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DETECTION OF DIAMONDS USING THE TAGGED NEUTRON METHOD

by

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BSc (Radiation and Health Physics) (University of Botswana)

A Dissertation Submitted to the Faculty of Sciences in Fulfillment of the
Requirements for the Award of the Master of Science in Physics of BIUST

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January, 2022

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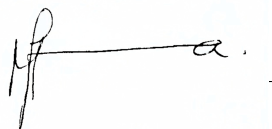
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Abstract

Current technologies for the detection and extraction of diamonds involve the repetitive mechanical crushing of kimberlite ore, resulting in the damage of real diamonds. In particular, the breakage of large diamonds into smaller constituents decreases their value drastically, resulting in huge financial losses to mining companies. This dissertation focuses on testing a newly developed technology for the detection of diamonds in the kimberlite ore prior to crushing processes, thus preventing breakage and retaining their value. This technology is based on the tagged neutron method which involves the irradiation of the ore by fast neutrons of 14.1 MeV and the detection of characteristic gamma rays from the inelastic scattering of neutrons. The criterion for diamond detection is the presence of increased carbon concentration within a certain area of the kimberlite sample.

This dissertation presents the results of the testing of the pilot prototype called Diamant-1000 where the neutron source is a portable neutron generator with inbuilt 256 channel alpha-detector from gallium arsenide. The characteristic gamma-rays are detected by a system of 22 bismuth germanium oxide detectors in coincidence with signals from the alpha detector. The Diamant-1000 prototype was tested on kimberlite ore from the Archangelsk mine in Russia and the Letseng mine in Lesotho. Diamond imitators ranging from 8 to 20 mm are inserted in the kimberlite stones ranging from 30 to 150 mm, which are placed in trays and passed through the neutron module. The minimum size of the diamond imitator detected was 8 mm (5 carats). The prototype had a throughput of about 1 ton/hour (1060 kg/hr) with false alarms rate of 3%. The probability to detect diamonds hidden in kimberlite ore of the range -150 + 100 mm is 95%, whereas for the kimberlite ore within the range of -90 + 40 mm the probability is 97%. The electric consumption of prototype is 0.6 kW/ ton of ore.

The study demonstrates that the tagged neutron method has the potential to be of significant benefit to mining industries in terms of profits, minimum waste and minimal impact on the environment. This method is especially valuable for preventing damages of large diamonds.

Acknowledgements

First of all, I would like to thank my supervisors **Prof. Gregory Campbell Hillhouse** and **Dr. Mikhail Sapozhnikov** for their guidance and support on this work. It was because of their patience that this work was completed.

I would also like to thank the **Joint Institute of Nuclear Research (JINR) in Dubna, Russia**, management for giving me the opportunity to join their members in the development of a tagged neutron sorter.

A special thank you to the **Debswana Diamond Company** for sponsoring my visit to the JINR. Without their support this work would not not have been possible.

Lastly, I would like to thank my family, friends and colleagues for their encouragement throughout my studies. To **Mr. Adolf Mogakolodi**, I thank you a lot for your input in proofreading my work.

Dedication

To my late father, Mr. Meja Sebele, who has been my support since I began my studies. May his soul rest in peace.

Scientific Output

Peer-reviewed Journal Publication

Yury Rogov, Vladislav Kremenets, Mikhail Sapozhnikov and **Motswakae Sebele** “Application of Tagged Neutron Method for Detecting Diamonds in Kimberlite”, MDPI Instruments, Vol. 4, 33, 1-11. Available online at <http://www.mdpi.com/2410-390X/4/4/33/pdf>; accessed on 01/04/2022.

Conference presentations

1. Invited oral presentation titled “Application of Tagged Neutron Method for Detecting Diamonds in Kimberlite” at the X Tastes of Nuclear Physics Conference, hosted virtually by the University of the Western Cape, South Africa, from 30 November to 4 December 2020. Available online at: <http://www.youtube.com/c/NicoOrce>; accessed on 01/04/2022.
2. Oral presentation titled “Application of Tagged Neutron Method for Detecting Diamonds in Kimberlite” at the South African Institute of Physics conference hosted virtually by the North-West University from 22-30 July 2021. Programme available online at: <https://events.saip.org.za/event/206/>; accessed on 01/04/2022.

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List of Abbreviations

ADC Analog to Digital Converter.

AMU Atomic Mass Unit.

BGO Bismuth Germanium Oxide.

DMS Dense Medium Separation.

FNAA Fast Neutron Activation Analysis.

GaS Gallium Arsenide.

JINR Joint Institute for Nuclear Research Dubna, Russia.

NAA Neutron Activation Analysis.

NaOH Sodium Hydroxide (Caustic Soda)

PCI Peripheral Component Interconnect

PFTNA Pulsed Fast-Thermal Neutron Analysis

PGNNA Prompt Gamma Neutron Activation Analysis

TNA Thermal Neutron Analysis

TNM Tagged Neutron Method

tph tonnes per hour

XRF X-ray Fluorescence

XRL X-ray Luminescence.

XRT X-ray Transmission

Introduction

Diamond is one of the most valuable minerals in the world. It is found mostly in igneous rocks such as kimberlite and alluvial deposits such as sand [1]. Although diamond can be found in other rocks, kimberlite is believed to be the main source [2]. Due to the hardness of kimberlite rock, the extraction of diamonds from kimberlite rock is a complex process that requires specialised equipment as well as energy and financial resources. After the initial crushing, which usually does not expose diamonds, the rock is subjected to secondary crushing that reduces the rock to millimeter sized particles so that the desired diamond detection method can be used. Various methods have been developed for the detection and extraction of diamond from kimberlite, including grease technology that has been used for decades, the dense medium separation method (DMS), X-rays fluorescence (XRF), X-ray luminescence (XRL), and the X-ray transmission (XRT) method which is the latest technology in the diamond mining industry [1, 3, 4].

Practically all techniques developed require that the diamond should be exposed from the kimberlite rock. These applications require that the kimberlite rock be crushed to millimeter sizes. Each recovery technique has its minimum and maximum particle size that it can sort. At present, none of the diamond detection techniques are suitable for the extraction of both small and large diamonds. For example, the XRT can recover large diamonds, but can not recover diamonds less than 4 mm in bulk sorting, hence a combination of two methods are usually used to complement each other [5]. This dissertation is dedicated to a new method of diamond detecting diamonds using the tagged neutron method.

1.1 Context and rationale

Botswana is a developing middle-income country whose economy is mainly based on natural resources, with diamonds contributing around 35% to 40% of the economy. The country has a long term vision “Vision 2036” which aims to transform the country’s economy from a middle-income to a high-income economy. The vision has four main pillars which are Sustainable Economic Development, Human Resource Development, Sustainable Environment and Governance, Peace and Security. Through sustainable economic development, the Government of Botswana aims to produce or generate more income from these natural resources with minimal negative impact on the environment.

The Botswana International University of Science and Technology, as a higher education institution, is mandated to help the government to achieve its goals through science and technology. The research focus areas of the institution are aligned with the pillars of the country’s Vision 2036 to produce innovative products and services for sustainable economic developments. This project is aligned with the latter vision and focuses on the development and application of an innovative technology to maximize income generation from the mining of diamonds for the benefit of the citizens of Botswana. In particular, the project aims to transfer the relevant skills, knowledge, and technologies to drive the knowledge-based economy of the country.

1.2 Problem statement

Diamond mining involves the repetitive crushing of kimberlite ore with crushers or grinders to a small size, depending on the top size of the diamond to be recovered. Each detection and recovery method has its minimal and maximal ore size in which diamonds can be detected. For example, the XRT method is limited to kimberlite ore size within the range of 8 to 60 mm in size [5]. Diamonds exist in parts per million (ppm) in the kimberlite ore [6, 7]. Due to this small fraction of existence, a lot of resources are used for processing the barren rock because of excessive crushing to kimberlite ore, the cooling of machines and containing dust [8]. Repetitive crushing does not only waste energy, but also exposes the diamond to a high risks of breakage or damage thus leading to a decrease in the economic value of diamonds [8, 9]. Given the high demand for diamonds in industries such

as jewellery manufacturing industries, research and manufacturing industries, and their scarcity on Earth, better mining methods need to be put in place to avoid damaging this scarce mineral. It is through these challenges currently faced by the diamond mining industry that led to this research project on the early rejection of barren kimberlite using the method of tagged neutrons. If this method is successful, it will reduce the chances of damaging the diamond and also increase chances of recovering large diamonds, and at the same time reducing production costs by saving energy and other resources.

1.3 Objectives

The general objective of this work is to gain a working knowledge and understanding of the physics principles for the application of a recently developed diamond-sorting technique based on the tagged neutron method with the goal of transferring this technology for the benefit of diamond mining industries in Botswana. The deployment of an optimal diamond sorter will allow for minimal damage and maximal recovery of diamonds, especially large diamonds that are of great value, while having a minimal negative impact on the environment. Furthermore, the successful application of this new diamond sorting technology will reduce the financial strains associated with processing barren ore and decrease the chances of diamond breakage that drastically reduces their value.

The specific objectives of this projects are to:

1. Test the new Diamant-1000 prototype that was recently developed by Diamant LLC at the Joint Institute for Nuclear Research in Dubna, Russia, for the detection of diamonds in kimberlite ore using the tagged neutron method; [\[10\]](#).
2. Determine the size of diamond that can be detected using the tagged neutron method;
3. Analyse data for the detection of diamonds in kimberlite extracted from the Archangelsk mine in Russia and the Letseng mine in Lesotho;.
4. Identify the suitability of deploying the tagged neutron method to Botswana

for identifying diamonds, particularly large diamonds, in a real-mining operation.

1.4 Dissertation outline

Chapter 2 presents an overview of the literature on the detection and recovery of diamonds from kimberlite ore. Chapter 3 focuses on the application of the tagged neutron method for the detection of diamonds in kimberlite ore. Chapter 4 presents the experimental setup for the Diamant-1000 prototype, including the components of the prototype, data acquisition system, and user interface. Results and discussion of the tagged neutron method are presented in Chapter 5. Chapter 6 contains the summary, conclusions and recommendations of this study.

Literature Review

This chapter provides an overview of the literature on the various methods used in the detection and recovery of diamonds from kimberlite ore.

2.1 Diamond detection methods

The detection of diamonds in kimberlite ore is mainly done through the use of X-rays, including X-ray fluorescence (XRF) method and X-ray transmission (XRT). The XRF method has long been used in the diamond mining industry, whereas the XRT method is a relatively new technique with the potential to detect large diamonds [11]. In addition, another new method called Mineral-PET (Positron Emission Tomography) has been developed to detect diamonds using positron emission tomography techniques. The above methods are discussed in subsections 2.2.1 to 2.2.3.

2.1.1 X-ray fluorescence method

The X-ray fluorescence (XRF) method uses X-rays to detect diamonds [12]. Mineral concentrates on a conveyor belt are exposed to X-rays. Diamonds fluoresce when exposed to X-rays, activating a photo-detector that triggers a jet of air to deflect diamond into a collector box. This technique is very effective in detecting small luminescent diamonds of about 1 mm in size. The challenge is with type II diamond

(diamonds with little or no nitrogen content) which do not produce light when exposed to X-rays. The XRF method also has problems in detecting diamonds that are covered with layers that prevent the production of light when exposed to X-rays. Furthermore, this method also fails to detect large diamond due to the X-ray absorption by thicker material, resulting in poor detection by photo-detectors [12]. The XRF method is a “hands off” technique and therefore offers a high level of security (from theft by human being) during the final recovery process. This method has been used in the diamond industry for many years.

2.1.2 X-ray transmission method

The X-ray transmission method (XRT) is a new separation technique that uses X-rays to detect diamonds both type I and II diamonds [11, 13]. Type I diamonds contain nitrogen atoms that fluoresce when exposed to X-rays, while type II does not contain nitrogen and hence does not fluoresce when exposed to X-rays. The XRT method can also detect diamonds that are completely obscured by rock or covered with a coating that prevents them from producing light when exposed to X-rays [11, 12]. Type II diamonds are very rare compared to type I. When X-rays penetrate through matter, they get attenuated, where the degree of attenuation depends on the elements present, the density and the thickness of the material, as well as the characteristics of X-rays. Each mineral absorbs high and low energy X-rays in different ways [4]. Systems based on the XRF method are completely enclosed thus offering a high level of security because of the “hands off” nature of the operating system.

The effectiveness of the XRT method in the diamond industry can be traced to the Karowe Mine owned by Lucara Diamond Corporation in Boteti in Botswana. This new technology enabled the company to detect and recover large diamonds including a 1758 carat diamond, a record breaking diamond which is the second largest diamond ever recovered in the world after the discovery of the 3106 carat Cullinan diamond discovered in South Africa in 1905 [13].

The XRT method represents the latest technology in the diamond mining industry and is capable of detecting large diamonds, including non-luminescent diamonds. In 2015, the Lucara Diamond Corporation detected and recovered 813 carat diamond called The Constellation. Within the same week, this corporation also detected and

recovered several large diamonds, including 296 carats, 183 carats, 374 carats, and the so-called Lesedi La Rona 1109 carat diamond [13].

It should be noted that although the Lucara Diamond Corporation succeeded in detecting and recovering these large diamonds in 2015, both The Constellation and Lesedi La Rona diamonds were crushed and the fragments were recovered. The 374 carat fragment of the Lesedi La Rona diamond was recovered a day after the recovery of Lesedi La Rona diamond. Had this diamond not been crushed, it would have weighed 1482 carats (almost 1500 carats) [13].

The Constellation and its fragments are believed to have been a gem of 1291 carats before it was crushed. Had it not been crushed, it could have been larger than the Lesedi La Rona which weighed 1109 carats. The Constellation was sold for 77.649 USD per carat bringing a total of 63.13 million USD [13]. Had this gem not been crushed, the company would have raised around 100.24 million USD for the sale of the 1291 carats diamond.

The Constellation and its fragments are shown in Figure 2.1. For the maximum and minimum size of diamond which can recovered by XRT see Table 2.1.



Figure 2.1: The Constellation and its fragments. Figure 2.1(a) shows the Constellation diamond while figure 2.1(b) and 2.1(c) show its fragments [13].

Visible damage can be seen to the two large diamonds in Figure 2.2. The cause of the damage is unknown, but it is suspected to have been damaged during blasting or during secondary crushing of the ore [13]. Figure 2.2 shows Lesedi La Rona (left) and The Constellation (right) with visible damage to both diamonds.



Figure 2.2: Lesedi La Rona (left) and The Constellation (right) showing the damaged sides [14].

2.1.3 Mineral-PET

Mineral-PET is a new technology to be used for the detection of diamonds. This method uses positron emission tomography (PET). In contrast to PET in medical diagnostics in which a radiotracer is used, in Mineral-PET gamma rays of energies between 22 and 27 MeV irradiate the carbon-12 isotope and transform it to a positron emitter isotope (carbon -11) through the following reaction:

$$^{12}\text{C}(\gamma, n)^{11}\text{C}, \quad (2.1)$$

where ^{12}C and ^{11}C are the carbon isotopes, γ is the incident gamma energy, and n is the emitted neutron.

The emitted positron annihilates with the electron producing back-to-back gamma rays of 0.511 MeV. A matrix of bismuth germanate (BGO) gamma ray detectors is used to detect the gamma rays. [8]. This technique has not been used in an actual mining situation. A diagram for the Mineral-PET method is included in Appendix B (see Figure B.2).

2.1.4 Tagged neutron method

The tagged neutron method (TNM) is a new method used for detecting diamonds [9, 10]. It uses fast 14.1 MeV neutrons to detect diamonds in kimberlite. This

method has not yet been used on an industrial scale in a real mining operation. Further details on TNM are given in Chapter 3.

2.2 Diamond recovery methods

Diamond recovery processes have advanced significantly from the traditional sorting to grease technology and caustic fusion dissolution. Although the recovery techniques have evolved, diamond mining procedure still remains the same. The ore is extracted from the ground, crushed and concentrated. One or more recovery techniques are used. This is because each recovery technique has its own challenges. These techniques include the dense medium separation technique, the grease technology and the caustic dissolution technique which are discussed in this section.

2.2.1 Dense medium separation method

The dense medium separation method (DMS) is used in combination with other diamond recovery techniques [11]. Although DMS is considered as a separating technique, it does not actually recover diamonds, but it makes it easier for the other separating methods such as the grease table method to be used. This method can be best described as a pre-concentration method used before any recovery methods [11], reducing worthless material in the mixture. This technique uses atomic density to separate materials: high density materials settle at the bottom while less dense material floats. Therefore, high density materials settle at the bottom of the centrifuge (hydrocyclones or cyclones) while lighter materials remain suspended in the mixture. Once the ore has been crushed to the desired size, water is added to the mixture. Ferrosilicon is added to the mixture to increase the density of the water solution. The resulting mixture is swirled in high and low speed in cyclones. High density materials settle on the walls of the cyclones while the lighter materials rise in the center of the cyclones and are sucked out, screened and the waste particles removed. After the lighter materials have been removed, a desired recovery technique is then used to recover diamonds from the concentrates.

2.2.2 Grease separating method

The grease method uses the physical properties of diamonds to separate them from the gangue [6]. Diamonds are hydrophobic and oleophilic, so they are attracted to grease while the gangue materials are washed away with water [6]. As a result of these characteristics, when wet ore concentrate flows over a greased table, diamonds stick on the table while the tailings are washed away with running water. The grease with diamonds is heated to melt it and the molten grease containing diamonds is cooled. Since water and grease do not mix, the grease remains suspended in the water while diamonds settle at the bottom of the container. This method has been used for decades and is still used today either as a single recovery method or in combination with other diamond recovery techniques such as X-ray sorting. This technique can recover approximately 98 percent of small diamonds and it is also useful for recovering non-luminescent diamonds which cannot be recovered using X-ray sorting [6]. However, this technique is labour intensive and uses a lot of water. In addition, it is not effective in recovering diamond with coatings, therefore it requires more time for feed preparation and diamonds must be exposed to the surface. Coated diamonds do not stick on grease, as a result they are flushed with running water together with the tailings which poses high risks of diamond loss. During the separation time, the grease must be maintained at the right temperature. The grease separation methods is “hands on” and hence poses the risk of diamond theft during the final recovery process.

2.2.3 Caustic fusion dissolution method

The caustic fusion dissolution method is used to recover micro-diamonds that are usually found in small samples of kimberlites during exploration [15]. Diamonds are resistant to caustic soda (NaOH), while other minerals found in kimberlites dissolve in NaOH, resulting in a 100 percent recovery rate. However this method is not suitable for large scale recovery in a mining environment, because dissolution of kimberlite rocks takes place over a long period (about 14 hours) and it is also labour intensive since large rocks have to be crushed to small pieces resulting in crushing large diamonds to small pieces which drastically reduces their value [15].

Tagged Neutron Method

The tagged neutron method (TNM) is a technique of elemental analysis method that provides information about the three-dimensional coordinates of the elemental concentration of a material [16]. This technique is better suited to analyse the composition of unknown materials [17]. Since neutrons are highly penetrative, they are able to provide more detailed information about the material under examination. This method is non-destructive, and therefore does not affect the chemical composition of the target. It relies on excitation and de-excitation of the material by neutrons [9, 18]. The de-excitation of the target nuclei leads to the emission of gamma rays whose energy is the characteristic of the target material, thus allowing the identification and quantification of the constituent elements [19].

The TNM technique is used in a wide range of applications in various fields such as medicine, environmental monitoring, forensic science and geology [17]. It is one of the most sensitive analytical techniques used in determining the elementary composition of a material [17].

This technique uses a deuterium accelerator that accelerates deuterons towards a tritium target. The interaction of the deuterons and tritium produces alpha and neutron particles with energies of 3.54 and 14.1 MeV, respectively. A position sensitive alpha detector measures the time-of-flight between alpha and prompt gamma ray signals from the neutron-carbon reactions and also provides the momentum and direction of the neutron [19]. Neutrons are uncharged particles and hence difficult to detect. Therefore, one detects the corresponding 3.54 MeV alpha particle emitted during the interaction (see Section 4.2 for neutron production in a neutron

generator). This dissertation focuses on the use of TNM to identify diamonds in kimberlite using carbon signals from prompt gamma rays from neutron-carbon reactions.

The TNM technique has two main processes, namely; the activation and detection processes which are discussed in the next section.

3.1 Activation process

When neutrons bombard the target material, they lose some of their energy in the target, resulting in the excitation of nuclei of the elements that are present. This process is represented by the following reaction equation:

$$n + A \rightarrow A^* + n', \quad (3.1)$$

where n is the incident neutron, A is the target nucleus, A^* is the excited nucleus, and n' is the scattered neutron.

Since the element of interest in kimberlite rocks is carbon (diamond is mainly carbon), only neutron-carbon reactions will be discussed under activation and detection. The incident neutrons scatter inelastically from carbon nuclei, losing energy which excites carbon isotopes as shown below:

$$n + {}^{12}\text{C} \rightarrow {}^{12}\text{C}^* + n', \quad (3.2)$$

where n is the incident neutron, ${}^{12}\text{C}$ is the carbon nucleus, and ${}^{12}\text{C}^*$ and n' are the excited carbon nucleus and the scattered neutron, respectively.

Since the incident neutron is fast, it is not absorbed by the target, but penetrates the target and deposits some of its energy, resulting in excitation of the nuclei which subsequently de-excites by the emission of gamma rays.

3.1.1 Detection process

The investigated material is bombarded with 14.1 MeV neutrons produced from the $^2H - ^3H$ reaction and emits characteristics prompt gamma rays as it de-excites to a stable nucleus [9, 20]. Each and every element produces a unique gamma spectrum that is used as a signature for the presence of elements of interest [9, 18, 17]. This is represented by the reaction equation shown below:

$$A^* \rightarrow A + \gamma, \quad (3.3)$$

where A^* is the excited nucleus, A is the de-excited nuclei and γ is the emitted gamma rays.

Since our element of interest is carbon, Equation 3.3 can be written as follows:

$$^{12}C^* \rightarrow ^{12}C + \gamma. \quad (3.4)$$

Each element emits characteristic gamma-ray radiation, so the presence of the element of interest can be determined by the emitted characteristic gamma rays if they are detected.

Neutrons are highly penetrative compared to X-rays, therefore a larger piece of ore can be irradiated resulting in the possibility to detect large diamonds in rocks. A comparison of TNM and XRT method is shown in Table 3.1 below. The notation $(-A + B)$ mm for ore size means that the largest dimension of the stone is smaller than A mm and the smallest dimension is larger than B mm.

Table 3.1: A comparison between tagged neutron method (TNM) and X-ray transmission (XRT) method in terms of throughput, minimum and maximum size of the ore as well as the stone to diamond size ratio.

	Tagged Neutron Method	X-ray transmission
Throughput(tph)	1	from 60 to 150
Minimum ore size (mm)	-30 +6	-14 +8
Medium ore size (mm)	-75 +40	-32+14
Largest ore size (mm)	-150 +100	-70
Stone/diamond size ratio	10	3

* Throughput for TNM is for experimental purposes and not for commercial use.

* Throughput for the XRT method depends on ore size: 150 tph for large ore, 100 tph for medium ore and 60 tph for the smallest ore size [11].

The XRT method is considered to be the best technique for detecting large diamonds, hence a comparison with TNM is shown in Table 3.1. From Table 3.1, it can be seen that TNM can detect larger diamonds as compared to the XRT method. The TNM method can also detect diamonds in ore that is ten times larger than the diamond as compared to XRT which can detect diamonds in ore that is three times larger than the diamond, see Appendix B, figs B.1 and B.3 for the main steps involved in TNM and XRT recovery processes.

3.2 Neutron sources

Neutron sources are available in different forms such as radioactive isotopes like californium (^{252}Cf) and americium (in the form of americium-beryllium (AmBe)), reactors and accelerator based devices such as neutron generators [21]. All neutrons are produced as fast neutrons (with energy in the range from 10 to 20 MeV). Neutron accelerators provide fast mono-energetic neutrons while radioactive sources can also produce gamma rays that accompany neutrons. These gamma rays increase the background making it impossible to use these sources in prompt gamma neutron activation analyses (PGNNA) [22].

Deuterium - deuterium ($^2\text{H} - ^2\text{H}$) and deuterium -tritium ($^2\text{H} - ^3\text{H}$) neutron generators are the most commonly used and are widely available [22]. In this project, a sealed deuterium-tritium ($^2\text{H} - ^3\text{H}$) neutron generator is used as a source of neutrons and therefore other sources are not be discussed in this dissertation. The choice of the neutron source depends on the incident neutron energy and the interaction mechanism of the incident neutron and the target material. Neutron interaction methods are discussed in Section 3.3. Fast neutrons have high penetrating power compared to thermal and cold neutrons. The characteristics of the deuterium-tritium generator are discussed in Chapter 4.

3.2.1 Deuterium-tritium neutron generator

Deuterium-tritium ($^2\text{H} - ^3\text{H}$) neutron generators are sealed sources because of the presence of harmful radioactive tritium (hydrogen isotope) [22]. Tritium spontaneously decays by beta minus-decay, to a stable helium isotope (^3He), an electron

and an antineutrino [23], as shown below:

$${}^3H \rightarrow {}^3He + e + \bar{\nu} + Q, \quad (3.5)$$

where, ${}^3H$ represents tritium, ${}^3He$ is the helium isotope, e is the electron, $\bar{\nu}$ is an antineutrino and Q is the energy released. Tritium has a half-life of 12.3 years [23, 24].

Deuterium-tritium (${}^2H - {}^3H$) neutron generators produce fast, mono-energetic neutrons [25]. These generators are widely used in various research fields. They have a large fusion cross-section and produce 14.1 MeV neutrons [25]. Neutron production occurs through the following reaction:

$${}^2H + {}^3H \rightarrow {}^4He + n + Q, \quad (3.6)$$

where, ${}^2H$ is the incident particle (deuterium), ${}^3H$ is the target particle (tritium), ${}^4He$ is the produced alpha particle and n is the emitted neutron particle while Q is the total amount of energy. Q produced is shared by the ${}^4He$ and n particles. When the ${}^3H({}^2H, n){}^4He$ reaction occurs, the total amount of energy before the reaction and after the reaction must be equal according to the law of conservation of energy. Therefore;

$$Q_i = Q_f, \quad (3.7)$$

where Q_i is the initial energy before the reaction which is given by:

$$Q_i = m_d c^2 + K_d + m_t c^2 + K_t, \quad (3.8)$$

where $m_d c^2$ and $m_t c^2$ are the rest energies of deuterium and tritium, while K_d and K_t are kinetic energies for deuterium and tritium respectively, and Q_f is the final energy after the reaction given by:

$$Q_f = m_{He} c^2 + K_{He} + m_n c^2 + K_n, \quad (3.9)$$

where $m_{He} c^2$ and $m_n c^2$ are the rest energies of emitted helium and neutron particle, while K_{He} and K_n are the kinetic energies of helium and neutron particle respectively.

Combining Equations 3.8 and 3.9, gives the following equation:

$$Q = [(m_d + m_t) - (m_{He} + m_n)]c^2 \quad (3.10)$$

where Q is the energy released.

It can be noted that the energy produced in this reaction is the result of the change in mass or mass difference between reactants and products which is calculated using Einstein's relation:

$$E = mc^2, \quad (3.11)$$

where m is mass in kg and c is the speed of light.

The mass difference Δ can be calculated as given below:

$$\Delta = M - A, \quad (3.12)$$

where M is the atomic weight of the nucleus and A is the mass number.

The mass difference for the reactants and products is: 13.1359 MeV for deuterium (^2H), 14.9500 MeV for tritium (^3H), 2.4248 MeV for the alpha particle (^4He) and 8.0714 MeV for neutron n [24], (see Appendix A, Table A.1 for selected nuclide). Using the mass difference to calculate the Q value yields 17.57 MeV using Equation 3.10.

In the $^3H(^2H, n)^4He$ reaction, the alpha particle and the neutron particle must have the same momentum [24, 26]. Using the law of conservation of momentum and calculating kinetic energy (K_n) of the produced neutrons, K_n gives a value of 14.05 MeV which corresponds to the value maximum energy value of $\equiv 14.1 MeV$ that is produced by the ING-27 neutron generator in a $^3H(^2H, n)^4He$ reaction while the kinetic energy of the alpha particles (K_{He}) is found to be 3.52 MeV. The neutrons produced by the ING-27 neutron generator are classified as fast neutrons [27, 28]: see Table 3.2 for classification of neutrons according to their kinetic energy.

Table 3.2: Neutron classification in terms kinetic energy.

Neutron Type	Kinetic Energy (eV)
Cold	<0.01
Thermal	0.01 to 0.30
Epithermal	0.30 to 10 000
Fast	>10 000 to 20 000 000
Relativistic	>20 000 000

3.3 Interaction of neutrons with matter

Since the tagged neutron method uses neutrons, the interaction of neutrons with matter is an important part of this work, and should be considered when designing the prototype for shielding. Neutrons are electrically neutral particles with a mass of 1.008 AMU which is slightly larger than the mass of a proton which 1.007 amu [29]. Although neutrons are stable in stable atomic nuclei, free neutrons undergo beta decay to a proton via the emission of an electron and an anti-neutrino with a half-life of 885 seconds (about 15 minutes), emitting gamma rays of 0.7823 MeV [30, 31, 32], as represented by the following equation;

$$n \rightarrow p + \beta^- + \nu_e + Q, \quad (3.13)$$

where, n is the neutron, p is the proton, β^- is the negative electron, ν_e is the anti-neutrino and Q is the energy released.

Neutrons interact with nuclei through the strong nuclear interaction. This interaction has a short range (within 10^{-13} cm of the nucleus), which means that the neutrons have to pass close to the nucleus for an interaction to occur [33]. The small size of the nucleus in relation to the atom results in neutrons having a low probability of interaction so they travel a considerable distance in matter before any interaction takes place, thus making neutrons extremely penetrative. When neutrons interact with matter, they can either be scattered or absorbed [34], depending on the incident energy of the neutron. Figure 3.1, summarises the interactions of neutrons with matter.

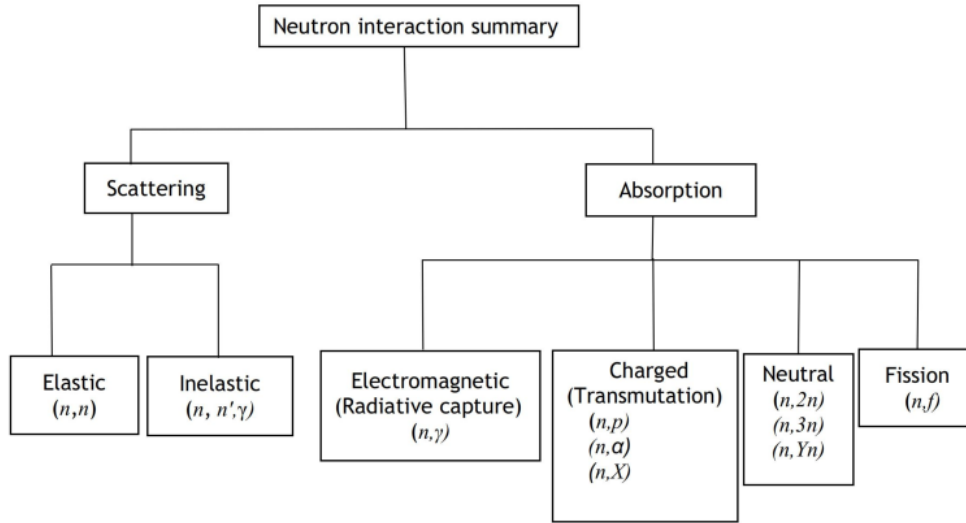


Figure 3.1: Schematic representation of the interaction of neutrons with matter. X represents charged particle emitted while Y is an integer number of neutrons emitted [34].

3.3.1 Neutron absorption

For neutron absorption to occur, fast neutrons must be slowed down (moderated) to thermal neutrons. When this happens, a compound nucleus is formed whereby nucleons in the target nucleus are rearranged. This compound nucleus will then undergo a variety of emissions or decays, such as (n, γ) , (n, p) , $(n, 2n)$ or (n, α) as shown in Figure 3.1 [24, 34].

3.3.1.1 Transmutation, Fission and Natural Reactions

Transmutation, fission and natural reactions occur when the incident neutron is absorbed by the target nucleus. When these reactions occur, the target nucleus is excited as in inelastic scattering reactions. In contrast to inelastic scattering in which gamma rays are emitted during de-excitation of the target, charged particles such as protons, alpha particles and deuterons are emitted. In fission, two fragments are emitted, one with relatively large and one with smaller mass.

3.3.1.2 Radioactive decay

Radioactive decay takes place when a nucleus is unstable. When a nucleus has excess neutrons, for example in the (n, γ) process, or conversely a proton excess as in the $(n, 2n)$ reaction, the nucleus undergoes a beta decay emitting an electron (beta-minus emission) or a positron (beta-plus emission). In beta-minus emission, a neutron is converted to a proton resulting in electron emission and an anti-neutrino to conserve lepton number and the energy Q . In this process, Q represents gamma rays. This process occurs spontaneously when the Q value is positive. The equation below represents the beta minus (β^-) emission:



where ${}^A_Z X^*$ is a neutron rich nucleus, ${}^A_{Z+1} Y$ is a daughter nucleus, (e^-) is a positron and $\bar{\nu}$ is an anti-neutrino.

In case of beta-plus (β^+) emission, excess protons such as those in $(n, 2n)$ reactions can decay through positron emission. This happens spontaneously as with the e^- emission when the Q value is positive. The beta-plus emission equation is shown by the reaction equation below:



where ${}^A_Z X^*$ is a proton rich nucleus, ${}^A_{Z-1} Y$ is a daughter nucleus, e^+ is a positron and ν is a neutrino.

In this reaction, a proton is converted to neutron and this is accompanied by the emission of a positron (e^+) and a neutrino (ν) in order to conserve charge and lepton number as shown in Equation 3.15 above. The positron produced annihilates with a free electron producing two gamma rays with an energy equivalent to 0.511 MeV which is equal to the rest mass of an electron or positron.

3.3.1.3 Radiative capture

Since fast neutrons are moderated to thermal energies, they can be captured by the nuclei of the target. When this happens, the atomic mass number of the target increases by 1 to form a compound nucleus of mass $(A + 1)$ in an excited state.

Figure 3.2, shows processes resulting from radiative capture of a neutron. In Figure 3.2, the excited nucleus so formed is shown to de-excite by the emission of delayed gamma-rays and a β particle.

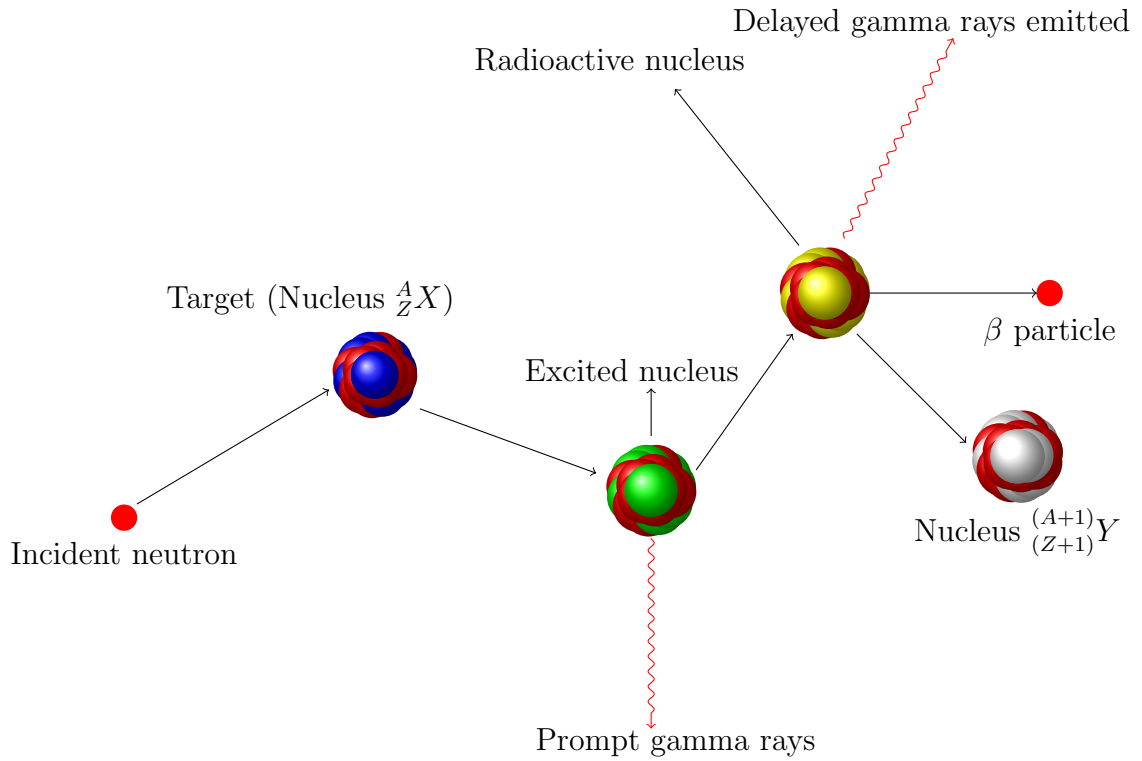


Figure 3.2: Radiative capture of a neutron: an incident neutron is absorbed by the target nucleus and the target becomes excited. The target nucleus de-excites by the emission of prompt gamma rays followed by a beta particle and delayed gamma rays.

The excitation energy in the compound nucleus is equivalent to the total of the kinetic energy of the incident neutron and the separation energy (the energy required to remove a particle or nucleon from the nucleus of an atom), defined as [35]:

$$S_n = [m(A) + m_n - (A + 1)]c^2, \quad (3.16)$$

where, $m(A)$ is the mass of the target nucleus, m_n is the mass of the incident neutron and $(A + 1)$ is the mass of the product nucleus.

The probability for this interaction to occur depends on the energy of the incident neutron and also on the separation energy of the neutron. The incident neutron should have a thermal energy of 0.025 eV. The neutron separation energy is about 8 MeV and this energy is released during the interaction in the form of gamma rays. The compound nucleus that is formed decays to ground state by emitting gamma rays. If, after the emission of gamma rays the nucleus $(A + 1)$ is still radioactive, it decays to stability in accordance with its half-life.

3.3.2 Neutron scattering

When neutrons penetrate matter, they collide with the atomic nucleus and scatter either elastically or inelastically.

3.3.2.1 Elastic scattering

Elastic scattering is more pronounced with slow neutrons than with fast neutrons. Since the kinetic energy of slow neutrons is very small, little energy is transferred to the target nucleus [36]. This type of interaction is known as a billiard-ball collision, in which kinetic energy and momentum are conserved [24, 37]. When this process occurs, no energy is transferred from the neutron to excite the target nucleus [24, 37]. The target gains the amount of kinetic energy that is lost by the incident neutron. The scattering angle is determined by the conservation of momentum. The kinetic energy of the recoiled nucleus depends on the recoiling angle of the nucleus. The maximum energy transferred from an incident neutron to the target nucleus occurs in head-on collision. The energy lost from the incident neutron is fully transferred to the recoiling nucleus. Figure 3.3 shows elastic scattering of neutrons.

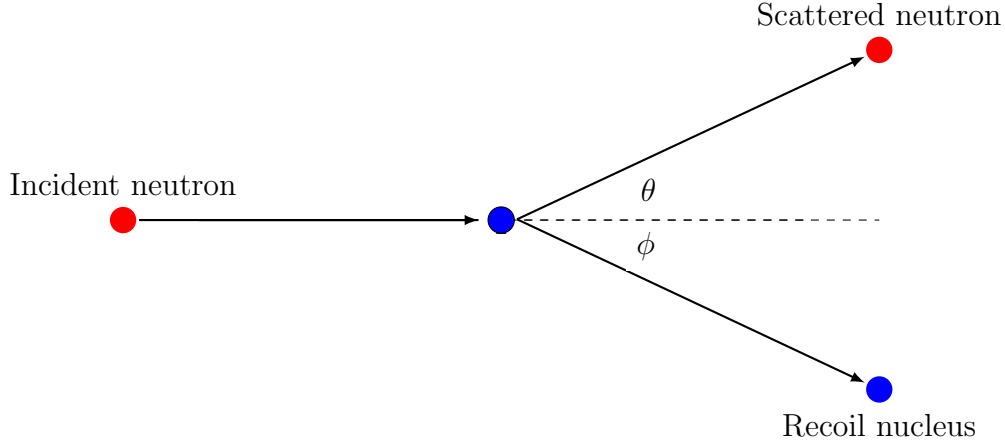


Figure 3.3: Schematic representation of the elastic scattering of a neutron with a nucleus.

When a neutron collides with the target, it is slowed down (moderated) to lower energy. The most efficient neutron moderators are elements with a low atomic mass number. Elements with a high atomic mass number, such as lead, only partially absorb the energy of the neutron. Elements with a low atomic mass number, the lightest of which is hydrogen, can absorb all the energy of the neutron because hydrogen consists of only a single proton with a mass that is almost equal to that of the neutron. The maximum amount of energy that an incident neutron can lose in a single collision and which is then absorbed by the target can be calculated using the following equation [38]:

$$E_R = \frac{4A}{(1+A)^2} E_n, \quad (3.17)$$

where E_R is the recoil nucleus energy, A is the atomic mass number of the target nucleus and E_n is the incident neutron energy.

If the scattering occurs at a certain angle, the maximum energy lost by a neutron can be calculated using the following equation:

$$E_R = \frac{4A}{(1+A)^2} (\cos \theta) E_n, \quad (3.18)$$

where θ is the scattering angle.

Table 3.3 shows the maximum fractional energy lost by the neutron in a single collision. The energy lost by the incident neutron is absorbed by the target nucleus. Table 3.3, clearly shows that elements with high atomic numbers are poor

absorbers of neutron energy. Hence elements with low atomic numbers are mainly used for shielding neutrons.

Table 3.3: Maximum fractional recoil energy lost by a neutron in a single collision with an element calculated using Equation 3.17.

Target nucleus	Mass number (A)	Fractional Recoil Energy (keV)
^1H	1	1.0000
^7Be	7	0.4375
^{10}B	10	0.3306
^{12}C	12	0.2840
^{16}O	16	0.2214
^{27}Al	27	0.1378
^{56}Fe	56	0.0689
^{118}Sn	118	0.0191
^{238}U	238	0.0167

3.3.2.2 Inelastic scattering

Inelastic scattering occurs with fast neutrons. The incident neutron must have enough energy to excite the target nucleus so that a sufficient amount of energy is transferred in a single collision. When this happens the recoil nucleus receives enough energy for the event to be detectable. During an inelastic scattering process, the incident neutrons collide with the target nucleus and transfer its energy to the target, leading to re-arrangement of the nucleons and resulting in the formation of a new compound nucleus in an excited state. The compound nucleus formed de-excites by emitting gamma rays and a neutron.

The gamma rays emitted by the test material can be classified into two categories, prompt and delayed gamma, according to their emission time. Figure 3.4 shows the inelastic scattering of a neutron in which only prompt gamma rays are emitted. After the emission of prompt gamma rays and a neutron, the excited nucleus returns back to its original state as shown in Figure 3.4.

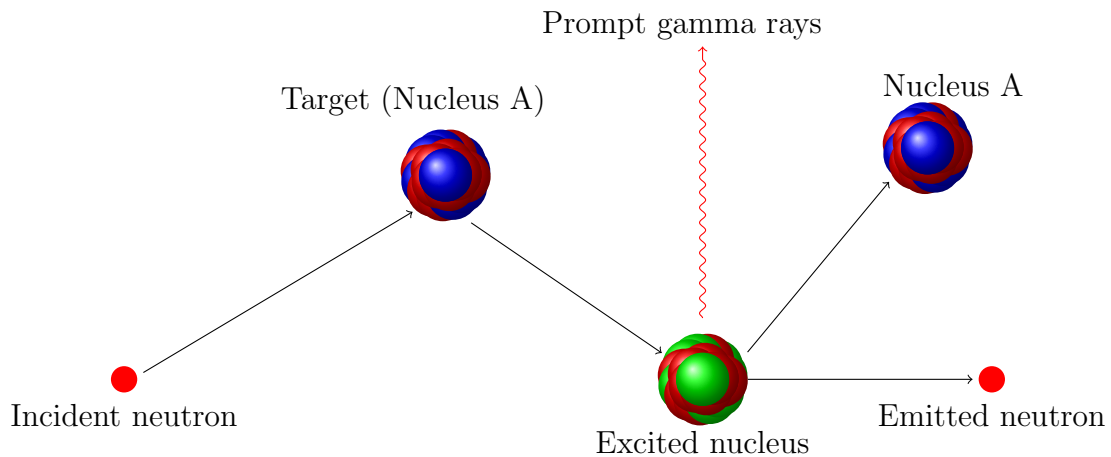


Figure 3.4: Prompt gamma ray emission in the inelastic scattering of a neutron. The incident neutron interacts with the target nucleus which de-excites through the emission of the prompt gamma rays and a neutron.

3.3.3 Delayed gamma-ray neutron activation analysis

After emitting prompt gamma rays, the sample may remain unstable. The unstable nucleus will then undergo radioactive decay (see Section 3.3.1.3 on radioactive decay) and then emits gamma rays to become stable. The gamma rays emitted

during this process are called delayed gamma emission. This process is commonly used in neutron activation analysis (NAA) when analyzing samples using thermal neutrons and it is suitable for the analysis of long-lived radionuclide [39].

3.3.4 Prompt gamma-ray neutron activation analysis

During irradiation with neutrons, the target de-excites by emitting prompt gamma rays. This method is suitable for short-lived radionuclides [39]. This type of neutron activation analysis is used to analyse the target material with fast neutrons. For example, the gamma rays emitted from kimberlite ore when searching for diamonds are prompt.

After the emission of prompt gamma rays, the nucleus can remain in an excited state and then emit beta particles (see Section 3.3.3 for delayed gamma ray emission).

3.3.5 Neutron cross-section

The neutron interaction rate depends on the incoming neutron flux and the cross-section for the interaction. The neutron cross-section determines the probability that a neutron will interact with other materials. It depends on the nucleus involved and also depends on the energy of the neutron. Neutron cross-sections are smooth functions at low energies while at high energies they are mostly dominated by resonances due to the presence of favorable states in the compound nucleus at these energies. Neutron cross-sections are divided into two types, microscopic cross-section and macroscopic cross-section [34].

3.3.5.1 Microscopic cross-section

The microscopic cross-section is considered to be effective area that the nucleus presents to the neutron for a reaction. A large effective area increases the probability of the reaction. When a mono-energetic beam of incident neutrons is directed to a target material of thickness (t), some neutrons are absorbed, some penetrate through the material without interacting with the target, while some neutrons are scattered and emerge with a reduced energy and a change of the original direction. [40]. The probability of occurrence of these events is determined by the cross-

section. The neutron cross-section depends on the energy of the incident neutron as well as the atomic weight and the atomic mass number of the target nucleus. If a mono-energetic beam of incident neutrons is directed onto a target material of thickness (t), area (α), the rate of reaction (R) that takes place is directly proportional to the parameters shown in Figure 3.5 [21].

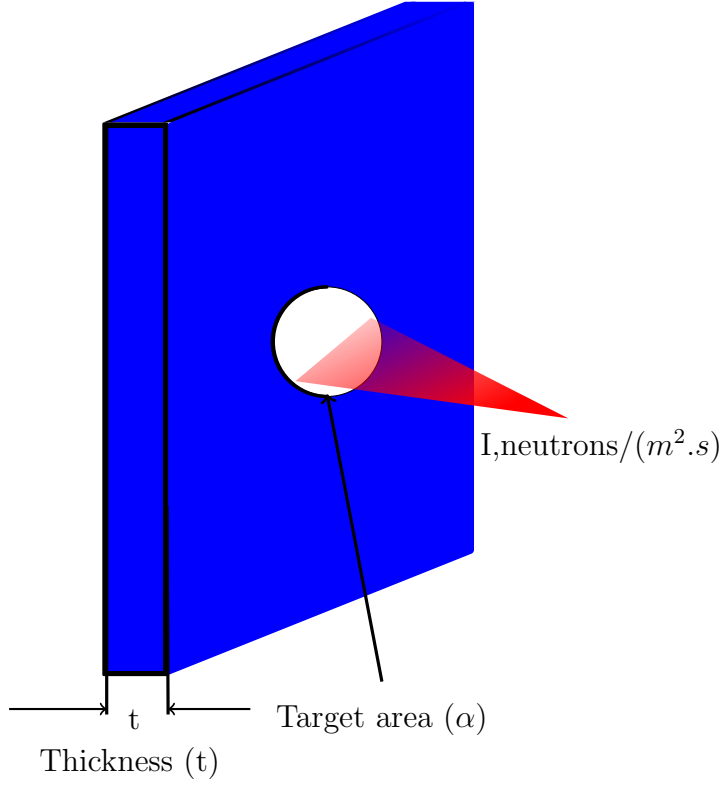


Figure 3.5: Mono-energetic neutron beam incident (I) on a target material of thickness (t).

The rate of reaction (R) is directly proportional to the cross-sectional area, the neutron velocity, and neutron density (ρ), the thickness of the target material and the density of the target, as represented by the equation below [21]:

$$R \propto \rho_n \alpha t v_n N, \quad (3.19)$$

where ρ_n is the neutron density, v_n is the neutron velocity, α is the cross-sectional area of the target, t is the thickness of the target and N is the density of the target material.

After introducing the constant of proportionality (σ), to the Equation 3.19, it can

be written as follows:

$$R = \sigma \rho_n \alpha t v_n N, \quad (3.20)$$

where all parameters have the same meaning as in Equation 3.19.

The cross-section and thickness of the material give the volume of the target. The number of neutrons reactions that occur per unit volume per second can be determined using the following equation:

$$R' = \frac{R}{\alpha t} = \frac{R}{v} = \sigma \rho_n v_n N \quad (3.21)$$

From Equation 3.21, making the constant of proportionality (σ) the subject of the formula, gives the following expression for the microscopic cross-section:

$$\sigma = \frac{R'}{\rho_n v_n N} \quad (3.22)$$

$\rho_n v_n$ gives the neutron flux (Φ). The microscopic cross-section is measured in barns where 1 barn is equal to 10^{-24}cm^2 or 10^{-28}m^2 and is given by Equation 3.23 [21, 24]:

$$\sigma = \frac{R'}{\Phi N} \quad (3.23)$$

The neutron cross-section is defined separately for each type of interaction that takes place [21]. These cross-sections are as follows:

1. σ_{el} for elastic scattering cross-section
2. σ_{in} for inelastic scattering cross-section
3. σ_a for absorption cross-section
4. σ_c for capture cross-section
5. σ_f for fission cross-section

The total cross-section is equivalent to the sum of all cross-sections [21, 40]. Since neutrons interact with the target either by scattering or absorption, the total cross-section can be given by the sum of the two interactions [36], which is shown in Equation 3.24 below:

$$\sigma_{total} = \sigma_{scat} + \sigma_{abs} = \sigma_{in} + \sigma_{el} + \sigma_f + \dots \quad (3.24)$$

3.3.5.2 Macroscopic cross-section

The interaction of a neutron within a certain volume of material does not only depend on the microscopic cross-section, but also on the number of nuclei in that volume. The macroscopic cross-section is the probability of a neutron reaction to occur per unit length traveled and is given by:

$$\sum_t = \sigma_t N, \quad (3.25)$$

where N is the atomic density of material (number of atoms of a given type per unit volume of the material) and σ_t is the microscopic cross-section of the nucleus.

For a compound that consists of different elements, the total macroscopic cross-section is the summation of the macroscopic cross-section for each element which is given by:

$$\sum = \sum_1 + \sum_2 + \sum_3 + \sum_4 + \dots \quad (3.26)$$

The atomic density N_i for an individual element is calculated using the following equation:

$$N_i = \frac{\rho N_A n_i}{M} ((n_1 \sigma_1) + (n_2 \sigma_2) + (n_3 \sigma_3) + \dots), \quad (3.27)$$

where ρ is the density of material in kgm^{-3} , N_A is Avogadro's number (6.022×10^{23} per mole), and M is the atomic weight in kg mole^{-1} while n_i is the total number of atoms present in one molecule.

From Equations 3.26 and 3.27, the macroscopic cross-section can be written as:

$$\sum = \frac{\rho N_a}{M} ((n_1 \sigma_1) + (n_2 \sigma_2) + (n_3 \sigma_3) + \dots), \quad (3.28)$$

3.3.5.3 Mean free path

When a mono energetic beam of neutron penetrates through the target of thickness (t), some neutrons are absorbed while others are scattered and change direction, and some emerge without interactions as they traverse through the target. Those that emerge without interaction are calculated using the equation below:

$$I_t = I_o e^{-\sum_{tot} t}, \quad (3.29)$$

where $\sum_{tot} = \sum_a + \sum_s$ represents the total macroscopic cross section and (t) is the thickness of the target material.

The average distance between two successive interactions is the mean free path which is calculated using the equation below [21]:

$$\lambda = \frac{1}{\sum_{tot}}, \quad (3.30)$$

where $\sum_{tot}$ is the total macroscopic cross section. Equation 3.30 can be written as:

$$I_t = I_o e^{\frac{-t}{\lambda}} \quad (3.31)$$

3.4 Interaction of gamma rays with matter

Gamma rays are electromagnetic radiation of nuclear origin with energies ranging from 100 keV to approximately 10 MeV energy with high frequency and short wavelength. They have no electrical charge and hence cannot be deflected by magnetic fields. Gamma rays are generated by the decay of unstable states in atomic nuclei from a higher energy state to a state of lower energy. [41]. When gamma rays interact with matter, absorption or scattering processes can occur. The absorption is proportional to the material density, the cross-section and the thickness of the material. The absorption process decreases exponentially with decreasing intensity and increasing distance from the source. The equation below shows the exponential attenuation of gamma rays by an absorber as a function of x , the distance from the incident surface [42]:

$$I_x = I_o e^{-\mu x}, \quad (3.32)$$

where $\mu = n\sigma$ is the absorption coefficient measured in cm^{-1} , n denotes the number of atoms per cm^{-3} of the material (atomic density), and σ represents the absorption cross-section in cm^2 .

Although there are many interactions, the most important are the photoelectric effect, Compton scattering and pair production [36]. All these interactions depend on the incident energy of the photons and the atomic number (Z) of the material they interact with. When the incident photon with low energy interacts with high

For high Z materials, the photoelectric effect becomes the main interaction process. Compton scattering is more pronounced for low Z materials and photons of intermediate energy, while pair production is dominant at high photon energy. Figure 3.6 shows the dependence of energy and atomic number for these three main types of gamma ray interaction processes with matter.

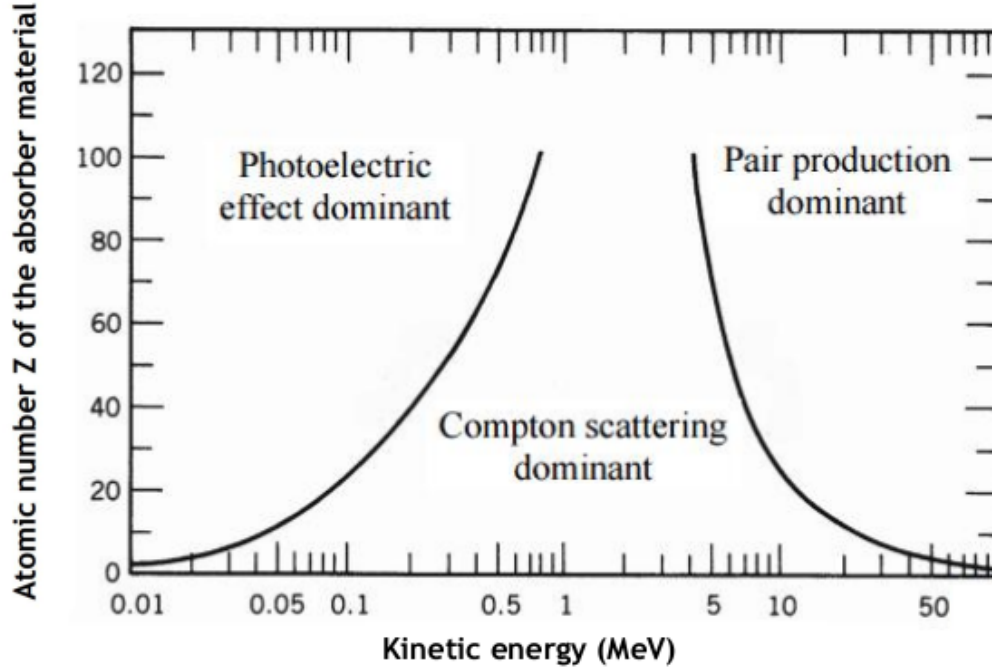


Figure 3.6: A graph of the atomic number Z of the absorber material versus kinetic energy showing three main types of gamma ray interaction with matter namely: the photoelectric effect, Compton scattering and pair production [43, 44, 45].

3.4.1 Photoelectric effect

In the photoelectric process, an incident gamma ray is absorbed by the atomic electron, and absorber electron is then ejected from the atomic shells (K-shells in most cases) [42] as shown in Figure 3.7.

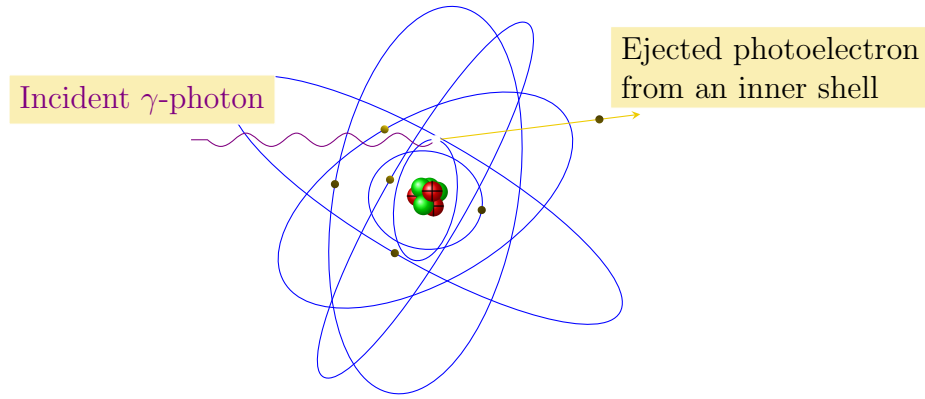


Figure 3.7: Photoelectric effect: an incident gamma ray is absorbed by an atomic electron resulting in emission of an inner electron.

The photoelectric effect occurs when the incident photon has more energy than the binding energy of the atomic electron. If the binding energy is greater than the incident photon energy, the electron cannot gain enough kinetic energy to escape from the shell [24]. In this interaction, the gamma rays transfer the following energy [42] to the electron: [24].

$$E_k = h\nu - E_b, \quad (3.33)$$

where E_k is the kinetic energy of the ejected electron, $h\nu$ is the incident photon energy, while E_b is the binding energy of the ejected electron.

After an electron is ejected, an electron vacancy is created and the atom becomes an excited ion. The vacancy caused by the ejection of the electron is quickly filled through electron capture of a free electron or through internal arrangements of the atomic shell structure, resulting in the emission of characteristics X-rays with energy equivalent to the energy difference between the two energy levels. An Auger electron can be ejected instead of X-rays resulting in de-excitation of the excited ion. [36].

The photoelectric probability depends on the gamma ray energy, the electron binding energy, and the atomic number of the nucleus. The probability is high in tightly bound electrons and usually takes place in K-shells. The probability of the photoelectric effect is given by the following equation below [24]:

$$\tau \propto \frac{Z^n}{E_y^{3.5}}, \quad (3.34)$$

where n is a value either 4 or 5 over the energy region of interest and τ is the photoelectric mass attenuation coefficient.

The dependence of the photoelectric absorption probability on the atomic mass number is the main reason for the use of high Z material in gamma shielding.

3.4.2 Compton scattering

As previously mentioned, Compton scattering occurs at intermediate energy (see Figure 3.6. When the incident energy increases, the probability for the photoelectric effect to occur decreases, while the probability for Compton scattering increases. In Compton scattering, the incident gamma ray loses part of its energy through interaction with an atomic electron [42], and the gamma ray is scattered at an angle from its initial direction with a reduction in kinetic energy as shown in Figure 3.8 [37, 36]. The ejected electron gains kinetic energy of a magnitude equal to the energy difference between the incident gamma energy and the deviated gamma ray. This amount of gamma ray energy after scattering is calculated using the Compton scattering formula [24, 36]:

$$\Delta\lambda = \frac{h}{(mc)(1 - \cos \theta)}, \quad (3.35)$$

where mc is the rest mass energy of an electron (0.511 MeV) and h is the Plank's constant. The maximum energy transfer occurs when θ is 180° . Figure 3.8 illustrates the Compton scattering process.

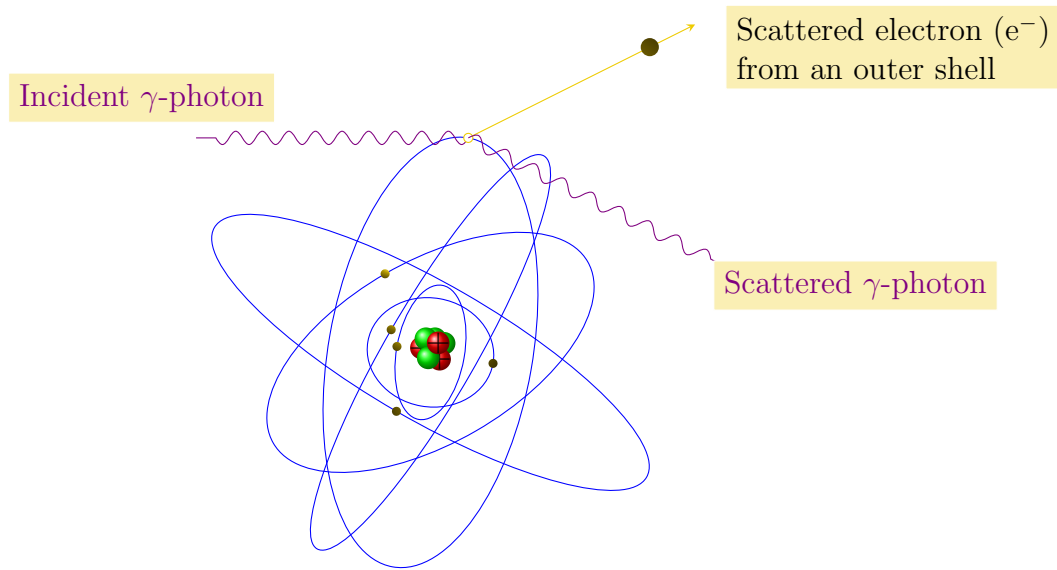


Figure 3.8: Compton scattering: the incident gamma photon interacts with an electron in the outer shell and scatters.

3.4.3 Pair production

Pair production occurs when a gamma ray interacts with the electromagnetic field surrounding the atomic nucleus and converts the gamma ray into a positron and an electron. For this to happen, the incident gamma ray must exceed more than twice the rest mass energy of an electron of 1.022 MeV (the sum of two rest mass energies of an electron or positron) [36]. The probability for pair production to occur increases with gamma ray energy and is the main decay mechanism in an absorber medium above 5 MeV energy [36]. The excess energy of the photon is conserved as kinetic energy and is shared between the positron and electron [36]. Figure 3.9 shows the production of the positron and electron in pair production.

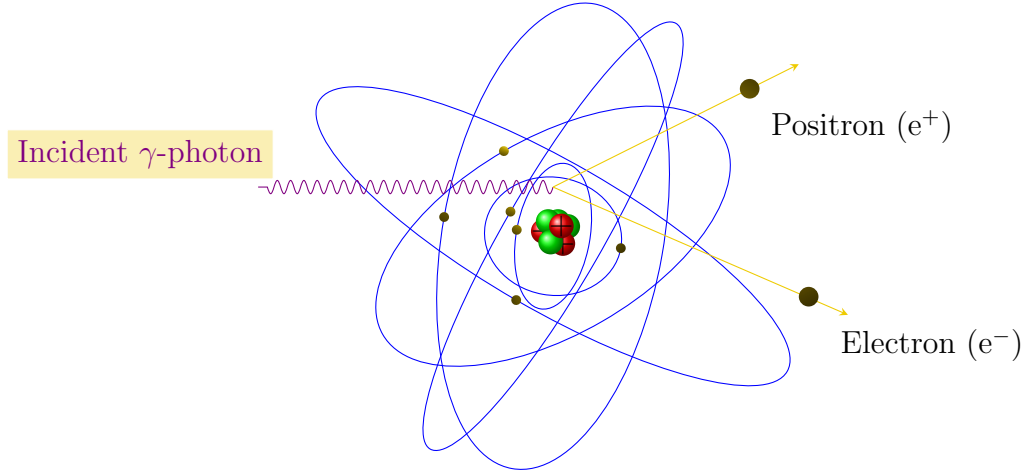


Figure 3.9: Pair production: an incident gamma interacts with the electron field surrounding the atomic nucleus converting the gamma ray into a positron and an electron.

3.4.4 Gamma detection

In this work, the choice of a gamma detector is crucial. The physical parameters and limitations of a gamma ray detector should be considered before making an appropriate selection. The detector should be made from a high Z material in order to detect photons with high energies. Furthermore, the detector able to resolve peaks of interest with minimum interference when the detector is irradiated with neutrons. Table 3.4 below shows a comparison between commonly used inorganic scintillators in the tagged neutron method in terms of their properties [46]. In this work, bismuth germanium oxide (BGO) detectors were selected as the best option (see Section 4.4 for more information about BGO detectors).

Table 3.4: Comparison of commonly used gamma detectors in terms of their properties.

Material	Peak Emission (nm)	Light yield (photons/MeV)	Density (g/cm ³)	Attenuation at 1.5 MeV	Hygroscopic	Energy Resolution at 661 keV	Decay Time (ns)
NaI(Tl)	415	38000-55000	3.67	114.7	Yes	7	250
BGO	480	8000-10 00	7.13	222.8	No	9.7-16	300
LaBr ₃	380	63 000	5.08-5.22	158.8	Yes	2.6-3.5	16

3.4.5 Detector efficiency

The most important feature of a detector is its ability to absorb the full gamma energy with sufficient energy resolution: this is very important for the accurate identification of elements from where the gamma originated. Gamma detectors with crystals made from materials with high atomic number are the best since they are able to absorb both low and high gamma rays. The ability of a detector to measure radiation depends on various parameters such as the detector geometry, and intrinsic and photo-peak efficiency of the detector. The total efficiency of a detector is a function of detector geometry and the probability that photons interact with the detector. The latter probability depends on the properties of the material from which the detector is made off, it is given by [47, 48]:

$$P = 1 - e^{-\epsilon_{total}}, \quad (3.36)$$

where ϵ_{total} is the absolute efficiency defined by the ratio of the number of counts recorded by the detector to the number of gamma rays emitted by the source, namely:

$$\epsilon_{abs} = \frac{N_C}{N_S}, \quad (3.37)$$

where N_C is the number of counts recorded by the detector while N_S is the number of gamma rays emitted by the source.

Geometric efficiency is a fraction of the total radiation quanta emitted by the radiation source impinging the radiation detector material [47] and depends on the size and shape of the detector, radioactive source, and the distance between the source and the detector.

Intrinsic efficiency is the ratio of the number of pulses recorded by the detector to the number of gamma rays impinging on the detector and is given by: [48]

$$\epsilon_{int} = 1 - e^{-\mu(E)x}, \quad (3.38)$$

where $\mu(E)$ is the linear attenuation coefficient and x is the thickness of the radiation detection material.

3.5 Neutron interrogation method

Neutron interrogation methods include thermal neutron analysis, pulsed fast-thermal neutron analysis, and fast neutron activation analysis. These interrogation methods use different neutron energies and therefore each of these methods has its unique advantages and disadvantages. The selection of the source is based on the element to be identified and the neutron flux produced by the neutron source.

3.5.1 Thermal neutron analysis

In thermal neutron analysis (TNA), neutrons with energies ranging from 0.01 to 0.3 eV. This method uses the characteristics of gamma rays produced by produced by different elements in the target material. The target is exposed to the thermal neutron flux which induces radioactivity in the target material through the (n,γ) interaction resulting in the emission of the gamma rays which can be analysed with a suitable detector. These gamma rays emanate from the compound nucleus formed after neutron capture reaction.

3.5.2 Pulsed fast-thermal neutron analysis

Pulsed fast-thermal neutron analysis (PFTNA) technique uses a combination of fast neutrons and thermal neutrons to reduce background radiation in fast neutron analysis. In this interrogation technique, neutrons are released in pulses, hence the name pulsed fast-thermal neutron analysis. Releasing neutrons in a short pulse allows for any gamma rays that are created during the de-excitation to reach the detector before the scattered neutrons that accompany them, allowing the gamma rays to be counted first. Gamma rays that are created due to neutrons interacting with the detector are not counted. Some fast neutrons are thermalized through multiple interactions with the target. Thermalized neutrons can be absorbed the same way as in the thermal interaction process and release prompt gamma rays. PFTNA performs two separate interrogations simultaneously (PFTNA and TNA interrogations). The neutron generator used in PFTNA is cycled to allow thermalisation between the pulses. For this to happen, the target is irradiated with fast neutrons for 10 μs and gamma rays are detected, thereafter the generator is switched off for 90 μs to allow neutrons to thermalise in the target and be absorbed

by the target nuclei and become excited. An excited nucleus will de-excite through the emission of gamma rays. This multiple detection of the target is most suitable for identifying the elementary composition of the target material.

3.5.3 Fast neutron activation analysis

The fast neutron activation analysis technique uses neutrons of high energy to analyse the target material. Irradiating the target with fast neutrons induces radioactivity in the target material through the inelastic scattering of the neutrons, resulting in excitation of the target material. The target de-excites by emitting characteristic gamma rays and neutrons of lower energy. This interaction per unit flux is lower than in TNA, because the cross-section is smaller [20].

3.5.4 Tagged neutron method

The tagged neutron method (TNM), also called associated particle imaging, uses fast neutrons of 14.1 MeV energy to analyse the target material. This technique detects prompt gamma rays produced when neutrons reacts with the target material, hence the name tagging. TNM can produce 10^7 to 10^8 neutrons per second [20]. The principles of TNM will be discussed in Chapter 4 (Tagged Neutron Facility). Table 3.5 shows a comparison of each interrogating method and the elements that can be identified by each method.

Table 3.5: Neutron interrogating method in terms of elements that can be detected by each technique [20].

Interrogating technique	Main reaction	Detected radiation	Primary elements
Thermal neutron analysis (TNA)	(n, γ)	Prompt gamma rays from neutron capture	Fe, S, Cl, Co (and others)
Pulsed fast thermal neutron analysis (PFTNA)	$(n, n' \gamma)$	Gamma rays from inelastic neutron scattering	C, N, O, Cl (and others)
Fast neutron activation analysis (FNAA)	$(n, n' \gamma)$	Gamma rays from Inelastic neutron scattering	C, N, O (and others)
Tagged neutron method (TNM)	$(n, n' \gamma)$	Gamma rays in coincidence with alpha particles	C, N, O (others)

3.6 Radiation protection

Radiation protection is aimed at reducing unnecessary exposure to ionising radiation in order to minimise detrimental effects of ionising radiation [49]. Protection of people and the environment is the most important aspect to consider when planning to use ionizing radiation. Human beings can be exposed to ionizing radiation either by external or internal exposure. Even though internal exposure can occur, its chances are minimal as compared to external exposure. Both internal and external exposures have similar radiation protection measures. Limiting exposure time, increasing the distance from the source, and shielding the source are the most fundamental principles to consider in order to achieve the As Low As Reasonably Achievable (ALARA) principle [50]. With the tagged neutron method, radiation protection is mainly focused on plant site location and tailings (see Sections 5.3 and 5.4).

Chapter 4

Tagged Neutron Facility

One of the objectives of this work is to understand and gain working knowledge of a tagged neutron prototype with a potential application in Botswana mining sector. This technique was developed by Diamant LLC in Russia for the detection of diamonds in kimberlite using 14.1 MeV neutrons [10].

This chapter firstly describes the tagged neutron facility, that is, the setup arrangement of equipment in the laboratory (Diamant-1000 prototype) and the components of the prototype. Lastly, this chapter looks at the data acquisition system as well as the user interface.

4.1 Diamant-1000 facility

The tagged neutron prototype is housed in a 6 m long transport container, consisting of a loader, conveyor belt, portable neutron generator (ING-27 neutron generator) with an inbuilt position-sensitive alpha detector, gamma detectors, data acquisition system, neutron module, and a selector.

The feeding hopper loads kimberlite ore in trays measuring 143 mm $\times$ 260 mm $\times$ 100 mm. These trays are transported to the inspection zone by conveyor belt where they are irradiated with a beam of 14.1 MeV neutrons. The sorter or selector automatically separates diamond-bearing ore from the barren ore. When a tray containing a diamond-bearing rock has been identified, the sorter automati-

cally closes the tailing path and opens the diamond path directing the tray to the concentrate. Figure 4.1, shows the arrangement of Diamant-1000 equipment in a shipping container.

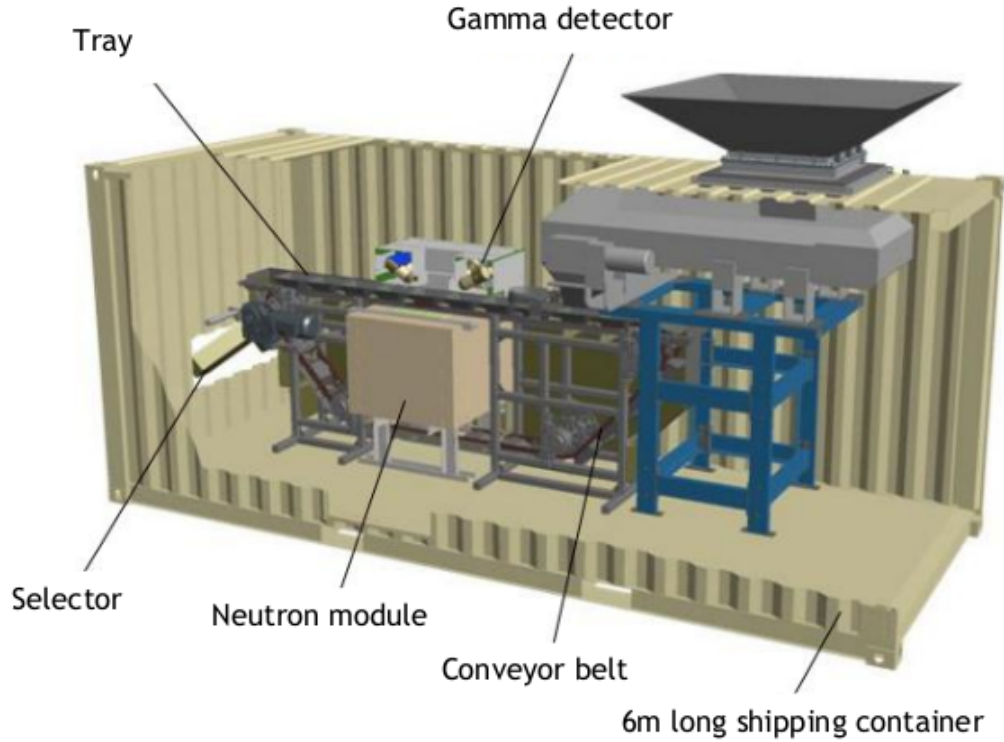


Figure 4.1: Diamant-1000 prototype equipment arrangement in a shipping container. Neutrons from the neutron module irradiate kimberlite ore in trays. The ore becomes excited and emits gamma rays which are detected by BGO gamma detectors placed above the trays above the neutron module.

The shielding material (comprising of iron and polyethylene) directs neutrons to the target (material being analyzed) and also prevents unnecessary neutron interaction with matter which might lead to an increase in background radiation: see Figure 4.2 for the schematic setup arrangement of this equipment. The iron shield is placed next to the generator to protect the BGO detectors from direct contact with neutrons. As neutrons interact with iron, they lose some of their energy (thermalise), and the thermalised neutrons are finally absorbed by the polyethylene shield.

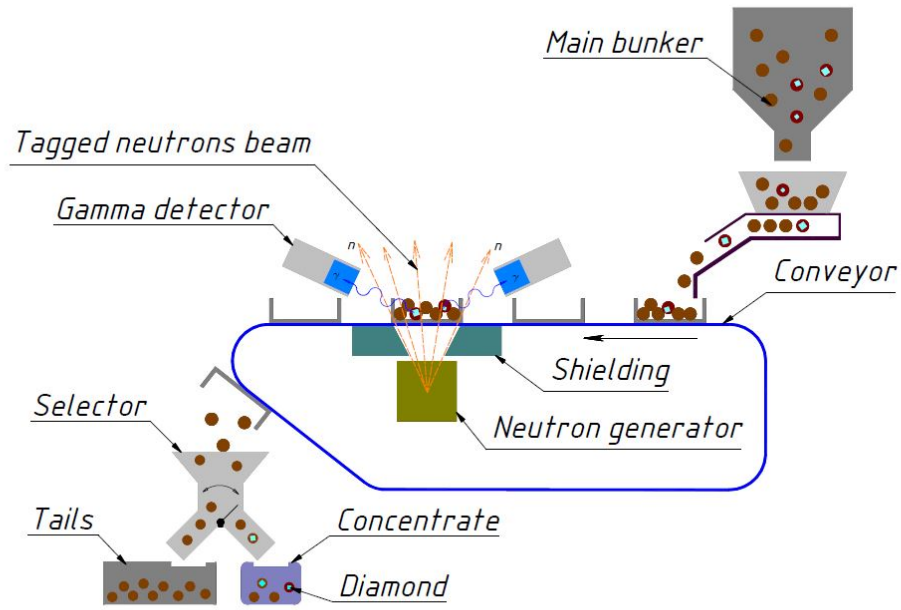


Figure 4.2: Schematic diagram of the Diamant-1000 prototype: the ore in the tray is irradiated with 14.1 MeV neutrons from the neutron generator, resulting in the excitation of kimberlite elements, and subsequent de-excitation through the emission of gamma rays which are detected by gamma detectors. A selector then separates diamond-bearing ore from the barren ore.

Kimberlite rock containing diamond imitator together with barren kimberlite are placed in a tray 40 cm away from the neutron generator and 30 cm away from the gamma detectors. Diamond imitators are made from synthetic diamond powders. The ore in the tray is irradiated with a beam of 14.1 MeV neutrons for 10 seconds. The geometry of the Diamant-1000 is given in Table 4.1.

Table 4.1: Geometry of the Diamant-1000 prototype.

Parameter	Geometry
L, distance to the tray (mm)	134
l, distance to the -detector (mm)	60
d, size of α -detector pixel (mm)	4
Matrix of α -detector	16×16
Dimension of α detector (mm)	64×64
Dimension of the inspected area (mm)	143×143
Dimension of voxel, D (mm)	8.93
Mass of ore in tray (kg)	2.5
Inspection time (s)	10
Throughput, t/h	1.5

4.2 Neutron generator

The ING-27 neutron generator is a sealed tube portable neutron generator with an inbuilt alpha detector [10]. The generator is sealed to protect humans from radioactivity of the tritium target. The generator produces 256-tagged beams of neutrons with a maximum energy of 14.1 MeV which corresponds to a speed of 5cm/ns and alpha particles of 3.52 MeV through the fusion-fission reaction ${}^3\text{H}({}^2\text{H}, n){}^4\text{He}$ [51]. Alpha particles and neutrons are emitted in opposite directions (the inclination angle is about 172° to 180°) [51].

This neutron generator is produced by the Dulkhov All Russian Research Institute of Automatics in Moscow and is used for remote detection and identification of explosives and other hazardous materials [52]. This generator provides a continuous neutron flux with an intensity of up to $5 \times 10^7 \text{s}^{-1}$, operates in DC mode and accelerates deuterium to 100 keV towards the tritium target with a power consumption of 40 W.

The tritium target is bombarded with deuterons resulting in the production of 14.1 MeV neutrons together with alpha particles of 3.54 MeV. These particles move in opposite directions as shown in Figure 4.3 below.

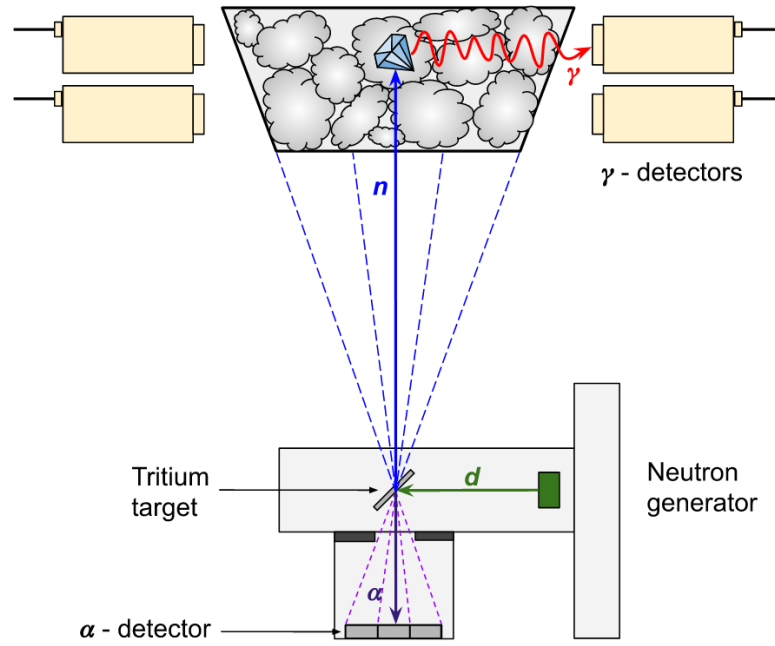


Figure 4.3: Schematic diagram of the tagged neutron method.

The size of the generator is $292 \times 199 \times 279$ mm. Table 4.2 summaries the characteristics of ING-27 neutron generator.

Table 4.2: Characteristics of ING-27 neutron generator.

Characteristics	Values
Maximum deuteron energy	80 keV
Minimum neutron energy (180 deg)	13.326 MeV
Neutron energy (83.9 deg)	14.17 MeV
Maximum neutron intensity	Up to 10^8 n/s
Number of pixels	64×64
Pixel area	4×4 mm ²
Central pixel to target spot distance	62 mm
Mode of operation	Continuous
Dc power supply	200 ± 5 V
Maximum power consumption	70 W
Operating lifetime @ 5×10^7 n/s	≈ 1500 h
Operating temperature	10 - 45 °C
Neutron emission unit (NEU)	$130 \times 279 \times 227$ mm
NEU exit steel windows thickness	0.8 mm
NEU weight	8 kg
Power supply and control unit dimensions	$351 \times 277 \times 99$ mm

4.3 Alpha detector

The alpha detector is an inbuilt gallium arsenide (GaAs) semiconductor alpha-based detector. This alpha detector is a double-stripped sided detector that has 16 perpendicular strips and 16 horizontal strips forming 16×16 matrix of 4×4 mm² elements with a total sensitive area of 32×32 mm². This position sensitive alpha detector was developed specifically for diamond detection. GaAs is relatively insensitive to overheating and has a better time resolution and detection efficiency as compared to the silicon detector [53]. This detector is located 62 mm away from the tritium target in the ING-27 generator [52]. Table 4.3 shows the comparison of alpha detectors found in different types of neutron generators.

Table 4.3: A comparison of different types of neutron generators in terms of alpha detectors, number of tagged beams and pixel size.

Type of neutron generator	Alpha detector material	Number of tagged beams	Pixel size (mm)	Spacing between pixels
ING-27	Silicon	9	10	1
Sodern, Euri-track	silicon	64	5.8	0.2
API-120		256	3	-
ING-27 (used in this work)	Gallium arsenide	256	4	0.1

The spatial size of the tagged neutron beam determines the size of the diamond that can be detected. For measuring the spatial size of the beam, a profilometer is used. The approximation of the spatial distribution of the beam produced from the 16 vertical and a single horizontal strip is shown in Figure 4.4 below. The approximation was done using the Gaussian function.

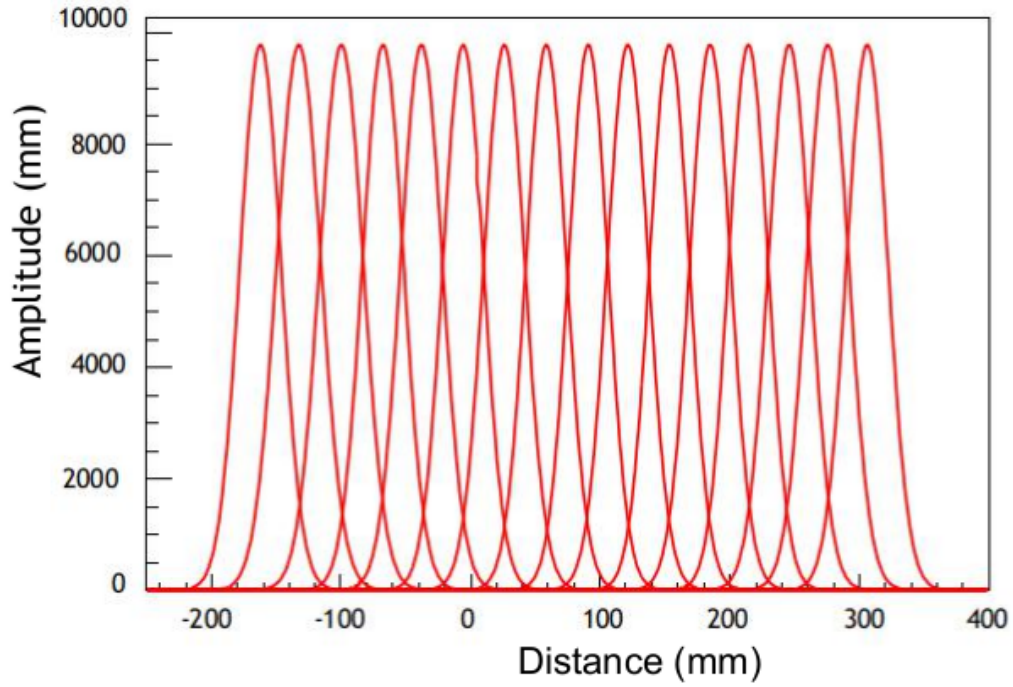


Figure 4.4: Approximation of the spatial distribution of the 16 tagged beams by Gaussian function. A graph of amplitude versus the distance of 16 tagged beams produced by the coincidences of the pulses from 16 vertical strips with a single horizontal strip of α -detector.

The beams had an average width of $\Gamma_x = (37.1 \pm 0.9 \text{ mm})$ and $\Gamma_y = (36.6 \pm 1.1 \text{ mm})$ at a distance of 570 mm from the neutron generator which is consistent with the value of 38 mm expected from a point like a deuteron beam hitting the tritium target from a distance of 570 mm. The beam is quite small with a thickness of about 1 - 2 mm.

4.4 Bismuth germanium oxide gamma detector

The characteristic gamma rays that are produced by the irradiated kimberlite rock are detected using 22 bismuth germanium oxide (BGO) gamma detectors with a thickness of 65 mm and a diameter of 76 mm, with an average energy resolution of $(4.5 \pm 0.1)\%$. These detectors have an energy resolution of 8 to 2.5% within the energy range of 1 to 12 MeV and have high efficiency for gamma-ray energy detection within the specified energy range and low sensitivity for detection of background neutrons [9]. Environmental conditions for this work require detectors that are less or non-hygroscopic. BGO detectors possess this property making them the best

choice (see Table 3.4 for more properties of BGO detectors). These detectors are shielded to prevent them from coming into direct contact with neutrons from the generator which may result in interference with the results due to an increase in background radiation. For better detection results, these 22 detectors are arranged in an array, placed in a rectangular array (with the longest sides having 7 detectors while the shortest have 4 detectors) facing each other as shown in Figure 4.5.

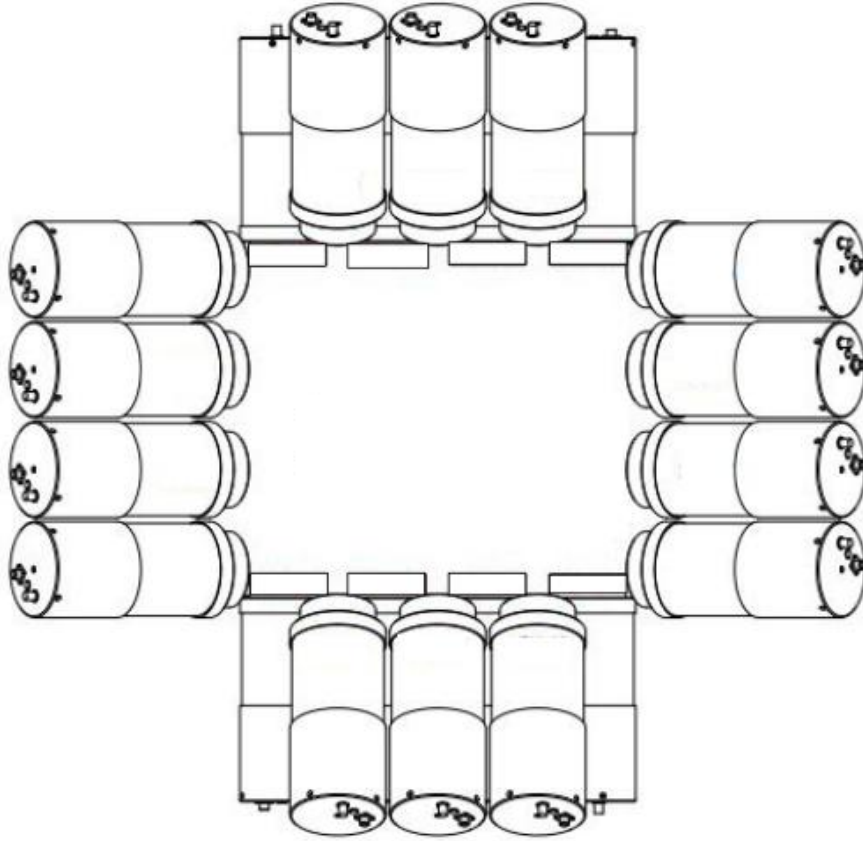


Figure 4.5: Arrangement of BGO detectors at the neutron module.

To measure the time and energy resolution of the detectors, a carbon plate was placed in a tray and irradiated. The energy resolution (Γ_E) for the gamma detectors ranged from $(4.2 \pm 0.1)\%$ to $(5.1 \pm 0.1)\%$ with an average Γ_E of $(4.5 \pm 0.1)\%$ while time resolution (Γ_t) ranged from (4.6 ± 0.1) ns to (5.5 ± 0.1) ns with an average Γ_t of (4.6 ± 0.1) ns as shown in Table 4.4. With an average time resolution of 4.6 ns, the should be placed closer to the neutron source.

Table 4.4: Energy and time resolution for 22 gamma detectors.

Gamma Detector No.	Energy Resolution (%)	Time Resolution (ns)
γ_0	4.7 ± 0.1	4.2 ± 0.1
γ_1	4.5 ± 0.1	4.2 ± 0.1
γ_2	4.6 ± 0.1	4.3 ± 0.1
γ_3	4.6 ± 0.1	5.1 ± 0.1
γ_4	4.5 ± 0.1	4.7 ± 0.1
γ_5	4.5 ± 0.1	4.7 ± 0.1
γ_6	4.4 ± 0.1	4.9 ± 0.1
γ_7	4.3 ± 0.1	4.7 ± 0.1
γ_8	5.0 ± 0.1	5.0 ± 0.1
γ_9	4.5 ± 0.1	4.7 ± 0.1
γ_{10}	5.0 ± 0.1	5.0 ± 0.1
γ_{11}	4.5 ± 0.1	4.5 ± 0.1
γ_{12}	4.5 ± 0.1	5.0 ± 0.1
γ_{13}	4.2 ± 0.1	5.0 ± 0.1
γ_{14}	4.8 ± 0.1	5.5 ± 0.1
γ_{15}	4.4 ± 0.1	5.3 ± 0.1
γ_{16}	4.7 ± 0.1	5.3 ± 0.1
γ_{17}	4.5 ± 0.1	5.5 ± 0.1
γ_{18}	4.4 ± 0.1	5.1 ± 0.1
γ_{19}	5.1 ± 0.1	5.4 ± 0.1
γ_{20}	4.8 ± 0.1	5.3 ± 0.1
γ_{21}	4.5 ± 0.1	4.6 ± 0.1
Average resolution	4.5 ± 0.1	4.6 ± 0.1

4.4.1 Time distribution

The amplitude of events recorded by gamma detectors during irradiation of the carbon plate is shown in Figure 4.6. In this figure, the blue line shows the Gaussian approximation while the red line shows the background around carbon line of 4.4 MeV. Since a large number of detectors (22 in total) were used, only the amplitude for one detector is shown here (detector number 1). In this figure, the detector had an energy resolution of $4.7 \pm 0.1\%$. The peak's position and width were determined using a Gaussian function while the background below the peak was determined using a linear equation. Vertical lines indicate the interval corresponding to the 4.4 MeV carbon line. When the range is large, the signal to noise ratio becomes high.

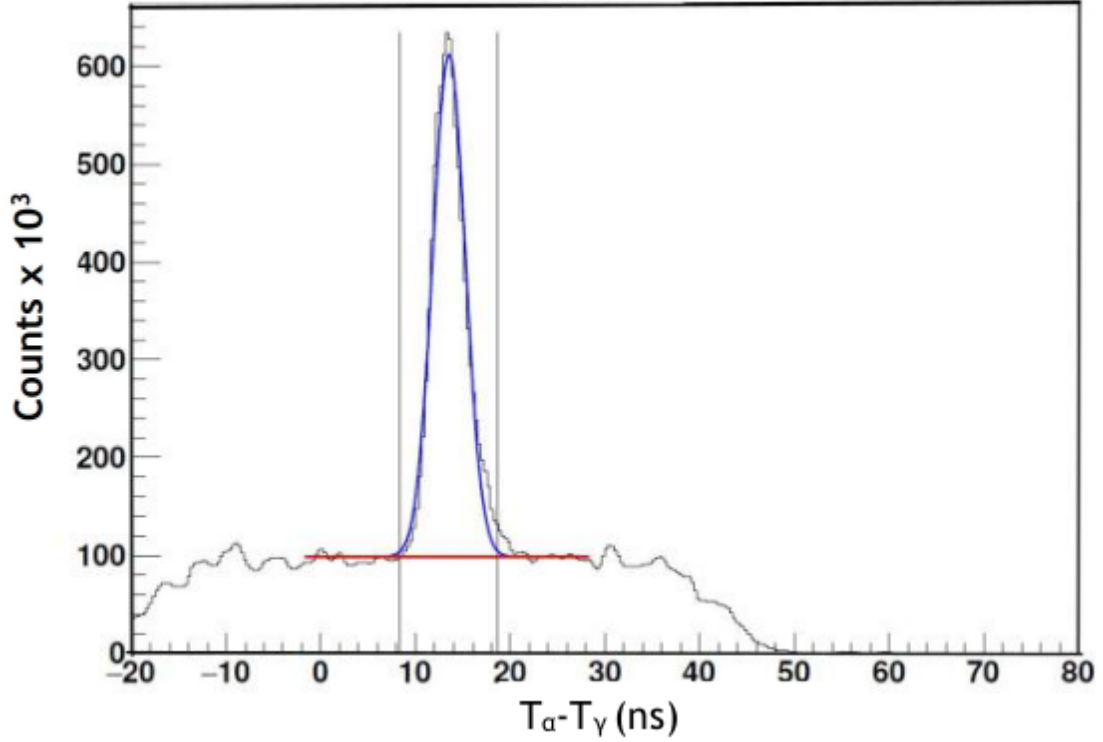


Figure 4.6: Time distribution signals from gamma detector number 1 for gamma quanta with energy around 4.44 MeV. $T_\alpha - T_\gamma$ is the time coincidence for the alpha and gamma signals in nanoseconds.

The time interval between detection of characteristic gamma rays from the carbon plate was measured in coincidence with alpha signals from the alpha detector. The time resolution for the coincidence detection was 4.6 ± 0.1 ns therefore the target should be closer to the neutron source.

The time of detection of gamma rays is used to locate the neutron-nucleus interaction point which is used to locate the target, while measurement of gamma-ray energy allows the elemental identification of the target. The energy spectrum of gamma quanta from irradiation of carbon forms a known peak at 3.93 MeV and 4.44 MeV (first escape and full-energy peak, respectively) which are used as a signature for the presence of diamond in kimberlite.

4.5 Data acquisition system

The data acquisition system for alpha and gamma detectors is designed as a single board of 32 inputs, which has the size of a standard peripheral component inter-

connect (PCI) card and can be inserted in the PCI- E slot of a personal computer (PC). The hardware data acquisition board utilizes a high-speed analog to digital converter (ADC) for sampling inputs signals. The system for registration of a signal from the alpha and gamma detectors is based on the principle of waveform digitalising with subsequent calculation of their time and amplitude characteristics.

Channels of digitalising signals from the detectors are built on the same scheme which includes an amplifier of 0-20 MHz band pass filter and analog to digital converter measurements interval of 10 ns (100 MHz). The digital code corresponding to each of the measurements is fed to an analog to digital converter logic circuit in which there is a definition of moments of α - γ coincidence in digital form.

The programmable scheme logic of the electronics board of data acquisition system organizes the reception of information from analog to digital control, allocates the coincidence moments definition, prepares a data buffer and provides its transfer to the PC memory, eventually the exact value of the pulse amplitudes and the time interval between them are calculated [9].

Special software for selection and signal processing extracts information about α - γ coincidence, determines triggered channel numbers, calculates a time interval between pulses and the area under the pulse envelope, defining thereby an amount proportional to the energy released in the detectors, and counts the pulses detected by each detector individually. These parameters are presented in real-time histograms and stored in data files [9].

4.6 The user interface

During the irradiation process, the interface shows 256 voxels with different colour codes. Each colour code represents a different level of localised excess carbon concentration in each of the 256 voxels. The grey colour represents empty regions of the tray: this is the region in which the statistics data was too low to be counted. The green colour represents regions in which excess localised carbon level was less than 3σ . This is also the region occupied by the ore sample. The red colour represents the region where the excess localised carbon level was larger than 3σ : this is the region occupied by the diamond imitator. The excess carbon levels can be as high as 7σ as shown in Figure 4.7 by the red colour.

0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
-1.16	-0.85	0.96	0.79	-0.58	-1.94	0.28	-0.65	0.68	-0.03	1.02	-0.19	-0.62	0.00	0.00	0.00
0.53	-0.24	-0.31	-1.73	0.60	0.00	0.65	-0.61	-1.03	0.28	0.30	0.35	0.81	1.57	0.00	0.00
-0.79	1.32	-0.04	2.22	0.81	0.19	-0.31	0.01	-1.54	-0.44	-1.18	-1.33	-0.83	-0.24	0.00	0.00
0.86	0.09	-1.17	-0.60	-1.36	-1.19	-0.02	-0.04	0.38	-0.43	-0.02	-0.77	-0.85	-0.22	0.00	0.00
0.03	-0.13	2.25	5.48	1.44	-0.72	0.31	0.73	-0.25	-1.11	-1.05	-1.41	-1.40	-0.84	0.00	0.00
-0.96	0.07	4.29	7.72	5.45	1.27	-1.18	0.02	0.08	-0.42	0.30	-1.09	-1.16	-1.40	0.00	0.00
0.00	0.00	0.00	3.71	0.00	0.22	-0.74	-0.13	-0.45	-0.95	0.04	-2.13	-0.36	-0.84	0.00	0.00
0.68	0.44	0.78	-2.36	-0.31	-0.27	0.07	-0.97	-0.30	-0.31	0.21	0.43	-1.09	0.03	0.00	0.00
-0.78	-0.56	-2.49	-0.84	-0.15	-0.86	-1.39	0.41	-0.49	-0.40	0.28	0.19	1.06	0.33	0.00	0.00
0.25	-0.64	1.07	0.09	-0.24	-0.09	-1.70	0.43	-0.40	0.53	0.38	-0.31	0.12	0.53	0.00	0.00
-1.29	-1.15	-0.72	0.67	-0.32	0.50	-2.45	1.03	-0.10	0.45	-0.89	-1.18	1.49	-1.68	0.00	0.00
0.46	-0.32	0.71	0.34	-0.14	-1.09	-1.90	0.60	1.53	-0.54	0.03	-1.16	1.68	0.48	0.00	0.00
-1.10	0.49	0.95	0.87	-0.04	0.45	-1.59	1.48	1.14	-0.06	-1.67	-2.18	1.06	-0.65	0.00	0.00
0.00	0.00	0.00	0.00	0.00	-0.26	2.81	1.07	0.40	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Figure 4.7: The user interface showing 90 mm kimberlite containing a 20 mm imitator. The grey or white colour shows an empty space of the tray, the green colour shows the region occupied by the kimberlite ore, while the red colour shows the region occupied by a diamond imitator.

Depending on the size of the imitator, diamond signals can be detected in more than one region as shown in Figure 4.7 or in one voxel as shown in Figure 4.8. For large imitators its signal can be detected in several voxels and may appear in different configurations as shown in Figure 4.7 with excess level as high as 7.7σ while for a small imitator the excess level can be 3.3σ . Detecting diamonds using the tagged neutron method makes it also possible to predict the size and position of diamonds as shown in Figures 4.7 and 4.8 respectively. Large diamonds are detected in more than one voxel as shown in Figure 4.7 while small diamonds (12 mm diamond imitator) are detected in one voxel as shown in Figure 4.8.



Figure 4.8: User interface showing a 30 mm Kimberlite with a 12 mm imitator. The green colour shows the region occupied by the kimberlite ore, the grey colour shows an empty space of the tray while the red colour shows the region occupied by the diamond imitator.

Results and Discussion

This section presents results for the identification of diamonds in kimberlite using the tagged neutron method described in the previous chapter. In this work, kimberlite from Archangelsk mine in Russia and the Letseng diamond mine in Lesotho was used. Kimberlite ore is placed in a tray and irradiated. The ore was arranged in a single layer in trays. A hole was drilled in the kimberlite ore and a diamond imitator was inserted in the hole. Diamond imitators were made from artificial diamond dust. The presence of a carbon signal in the kimberlite spectrum is used as an indication of the presence of diamond in kimberlite ore. Results include the minimal and maximal size of the diamond that can be detected using the tagged neutron method and the probability of false alarm signals. Radiation damage to the diamonds and radioactivity of tailings are also presented.

5.1 Diamond detection

Kimberlite ore containing a diamond imitator was placed in a tray and irradiated with 14.1 MeV neutrons (see Figure 5.2 for the ore arrangement in a tray): diamond imitators were made from artificial diamond dust. Diamond is mainly carbon, therefore detection of diamond is done by the detecting excess of carbon concentration in some regions of the ore. The gamma spectrum of carbon displays peaks at 3.9 and 4.4 MeV (first and second escape peaks, respectively) as shown in Figure 5.1. None diamond carbon is homogeneously distributed in the ore, therefore localised carbon excess can be easily detected. The appearance of a carbon spectrum

within the kimberlite spectrum is used as an indication of the presence of diamond in kimberlite stones. The spectrum in Figure 5.1, shows carbon peaks for carbon plate at 3.9 and 4.4 MeV, where the 4.4 MeV peak is dominant.

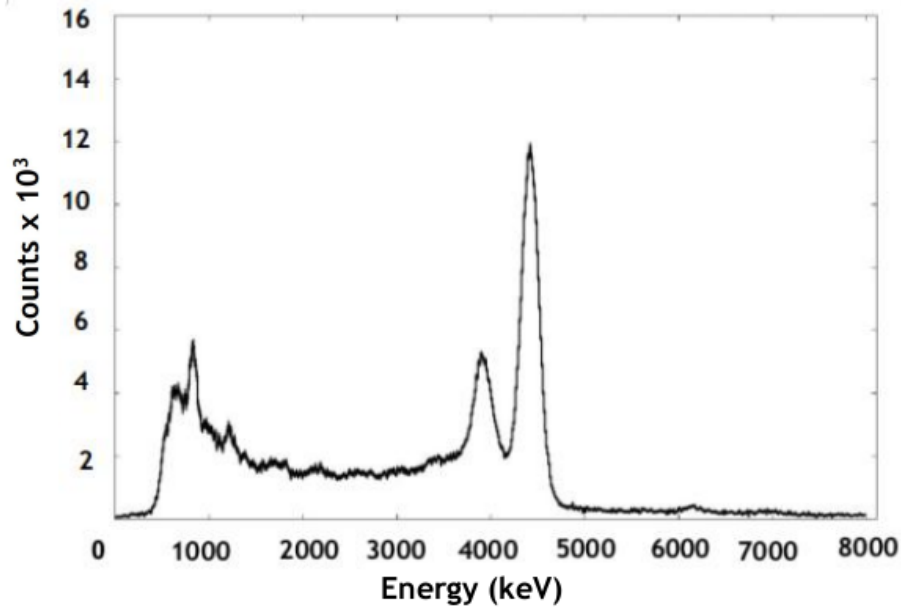


Figure 5.1: Gamma spectrum for a carbon plate, the spectrum is dominated by the single-escape peak with energy 3.9 MeV and the full-energy peak at 4.4 MeV.

5.2 Ore irradiation

To check the effectiveness of the tagged neutron method for detecting diamonds, kimberlite ore from different mines was irradiated with 14.1 MeV neutrons (100 samples from the Archangelsk mine in Russia and 70 samples from the Letseng diamond mine in Lesotho). These samples ranged from 6 mm to 150 mm in diameter. Although these samples were from kimberlites, their mineral content is not the same. Letseng kimberlite ore contains less carbon content compared to ore from the Archangelsk mine: their spectra are shown in Figures 5.3 and 5.4.

The ore was arranged in a single layer in a tray as shown in Figure 5.2 and irradiated with a beam of neutrons flux with an intensity of $3.2 \times 10^7 s^{-1}$. Each tray had a single ore with a drilled hole to insert a diamond imitator. Figure 5.2 shows how the ore was arranged in a tray.



Figure 5.2: Ore arrangement in a tray. A hole is drilled in a kimberlite ore and a diamond imitator inserted in the hole.

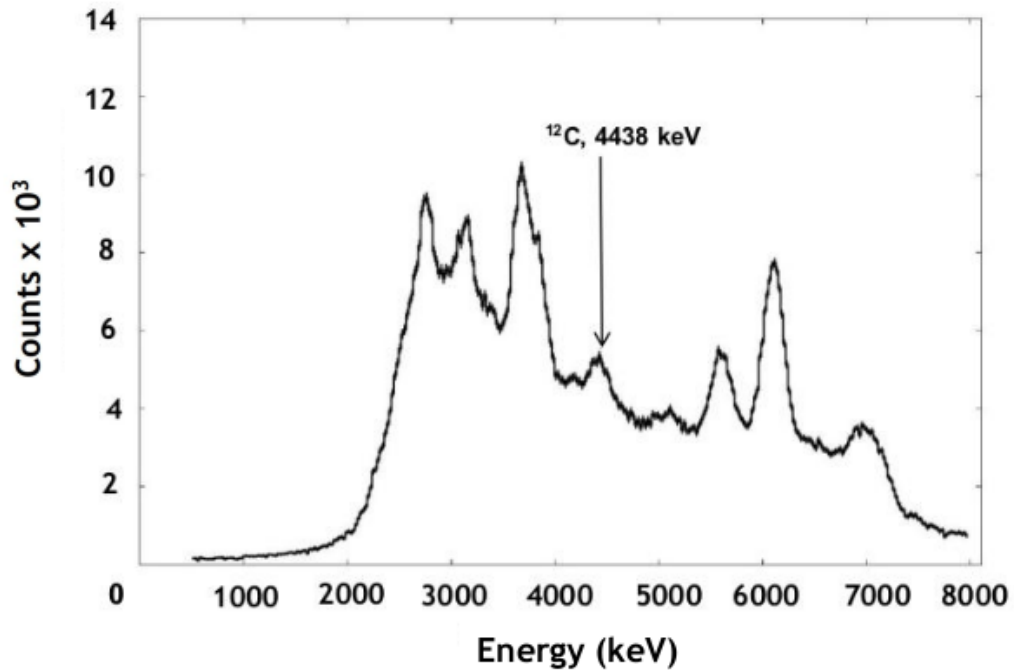
5.2.1 Kimberlite spectrum

Kimberlite consists of many compounds and elements. These elements are activated during irradiation of the kimberlite and emit prompt gamma rays which may result in the increase of the background radiation. Some of these elements like oxygen emit gamma rays with energy closer to the gamma lines of carbon. The compounds listed in Table 5.1 are major and trace compounds found in most kimberlite (see Figures 5.3 and 5.4 for the kimberlite spectra).

Table 5.1: Minimum and maximum percentages of major and trace compounds found in kimberlite ore [54].

Compound/Element	Minimum %	Maximum %
SiO ₂	25.6	36.4
Al ₂ O ₃	2.50	5.10
TiO ₂	1.20	3.40
Fe ₂ O ₃	3.90	6.78
MnO	0.10	0.19
MgO	17.0	28.6
CaO	7.60	21.4
Na ₂ O	0.12	0.48
K ₂ O	0.34	2.10
P ₂ O ₅	0.70	1.10

When the ore is irradiated with fast neutrons, the kimberlite elements become excited and de-excite through the emission of gamma rays. Figure 5.3 shows the kimberlite spectrum for the Russian (Archangelsk mine) kimberlite, while Figure 5.4 shows the spectrum for Lesotho (Letseng) kimberlite. From these two spectra, one can see that the carbon peak in the Archangelsk kimberlite spectrum is very high, but a bit smaller in the Letseng kimberlite spectrum.

**Figure 5.3:** Gamma ray spectrum from irradiated Archangelsk kimberlite ore without a diamond imitator.

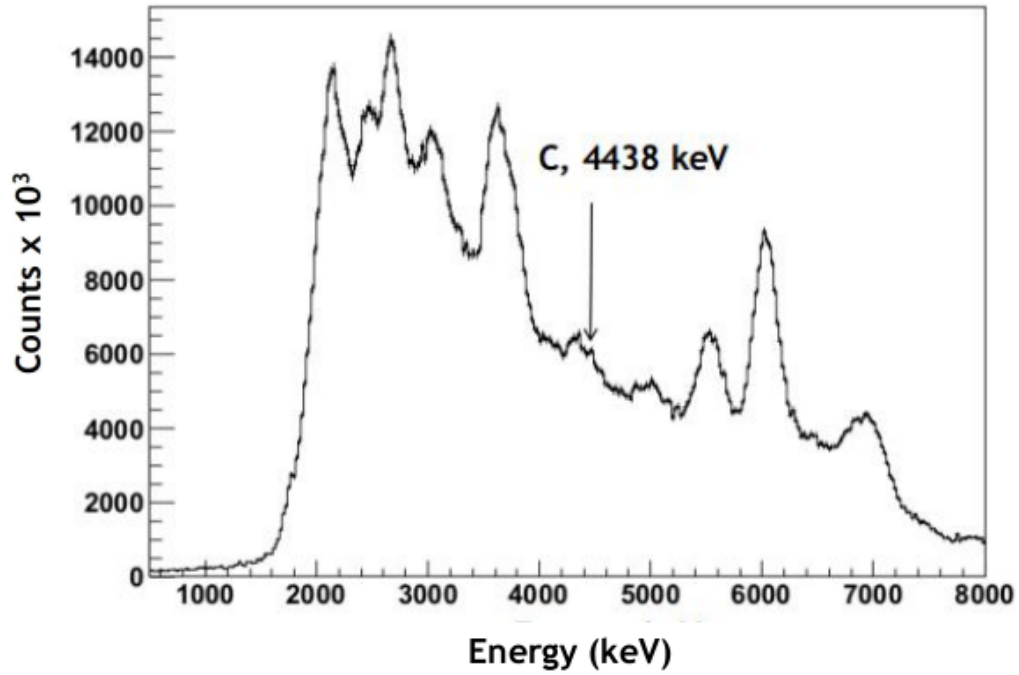


Figure 5.4: Gamma ray spectrum from irradiated Letseng kimberlite ore without a diamond imitator.

Since kimberlite contains many elements, the spectra shown in Figures 5.3 and 5.4 only show gamma lines for elements within the range of 2500 keV to 8000 keV; therefore, low energy spectra for elements such as magnesium are not shown. Both spectra are dominated by oxygen lines (at 3.8, 6.13, 6.92 and 7.12 MeV).

5.2.2 Kimberlite spectrum with a diamond imitator

In this work, diamond imitators were used as a substitute for diamonds. The peak around the carbon region is small in Letseng kimberlite ore compare to the Archangelsk kimberlite ore. Letseng ore has almost no carbon content resulting in minimal interference with the results. The black line in Figure 5.5 shows the averaged spectrum of all voxels in a tray, while the red line shows the spectrum in a voxel where the excess of carbon is observed. In Figure 5.6, the black line shows the averaged spectrum of all voxels in a tray while the blue line shows the spectrum in a voxel where the excess of carbon is observed.

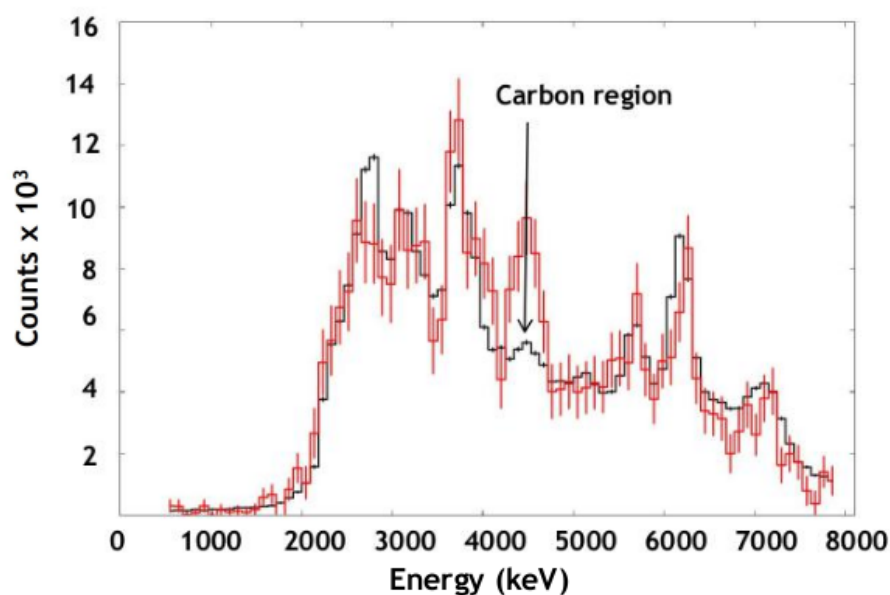


Figure 5.5: Gamma-ray spectrum for irradiated Archangelsk kimberlite ore (90 mm) containing a 20 mm diamond imitator: black lines shows the kimberlite spectrum, while red lines shows the spectrum in a voxel where the excess of carbon is observed.

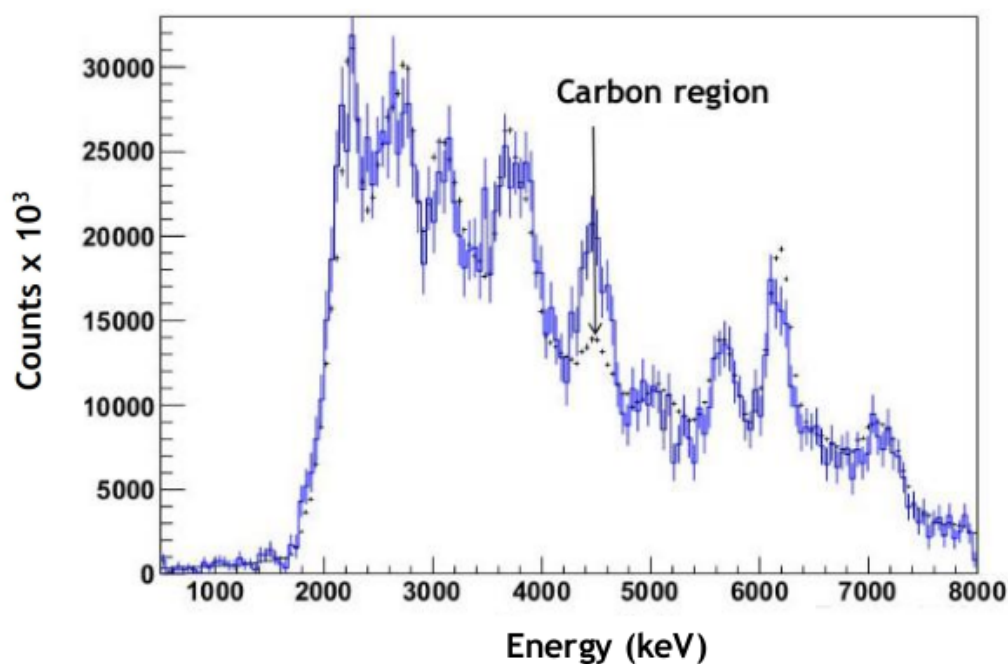


Figure 5.6: Gamma ray spectrum for irradiated Letseng kimberlite ore (90 mm) containing a 20 mm diamond imitator. Black dotted lines shows the kimberlite spectrum while the blue line shows the spectrum in a voxel where the excess of carbon is observed.

5.2.3 False alarm probability

A false alarm is the appearance of a diamond signal in empty kimberlite ore without diamonds. To determine the false alarm rate, empty kimberlite (kimberlite without diamond imitator) was irradiated.

Since diamonds exist in parts per million (ppm) in kimberlite, the separator mainly works with the barren ore such that the probability of finding an open diamond in the kimberlite ore is quite small. The selection criterion should minimise the false alarm as much as possible. This selection criterion is based on an excess of the local concentration of carbon in the spectrum of certain voxel in comparison with the carbon concentration averaged over the whole sample. The value of the excess concentration level of carbon (σ) is calculated using the equation below:

$$\sigma = (C - \overline{C}) / \sqrt{(\varepsilon_{\overline{C}}^2 + \varepsilon_C^2)}, \quad (5.1)$$

where C is the number of events in carbon line region in a certain voxel, $\overline{C}$ is the average number of events in carbon line region in measured sample, and $\varepsilon_{\overline{C}}^2$ and ε_C^2 denote the corresponding statistical errors.

The false alarm probability for the Letseng and Archangelsk mine kimberlites are given in Table 5.2. The results show that Letseng kimberlite has a lower false alarm probability compared to the Archangelsk mine ore. This result is in line with the expected results since Letseng ore has less carbon content as compared to Archangelsk mine ore.

Furthermore, the results in Table 5.2 show that false alarm probability decreases with increasing σ detection parameter. When σ is 3.13, the false alarm probability for Archangelsk ore is around 3%, while it is 0% for Letseng ore. The results in Table 5.2, show that for kimberlite ore with little carbon content such as Letseng ore, the σ level can be as low as 2.88 and at this σ value, the false alarm probability can be around 1%, whereas it can be 14% for Archangelsk ore at the same σ value.

Table 5.2: False alarm probability for irradiating kimberlite ore at different excess (σ) levels.

Criterion of diamond signal in terms of statistical error (σ)	False alarm probability	
	False alarm for Archangelsk ore	False alarm for Letseng ore
2.38	0.59	0.67
2.50	0.47	0.31
2.63	0.36	0.14
2.75	0.21	0.04
2.88	0.14	0.01
3.00	0.07	0.00
3.13	0.03	0.00

The increase in σ level results in the false alarm approaching zero for all types of kimberlite as shown in Figure 5.7. Since the objective was to detect both small and large diamonds, a suitable σ was found to be around 3.13 for Archangelsk mine and around 3 for Letseng ore. At values larger than 3.13, the false alarm is almost zero for all types of kimberlite.

With high σ levels, the chance of missing diamonds less than 8 mm increases, while at low σ level the false alarm probability increases (chances of missing diamonds, both less than 8 mm and larger than 8 mm increases). Therefore, the chance of missing small diamonds and the false alarm must be balanced.

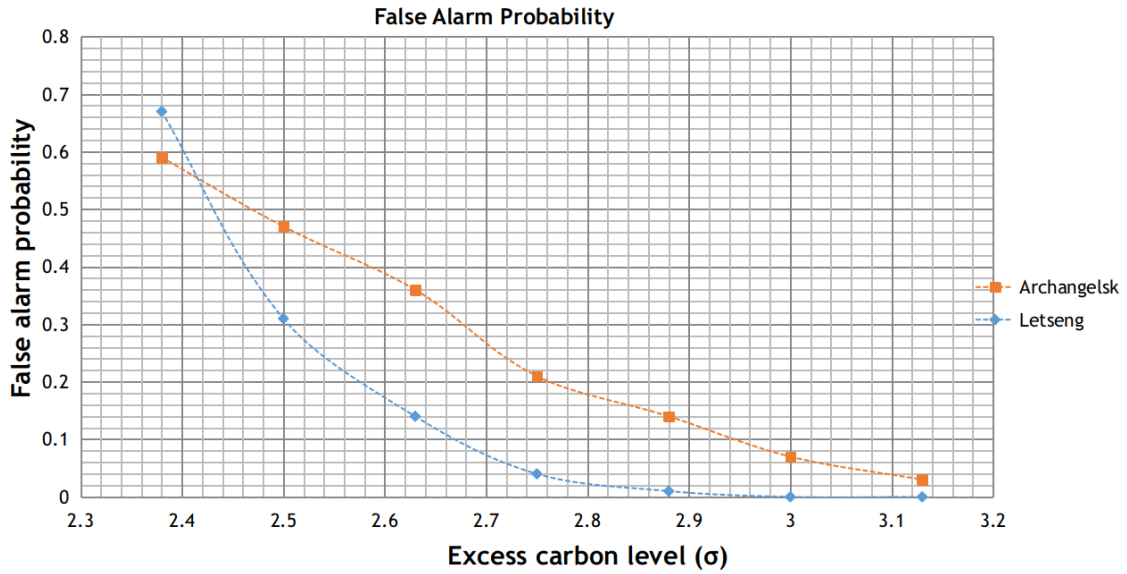


Figure 5.7: False alarm probability versus excess carbon level (σ): the false alarm probability decreases with an increase of σ level in both Letseng and Archangelsk kimberlite ore.

5.2.4 False alarm

When the kimberlite ore is irradiated for more than 10 seconds, the false alarm probability at 3σ becomes higher than 3%. In order to have a minimal false alarm of 3% or less, it means the σ level must be increased to a value greater than 3σ , for example about 3.7σ but this leads to increasing the time of irradiation up to 80 seconds. Increasing the σ value increases the chances of missing small diamonds. Figure 5.8, shows the false alarm when irradiating kimberlite for different exposure times (see Appendix C).

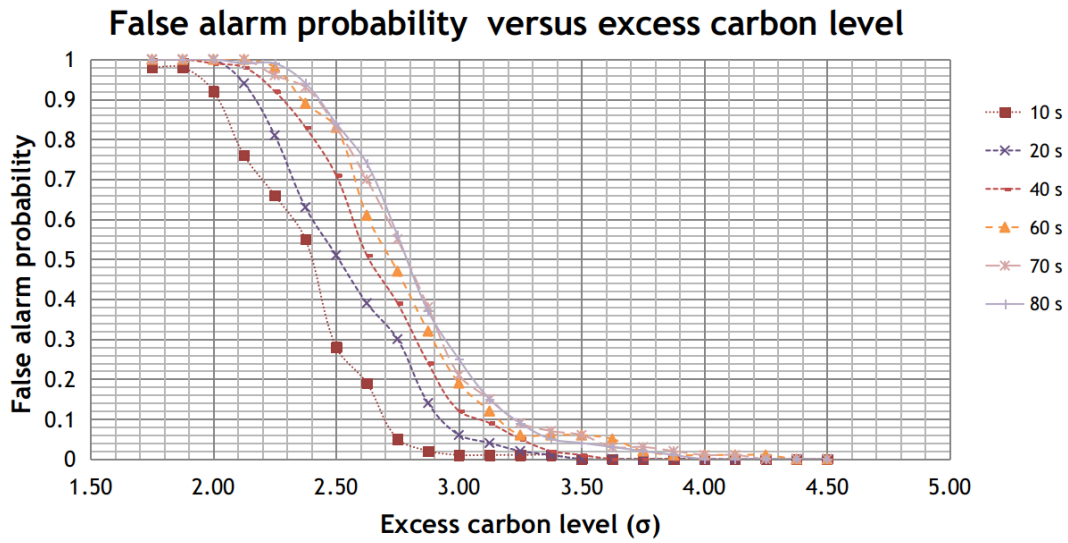


Figure 5.8: False alarm decreases with minimal exposure time, for example when kimberlite ore is irradiated at 80 seconds, false alarm becomes high unlike irradiating the ore for 10 seconds.

5.2.5 Diamond detection probability

To check the efficacy of the TNM technique, tests were carried out in different ore sizes with diamond imitators of different sizes. The size of the ore ranged from 30 to 150 mm, while the size of imitators ranged from 8 mm to 20 mm. Diamond imitators were made of artificial diamond dust with diameters equivalent to their heights.

The diamond detection probability depends on the size of the ore. Ore composition has little influence on diamond detection probability because the tagged neutron method is based on localized carbon and not homogeneously distributed carbon in the ore. Table 5.3 shows the detection probability in different sizes of ore: the tagged neutron is capable of successfully detecting both small (8 mm diamonds) and large diamonds (20 mm diamonds). The minimal diamond imitator detected was 8 mm in kimberlite ore of $-30 + 6$ mm, while the maximum was a 20 mm diamond imitator in kimberlite ore within the range of $-150 + 100$ mm. Results show that the TNM has a probability of 95% to detect 20 mm diamonds in kimberlite ore within the range of $-150 + 100$ mm.

Table 5.3: Probability to detect diamond in kimberlite stones of different sizes: the notation $(-A + B)$ mm for ore size means that the largest dimension of the stone is smaller than A mm and the smallest dimension is larger than B mm.

Size of diamond imitator, (mm)	Size of kimberlite stones, (mm)	Detection probability
8	-30 +6	0.96
12	-75 +40	0.96
16	-75 +40	0.99
16	-90 +40	0.97
20	-150 +100	0.95

5.3 Radiation damage to the diamonds

Effects of irradiating diamonds with fast neutrons of 14.1 MeV were studied at the Veksler and Baldin High Energy Physics Laboratory at the Joint Institute for Nuclear Research in Dubna, Russia [55]. The properties of diamonds, including the state of aggregations, colour, and nature of cleavages, was studied before irradiation. No significant damage to the diamonds was observed after exposing the diamonds to 14.1 MeV neutrons for about 10 seconds (the time needed to irradiate the kimberlite) [55].

5.4 Tailings

Tailings are waste materials (barren kimberlite ore) that remain after the selection of diamond bearing ore for processing. After the emission of prompt gamma rays, the kimberlite rock may remain excited, and subsequently de-excite through emission of the beta particles, followed by gamma rays (delayed gamma rays). It is worth noting that in detecting diamonds using the TNM technique, no radioactivity was detected after the emission of prompt gamma rays.

5.5 Tagged neutron feasibility and applicability

The early detection of diamond-bearing rocks and rejection of non-diamond bearing rocks without any loss of diamond-bearing rocks is very crucial in the diamond mining process, reducing the load capacity and processing time. After rejecting

barren kimberlite rocks, an assiduous (chemical or mechanical) method can be used for further processing of the fertile kimberlite without or with minimal damage to the diamonds.

5.6 Mir mine TNM results

The TNM technique has been tested at the Mir mine in Russia where it proved to be effective in detecting diamonds within the kimberlite ore. For the Mir mine tests, 33 kimberlite ore samples with masses ranging from 2 to 5 kg were irradiated. Out of 33 samples provided, one sample had excess carbon greater than 3σ . With further processing of the sample with excess carbon level greater than 3σ , two diamonds inclusions with diameters up to 7 mm consisting of small diamonds crystals were discovered. Figure 5.9 shows two diamond inclusions detected using the tagged neutron technique at the Mir mine. These results show that the tagged neutron method can in principle be successfully used in the diamond mining industry.



Figure 5.9: Two diamond inclusions detected at the Mir mine using the tagged neutron method.

Summary, Conclusions, Final Remarks and Recommendations

This chapter presents the summary, conclusions, final remarks, and recommendations of this dissertation.

6.1 Summary

This dissertation focused on the detection of diamonds in kimberlite using the tagged neutron method. Chapter 1 described the context rationale, problem statement, and objectives associated with this project. Chapter 2 provided an overview of current technologies and methods for detecting and recovering diamonds from kimberlite ore. Chapter 3 covered the principles underlying the tagged neutron method and the generation of neutrons for identifying diamonds in kimberlite. Chapter 4 described the components of tagged neutron prototype tagged including the data acquisition system and user interface. Chapter 5 presented the results obtained from using the tagged neutron prototype in detection of diamonds within the kimberlite ore.

6.2 Conclusions

The main objective of this project was to test the newly developed prototype (Diamant-1000 prototype) at the Joint Institute for Nuclear Research in Dubna, Russia, for the detection of diamonds in kimberlite using the tagged neutron method and to check the suitability of eventually deploying this prototype in Botswana for identifying diamonds in real-mining operations. The study was carried out using kimberlite ore from the Archangelsk mine in Russia and the Letseng mine in Lesotho. These kimberlites ores contained different carbon content (Archangelsk kimberlite ore had more carbon content while Letseng kimberlite ore had little or no carbon content). These ores were placed in trays and irradiated with 14.1 MeV neutrons. The probability to detect diamonds hidden in kimberlite ore within the range of $-150 + 100$ mm was found to be 95%. In kimberlite ore ranging from $-90 + 40$ mm, the probability of detecting hidden diamond was found to be 97%. The prototype had a throughput of about 1 ton/hour (1060 kg/hr) with an electric consumption of 0.6 kW/ ton of ore. This prototype has 22 gamma detectors and a neutron generator with 256- α channels. The minimal size of the diamond imitator detected was 8 mm (5 carats). The false alarm probability of the prototype was 3%. The throughput, however, of 1060 kg/hr is small when compared to the 30 - 50 tonne/hour which is needed for practical application.

The study demonstrates that the tagged neutron method has the potential to be of tremendous benefit to mining industries in terms of profits, minimum waste and minimal impact on the environment. This method also has the potential to detect large diamonds (diamonds larger than 20 mm) ore of 150 mm size before the secondary crushing stage.

This method can detect both Type A and Type B diamonds of all sizes, therefore allowing this technique to serve as a single recovery method. The tagged neutron method also produces less waste products compared to other recovery techniques such as the X-ray transmission technique. Deployment of the tagged neutron method by diamond mining companies will reduce the financial burden that comes with processing barren ore: only diamond bearing ore is selected for further processing. The downstream yield will be reduced with only about 3% of the final products being the barren kimberlite ore.

A project for a conveyor belt separator is currently under preparation for handling a throughput of 5 tonnes/hour which is 5 times higher than the current setup. Although the throughput is still low, this will be the starting point for a commercial tagged neutron separator that could be used in real diamond mining setup.

6.3 Final Remarks and Recommendations

The visit of the author of this dissertation to the Joint Institute for Nuclear Research (JINR) in Dubna, Russia, to work with the Diamant group on using the recently-developed tagged neutron prototype (Diamant-1000) for detecting diamonds in kimberlite, provided invaluable knowledge and experience for a young Motswana student to be part of an international collaboration with the potential to transfer the technology to Botswana for application in the diamond mining industry. Indeed, through this project, the Botswana International University for Science and Technology has forged closer ties with diamond industries in Botswana as well as the JINR, with opportunities now becoming available for more students and staff to collaborate with experts on developing techniques for the extraction of diamonds. Further opportunities exist for deploying the tagged neutron method extraction of other minerals, such as phosphates and coal, in Botswana.

The next step will be to procure the Diamant-1000 prototype to Botswana for education, training, skills development, exploitation, and further development of this innovative technology for the extraction of large diamonds and potential diversification to other local mining sectors. In particular, the local diamond mining companies could benefit in the long term from further developments in the following areas:

1. Grade evaluation and sampling without comminution, concentration, and recovery;
2. Sorting and rejection of non-diamond bearing rocks from the processing stream to significantly reduce the size, footprint, sustainability requirements, and capital, which are the ambitions of any major extraction company for mines and plants of the future;

3. Small, marginal operations as a sorter to reduce capital and processing costs as per the previous item above.

If everything goes according plan,the Diamant-1000 prototype will be procured and transferred to Botswana by the end of 2022.

Appendix A

Selected Nuclides Data

Data for selected nuclides are shown in Table A.1 Nuclide data give useful information about the nuclide. Table A.1, also helps in predicting the amount of energy released or absorbed during reactions like the $d - t$ reaction in ING-27 [24].

Table A.1: Selected Data on Nuclide

Nuclide	Natural abundance(%)	Mass difference $\Delta = M - A$ (MeV) (at.mass-at.massNo.)	Type of decay	half-life	Major Radiations, Energies (MeV) and Frequency per Disintegration (%)
^1_0n	-	8.0714	β^-	12 min	β^- : 0.78 max
^1_1H	99.985	7.2890	-	-	-
^2_1H	0.015	13.1359	-	-	-
^3_1H	-	14.9500	β^-	12.3 years	β^- : 0.0186 max, no γ
^3_2He	0.00013	14.9313	-	-	-
^4_2He	99.99+	2.4248	-	-	-
$^{11}_6\text{C}$	-	10.648	β^+ 99+%,EC 0.2%	20.38 min	β^+ : 0.960 max(avg 0.386), γ :0.511(200%, $\gamma^\pm$)
$^{12}_6\text{C}$	98.892	0	-	-	-
$^{14}_6\text{C}$	-	3.0198	β^-	5730 years	0.156 max (avg 0.0495), no γ

(adapted from Atoms,Radiation and Radiation Protection [24])

Diamond Recovery Processes

The main steps in diamond recovery for both XRT, Mineral-PET and Tagged Neutron Method are shown in Figure B.1, B.2 and B.3.

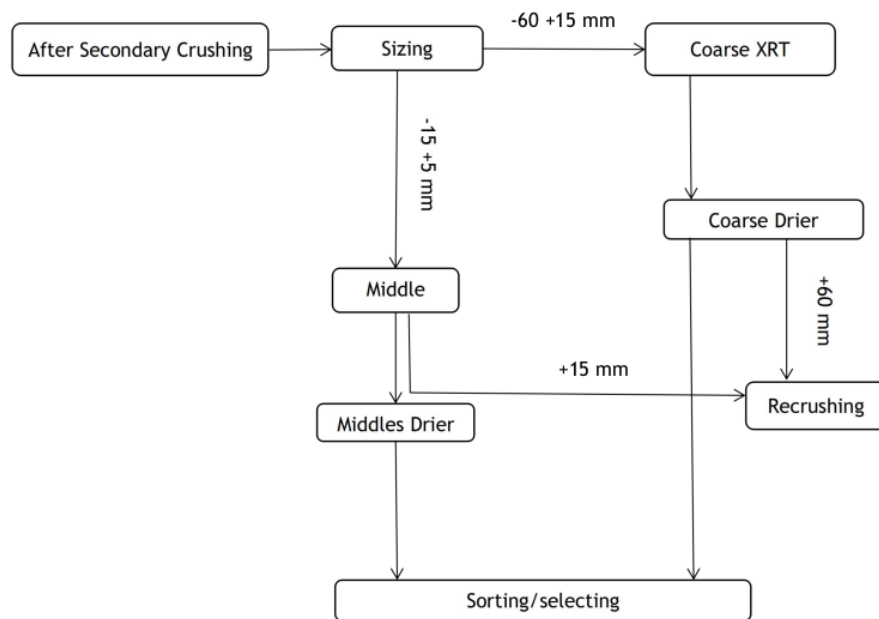


Figure B.1: A simplified for XRT recovery process; adapted from application of diamond size frequency [11, 12].

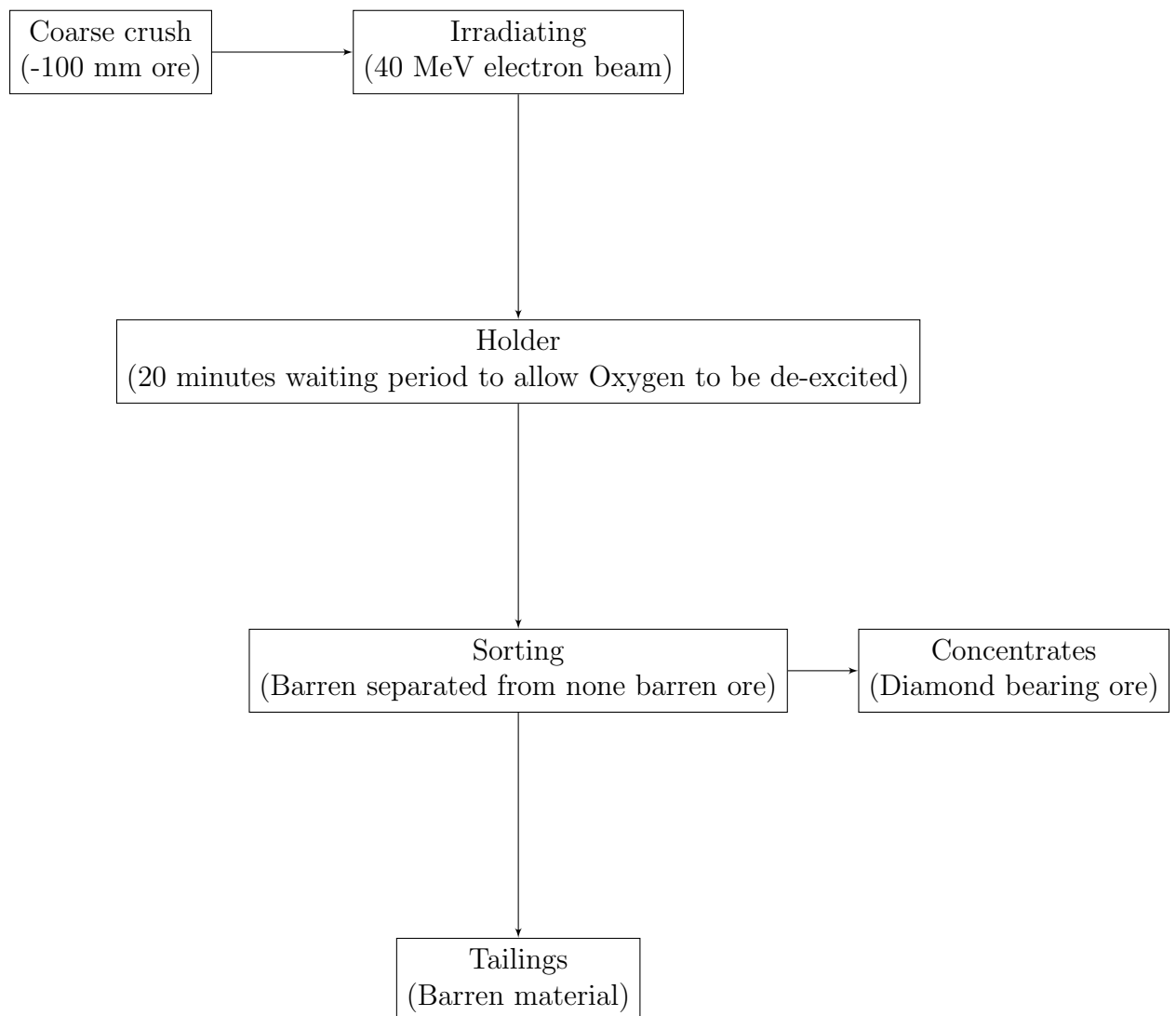


Figure B.2: Mineral-PET detection process; adapted from The Mineral-PET diamond discovery technique [56].

N.B: -100 mm means the maximum size of the ore is less than 100 mm.

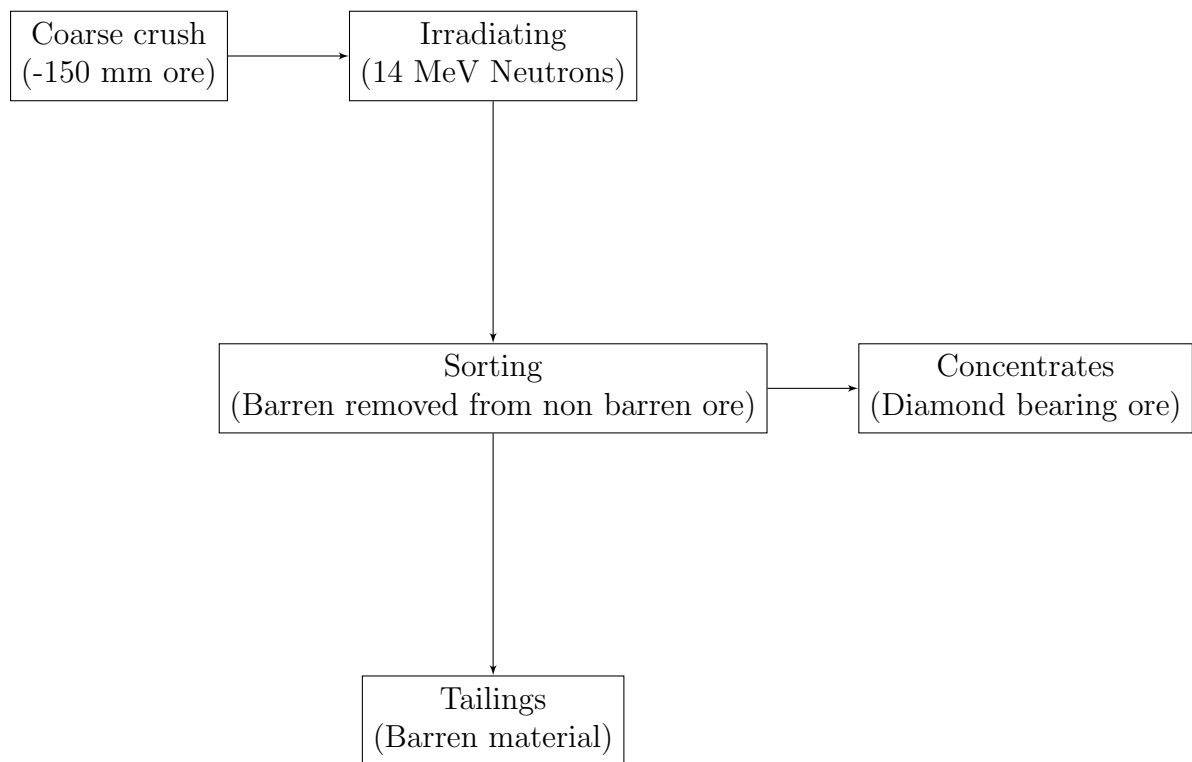


Figure B.3: Tagged Neutron Method recovery process.

N.B: -150 mm means the maximum size of the ore is less than 150 mm.

Appendix C

Letseng and Archangelsk Mine Kimberlite Results

Results of irradiating ore at different intervals and at different excess carbon level (σ) are presented in this appendix. Results for Archangelsk kimberlite ore are in Tables [C.1](#) and [C.2](#) while for Letseng kimberlite ore are in Tables [C.3](#) and [C.4](#).

Table C.1: Archangelsk mine kimberlite ore irradiated at different excess level (σ) at different time interval. Time measured in seconds (s).

σ	5 s	10 s	15 s	20 s	25 s	30 s
1.75	0.56	0.98	0.99	1.00	1.00	1.00
1.88	0.48	0.98	0.98	1.00	1.00	1.00
2.00	0.32	0.92	0.95	1.00	1.00	1.00
2.13	0.20	0.76	0.90	0.94	0.98	0.99
2.25	0.12	0.66	0.77	0.81	0.90	0.92
2.38	0.08	0.55	0.61	0.63	0.73	0.78
2.50	0.04	0.28	0.40	0.51	0.61	0.68
2.63	0.02	0.19	0.22	0.39	0.43	0.47
2.75	0.01	0.05	0.15	0.30	0.26	0.33
2.88	0.01	0.02	0.08	0.14	0.14	0.21
3.00	0.01	0.01	0.05	0.06	0.08	0.12
3.13	0.00	0.01	0.02	0.04	0.02	0.05
3.25	0.00	0.01	0.02	0.02	0.00	0.00
3.38	0.00	0.01	0.01	0.01	0.00	0.00
3.50	0.00	0.00	0.00	0.00	0.00	0.00
3.63	0.00	0.00	0.00	0.00	0.00	0.00
3.75	0.00	0.00	0.00	0.00	0.00	0.00
3.88	0.00	0.00	0.00	0.00	0.00	0.00
4.00	0.00	0.00	0.00	0.00	0.00	0.00
4.13	0.00	0.00	0.00	0.00	0.00	0.00
4.25	0.00	0.00	0.00	0.00	0.00	0.00
4.38	0.00	0.00	0.00	0.00	0.00	0.00
4.50	0.00	0.00	0.00	0.00	0.00	0.00

Table C.2: Archangelsk mine kimberlite ore irradiated at different excess level (σ) at different time interval. time measured in seconds (s).

σ	35 s	40 s	45 s	50 s	60 s	65 s
1.75	1.00	1.00	1.00	1.00	1.00	1.00
1.88	1.00	1.00	1.00	1.00	1.00	1.00
2.00	0.99	0.99	1.00	0.99	1.00	1.00
2.13	0.98	0.98	0.97	0.98	1.00	1.00
2.25	0.93	0.92	0.92	0.93	0.98	0.98
2.38	0.80	0.83	0.83	0.87	0.90	0.89
2.50	0.67	0.71	0.71	0.72	0.77	0.83
2.63	0.48	0.51	0.54	0.44	0.63	0.61
2.75	0.33	0.39	0.36	0.28	0.46	0.47
2.88	0.18	0.24	0.22	0.17	0.31	0.32
3.00	0.13	0.12	0.14	0.12	0.21	0.19
3.13	0.06	0.09	0.10	0.09	0.15	0.12
3.25	0.05	0.05	0.08	0.05	0.09	0.06
3.38	0.05	0.02	0.06	0.04	0.04	0.06
3.50	0.02	0.01	0.02	0.03	0.03	0.06
3.63	0.01	0.00	0.02	0.02	0.02	0.05
3.75	0.00	0.00	0.01	0.01	0.02	0.02
3.88	0.00	0.00	0.00	0.00	0.01	0.01
4.00	0.00	0.00	0.00	0.00	0.01	0.01
4.13	0.00	0.00	0.00	0.00	0.01	0.01
4.25	0.00	0.00	0.00	0.00	0.01	0.01
4.38	0.00	0.00	0.00	0.00	0.01	0.00
4.50	0.00	0.00	0.00	0.00	0.00	0.00

Table C.3: Letseng mine kimberlite ore irradiated at different excess level (σ) at different time interval. time measured in seconds (s).

σ	10 s	20 s	30 s	40 s	50 s	60 s	70 s	80 s
1.00	0.00	0.00	0.7	0.99	1.00	1.00	1.00	1.00
1.25	0.00	0.00	0.43	0.97	1.00	1.00	1.00	1.00
1.50	0.00	0.00	0.29	0.90	0.99	1.00	1.00	1.00
1.75	0.00	0.00	0.20	0.66	0.89	0.93	0.99	0.99
2.00	0.00	0.00	0.07	0.30	0.67	0.69	0.80	0.77
2.25	0.00	0.00	0.05	0.18	0.31	0.39	0.44	0.54
2.50	0.00	0.00	0.01	0.09	0.14	0.14	0.26	0.27
2.75	0.00	0.00	0.00	0.06	0.04	0.10	0.11	0.10
3.00	0.00	0.00	0.00	0.00	0.01	0.03	0.03	0.04
3.25	0.00	0.00	0.00	0.00	0.00	0.03	0.03	0.04
3.50	0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.03
3.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01
4.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table C.4: Letseng mine kimberlite ore irradiated at different excess level (σ) at different time interval. time measured in seconds (s).

σ	90 s	100 s	110 s	120 s	130 se	140 s	150 s	160 s
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.25	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.50	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.75	0.99	0.96	0.97	0.97	0.99	0.98	0.97	1.00
2.00	0.83	0.87	0.87	0.86	0.86	0.91	0.89	0.83
2.25	0.57	0.61	0.61	0.69	0.69	0.67	0.63	0.61
2.50	0.27	0.23	0.34	0.31	0.33	0.34	0.40	0.33
2.75	0.09	0.04	0.10	0.13	0.13	0.17	0.13	0.13
3.00	0.02	0.04	0.04	0.03	0.06	0.03	0.03	0.03
3.25	0.03	0.03	0.03	0.01	0.01	0.01	0.01	0.01
3.50	0.03	0.01	0.01	0.01	0.01	0.01	0.01	0.01
3.75	0.01	0.01	0.01	0.01	0.00	0.00	0.00	0.00
4.00	0.01	0.01	0.01	0.01	0.00	0.00	0.00	0.00
4.25	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00
4.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6.25	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

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