit material diffractograms (e.g. TNT, PETN) were evaluated in the presence of those from benign materials (e.g. talcum powder, sucrose). The diffraction patterns produced by the peak tracking method were checked by a simple algorithm that searched a limited threat database of diffraction patterns to determine if an illicit material was present. Receiver operator characteristic (ROC) curves were created to compare the true positive (sensitivity) and false positive ($1 - \text{specificity}$) rates. This demonstrates how often threats were correctly identified and how often benign materials are identified as a threat (false alarm), respectively. The number of Bragg peaks matching those of a threat material was used as the discriminating

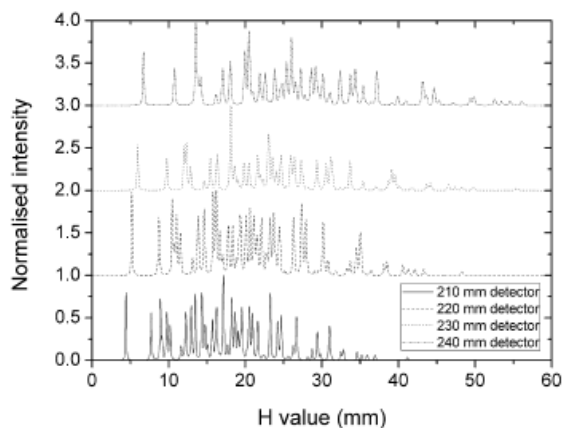


Fig. 2. Simulation of the diffraction patterns observed on a series of linear arrays arranged normal to the primary X-ray beam path at 210, 220, 230 and 240 mm from the source when spatially separated samples of TNT and talcum powder are placed in the beam.

threshold to produce the ROC. A summary of these results can be found in Sections 3.1.2 and 3.1.3. The reader may be interested in the false negative rate (e.g. how often the system fails to identify a threat). This is $1 - \text{true positive rate}$.

2.2. Experimental parameters

We have undertaken a systematic series of experiments incorporating samples placed at different distances from a detector and resultant diffraction patterns collected at increasing sample to detector distances. The X-rays were produced by a Philips PW1830 X-ray generator incorporating a sealed, long fine focus X-ray tube with a molybdenum target. The accelerating voltage and current were 40 kV and 30 mA, respectively. A PIXIS 1024 × 1024 16 bit CCD camera with a phosphor screen was used in a raster scan mode to collect the scattered photons. A 4 mm thick brass plate with 0.66 mm diameter aperture was used to collimate the primary X-ray beam into a pencil beam. Finally, a set of Thorlab stages were used to translate the detector along the primary beam path.

2.3. Materials

In the example shown in Section 3.2, sheet aluminium (approximately 0.2 mm thick) and aluminium oxide (approximately 0.1 mm thick) were used as they present characteristic diffraction maxima over a typical angular range. Relatively thin samples were chosen due to the low energy X-rays employed as well as its subsequent affect on Bragg maxima width (presented in Section 3.1.2 and discussed in Section 4). In addition, these two materials are chosen as it would be difficult to differentiate between them by applying elemental techniques, which helps to demonstrate the specificity achievable with a scatter based system.

3. Results

3.1. Simulations

3.1.1. The premise

Fig. 2 illustrates the simulated patterns created from a spatially separated mixture of trinitrotoluene (TNT) and talcum powder (each at 0.1 mm elongation) when viewed at differing detector positions. The change in peak position (ΔH) is a function of 2θ and the materials' position along the primary X-ray beam path. Apply-

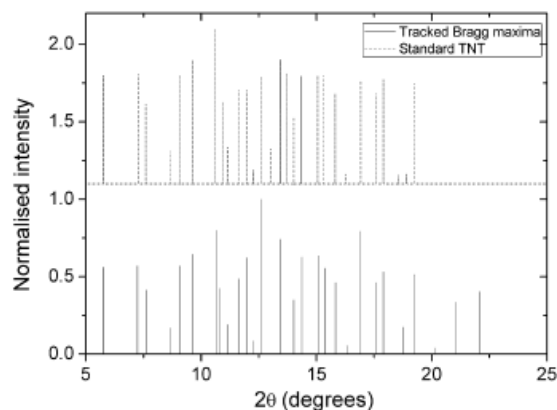


Fig. 3. Plot of a grouping of Bragg peaks tracked through the patterns illustrated in Fig. 2 believed to have a common origin. The pattern corresponds well with TNT.

ing the peak tracking method described in Section 1.3 results in many possible calculated 2θ values and positions. Two of these groups of Bragg peaks are demonstrated in Figs. 3 and 4. The peak positions were a good match to those of standard TNT and talcum powder proving to be the most likely candidate as determined by a conventional diffraction matching algorithm (Crystallographica Search-Match®) that searched approximately 200,000 diffraction patterns. Thus, from a mixture of diffraction patterns arising from differing sample positions, the algorithm was capable of discriminating between materials and locating their relative positions with respect to the detector.

3.1.2. Effect of elongated samples

The peak tracking method is dependent upon the algorithm's ability to correctly identify peak positions at the detector. When samples are elongated along the primary beam axis the diffraction patterns, when collected in transmission mode, are known to be problematic. This is because broadening in the diffraction maxima caused by scattering from the front surface right through to the back surface of the sample is observed [10]. This reduces the precision in peak location and increases the overlapping likelihood. Fig. 5A illustrates the change in shape of an ROC curve as the samples under inspection are elongated. Each individual simulation contained two materials (chosen at random from a small refer-

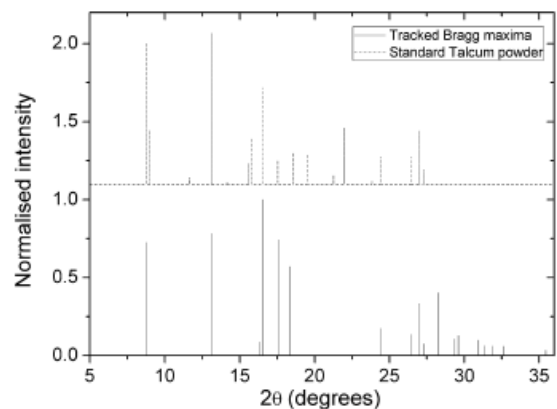


Fig. 4. Plot of a grouping of Bragg peaks tracked through the patterns illustrated in Fig. 2 believed to have a common origin. The pattern corresponds well with Talcum powder.

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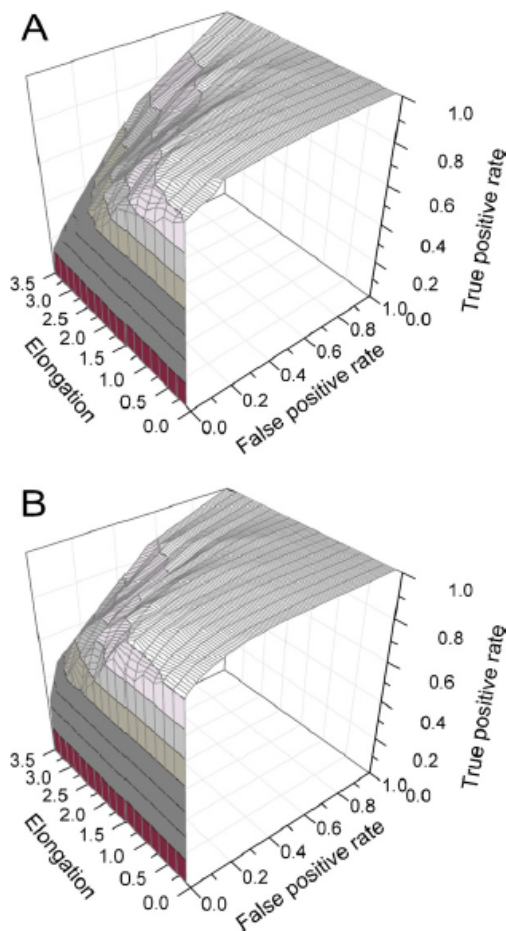


Fig. 5. Surface A illustrates the change in shape of the ROC curve with elongation of the sample materials. Simulated detectors were placed at 210, 220, 230 and 240 mm. Surface B illustrates the same arrangement except the simulated detectors were placed at 220, 240, 260 and 280 mm.

ence library of benign and illicit materials) at a specified elongation assigned to a random location within the inspection volume. Theoretical detectors were placed at 210, 220, 230 and 240 mm from the source. The ROC curve demonstrates the systems ability to reliably identify a threat material. The number of peaks required to identify the threat was used as the discriminating threshold. The simulation was repeated approximately one thousand times. The results of this simulation are discussed in Section 4.

3.1.3. Effect of detector distance

Increasing the sample to detector distance could mitigate against the reduced resolution offered by elongated samples. This is because the concomitant increase in relative diffraction peak width would be less than that of the corresponding positional change. Similarly to Fig. 5A (described in Section 3.1.2), Fig. 5B illustrates an ROC surface with the same simulation settings except the detector distances were increased from 210, 220, 230 and 240 mm to 220, 240, 260 and 280 mm. The change in volume bounded by the surface is discussed in Section 4.

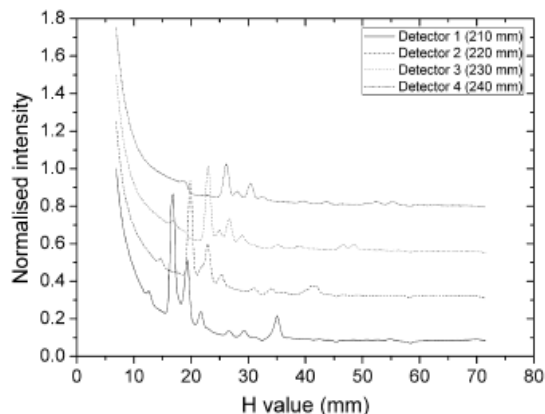


Fig. 6. Radially integrated diffraction patterns of spatially separated aluminium and aluminium oxide measured normal to the primary beam at specified distances along the primary beam path.

3.2. Experimental proof of principle

To evaluate the methods empirically sheets of aluminium and aluminium oxide were placed at different locations along the primary beam path to simulate objects at differing locations in a volume. The diffraction patterns were then measured at four different detector positions relative to the inspection volume. The intensities within each pattern were radially integrated to produce the 1D scattering patterns illustrated in Fig. 6. It should be noted that evaluating thin samples with large crystallites can result in incomplete debye rings. Radially integrating the pattern from a 2D detector mitigates against this effect.

Similarly to that demonstrated in Section 3.1.1 the Bragg maxima recorded at each detector position were evaluated to identify those with behaviour consistent with Eq. (2). The results are illustrated in Figs. 7 and 8. These patterns correspond well to those produced by aluminium and aluminium oxide. Some of the standard peaks from aluminium and aluminium oxide are missing in the tracked patterns; this effect is caused by the peak overlapping phenomenon mentioned previously. An additional compounding consideration is that any preferred orientation affects may cause peak amplitudes to diminish below the noise level of the detector.

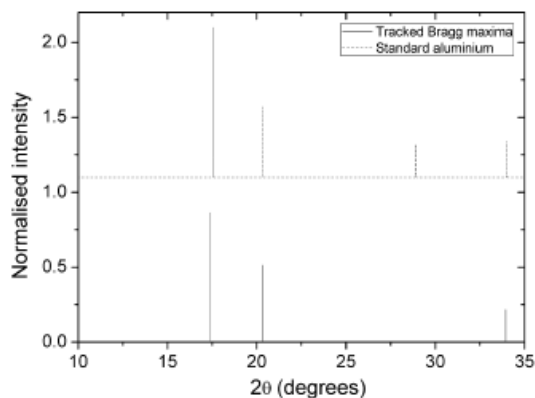


Fig. 7. Plot of a grouping of Bragg peaks tracked through the diffraction patterns illustrated in Fig. 8 which are believed to have a common origin. The pattern corresponds well with aluminium.

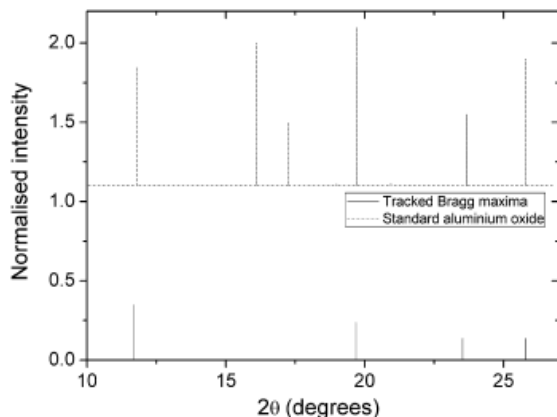


Fig. 8. Plot of a grouping of Bragg peaks tracked through the diffraction patterns illustrated in this figure which are believed to have a common origin. The pattern corresponds well with aluminium oxide.

4. Discussion and conclusion

A novel and versatile rationale capable of identifying the angles of scattered radiation (2θ) as well as the positions of diffracting materials (sample position) has been proposed and validated in the laboratory. The technique would lend itself well to the characterisation of materials within a volume such as those required in an airport screening environment because a prior knowledge of the sample position is not required. Multiple unknown materials that are spatially separated along the primary X-ray beam path can also be resolved simultaneously. Therefore, it could be used to increase the efficiency of mail screening systems as multiple letters (located at differing depth planes) could be evaluated at once. Perhaps one of the more interesting aspects of this technique is that the spatial differentiation is achieved without the use of post-sample collimation. Such collimation can result in a loss of as much as 99% of the scattered beam flux [4] and such systems are thus inherently inefficient. Another related consideration is the approach in deriving the sample position. For systems that use post-sample collimation, the sample position is at the point of intersection defined by the primary and diffracted beam collimators. As such the precision to which the sample position can be identified is constrained by the engineering and mechanical tolerances of the collimators. Our approach has no such limitation.

One of the main confounding factors of this technique is that of samples elongated along the primary beam. If the sample(s) are elongated along the primary beam axis then in transmission diffraction maxima broadening is observed. This impedes the performance of the peak tracking method. The extent to which the

performance is affected is illustrated by the ROC surface Fig. 5A. As the samples are elongated the ROC flattens nearing the no-discrimination line. To combat the loss in resolution introduced by elongated samples Section 3.1.3 evaluates the idea of increasing the sample to detector distances. The rate at which the Debye cone propagates with respect to P is greater than the diffraction maxima broadening introduced by the diverging pencil beam. For this reason increasing the sample to detector distances acts as a positive optical lever. The result of increasing the sample to detector distance is seen best in Fig. 5B. ROC curves are compared typically using their respective areas. As we are evaluating a third variable (elongation) it is appropriate to compare the respective volumes bounded by the ROC surfaces, where a volume of 100% would provide perfect discrimination and 50% would provide no discrimination. Fig. 5A has a volume of 84% whereas Fig. 5B has a volume of 89%. We therefore infer that increasing the sample to detector distances should help alleviate some of the issues encountered by elongated samples. Further, such increases in physical dimensions may well be a natural requirement for some applications of our technique. This preliminary work is part of an ongoing programme to incorporate X-ray diffraction based materials discrimination into an imaging technique. As a next step we intend to combine this approach with the novel tomographic approach afforded by kinetic depth effect X-ray imaging [12].

Acknowledgments

This ongoing programme of work is funded by EPSRC and is in collaboration with the UK Home Office Scientific Development Branch (HOSDB) and the US Department of Homeland Security (DHS).

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Combined X-ray diffraction and kinetic depth effect imaging

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Abstract: In this preliminary study we present a depth resolved transmission image sequence of an object combined with the materials discriminating ability of angular dispersive X-ray diffraction. Volumes within the object giving rise to diffraction patterns matched to a library of specific materials have been encoded visually within the images. The intensity of these highlighted areas has been weighted based on the certainty of the match. Both the theory and experimental proof of principle have been demonstrated. Considerations pertaining to a “scaled up” version of this technique are also discussed.

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OCIS codes: (110.7440) X-ray imaging; (050.1940) Diffraction; (100.2980) Image enhancement.

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1. Introduction

The development of rapid 3 dimension imaging for the reconstruction of volumetric information has impacted within an increasing number of fields including materials science, medicine and security screening. However, the aim of such imaging is often to identify the distribution (or presence) of a specific material or range of materials within a volume and this is generally not available through current imaging modalities. For example, the screening of luggage to detect the presence of prohibited materials is a problem that still lacks a comprehensive solution.

Whereas absorption based image acquisition methods have developed into high resolution, volumetric visualisation tools such as stereoscopy [1, 2], computerized tomography (CT) [2, 3] and kinetic depth effect X-ray (KDEX) techniques [4], there has been little concomitant development of complementary chemical identification methodologies. The demand for such methods is well demonstrated, for example, with the need to identify explosives within luggage [5–7] that may have an innocuous shape. Qualitative materials discrimination is currently mostly limited to dual-energy imaging which has also been applied recently to very high spatial resolution phase contrast imaging [8, 9], although material's identification methods remains under development.

X-ray diffraction is a well established technique that is routinely used within laboratories for materials identification. It has been shown to be an effective probe for chemically identifying concealed explosives [10–12] and can identify biochemical changes accompanying the onset of cancer [13, 14]. The 'fingerprint' scattering signatures [12] produced from a diffraction experiment can thus be used as a highly specific marker of particular materials even within a mixture of several materials. High resolution, 3D chemico-structural probes have been developed to characterise microstructures for both static [15] and dynamic studies [16]. Perhaps the most advanced diffraction imaging system is 'TEDDI' [17] which provides tomographic diffraction imaging and has evolved to accommodate relatively high X-ray energies. The 'gold standard' for definitive materials analysis from diffraction employs monochromatic X-rays and an angularly dispersive data collection approach. In contrast, often when diffracted X-rays have been used previously to complement absorption imaging an energy dispersive approach has been adopted [14, 17, 18]. Here characteristic diffraction patterns are obtained from individual locations within an inspection volume that have been illuminated by white radiation. Restricting the directionality of the primary and scattering X-rays via collimation provides positional information at the expense of overall beam flux [19].

We have been pursuing alternative angular dispersive configurations [20, 21] (because of their inherently superior specificity [22]) that crucially do not require scattering collimation. The limitation of such chemically specific techniques applied in isolation is that not all prohibited materials are chemically distinct. Knives and scissors are not readily distinguished from innocuous metals based on their chemical structure. Therefore any chemical technique needs to complement conventional imaging technology rather than replace it.

Our study demonstrates the first X-ray image sequences of an inspection volume obtained via the imaging technique, KDEX that has simultaneously been interrogated by X-ray diffraction. Areas giving rise to diffraction patterns matched to a library of specific materials have been encoded visually within the images. Furthermore the intensity of these highlighted areas has been weighted based on the certainty of the match. It should be noted that this preliminary study has been conducted with molybdenum radiation (K alpha $\approx 17\text{keV}$) which is somewhat lower energy than tungsten radiation (K alpha $\approx 59\text{keV}$) traditionally employed for baggage and medical imaging. However, the benefits demonstrated by this work may one day translate to a scaled up system operating at higher energies. Some of the issues involved in a scale up have been addressed in section 4.

1.1 Principles of X-ray coherent scatter

The majority of materials are polycrystalline or semi-crystalline. For this reason a powder diffraction model has been adopted employing a kinematic approximation. If a mono-energetic X-ray beam strikes a material with long range order, then a proportion is absorbed and then remitted isotropically. Constructive and destructive interference effects result in radiation being scattered at characteristic angles [23]. These angles are directly related to interplanar spacing's (d) within the material and are governed by Braggs law. Once the d -spacings of a material have been determined these are matched against a reference library to provide an identification. An important consideration is the experimental geometry and, in particular, the relative positions of sample and detector. If the relationship between the incident beam direction, the sample position and the detector position is ill described then the correct angles of diffraction cannot be calculated.

1.2 Determining the spatial origin of scatter signatures

We have employed an open collimation system and thus Bragg maxima from any diffracting material illuminated by the primary pencil beam can subsequently strike the detector. However, for objects extended along the primary beam Bragg maxima from multiple unknown positions may contribute to the resulting diffraction pattern. Previous work [21] has examined a possible solution that uses an X-ray detector translated to a series of positions along the primary beam path (illustrated in Fig. 1b). A peak tracking algorithm is then applied that searches the patterns for Bragg maxima positions that linearly propagate through the detector positions. The gradient of these linearly propagating Bragg maxima positions provides the diffraction angle (2θ). Back projecting the linear propagation provides the origin within the inspection volume. As a result the sample to detector distance of any diffracting materials (sample position) and Bragg diffraction angle can be determined simultaneously. A more detailed description of this approach can be found in Dicken et al [21].

1.3 Principles of kinetic depth effect X-ray (KDEX)

KDEX provides the ability to visualise an inspection volume in 3D and is therefore comparable to CT. However CT scanners often require many projections to reconstruct a volumetric image [2, 4] and this is costly in time. KDEX operates similarly in that it requires multiple views to provide its 3D effect, however the number of images it requires is typically an order of magnitude lower and there is no reconstruction penalty. A parallax effect is brought about by differences in angular velocity of individual objects within an inspection volume. Thus depth perception is afforded through this parallax [4]. Another important consideration is that the linear detector arrays used to produce the primary images can be arranged within an inspection tunnel in such a manner as they do not increase data acquisition time from that of a single detector system [4].

KDEX and X-ray diffraction then provide significantly different, complementary information concerning characteristics of any inspection volume. This work seeks, for the first

time, to combine these approaches and realise an unprecedented, materials specific imaging technique.

2. Method and materials

A 3D small scale phantom object was constructed that contained an alumina component (the 'target' material) as well as other scattering and attenuating objects overlapped in depth (illustrated in Fig. 2). X-rays were produced by a Philips PW1830 X-ray generator incorporating a sealed, long fine focus X-ray tube with a molybdenum target. A cooled (-40°C) PIXIS 1024 x 1024 16bit CCD camera with a $\text{Gd}_2\text{O}_2\text{S:Tb}$ phosphorous screen was used to detect the X-rays. A selection of Thorlab and Sigma-koki translation stages were used to move various components namely the phantom and camera through multiple axes. To obtain the KDEX image sequences, multiple images were collected after the object was rotated through 1° increments from $+15^{\circ}$ to -15° of the rotation axis whilst the X-ray source and detector remained stationary (illustrated in Fig. 1a). This is equivalent to the traditional KDEX geometry where the object remains stationary and source and detector are rotated [4]. The X-ray diffraction patterns were collected by collimating the X-rays into a pencil beam with a 4 mm thick brass plate and 0.66 mm diameter aperture (illustrated in Fig. 1b). A set of translation stages were then used to systematically move the sample across the pencil beam. The PIXIS camera was used in raster scan mode to collect the diffraction patterns propagating normal to the primary beam path. This operation was repeated at differing distances along the primary beam path so that multiple diffraction patterns could be collected for each interrogated point on the sample. 28 points (equating to a 4×7 grid) on the sample were inspected. Four diffraction patterns were collected per point. It should be noted that all diffraction patterns were collected from the inspection volume when it was at 0° . Camera exposure times were quite different for the KDEX and diffraction aspects (4 seconds per image and 3 minutes per diffraction pattern respectively). This is because only a small proportion ($\ll 1\%$) of the primary beam is coherently scattered. It should also be noted that although currently each aspect is collected consecutively, it is hoped that sophisticated collimation could enable both aspects to be obtained simultaneously in the future.

For each diffraction interrogation point the four diffraction patterns were used by the peak tracking algorithm to determine both 2θ and position for each source of diffracted X-rays. These were subsequently compared to a limited material diffraction library to identify the materials. The number of Bragg maxima matching those of a target material for a particular location in a depth plane was used as an index of match certainty.

Material matches were then mapped onto all of the perspective images within the KDEX sequence using the peak tracking algorithm and a simple trigonometric function. Relative distances within the image sequence were calibrated using an internal standard (ruler apparent in the top left hand corner of Figs. 5 and 6).

To colour encode those positions identified as containing a target material the 16 bit tiff files were split into RGB components and the pixel values in the red channel were then multiplied by a weighting factor based on the number of target Bragg maxima identified in that region. In doing so the intensity of red reflects the index of match.

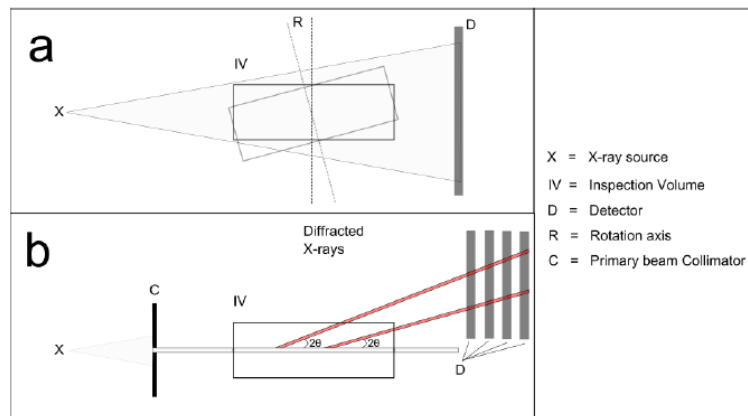


Fig. 1. A schematic illustrating the KDEX configuration (a) where the inspection volume is illustrated normal to the primary beam and a superposition at a given degree of rotation. The diffraction configuration (b) is also illustrated with the same detector positioned at differing distances along the primary beam path.

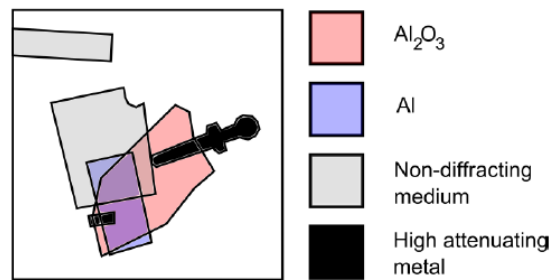


Fig. 2. Schematic illustrating the material composition of objects within the phantom.

3. Results

3.1 Peak finding algorithm

Examples of diffraction patterns measured at one interrogation point with different detector positions are shown in Fig. 3. The change in peak position between each detector location is a function of (i) the scattering angle and (ii) the diffraction source location along the primary beam. Figure 4 illustrates the positions (scattering angle) of a group of Bragg maxima tracked to a single location (± 0.5 mm of each other) on the primary beam path within the inspection volume. These tracked Bragg maxima positions compare well with those for Al_2O_3 also demonstrated in Fig. 4 (the target in this example). Interrogation points that passed through other scattering materials (i.e. Al) also gave rise to diffraction signatures but were tracked to a different location in the inspection volume. It should also be noted that although in this example only one material is targeted and identified the technique has been shown previously to resolve multiple unknown materials overlapped in depth [21].

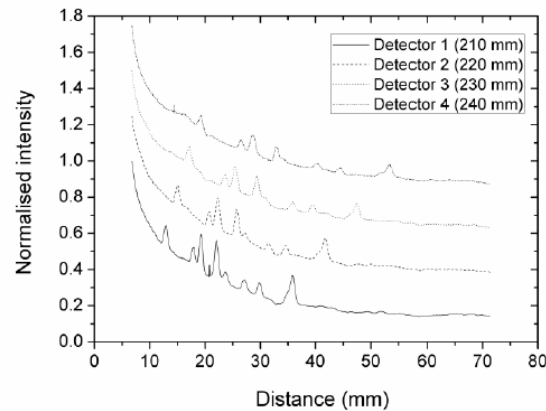


Fig. 3. Four diffraction patterns obtained from a single interrogated point in the inspection volume collected at specified distances along the primary beam path.

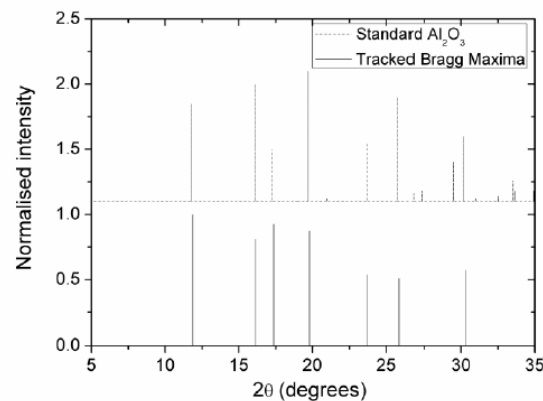


Fig. 4. Bragg maxima tracked to a single location in depth using the diffraction patterns illustrated in Fig. 3 and the peaking finding algorithm. The Bragg maxima correlate well with Al_2O_3 .

3.2 Combined KDEX and diffraction imagery

The 0° KDEX image is illustrated in Fig. 5a and the diffraction inspection points are superimposed in Fig. 5b. Bragg maxima for each interrogated point were compared to a limited target database. Four points (indicated in Fig. 5c) were found to contain Bragg maxima consistent with the target material (Al_2O_3) at a specific depth within the object. It should be noted that these four points were the only ones to pass through the target material. The areas directly adjacent to the interrogated points with diffraction peaks corresponding to the target material were tagged. The red channel intensity values in these tagged areas were increased depending on the number of peaks that match the target diffraction pattern. Thus a greater red intensity indicates a greater confidence in the match between the unknown and

target material. Positions of the tagged areas were recalculated for the other KDEX perspectives using a trigonometric function and the position of the diffracting material produced by the peak tracking algorithm. This results in the tagged areas following the target material position within the transmission images. This effect is best appreciated when the images are demonstrated in a movie format. Figure 6 illustrates snapshots of the accompanying video at differing perspectives.

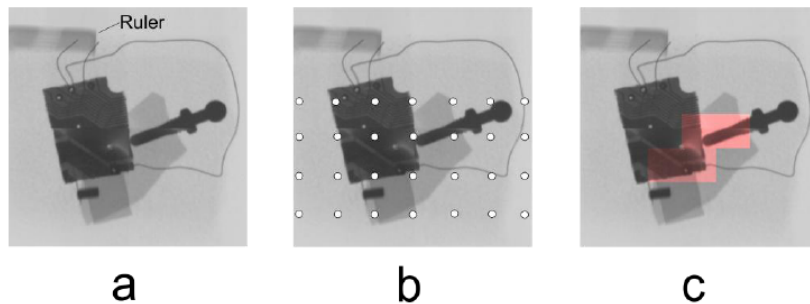


Fig. 5. A transmission image of the inspection volume at 0° rotation (a). The locations of the 28 interrogated positions that were illuminated by pencil beams (b). The tagged regions illustrating areas identified to contain threat materials (c). The end on projection of a ruler can be seen in the top left hand corner of each image, this can be better appreciated in the accompanying video.

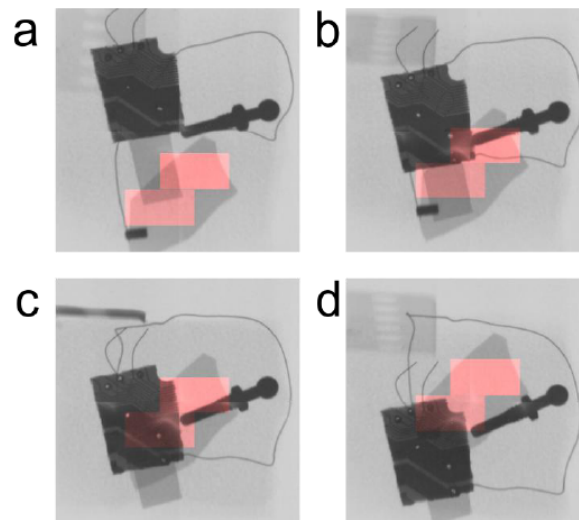


Fig. 6. Selected transmission images from a KDEX image sequence. Images are from -15° (a), -5° (b), $+5^\circ$ (c) and $+15^\circ$ (d) rotation of the inspection volume. Areas reported to have a target material based on their diffraction pattern have been colour encoded in red. The intensity of the red channel in the tagged areas reflects the certainty with which the area is believed to contain the target. ([Media 1](#))

4. Discussion and conclusion

We have demonstrated that the 3-dimensional capability of the X-ray imaging technique, KDEX, has been combined with the material discriminating abilities of X-ray diffraction. The

fusion of these differing, complementary techniques has shown the potential to be a powerful diagnostic tool. The benefits of KDEX (i.e. the increase in morphological recognition performance in the presence of complicated structures) over conventional 2-dimensional (static) transmission imagery is realised with the simultaneous addition of chemically specific spatially resolved information. Potential applications are widespread (e.g. security screening). The radiographic identification of, for example, wiring within a bag, may be further and simultaneously investigated by chemico-structural information provided by associated diffraction signatures which could indicate a threat.

This preliminary study has demonstrated great promise for a new diagnostic tool. However there are several issues that still need to be addressed. Coherently scattered X-rays are of several orders of magnitude lower in intensity than that of the principal X-ray beam. Efforts have been made here to increase the scattering flux by removing the need for scattering collimation. However data acquisition times probably require reducing further if the approach is to be exploited within real world applications. For such applications developments in detector and source technology must be made, although simple beam optics such as those demonstrated by Rogers et al [24] may provide a cost effective solution. Applications may also require a higher X-ray energy than that used here e.g. penetration in security screening applications requires typically $>60\text{keV}$. This energy "scale up" provides its own challenges, in particular a decrease in angular resolution. Moving the detectors further from the diffracting material should act as a positive optical lever (thus mitigating against the loss in angular resolution), however this is at the expense of the system sensitivity (also mentioned earlier) given the inverse squared law and additional air absorption. Presumably larger and/or more sensitive detectors would then be required. These technological improvements and modifications will need to be investigated for energy scale up, although niche markets involving the inspection of smaller attenuating volumes containing poly or semi-crystalline targets may find this technique valuable.

Acknowledgements

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8.6 Poster Presentations

Title	The Separation of X-ray Diffraction Patterns for 3D Imaging
Location	Cranfield Health Student Symposium, Cranfield University, Bedfordshire, UK.
Date	October 2009

Title	Scatter Enhanced X-ray Imaging
Location	Denver National X-ray Conference, Colorado USA.
Date	May 2010

Cranfield Health

The Separation of X-ray Diffraction Patterns for 3D Imaging

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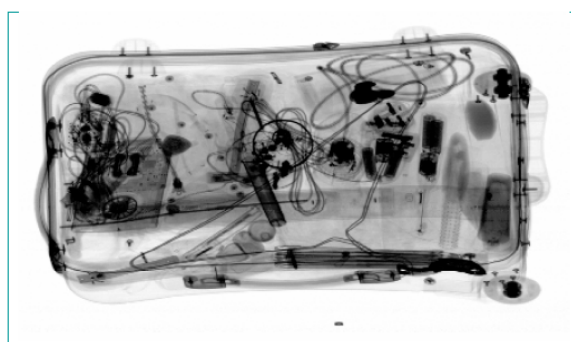
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Introduction

This programme of work demonstrates the first steps toward the development of a new screening technique designed to identify concealed threats such as knives and explosives. On completion this will have unprecedented specificity brought about by the amalgamation of two existing techniques. The 3D power of Kinetic Depth Effect X-ray (KDEX) will be combined with the material discriminating capabilities of X-ray diffraction.

KDEX



Methods/Materials

X-ray diffractograms typically describe a single known point in space. Kinetic Depth Effect X-ray provides a form of volumetric imaging. If a volume is considered then any diffraction pattern obtained may be caused by the summation of diffraction signal from several unknown locations simultaneously. This makes the interpretation of the pattern problematic.

This body of work utilises a multiple view approach to separate combined diffraction patterns into their component patterns as well as their respective locations within the volume. The discrepancy in peak position between the diffractograms can be used to calculate the location of the diffracting material and the true peak angle. These are then compared against a database of known threat materials.

Applications

It is conceivable that this technology could also be employed in the following areas.

- Security Screening
- Medical Imaging
- Industry Quality Assessment

Results

Figure 1. The summation of two X-ray diffractograms from different locations within the inspection volume.

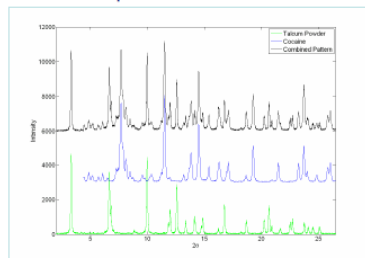


Figure 2. The combined diffraction pattern detected at different rotation positions (views).

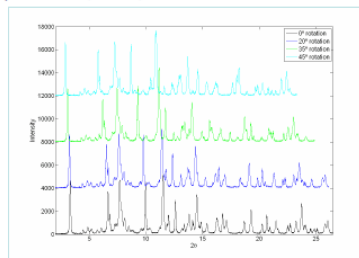
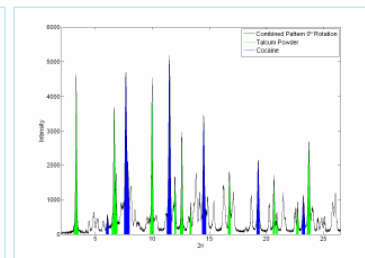


Figure 3. Peaks separated according to their predicted location within the inspection volume.



Conclusions

The rate at which diffraction peaks modify their position in a number of diffractograms collected from different rotations (views) can be used to separate combined patterns. The origins of each peak (i.e. sample position) can also be predicted. Predictions are accurate to within 10% for peak angles below 30° 2θ. This is likely given the radiation energies required in a screening environment.

Scatter Enhanced X-ray Imaging

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Abstract

We present for the first time Kinetic depth effect X-ray Imagery (KDEX) combined with materials specific colour coding obtained via angular-dispersive X-ray diffraction. A novel diffraction sensor geometry (designed to identify the origin of scatter signatures in depth) was applied in raster-scan mode to identify threat materials in the inspection volume. Scatter patterns matching those of threat materials were then “flagged up” in the KDEX imagery to alert the screener.

Context

The security screening industry is faced with the problem of developing technology that can readily detect concealed threats prior to their placement onboard an aircraft. KDEX has been shown to improve the detection performance of typical shapes by providing the screener with 3D information via motion parallax [1]. Explosives however, present a new problem in that they may have an innocuous shape. X-ray diffraction is an effective probe for detecting explosives [2]. The amalgamation of both techniques should have a synergistic effect on threat recognition.

Method

Before a threat material can be identified and thus accurately represented within KDEX imagery we must transform any measured diffraction pattern to that arising from a known spatial origin within the inspection volume. We have employed a novel sensor geometry to obtain diffraction patterns at differing distances along the primary beam path (demonstrated in figure 1). Possible 2θ solutions are back-projected through peak positions on each scatter sensor. The tangent of the gradient formed from the change in peak position between detectors provides the true 2θ values. The sample position is the point on the primary beam that would be intersected by a straight line originating from the peak position on the CCD (at a given detector position) travelling with respect to its measured 2θ .

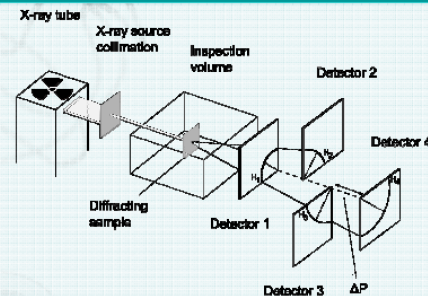


Figure 1. Geometry of the X-ray diffractometer with four detectors superimposed at different positions along the primary X-ray beam path

Results

A inspection volume was raster scanned (with a 7×7 grid) with a pencil beam in transmission mode. Four diffraction patterns were collected at differing distances along the primary beam path for each grid coordinate (similarly to figure 1). The back-projection method was applied at each grid coordinate to identify threat materials in each depth plane (demonstrated in figures 2 and 3). Grid coordinates demonstrating Bragg maxima that match our threat material (Al_2O_3 was used for convenience) were then “flagged up” in red in the transmission imagery. KDEX typically depicts the volume at alternate perspectives to provide a depth sensation. As such the flagged up areas are re-mapped in the alternate perspectives according to the suspect materials calculated location in depth. A selection of these views (A – D) are demonstrated in figure 4.

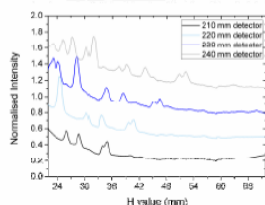


Figure 2. Four diffraction patterns collected along the primary beam path for a single grid coordinate in the inspection volume.

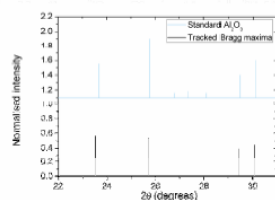


Figure 3. Bragg maxima tracked through the diffraction patterns in figure 2. Peaks correspond well with our threat material (Al_2O_3).

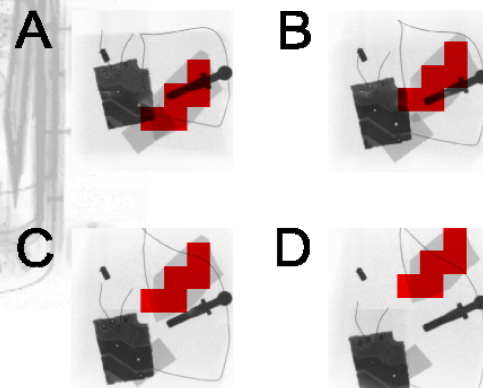


Figure 4. Extracts from a colour encoded KDEX image sequence. A–D demonstrate differing perspectives of the inspection volume. Areas found to harbour a suspect material have been “flagged up” in red.

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Conclusion

Kinetic depth effect X-ray imagery has been united with the materials discriminating ability of angular dispersive X-ray diffraction for the first time. Such a technique promises to enhance the detection of threats such as knives as well as explosives in a cluttered environment. Such a technique could have widespread application in the aviation industry.

Acknowledgments

This ongoing programme of work is funded by EPSRC and is in collaboration with the UK Home Office Scientific Development Branch (HOSDB) and the US Department of Homeland Security (DHS).

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