

# Scientific Review on theory and Mechanism of Neutron Activation Analysis in Geological and Environmental Samples

<sup>1</sup>BUHARI SAMAILA, <sup>2</sup>SHEHU ALHASSAN ALHJI

<sup>1</sup>Department of physics with electronics, Federal University Birnin Kebbi P.M.B 1157

<sup>2</sup>Department of Physics, Waziri Umaru Federal Polytechnic Birnin Kebbi, P.M.B 1034  
Kebbi, Nigeria.

Corresponding Author: [kawara002@gmail.com](mailto:kawara002@gmail.com)

Co-author: [Alhassanalhajis@gmail.com](mailto:Alhassanalhajis@gmail.com).

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**Abstract:** This study described the theory and mechanism for the simultaneous determination of major, minor and trace elements involved in Neutron Activation Analysis, applied to geological and environmental samples as a sensitive and non-destructive tool. Using Neutron Source, the gamma-rays emitted due to neutron capture have been applied for the investigation of the elemental constituents and concentrations of major, minor and trace elements in such samples. Neutron Activation Analysis is of increasing importance in the field of element characterization in environmental and geological samples for a number of reasons that were documented and reviewed by several scientific researchers. NAA serve in the local industry and economy in each regional area of the country. It presents a sensitive multi-element technique which is valuable for major, minor and trace element analysis due to its precision and accuracy. The basic theory and detailed information on how NAA works in the analysis of above samples was presented in this paper.

**Keywords:** mechanism, Neutron Activation Analysis, geological and environmental samples.

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## 1. INTRODUCTION

Neutron Activation Analysis (NAA) is an analytical technique based on the measurement of characteristic radiation from radionuclide formed directly or indirectly by neutron irradiation of the material of interest (Asres Yihunie Hibstie 2012). The major objective of this work is to describe the theory and mechanism in identify the various major, minor and trace elements present in the geological and environmental samples. Neutron Activation Analysis (NAA) was first proposed in 1936 by George Charles de Hevesy when he found that materials containing rare metals became radioactive after being exposed to neutron sources. De Hevesy then began to implement this technique to produce radioisotopes as medical biological tracers. Since then, the NAA has been used for a wide variety of applications. In archeology, NAA is frequently used to fingerprint the artifacts in verifying their authenticity and track their origins. One example illustrating the effectiveness of the NAA in fingerprinting the archeological samples is found in 2009, at the University of Mainz in Germany where the group of scientists irradiated 20 samples from reverse glass paintings to identify the place and date of origin. Through both, long term and short term irradiations, in a TRIGA Mark II reactor, isotopes from 23 elements were revealed which were used to characterize the glass matrix composition. The glass compositions were compared to other glasses from known locations and periods in validating their origins (Todd H, 2012). NAA is often applied to criminal forensics. For example, the death of Napoleon Bonaparte was investigated using the NAA to determine if he died from arsenic poisoning. Hairs, which were taken immediately after his death, were exposed to neutrons and analyzed through the NAA to determine the arsenic concentrations. The results indeed verified that Napoleon had received significant doses of arsenic prior to his death.

NAA is also used to investigate bullets and gunshot residues at crime scenes. For example, in 1964 NAA was used to investigate the assassination of President John F. Kennedy. Bullet fragments recovered from the crime scene as well as

the rifle of Lee Harvey Oswald were irradiated in a reactor at the Oakridge National Laboratory to analyze if the compositions matched (Guinn, 1979). The NAA report was included in the official investigation report issued by the Warren Commission, which confidently identified Oswald as the President's assassin (Todd H, 2012). With the development of high-power neutron source, precise and fast neutron detectors and sophisticated electronics, NAA has achieved ultralow minimum detection limit (MDL). As a result the method has turned out to be one of the best choices for quantification of trace elements in a wide variety of samples such as geological, archaeological, biological and environmental samples (Maitreyee Nandy 2018). Different forms of NAA offer good choice over other techniques for application in geological and environmental samples. NAA is used for quantitative analysis of trace elements in the samples.

### Neutron Source

There are two commonly source of neutrons, these include: isotopic source and reactor source. The most practical neutron source for NAA is a nuclear reactor, which produces neutrons via the nuclear fission process. Reactors are large devices in which fissionable material such as  $^{235}\text{U}$  is fissioned into two nuclides with simultaneous emission of neutrons that induce further fissions in a chain reaction. The cross-section for neutron induced fission of fissionable nuclides increases with decreasing energy of the neutrons. In order to increase the neutron activity, moderators that slow down neutrons are added to reactors. To reflect back some of the neutrons that are leaked from the reactor core, reflectors are used. The fission process releases large amounts of energy so the system must be cooled by a coolant either liquid or gas (Esam Abd-Elhady M. H 2012).

### Types of Neutron Activation Analysis

NAA is the most common form of activation analysis. The activation reaction is induced by the interaction of a neutron with the nucleus of an analyte element. Depending on the energy of the incident neutron and the reaction cross-sections of the target elements, different types of reactions can take place, leading to activation products (Esam Abd-Elhady M. H 2012).

#### *Thermal Neutron Activation Analysis (TNAA)*

Thermal Neutron Activation Analysis (TNAA) is carried out using thermal neutrons. They are obtained after good moderation of neutrons from any neutron source. Due to the  $1/v$  - dependence of the capture cross section, thermal neutrons have the greatest probability of interacting with most target elements. For this reason, TNAA is the most simple and universal activation analysis technique and remains the workhorse in the activation analysis laboratory (Esam Abd-Elhady M. H 2012)

#### *Epithermal Neutron Activation Analysis (ENAA)*

Epithermal neutrons ( $\sim 0.5$  eV to  $\sim 0.5$  MeV) serve as activation particles in this special type of NAA, which is based on the selective activation of certain nuclides the cross sections of which exhibit strong individual resonances in the lower part of this energy range. Special facilities to enhance ENAA reactions over thermal NAA are employed in the irradiation locations. To suppress the thermal neutron activation of the matrix elements, the sample is covered with a thermal neutron absorber, such as cadmium (Cd) or boron. As a result, high sensitivities can be attained despite the relatively low activity induced by the epithermal neutrons (Esam Abd-Elhady M. H 2012).

#### *Fast Neutron Activation Analysis (FNAA)*

Neutrons with energies greater than  $\sim 0.5$  MeV are called fast neutrons. The fission spectrum of a light-water-moderated reactor provides as many fast neutrons as thermal neutrons. Therefore, fast neutron activation of certain elements via (n, p) reactions is a very selective technique, complementary to thermal and epithermal NAA. A typical example is Fe, for which the  $^{56}\text{Fe} (n, p)^{56}\text{Mn}$  activation reaction produces a better gamma-ray emitter than the thermal capture reaction. A common and particularly useful form of (FNAA) is the instrumental analysis with 14 MeV neutrons that are produced by small accelerators known as neutron generators (Esam Abd-Elhady M. H 2012).

#### *Prompt Gamma Neutron Activation Analysis (PGNAA)*

Prompt gamma neutron activation analysis (PGNAA) is an instrumental procedure that makes use of the prompt gamma-ray emissions that occur immediately after neutron capture during the de-excitation of the newly formed compound nucleus. The capture can produce nuclear states with energies up to about 1 MeV above the ground state, which usually

decay through a cascade of gamma rays. The complexity and broad energy range of these spectra distinguishes PGNA from most other NAA procedures. The main advantage of PGNA is the ability to obtain gamma-ray spectra from the neutron capture of nuclides that do not produce radioactive isotopes through the  $(n, \gamma)$  reaction. PGNA favorably complements NAA through efficient reactions with the light elements that generally do not produce radio nuclides, and also a number of higher Z elements with high cross sections the normal decay products of which may be difficult to measure in the presence of high activities induced in certain matrices. The light elements most often investigated are H, B, C, N, Si, P, S, and Cl; and the heavier elements are Cd, Sm, and Gd.

Other types of NAA include short time neutron activation analysis (STNAA) and radiochemical neutron activation analysis (RNAA). In STNAA, the gamma ray measurements are performed after short irradiation and decay times to determine elemental contents of isotopes characterized with short or medium half life times. RNAA is used in the case where the induced radio nuclides of trace elements are masked by matrix activity. The resulting radioactive sample is chemically decomposed and the radionuclides are isolated by chemical separations into a single or several fractions free of interfering radioactivities. Although RNAA has the advantage of high sensitivities for many elements, it has some disadvantages: it is destructive, time consuming and the radiochemical procedures are complicated (Esam Abd-Elhady M. H 2012).

## 2. METHODOLOGY

### *Geological sample preparation*

The geological sample should be dried, grounded and sieved with a 500  $\mu\text{m}$  mesh or any other sieving equipment and homogenized. The small particles size then prepared and wrapped in thin aluminum/polythene foils with other binding agents, according to the number required to be irradiated. The mass of each wrapped sample will be determined using electronic weighing balance. The sample preparation is based on short, medium and long irradiation period. For the short irradiations, each of the wrapped analyte samples will be put into a separate polyethylene capsule and heat-sealed using a hot soldering iron. For the medium and long irradiations the individually wrapped analyte samples were put into two polyethylene capsules, that is, in each capsule and also heat-sealed. The difference in irradiation methodology is to account for differences in the half-lives of possible radio nuclei, which will be produced by the irradiation. A standard reference material, from the centre will also prepare and analyzed to evaluate the accuracy of the analytical technique. The analyte samples of the standard reference material (SRM) each weighed and wrapped in thin polyethylene foil. Each sample of SRM materials was put into a separate polyethylene capsule and heat-sealed, for short, medium and long irradiation (Baafuo Osei, 2002).

### *Soil/rocks samples collection and preparation*

The geological samples (Soil/rock) should be collected with a pair of gloves, if it is the soil samples should be collected with hand auger and plastic shovel and transferred into pre-cleaned polyethylene bags. After taking each sample, the plastic shovel and the auger were washed with clean water and the hand gloves replaced to avoid contaminations. Great care must be taken into account not to introduce contaminants that affect the samples to be irradiated such as sodium, chlorine and others. The same procedure should be followed when collecting the rock samples. All the samples would be doubly bagged and boxed for safe transport from the field to the laboratory. At the laboratory, the soil/or rock samples brought from the field would be spread on pre-cleaned polyethylene sheets for <gloved-hand-picking> of stones and plant materials. Then the samples are stored in pre-cleaned polyethylene capped bottles and handled with extreme care until the moisture is completely removed from the samples' surface (if any). Just after removing the stones and vegetation, samples are dried up at room temperature, sieved, placed in the plastic container and left for a certain days to remove moisture and water content. Dried samples are pulverized into powder form, homogenized, weighed and placed in a plastic container for measurement. The powdered samples would be re-grounded again using local grinding machine, homogenized and packed in plastic containers, weighed, wrapped in polyethylene films. Finally, a refinement will be done using portable mortar with pestle and then the homogenized samples are weighed, packed and sealed tightly with cup-holder. The empty sample containers were first washed with tap water, dried, weighed and filled with soil/rock samples and weighed again. To do such activities clean instruments such as knife, file, tweezers, etc., and clean plastic gloves are needed. The containers were closed by plastic tapes wrapped over the caps. The Sample procedures will be applied for the reference materials. The samples and standards are irradiated to certain period of time to achieve secular equilibrium. Samples and standards are bundled and sandwiched together and irradiated at a thermal flux. Because of the low detection limits for many elements, sample collection and preparation is critical. Virtually any solid material can be analyzed by this method,

but there are geometric constraints: the sample must fit into the irradiation vials and should present a consistent geometry. Before selecting material for analysis, consideration should be given to the geometrical factors (Asres Y. H. 2012).

#### *Environmental Sample preparation*

The samples, such as sediments, biological tissue, air filters and sea and fresh water salts are normally weighed and heat sealed in cleaned polyethylene, polypropylene or high purity quartz vials ranging in size from 1 cc to 10 cc. Sample size can range from a few milligrams to tens of grams depending upon the sensitivity required and sample size available. Desiccation of sediments and biological tissue prior to encapsulation is accomplished by either freeze-drying or oven drying at 105°C. Special precautions are necessary when drying samples for trace Hg analyses to avoid losses by volatilization. Natural waters, especially seawater, must usually be evaporated to dryness if the samples are to be irradiated for more than several minutes, since gaseous radiolysis products (H<sub>2</sub>, O<sub>2</sub> and other gases) are generated and produce high pressures which rupture the plastic irradiation vials. Problems due to pressure build-up are dependent upon the length of the irradiation, and almost any kind of sample can be irradiated for a few minutes. Desiccation can be accomplished by either freeze drying or slowly evaporating the water under an infrared heat lamp. Again, great care must be taken not to volatilize Hg and other volatile elements during these drying operations. Water samples can be neutron irradiated directly, without evaporation, if the water is sealed in quartz vials. However, in the case of seawater, the radiolysis products build up dangerously high pressures which have even shattered sealed quartz vials and this method is not recommended for routine use. In addition, quartz contains much higher levels of impurities than polyethylene or polypropylene and is a potential source of contamination. Tanner et al. (1972), and Brune and Landstrom (1966) have described a method for neutron irradiating 30 to 80 ml samples of fresh water in a frozen state in polyethylene containers as a means of preserving the samples prior to and during the neutron irradiation. It is important that samples in a given set be encapsulated in reproducible, identical geometries so that all samples will be neutron irradiated in a known orientation (Robertson D *et al*, 1974). Once prepared, the samples are submitted to the following methods, depending on the objective:

#### *Instrumental neutron activation analysis*

This method is the application of the neutron activation technique without requiring any chemical process neither during preparation nor during the analysis

#### *Comparative instrumental neutron activation analysis*

In this method, standards of the element or elements to be analyzed are included in each group of samples to be studied and the elemental concentrations are calculated by linear regression, based on standard measurements (Dovenir F *et al*, 2009).

#### *Parametric methods - k<sub>0</sub>-instrumental neutron activation analysis*

This method is an alternative method used when one is introduced to the NAA method because it is more didactic for beginners or when the geometry of the samples is not punctual. This method uses sodium as comparator, the values for  $f$  and  $\alpha$  are average values for the Carousel Facility, each sample is irradiated in one irradiation channel and all samples and comparators are irradiated simultaneously (Dovenir F *et al*, 2009)

#### *Delayed fission neutrons analysis*

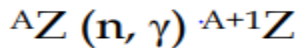
The Delayed Fission Neutron Activation method applied is a specific method for uranium analysis when the samples are irradiated with thermal neutrons. A pneumatic transfer system used for irradiation is located outside the core where the contribution of the thermal neutron flux to the total neutron flux is higher than in the carousel facility. The system irradiates measures and calculates the results automatically. The final result is calculated by linear regression. Others include: *Chemical Neutron Activation Analysis, Radiochemical Neutron Activation Analysis*

*Radiochemical and Chemical Neutron Activation Analysis*, (Dovenir F *et al*, 2009).

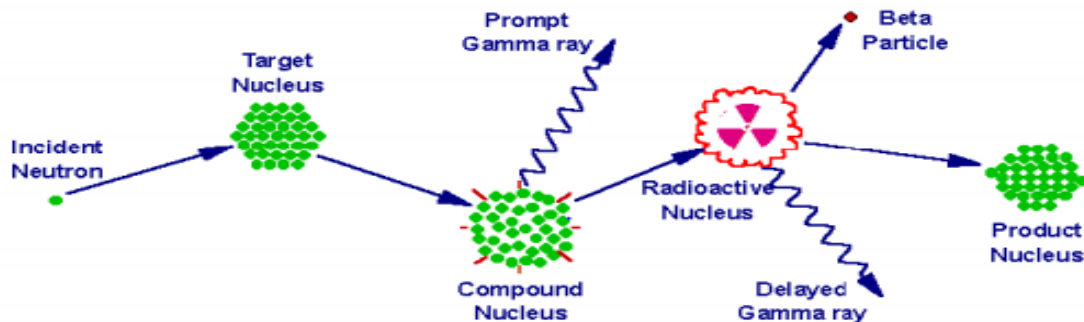
#### **Sample Irradiation**

In typical NAA, stable nuclides (<sup>A</sup>Z, the target nucleus) sample undergo neutron capture reactions in a flux of (incident) neutrons. The radioactive nuclides (<sup>A+1</sup>Z, the compound nucleus) produced in this activation process usually decay by emission of a beta particle (β<sup>-</sup>) and gamma ray(s) with a unique half-life. A high-resolution gamma-ray spectrometer is used to detect these 'delayed' gamma rays in the presence of the artificially induced radioactivity in the sample for both

qualitative and quantitative analysis. The sequence of events that occur during the most common type of nuclear reaction used for activation analysis is shown in Fig. 2. The incident neutron hits the target nucleus, which captures the neutron and is converted into a compound nucleus. The latter immediately emits radiation called prompt gamma radiation and forms the radionuclide, which then kicks out a beta particle and emits the delayed gamma radiation (since it is emitted after some time delay), forming the product nucleus (David T 2004).



Where  $n$  is a neutron,  $\gamma$  is a gamma photon, and  $A$  is the atomic mass. And also by comparing the specific activities induced in the standards and unknowns provides the basis for computation of elemental abundances (Lianne R. Lobo, 2013).



**Fig .1: Diagram Illustrating the Process of Neutron Capture by a Target Nucleus Followed By the Emission of Gamma Rays (Borut Smodiš, 2015). If NAA is conducted directly on irradiated samples it is termed Instrumental Neutron Activation Analysis (MAMUNU, MUHAMMADU INUW 2015).**

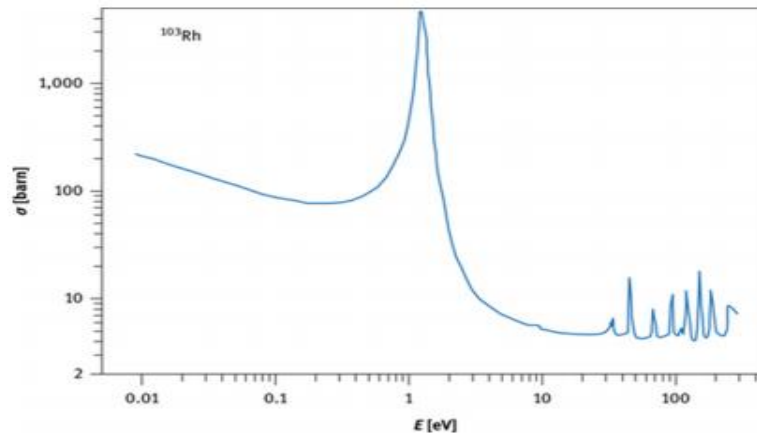
**Principle of Neutron Activation Analysis**

An atomic nucleus that is hit by a neutron ( $n$ ) may undergo a variety of nuclear reactions, depending upon the energy of the neutron. At low-to-intermediate kinetic energies, below about one million electron volts (MeV),  $(n, \gamma)$  reactions prevail, meaning that the neutron is captured by the nucleus which then emits a  $\gamma$ -ray. Higher kinetic energies of the neutron cause reactions such as  $(n, p)$ , by which the neutron is captured and a proton  $p$  is emitted, or  $(n, \alpha)$ , which indicates that an  $\alpha$ -particle (a helium nucleus) is emitted. As neutrons do not carry an electric charge, they experience no repulsion by an atomic nucleus. For this reason, neutrons with very small kinetic energies can be captured by a nucleus. In fact, neutrons, thermal are most commonly utilized in NAA. Thermal neutrons carry kinetic energies less than about 0.1 eV ( $1\text{eV} = 1.602 \times 10^{-19} \text{ J}$ ). Using the relation  $E = k_B \times T$ , where  $k_B$  is Boltzmann’s constant ( $8.617 \times 10^{-5} \text{ eV K}^{-1}$ ), it was calculated that a temperature of 300 K corresponds to an energy of just 0.026 eV. If a target which contains atomic nuclei of kind  $X$  is exposed to a beam of neutrons (or other particles), the number  $N$  of nuclei  $Y$ , formed by a reaction, can be calculated:

$$\frac{dN_Y}{dt} = \sigma \times \Phi \times N_X \dots\dots\dots I$$

where  $dN_Y/dt$  is the production rate of  $Y$ ,  $\sigma$  is the cross section, neutron ( $\text{cm}^2$ ),  $\Phi$  is the neutron flux ( $\text{cm}^{-2} \text{ s}^{-1}$ ), assumed to remain constant across the target, and  $N_X$  the number of nuclei  $X$ , also assumed to remain essentially constant. The cross section  $\sigma$  indicates the probability of the reaction to occur. It is commonly associated with the apparent cross section of nucleus  $A$  for a neutron that approaches the nucleus. Historically, the word “barn, neutron cross section” has been coined as the unit for the cross section where 1 barn (b) =  $10^{-24} \text{ cm}^2$ ; 1 b corresponds approximately to the cross section of a uranium nucleus. For thermal neutrons,  $\sigma$  decreases approximately as a function of  $1/v$ , where  $v$  is the speed of the neutron. The slower the neutron is, the larger is the probability that it is captured by a nucleus. Above the thermal energy range, there is the range of epithermal energies up to about 100 eV. This range is characterized by sharp increases in  $\sigma$  at well-defined energies that correspond to resonances, the formation of a compound nucleus in discrete excited states (Figure 2).





**Figure 2: Dependence of the neutron-absorption cross section  $\sigma$  upon the neutron energy displaying the decrease of  $\sigma$  with neutron energy for thermal neutrons and neutron capture resonances at epithermal energies (Heinz-Günter Stosch 2016).**

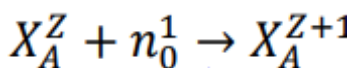
Neutron capture of an atomic nucleus results in the formation of an isotope of the same element in an excited state. A prompt  $\gamma$ -ray is released immediately and carries away part or all of the excitation energy, depending upon the existence and stability of excitation levels. Prompt  $\gamma$ -ray NAA is one of the methods of neutron activation. It requires the detector to be set up close to the neutron source, for example, a reactor. It may be the method of choice if only stable isotopes of an element are produced during neutron irradiation or if only radioactive isotopes with very short half-lives are generated. In many cases, Y is a radioactive isotope that decays with its characteristic half-life  $t_{1/2}$  into a stable isotope or into another radioactive isotope of a different element. The  $\beta^-$  decay results in the formation of an element with a higher atomic number  $Z + 1$  by emission of an electron or to an isotope of an element with a lower atomic number  $Z - 1$  by the nucleus capturing an orbiting electron, more rarely by emission of a positron from the nucleus ( $\beta^+$  decay). Half-life and decay constant  $\lambda$  are related by

$$t_{1/2} = \frac{\ln(2)}{\lambda} \dots\dots\dots \text{II}$$

At least part of the decays by  $\beta^-$ ,  $\beta^+$  or electron capture results in the formation of a nucleus in an excited state. The excitation energy is immediately released by emission of one or more  $\gamma$ -rays with characteristic energies. These  $\gamma$ -rays are registered and evaluated in NAA (Heinz-Günter Stosch 2016).

**Neutron Activation Analysis Theory and Physics**

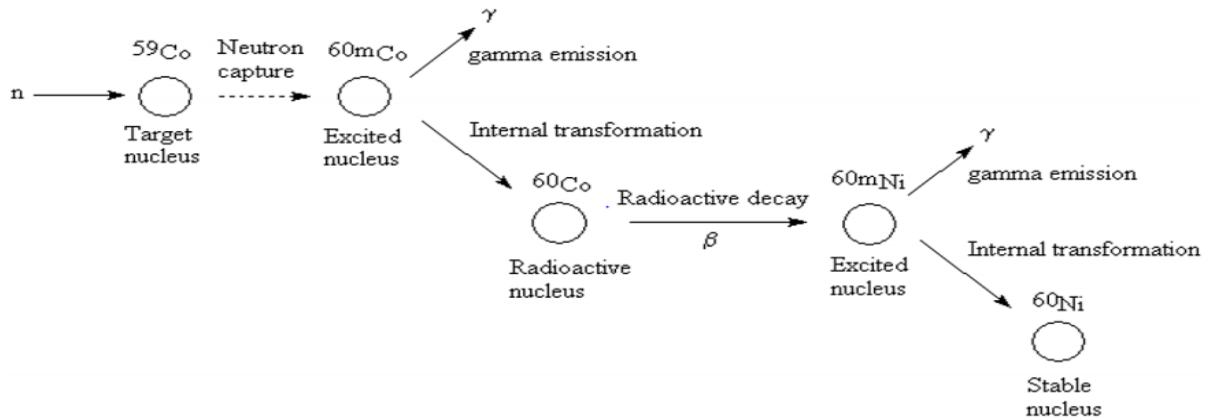
The basic idea of neutron activation analysis is that irradiation by neutrons induces nuclear reactions in the nuclei of the analyte, and the reaction products (which usually are radioactive) emit radiations characteristic of the identity and quantity of the elements to be determined. Among other possibilities of irradiation by charged particles or gamma rays, irradiation by neutrons is the best choice since they have no charge and can be deeper penetrates into the investigated sample (Esam Abd-Elhady Mohamed Hammad 2012). Neutrons interacting with a nucleus of an atom may either be scattered or captured to form a compound nucleus. One of these interactions is neutron capture, in which a neutron is absorbed by the nucleus. The capture thus increases the mass number of the nucleus:



This newly generated isotope is usually unstable. Neutron activation converts nuclide into new isotope. The ability of a nucleus to capture a neutron depends upon the nuclear properties of the nucleus and probability of neutron capture. It is completely independent of all chemical, electrochemical and magnetic properties of the nucleus, which makes NAA virtually independent of chemical and electromagnetic properties of the examined sample.

Many nuclides, upon capturing a neutron, become unstable and undergo a nuclear transmutation and or radioactive decay in becoming stable. Such radioactive decays can involve one or more of the following modes: beta emission, positron

emission, electron capture and/or isomeric transition. Each of these events can be accompanied with the emission of one or more gamma photons, as illustrated in **Figure 3**. These high energy photons can be detected and counted using gamma spectroscopy detectors.



**Figure 3: Neutron capture by Co-59 and radioactive decay of the former Co-60**

Decaying nuclides emit signature gamma photons, which are used to identify the isotope. Most of the activated isotopes during NAA produce gamma rays as they decay, which makes them easy to detect. Nuclear events such as radioactive decay or neutron interactions are characterized by the probability of occurrence. Measured nuclear properties such as half life and cross section determine the probability of whether an event will occur over time. Equation 2, defines the probability of decay for a single nuclide called decay constant ( $\lambda$ ):

$$\lambda = \frac{\ln 2}{t_{\frac{1}{2}}} \dots\dots\dots 2$$

$t_{\frac{1}{2}}$ : half life (sec)       $\lambda$ : decay constant

The product of the decay constant and the number of radioactive nuclei of a given isotope, gives the total number of probable decays per second, which is the activity of that nuclide, and expressed in the unit of Becquerel's (Bq) is equal to:

$$A = \lambda n \dots\dots\dots 3$$

$A$ : total activity (Bq)       $n$ : number of atoms (atoms)

$$\frac{dn}{dt} = -\lambda n \quad A(t) = A_0 e^{-\lambda t} \dots\dots\dots 4$$

$t$ : decay time (sec)

The probability for a neutron to interact with a nucleus is expressed as microscopic cross section ( $\sigma$ ). This property is represented as a cross sectional area measured in barns. One barn is equal to  $10^{-24} \text{ cm}^2$ . Smaller cross section units is defined as an outhouse ( $10^{-30} \text{ cm}^2$ ) and a shed ( $10^{-48} \text{ cm}^2$ ); these units are rarely used (The Language of the Nucleus). An integrated cross section over a number of nuclei (N) in a target per unit volume is defined as macroscopic cross section ( $\Sigma$ ):

$$\Sigma = N\sigma \dots\dots\dots 5$$

$\Sigma$ : macroscopic cross section  $\left(\frac{1}{\text{cm}}\right)$        $N$ : atom density  $\left(\frac{\text{atoms}}{\text{cm}^3}\right)$

$\sigma$ : microscopic cross section  $\left(\frac{\text{cm}^2}{\text{atom}}\right)$

Neutron cross section is not a constant value, but a variable dependent on the speed (or energy) of the interacting neutron and on the nuclear properties of the target material. The neutron capture cross section tends to increase as the neutron energy decreases. Therefore, the probability that a nucleus will capture a slower neutron is usually higher than the probability of capturing a fast neutron. However, there are exceptions, especially in the resonance region where large cross sections for capture can be observed (Todd Hansen Sherman 2012)

$$A_D(t_{decay}) = \phi n \sigma (1 - e^{-\lambda_D t_{irr}}) \dots\dots\dots 6$$

$A_D(t_{decay})$ : activity of activated daughter isotope (Bq)

$\lambda_D$ : daughter decay constant  $\left(\frac{1}{sec}\right)$

Equation 5 and 6 are assuming the mono-energetic neutrons. More accurate activation equation taking into account a range of neutron energies is obtained by integrating over all the energies:

$$R = n \int_0^{\infty} \phi(E) \sigma_{absorption}(E) dE \dots\dots\dots 7$$

$R$ : reaction rate  $\left(\frac{\text{neutron interactions}}{sec}\right)$

$$A_D(t_{irr}) = (1 - e^{-\lambda_D t_{irr}}) \sum_{i=0}^N n_i \phi_i \sigma_{i,absorption} \quad \text{or}$$

$$A_D(t_{irr}) = n_P (1 - e^{-\lambda_D t_{irr}}) \int_0^{\infty} \phi \sigma_{absorption} dE \dots\dots\dots 8$$

$n_P$ : number of parent atoms (atoms)

Combining Equation 3 and 6 the activity of the remaining generated daughter isotopes after irradiation and decay is obtained as follows:

$$A_D(t_{decay}) = \phi n \sigma (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}} \dots\dots\dots 9$$

The absorption cross sections ( $\sigma$ ) are affected by the temperature of the sample material; therefore

$$\sigma(T) = \frac{\sqrt{\pi}}{2} \sigma \sqrt{\frac{293}{T}} \dots\dots\dots$$

$\sigma$ : microscopic cross section at 293 K  $\left(\frac{cm^2}{atom}\right)$   $T$ : temperature (K)

The number of parent atoms can be calculated from:

$$n_P = m \frac{N_A}{A_m} A_{\%} \dots\dots\dots 11$$

$m$ : mass (g)

$N_A$ : Avogadro's number  $\left(\frac{\text{atoms}}{\text{mole}}\right)$   $A_m$ : atomic mass  $\left(\frac{g}{\text{mole}}\right)$

$A_{\%}$ : atomic abundance ratio



The modified NAA equation containing all the variables based on the “one group” activation theory is:

$$A_D(t_{decay}) = \phi \left[ \frac{\sqrt{\pi}}{2} \sigma_P \sqrt{\frac{293}{T}} \right] m \frac{N_A}{A_m} A_{\%} (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}} \dots\dots\dots 12$$

$\sigma_P$ : parent microscopic absorption cross section at 293 K ( $cm^2$ )

If the activity of the isotope, at a given time (t), is measured and the irradiation time and flux are known, then the mass of the original parent element (m), mass concentrations (C), as well as the number of original atoms (n<sub>P</sub>) can be calculated using the modified NAA relations as follows:

$$n_P = \frac{A_D(t_{decay})}{\phi \sigma_P (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}}} \dots\dots\dots 13$$

$$n(\text{atoms}) = \frac{A \left( \frac{\text{decays}}{\text{sec}} \right)}{\phi \left( \frac{\text{neutrons}}{\text{cm}^2 * \text{sec}} \right) \sigma \left( \frac{\text{cm}^2}{\text{atom}} \right) 1 \text{ absorption} \left( \frac{\text{atom}}{\text{neutron}} \right) 1 \text{ decay} \left( \frac{\text{decay}}{\text{atom}} \right)} \dots\dots\dots$$

$$m = \frac{A_D(t_{decay})}{\phi \sigma_P \frac{N_A}{A_m} A_{\%} (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}}} \dots\dots\dots 15$$

$$C = \frac{A'_D(t_{decay})}{\phi \sigma_P \frac{N_A}{A_m} A_{\%} (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}}} \dots\dots\dots 16$$

C: mass concentration  $\left( \frac{g}{g} \text{ or } \frac{g}{mL} \right)$

$A'_D$ : specific daughter isotope activity  $\left( \frac{Bq}{g \text{ or mL}} \right)$

Many of the activated isotopes only decay or transmute once to reach a stable state, but others go through multiple decays or combinations of transmutations and decays to reach a stable state (Todd Hansen Sherman 2012).

### 3. RESULT AND DISCUSSION

Finding the number of parent atoms in the samples ( $n_p$ )

The number of parent atoms in the samples can be calculated using the granddaughter isotopes:

$$\frac{d n_G(t)}{dt} = \phi \sigma_P n_P - \lambda_D n_D(t) - \lambda_G n_G(t) \dots\dots\dots 17$$

$n_D$ : number of daughter atoms (atoms)

$\lambda_G$ : granddaughter decay constant  $\left( \frac{1}{\text{sec}} \right)$

$n_G$ : number of granddaughter atoms (atoms)

$$A_G(t) = \frac{n_P(0) \sigma_P \phi}{(\lambda_D - \lambda_G)} \left[ \lambda_D (1 - e^{-\lambda_G t_{irr}}) e^{-\lambda_G t_{decay}} - \lambda_G (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}} \right] \dots\dots\dots 18$$

$A_G$ : activity of granddaughter isotope (Bq)

$$n_P = \frac{A_G(t_{decay})}{\frac{\sigma_P \Phi}{(\lambda_D - \lambda_G)} [\lambda_D (1 - e^{-\lambda_G t_{irr}}) e^{-\lambda_G t_{decay}} - \lambda_G (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}}]} \quad \dots\dots\dots 19$$

$$C = \frac{A'_G(t_{decay})}{\frac{\sigma_P \Phi}{(\lambda_D - \lambda_G)} \frac{N_A A_0}{A_m} [\lambda_D (1 - e^{-\lambda_G t_{irr}}) e^{-\lambda_G t_{decay}} - \lambda_G (1 - e^{-\lambda_D t_{irr}}) e^{-\lambda_D t_{decay}}]} \quad \dots\dots\dots 20$$

$A'_G$ : specific granddaughter isotope activity  $\left(\frac{Bq}{g \text{ or } mL}\right)$

In the case of nonmonoenergetic neutrons, the mass of the parent element can still be calculated without knowing the neutron energy spectrum. This can be achieved by irradiating the sample together with the unknown sample material, and using the assayed standards containing known concentrations of the same element assuming they are all exposed to same neutron beam. The mass/concentration of the standard can be multiplied by the ratio of the activity of the unknown sample over the activity of the standard. The result is the mass/ concentration of the unknown sample. By using a certified standard the need to integrate over all neutron energies, as well as the neutron cross sections, can be avoided altogether (Overview of Neutron Activation Analysis) as follows:

$$\frac{m_{Standard}}{m_{Sample}} = \frac{A_{Sample}}{A_{Standard}} \quad \dots\dots\dots 21$$

Mass concentration of the samples

The mass concentration of the sample can be calculated using the expression below:

$$C_{Sample} = C_{Standard} \frac{m_{Standard}}{m_{Sample}} \frac{A_{Sample}}{A_{Standard}} \quad \dots\dots\dots 22$$

The relative method is precise and accurate as long as the standard materials are available and similar to the sample composition

**Interaction of gamma ray with matter (detector)**

Gamma ray photons are uncharged and create direct ionization or excitation of the material through which they pass. The detection of  $\gamma$ -rays is therefore depending on causing the  $\gamma$ -ray photon to undergo an interaction that transfers all or part of the photon energy to an electron in the absorbing material. Although a large number of possible interaction mechanisms are known for  $\gamma$ -rays in matter, only four major types play an important role in radiation measurements. They are: a) Photoelectric Effects b) Compton Scattering c) Pair production d) Positron annihilation. All these processes partially or completely transfer gamma ray energy to electrons in the atom of the interacting medium. These processes are strongly on photon energy and the atomic number Z of the interacting material. Thomson scattering and others are much less important and so ignored in detection process. A brief description of the major processes is given below (SHEIKH MD. ANOWAR HOSSAIN 2018)

**The Photoelectric Effect**

A photon of relatively low energy (less than 1 MeV) may transfer all its energy to a tightly bound electron in an inner shell, causing the electron to be ejected from the absorber atom. The ejected electron, known as a photoelectron will move through the absorber causing secondary ionization and excitation. For typical photon energies, the most probable origin of the photoelectron is the innermost electron orbit or K shell. The absorber atom is left in an excited state with vacancy in one of its inner shells. The vacancy is quickly filled by the capture of a free electron from the absorber, or by rearrangement of electrons from other shells of the atom. In the latter case, electrons move from a higher energy shell to fill the vacancy; as they do so, energy is released in the form of characteristic x-rays. In a few case, the x-rays produced by the above process will interact with an outer shell electron and cause it to be ejected from the atom. It has a low energy and is known as an auger electron. Note that the photoelectric effect is most likely to occur in materials with a high atomic number, so a material such as lead (Z=82) makes a useful shielding material for low energy photons. The photoelectric

effect is relatively unimportant in low Z materials such as aluminum (SHEIKH MD. ANOWAR HOSSAIN 2018). Figure 3.1 shows the process of photoelectric effect and Figure 3.2 shows the production of auger electrons.

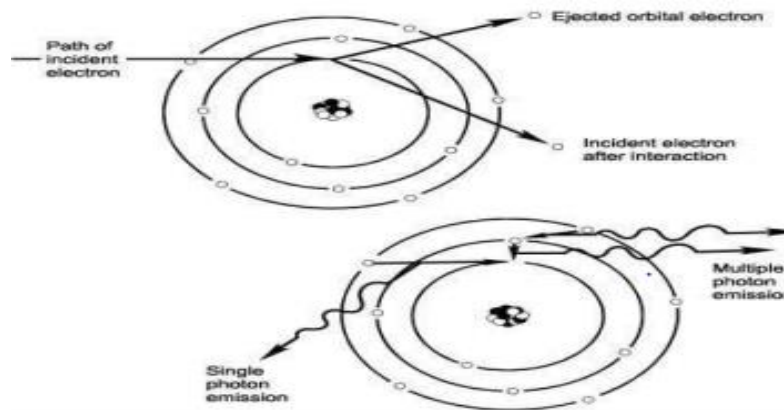


Figure 3.1: Photoelectric emission

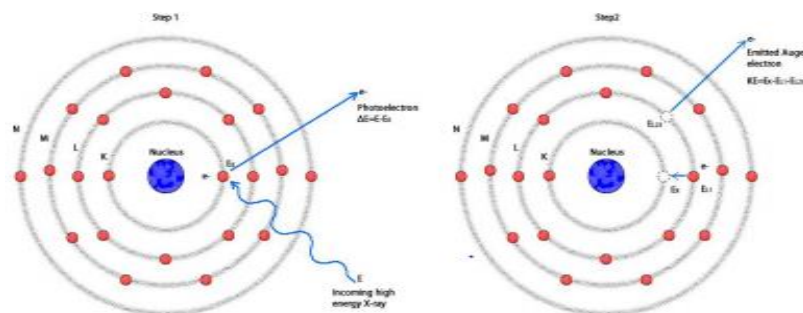


Figure 3.2: Auger electrons.

### Compton Scattering

Compton scattering involves a collision between a photon and an outer shell electron in which only part of the photon energy is transferred to the absorber atom. The electron is released from the atom (primary ionization) and will continue moving through the absorber causing secondary ionization and excitation. The photon is scattered with reduced energy and may also continue to interact with other absorber atoms. Figure 6 shows an incoming photon releasing an outer shell electron and being scattered with a reduced energy. The angle at which the photon is scattered depends on its original energy and the energy transferred to the electron. Low energy photons transfer very little energy to the released electron and scattered through large angles. However, high energy photons (10 to 100 MeV) transfer most of their energy to the released electrons and are not scattered very much. Compton scattering is most important for photon energies between about 0.2 and 5.0 MeV, and predominates in absorbers with higher values of Z (SHEIKH MD. ANOWAR HOSSAIN 2018).

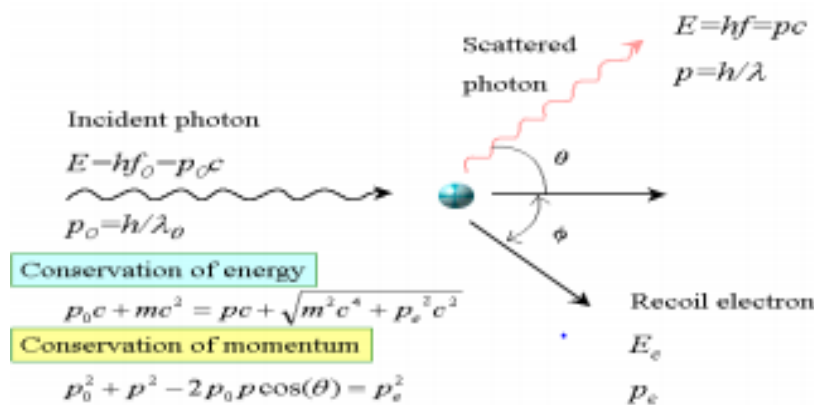
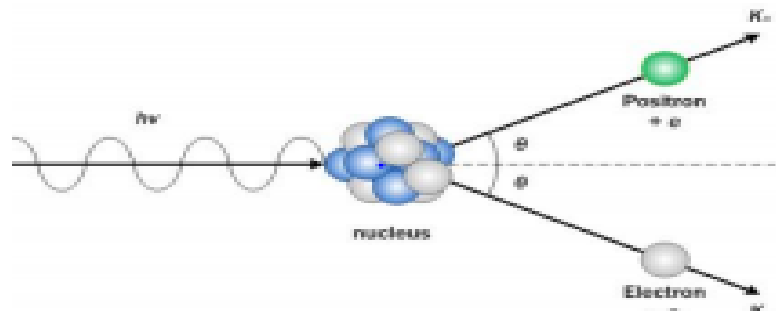


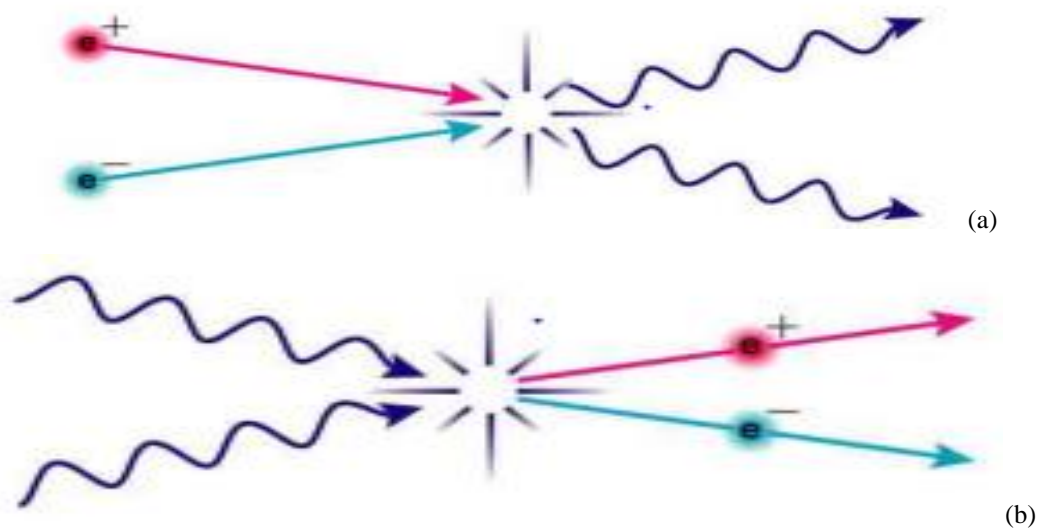
Figure 3: Compton scattering of gamma radiation.

**Pair production**

Pair production takes place when a photon with energy greater than 1.02 MeV interacts with the strong electric field close to the heavy nucleus of an absorber atom and produces two particles, an electron and a positron. The energy value of 1.02 MeV is the energy equivalent of the total mass of positron-electron pair and any photon energy in excess of this value provides kinetic energy to the electron and positron and may also cause the nucleus of the target atom to recoil. The electron and positron then move off and lose kinetic energy through secondary ionization. A positron cannot exist without kinetic energy so when it has lost all its energy it will combine with an absorber atom electron in a process called annihilation. In this process, the two particles destroy each other and are converted into two annihilation photons, each of 0.51 MeV. These photons are emitted in opposite directions from each other. Figure 3.3 shows the Compton scattering of gamma radiation and Figure 3.4 shows the pair production. Fig 3.5 shows the whole process of pair production and annihilation in the electric field around the nucleus of a lead atom. For photons with energies above the threshold, the probability that pair production will take place increase with atomic number of the absorber.



**Figure 4: Pair Production.**



**Figure 5: Pair Production and Positron Annihilation process.**

The effect also increases with photon energy, slowly from 1.02 to 5 MeV and more rapidly above that. Pair production is the most likely interaction for high energy photons in materials with a high atomic number.

**Positron Annihilation**

If pair production occurs, the positron slows down in the material by successive collision. When the positron comes to rest, it combines with nearby electron and then annihilates, producing two 0.511 MeV photons emitted in opposite directions due to energy and momentum conservation law.



### Detection of Gamma Rays using HPGe detector

After irradiation the samples and standards are placed on the HPGe detector. For the detection of the gamma rays emitted from the experimental samples an experimental arrangement was established that includes a High Purity Germanium (HPGe) detector, a Digital Spectrum Analyzer DSA-1000 with Canberra Detector Interface Module (DIM) containing a high voltage power supply, a pre-amplifier, analog to digital converter (ADC) and PC based Multi-Channel Analyzer (MCA) software-Genie 2000 etc. Figure 3.6 shows block diagram of gamma ray detection arrangement (SHEIKH MD. ANOWAR HOSSAIN 2018).

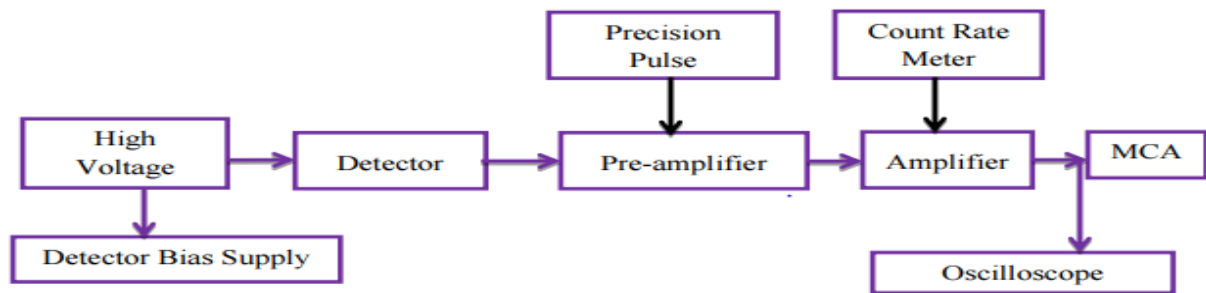


Figure 6: shows block diagram of gamma ray detection arrangement

## 4. CONCLUSION

In present study, the essential information on how NAA work in the analysis of major, minor and trace element in geological and environmental samples have been assessed. The position of NAA among the analytical techniques is still strong despite some weaknesses like the necessity of having access to a nuclear reactor, long analysis time, etc. and NAA is, with no doubt, a valuable method in geological and environmental samples analysis, especially important in certification of reference materials.

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