THE DETERMINATION OF k_0 – VALUES OF SOME DISCREPANT NUCLIDES USING STANDARD REFERENCE MATERIALS WITH NIRR-1 FACILITIES

BY

AVAA, AUNGWA ABRAHAM
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DECLARATION

I declare that the work in the thesis entitled 'The determination of k_0 – values of some discrepant nuclides using standard reference materials with NIRR-1 facilities' has been performed by me in the Department of Physics under the supervision of Prof. S.A Jonah and Dr. U Sadiq.

The information derived from the literature has been duly acknowledged in text and a list of references provided. No part of this thesis was previously presented for another degree or diploma at any university.

Avaa, Aungwu Abraham November 3, 2010

_____	_____	_____
Name of student	Signature	Date

CERTIFICATION

This thesis entitled "THE DETERMINATION OF K_0 – VALUES OF SOME DISCREPANT NUCLIDES USING STANDARD REFERENCE MATERIALS WITH NIRR-1 FACILITIES" by **Avaa, Aungwu Abraham** meets the regulations governing the award of the degree of Master of Science of Ahmadu Bello University, Zaria, and is approved for its contribution of knowledge and literary presentation.

_____ Date _____

Prof. S.A Jonah
Chairman, Supervisory Committee

_____ Date _____

Dr. U Sadiq
Member, Supervisory Committee

_____ Date _____

Dr. N Rabiu
Head of Department

_____ Date _____

Prof. Adebayo .A. Joshua
Dean, Postgraduate School

DEDICATION

This thesis is dedicated to Mr. and Mrs. Omo-erigbe

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ABSTRACT

The k_0 –values of the following nuclides (Ca-49, Ba-131, Cd-115, In-116m, Cd-116m_2, Co-60, Cs-134, Ir-192, Hg-197, Se-75, Gd-153, Gd-159, Pd-109, Ag-110m, Tm-170, Re-186, Sm-153, S-36, U-239) suspected to have discrepant k_0 –values were determined using Certified Reference Materials, (SMELS I, III and NIST 1633b Coal fly Ash). The result 1.10E-4 for ^{49}Ca at 3084keV, 1.95E-5, 3.43E-6, 6.45E-5 for ^{131}Ba at the energy of 373, 486, and 496keV 4.70E-4 for ^{115}Cd at 528keV; 1.66E-1 for ^{153}Sm at 103keV; 6.88E-3, 1.38E-3 for ^{75}Se at 136 and 401.73keV respectively; 7.27E-1, 1.54E+0, 2.28E+0 for ^{116m}In at 418.9, 1097 and 1293keV respectively; 4.00E-2, 7.32E-2, 4.67E-1, 3.93E-2 for ^{134}Cs at 562, 569, 602 and 802keV respectively were gotten. The student t-test which is the statistical analysis was used to compare the result of this work with the recommended data. The result of the comparison at 5% confidences intervals showed no significance difference which reveals the accuracy of the k_0-standardisation adopted in Nigeria Research Reactor -1 (NIRR-1).

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ABBREVIATION

BDL - Below Detection Limit

CERT - Centre for Energy Research and Training

Df - Degree of Freedom

GHARR-1- Ghana Research Reactor-1

HPGe - High purity Germanium

IAEA - International Atomic Energy Agency

MCA – Multi Channel Analyser

MNSR - Miniature Neutron Source Reactor

NAA - Neutron Activation Analysis

NIRR-1 - Nigeria Research Reactor-1

NA - Not Available

RNAA - Reactor Neutron Activation Analysis

SMELS - Synthetic Multi-elemental Standard

CHAPTER ONE

1.0 Introduction

The interest in the role of trace, minor, and major elements in biology, environmental research, geology, and technology (to ensure safety) for the socio economic development of several countries has led to an increasing need for multi element analysis on a large number of samples. Various standardization methods have been used in neutron activation analysis (NAA), namely: Absolute, Relative or Single comparator. In the absolute standardization, the physical parameters characterizing an element were determined by independent methods, their imprecision' would add up when calculating the amounts of elements (Dung and Hien; 2003) leading to large systematic errors. In the relative standardization, the unknown sample is irradiated together with a standard or reference material containing a known amount of the element of interest. The standard reference material is measured under the same counting condition as the sample. This standardization method is still being regarded as one of the methods which has the highest accuracy of NAA (Dung and Hien; 2003). However, it is not suited for multi-element analysis with regards to the experimental workload and the impossibility to quantify unexpected elements. The single comparator standardization makes multi-element analysis with NAA feasible via the k_0-factors, which are experimentally determined by co-irradiation of a standard and a single comparator.

The k_0-standardization method of NAA (k_0-NAA) launched in the mid 1970s (Simonits et al; 1975) is able to overcome the above mentioned disadvantages. It has

been developed as absolute standardization, where the unreliable nuclear data are replaced by accurately measured composite nuclear constants, k_0-factors. The k_0-NAA method involves the co-irradiation of sample with a neutron flux monitor such as gold, and calculating the nuclear constant called k_0 by imputing certain experimental parameters. The method is widely accepted for multi-element NAA with research reactors because it allows flexibility with respect to variations in neutron spectrum parameters and gamma- ray counting geometry (Jonah et al; 2005). The adoption of this method brought about elimination of standards for each element and the inaccuracy caused by poorly prepared in house standards. There is however a need to ascertain the accuracy of the k_0-values used in k_0-NAA by continuous measurement because the use of inaccurate k_0-value will result to a systematic error which is linearly transferred to the analytical result.

This present work is aimed at determining the k_0- values of the discrepant nuclides using Synthetic Multi - element Standards (SMELS), which is the reference materials recommended by the International k_0-user Community for the validation of k_0 –NAA method and the standard reference materials , NIST-1633b (Coal Fly Ash) .

1.1 **Research Problem**

The k_0 –NAA method has been adopted by many Nuclear Research Centers including Nigeria for multi- element analysis. However, some discrepancies were observed in the data of the following nuclides,Ca-48, Ba-131, Cd-115 and In-116m using k_0 –NAA (Wispeleaere and De Corte;2003) Recommendation was made to re-evaluate the corresponding k_0- values or true-coincidence correction factors. Problems

were also outlined related to the inaccuracy of the k_0 –factors of ^{131}Ba, ^{115}Cd, and ^{115m}In (Simodis et al; 1994). The following nuclides (^{48}Ca, ^{131}Ba, ^{115}Cd, ^{116m}In, $^{116m2}Cd$, ^{60}Co, ^{134}Cs, ^{192}Ir, ^{197}Hg, 75Se, ^{153}Gd, ^{159}Gd, ^{109}Pd, ^{110m}Ag, ^{170}Tm, ^{186}Re, ^{153}Sm, ^{36}S, ^{239}U) were suspected to have discrepant k_0 –values. Recommendations were to re-measure them by the International k_0 –user community.

The calculation of k_0- values using nuclear data from the literature gave results that differ significantly from the recommended data, these are nuclides suspected to be discrepant in their k_0- values by the international k_0 user community. Hence the need to determine experimentally the k_0- values of the discrepant nuclides.

1.3 Justification of Study

The large deviations that appear in the comparison of calculated k_0- values using randomly chosen nuclear data from the recommended k_0- values in Table 4.4 reveals the unreliability of the nuclear data. These nuclear data are compilation made several years ago and they turn out to be obsolete (Moens et al; 1983). It has also been reported that good accuracy would be obtained from continuous updating of the k_0- values

Table 1.1, shows the result of measured k_0- values and its accuracy when compared to the recommended data by De Corte and Simonits, (2003), St- Pierre and Kennedy, (2003, 2006).

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Table, 1.1: Re-measured k_0-values of discrepant nuclides

Target nuclides	Gamma energy (Kev)	k_0 -values (Kennedy and St-Pierre, 2003)	Recommended k_0 –values (De Corte and Simonits, 2003)	% dev.	k0-values (St-Pierre and Kennedy,2006)	% dev.
^{115m}Cd	336	6.59E-4	7.73E-4	14.7	7.20E-4	6.9
^{110m}Ag	884	2.98E-2	2.69E-2	-10.8	2.83E-2	-5.2
^{109}Pd	88	1.44E-3	1.71E-3	15.8	1.52E-3	11.1
^{76}As	559	4.67E-2	4.83E-2	-3.3	4.75E-2	1.7
^{88}Rb	1836	1.43E-4	1.57E-4	-8.8	1.55E-4	1.3
^{85}Sr	514	9.91E-5	9.15E-5	8.3	9.33E-4	-919
^{116m}In	1097	1.73E-0	1.60E-0	8.1	1.66E-0	0
^{128}I	443	1.15E-2	1.12E-2	3.0	1.19E-2	-6.3
^{134}Cs	796	4.04E-1	4.15E-1	-2.8	4.02E-1	3.1
^{134m}Cs	127	5.63E-3	5.48E-3	2.8	5.76E-3	-5.1
^{170}Tm	84	3.41E-2	3.26E-2	4.6	3.45E-2	4.7
^{182}Ta	1221	6.30E-2	6.45E-2	2.3	6.28E-2	2.6
^{186}Re	137	4.60E-2	4.33E-2	-6.2	4.48E-2	-3.5

From Table 1.1, it is observed that the uncertainties could be reduced by more accurate measurements. In this work, some of the discrepant k_0-values were re-measured.

The Nigeria Research Reactor-1 (NIRR-1) is a 31kW Miniature Neutron Source Reactor (MNSR) designed mainly for Neutron Activation Analysis (NAA) and limited Radioisotope production. It attained criticality in 2004. It has a stable maximum neutron flux (1×10^{12} n.cm^{-2}s^{-1}) during steady state operation and has a burn-up of < 1 % for the entire life time of core. This keeps the neutron spectrum parameters constant for the entire period (Jonah et al; 2009). This is an advantage of the reactor for adaptation of k_0-method and it gives the basis for which the k_0-values of the discrepant nuclides can be measured and result obtained is expected to be in good agreement with the certified values.

1.3 Objectives of this study

This study is aimed at achieving the followings:

- Calculate the k_0 –values using nuclear data in the literature
- Re- measure the k_0-values of the following discrepant nuclides Ca-48, Ba-131, Cd-115, In-116m, Cd-116m2, Co-60, Cs-134, Ir-192, Hg-197, Se-75, Gd-153, Gd-159, Pd-109, Ag-110m, Tm-170, Re-186, Sm-153, S-36, U-239) using Certified Reference Materials (CRM) with NIRR-1 facilities
- Contribute to the k_0-Nuclear Data library and recommend k_0- values for nuclides whose values are discrepant
- Establish some of the possible reasons for such discrepancies and make appropriate recommendations

1.4 Previous Work

The k_0 – standardization technique of NAA has been developed in 1970s (Simonits et al, 1975) for multi elemental analysis. The k_0-based NAA involves the simultaneous irradiation of a sample and a neutron flux monitor, such as gold, and the use of a composite nuclear constant called k_0 – factor. The k_0 – factor is independent of irradiation and measuring conditions. This technique eliminates the need of using multi element standards (Simonits et al. 1975).

Moens et al, (1983) reported that the method was applied by El Nimr (1981) for the determination of k_0-factors for 44 (n, γ) reactions and the average uncertainty was found to be 2%. From a comparison of the measured result and the calculated values a reasonable agreement was attained. Observed differences were non-systematic and below 5%. The average deviation amounted to $\sim$ 3% which is fully acceptable keeping in mind the experimental uncertainties of the compared k_0- values. This agreement shows the accuracy of the k_0 – factors obtained by Moens et al, (1983) and the one published by IAEA, (1974).

In a related development, the standardization of k_0- NAA was carried out at Ghana Research Reactor -1 Centre (Nyarko et al, 2003) using Certified Reference materials for the validation of the method with gold as the comparator. The result of the k_0-values obtained for these nuclides (Al, Ca, Fe, Mg, Na, K, Mn, Cr, As, Co, Cu, V, I) were in agreement with the recommended k_0-values in literature, however, the % uncertainty for some nuclides were over 5% when compared with the certified values in (Moens etal, 1983).

Also Kennedy and St-Pierre, (2003) in their quest to verify the accuracy of the k_0 method for all elements with high Q_0 values; Standards of 40 elements were prepared and irradiated in the inner and outer irradiation sites of the Ecole Polytechnique SLOWPOKE reactor and then counted 1cm from a 29% Germanium Detector and the concentration of each element was calculated from the gamma-ray spectrum using the k_0- method. For 13 nuclides with high Q_0- values the results differed from expected amounts by more than 5% which suggests that measurement of new k_0 and Q_0-values are needed. However, in the work of St- Pierre and Kennedy; (2006), the k_0 and Q_0-values for 14 nuclides were re-measured. Their results for ^{76}As, ^{88}Rb, ^{85}Sr were in good agreement with the literature values. Similarly, the results for ^{115} Cd, ^{128}I, ^{109}Pd, ^{110m}Ag, ^{116m}In, ^{134}Cs, ^{134m}Cs, ^{131}Ba, ^{170}Tm and ^{186}Re for some gamma ray peaks seems to be in good agreement with literature values while the k_0- values for the following nuclides ^{115m}Cd, ^{170}Tm, ^{186}Re, ^{134m}Cs, ^{110m}Ag, ^{182}Ta, ^{131}Ba at different energy peaks disagree with the literature values. Hence they concluded that the uncertainty could be reduced by more accurate measurement.

Furthermore, the k_0 – INAA method has been used for elemental analysis with NIRR-1 irradiation and counting facilities (Jonah et al; 2009). The synthetic Multi-element standards (SMELS) types I, II and III as well as standard references materials were analyzed for the validation of the method in NIRR-1, NAA laboratory. Result obtained show good agreement with recommended values in the certificates and confirm the suitability of the k_0-IAEA programme for elemental analysis with low power nuclear research reactor, such as the MNSR that exhibit stable neutron spectrum

parameters for the entire core lifetime. This is an advantage for the determination of k_0-values, since the measurement can be carried out over a long period of time under stable conditions. Therefore, the present work was carried out to re-evaluate the k_0-values of the twenty discrepant nuclides whose values do not agree with the recommended data in literature.

CHAPTER TWO

LITERATURE REVIEW

2.1 Neutron Sources

Neutrons do not exist freely in nature and so are produced based on either spontaneous fission or on nuclear reactions. Some of the sources especially the ones available in CERT are described below:

2.1.1 Neutron Generators

Neutron Generators like the one available in CERT, A.B.U. Zaria uses the reaction ^{2}H (d,n)^{4}He with Q = 17.58 MeV to produce mono-energetic neutrons of energy 14 MeV. Their basic operating principle is based on the fact that charged particles are accelerated to an adequate energy and then directed to strike a target. (Knoll, 1979)

2.1.2 Isotopic Neutron Sources

The source comprises of an alpha-radiation material closely packed with neutron emitter like beryllium. The reaction producing the neutrons is:

$$^{4}_{2}\alpha + ^{9}_{4}Be \rightarrow ^{12}_{6}C + ^{1}_{0}n$$

With Q = 5.7 Mev, typical α-emitters are ^{226}Ra [Eα= 4.78 Me], ^{241}Am [Eα = 5.48meV] ^{239}Pu [Eα=5.16 MeV] and ^{210}Po [Eα=5.30 MeV]; the mixtures are known as Ra-Be, Pu-Be, Po-Be and Am-Be, neutron sources. The α-particles experience considerable energy loss before interacting with Beryllium, and as a result the energy spectral of the neutrons produced is continuous.

2.1.3 Nuclear Reactors.

Nuclear reactors with high fluxes of neutrons from fission of Uranium-235 give the most intensive irradiations better than Neutrons Generators and Isotopic sources and hence, have the highest available sensitivities for Neutron Activation Analysis (NAA).

The neutron energy distributions of reactors vary considerably in relation to the type of reactor and between irradiation channels within a reactor. Fission produced neutrons have energies ranging up to 15 MeV with an average energy of about 2 MeV (IAEA, 1990).Through elastic collisions with moderator nuclei, the fission neutrons rapidly become thermalised creating a broad energy distribution consisting of three principal component- thermal, epithermal, and fast. The Nigeria Research Reactor-1 (NIRR-1) is the source of neutrons for this work.

2.2 Description of NIRR-1

The Nigeria Research Reactor-1 (NIRR-1) which is a Miniature Neutron Source reactor (MNSR) is designed mainly to serve as a neutron source. It is a tank-in pool type reactor with 90% enriched Uranium as fuel, light water as moderator and coolant, and metallic beryllium as reflector. It has a nominal thermal power rating of 31KW and a built-in clean cold core excess reactivity of 3.77mk measured during the on-site zero power and criticality experiments. The reactor can operate for 4.5 hours at full power, equivalent to a neutron flux of 1×10^{12} n/cm^2.s. The reactor can run for over ten years under these conditions with a burn-up of < 1%. The Table below shows the main specifications of NIRR-1.

Table 2.1: Reactor Specification

Reactor type	Tank-in pool
Nominal thermal power	31kW
Fuel type	U-Al alloy >90% enrichment
Number of fuel elements	347 (with 248mm in length, active length is 230mm and diameter of fuel meat being 4.3mm)
Loading of 235U in the core	2.88grams per pin
Reflector	Metallic beryllium
Coolant and moderator	Light water
Number of control rod	1
Control rod material	Cd
Neutron irradiation channels	5 inner and 5 outer
Thermal neutron flux(normal condition)	Inner irradiation channels $1 \times 10^{12} n/cm^2.s$ Outer irradiation channels $\sim 5 \times 10^{11} n/cm^2.s$
Cd ratio of Au	2.12±0.02 (inner)
Thermal- epithermal flux ratio	19.2±0.5 (inner) 48.3±3.3 (outer)
Neutron temperature	333.7K
Cladding material	Al alloy

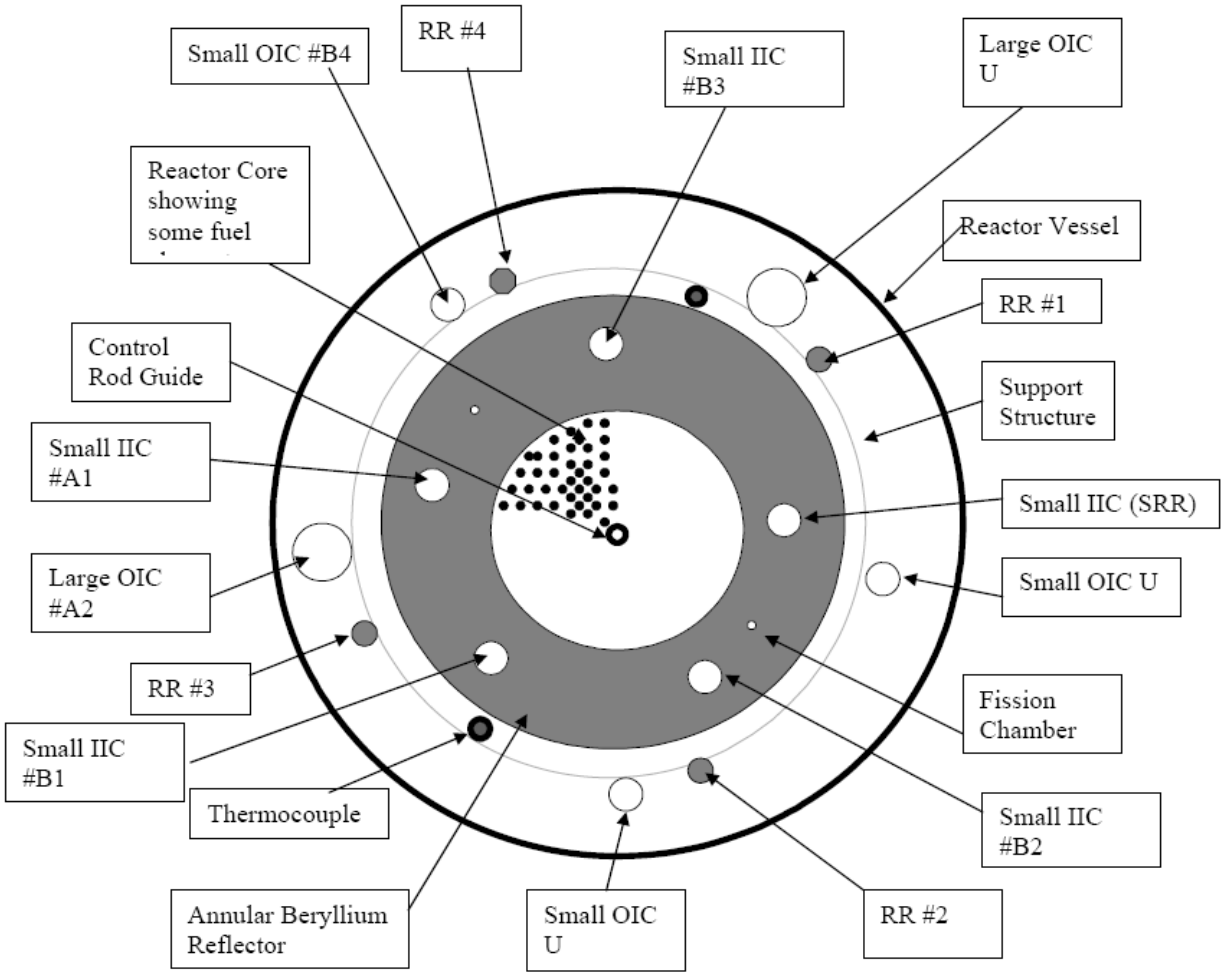


Fig. 2.1 Lay-out of NIRR-1 core showing irradiation channels and control devices

2.2.1. Reactor Complex

The reactor complex is composed of reactor core of cylindrical fuel assembly.NIRR-1 has only one control rod which performs the functions of safe startup, shutdown, power regulation and reactivity control of the reactor. The reactor core assembly is surrounded by beryllium reflectors located at the bottom of the reactor vessel. It also has in-pile detector, irradiation channel, moderator, and coolant as well as reactor vessel as shown in fig.2.1.

2.2.2. Control System and Control console

The control system of the reactor can be linked to its nervous system. It is used for indicating the reactor conditions and commanding reactor operations (like: normal and abnormal, start up and shut down). Because NIRR-1 has inherent safety and stabilizing characteristics, so the whole control system can be simplified greatly. The control system includes control console, neutron detector, and drive mechanism for control rod, and control rod movement.

2.2.3. Auxiliary System of NIRR-I

In order to ensure safe operation of the reactor and radiation exposure of personnel in working site, some auxiliary systems are arranged for NIRR-1. They are: reactor water purification system and pool water purification system, which guarantee the quality of reactor water and pool water, reactor gas purge system, thermal hydraulic parameter monitoring system and pool water cooling system.

2.2.4. Irradiation System

The irradiation facility of NIRR-1 consists of five small rabbit tubes installed inside the be annulus and are known as the inner irradiation channels. Three small and two large rabbit tubes installed outside of the Be annulus are known as the outer irradiation channels. Six out of the ten irradiation sites are presently connected to the pneumatic transfer systems. There are two sets of transfer systems, the pneumatic transfer system type A, which is suitable for the irradiation of middle and long-lived nuclides. The other is a multi-functional transfer system simply known as the pneumatic transfer system B. It is used to send a sample into the reactor and retrieved it (Jonah and Balogun). In order to optimize utilization of NIRR-1 for k_0-NAA, experimental procedures were developed for the characterization of the irradiation channels. At present, the irradiation channels B2, B4, A1 and A2 have been characterized and the neutron spectrum parameters have been determined (Jonah et al, 2009)

2.3 General Principle of Neutron Activation Analysis (NAA)

Neutron Activation Analysis (NAA) is a sensitive analytical technique useful for performing both qualitative and quantitative multi-element analysis of major, minor, and trace elements in samples from almost every conceivable field of scientific or technical interest. For many elements and applications, NAA offers sensitivities that are superior to those attainable by other methods. In addition, because of its accuracy and reliability, NAA is generally recognized as the " referee method" of choice when new procedures are being developed or when other methods yield results that do not agree (Glascock, 2000).

Fig.2.2 gives an illustration of the sequence of events occurring during the most common type of nuclear reaction used for NAA, namely the neutron capture or (n,γ) reaction. When a neutron interacts with the target nucleus via a non-elastic collision a compound nucleus forms in an excited state. The compound nucleus will instantaneously de-excite into a more stable configuration through emission of one or more characteristic prompt gamma rays. This new configuration yields a radioactive nucleus which also de-excites (or decays) by emission of one or more characteristics delayed gamma ray (Glascock, 2000)

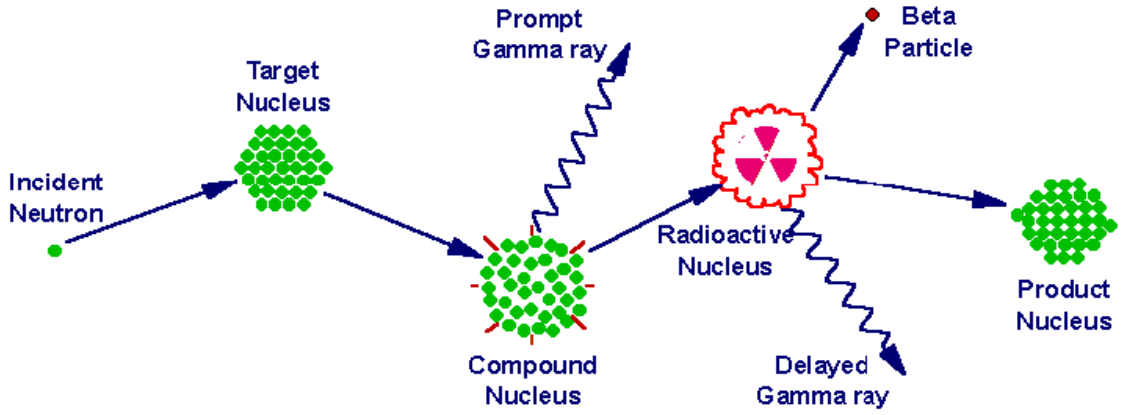


Fig 2.2 Diagram illustrating the process of neutron capture by a target nucleus followed by the emission of gamma rays.

2.3.1. NAA Procedure

The NAA procedure may involve some or all of the following steps:

I. Sampling

II. Pre-irradiation sample treatment (such as cleansing, drying or ashing, pre-concentration of elements of interest or elimination of interfering elements, sub-sampling and packing)

III. Irradiation (and prompt γ-ray counting in PGNAA)

IV Radiochemical separation (only in RNAA)

V Radioactivity measurement

VI Elemental concentration calculation

VII Critical evaluation of results and preparation of the NAA report

2.3.2. Forms of NAA

The following are the forms of NAA:

I. Instrumental neutron activation analysis (INAA)

II. Radiochemical neutron activation analysis (RNAA)

III. Epithermal neutron activation analysis (ENAA)

IV. Prompt gamma-ray neutron activation analysis (PGNAA)

V. Cyclic neutron activation analysis (Cyclic NAA)

However, with respect to the time of measurement, NAA falls into two categories;

1. PGNAA, where measurements take place during irradiation or
2. Delayed Gamma-ray Neutron Activation Analysis (DGNAA), where the measurements follow radioactive decay (Glascock, 2000).

The need for a fast computerized analytical method for Instrumental Neutron Activation Analysis (INAA) is obviously in view of the wide application of the technique in multi-element analysis. The availability of standard reference materials from agencies like the National Institute of Standard and Technology (NIST), United States Geological Service (USGS), The International Atomic Energy Agency (IAEA) and other national bodies make the distribution of Certified Reference Materials fairly easy for use. However, reinvestigations of these method by many workers (Greenberg, 1987, Glascock,.1993; Molnar et al., 1993) have revealed departures from the certified values leading to the use of consensus values for these CRMs as against their certified values (Flanagan, 1973).

2.4 Absolute method of NAA

In the absolute method, the concentration of elements in a sample can be calculated with the activation and decay equation. [2.1] and [2.2] provided the reaction rates, detector efficiency curve, half-lives, and decay schemes of radionuclide are known.

$$D = (NWF/M)*R\ [1-\exp(-\lambda Ti)] \qquad 2.1$$

$$A = \varepsilon TD \exp(-\lambda Td) \qquad 2.2$$

Where,

N = Avogadro's number, 6.033×10^{23} atoms/mole

W = weight of element irradiated in grams

F = fractional abundance of target isotope

M = atomic weight of element

R = nuclear reaction rate per nucleus of target isotope

λ = decay constant

T = branching ratio of identifying gamma ray

ε = detector photo peak efficiency

Ti = irradiation time

Td = decay time

The reaction rate can be determined by measuring the neutron fluxes with monitor elements like Au, which is co- irradiated with samples. If the nuclear parameters are precisely known, then this method can yield accurate concentrations for elements in the sample. In practice, however, uncertainties in the nuclear parameters can easily affect the accuracy for determining the concentration of an element of interest (IAEA, 1990).

2.5 The Relative method of NAA

This method is generally accepted as the most accurate way to quantify element concentrations. In this method, samples are irradiated simultaneously with standards containing known amounts of elements. After irradiation, both samples and standard are measured under identical geometrical conditions with the same detector. This procedure eliminates uncertainties in the nuclear parameters, detector efficiencies, etc. and reduces the NAA equation for each element to its simplest form:

$$\frac{Ws}{W_{Au}} = \frac{A_S}{A_{AU}} \frac{esp(+\lambda T_{DS})}{esp(+\lambda T_{DAu})} \qquad 2.3$$

Where

Ws = weight of element being sought in sample

W_{Au} = weight of the element in standard

T_{DS} = decay time for the radioisotope measured

T_{DAu} = decay time for the standard

A_S = activity of radioisotope measured

A_{AU} = activity of standard

In the multi- elemental analysis of a sample, the comparator method requires a large number of individual element standards or use of a well-characterized multi-element standard (IAEA, 1990). Preparation and irradiation of individual standards for each element are time consuming and expensive; and the use of reference materials is not always practical because of the limited accuracy for certain elements or the difference between sample and standard matrices. As a result the k_0 –standardization method was developed.

2.6 k_0 - Standardization method

The concept of k_0 – standardization in NAA, one of the most frequently used single comparator method is based on co-irradiation of the sample and a neutron fluence rate monitor and on using an experimentally determined composite nuclear constant

[k_0.n(a)] (Kucera et al, 1983) the establishment of this method is to hold the following merits:

I. The troublesome preparation of chemical standards can be avoided

II. The radioactivity counting time for standards can be saved

III. Quantitative results for elements, but absent in chemical standards can be obtained

IV. Some nuclear parameters, e.g. the cross- section of neutron capture reaction and the branching ratio of the gamma-ray emitted by the radioactive nuclide, can be re-evaluated

The k_0-factors are intermediary in the calculation of the concentration of the elements thus yielding good traceability, flexibility and adaptability to computerization (Ewa, 2002). The theory of the method, its basic equations is hereby reviewed.

The induced activity A on a sample of mass *m* irradiated in a reactor under the flux of both thermal and epithermal neutrons is given (Sansoni, 1985) theoretically as

$$A = \left(\phi_{th}\delta_{th} + \phi_e I_0\right)m * \frac{N_{av}}{M} * \theta * S * D * C \qquad 2.4$$

Where,

ϕ_{th} = thermal neutron flux

ϕ_e = epithermal neutron flux

σ_{th} = thermal absorption cross section for neutron (velocity 2200m/s)

I_0 = resonance integral

N_{av} = Avogadro's constant

M = Atomic mass of irradiated element

θ = Isotopic abundance of target nuclide

S = $\left(1 - e^{-\lambda t_i}\right)$ = saturation factor with t_i = irradiation time

D = $e^{-\lambda t_d}$ = decay constant with t_d = decay time

C = $\left[\frac{1 - e^{-\lambda t_c}}{\lambda}\right]$ = counting time correction with t_c = counting time

λ = $\frac{\ln 2}{T_{1/2}}$ = decay constant

The activity of the irradiated samples for any nuclide with a gamma emission probability γ when counted for a duration t_m by a detector of efficiency ε_γ is given as;

$$A = \frac{N_P}{t_M * \gamma * \varepsilon_\gamma} \qquad 2.5$$

Where,

Np is the net photo-peak counted at specified energy for the nuclide of interest. Combining equation [2.4] and equation [2.5] we have,

$$\frac{N_P}{S * D * C * m} = (\phi_{th}\sigma_{th} + \phi_e I_0) * \frac{N_{av}}{M} * \theta * \gamma * \varepsilon_\gamma * t_m \qquad 2.6$$

Where all the parameters retain their usual meaning,

If the specific intensity for the irradiated sample is designated as I_{sp} then;

$$I_{sp} = \frac{N_P / t_m}{S * D * C * m} \text{ and } I = \frac{N_P / t_m}{S * D * C} \qquad 2.7$$

Substituting equation [2.7] into [2.6], equation [2.6] can be rewritten for the irradiated sample as;

$$I_{sp} = (\phi_{th}\sigma_{th} + \phi_e I_0) * \frac{N_{av}}{M} * \theta * \gamma * \varepsilon_{\gamma} \qquad 2.8$$

Also rewriting equation [2.8] for a co-irradiated Gold as the comparator gives;

$$I_{sp(Au)} = (\phi_{th}\sigma_{th} + \phi_e I_0)_{Au} * \frac{N_{av}}{M_{Au}} * \theta_{Au} * \gamma_{Au} * \varepsilon_{\gamma\,Au} \qquad 2.9$$

The infinitely dilute resonance integral I_0 is defined (Glascock, 1988) as:

$$I_0 = \int_{E_{Cd}}^{\infty} \sigma(E)\frac{dE}{E} \qquad 2.10$$

By definition of k_0 –factors using equation [2.8] and [2.9] give the ratio of specific intensities (I_{sp}) for any given nuclide compared with that of Gold ($I_{sp\,Au}$) as;

$$k = \frac{I_{SP}}{I_{SP(Au)}} = \frac{(\phi_{th}\sigma_{th} + \phi_e I_0)\frac{N_{av}}{M} * \theta * \gamma * \varepsilon_{\gamma}}{(\phi_{th}\sigma_{th} + \phi_e I_0)_{AU}\frac{N_{av}}{M_{AU}} * \theta_{AU} * \gamma_{AU} * \varepsilon_{\gamma_{AU}}} \qquad 2.11$$

Hence,

$$k = \frac{M_{Au} * \theta * \gamma * \sigma_{th} * \varepsilon_{\gamma}(f + Q_0)}{M * \theta_{Au} * \gamma_{Au}\sigma_{th(Au)}\varepsilon_{\gamma}(f + Q_0)_{Au}} \qquad 2.11$$

where,

f = thermal to epithermal flux ratio

Q_0 = the ratio of resonance integral to thermal cross-section

Equation [2.11] yields the k-factors for any given nuclides of interest and f determines the ratio of the flux

$$f = \left(\frac{\phi_{th}}{\sigma_e} \right)$$

Since the k_0-formalism is written on the Hogdah-convention (Hogdah, 1962). The nuclear constant Q_0 is defined here as the ratio of I_0/σ_{th} . Having known the k_0-factor for a given nuclide of interest from intensities, the mass (m) of the irradiated sample could be obtained as follows:

$$m = \frac{I}{kIsp_{Au}}$$

From equation [2.11] an expression for k_0, which is a nuclear constant independent of irradiation and measurement conditions could be thus:

$$k_0 = \frac{M_{Au} * \theta * \gamma * \sigma_{th}}{M * \theta_{Au} * \gamma_{Au} \sigma_{th(Au)}} \qquad 2.12$$

These parameters are taken from literature and used to calculate the k_0-values. The k_0-factors can also be determined experimentally using equation [2.13] which is written in the Hogdahl formalism.

$$k_{0\cdot Au} = \frac{(N_P / WtmSDC)_a f + Q_{0.Au}(\alpha)\varepsilon_\gamma}{(N_P / WtmSDC)_{Au} f + Q_{0.a}(\alpha)_{Au}\varepsilon_{\gamma\,Au}} \qquad 2.13$$

Where,

$Q_0(\alpha)$ = the ratio of resonance integral to thermal cross-section corrected for non-ideal epithermal neutron flux, while all the parameters retain their usual meaning The flux ratio f, is gotten from the expression,

$$f = (R_{cd} - 1)I_0(\alpha)/\sigma_{th} = (R_{cd}-1)\, Q_0(\alpha) \qquad 2.14$$

With cadmium ratio given as:

$$R_{cd} = \frac{Aspb}{Asp\ ,\ cd} \qquad 2.15$$

Where,s

R_{cd} = cadmium ratio

$I_0(\alpha)$ = resonance integral

σ_{th} = thermal neutron capture cross-section

A_{spb} and A_{sp},cd are the specific count ratio of the bare and cadmium covered target of gold.

To determine the ratio of resonance integral to thermal cross-section, the cadmium ratio of the monitor is used. The equation that relates the two parameters is given by:

$$Q_0(\alpha) = f/F_{cd}\, R_{cd}-1 \qquad 2.16$$

Where,

F_{cd} = cadmium transmission factor for epithermal neutrons

The concentration of an element in the sample for a stable 'ideal' epithermal neutron flux could therefore be written in terms of k_0 as :

$$C(ppm) = \frac{(N_P / WtmSDC)_a}{(N_P / WtmSDC)_{Au}} \cdot \frac{1}{k_0} \cdot \frac{f + Q_{0.Au}(\alpha)}{f + Q_{0.a}(\alpha)} \cdot \frac{\varepsilon_{P.Au}}{\varepsilon_{P.a}} \qquad 2.17$$

The k_0 can alternatively be determined by co-irradiating the element and the comparator both with and without Cd-cover (Moens et al, 1983).The equation for determining the k_0-values is given as:

$$k_0 = \frac{A_{sp.a} - (A_{sp.a})Cd}{Asp.Au - (Asp.Au)Cd} \frac{\varepsilon_{p.Au}}{\varepsilon_{p.a}} \qquad 2.18$$

The k_0-values gotten from the use of equation [2.18] is neither influenced by the non-ideality of the epithermal spectrum (Moens et al, 1983) nor by epithermal self-shielding and nuclear interferences caused by fast neutron reaction and independent of nuclear data.

2.6.2 Full-Energy peak detection efficiency (ε_p)

All radiation detectors will, in principle, give rise to an output pulse for each quantum of radiation which interacts within its active volume. For uncharged radiations such as gamma rays or neutrons must first undergo a significant interaction in the detector before detection is possible Knoll (1979). Because these radiations can travel large distance between interactions, detectors are often less than 100 percent efficient

Knoll (1979). It then becomes necessary to have a precise figure for the detector efficiency in order to relate the number of pulses counted to the number of neutrons or photons incident on the detector.

In the field of neutron activation analysis and gamma ray spectroscopy, there is the need to have accurate knowledge of the efficiency of the detection system for the source- detector geometry. Two ways of calculating detector's efficiency are via the fixed source-detector geometry and the variable source-detector geometry respectively (Medhat et al ;2001).

A. The fixed sample-detector geometry

For the fixed sample- detector geometry, the full energy peak efficiency (ε) depends only on the gamma-ray energy (E) that is, $\varepsilon = \varepsilon$ (E). However, the energies of the gamma active radio-nuclides produced by thermal neutron activation are in the range of 20-4000keV (Osae et al; 1999). Since there are not sufficient gamma-ray standard sources to cover this wide range, the most convenient method for the determination of the efficiency within this energy range is by interpolation. This method involves choosing as primary standards, some radio-nuclides of relatively long half-lives with known emission probabilities and well resolved gamma energy peaks within the energy range (Osae et al; 1999). Using these standard sources the full energy peak efficiency of the detection system can be obtained by dividing the detected count rate by the emission rate.

$$\varepsilon = \frac{\text{count rate count/sec}}{\text{emission rate em/sec}} \qquad 2.19$$

Where, detected count rate = $\frac{\text{net peak count}}{\text{Live time}}$ 2.20

A theoretical curve can then be fitted to the experimental data from which the efficiency at any energy within the range can be obtained by calculations. This method was adopted for this work.

B. The variable sample-detector geometry

For the variable sample-detector geometry, the full energy peak efficiency of the detector system does not depend only on the energy of the gamma ray (E) but also on the sample- to –detector distance (d) that is ; $\varepsilon = \varepsilon(d,E)$. By adopting the fixed sample-detector geometry approach, experimental measurements were made at different source-detector distances. The measured efficiencies, as a function of E, are plotted on the same graph and a family of curves is obtained, each curve representing a different d distance from the detector surface (Osae et al; 1999).

2.6.3 Q_0 : This stand for the ratio of the resonance integral to the 2200m/s cross section (**$Q_0 = \frac{I_0}{\sigma_0}$**), in order to avoid errors introduced on the RNAA results by using Q_0,s instead of the correct $Q_0(\alpha)$,s. It is essential to perform appropriate corrections for $\alpha \neq 0$ (De Corte et al, 1981).

2.6.4. Neutron Spectrum Parameters *f* and α

The epithermal neutron flux shape factor (α) and sub-cadmium-to-epithermal neutron flux ratio (f) are important parameters used in the determination of k_0-values using equation [2.13]. The parameters are dependent on each irradiation facility and

must be determined for standardization and application purposes. The value of α can be positive or negative depending on the reactor configuration which corresponds to 'softened' or 'hardened' epithermal neutron spectrum compared to the ideal one (Acharya and Chatt; 2003), it is a factor that correct for non-ideal conditions arising from resonances. The flux monitors such as Au, Zn and Zr can be used for the evaluation of these parameters. The parameters f and α vary from one reactor to another as well as among the different irradiation sites in a given reactor.

For NIRR-1, the two parameters have been determined for irradiation position (inner and outer irradiation channels) for the implementation of the k_0-method (Jonah et al, 2005). This was done using the "Cd-ratio for multi-monitor" because of the stable neutron flux characteristics of the reactor. Other reactors with the same configuration and moderator e.g, the Canadian SLOWPOKE, GHARR-1 and Chinese MNSR, have adopted the same method. The advantage of this method is that in the determination of cadmium ratio method only the peak area ratio is taken into account as such no input data on the detector efficiencies are required as in the case of multi isotope or bare resonance method (Akaho et al, 2002).

The result obtained by (Jonah et al,2005) for neutron spectrum parameters for the inner irradiation channel (B2) and outer irradiation channel (B4) as representations of all the irradiation channels since they are symmetrical with respect to the core is consistent with the measured values available from SLOWPOKE and other MNSR except for the deviation in the α value for the inner irradiation channel of GHARR-1 as seen in table 2.1

Table 2.2, Comparison of f and α -values in irradiation channels of Slowpoke and MNSR facilities

Facility	Inner irradiation channel		Outer irradiation channel	
	f	α	f	α
MNSR, NIRR-1:Zaria, Nigeria 90% ^{235}U	19.2±0.5	-0.052±0.002	48.3±3.3	+0.029±0.003
MNSR,GHARR-1:Accra,Ghana 90% ^{235}U	18.8	-0.104	49	-0.0261
MNSR, Protyp. Beijing, China 90% ^{235}U	19.8	-0.009	58.5	+0.023
Slowpoke-2.DUSR.Halifax, Canada. 20%^{235}U	Site#2: 18.8 Site#3: 18.9	-0.0425 -0.0422	57.1	-0.0098
Slowpoke-2,EcolePolytechnic, Montreal, Canada. 20%^{235}U	19.8	-0.052	55.4	-0.006

In order to optimize utilization, additional neutron spectrum parameters such as the Maxwellian neutron temperature, T_n, the modified spectral index, ($r(\alpha)\ T_n/T_0$) and the ratio of thermal-to-fast neutron f_T were determined for NIRR-1 (Jonah et al, 2009) as shown in table 2.3

Table 2.3, Neutron spectrum parameters of four connected NIRR-1 irradiation channels determined at a steady state power level of 15.5kw

Parameters	Inner		Outer	
	A1	B2	A2	B4
α	-0.046±0.005	-0.052±0.002	0.024±0.002	0.029±0.003
f	18.4±0.3	19.2±0.5	49.5±0.1	48.3±3.3
$\frac{\phi_{th}}{\phi_f}(f_F)$	5.2±0.2	5.0±0.2	15.0±0.8	15.5±0.9
($r(\alpha)\ T_n/T_0$)	0.047±0.02	0.046±0.003	0.018±0.001	0.018±0.01
T_n(°C)	52.3±2.6	60.6±3.7	40.4±2.4	44.5±2.7

Results of f and α from table 2.3 show that the outer irradiation channels are better thermalized in comparison with the inner channels. This is because of the compact nature of the core, designed with relatively low critical mass and low value of HE ^{235}U atoms in the fuel lattice (Jonah et al, 2009). In the present work, the neutron spectrum parameters in Tables 2.2 and 2.3 for NIRR-1 were used, since the reactor is just in its first fuel cycle, the parameters are stable.

2.7 Uncertainty in RNAA

2.7.2. Uncertainty

The sources of standard uncertainty δ_I in RNAA can be grouped according to the individual steps of analysis into three categories as follows:

1. Sample and comparator preparation U_i
    a. Mass determination of a sample
    b. Mass determination of comparator
    c. Mass changes of samples due to moisture uptake during weighing

2. Irradiation, U_2
    a. Irradiation geometry differences
    b. Neutron self shielding / scattering differences
    c. Duration of irradiation
    d. Nuclear reaction interferences
    e. Neutron spectrum variations in time and space
    f. Volatilization losses during irradiation

3. gamma- ray spectrometry measurement, U_3

a. counting statistics

b. counting geometry differences

c. random coincidences

d. cascade summing

e. dead-time effects

f. decay-timing effects

g. duration of counting

h. peak integration method

i. γ-ray self-shielding (Kucera et al, 1999)

In this work, not all uncertainty sources were considered, since the contribution of some is negligible, However, the uncertainty sources ranging from neutron spectrum parameters (f and α) and γ- ray spectrometry (peak area analysis) have been identified.

The combined standard uncertainty δ_c can be calculated according to equation [2.22]

$$\delta_c = \sqrt{\frac{\sum U_i^2}{n}} \qquad 2.22$$

Where, U_i is the uncertainties of the individual uncertainty sources expressed as:

$$u\% = \frac{\delta_u}{U} * 100 \qquad 2.23$$

CHAPTER THREE

MATERIALS AND METHOD

3.1 Sample preparation

Synthetic Multi-elemental Standards (SMELS) type I and III as well as Coal Fly Ash NIST 1633b are the reference materials used for this work because they contain the nuclides of interest and also, they are the reference materials recommended for all k_0-users by IAEA.

The SMELS material was made based on a phenol formaldehyde resin (Bakelite) that was spiked with 33 different materials. It has been shown that SMELS material is unique material for the k_0-NAA users because of the following reasons:

- it can be used for short, medium, and long lived radio-isotopes
- It is a material with well characterized inhomogeneity with a between-bottle reproducibility of less than 1% for most elements
- It is radiation stable even at very high fluxes
- SMELS Type III contains Au and Zr, thus also allowing the direct determination of f and α.
- It can also be used as a quality control material to monitor the irradiation facility and the detector, thus replacing the traditional flux monitors.

NIST 1633b is a bituminous coal fly ash that was sieved through a nominal sieve opening of 90 μm (170 meshes) and then blended to assure homogeneity. This material is considered to be stable.

Accurately weighed amounts of samples and standard of approximately 0.2g for SMELS Type I, III and 0.1529g, 0.1856g for Coal fly ash were wrapped in polyethylene films, which have been cleaned appropriately. Two samples each were prepared for long and short irradiations.

3.2 Irradiation of samples

Irradiation of samples was done using Nigeria Research Reactor-1 (NIRR-1) operating at half full power of about 15kW with neutron flux 5×10^{11}n/cm^2.s. Two regimes of irradiation depending on the half - life of product radionuclide were adopted. The short irradiation was performed in the outer irradiation channel B_4, where the population of fast neutron flux is small thereby minimizing interference caused by threshold reactions with an irradiation time of 1 minute. For elements that produce long-lived nuclides, the irradiation was performed for 6 hours in the inner irradiation channel B_2, where the magnitude of the thermal neutron flux is highest.

3.3 Gamma- ray spectrometry measurement

After irradiation, induced activities in detector foils were measured on a GEM-30195 HPGe Coaxial, vertical dip-stick detector (Ortec), which has a relative efficiency of 30% and resolution of 1.95Kev at 1-33Mev, ^{60}Co. A MAESTRO emulation software compatible with Multi-channel Analyzer (MCA) was used for peak identification and evaluation. The efficiency curves of the detector system at near and far source-detector geometries have been determined by standard gamma-ray sources in the energy range of 59.5-1332 keV, the efficiency at any energy within the range is obtained by

interpolation and calculations. In the case of nuclides with energy outside the range (Ca-49 ; 3084kev) the efficiency curve was extended to 4000 keV by extrapolation

3.3.1 Counting of Irradiated Samples

Two gamma-ray measuring procedures were employed for the counting of short-lived nuclides. The first procedure, S_1 is performed at sample-detector geometry of 5cm for 10minutes, after a delay time of 2-15 minutes. The second procedure, S_2 is performed also 10minutes but a sample detector distance of 2cm following a waiting time of 3hours, 35minutes. For the long-lived nuclides, the gamma-ray measurements are performed at sample-detector geometry of 1cm for 30minutes after a waiting period of 5-6 days, which has been designated as protocol, L1. The second protocol, L2 gamma-ray measurement is performed at the same geometry for 1 hour, after a delay time of 9-10 days.

3.4. **Full energy peak efficiency calibration (ε_p)**

Standard gamma ray sources (Am^{241}, Eu^{152}, Ra^{226}, Cs^{137}, Co^{60}) were used to determine efficiencies at the different geometries. They were counted at 1cm and 5cm, for 1000sec and 1800sec.And the efficiency was calculated using equation [2.19]. A theoretical curve was then fitted to the experimental data from which the efficiency at any energy within the range were obtained by interpolation, and for energies outside the range, a trend line was developed using polynomial of order 2 to obtain efficiency of energies that are not within the range by extrapolation.

3.5 **Calculations**

The activation product when assayed by gamma-ray spectrometry, the net peak area (Np) corresponding to the photo peak, was calculated by summing the counts under the peak and subtracting the linear Compton background. The specific count rate (A_{sp}) for the activation product of the element of interest is given by,

$$A_{sp} = \frac{Np}{S.D.C.Wtc} \qquad 3.1$$

Where,

S = $1-e^{-\lambda t_{irr}}$, t_{irr} is the irradiation time

D = $e^{-\lambda t_d}$ = decay factor with t_d decay time

C = $1 - e^{-\lambda tc}$ = counting time correction with tc = counting time

$\lambda = \frac{ln2}{T_{1/2}}$ = decay constant

w = mass of the element

Similarly, the specific count rate for the comparator (gold) is written as

$$A_{sp.Au} = \left(\frac{Np.}{S.D.C.W.tc}\right)Au \qquad 3.2$$

The k_0- values were gotten by imputing the neutron flux parameters, full energy peak detection efficiency, and the specific activities obtained from equation [3.1) and [3.2] into equation [2.13] using MICROSOFT EXCEL.

The combine standard uncertainty in the experimental k_0-values was calculated by multiplying equation [2.22] with the k_0-values which is expressed as

$$\delta(k0) = k0\delta c \qquad \textbf{3.3}$$

Where,

The uncertainties sources in this work are due to the net peak count, f and α..

The result of this work was compared with the recommended data using the student "t-test" , the expression for the t-test is shown below:

$$t = \frac{\bar{d}}{S_d/\sqrt{n}} \qquad \textbf{3.4}$$

Where,

$\bar{d}$ = average difference given as: $\bar{d} = \frac{\sum d_i}{n}$ 3.5

$$S_d^2 = \frac{\sum (d_i - \bar{d})^2}{N-1} \qquad 3.6$$

The hypothesis for the t-test is stated as follows:

H_0: all $d_i = 0$; there is no difference between the measured k_0-value and the recommended values.

H_1 : all $d_i \neq 0$; there is a significant difference between the measured and the recommended values.

This is a null and alternative hypothesis, this means for the null hypothesis, when $t\text{-}_{cal.} > t_{critical}$ reject H_0 and for the alternative hypothesis, when $t\text{-}_{cal.} < t_{critical}$, accept H_0.

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 Efficiency Calibration

The full energy peak efficiency of the detector was determined using standard gamma-ray- sources such as ^{241}Am, ^{152}Eu, ^{226}Ra, ^{137}Cs, and ^{60}Co. The net peak counts were made on HPGe Coaxial Detector which has an energy resolution of 1.95keV at 1332.5keV belonging to ^{60}Co radio nuclide and relative efficiency of 30%. The associated electronics consist of an HV Power supply model 459, an amplifier model 485, a pulse generator model 480 and a NIM bin (all manufactured by ORTEC Industries Inc). The measured efficiencies using equation [2.19] for the energy range of 59.5-1332.5 and a source-to-detector distance of 1cm and 5cm are shown in Table, 4.1 and 4.2 respectively. Table, 4.3 shows the efficiency values for the nuclides whose k_0-values were measured. These values were evaluated by interpolation and extrapolation from the efficiency curve of 1cm and 5cm presented in fig. 4.1 and 4.2 respectively. The curve shows that efficiency decreases with increasing energy.

Table 4.1 determined efficiency at 1cm geometry

Radio nuclides	Energy (keV)	Peak counts (counts)	Live time (sec)	Count rate (count/s)	Initial act.(Bq)	Half life(sec)	Decay time (sec)	Decay const	B.Ratio (%)	Present Act. (Bq)	Emm. Rate	Efficency
Am-241	59.54	138,391	1,000	138.39	36,500	1.36E+10	1.69E+08	5.08E-11	0.3530	36,188.66	12,774.60	0.0108
Eu-152	121.78	419,662	1,000	419.66	38,000	4.26E+08	1.67E+08	1.63E-09	0.2821	28,942.08	8,164.56	0.0514
Ra-226	186.10	47,704	1,000	47.70	37,400	5.05E+10	1.69E+08	1.37E-11	0.0341	37,313.55	1,271.65	0.0375
Ra-226	241.90	79,447	1,000	79.45	37,400	5.05E+10	1.69E+08	1.37E-11	0.0708	37,313.55	2,642.55	0.0301
Eu-152	244.70	75,149	1,000	75.15	38,000	4.26E+08	1.67E+08	1.63E-09	0.0742	28,962.43	2,149.88	0.0350
Ra-226	295.20	183,192	1,000	183.19	37,400	5.05E+10	1.69E+08	1.37E-11	0.1810	37,313.55	6,753.75	0.0271
Eu-152	344.20	223,062	1,000	223.06	38,000	4.26E+08	1.67E+08	1.63E-09	0.2641	28,962.43	7,648.98	0.0292
Ra-226	351.90	344,135	1,000	344.14	37,400	5.05E+10	1.69E+08	1.37E-11	0.3510	37,313.55	13,097.06	0.0263
Eu-152	411.00	12,901	1,000	12.90	38,000	4.26E+08	1.67E+08	1.63E-09	0.0230	28,962.43	666.43	0.0194
Ra-226	609.30	240,872	1,000	240.87	37,400	5.05E+10	1.68E+08	1.37E-11	0.4426	37,313.78	16,515.08	0.0146
Cs-137	661.60	603,623	1,000	603.62	36,900	9.52E+08	1.67E+08	7.28E-10	0.8462	32,668.52	27,644.10	0.0218
Ra-226	768.40	16,181	1,000	16.18	37,400	5.05E+10	1.69E+08	1.37E-11	0.0474	37,313.55	1,768.29	0.0092
Ra-226	1120.30	41,967	1,000	41.97	37,400	5.05E+10	1.69E+08	1.37E-11	0.1469	37,313.55	5,481.36	0.0077
Co-60	1173.20	205,228	1,000	205.23	38,400	1.66E+08	1.67E+08	4.17E-09	0.9988	19,118.83	19,095.89	0.0107
Co-60	1332.50	186,581	1,000	186.58	38,400	1.66E+08	1.67E+08	4.17E-09	0.9998	19,118.83	19,115.01	0.0098

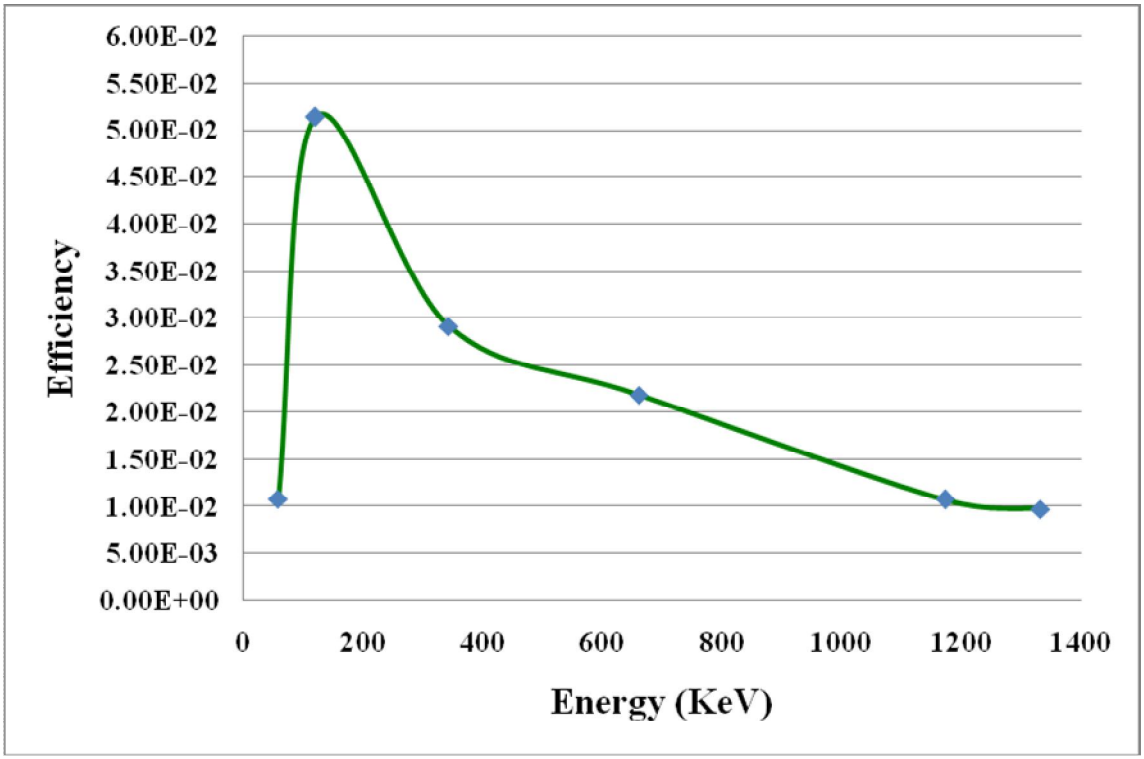


Fig 4.1. Efficiency at 1cm geometry

Table 4.2: determined efficiency at 5cm geometry

Radio nuclides	Energy (keV)	Peak counts (counts)	Live time (sec)	Count rate (count/s)	Initial act.(Bq)	Half life(sec)	Decay time (sec)	Decay const	B.Ratio (%)	Present Act. (Bq)	Emm. Rate	Efficency
Am-241	59.54	219,318	1800	43.86	36,500	1.36E+10	1.69E+08	5.08E-11	0.3530	36,188.66	12,774.82	0.0034
Eu-152	121.78	688,468	1800	137.69	38,000	4.26E+08	1.67E+08	1.63E-09	0.2821	28,942.08	5,359.90	0.0257
Ra-226	186.10	107,093	1800	21.42	37,400	5.05E+10	1.69E+08	1.37E-11	0.0341	37,313.55	1,271.65	0.0168
Ra-226	241.90	163,058	1800	32.61	37,400	5.05E+10	1.69E+08	1.37E-11	0.0708	37,313.55	2,642.56	0.0123
Eu-152	244.70	139,538	1800	27.91	38,000	4.26E+08	1.67E+08	1.63E-09	0.0742	28,962.43	1,410.37	0.0198
Ra-226	295.20	384,486	1800	76.90	37,400	5.05E+10	1.69E+08	1.37E-11	0.1810	37,313.55	6,753.79	0.0114
Eu-152	344.20	401,968	1800	80.39	38,000	4.26E+08	1.67E+08	1.63E-09	0.2641	28,962.43	5,017.90	0.0160
Ra-226	351.90	629,625	1800	125.93	37,400	5.05E+10	1.69E+08	1.37E-11	0.3510	37,313.55	13,097.12	0.0096
Eu-152	411.00	27,778	1800	5.56	38,000	4.26E+08	1.67E+08	1.63E-09	0.0230	28,962.43	437.19	0.0127
Ra-226	609.30	529,934	1800	105.99	37,400	5.05E+10	1.68E+08	1.37E-11	0.4426	37,313.78	16,515.06	0.0064
Cs-137	661.60	980,014	1800	196.00	36,900	9.52E+08	1.67E+08	7.28E-10	0.8462	32,668.52	15,612.39	0.0126
Ra-226	768.40	35,405	1800	7.08	37,400	5.05E+10	1.69E+08	1.37E-11	0.0474	37,313.55	1,768.30	0.0040
Ra-226	1120.30	108,644	1800	21.73	37,400	5.05E+10	1.69E+08	1.37E-11	0.1469	37,313.55	5,481.39	0.0040
Co-60	1173.20	412,656	1800	82.53	38,400	1.66E+08	1.67E+08	4.17E-09	0.9988	19,118.83	19,130.29	0.0043
Co-60	1332.50	385,765	1800	77.15	38,400	1.66E+08	1.67E+08	4.17E-09	0.9998	19,118.83	19,149.45	0.0040

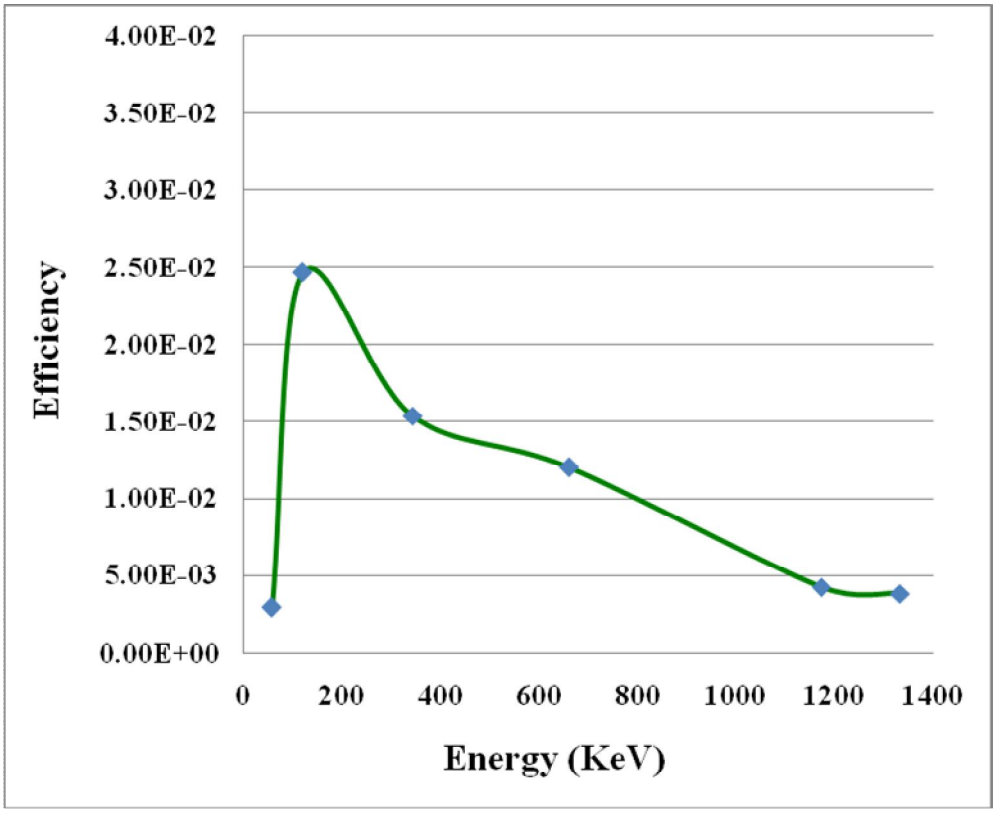


Fig. 4.2: Efficiency curve at 5cm geometry

Table 4.3: Determined efficiency values for nuclides whose k_0-values were re-Measured

Formed isotope	**Gamma energy**	Eff. @ 1cm	Eff. @ 5cm
Au-197			
	411.8	1.32E-02	3.18E-3
Ca-49(S1)	3084		1.20E-03
Ba-131(L2)	373	2.37E-03	
	486	2.49E-03	
	496	2.50E-03	
Cd-115(L2)	528	2.53E-03	
Sm-153(L1)	103		2.10E-03
Co-60m(L2)	1332	9.80E-03	
Se-75(L2)	121	5.10E-02	
	136	5.08E-02	
	401.73	2.60E-02	
In-116m(S1)	418.9		1.48E-02
	1097		5.50E-03
	1293		3.75E-03
In-116m2(S1)	162		2.55E-02
Cs-134m(L2)	127	5.20E-02	
Cs-134(L2)	562	2.40E-02	
	569	2.30E-02	
	602	2.20E-02	
	802	1.80E-02	

4.2 Calculation of k_0-values

Table 4.5, shows calculated k_0-values using equation [2.12] with nuclear data presented in Table 4.4. The calculated k_0-values were compared with recommended values, significant deviations ranging from -10.2% -98.7% were observed. This large

deviation demonstrates the unreliability of randomly chosen nuclear data reported by MOENS et al (1983).

4.3 Measured k_0-values

Table 4.6 and 4.7 present measured k_0-values from Coal Fly Ash and SMELS respectively, the Q_0-values used were taken from literature and corrected for non-ideality of epithermal neutron flux. Specific activities were computed by substituting the measured peak area into equation [3.1] and [3.2], the k_0-values were calculated by imputing the specific activities into equation [2.13] using MICROSOFT-EXCEL. For [^{49}Ca (3084keV)] the value was 1.10E-04. At gamma-ray energy of 124, 216keV and 620keV respectively for ^{131}Ba, no peak was identified because it was below detection limit, however at the energy of 373-496keV, the values were 1.95E-05, 3.43E-06, and 6.45E-05 respectively.

For SMELS : nuclides like [^{75}Se(121keV)], [^{116m2}In(162keV)], [^{134m}Cs(127keV)], [^{170}Tm (84keV)], and [^{60m}Co(59 and 1332keV)] were below detection limit. However, the long-lived isotope, ^{134}Cs at 569keV gave k_0-values of 7.13E-02 and 7.32E-02 for type I and III respectively while at 562keV, 604keV and 795.6keV, the values were 4.00E-02, 4.67E-01, and 3.93E-02 respectively .

Table 4.8 gives the comparison of the measured k_0-values with the recommended data and calculated values as well as their uncertainties. The major source of uncertainty in this work is that due to peak area analysis and from determination of overall, f and α values used. The results of the experimental values compare well with the recommended data for most nuclides except for ^{153}Sm whose

recommended k_0-value of 2.31E-1 deviate from the measured value of 1.66E-1, which represent a deviation of 28%. The reason is due to the discrepant nuclear data used in the calculation of k_0-value for this nuclide. However, the experimental values differ significantly with the calculated values showing how unreliably the randomly chosen nuclear data is.

Table 4.9 contains the statistical analysis of the measured k_0-values. A two-tailed student "t- test" was used to test for the level of significance; a confidence interval of 5% was chosen which represent $t_{0.025.}$ From the t-table (abridged from Table 3 of Fisher and Yates), for 5% level of significance and 14 degree of freedom (df), t is given as: $t_{0.025,14} = 2.145$, this value is greater than the calculated t-value of 0.038339, it was concluded that the result was not significant at 5% level. That is to say the null hypothesis is accepted.

Table 4.4: Nuclear data used for the calculation of k_0-values

Target nuclide	Formed isotope	$T_{1/2}$	E_γ(keV)	Ma	γ_a	θ_a	δ_a	Q_0
Cd-114	Cd-115	53.46	528	112.41	27.5	2.87E-1	2.30E-1	39.6
Cd-114	Cd-115m	4.486h	336	112.41	46.1	2.87E-1	3.60E-2	39.6
Ir-193	Ir-194	73.83d	296	192.22	28.3	3.73E-1	6.24E+2	3.5
			308	192.22	29.3	3.73E-1	6.24E+2	3.5
			316	192.22	83.1	3.73E-1	6.24E+2	3.5
			468	192.22	47.6	3.73E-1	6.24E+2	3.5
Hg-196	Hg-157	2.67d	77	200.59	18.2	1.40E-2	2.99E+3	0.5
Se-74	Se-75	119.8d	121	78.96	17.2	9.00E-3	5.04E+1	11.5
			136	78.96	59.0	9.00E-3	5.04E+1	11.5
			265	78.96	59.1	9.00E-3	5.04E+1	11.5
			279	78.96	25.2	9.00E-3	5.04E+1	11.5
			401	78.96	11.5	9.00E-3	5.04E+1	11.5
Gd-152	Gd-153	241.6d	97	157.25	27.5	2.00E-3	8.11E+2	0.8
			103	157.25	19.5	2.00E-3	8.11E+2	0.8
Gd-158	Gd-159	18.56h	363	157.25	11.0	2.48E-1	2.34E+00	31.9
Ba-132	Ba-131	11.5d	124	137.33	29.1	1.06E-3	7.54E+00	21.3
			134	137.33	2.2	1.06E-3	7.54E+00	21.3
			216	137.33	19.9	1.06E-3	7.54E+00	21.3
			373	137.33	13.3	1.06E-3	7.54E+00	21.3
			486	137.33	1.9	1.06E-3	7.54E+00	21.3
			496	137.33	43.7	1.06E-3	7.54E+00	21.3
			620	137.33	1.6	1.06E-3	7.54E+00	21.3
Pd-108	Pd-109	13.46h	88	106.42	3.7	8.66E+00	2.68E+1	26.8
Pd-108	Pd-109m	39.6s		106.42	3.7	2.00E-1	1.10E+1	11.0
Ag-109	Ag-110m	249.8d	658	107.87	94.6	4.82E-1	3.97E+00	16.0
			764	107.87	22.2	4.82E-1	3.97E+00	16.0
			884	107.87	72.6	4.82E-1	3.97E+00	16.0
			1384	107.87	24.3	4.82E-1	3.97E+00	16.0
In-115	In-116m	54.15m	417	114.82	30.0	9.57E-1	1.54E+2	16.8
			1097	114.82	56.2	9.57E-1	1.54E+2	16.8
			1293	114.82	85.1	9.57E-1	1.54E+2	16.8
In-115	In-116m2	2.18s	162	114.82	100.0	9.57E-1	1.54E+2	16.8
Cs-133	Cs-134m	2.91h	127	132.9054	12.6	1.00E-1	2.74E+00	11.8
Cs-133	Cs-134	2.062y	563	132.9054	8.4	1.00E-1	3.04E+1	12.7
			569	132.9054	15.4	1.00E-1	3.04E+1	12.7
			605	132.9054	97.5	1.00E-1	3.04E+1	12.7
			796	132.9054	85.3	1.00E-1	3.04E+1	12.7
			802	132.9054	8.7	1.00E-1	3.04E+1	12.7
Tm-169	Tm-170	128.6d	84	168.9342	3.3	1.00E-1	1.07E+2	14.5
Re-185	Re-186	90.64h	137	186.2070	8.5	3.74E-1	1.06E+2	15.4
Sm-152	Sm-153	46.5h	103	150.36	37.0	2.67E-1	2.20E+2	14.4
S-35	S-36	5.05m	3103	32.066	94.0	2.00E-4	1.60E-1	1.1
Ca-48	Ca-49	8.71m	3084	40.0780	92.1	1.87E-3	1.12E+00	0.5
Co-59	Co-60m	10.47m	59	59.9332	2.0	1.00E+00	2.00E+1	2.0
			1332	59.9332	0.3	1.00E+00	2.00E+1	2.0
U-238	U-239	23.54m	75	238.0289	50.0	9.93+1	2.71E+00	103.4

Table 4.5.Comparison of calculated k_0-values with recommended values as published by DE CORTE AND SIMONITS (2003)

Target nuclide	Formed isotope	$T_{1/2}$	E_γ(keV)	Cal. k_0-values	Recom. values	% dev.
Csd-114	Cd-115	53.46	528	4.27E-4	4.77E-4	10.5
Cd-114	Cd-115m	4.486h	336	1.12E-4	7.73E-4	85.5
Ir-193	Ir-194	73.83d	296	9.06E-1	NA	
			308	9.38E-1	NA	
			316	2.66E+00	NA	
			468	1.52E+00	NA	
Hg-196	Hg-157	2.67d	77	1.00E-1	NA	
Se-74	Se-75	119.8d	121	2.61E-3	1.94E-3	-34.6
			136	8.96E-3	6.76E-3	-32.5
			265	8.97E-3	7.11E-3	-26.2
			279	3.83E-3	3.00E-3	-27.6
			401	1.75E-3	1.43E-3	-22.1
Gd-152	Gd-153	241.6d	97	7.50E-3	5.86E-3	-27.9
			103	5.32E-3	4.21E-3	-26.3
Gd-158	Gd-159	18.56h	363	1.07E-3	8.49E+0	-26.4
Ba-132	Ba-131	11.5d	124	4.48E-5	3.90E-5	-14.8
			134	3.35E-6	3.24E-6	-35.0
			216	3.06E-5	2.75E-5	-11.3
			373	2.05E-5	1.92E-5	-65.6
			486	2.89E-6	3.44E-6	15.9
			496	6.72E-5	6.48E-5	-37.4
			620	2.42E-6	2.34E-6	-32.1
Pd-108	Pd-109	13.46h	88	2.12E-3	NA	NA
Pd-108	Pd-109m	39.6s		4.91E-5	1.71E-3	97.1
Ag-109	Ag-110m	249.8d	658	4.44E-2	3.50E-2	-26.7
			764	1.04E-2	8.27E-3	-25.9
			884	3.40E-2	2.69E-2	-26.5
			1384	1.14E-2	9.12E-3	-24.9
In-115	In-116m	54.15m	417	1.02E+00	7.54E-1	98.7
			1097	1.91E+00	1.60E+0	-19.2
			1293	2.89E+00	2.29E+0	-26.1
In-115	In-116m2	2.18s	162	3.39E+00	NA	NA
Cs-133	Cs-134m	2.91h	127	6.87E-4	5.48E-3	87.5
Cs-133	Cs-134	2.062y	563	5.07E-2	4.14E-2	-22.5
			569	9.31E-2	7.34E-2	-26.9
			605	5.89E-1	4.76E-1	-23.8
			796	5.16E-1	4.15E-1	-24.3
			802	5.28E-2	4.11E-2	-28.4
Tm-169	Tm-170	128.6d	84	3.46E-2	3.26E-2	-67.4
Re-185	Re-186	90.64h	137	4.77E-2	4.33E-2	-10.2
Sm-152	Sm-153	46.5h	103	3.82E-1	2.31E-1	-65.4
S-35	S-36	5.05m	3103	2.48E-6	1.96E-6	-26.5
Ca-48	Ca-49	8.71m	3084	1.27E-4	1.01E-4	-26.0
Co-59	Co-60m	10.47m	59	1.80E-2	1.51E-2	-19.2
			1332	2.21E-3	1.75E-3	-26.0
U-238	U-239	23.54m	75	1.49E+00	1.33E-2	-11.1

NA: not available

Table 4.6: Experimental k_0-values using from Coal Fly ash (1633b)

Formed isotope	Energy	Q_0	α	$\overline{E_r^a}$	$Q_{0,a}(\alpha)$	Q_0, Au(α)	F	A_{sp}	$A_{sp.Au}$	ε_p .Au	ε_p .a	$k_{0.Au}(a)$
	(kev)									3.18E-3		
Ca-49(S1)	3084	0.45	0.029	1.131071	0.41708	16.972	48.3	3.97E+05	4.37E+08	3.18E-3	1.20E-03	1.10E-04
Ba-131(L2)	124	24.8	-0.05	0.801839	30.8878	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
	134	24.8	-0.05	0.801839	30.8878	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
	216	24.8	-0.05	0.801839	30.8878	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
	373	24.8	-0.05	0.801839	30.8878	16.972	19.2	1.16E+04	4.36E+08	1.32E-2	2.37E-03	1.95E-05
	486	24.8	-0.05	0.801839	30.8878	16.972	19.2	2.14E+03	4.36E+08	1.32E-2	2.49E-03	3.43E-06
	496	24.8	-0.05	0.801839	30.8878	16.972	19.2	4.03E+04	4.36E+08	1.32E-2	2.50E-03	6.45E-05
	620	24.8	-0.05	0.801839	30.8878	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
Cd-115(L2)	528	32.4	-0.05	0.801839	40.366	16.972	19.2	3.54E+05	4.36E+08	1.32E-2	2.53E-03	4.70E-04
Hg-197(L2)	77	0.49	-0.05	0.801839	0.56999	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
Gd-153(L2)	97	0.77	-0.05	0.801839	0.91919	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
	103	0.77	-0.05	0.801839	0.91919	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
Sm-153(L1)	103	14.4	-0.05	0.801839	17.9176	16.972	19.2	6.49E+07	4.36E+08	3.18E-3	2.10E-03	1.66E-01
Tm-170(L2)	84	13.7	-0.05	0.801839	17.0446	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
U-239	75	103.4	-0.05	0.801839	128.912	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
Co-60m	59	2	-0.05	8.02E-01	2.45316	16.972	19.2	BDL	1.70E+04	1.32E-2		BDL
	1332	2	-0.05	8.02E-01	2.45316	16.972	19.2	BDL	4.36E+08	1.32E-2	BDL	BDL

BDL (below detection limit)

Table 4.7: k_0-values from SMELS type I and III

Target nuclide	**Gamma energy**	**Q_0**	**A**	E_γ	$\overline{E_\gamma}$	$Q_{0,a}(\alpha)$	**Q_0, Au(α)**	f	A_{sp}	$A_{sp.Au}$	ε_{p} .Au	ε_{p} .a	$k_{0.Au}(a)$
Type III													
Se-75	121	10.8	-0.052	29.4	8.39E-01	1.29E+01	1.69E+01	19.2	BDL		1.32E-2	5.10E-02	BDL
	136	10.8	-0.052	29.4	8.39E-01	1.29E+01	1.69E+01	19.2	1.04E+02	6.73E+02	1.32E-2	5.08E-02	6.88E-03
	401.73	10.8	-0.052	29.4	8.39E-01	1.29E+01	1.69E+01	19.2	2.09E+01	6.73E+02	1.32E-2	2.60E-02	1.38E-03
In-116m	418.9	16.8	0.029	1.56	1.01E+00	1.66E+01	1.69E+01	48.3	5.09E+04	6.73E+02	3.18E-03	1.48E-02	7.27E-01
	1097	16.8	0.029	1.56	1.01E+00	1.66E+01	1.69E+01	48.3	1.08E+05	6.73E+02	3.18E-03	5.50E-03	1.54E+00
	1293	16.8	0.029	1.56	1.01E+00	1.66E+01	1.69E+01	48.3	1.60E+05	6.73E+02	3.18E-03	3.75E-03	2.28E+00
In-116m2	162	16.8	0.029	1.56	1.01E+00	1.66E+01	1.69E+01	48.3	BDL	6.73E+02	3.18E-03	2.55E-02	BDL
Cs-134m	127	11.8	-0.052	9.27	8.91E-01	1.33E+01	1.69E+01	19.2	BDL	6.73E+02	1.32E-02	5.20E-02	BDL
Cs-134	562	12.7	-0.052	9.27	8.91E-01	1.43E+01	1.69E+01	19.2	6.30E+02	6.73E+02	1.32E-02	2.40E-02	4.00E-02
	569	12.7	-0.052	9.27	8.91E-01	1.43E+01	1.69E+01	19.2	1.15E+03	6.73E+02	1.32E-02	2.30E-02	7.32E-02
	602	12.7	-0.052	9.27	8.91E-01	1.43E+01	1.69E+01	19.2	7.37E+03	6.73E+02	1.32E-02	2.20E-02	4.67E-01
	802	12.7	-0.052	9.27	8.91E-01	1.43E+01	1.69E+01	19.2	6.20E+02	6.73E+02	1.32E-02	1.80E-02	3.93E-02
Tm-170	84	13.7	-0.052	4.8	9.22E-01	1.49E+01	1.69E+01	19.2	BDL	6.73E+02	1.32E-02	BDL	BDL
Co-60m	59	2	-0.052	136			1.69E+01	19.2	BDL	6.73E+02	1.32E-02	BDL	BDL
	1332	2	-0.052	136	7.75E-01	2.52E+00	1.69E+01	19.2	BDL	6.73E+02	1.32E-02	9.80E-03	BDL
Type I													
Cs-134	569	12.7	-0.052	9.27	8.91E-01	1.43E+01	1.69E+01	19.2	2.89E+02	1.73E+02	1.32E-02	2.30E-02	7.13E-02

Table 4.8: Comparison of experimental values and calculated k_0-values with recommended values

Formed Isotope	Energy (kev)	**Recom. k_0-values**	**Expt. k_0-values**	**%dev**	**cal. k_0-values**	**Expt. k_0-values**	**%dev**
Ca-49(S1)	3084	(1.01±0.9)E-04	(1.10±1.4)E-04	-8.7	1.27E-04	(1.10±1.4)E-04	-15.6
Ba-131(L2)	124		BDL			BDL	
	134		BDL			BDL	
	216		BDL			BDL	
	373	(1.92±0.4)E-05	(1.95±0.6)E-05	-1.7	2.05E-05	(1.95±0.6)E-05	-5.0
	486	3.44E-06	(3.43±0.2)E-06	0.2	2.89E-06	(3.43±0.2)E-06	15.8
	496	(6.48±0.2)E-05	(6.45±0.2)E-05	0.5	6.72E-05	(6.45±0.2)E-05	-4.3
	620		BDL			BDL	
Cd-115(L2)	528	(4.77±1.3)E-04	(4.70±1.3)E-04	1.5	4.27E-04	(4.70±1.3)E-04	9.1
Hg-197(L2)	77		BDL			BDL	
Gd-153(L2)	97		BDL			BDL	
	103		BDL			BDL	
Sm-153(L1)	103	(2.31±0.4)E-01	(1.66±0.1)E-01	28.0	3.82E-01	(1.66±0.1)E-01	-130.0
Tm-170(L2)	84		BDL			BDL	
U-239	75		BDL			BDL	
Co-60m(L2)	59	1.51E-02	BDL		1.80E-02	BDL	
	1332	(1.75±1.4)E-03	BDL		2.21E-03	BDL	
Se-75(L2)	121	1.94E-03	BDL		2.61E-03	BDL	
	136	(6.76±1.1)E-03	(6.88±1.1)E-03	-1.9	8.96E-03	(6.88±1.1)E-03	-30.1
	401.73	(1.43±0.8)E-03	(1.38±0.7)E-03	3.2	1.75E-03	(1.38±0.7)E-03	-26.4
In-116m(s1)	418.9	(7.54±1.1)E-01	(7.27±0.9)E-01	3.6	1.02E+00	(7.27±0.9)E-01	-40.3
			(1.54±1.2)E+0	4.0			-34.3
	1097	(1.60±1.3)E+00	0		1.91E+00	(1.54±1.2)E+00	
	1293	2.29±0.8	2.28E+00	0.3	2.89E+00	2.28E+00	-26.6
In-116m2(s1)	162	NA	BDL		3.39E+00	BDL	
Cs-134m(L2)	127	(5.48±1.7)E-03	BDL		6.87E-04	BDL	
Cs-134(L2)	562	(4.14±1.7)E-02	(4.00±1.4)E-02	3.4	5.07E-02	(4.00±1.4)E-02	-26.8
	569	(7.34±1.5)E-02	(7.32±1.5)E-02	0.3	9.31E-02	(7.32±1.5)E-02	-27.2
	602	(4.76±2)E-01	(4.67±1.8)E-01	1.8	5.89E-01	(4.67±1.8)E-01	-26.0
	802	(4.11±2)E-02	(3.93±1.8)E-02	4.3	5.28E-02	(3.93±1.8)E-02	-34.2
Tm-170(L2)	84	3.26E-02	BDL		5.46E-02	BDL	

Table 4.9: Comparison of k_0-values of this work with recommended data using "t-test"

	Mean	N	Df	St. Deviation	Variance	T
Expt.k_0-Rec.k_0-values	3360	15	14	175279.8	350559.5	0.038339
Expt. Unc-Rec. Unc.	-0.8	15	14	0.01419	0.028381	-112.752

In Table 4.10, the k_0-values from this work were compared with the measured and re-measured k_0-values by St-Pierre and Kennedy (2003, 2006), their uncertainties were higher than that of this work for almost all the nuclides except Cs^{134} at gamma line of 602 keV and 802keV. The reason for the difference may be mostly coming from the type of detector used in each case. These uncertainties are however in agreement with recommended uncertainties in Table 4.8 discussed earlier. This shows the accuracy of the result obtained in this work which is in agreement with the recommendation of (MOENS et al, 1983) that uncertainties could be reduced by more accurate measurements.

Table 4.10: Measured k_0-values compared with literature values by St-Pierre and Kennedy

formed isotope	T1/2	Energy (keV)	2003	2006	This work
Cd-115	53.46h	527.9	6.59E-04	(4.49±1.5) E-04	4.70E-04(1.3)
In-116m	54.15m	417		(7.86±1.7)E-01	7.27E-01(0.9)
		1097	1.73E+00	(1.66±1.9)E+00	1.54E+00(1.2)
		1293		(2.40±1.9)E+00	2.28E+00(0.7)
Cs-134	2.062yr	562		(3.93±1.7)E-02	4.00E-02(1.6)
		569		(7.11±1.7)E-02	7.32E-02(1.5)
		602		(4.57±1.7)E-01	4.67E-01(1.8)
		802		(4.11±1.9)E-02	3.93E-02(1.9)
Ba-131	11.53d	373		(1.81±)E-05	1.95E-05(0.6)
		496		(6.19±1.4)E-05	6.45E-05(0.2)

CHAPTER FIVE

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

In order to determine the k_0-values, the full energy peak efficiency at 1cm and 5cm geometry of the HPGe detector was determined using gamma ray sources, and an efficiency curve at the respective geometries were fitted to obtain the efficiencies of the discrepant nuclides.

The k_0 –values of the of the following nuclides (Ca-48, Ba-131, Cd-115, In-116m, Cd-116m2, Co-60, Cs-134, Ir-192, Hg-197, Se-75, Gd-153, Gd-159, Pd-109, Ag-110m, Tm-170, Re-186, Sm-153, S-36, U-239) suspected to be discrepant were calculated using randomly chosen nuclear data, results obtained shows much deviation when compared with recommended data. This shows how unreliable M, θ_0, σ_0, and γ. The k_0-values of the following discrepant nuclides; Ca-49, Ba-131, Cd-115, In-116, Sm-152 , Se-75, Cs-134 were also re-measured using Certified Reference Materials, (SMELS I, III and NIST 1633b Coal fly Ash), result obtained show very good agreement with the recommended data, which shows the accuracy of the method and facilities used for this work.

To further support the accuracy of this measurements a statistical analysis (two tailed student t-test) was used to compare the results of this work with the recommended data. The result at 5% confidence interval affirms the null hypothesis. Meaning there is

no significance difference with the re-measured k_0 –values and the recommended data. This shows the usefulness of the k_0-standardisation method adopted in NIRR-1 Laboratory.

5.2 Recommendation

Based on the discrepancies observed in the calculated k_0-vaues there is need to update the nuclear data for M, θ_0, σ_0, and γ. Also considering the accuracy of the re-measured k_0-values of this work can be added to the data library of NIRR-1. Further investigations should be conducted to determine k_0 –values of ^{153}Sm at gamma-ray energy of 103.5keV and other nuclides on the recommendations of the International k_0-user Community.

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APPENDIX

APPENDIX A: Determined specific activity from measured net peak counts (1633b)

Formed Isotope	Energy (kev)	Net peak	T1/2 (sec)	Td (sec)	γ	tc (sec)	t_{irr}(sec)	W	C	S	D	Asp
Ca-49(S1)	3084	63725	522.6	660	1.33E-3	600	60	0.1529	5.49E-1	7.65E-2	4.17E-1	3.97E+4
Ba-131(L2)	124	BDL	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	BDL
	134	BDL	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	BDL
	216	BDL	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	BDL
	373	35160	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	1.16E+4
	486	26429	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	2.10E+3
	496	55603	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	4.03E+4
	620	BDL	993600	854940	6.97E-7	3600	21600	0.1856	2.51E-3	1.50E-2	5.51E-1	BDL
Cd-115(L2)	528	10482	192456	854940	3.60E-6	3600	21600	0.1856	1.29E-2	7.48E-2	4.60E-2	3.54E+5
Hg-197(L2)	77	BDL	85680	854940	8.09E-6	3600	21600	0.1856	2.87E-2	1.60E-1	9.93E-4	
Gd-153(L2)	97	BDL	2.1E+07	854940	3.34E-8	3600	21600	0.1856	1.20E-4	7.20E-4	9.72E-1	
	103	BDL	2.1E+07	854940	3.34E-8	3600	21600	0.1856	1.20E-4	7.20E-4	9.72E-1	
Sm-153(L1)	103	172146	167400	494940	4.14E-6	1800	21600	0.1856	7.42E-3	8.55E-2	1.29E-1	6.30E+6
Tm-170(L2)	84	BDL	1.1E+07	854940	6.24E-8	3600	21600	0.1856	2.25E-4	1.35E-3	9.48E-1	BDL
U-239	75	BDL	1407	494940	4.93E-4	3600	21600	0.1856	8.30E-1	1.00E+0	1.35E-6	BDL
Au-198	411.8	20159	232848	494940	2.98E-6	3600	21600	0.1856	1.07E-2	6.23E-2	2.29E-1	2.55E+3
Co-60	59	BDL	6282	854940	1.10E-4	3600	21600	0.1856	3.28E-1	9.08E-1	1.10E-41	BDL
	1332	BDL	6282	854940	1.10E-4	3600	21600	0.1856	3.28E-1	9.08E-1	1.10E-41	BDL

APPENDIX B: Determined specific activity from measured net peak counts

(SMELS, Type III)

Formed nuclide	E(kev)	Npa	T1/2	td	λ	tc	t_{irr}(sec)	W	C	S	D	Asp
Se-75(L2)	121	BDL	10350720	864000	6.70E-8	3600	21600	0.2	2.41E-04	1.45E-03	9.44E-01	BDL
	136	4.17E+04	10350720	864000	6.70E-8	3600	21600	0.2	2.41E-04	1.45E-03	9.44E-01	1.04E+02
	401.73	3.42E+06	10350720	864000	6.70E-8	3600	21600	0.2	2.41E-04	1.45E-03	9.44E-01	2.09E+01
In-116m(s1)	418.9	7.16E+05	3249	120	2.13E-4	600	60	0.2	1.20E-01	1.27E-02	9.75E-01	5.09E+04
	1097	1.51E+06	3249	120	2.13E-4	600	60	0.2	1.20E-01	1.27E-02	9.75E-01	1.08E+05
	1293	2.25E+06	3249	120	2.13E-4	600	60	0.2	1.20E-01	1.27E-02	9.75E-01	1.60E+05
In-116m2(s1)	162	BDL	2.18	120	3.18E-1	600	60	0.2	1.00E+00	1.00E+00	2.67E-17	BDL
Cs-134m(L2)	127	BDL	10476	864000	6.62E-5	3600	21600	0.2	2.12E-01	7.60E-01	1.51E-25	BDL
Cs-134(L2)	562	6.13E+04	65027232	864000	1.07E-8	3600	21600	0.2	3.84E-05	2.30E-04	9.91E-01	6.30E+02
	569	3.63E+04	65027232	864000	1.07E-8	3600	21600	0.2	3.84E-05	2.30E-04	9.91E-01	1.15E+03
	602	2.07E+03	65027232	864000	1.07E-8	3600	21600	0.2	3.84E-05	2.30E-04	9.91E-01	7.37E+03
	802	1.75E+03	65027232	864000	1.07E-8	3600	21600	0.2	3.84E-05	2.30E-04	9.91E-01	6.20E+02
Tm-170(L2)	84	BDL	11111040	864000	6.24E-8	3600	21600	0.2	2.25E-04	1.35E-03	9.48E-01	BDL
Co-60m(L2)	59	BDL	6282	864000	1.10E-4	3600	21600	0.2	3.28E-01	9.08E-01	4.04E-42	BDL
	1332	BDL	6282	864000	1.10E-4	3600	21600	0.2	3.28E-01	9.08E-01	4.04E-42	BDL

APPENDIX C: percentage points of the t distribution(abridged from Table III of Fisher and Yates)

v/α	0.25	0.2	0.15	0.1	0.05	0.025	0.01	0.005	0.0005
1.00	1.00E+00	1.38E+00	1.96E+00	3.08E+00	6.31E+00	1.27E+01	3.18E+01	6.37E+01	6.37E+02
2.00	0.816	1.061	1.386	1.886	2.92	4.303	6.965	9.925	31.598
3.00	7.65E-01	9.78E-01	1.25E+00	1.64E+00	2.35E+00	3.18E+00	4.54E+00	5.84E+00	1.29E+01
4.00	0.741	0.941	1.19	1.533	2.132	2.776	3.747	4.604	8.61E+00
5.00	0.727	9.20E-01	1.156	1.48E+00	2.015	2.57E+00	3.365	4.03E+00	6.859
6.00	0.718	9.06E-01	1.134	1.44E+00	1.943	2.45E+00	3.143	3.71E+00	5.959
7.00	0.711	0.896	1.119	1.415	1.895	2.365	2.998	3.499	5.405
8.00	0.706	8.89E-01	1.108	1.40E+00	1.86	2.31E+00	2.896	3.36E+00	5.041
9.00	0.703	0.883	1.1	1.383	1.833	2.262	2.821	3.25	4.781
10.00	0.7	8.79E-01	1.093	1.37E+00	1.812	2.23E+00	2.764	3.17E+00	4.587
11.00	0.697	0.88	1.088	1.36E+00	1.796	2.20E+00	2.718	3.11E+00	4.437
12.00	0.695	0.873	1.083	1.356	1.782	2.179	2.681	3.055	4.318
13.00	0.694	0.87	1.079	1.35E+00	1.771	2.16E+00	2.65	3.01E+00	4.221
14.00	0.692	0.87	1.076	1.345	1.761	2.145	2.624	2.977	4.14
15.00	0.691	0.87	1.074	1.34E+00	1.753	2.13E+00	2.602	2.95E+00	4.073
16.00	0.69	0.87	1.071	1.34E+00	1.746	2.12E+00	2.583	2.92E+00	4.015
17.00	0.689	0.86	1.069	1.333	1.74	2.101	2.567	2.898	3.965
18.00	0.688	0.86	1.067	1.33E+00	1.734	2.09E+00	2.552	2.88E+00	3.922
19.00	0.688	0.86	1.066	1.328	1.729	2.086	2.539	2.861	3.883
20.00	0.687	0.86	1.064	1.33	1.725	2.09E+00	2.528	2.85E+00	3.85
21.00	0.686	0.86	1.063	1.323	1.721	2.08E+00	2.518	2.83E+00	3.819
22.00	0.686	0.86	1.061	1.32E+00	1.717	2.074	2.508	2.819	3.792
23.00	0.685	0.86	1.06	1.319	1.714	2.07E+00	2.5	2.81E+00	3.767
24.00	0.685	0.86	1.059	1.32E+00	1.711	2.064	2.492	2.397	3.745
25.00	0.684	0.86	1.058	1.316	1.708	2.06E+00	2.485	2.79E+00	3.725
26.00	0.684	0.86	1.058	1.32E+00	1.706	2.06E+00	2.479	2.78E+00	3.707
27.00	0.684	0.86	1.057	1.314	1.703	2.052	2.473	2.771	3.69
28.00	0.683	0.86	1.056	1.31E+00	1.701	2.05E+00	2.467	2.76E+00	3.674
29.00	0.683	0.65	1.055	1.311	1.699	2.045	2.462	2.756	3.659
30.00	0.683	0.85	1.055	1.31E+00	1.697	2.04E+00	2.457	2.75E+00	3.646
40.00	0.681	0.85	1.05	1.30E+00	1.684	2.02E+00	2.423	2.70E+00	3.551
60.00	0.679	0.848	1.046	1.296	1.671	2	2.39	2.66	3.46
120.00	0.677	0.85	1.041	1.29E+00	1.658	1.98E+00	2.358	2.62E+00	3.373
∞	0.674	0.842	1.036	1.282	1.645	1.96	2.326	2.576	3.291