SPECTRUM STRIPPING ANALYSIS OF THE INHERENT SPECTRAL PERFORMANCES OF CHEMICAL VAPOR DEPOSITION (CVD) DIAMOND DETECTORS EXPOSED TO ALPHA-PARTICLE RADIATION

BY

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DEPARTMENT OF PHYSICS

FACULTY OF SCIENCE

AHMADU BELLO UNIVERSITY, ZARIA, NIGERIA

OCTOBER, 2012.

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(M.Sc/SCIEN/10121/2007-2008)

A THESIS REPORT SUBMITTED TO THE POSTGRADUATE

SCHOOL, AHMADU BELLO UNIVERSITY, ZARIA,

IN PARTIAL FULFILMENT OF THE REQUIREMENT FOR THE AWARD OF MASTERS OF SCIENCE (M.Sc.) IN RADIATION BIOPHYSICS

DEPARTMENT OF PHYSICS

FACULTY OF SCIENCE

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OCTOBER, 2012.

DECLARATION

I Abdullahi Muhammad Vatsa hereby declare that this thesis is titled "**Spectrum Stripping Analysis of the Inherent Spectral Performances of Chemical Vapor Deposition (CVD) Diamond Detectors Exposed to Alpha-Particle Radiation**" is my original work and has not been submitted previously for higher degree. All quotations and sources of information are duly acknowledged.

_____	_____	_____
Abdullahi Muhammad Vatsa	Signature	Date

CERTIFICATION

The thesis entitled "**Spectrum Stripping Analysis of the Inherent Spectral Performances of Chemical Vapor Deposition (CVD) Diamond Detectors Exposed to Alpha-Particle Radiation**" by ABDULLAHI MUHAMMAD VATSA meets the regulation governing the award of Master of Science in Radiation Biophysics of the Ahmadu Bello University, Zaria and is approved for its contribution to knowledge literary presentation.

Dr. Y. I. Zakari (Major Supervisor)	Signature	Date
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Dr. U. Sadiq (Head of Department)	Signature	Date
Prof. A. A. Joshua (Dean, Postgraduate School)	Signature	Date

DEDICATION

This piece of work is dedicated to Allah (SWT) and to my sweet, dear, loving, caring and very large family.

ACKNOWLEDGEMENT

I am indeed grateful to Allah (SWT) for seeing me through this programme. My sincere thanks goes to my lovely parents Alhaji and Hajia Muhammad Sule Vatsa, my step mothers, my entire family, my brothers and sisters, uncles, aunts, cousins, nieces, my wife-Salamatu Ibrahim and son-Abdussabur Abdullahi for their encouragement, moral and financial support all through the period of my study.

I wish to acknowledge the support, encouragement and useful suggestions given to me by my supervisor Dr. Y.I Zakari, whom I owe a great debt for painstakingly guiding and accommodating me all through despite tide schedules, which made my efforts acceptable to this prestigious university (ABU). My heartfelt appreciation also goes to the Head of Department, Physics; and who was my minor supervisor Dr. U. Sadiq for his tireless pursuit of academic excellence in the department and has been a source of inspiration throughout the programme. I pay tribute to the entire academic and non-academic staff members of Physics Department. May Allah (SWT) reward you all abundantly.

I wish to acknowledge several buddies/friends/mentors like Dr. B. I. Tijjani, Dr. Y. Ahmed, Dr. T. O Ahmed, Dr. Abdulmalik Amouka, Raimi Jimoh, N.N. Garba, Abubakar Khalid Abubakar (A.K.A), Abdussalam Galadima Isma'il, Mal. Rabiu Adamu, Mal. Muhammad Lawal, Mal. A.U. Abubakar, Mal. Bashir Jatau, Aminu Isma'il. They all tolerated me and shared in the defeats and triumphs accompanying the research efforts and also to those whose names were not mentioned I truly appreciated everything.

Finally, my thanks go to everyone who has helped to ensure the success of this thesis. May Allah bless you all, amin.

ABSTRACT

The research put forth the outcome of the spectrum stripping analysis used to remove the defects and background noise levels from the experimental alpha spectra of nine Chemical Vapour Deposition (CVD) diamond wafers when used as detectors of α-particles radiation. The spectra, each with different spectral width/peak broadening attributable to the presence of defect/impurity concentration were analyzed for statistical noise and fluctuations. The objective was to provide a spectra resolution necessary for comparative spectrometric analysis to be performed on the detectors under investigation. The detectors average leakage current was measured to be 5.2pA for Single Crystal (SC); 21.5pA for Detector Grades (DG); and 5.2pA for Optical Grades (OG) and its contribution to the noise level analyzed. The poor spectra resolution was reported to be due not only to difference in defects concentration but also to background statistical effect and noise levels. The observed effect of defects and background effects on the experimentally acquired spectrum was analyzed using a High Performance Language MATLAB. An average spectrum peak resolution of 9.5% for DG, 67.4% for OG and 1.7% for SC was calculated. The average absolute efficiency of 0.19% for DG, 0.15% for OG and 0.12% for SC, full energy peak efficiency of 87% for DG and OG, 96% for SC and the particle range from the alpha particle range-energy plot were all analyzed and the results compared. The results show an improvement on the experimental observations without affecting the detectors inherent characteristic performance.

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Appendix I

CHAPTER ONE

1.0 INTRODUCTION

Historically, the first artificial synthesis of diamond was achieved in 1953 (although the work was never published) at Alemanna Elektris Svenska Aktiebolaget (ASEA). In 1955 the General Electric scientist published an article where it reported the synthesis of artificial diamond crystal through technique based on the use of high pressures and temperatures. The first growth of diamond at low pressure was realized by W. Eversole in the early fifties of last century (first group ASEA) who synthesized diamond particles using arcs of graphite, thus showing that the diamond can be produced in the region of the metastable phase diagram of carbon. From these efforts, much progress has been made, especially in recent years. The development methods for the synthesis of diamond CVD (Chemical Vapour Deposition) was the result of discovery and interaction among Americans, Russians and Japanese research groups. Understanding the role played by hydrogen atoms and molecules in the process of nucleation and growth over the years has led to significant improvements in outcomes such as the increase in the rate of growth, the suppression of nucleation of carbon graphite and the possibility of nuclear crystals of diamond on different substrates by the same diamond. This last feature of the CVD process allows the growth of polycrystalline diamond on substrates of different material (e.g. Si) for growth surfaces extended to several square centimeters and thicknesses ranging from few microns to at least an inch.

The mechanism that leads to the growth of CVD diamond has not been fully elucidated. The most reliable theories assume a simultaneous deposit of hybridized sp^3 carbon (diamond) and sp^2 (graphite), but the presence of hydrogen facilitates the removal of

graphite. The formation of diamond was facilitated by hydrogen because it reacted on the surface of the film with the hybridized sp^2 free carbon bindings, forming a uniform layer of saturated hydrocarbons which prevents a single species of atoms to go and settle on the surface. This theory was supported by the observation that if you remove the layer of hydrogen, there is a rearrangement of atoms on the surface (Almaviva, 2010).

With the advent of CVD techniques for growing polycrystalline diamond in the early nineties, the latent interest in diamond radiation detectors was revived. The material proved capable of detecting various types of radiation, starting from the visible and ultraviolet wavelength ranges, reaching far into the X-ray and gamma-radiation regions. Diamond is also known as a thermo-luminescent (TL) material for α, β, and γ radiations. Its properties: as thermoluminescence and photoluminescence dosimeters, scintillation detectors, and ionization chambers (photoconductive detectors) for detecting ionizing radiations has been documented (Keddy and Nam, 1993). Due to its extreme hardness, and hence, its ultimate resistance to radiation damage, the diamond has an important advantage over other solid-state materials for applications to heavy charged particles and high-energy, minimum ionizing particles from accelerators (Souw and Meilunas, 1997).

Recent developments in diamond synthesis have enabled the production of detector grade polycrystalline diamond wafers and high-purity single-crystal plates. These materials enable diamond to be exploited in a range of sensing applications. Diamond can be used as a solid-state ionizing chamber for environmental radiation measurement. This technology was being researched by a number of universities and scientific groups with growing success. Until recently, two of the factors limiting exploitation of this emerging technology

were the availability of material and its subsequent processing to provide industry and research groups with packaged sensors.

Natural diamonds have been used since the 1920s, but rarity and high cost have limited their use to research and a small number of medical dosimetry applications. The availability of high purity, high quality synthetic diamond enables, for the first time, the fabrication of diamond detectors in volume without the drawbacks associated with natural diamonds. One field of future applications of CVD (chemical vapor deposition) diamond could be particle detection in high energy physics experiments, where fast, radiation-hard detectors are required (Friedl, 1999). Tremendous attempt have been made to develop CVD diamond alpha particle detectors. However, more attention has been paid in the improvement of the detector performance. The polycrystalline nature of films makes the detector performance strongly depend on the microstruction of the CVD diamond films (Wang *et al.,* 2005).

Despite the large interest in this material very little information was available of the electrically active defect of CVD single crystal diamond. In particular, in most cases, undesirable effects as very slow response time or pumping effects have been observed indicating the presence of defect in the material (Balducci *et al.*, 2005, Mavunda *et al.,* 2008). The exceptional properties (such as: electronic, optical, mechanical, etc) of synthetic diamond film grown by CVD techniques give excellent features in terms of high radiation resistance, low leakage current, high temperature operation, high chemical inertness, and tissue equivalent. Thus, it has been expected that CVD diamonds radiation detectors can be used for many application in several fields, among which there is the realization of nuclear detectors which are being actively investigated. The high resistivity permit a simple

structure, and the very high band gap result in an extremely low number of free carriers leading to very low noise and power dissipation, while the high carrier mobilities and saturation field allow fast charge collection (Wang *et al.,* 2005).

An alpha-particle going through any material will have interactions affecting its detection in two ways: first, the energy spectrum is distorted because of the energy loss caused by the interactions in any mass interposed between source and detector. Second, a particle entering the active detector volume will interact there at least once and will be detected, i.e. the efficiency is practically 100 percent (Tsoulfanidis, 1983).

Since any energy loss outside the detector is undesirable, the task of the experimenter is to design a spectrometer with zero mass between the source and the detector. Such an ideal system cannot be built, and the only practical alternative is a spectrometer that results in such a small energy loss outside the detector that reliable corrections can be applied to the measured spectrum.

In certain measurements, the particles do not stop but they go through it and emerge with only a fraction of their energy deposited in the detector. Then a correction to the spectrum of the existing particles will have to be applied because of energy straggling, a term used to describe the statistical fluctuations of energy loss. Energy straggling should not be confused with the statistical effects that result in the infinite energy resolution of the detector (Tsoulfanidis, 1983).

The use of radiation as a clinical diagnostic tool has increased in recent years. For a greater control over treatment from the radiation beams, it will be pertinent to have an improved system of measuring the radiations. The need to put forth an improved comparative evaluation of the inherent spectrometric performances of CVD diamond detectors exposed to α-particles radiation propels this work. This is to provide a spectral resolution of the various grades of the CVD diamond. An understanding of the types of radiation interaction likely to be encountered requires a thorough and quantitative knowledge of the activity of the source, the type, and in the case of poly-energetic radiations, the energy range of the spectrum of all the sources likely to be used for the analysis (Seelentag *et al.,* 1979).

From the work of Mavunda *et al*. (2008), the analysis has established that a spectrum resolution is an indication of the quality of the detector material. The contributions of the fluctuations and pile-up in the signal set important limits on the detector's performance. This work examined the energy spectrum of the alpha-particle radiation sensed by the diamond detectors with a view to provide a spectrum independent of the detection system used and free from defects and impurities; using a High Performance Language (MATLAB) to design a model for stripping off the unwanted responses acquired from the experimental performances of the sensing devices (diamond wafers).

1.1 Statement of Research Work

The detector performance and stability are determined by structural imperfections and impurities in the material, which influence the induced current signal. In polycrystalline diamond detectors it was known that grain boundaries act as charge trapping centers,

severely limiting the spectroscopic performance (Lohstroh *et al.,* 2007). Diamond slabs grown by Chemical Vapour Deposition (CVD) comprised of many diamond crystallites of different orientations and separated by grain boundaries. These may affect the electrical properties of the material by acting as traps and/or recombination centres, thus degrading the charge collection efficiency (Beckman *et al*., 1997). Again, devices based on single-crystal high-pressure, high-temperature diamond (HPHT) was found to be limited by the small area of the available substrates. Moreover, homoepitaxial growth does not rule out completely the presence of defects as in natural diamond or in other single crystal materials. The study of defects with many different solid state and nuclear techniques has given a deeper insight on the physics of the transport in CVD diamond (Mavunda *et al.,* 2008), yet a thorough picture of defects and their influence on carrier lifetime in CVD diamond was lacking (Mersi *et al*., 2004).

These observed limitations, indeed jeopardize or pose detrimental effects on the use of diamond detectors for spectrometric or dosimetric performances. To improve on these limitations a correction is needed on the number of electron – ion pairs created as a function of the ionization energy of the diamond detector and energy deposited. To successfully correct these problems, analytical examination of the experimentation shall be generated from the observed spectrum. This will enable the formulation of stripping algorithm for the work. In order to increase the assurance of this work, the analytical formulation (program algorithm) shall be coded using a High Performance Language (MATLAB) to show the stripping process.

1.2 Justification of the Research Work

Severe detrimental effects of defect levels may occur in diamond. These effects can significantly alter the detection properties of devices. In fact, it was clear that the response of devices was reduced through defects and impurities (Bergonzo *et al.,* 2001). If the device state is to be optimized either due to ionization mechanism (trapping), or a significant rise in temperature (trap release), the internal electric field as well as the carrier lifetime of the device can be modified thereby influencing the detector response to give a better result for the problem. This work shall make use of MATLAB to optimize the observed experimental spectra (Mavunda *et al.,* 2008) of the three grade of CVD diamonds used as detection elements.

1.3 Aim and Objectives of the Research Work

The aim of this work is to find out the true or exact detector response (performance) on optimizing the peak resolution from the experimentally observed spectrum of three broadly classified types of diamonds (detector grade, optical grade and single crystal) that were produced by Chemical Vapor Deposition (CVD) as they responded to

α- particle radiation when used as detection elements.

The objectives of this work are:

- To identify reasons for poor spectrometric performance of three grades of CVD diamond detectors.
- To strip off pile-up, background noise or fluctuations, ionization and excitation energy losses effects.

- To determine a spectral resolution needed for spectrometric analysis to be performed.
- To improve on the poor grade diamond wafer that can be economically use for alpha spectrometry.
- Compare the experimental and strip spectra in terms of peak efficiency and resolution.

1.4 Scope of the Research Work

With the progress in the technology of personal computers, PC-based software packages started to play a key-role in the control, acquisition and validation of the data in any nuclear experiment. Because of the range of applications of alpha-particle spectrometry, analysis software packages were among the most used software in any nuclear laboratory. They were being used in such important applications as environmental studies, low level monitoring, radioactive waste analysis and safeguards (Blaauw *et al.,* 1999).

Alpha-particle spectra were usually very complex to analyze. Only a few radionuclides emit monoenergetic alpha particles and very often two or more lines from the same radionuclide overlap. Most alpha emissions have energies between 4 and 6MeV, and with the energy resolution achievable in realistic conditions, a typical alpha-particle spectrum contains one or several groups of overlapping lines. The difficulty of the analysis depends on the amount of information that one tries to obtain from the spectrum and reaches its maximum when a full analysis was performed to obtain the energies and intensities of all spectral components (Blaauw *et al.,* 1999).

This work intends to use a high performance language (MATLAB) to code a simple stripping procedure from the analytical formulation that is formed considering the measures of likelihood as the three broadly classified CVD diamonds detectors interact with α-particle radiation.

CHAPTER TWO

2.0 LITERATURE REVIEW

A radiation detector can yield important information on the nature of the incident radiation or the behavior of the detector itself and is therefore one of the most important functions to be performed in nuclear measurements. The rapid advance in detector development and radiation spectrometry/dosimetry over the past decades would have been impossible without the parallel development of reliable instruments to record the spectra from these instruments under a variety of conditions (Knoll, 2000).

The use of diamond as radiation sensing device (detector) have been reported by various authors: In a work carried out by Bergonzo *et al.* (1999) on thin film diamond alpha detectors for dosimetry applications. Diamond was seen as a resilient material with rather extreme electronic properties. As such it was found an interesting material for the fabrication of high performance solid state particle detectors. However, the commercially accessible form of diamond, grown by chemical vapour deposition (CVD) methods, was polycrystalline in nature and often displays rather poor electrical characteristics. Their work considered the way polycrystalline (diamond films) materials can be used as alpha particle dosimeters. One approach adopted has been to reduce the impurity levels within the feed-stock gases that were used to grow the diamond films.

High collection efficiency has been demonstrated for alpha detectors fabricated from CVD polycrystalline diamond films by optimizing film growth conditions and the geometric structure of the device. Diamond films with a thickness of 100mm have demonstrated a

charge collection efficiency of 40% when in a sandwich configuration, whilst collection efficiencies as high as 70% have been achieved with planar device designs.

Bergonzo *et al.* (2001) investigated the growth of high quality chemically vapour deposition (CVD) polycrystalline diamond as well as specific material and device processing. CVD diamond films were grown using the microwave plasma enhanced technique. Deposition processes were optimized according to the application requirements. This includes the synthesis of films with high sensitivity, with weak optical absorption in the ultra-violet visible domain or with short carrier lifetime. The synthesis of CVD diamond has led to significant improvements in its electronic properties.

One of the inherent problems with diamond was the presence of defect levels altering the detection characteristics: these may be the cause of an observed instability of the device responses. Their work has found, however, that it was possible to moderate these trends through the fine-tuning of the growth conditions and of the device preparation steps. Films with thicknesses ranging from 5 to 500μm have been used for detector fabrication. The role of post-growth treatments and the contact formation procedure was also extensively studied, leading to significant improvements of the detector characteristics.

In yet another work by Bergonzo *et al.* (2002) chemically vapour deposition (CVD) diamond was found a remarkable material for the fabrication of radiation detectors. In fact, there exist several applications where other standard semiconductor detectors do not fulfil the specific requirements imposed by corrosion, hot and/or high radiation dose environments. Developing on a CVD diamond-based detector through improvements in the

material synthesis as well as on device fabrication especially concerning the synthesis of films that do not exhibit space charge build up effects, which were often encountered in CVD diamond materials and that are highly detrimental for detection devices was their concern.

Kaneko *et al.* (2003) carried out successfully an investigation of a radiation detector made of a diamond single crystal grown by a chemical vapor deposition (CVD) method. The diamond single crystal, having a size of $2.0\times2.0\times0.7mm^3$, was then applied to the detector. The CVD method was able to reduce nitrogen and boron impurities in the diamond crystals more effectively than a high-pressure and high-temperature (HP/HT) method. Consequently the detector had high-energy resolution of 0.4% for 5.486MeV particles from ^{241}Am source. The behavior of charge carriers in the detector could not be adequately investigated because the detector had a partial depression layer and strong rectification. Ionization energy of the CVD diamond was determined to be 16.1eV; this value was higher than that of the HP/HT type IIa diamond, i.e., 13.1eV. This result shows the potential of CVD diamond single crystals because an ideal diamond should have higher ionization energy than the HP/HT type IIa diamond that had the influence of impurities.

In a radiation hardness and linearity studies of CVD diamonds by Behnke *et al.* (2003) the behavior of CVD diamonds under intense electromagnetic radiation and on the response of the detector to high density of deposited energy were studied. In their work, diamonds detectors have been found to exhibit a linear response to better than 2% for energy depositions of up to $250GeV/cm^3$. Properties of CVD diamond detectors like their radiation hardness, their characterization using the Thermally Stimulated Current (TSC) method and

their linearity response to large amount of electromagnetic deposited energy, have been studied.

Fidanzio *et al.* (2004) examined the performance of a radiation detector based on a 43μm thick polycrystalline diamond film. It was obtained that hot filament CVD that has been tested under realistic clinical conditions of irradiation generated by conventional photon and electron beams generally used for radiotherapy purposes. The linearity of the collected charge with the dose was verified and the sensitivity per unit of the detector sensitive volume was evaluated to be $20nCGy^{-1}$ mm^{-3} with an applied electric field of 23kV/cm. A correction factor $\Delta = 0.94$ was found for the detector current dependence on the dose rate working under the conditions of maximum signal-to-noise ratio. However, even a $\Delta = 1.00$ may be obtained when working with higher polarization electric field but at the expense of a lower signal-to-noise ratio. When the diamond detector signals were corrected for the dose rate dependence, the Percentage Depth Signal and the Output Factor values were in agreement with the results obtained with an ionization chamber well within the experimental uncertainty.

Guerrero *et al.* (2004) worked on the requirement for synthetic diamond devices for radiotherapy dosimetry application and put forth that, CVD diamond was a remarkable material for the fabrication of radiation detectors. One particular interest of diamond stands in the fact that its atomic number was close to that of human tissues. This implied that the response of a diamond device to radiation was close to that received by the human body. It thus enabled the straightforward measurement of the dose for radiotherapy applications. However, this required high reproducibility and linearity. It was widely observed that

radiation exposure was modifying the initial performances of diamond detectors and priming devices was therefore required to obtain the required linearity. However, the nature of defects in the material strongly influences the type of priming required (Guerrero *et al.,* 2004). Priming defines the pre-irradiation that enables to fill up the shallow as well as deep levels in order to stabilize the photo-sensitivity of the device.

CVD diamond was studied as an ionization chamber and thermoluminescence (TL) dosimeter for radiotherapy application. The technology has demonstrated that current grade poly-crystalline material grown using the CVD technique was a good candidate for this application even though improvements can be expected from the growth aspects as well as from the optimization of the measurement protocol. Diamond ionization chambers remain stable and reproducible as long as the duration between irradiations was kept short. Diamond TL dosimeters are luminescent in the same spectral region as Lithium Fluoride (LiF) but linearity to the dose response can be improved by addressing the impurity concentration in the material.

In the work carried out by Kagan (2005), "Diamond radiation detectors may be forever!" Chemical Vapor Deposition (CVD) diamond particle detectors were developed as a radiation tolerant replacement or upgrade to present detectors in extreme radiation environments. He grew large polycrystalline CVD wafers which were of very high quality, investigated the radiation hardness of the material up to fluences of 3×10^{15}p/cm^2. Their radiation resistance was beyond that required for 10years of Large Hadron Collider (LHC) operation. Polycrystalline CVD diamond trackers have achieved a signal-to-noise of 60:1 with 2μs shaping time and signal-to-noise of 8:1 with 25ns peaking time. Single-crystal

CVD diamond was also developed. The results indicated that diamond detectors can be constructed and operated for many years in high radiation areas with sufficiently high efficiency and spatial resolution.

Wang *et al.* (2005) reported on the transient response of room temperature pulses generated from alpha particles (i.e. alpha particle irradiation) from a chemical vapor deposition (CVD) polycrystalline diamond detector. For transient signals dominated by electron transport only prompt pulse shapes were observed with an average rise time of 160ns limited by the preamplifier rise time. For transient signals dominated by hole, transport of significant slow components were observed in the majority of pulses due to thermal emission of charge from shallow hole traps. These slow pulses were observed from the device when in its as-grown state, without any previous 'priming'. Two separate slow components were identified from the hole pulses, with average rise times of 600ns and 1–10μs, respectively. These data suggested that room temperature electron transport in polycrystalline CVD diamond was prompt, with no evidence for thermal de-trapping. In contrast, hole transport at room temperature contains a significant delayed component due to thermal emission of holes from at least two bands of shallow defects.

In the work by Betzel *et al.* (2010) on the operating parameters of CVD diamond detectors for radiation dosimetry. A prototype clinical radiation detector based on commercially available single crystal diamond film made via chemical vapor deposition was investigated to determined optimal operating parameters for clinical dosimetry. This work examined how changes in applied electric field affected dosimetric performance, and determined a preferred operating voltage within the limits of clinical dosimetry equipment. Dosimetric

analysis included leakage current, response dynamics such as rise and fall times, sensitivity, polarity and dependence on dose and dose rate.

The results of this work indicated a preference for using a setting of 62.5V due to its minimal rise time of 2s, including a faster time to equilibrium, yet sufficient sensitivity of $37nCGy^{-1}$, which was nearly independent of polarity. At this voltage, a wider range of dose may therefore be recorded with charge collecting instruments than at higher voltages. Leakage currents were negligible (< 2.5pA). The highest voltage setting of 248.0V resulted in the highest sensitivity of $235nCGy^{-1}$, but corresponding rise times of $\geq$ 7s may limit the use of this detector.

2.1 Sources and Attributes of Alpha particle

Alpha particles are positively charged and highly ionizing form of corpuscular radiation that consist of two protons and two neutrons bound together into a particle identical to a helium nucleus, which is produced in the process of alpha decay. The fundamental interactions responsible for alpha decay are a balance between the electromagnetic force and nuclear force. Alpha decay results from the Coulomb repulsion between the alpha particle and the rest of the nucleus, which both have a positive electric charge, but which is kept in check by the nuclear force (*en.wikipedia.org)*.

Alpha particles are commonly emitted by all of the larger radioactive nuclei such as uranium, thorium, actinium, and radium, as well as the transuranic elements (e.g. Americium-241, polonium-210, etc). Unlike other types of decay, alpha decay as a process must have a minimum-size atomic nucleus that can support it. The process of emitting an alpha sometimes leaves the nucleus in an excited state, with the emission of a gamma ray thus removing the excess energy (*en.wikipedia.org)*.

The energy of alpha particles emitted varies, with higher energy alpha particles being emitted from larger nuclei, but most alpha particles have energies of between 3 and 7 MeV (mega-electron-volts), corresponding to extremely long to extremely short half-lives of alpha-emitting nuclides, respectively. This energy is a substantial amount of energy for a single particle, but their high mass (6.644657×10^{-27} kg) means alpha particles have a lower speed than any other common type of radiation (β particles, neutrons, etc). Because of their charge and large mass, alpha particles are easily stopped by materials, and they can travel only a few centimetres in air. They can be stopped by tissue paper or the outer layers of human skin (about 40 micrometres, equivalent to a few cells deep) (*en.wikipedia.org*).

2.2 Physics of Alpha Particle Interaction with Matter

When radiation is composed of charged particles such as alpha-particle, the main quantity characterizing interaction of radiation with matter is the average decrease of particle kinetic energy per unit path length. This quantity is called the stopping power of the medium and is denoted S. An alternative notation is $-\mathrm{d}E/\mathrm{d}x$ or $|\mathrm{d}E/\mathrm{d}x|$ (such notation reflects the

mathematical meaning of the stopping power: it is opposite to the derivative of particle energy E relative to the traveled path x). The main mechanism of the energy loss of charged particles (and electrons with energies of the order of a few MeV or less) is ionization, excitation or radiative losses of the atoms of matter (Poškus, 2010).

2.2.1 Excitation, Ionization, and Radiative Losses

Energetic charged particles all interact with matter by electrical (i.e., coulombic) forces and lose kinetic energy via excitation, ionization, and radiative losses. Excitation and ionization occur when charged particles lose energy by interacting with orbital electrons. Excitation is the transfer of some of the incident particle's energy to electrons in the absorbing material, promoting them to electron orbitals farther from the nucleus (i.e., higher energy levels). In excitation, the energy transferred to an electron does not exceed its binding energy. Following excitation, the electron will return to a lower energy level, with the emission of the excitation energy in the form of electromagnetic radiation or Auger electrons. This process is referred to as de-excitation. If the transferred energy exceeds the binding energy of the electron, ionization occurs, whereby the electron is ejected from the atom. The result of ionization is an ion pair consisting of the ejected electron and the positively charged atom. Sometimes the ejected electrons possess sufficient energy to produce further ionizations called secondary ionization. These electrons are called delta rays.

In alpha particle interaction with the detector, energy is lost in discrete amount due to ionization and excitation; thereby leading to the observed statistical fluctuations or pile-up

and collisional energy losses while interacting with the orbital electrons. The deflection of the α-particle by a negligible angle as it interacts with the detector brings about scattering of ions. In practice, most radioactive sources emit a broad beam of radiation such that the detector registers not only events from the incident beam but also those scattered through large angles, and these effects must be included in computing detector response (Coursey, 2000).

Another consideration of utmost important is the energy range, which depends on how the particle interacts, either the particle follows very nearly a straight path or scattered or concentrated (pile-up) in the detector volume. The diamond wafers used have dimensions of up to $5 \times 5 \times 0.1 mm^3$ in volume. Two quantitative parameters used to describe inelastic interactions of charged particles as they slow down in matter are stopping power and range. The range of a charged particle of a given energy in a material is an important parameter in designing detectors. In general, a charged particle such as alpha-particle does not travel in a straight line as it slows down. However, the total rectified path length traveled by the alpha-particle is called the mean range (Coursey, 2000).

More so, electrons with high enough energy (in the MeV region) can create ion-pairs (ionization and excitation). Energetic electrons can further produce ions themselves by collision known as secondary electrons. When fast electrons interact in matter (detector) part of their energy is converted into electromagnetic radiation in the form of bremsstrahlung. The fraction of the electron energy converted into bremsstrahlung increases with increasing electron energy and is largest for absorbing materials of high

atomic number (Knoll, 2000). To determine the energy loss by the particle, we must include the primary and the secondary electrons as well as the atomic excitations.

2.2.2 Alpha Particle Ionization and Range

As charged particle (e.g. alpha particle) transverses a material, the particle slows as it loses kinetic energy. The loss of energy by charged particles traveling through a material is broken into two components based on the mechanism of energy transfer, either collisional or radiative energy loss. The total stopping power is

$$\frac{dE}{dx} = \left(\frac{dE}{dx}\right)_{col} + \left(\frac{dE}{dx}\right)_{rad} \quad (2.1)$$

where $(dE/dx)_{col}$ is the electronic energy loss due to Coulomb interactions (*i.e.*, the ionization and excitation), and $(dE/dx)_{rad}$ is the nuclear energy loss (*e.g.*, due to emission of Bremsstrahlung or Cerenkov radiation, and nuclear interactions).

2.2.3 Alpha Particle Tracks

A distinction between heavy charged particles (e.g. alpha particles) and electrons is their paths in matter. Electrons follow tortuous paths in matter as the result of multiple scattering events caused by coulombic deflections (repulsion and/or attraction). The electrons often exhibit a sparse non-uniform ionization track. On the other hand, the larger mass of a heavy charged particle results in a dense and usually linear ionization track. The path length of a particle is defined as the actual distance the particle travels. The range of a particle is defined as the actual depth of penetration of the particle in matter. The path length of the electron almost always exceeds its range, whereas the straight ionization track of a heavy charged particle results in the path and range being nearly equal. Note: The change of the direction of particle motion is called scattering (Poškus, 2010).

2.2.4 Estimation of Alpha Particle Range

It is not possible to fund range experiments for all possible incident particles, all possible absorbing materials at all possible energies. In order to estimate the range or energy loss characteristics of a particular example we must assume the validity of the Bethe – Bloch formula and assume that the stopping power per atom of compounds or mixtures is additive.

The latter assumption is known as the Bragg - Kleeman rule (Knoll, 1989). The formula relates the range of particles in a material for which we have no energy loss or range information if we have the range information in another material. The approximation becomes less valid as the separation in atomic weights increases. This rule is thus defined as: -

$$\frac{R_d}{R_a} = \frac{\rho_a \sqrt{A_d}}{\rho_d \sqrt{A_a}} \qquad (2.2)$$

where R_d and R_a are the ranges in the detecting medium and air, ρ_a and ρ_d are densities of air and the detector material and A_a and A_d are the atomic weights of air and the detector material.

This rule follows from the fact that the range (cm) is inversely proportional to the density and taking ratio of ranges, one known and the other unknown. The atomic weight part is an approximation and the guts of the Bragg - Kleeman rule. The estimation of the particles range in a given material is a two step process: -

- Determine the range in air, then
- Determine the range in the material given the Bragg – Kleeman rule.

The range of alpha particles (cm) depends upon the material and the energy of alphas. The first part takes care of energy in air; the second part takes care of the difference in materials between air and the material of choice. Note; the limitations of this rule were ignored.

2.2.5 Determination of the Range Alpha in Air

In order to compute the range of alpha particles in any material, we must use empirical equation for the range of alpha particles in air. The following equation for range (cm) is valid for alphas in air in the energy range $4 < E < 15\ MeV$ (Lapp and Andrews, 1948).

$$R_a = (0.005E + 0.285)E^{\frac{3}{2}} \quad ; R_a \text{ in cm} . \tag{2.3}$$

A simpler equation for range (cm) for alpha in air in the energy range $4 < E < 8\ MeV$ (Sorenson and Phelps, 1980).

$$R_a = 0.325E^{\frac{3}{2}} \quad ; R_a \text{ in cm} \tag{2.4}$$

2.2.6 Scattering of Alpha Particle

Scattering refers to an interaction resulting in the deflection of a particle or photon from its original trajectory. A scattering event in which the total kinetic energy of the colliding particles is unchanged is called elastic. When scattering occurs with a loss of kinetic energy (i.e., the total kinetic energy of the scattered particles is less than that of the particles before the interaction), the interaction is said to be inelastic. For example, the process of ionization can be considered an elastic interaction if the binding energy of the electron is negligible compared to the kinetic energy of the incident electron (i.e., the kinetic energy of the ejected electron is equal to the kinetic energy lost by the incident electron). If the binding

energy that must be overcome to ionize the atom is significant (i.e., the kinetic energy of the ejected electron is less than the kinetic energy lost by the incident electron), the process is said to be inelastic.

2.2.7 Radiative Interactions-Bremsstrahlung

Electrons can undergo inelastic interactions with atomic nuclei in which the path of the electron is deflected by the positively charged nucleus, with a loss of kinetic energy. This energy is instantaneously emitted as ionizing electromagnetic radiation (x-rays). The energy of the x-ray is equal to the energy lost by the electron, as required by the conservation of energy. The radiation emission accompanying electron deceleration is called bremsstrahlung, a German word meaning "braking radiation". When the kinetic energy of the electron is low, the bremsstrahlung photons are emitted predominantly between the angles of 60 and 90 degrees relative to the incident electron trajectory. The probability of bremsstrahlung emission per atom is proportional to Z^2 of the absorber. Energy emission via bremsstrahlung varies inversely with the square of the mass of the incident particle. Therefore, protons and alpha particles will produce less than one-millionth the amount of bremsstrahlung radiation as electrons of the same energy.

2.3 Spectrometry

The result of a direct spectrometric process is a pulse height distribution not a photon spectrum. A device consisting of a detector, an amplifying system (amplifier) and a multichannel analyzer is principally a spectrometer (Seelentag and Panzer, 1979). Radiation

detectors are frequently used for measurements of radiation energy spectrum. Such measurements comprise the field of radiation spectroscopy. Spectroscopy is the application of radiation detector to measure the energy distribution or response function of the incident radiation. For the spectrum of a detector response function (Fig: 2.1), larger width corresponds to worse energy resolution and this makes it obvious that the widths of those peaks are different. A large width of a maximum means that the spread (fluctuations) of pulse was large, even though each interaction of a particle with the detector caused the same amount of energy to be transferred to the detector material. A smaller width correspond to a good or better energy resolution, which means that the detector can resolve two close peaks of the spectrum more easily (Knoll, 2000).

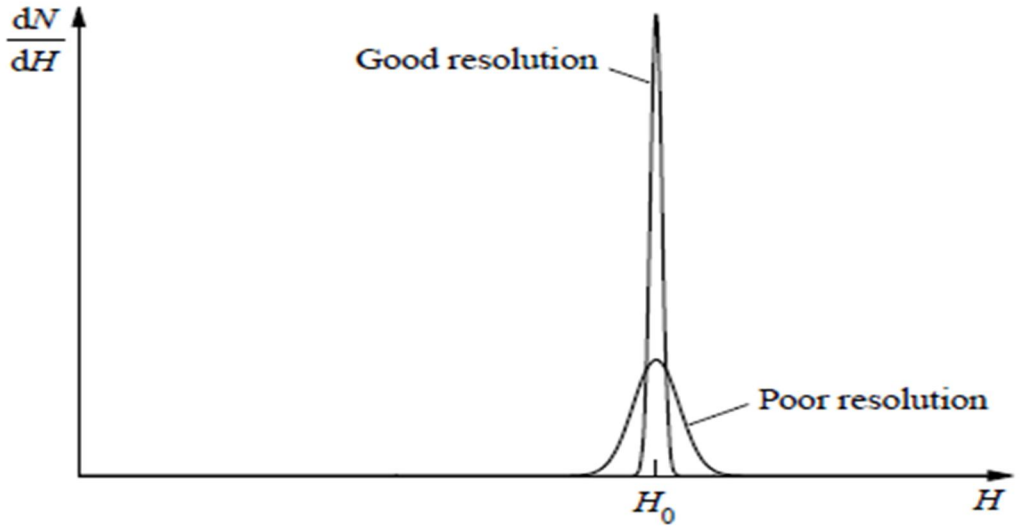


Fig. 2.1 Examples of response functions for detectors with relatively good resolution and relatively poor resolution.

The difference in peak resolution could partly be caused by the spread of the electric field along the material due to the polycrystalline nature of the material (diamond wafers) with non-uniform geometry and thickness (Tromson *et al.,* 2000). Also partly due to their state of modification either because of the ionization mechanism (trapping) or because of a

significant elevation in temperature (trap release), induced space charge build up (pile-up), the internal electric field in the device as well as the carrier lifetime lead to the observed spectral asymmetric broadening (Bergonzo *et al.,* 2002; Souw and Meilunas, 1997).

The statistical fluctuations/noise of pulses reflected the width of the response function and entails the most dominant source of fluctuations. This width usually measured at half-height of the peak (that is why it is abbreviated FWHM: "full width at half maximum"). If the peak should be Gaussian in shape, then FWHM can be related to a standard deviation. So that on the average it accounts for the number of charge carriers (ion-pairs) and assuming that the formation of each charge carrier is a Poisson process. This allows the inherent fluctuations to be calculated. The energy resolution is defined as the ratio of the width at half-maximum to the average pulse height (Knoll, 2000).

Alpha-particle spectroscopy demands the application of carefully designed techniques since alpha-particles continuously lose their energy as they travel through matter. Thick specimens cause a degradation of the final energy resolution affecting the quality of alpha spectra containing several overlapping peaks. It was nevertheless possible to employ sophisticated computer algorithms to resolve these close-lying alpha energies and obtain valuable information, from energies and intensities of all spectral components for nuclear decay data (Hurtado *et al.*, 2008).

2.4 Dosimetry

Radiation dosimeter is a device, instrument or system that measures or evaluates, either directly or indirectly, the dosimetric quantities or related quantities of ionizing radiation. Measurement of a dosimetric quantity is the process of finding the value of the quantity

experimentally using dosimetry systems. The result of a measurement is the value of a dosimetric quantity expressed as the product of a numerical value and an appropriate unit.

To function as a radiation dosimeter, the dosimeter must possess at least one physical property that is a function of the measured dosimetric quantity and that can be used for radiation dosimetry with proper calibration. In order to be useful, radiation dosimeters must exhibit several desirable characteristics. For example, in radiotherapy exact knowledge of both the absorbed dose to water at a specified point and its spatial distribution are of importance, as well as the possibility of deriving the dose to an organ of interest in the patient. In this context, the desirable dosimeter properties will be characterized by accuracy and precision, linearity, dose or dose rate dependence, energy response, directional dependence and spatial resolution (Almaviva, 2010).

2.5 Diamond and Diamond Detectors

Diamond is a material with a set of very unique characteristics. It is mainly known as a gem, but also for its hardness. Diamond is composed of carbon atoms arranged in the tetrahedron diamond lattice. The atoms stick together through strong sp^3 σ-type bonds. The small carbon atoms give a very dense, but low weight lattice. These facts give reason for the extraordinary characteristics of diamond. There is a third property that is not so well known; diamond shows extremely high thermal conductivity while it is electrically insulating. Besides that, diamond has the reputation of being radiation hard since the 1950s, but only recently this has been examined systematically using modern irradiation facilities.

These facts give reason for the extraordinary characteristics of diamond. The thermal conductivity of diamond is the highest of any material known; at room temperature it is five times higher than that of copper. Even more, it is coupled with electrical insulation, which is a very rare combination in nature. Specially treated synthetic diamond crystals conduct heat even better. Therefore, diamond heat sinks are used, e.g., in Pentium Processors, where a huge amount of thermal power has to be dissipated in a very small volume. Diamond is the hardest mineral known; therefore it is used for drilling and cutting applications (Friedl, 1999).

The main demerit of the natural diamond detectors was a high cost due to extremely rare detector grade natural diamond (Type IIa), which limits the availability of these detectors and moreover, electronic properties of diamond stones within the Type IIa category can vary strongly. Therefore, the production of sufficiently cheap diamond plates with sizes at least 4mm and thickness 50-500 µm with sufficient quality to build the detector was rather an urgent problem (Akulinichev *et al.,* 2003).

With a band gap of E_g = 5.5eV, diamond is regarded as an insulator. This implies negligible intrinsic carrier densities even at room temperature; allowing to operate intrinsic diamond film as a detector. Electrodes are applied to the diamond film on opposite sides to form Ohmic contacts. As there is no pn-junction, the polarity of the electric field is irrelevant. The dark current of the diamond samples, including both bulk and surface currents, is less than $1nAcm^{-2}$ at an electric field of $1V\mu m^{-1}$. According to the high carrier mobilities in diamond, the charge collection is very fast, taking about 1ns in detectors of approximately

500µm thickness. It has been shown that CVD diamond detectors are able to count heavy ion rates above $10^8 cm^{-2}\ s^{-1}$ with a single readout channel.

2.5.1 Types of Diamond

In the early 20th century, natural diamonds were divided into type I, containing nitrogen impurities, and type II, relatively free of nitrogen. Later, by refining the analysis methods, subgroups were introduced to the type terminology as shown in table 2.1. Natural diamond, which was found mainly as type Ia, was not applicable as a detector because of its nitrogen impurities. Reasonable detector material, synthesized in the CVD process, must contain less than 1ppm of nitrogen (type IIa). With natural or synthetic boron implantation, p-type semiconducting behavior was introduced to the material.

Table 2.1: - The Diamond Type Terminology

Type	Impurities	Comment
Ia	Aggregated nitrogen up to 2500ppm	Most natural diamonds
Ib	Substitutional nitrogen up to 300ppm	Most synthetic diamonds
IIa	Substitutional nitrogen < 1ppm	Detector material
IIb	Boron doped	p-type semiconductor

2.5.2 CVD Technique

Diamond detectors are grown in the chemical vapor deposition (CVD) process. A small fraction of hydrocarbon gas, such as methane, is mixed with molecular hydrogen and

oxygen gas. When the gas mixture is ionized, carbon based radicals are reduced and settle on a substrate, usually silicon or molybdenum, and link together with σ-type bonds, forming a diamond lattice.

The properties of the diamond grown in this process depend on the gas mixture, temperature and pressure. Although this is an easy principle, the growth process is extremely difficult to control in order to grow material suitable for detector application; the parameters are not constant throughout the process. The growth speed is typically about $1\mu mh^{-1}$. There are several types of CVD detectors, which differ in the way the gas is ionized; e.g., this is done by microwaves or by a heating wire. After the growth process, the substrate is etched from the diamond film, which is then cut and cleaned.

Initially, there is a large number of small crystal seeds on the substrate, each oriented individually. As deposition continues, the grains grow together, forming columnar single crystals with grain boundaries between. On the substrate side the lateral grain size is very small (in the order of micrometers), while the size continuously increases in the growth direction, reaching a diameter in the order of 100µm with a diamond film thickness of 500µm (Friedl, 1999).

It is generally believed that the following conditions are required for chemical vapor deposition of large-grain, high-quality diamond films (Regel and Wilcox, 2001):

1. A substrate on which diamond can nucleate and grow.
2. A substrate temperature of approximately 700 –1000◦C.
3. Atomic hydrogen at the growth surface.

4. A suitable hydrocarbon species at the growth surface.

2.5.3 CVD Diamond

The most common method for the production of diamond to be used in electronic applications is the chemical Vapor Deposition (CVD) technique due to the low cost of this method and the possibility to deposit diamond films in a controlled and reproducible way (Balducci *et al.*, 2005). CVD diamond has a number of properties which make it an attractive material for solid-state detector applications. Its large band gap (5.5eV) and large displacement energy (42eV/atom) makes it a material that should be inherently radiation tolerant with very low leakage currents. Its small dielectric constant (5.7) makes it a material with low detector capacitances and, thereby, low-noise performance of the associated front-end electronics. Its large thermal conductivity (between four and five times that of copper at room temperature) makes it a material whose devices may be operated without cooling. Although diamond appears ideal in many respects, it does have one major limitation: the large band gap which produces many of its outstanding properties limits the number of electron–hole pairs created when a particle traverses it (Kagan, 2005).

More so, these unique properties of diamond propels the use diamond materials as detectors of ionizing radiation operating in conditions imposing special requirements to stability of measurement of a dose, for example in medical installations for radiotherapy. Initially, natural diamonds with suitable electronic properties were used in radiation detection. This promising technology for synthesis of diamond materials, allows growing diamond material plates in controllable vacuum with specified thickness and sizes, which are determined by

sizes of substrates and duration of the process. However, the CVD diamond has a polycrystalline structure with crystallites sizes about 10-20% of thickness of grown plates, and crystallites bounders could act as the traps and decrease the charge collection efficiency (Akulinichev *et al.,* 2003).

CVD Diamond allows the practical and cost-effective realization of hard detectors for nuclear radiation. The growth of high-quality CVD layers on low-quality single-crystal HPHT diamond was also very promising. More so, CVD polycrystalline diamond now yields an average carrier drift length (charge collection distance or CCD) of about 250μm: This figure of merit together with the extremely low leakage current has allowed preparing diamond devices with fast electronics and a Signal-to-Noise (S/N) ratio of 10/1. However, polycrystalline natures of the material have slackened the efficiency in improvement (Mersi *et al*., 2004).

2.5.4 Radiation Detection with Diamond

When a heavy charged particle (e.g. alpha-particle) traverses a diamond wafer, energy is mainly transferred due to Coulomb interactions between the particle and the atomic electrons in the diamond wafer. In the diamond wafers with an atomic lattice, which can be described by the band model, the electrons are excited from the valence to the conduction band when the particle transfers enough energy. This process is known as electron-hole generation. At very high incident particle energies, bremsstrahlung radiation is emitted when collisions occur (Friedl, 1999).

The principle relied on the creation of electron–ion pairs within the diamond from the interaction of the incident particles or photons to be detected. The local displacement of these carriers driven by an electric field in the material will induce a transient signal on the device electrodes. A typical configuration consists of applying an electric field through the volume of the diamond layer in a sandwich configuration where electrical contacts are deposited on both sides. In the case of very thin devices, the wafer onto which was induced the growth may be kept as the back contact.

The electronic properties of diamond can be assessed from the measurement of the detection response characteristics under charged particles. A figure of merit deduced from this measurement was the charge collection efficiency Z defined as the ratio of the electrical charge Q_{ind} induced in the external circuit to the charge generated by the interaction of the incident particle in the detector Q_0: This ratio was assumed to be equal to that of the mean charge collection distance of free carriers, i.e. the $\mu\tau E$ product, by the detector thickness L. $\eta = \mu\tau E/L$, under the approximation of a uniform field profile distribution through the material and if L is large with respect to $\mu\tau E$:This has motivated several studies that led to materials exhibiting $\mu\tau E$ products as high as 200mm measured under minimum ionization particles, with special interests for the detection of high energy particles where the deposited energy is proportional to the detector thickness. In particular, it was found that the collection distance increases with the diamond layer thickness. However, in the case of short-range particles, the energy is deposited within the first few microns of the material, and there is here a strong interest in keeping the thickness low for high collection efficiency. This also tends to reduce polarization effects, i.e. the non-homogeneous

modification of the material properties due to localized trap filling on the entrance side of low range particles (Bergonzo *et al.,* 2002).

CHAPTER THREE

3.0 MATERIALS AND METHODS

3.1 Analytical Details

The data used in this thesis originates from an experimental in the Republic of South Africa. The experiment took place at Radiation and Health Physics Unit, DST/NRF Centre of Excellence in Strong Materials, and was performed by Mavunda *et al.* (2008) from the School of Physics, University of the Witwatersrand, Johannesburg. The goal was to characterize nine CVD diamond samples classified as electrical grade, optical grade and single crystal for used as a radiation probe.

The CVD diamond wafers were characterized and information about the level and types of impurities was source for different grades of the diamonds identified by the supplier as optical grade samples (OG1,OG2, OG3, OG4), detector grade samples (DG1, DG2,DG3, DG4), all polycrystalline, and a single crystal (CVD-SC) type. The diamonds wafers were all of dimensions $5\times5\times0.5mm^3$ and $5\times5\times0.1mm^{3,}$ for the polycrystalline and single crystal structures respectively. Each of the wafers were assayed, placing each in the magnetic field of a Bruker microwave bridge ESP 380 – 1010 coupled to a ESP 380 – 1020 controller and an ESP 300E ESR unit.

The CVD diamond wafers were placed 2.2cm from a 3.7×10^6 $Bqcm^{-2}$ Americium-241(or Am-241) alpha source for the acquisition of alpha spectrum using PC-based commercial software APTEC. The numbers of α-particles impinging on the surface of the wafers per second were estimated to have a value of 4.84×10^4. The CVD diamond wafers were probed with a bias voltage (100V), applied across the opposing metalized surfaces, and then

connected to a preamplifier (locally designed). The whole systems, including the alpha source were placed in a vacuum chamber. The preamplifier coupled to an oscilloscope for the pre-assessment of the signal, and a personal computer with APTEC software for data acquisition, processing and analysis. The alpha energy peak (5.48MeV) value was equated with the peak value of each spectrum to assess the energy per channel for a plot of the calibrated graph to be examined. The obtained experimental data from the detector's response (performance) in connection to the effect of their defects have been reported (Mavunda *et al.,* 2008). In order to examine the difference in noise level of the detector, the leakage current was measured.

The alpha particle spectrum of each of the CVD diamond detectors were analyzed for peak resolution, peak efficiency and absolute efficiency. The alpha spectrums, each experimentally acquired and calibrated were plotted, examined for the total counts. The counts were normalized to the exposure time of irradiation. A software package ORIGIN (6.1) was used for both baseline insertion and background subtraction. The baseline was inserted and modified so as to vide a better Gaussian fit owing to the asymmetric nature of the polycrystalline spectra (Mavunda *et al.,* 2008).

To correct for (strip off) the collisional and radiation losses, background noise and/or fluctuations buried in the observed experimental spectra of the three types of broadly classified diamonds that were produced by Chemical Vapor Deposition (CVD) as they responded to α-particle radiation when used as detection elements, the following will be required:

- The physics of charged particle interaction with matter (diamond wafers);

- A programming language (MATLAB);
- A computer;
- Adequate power supply

3.2 Stripping Methodology

Alpha particles (or charged particles) produced in nuclear scattering reactions will lose energy as they escape the target. The amount of energy lost depends on the particle species and the composition and thickness of the target through which the particle must pass. If the energy losses are significant, particles may stop in the target and the measured spectrum will be a distortion of the true spectrum. To correct for these losses (energy and particle), a method must be sort for, to unfold the information in the measured spectrum to yield the true spectrum (Rezentes *et al.,* 1995).

It has been found that the presence of defects and impurities in diamond and other synthesized diamonds (CVD diamonds) could be detrimental for device applications. This affects the response of diamond to radiation in different forms (these differences could either be peak resolution, asymmetric broadening, or sensitivity of the spectrum). The observed alpha responses of most devices in different forms was either because of the ionization mechanisms (trapping) or because of a significant elevation in temperature (trap release), the internal electric field in the device as well as the carrier lifetime and charge collection efficiency (Bergonzo *et al.,* 2002 and Mavunda *et al.,* 2008). All these different spectrum responses indicated the quality of the detector material to perform as a radiation sensing device (i.e. either a dosimeter or a spectrometer). To put forth a good stripping

methodology, it will pertinent to analyze the problem by looking at the interaction of alpha particle with radiation sensing material (diamond wafers).

Charged particles (e.g. alpha particle) interact with matter (diamond) in many different ways. These could include: inelastic collisions involving ionization and excitation of atomic electrons of the material, inelastic collisions involving bremsstrahlung (photon) production in the field of the atom, elastic scattering in the field of the atom etc (Coursey, 2000). Incident radiation (external electron beam) can consist of heavy charged particle. The interactions of these particles in the target will give rise to the excited atoms necessary for the emission of characteristic X-rays. Alpha particle emitted by radioisotopes sources are the most convenient source of the incident particle due to their convenient half-lives and relative freedom from contaminant electromagnetic radiation. Alpha particle excitation avoids the complication of bremsstrahlung associated with electron excitation and is therefore capable of generating a relatively clean characteristic X-ray spectrum (Knoll, 2000). Thus this may mean that, there are traces of bremsstrahlung (photon) production during an interaction in the charged particle (alpha particle) field perhaps in small amount.

To realize the needed correction (stripping) on the observed experimental spectrum, the combining measures of likelihood of an incident particle in a given energy interval producing an electron-ion-pair and the likelihood of the scattered (interaction losses) and pile-up fraction also deposited in the diamond wafers must be considered. Considering the possible interactions of an incident alpha particle with the detector material, the following probabilities need to be understood:

- The likelihood of an incident alpha-particle interacting and get deposited within the detector volume is measured.
- The likelihood of an ion-pair is produced (by ionization) due to energetic electrons (secondary eletrons) and result to subsequent release of bremsstrahlung (Compton Scattering) to deposit Compton recoil energy is formulated.
- The likelihood of the excited ions (pile-up and fluctuations) detected by the detector is computed.

The three measure of likelihood above would make it possible to calculate the fractional error. Further examining the acquired experimental spectra, the peak resolution can be enhanced. To do this, the observed electron-ion pair produced is stripped off in order to eliminate pile-up and the electronic noise from the background. Interactions of charged particle with a detector can be defined by measure of likelihood (Seelentag and Panzer, 1979) but a more useful definition is in terms of the stopping power or number of ion pairs formed per unit distance (specific ionization).

3.3 Data Analysis

3.3.1 Ion-Pair Formation per Alpha Particle

An ion-pair may be described as the formation of a positively charged ion and an equally charged negative ion (most usually an electron) resulting from an atomic interaction of an incident radiation particle or ray with a target neutral atom or molecule or medium. The average specific ionization of an alpha particle from ^{241}Am passing through a detector is a function of four parameters;

1) Kinetic energy, of alpha particle,

2) Velocity, of alpha particle,

3) Range of alpha particle in diamond, and

4) Ionization potential including secondary electron ionization.

The statistical average of ion pairs formed along the path of an alpha particle varies with the four above parameters. The initial kinetic energy of the alpha particle from decay of the ^{241}Am nucleus approaches relativistic velocities but began to decrease with distance (path length) because of elastic collisions and electron stripping interactions. The alpha-particle with an initial charge of 2+ and large mass (4 AMU) has a high specific ionization and began to slow down to a kinetic energy level of ~1MeV. This energy level is comparable to the energy of the orbital velocity of the outer K-shell electrons of the target wafers. At this velocity (kinetic energy) the alpha particle from decay of ^{241}Am began to experience inelastic collisions with the detector, which give rise to nuclear deceleration rather than electronic slow down. It was during this inelastic collision phase that optimum excitation might occur.

The range of an alpha-particle (emitted from the decay of ^{241}Am) in the diamond detector can be calculated using Bragg-Kleeman rule having known its range in air. For a given energy of an alpha particle and its range in the diamond wafers the specific ionization or number of ion-pairs formed per cm of travel can be calculated using:

$$I(E_i) = \frac{E_i(KeV)}{W \times R_{di}} \left(ionpairscm^{-1}\right) \qquad (3.1)$$

where E_i = incident energies of the alpha-particle, W = average energy expended to create an ion or electron-hole pair in diamond (for diamond the value of 12.8eV/ion-pair was reported (Pomoski *et al.,* 2005)), R_{di} = range of alpha particle in the detecting medium, i= 1, 2, 3, ------, n integers referring to each spectrum energy. Equation (3.1) shall be used to

calculate the number of ion-pairs that will be created for each 12.8eV energy (E_i) interacting with the diamond detector.

3.3.2 Fractional Energy of the Alpha Particles Lost As Radiation

Since a photon (bremsstrahlung) might be created in the field of a charged particle, the likelihood of the bremsstrahlung emitted interacting with the field to create an ion-pair has to be formulated. Alpha particle with energies below the value 12.8keV may have less close encounter with the atoms of the detector and hence such energies transferred to the atom causes excitation interaction which is dissipated in molecular vibrations, atomic emission of infrared, visible or ultra-violet radiations (Sorenson, 1938). Thus the small but not negligible bremsstrahlung produced due to energetic electrons (delta rays), internal conversion and Auger effect in the alpha particle interaction may be the major sources of the background counts observed below 1.02MeV energy of the experimental alpha spectra; and as to be accounted for to achieve the needed correction. Within the observed 1.02MeV energy range the most probable bremsstrahlung interaction is Compton scattering of the photon with the free electrons to deposit recoil energy. To account for the background ion-pairs due to energetic electrons and collisional losses of the Compton interaction of the bremsstrahlung with free electrons to recoil energetic electrons with maximum recoil energy of 1.022MeV deposited in the detector as sensed by diamond has to be formulated (Knoll, 2000).

The amount of energy transferred to the detector varies with the angle of deflection. As θ approaches zero, none of the energy is transferred. The maximum amount of energy is transferred when θ approaches 180^0. If each of the electron-ion pairs created has a kinetic

energy of m_oc^2 = 511keV or $e^+ = E_i - 0.511keV$ (knoll, 2000); thus, the fractional energy input of the alpha particles lost as radiation to create the electron-ion pairs that interact with bremsstrahlung can be expressed as

$$^1/_{E_{min}} - {}^1/_{E_i} = {}^2/_{1022} \quad (3.2)$$

Or

$$E_{min} = {}^{E_i}\Big/_{\left(1 + \frac{2E_i}{1022}\right)} \quad (3.3)$$

where E_{min} is the minimum amount energy of the outgoing photon which escapes the detector or lost as radiation.

Therefore, the energy that may have contributed to ion produced charges can be expressed as:

$$\text{i.e. } E_{max} = E_i - E_{min} \quad (3.4)$$

Or

$$E_{max} = \frac{2E_i^2}{1022 + 2E_i}\ (keV) \quad (3.5)$$

where the factor 1022keV of the scattering equation is the energy of two electron rest mass $2m_ec^2 = 2 \times 511keV$ (Knoll, 2000).

3.3.3 Determination of Fractional Error

Another factor related to the detector performance is the full energy peak. When the thickness of the detector is considerably larger than the maximum penetration distance of the alpha particles, the response function consists of a full energy peak plus a continuum of

lower amplitude events. The full energy peak correspond to all those alpha particles that are fully stopped within the active volume of the detector. The continuum represents partial energy loss events, which can corresponds to backscattering of alpha particles that re-emerge from the incident surface without having lost all their energy or the escape of bremsstrahlung photons generated along the track of an electron which otherwise may be fully stopped. A detector, whose intrinsic thickness exceeds that of the maximum penetration distance of alpha particles of interest, makes a very suitable alpha particle spectrometer (knoll, 1989).

Only a fraction $I(E_{max})$ of the number $I(E_i)$ of ion-pairs created for each of the 12.8eV energy interacting with the diamond detector is counted in the correct energy channel. The ratio is usually referred to as the full energy efficiency or spectrum correction factor (Seelentag and Panzer, 1979). Thus, the spectrum correction factor or full energy peak efficiency of the detector is formulated as:

$$f_{pi} = \frac{I(E_i) - I_{(max)i}(E_{max})}{I(E_i)} \tag{3.6}$$

where $I_{(max)i}(E_{max})$ = the number ion-pairs associated with radiation and collisional losses (Fewell *et al.,* 1977). This is calculated using the difference between E_i and E_{max}.

Thus, for the formulation of the detector efficiency characteristics due to the alpha particle interactions, the full energy peak efficiency values obtain using Eqs.(3.5) to (3.6) shall be used to calculate the inherent fractional error from the experimentally acquired data i.e.

$$f_{Ei} = 1 - f_{pi} \tag{3.7}$$

3.3.4 Expression for Stripped Counts

Corrections due to electronics and statistical noise are estimated assuming a Poisson distribution so that the fluctuations (excitations) and pile up are characterized by a standard deviation in the number of charge carriers. This correction takes care of the 0.05% factor from collisional loss (Sorenson, 1938). The probability of observing n number of charge carriers in a period of Δt is given a Poisson distribution as:

$$P\,(n, \Delta t) \;=\; \frac{m^n}{n!} e^{-m} \tag{3.8}$$

where m is the average number of charge carriers in the period Δt.

The standard deviation of the distribution is then given as:

$$\delta \;=\; \sqrt{m} \tag{3.9}$$

The corrected number of ion-pairs or stripped (ions) counts expression that takes into account all the ion-pairs produced either by collisional, radiation losses and excitations that have to be stripped off the spectrum is given as:

$$I_{pi} = \left| I_0(E_i) - \left(I_0(E_i) \cdot f_{Ei} + 2\sqrt{I_0(E_i).f_{Ei}} \right) \right| \tag{3.10}$$

where $I_0(E_i)$ = the experimental counts (ions) at energyE_i, f_{Ei} = the fractional error causing the background effects (collisional and radiation losses), pile up effect and fluctuations (excitations) observed in the spectrum and it is defined based on the spectrum correction factor as$f_{Ei} = 1 - f_{pi}$, $I_0(E_i).f_{Ei}$ = the mean or average number of ion-pairs, $2\sqrt{I_0(E_i).f_{Ei}}$ = the standard deviation of the ion-pairs due to collisional loss that causes the pile up effects and fluctuations (excitations), the factor 2 is introduced to account for each ion-pairs created.

3.5 Determination of Efficiency

Efficiency is an important parameter of any given detector. The efficiency of a detector is the proportionality constant which relates the activity of the source being counted and the number of counts observed. The true activity of sources can be calculated by the efficiency of the detector. The efficiency of a detector changes with the physical change of counting system and the environment that surrounds it, therefore counting must keep constant throughout the experiment.

There are various kinds of efficiency, which are in common uses, some of which are: absolute efficiency, intrinsic efficiency, relative efficiency and full energy photo peak efficiency. Almost all of these efficiencies are significant parameter in practical spectrometry. Full energy photo peak efficiency is a basic parameter of the detector and is independent of the detector geometry. In the present work, we have calculated the peak efficiency of diamond wafer using the relation:

$$\varepsilon_p(E) = \frac{N_p(E)}{F(E)} \times 100\% \qquad (3.11)$$

where $N_p(E)$ is the number of counts in the full energy peak corresponding to energy $E(N_p(E))$ and $F(E)$ is the number of alpha particles with energy E emitted by the source.

The absolute efficiency of the CVD diamond detectors, defined as the number of counts recorded divided by the number of alpha particle emitted by the source was applied. It is thus given as:

$$\varepsilon_{abs} = \frac{no.of\ counts\ recoded}{no.of\ alpha\ particle\ emitted\ by\ source} \qquad (3.12)$$

It is a standard practice to use the Full-Energy Width at Half the Maximum peak count (FWHM) above the background as the measure of the photopeak width. This is proportional to the standard deviation of the distribution's average value (centroid). The energy resolution of a detector is a measure of its ability to resolve small differences in the energy of incident alpha particles. It is defined as:

$$R = \frac{FWHM}{H_0} \times 100\% \qquad (3.13)$$

where FWHM is the Full width of the full-energy peak at half maximum height expressed as the number of channels, and H_0 is the channel number corresponding to the peak centroid.

CHAPTER FOUR

4.0 RESULT AND DISCUSSION

4.1 Results

The figures 4.1 – 4.9 displayed the spectrum showing the variation in the experimental and corrected alpha counts/sec. with energy (KeV) for each of the detectors. Each of the figures consists of two plots, the experimental plot and the stripped plot after correction. The Detector Grade CVD diamonds (Fig. 4.1 to Fig. 4.4) spectra are observed to have a better and more pronounced peak value, while the Optical Grade CVD diamonds (Fig. 4.5 to Fig. 4.8) spectra are observed to be broad; each without a well defined peak value.

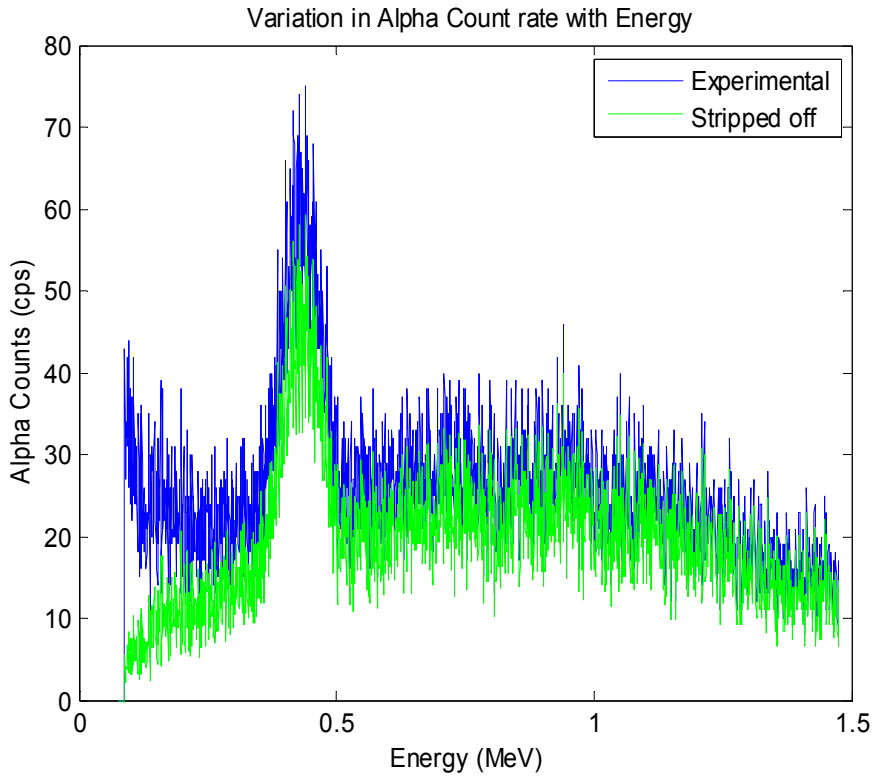


Figure 4.1: Variation of alpha count rate with energy for a Detector Grade CVD diamond (DG1)

4.2 Discussions

Table 4.1: - Spectral Analysis of the three broadly classified grades of CVD Diamonds

	Alpha Counts (cps)		Absolute Efficiency (%)		FWHM (KeV)		Average Distribution's Value (Centroid)		Spectrum Peak Resolution		Spectrum Peak Efficiency (%)	
Detector Type	EXP.	STRD.	EXP.	STRD.	EXP.	STRD.	EXP.	STRD.	EXP.	STRD.	EXP.	STRD.
DG1	26.06	19.43	0.053843	0.040145	110.3	42.1	447.5	437.85	0.24648	0.096152	80	
DG2	60.91	49.74	0.125847	0.102769	141.8	117.5	804.2	809.95	0.176324	0.145071	80	
DG3	334.7	294.3	0.691529	0.608058	10	8.5	1056	1066	0.009469	0.007973	82	
DG4	14.68	10.98	0.030331	0.022686	104.1	95.9	735.4	740.15	0.141565	0.129568	80	
OG1	64.59	46.41	0.13345	0.095888	275.8	148.6	355.4	405.6	0.776027	0.366371	60	
OG2	92.1	71.38	0.190289	0.147479	519.4	354.1	435.6	454.45	1.145062	0.779184	60	
OG3	152.6	125	0.315289	0.258264	650.6	606.2	596.7	691.3	1.09033	0.876899	60	
OG4	56.94	42.39	0.117645	0.087583	553.4	322.7	441.4	478.15	1.253738	0.674893	60	
SC	60.06	56.57	0.124091	0.11688	0.32	0.22	18.36	18.46	0.017429	0.011917	80	

DG represents Detector Grade wafers, **OG** represents Optical Grade wafers, **SC** represent Single Crystal wafer, **EXP.** represent Experimental Values and **STRD.** represent Stripped Values

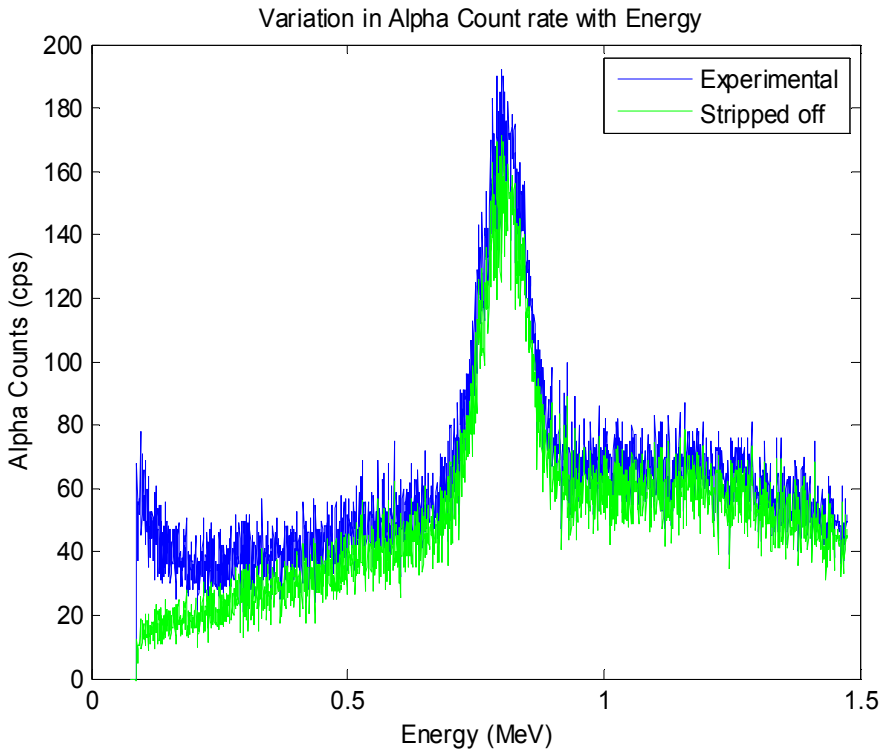


Figure 4.2: Variation of alpha count rate with energy for a Detector Grade CVD diamond (DG2)

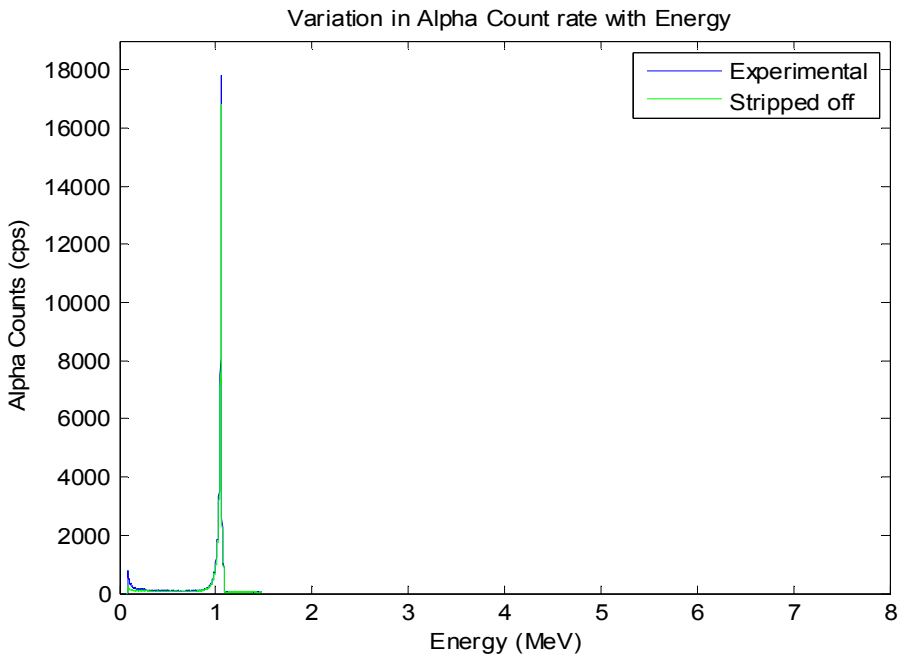


Figure 4.3: Variation of alpha count rate with energy for a Detector Grade CVD diamond (DG3)

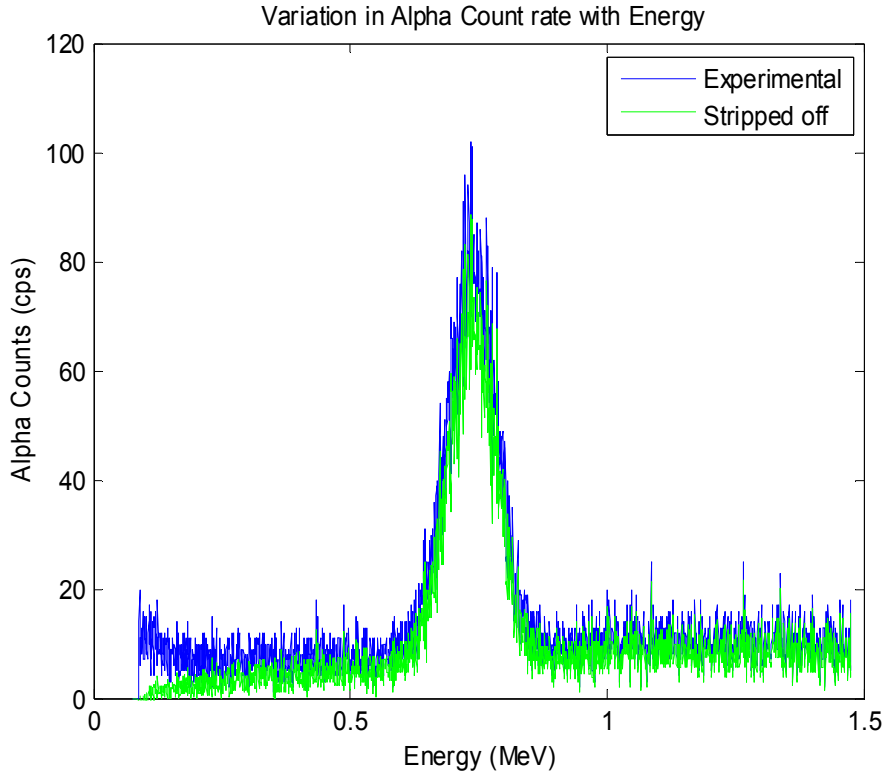


Figure 4.4: Variation of alpha count rate with energy for a Detector Grade CVD diamond (DG4)

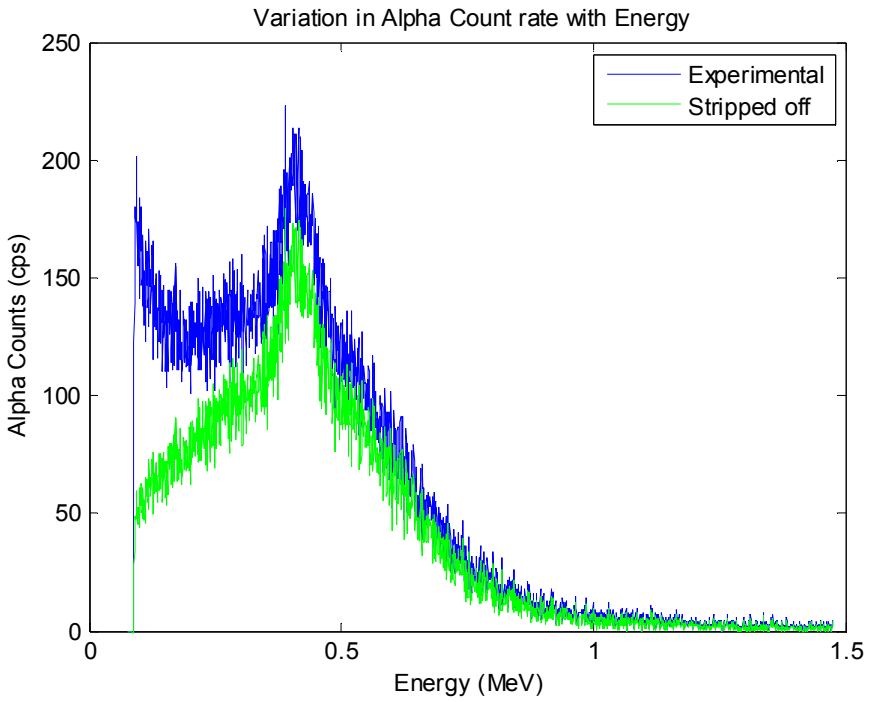


Figure 4.5: Variation of alpha count rate with energy for an Optical Grade CVD diamond (OG1)

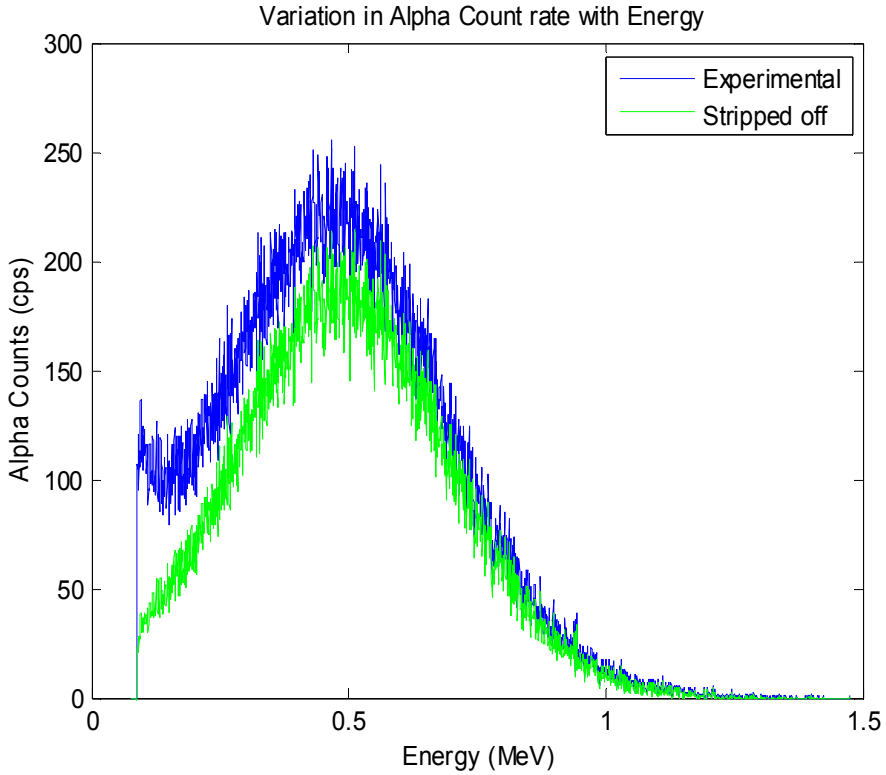


Figure 4.6: Variation of alpha count rate with energy for an Optical Grade CVD diamond (OG2)

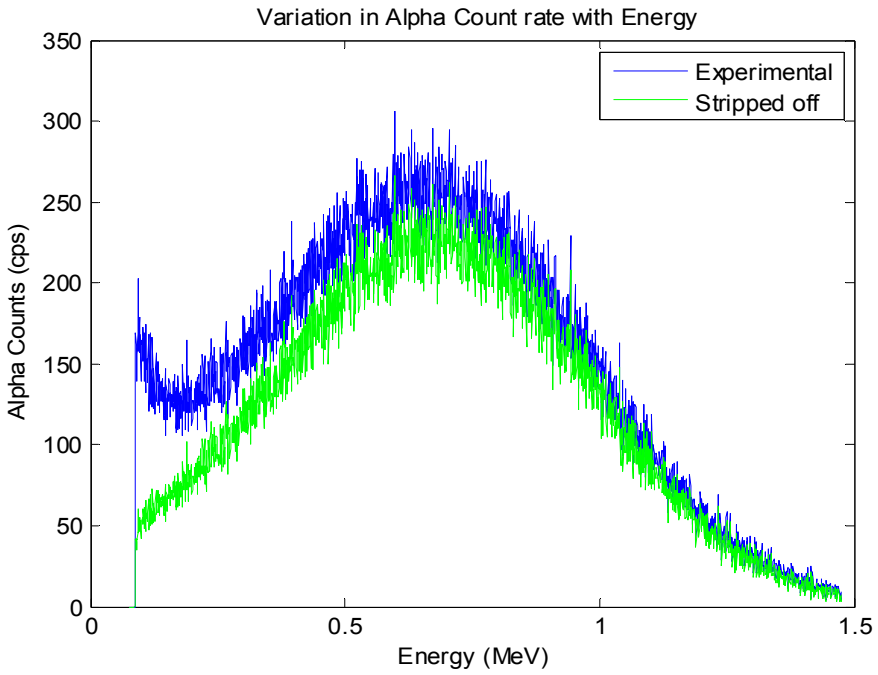


Figure 4.7: Variation of alpha count rate with energy for an Optical Grade CVD diamond (OG3)

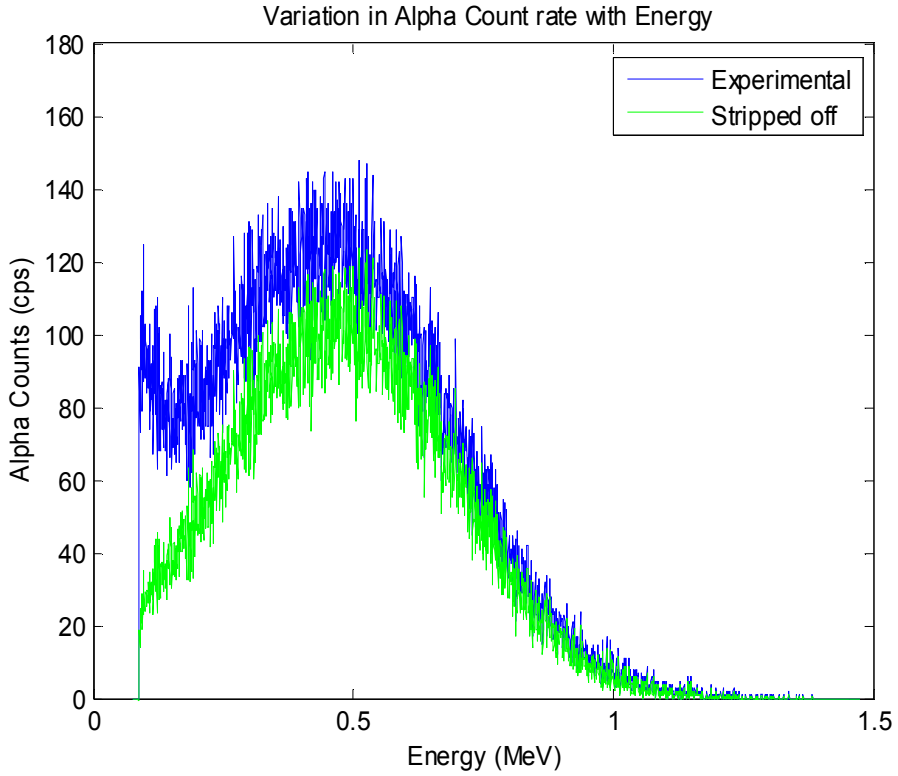


Figure 4.8: Variation of alpha count rate with energy for Optical Grade CVD diamond (OG4)

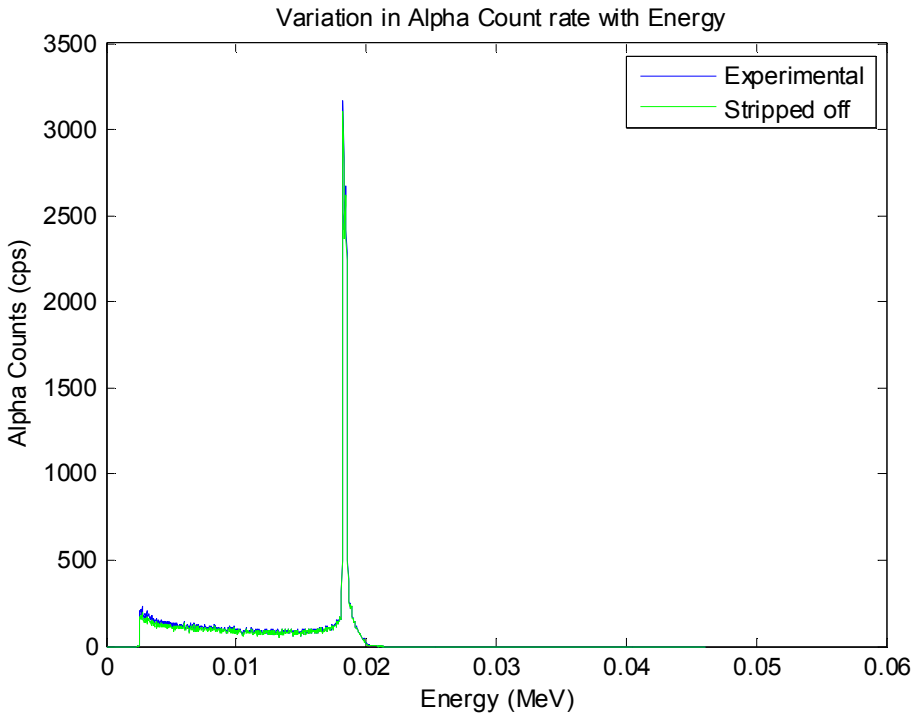


Figure 4.9: Variation of alpha counts rate with energy for a Single Crystal CVD diamond (SC)

The stripping methodology put forth made it possible to resolve fine details in the incident energies of the alpha-particle radiation and that has obviously improved the width of the spectra. Spectroscopically, the performance observed for the entire detectors illustrates two possibilities: a response around the mean spectrum height (good resolution) and a response with inferior performance (poor resolution). Both spectrum responses were centered at the same mean value (centroid), with the width of the spectrum in the poor performance case being greater. For instance, the spectrum width in blue for DG2 (Fig: 4.2) reflected the fact that a large amount of fluctuation was recorded from counts to counts even though the same energy was deposited in the detector for each event. Stripping off these fluctuations and defects/impurities made the spectrum width in green small. In comparison, DG3 (Fig: 4.3) become smaller and the peak approach a sharp spike after the removal of defects levels and fluctuations.

The influence of the pair produced counts that causes asymmetry in some of the spectra (OG2 (Fig: 4.6) and OG3 (Fig: 4.7) in particular) could be related to the defects levels of the wafer. Below 0.3MeV energy the asymmetric natures of the spectra were most pronounced with the detector and optical wafers; least affects the single crystal (Fig: 4.9) and DG3 wafers with least defect concentration type that could act as source of radiation losses.

Of note was the observation that the stripped alpha counts values with improved peak resolution has shifted from the alpha centroid for all the detectors. This perhaps owe to the improvement achieved in reducing the asymmetric nature of the diamond wafers. The measured average leakage current of 5.2pA for Single Crystal (SC); 21.5pA for DG; and 54.2pA for OG could be part of the reason for the observed higher noise (fluctuation) in the optical and detector grade polycrystalline. The fact that less of the noise was measured in the Single Crystal (Fig: 4.9) may be the reason for it better peak resolution and hence that of the Detector Grade (DG3) wafer (Fig: 4.3).

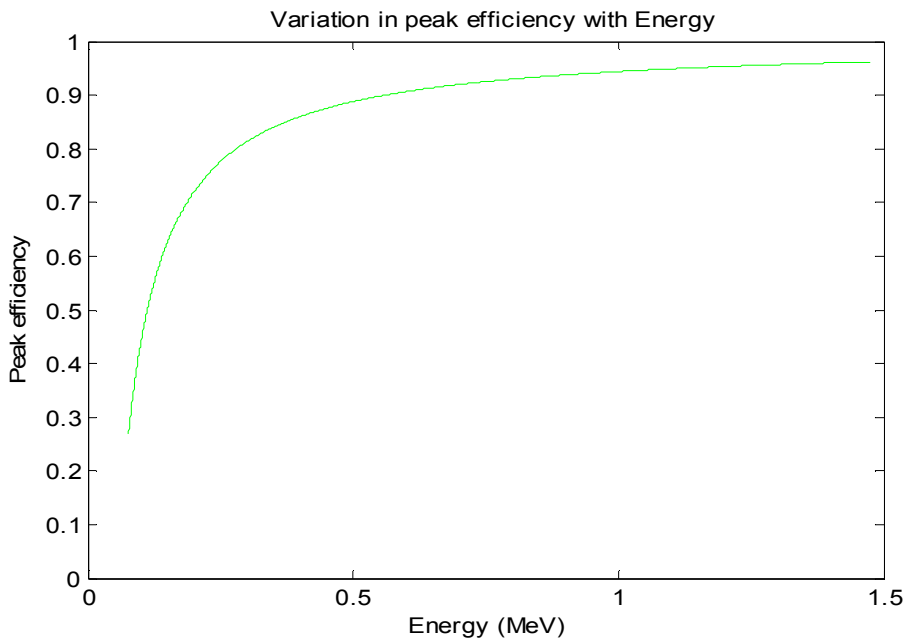


Figure 4.10: Variation of the CVD diamond detector peak efficiency with energy

Fig: 4.10 show a relationship between the detector's full energy peak efficiency with alpha particle energy; this relationship is the same for all the different CVD grades. The detector efficiency is seen to rise linearly with increase in energy between 0 and 0.3MeV due to the creation of electron-ion pair of the alpha particle interaction with the nucleus of the detector atoms that result in bremsstrahlung electromagnetic radiation. The effect saturates to a value of 1 and this is an indication that the detector peak efficiency is not affected within the saturation energy range but for collisional and other statistical losses.

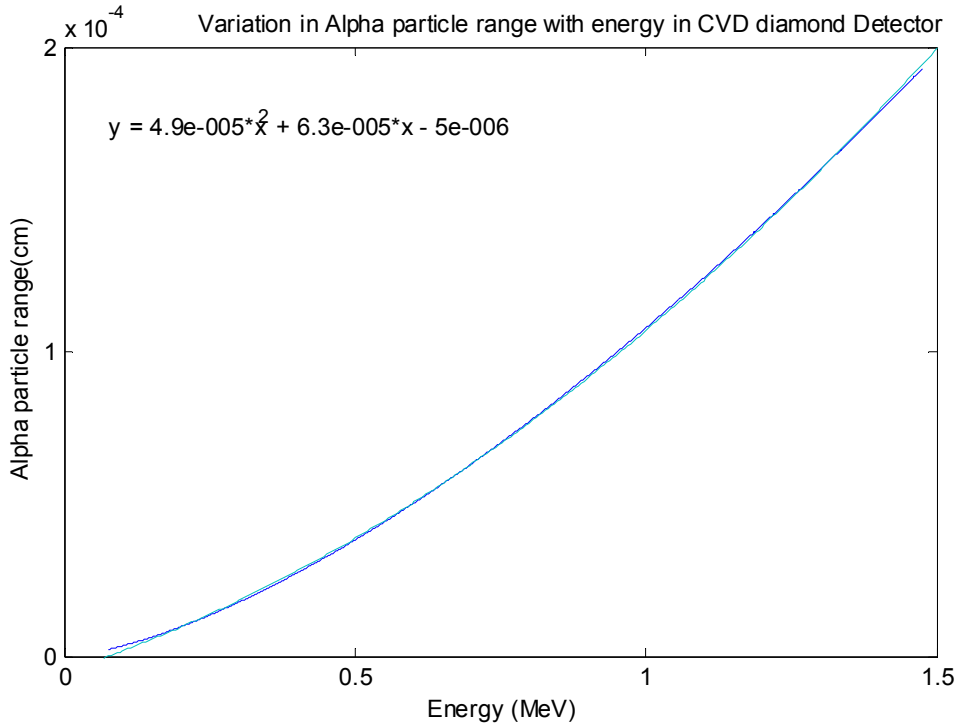


Figure 4.11: Variation in Alpha particle range in CVD diamond detector with energy

Fig: 4.11 show a range – energy relationship for alpha particle interaction with the CVD diamond. This was used to determine the range of alpha particle in each of the different CVD grades at different energies and to confirm that the interaction is a surface effect. For energy inputs up to 1.29MeV the polynomial equation obtained from the data points shows that alpha interact with the outer surface of the detectors (zero range).

Table 4.1 shows both the experimental and the stripped (analytical) analyzed data using the high performance language (MATLAB). The experimental spectrum Alpha Count rate was observed to be closely related to the stripped spectrum data but with slight differences (first two columns). The Detector Grade (DG) stripped data averagely maintained an absolute peak efficiency value of approximately 0.19% and the Single Crystal (SC) maintained an approximate value of 0.12%; the DG value slightly lower than the average experimental value of 0.23% and SC value remained the same. To the Optical Grade (OG) stripped data the average absolute peak efficiency value of approximately 0.15% as compared to the experimental average value of 0.19% were observed. A factor that possibly affects the changes observed could be due to the speed of analysis. This was attributed to the coding language (MATLAB) having lost track of data points during plotting. Be as it may, the technique put forth a good comparison of the CVD diamond detectors performance as alpha detectors.

More so, the table provides the spectrum peak resolutions and peak efficiencies for the experimental stripped data. These were obtained using the FWHM values acquired for both the experimental and stripped off spectrum generated by the High Performance Language (MATLAB). The Detector Grade and Optical Grade wafers indicate peak-efficiency performance values of 87% each, which in comparison with the Single Crystal wafer value of about 96%; while 80% each were the results observed experimentally for Detector and Single Crystal Grades and 60% for the Optical Grade. The Detector Grade stripped data averagely maintained a spectrum peak resolution value of 9.5% and the Single Crystal maintained an average value of 1.7%; the experimental values of 14.3% and 1.7% were

observed respectively. The Optical Grade stripped data have an average spectrum peak resolution value of 67% as compared to the experimental average value of 107%.

Again, to be specific, the Detector Grade wafers have an optimized spectrum resolution from 24.6% to 9.6% for DG1 (poorest peak) and 0.95 % for DG3 (best peak), with a close performance in the Single Crystal (SC) are observed. The stripped result of the Optical Grade wafers gave improved peak resolution values of between 78% (for OG2) to 37% (for OG1) as against the experimental results while the Detector Grade diamond was between 0.71% (for DG3) to 14.5% (for DG2). In comparison the peak resolution for the Single Crystal is about 1.2%.

CHAPTER FIVE

5.0 CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

The spectra obtained from the interactions of alpha particle with CVD diamond wafers were analyzed. The inherent effects due to defects/impurities present in the wafers were corrected for. A technique has been sorted for obtaining a spectrum independent of the detecting system.

In conclusion, the methodology adopted on each of the measured alpha spectrum had noticeable improvements on peak resolution, absolute efficiency and peak efficiency of the different CVD grades. The spectrum stripping analysis were observed to have influenced the broadness of the Optical Grade CVD detectors than on the narrower and better resolved spectra of the Detector Grade and Single Crystal. The improvement on the Optical Grade shows an average spectrum resolution value of 0.67 and this was consistent to the value of 0.7 reported in some work (Berdermann *et al.,* 2004 and Pomorski *et al.,* 2005). Thus, confirming that Optical Grade can be a good choice wafer for use in spectrometry. The optimized spectrum peak resolutions of the Detector Grade wafers (0.096 to 0.008), where the value 0.008 was lower than the 0.01 to 0.03 range values reported by Wang et al. (2005); confirming the possible choice of such material for alpha spectrometry. The comparative spectrum stripping analysis adopted has provided improved information of the actual performance characteristics of the different diamond grades when used for alpha sensing applications. For the polycrystalline wafer the observed differences in the experimental and stripped methods could be attributed to the high leakage current, giving rise to statistical fluctuation and noise. Generally the CVD diamond wafers were observed

on exposure to alpha particles with energy of 5.48MeV to have low leakage current (between 5.2pA and 54.2pA) and about 87% full energy peak efficiency for the Detector Grade and for the Optical Grade wafers each.

5.2 RECOMMENDATIONS

Based on the outcomes of the stripping analysis, this work makes the following recommendations:

- The chemical Vapor Deposition (CVD) technique used for the synthesis of diamonds should be further investigated to enhance the production of CVD polycrystalline diamonds with less or no presence of intrinsic defects/impurities and extrinsic contaminants or dopants, mostly prominently nitrogen, silicon, boron and phosphorus, as well as different structural morphologies.
- Faulty circuitry, too sensitive gain setting, high humidity and variable line voltage or transient of the detecting system should be checked before carrying out the experiment in order to avoid background and/or statistical fluctuations.
- Software that can code all the probable interactions of alpha particle with CVD diamond grades shall go a long way in bringing out completely the true and exact information needed for the much talked about low energy Physics diamond detectors.

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APPENDIX I

Stripping program

```
% Date 3rd July 2012
% Declaration of constants to be used

W = 1.28eV;
E = DG1(:,1)/1000MeV;
Io = DG1(:,2)cps;
rhod = 3.54/1000Kg/cm^3;
rhoa = 1.293e-3/1000Kg/cm^3;
Ad = 12.0107/1000Kg/mol;
Aa = 14.6/1000Kg/mol;

% Specific ionization due to the interaction of alpha particle with diamond

Ra = 0.325*(E.^(3/2));
Rd = Ra*(rhoa/rhod)*sqrt(Ad/Aa)
I = E/(Rd.*W);
I = I(:,2048)

% Energy that have contributed to the ion produced charges

Ec = (2.*E.^2)./(2.*E) + 1022

% Specific ionization associated with the radiation and collisional losses

e = Ec-E
Rc = 0.325*(e.^(3/2))
Rdc = Rc*(rhoa/rhod)*sqrt(Ad/Aa)
Ic = e./(W*Rdc)

% the spectrum correction factor of the detector

fp = (I-Ic)./I
fe = 1-fp

% the correction of specific ionization that takes into account all the ion pairs produced either by collisional, radiation losses and excitation

b = ((Io.*fe)+(2.*sqrt(Io.*fe)))
Ip = Io-b

% Plot of the experimental and stripped counts

plot(E,Io,'b-');hold on
plot(E,Ip,'r-');grid on;
title('Variation in Alpha count rate with Energy')
xlabel('Energy (KeV)')
ylabel('Alpha Counts (cps)')
legend ('experimental','stripped')
```