CHARACTERIZATION OF THE MODERATED NEUTRON FIELD AROUND AN ISOTOPIC Am-241/Be NEUTRON SOURCE

BY

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DECLARATION

I declare that this thesis entitled "Characterization of the moderated neutron field around an Isotopic Am-241/Be neutron source", is my own work and has not been submitted in any form for another degree or diploma to any other University or institution. Information derived from published and unpublished work of other workers has been acknowledged in the text.

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CERTIFICATION

This thesis entitled "Characterization of the neutron field around an Isotopic Am-241/Be neutron source" by Mr. Amidu Olalekan MUSTAPHA meets the regulations governing the award of the degree of Master of Science of Ahmadu Bello University, and is approved for its contribution to knowledge and literary presentation.

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DEDICATION

To all friends and relations

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ABSTRACT

The spatial variation of the neutron field in the cylindrical cavity above a 5-curie Am-241/Be source, installed in a paraffin wax 'howitzer' has been characterised using the neutron activation analytical method. The neutron flux monitors used are aluminium and indium foils. A third order polynomial was obtained from a fit of the axial neutron flux distribution curve, and the application of this polynomial, in estimating the average neutron flux within a bulk sample that is being irradiated, was discussed. For the sake of comparison, a theoretical estimation of the neutron flux distribution was also carried out, using the elementary diffusion equation. Finally, the work discussed the unique application of isotopic neutron sources in quality control, using the fast neutron thermalisation method.

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CHAPTER ONE

INTRODUCTION

Since its conclusive discovery by James Chadwick in 1932, the neutron has proved to be a unique tool for investigating the structures of nuclei, atoms, molecules, solids, and liquids. Its characteristics, which include overall neutrality, radioactivity, and possession of magnetic moment, contribute interest to its study as well as its use at a more practical level. For example, at thermal energies, the wavelength of the neutron wave is such that it is mainly the atomic nuclei which act as its scatterers. Thus neutrons are being used (Bacon, 1969)to study the innermost structures of materials.

One of the major applications of neutrons in the study of materials is neutron activation analysis (NAA), developed by G. Von Hevesy and H. Levy in 1936 (Garg and Batra, 1986) and it was first used in the analysis of Dysprosium, in rare earths, using an isotopic neutron source (IAEA, Isotopic, 1988). Since then NAA has been widely used in various fields such as geology, medicine, agriculture, electronics, metallurgy, criminology, and petroleum industry (Garg and Batra, 1986). When NAA is compared with other instrumental analysis such as gravimetric,

calorimetric, spectrographic, or mass spectrometry, its sensitivity is usually shown to be better, by a factor of 10, than most other methods (EG & G ORTEC, 1984).

In the light of its numerous uses and applications, it becomes necessary to produce neutrons in large quantities, since they do not exist freely, for long, in nature. For example, neutrons half life against beta decay is 11.7 minutes (Beckutz and Wirtz, 1964). The Nuclear Reactors provide high intensities of neutrons, which, in turn, lead to high sensitivity in analytical results. In this respect, nuclear reactors are closely followed by the Neutron Generators. These large and high intensity neutron sources are, however, very expensive and, as such, they are usually out of reach of many universities and research centres, especially in the developing countries (IAEA Isotopic, 1988). The isotopic neutron sources provide a cheaper means of neutron production, although with flux outputs many orders of magnitudes below what is obtainable from reactors and generators. Again, the isotopic neutron sources have certain intrinsic advantages over the other types of neutron sources (Garg and Batra, 1986), some of which have some unique applications. One of such is in the area of process or quality control of certain industrial products, such as liquids flowing in pipes (Ceichanowski et al, 1988), and bulk materials moving

on conveyor belts.

NAA with isotopic neutron sources is mostly carried out with thermal neutrons using the radioactive absorptions (n, γ) reaction, although fast and epithermal neutrons, are also being used (IAEA Isotopic, 1988). Since isotopic neutron sources are originally fast neutron sources, the fast neutrons from them are often moderated to thermal energies by installing the sources in moderating media usually made up of hydrogenous materials like water, paraffin, polyethelene, etc. A special moderating geometry designed with a view of producing a large flux of thermal neutrons, preferably in one direction, is called the "Howitzer" (Fink, 1936 and Cox et al, 1968). In the present work, thermalization of the fast neutrons from the 5 Curie Am/Be source was achieved by a nearly 4π immersion of the source in its transportation container made of paraffin wax. This container, after modifications (with wax and perspex) partly based on the specification of Cox et al (1968), now constitute a neutron Howitzer.

One of the inherent problems of the neutron fields obtainable around an isotopic neutron source which is surrounded by moderating material, is the existence of (axial and radial) flux gradients within the moderating medium (irradiation region). Therefore, for any meaningful activation analysis (absolute or

relative), there is need for proper characterization of the neutron field around the source, so that the actual neutron flux experienced by an irradiated sample is known (in case of absolule NAA), or, the ratio of the fluxes in the sample to that in the standard during irradiation is also known, for comparison, in the case of relative NAA.

The neutron field around the Am/Be source used in the present work was characterized by monitoring the neutron flux variation within the irradiation cavity using the foil activation method.

A major impetus to the neutron field characterization as carried out in this work, is the need for a means of estimating the average neutron flux experienced by a bulk sample at a specified geometry during irradiation for the purpose of absolute or relative analysis. In this regard, a relation was obtained which, when evaluated, could yield an approximate value of the average neutron flux within a bulk sample of known dimension. A theoretical calculation of the thermal flux distribution was also carried out using the two group diffusion theory for a point source of fast neutrons (Beckurts and Wirtz, 1964).

The possibility of adapting the characterised irradiating facility to carry out some of the unique applications, notable of isotopic neutron sources (Garg and Batra, 1986), especially the process control, and the on-stream analysis of refinery products, was considered. The detecting and counting instrumentation used in this aspect of the work consisted of a 2" x 2" NaI (Tl) sodium iodide detector coupled with a Multichannel Analyser, bearing in mind the need for compactness and easy mobility.

CHAPTER TWO

CHARACTERISTICS OF NEUTRONS

2.1 FUNDAMENTAL PROPERTIES OF NEUTRONS

CHARGE

The neutron is assumed to be entirely neutral, though experimental observation of a spherically symmetrical scattering of thermal neutrons by Xenon gas, made by Fermi and Marshall (Thewlis, 1961), have been interpreted as resulting from a net neutron charge of about 10^{-18} electron charge.

MAGNETIC MOMENT

The experimentally measured value of the neutron's Magnetic moment is μ = 1.913148 +/- 0.000066 NM (Nuclear Magnetons). It is the fact that neutron possesses a magnetic moment, which could only result from some circulation of charged particles, that necessitated the consideration that for a fraction of time the neutron could exist as a concentrated positive charge surrounded at a relatively large distance by a cloud of equal negative charge (Bacon, 1969 and Morrison, 1951). Thus the assumption of zero net charge of neutrons could indicate an equal number of positive and negative charges.

STABILITY

A free neutron itself is unstable, it undergoes beta decay into an electron, a proton, and a neutrino. (Beckurts and Wirtz, 1964).

$$n \longrightarrow P + \beta^{-} + \bar{\nu} \qquad \text{..... 2.1}$$

With a beta energy $E_{\beta} = 782$ KeV

This could however be considered as long lived in light of the types of processes in which the neutrons are involved, and it is only in this regard that the neutron could be considered to be a stable particle.

MASS

Neutron's mass of 1.008665u(1.674663×10^{-24}g) is slightly more than that of proton which is 1.007276u (1.672357×10^{-24}g) (beckurts and Wirtz, 1964). The values were obtained by indirect methods, one of such methods is the measurement of the gamma ray emitted when slow neutrons are captured in hydrogen. This gives the binding energy (E_d) of the deuteron formed and consequently its mass (M_d), and from the known mass of hydrogen (M_p), the neutron mass (M_n) is determined. These are related by the equation;

$$M_d = M_n + M_p - E_d/C^2 \qquad \text{..... 2.2}$$

where $E_d = 2.25$ MeV.

SPIN

Neutron spins about an axis through its centre of mass with an angular momentum of $1/2 \times \frac{h}{2}$. It is thus ascribed a spin of 1/2 . Neutron is therefore a Fermion (Bacon, 1969; and Beckurts and Wirtz, 1964).

2.2 CLASSIFICATION OF NEUTRONS

Neutrons are broadly classified into three major groups according to their energies.

i) Slow or low Energy neutrons ($E_n < 1$ KeV)

ii) Intermediate energy neutrons (1 KeV $< E_n <$ 500 KeV), and

iii) Fast or high energy neutrons ($E_n >$500 KeV).

The above groups are further subdivided in a more detailed classification as shown on table 2.1

Table 2.1 Classes of Neutrons (Thewlis, 1961)

Neutron Type	Energy (eV)	Temperature (K)	Velocity (m/s)	Wavelength (m)
Cold	0.001	11.6	4.37×10^{7}	9.04×10^{-10}
Thermal	0.0253	295.0	2.20×10^{3}	1.80×10^{-10}
Epithermal	11.0	1.16×10^{4}	1.139×10^{4}	2.86×10^{-11}
Resonance	100.0	1.16×10^{6}	1.39×10^{5}	2.86×10^{-12}
Intermediate	10^{4}	1.16×10^{8}	1.39×10^{5}	2.86×10^{-13}
Fast	10^{6}	1.16×10^{12}	1.39×10^{7}	2.86×10^{-14}
High Energy	10^{8}	1.16×10^{12}	1.29×10^{8}	2.79×10^{-15}
Relativistic	10^{10}	1.16×10^{14}	2.99×10^{8}	1.14×10^{-16}

2.3 INTERACTIONS OF NEUTRONS WITH MATTER

Being uncharged particles, neutrons' interaction with matter through electromagnetic force is negligibly small, although it occurs, for example the interaction between neutrons and electrons arises (Beckurts and Wirtz, 1984) from their magnetic dipole moments when they are at close distances from each other. Mostly, neutrons interact with matter through nuclear reactions.

Collisions between neutrons and nuclei can either result in the neutrons bouncing off, or their sticking tightly to the nuclei. Whichever one takes place depends, partly, on the speed (energy) of the incident

neutrons (Table 2.2). Generally, neutron's reaction with nuclei can either be described as DIRECT, or INDIRECT nuclear reactions.

2.3.1 INDIRECT NUCLEAR REACTION

This refers to any reaction in which the incident neutron unites with the target nucleus to form a compound nucleus which may exist in an excited state for some time (usually less than 10^{-17} sec.) with an excitation energy which is equal to the sum of the kinetic and the binding energies of the captured neutron. The excited compound nucleus can then decay in a number of ways, some of which are enumerated below;

Compound Elastic Scattering or Resonance Scattering (n,n).

This refers to the reaction where a neutron of the same energy as the originally captured neutron is emitted by the compound nucleus. The reaction is an elastic collision process when in the nucleus - neutron centre of mass system there is no change in the neutron's energy.

Inelastic Scattering (n,n')

A neutron of kinetic energy less than that of the incident (absorbed) neutron can also be emitted, this leaves the residual nucleus in an excited state.

Subsequently, it can de-excite by gamma ray emission e.g.

$$^{115}In(n,n')In^{115} \quad \ldots\ldots\ 2.3$$

And for a higher incident neutron energy, the inelastic scattering can lead to emission of two neutrons in the form: A (n,2n') B.

Radioactive capture (n γ)

This refers to the case in which the compound nucleus de-excites by emitting one or more gamma-rays. Examples are;

$$^{27}Al + {}^{1}n \longrightarrow {}^{28}Al + \gamma$$
$$^{1}H + {}^{1}n \longrightarrow {}^{2}D + \gamma \qquad \ldots\ldots\ 2.4$$

This process can take place with nearly all elements and it is often more effective with slow neutrons (Garg and Batra, 1986). The compound nucleus is always an isotope of the original element.

Charged Particle Emission

For sufficiently high excitation energy the compound nucleus can de-excite by emitting charged particles; (n, α), n,p), (n,2p), (n,d), (n,t), etc.

Examples:

$$\left.\begin{array}{l} {}^{6}Li + n \longrightarrow {}^{3}H + \alpha + 4.786\ MeV \\ {}^{3}He + n \longrightarrow {}^{3}H + P + 0.764\ MeV \end{array}\right\} \quad \ldots\ldots 2.5$$

<u>Neutron Induced Fission (n,f)</u>

The compound nucleus could break into two almost equal nuclei with the emission of more neutrons. This reaction is most common with heavy target nuclei, and the reaction is usually accompanied by energy release, e.g.

$$^{235}U + n \longrightarrow {}^{236}U \longrightarrow {}^{139}La + {}^{95}Mo + 2n + 200MeV \quad \ldots\ldots 2.6$$

Table 2.2 Neutron reactions with nuclei.

Neutron Energy Band (eV)	Neutron Reactions with Nuclei	
	25 < A < 80	80 < A < 240
$0 - 10^3$	(n,n), (n,γ)	(n,γ), (n,n), (n,f)
$10^3 - 0.5\times10^6$	(n,n), (n,γ)	(n,n), (n,γ)
$0.5\times10^6 - 1\times10^7$	(n,n) (n,n'), (n,p), (n,α)	(n,n), (n,n') (n,p), (n,γ)
$10^7 - 5\times10^7$	(n,2n), (n,n') (n,n) (n,p) (n,np), (n,2p) (n,α).	(n,2n), (n,n') (n,n), (n,p), (n,np), (n,2p) (n,α).

2.3.2 DIRECT NUCLEAR REACTION

As the name implies these are nuclear reactions that proceed directly without any compound nucleus formation. A good example of such reactions is the Direct Elastic Scattering. It is frequently identified with potential scattering which is the deflection (scattering) of incident neutron by the (real) average nuclear potential in the target nucleus.

While the direct scattering can always occur, a compound nucleus can only be formed (in an indirect reaction) if the sum of the binding and kinetic energies of the incident neutron correspond to an excited state of the compound nucleus (Beckurts and Wirtz, 1964). This preferrential absorption of specific energies by certain nuclei is also referred to as resonance reaction (scattering).

2.4 NUCLEAR REACTION CROSS SECTION.

If a beam of neutrons is incident on matter, a variety of reactions could take place simultaneously, a particular reaction might occur and it might not. Although it is impossible to predict, in advance , the fate of an individual neutron, it is possible to state, after investigation, the percentage or fraction of occassions, or encounters, for which each mode of reaction will occur. Thus for each mode of behaviour we can state the "probability" that any particular kind

of interaction between a neutron and a nucleus will occur.

If J is the number of neutrons crossing a unit surface area of a material perpendicularly per second, then the number of interaction ψ (scattering and absorption), in the material per second per unit volume is directly proportional to both J and N, where N is the number of identical atomic nuclei in the material per unit volume i.e.

and

$$\psi \propto JN$$

$$\psi = \sigma JN \qquad \text{..... 2.7}$$

The proportionality constant σ is called the cross section and it is used to describe the "STRENGTH" of a particular interaction. Indeed, the cross section is the probability that any particular kind of interaction between a neutron and an atom will occur.

From equation 2.7 it follows that σ has the dimension of area, though the unit Barn is frequently used:

$$1 \text{ barn} = 10^{-24} cm^2$$

The total probability for a neutron to intereact is given by the sum of the individual cross sections

(IAEA, Isotopic, 1988 and Leo, 1987);

$$\sigma_{total} = \sigma_{elastic} + \sigma_{inelastic} + \sigma_{capture} + \sigma_{fissions} + \ldots \quad \ldots\ldots 2.8$$

Another significant fundamental quantity of nuclear reaction is called Macroscopic cross section given by

$$N\sigma = \sum \quad \ldots. 2.9$$

It specifies the cross section per unit volume. It's reciprocal is also of physical significance known as the mean free path :

$$\lambda = 1/\sum \quad \ldots\ldots 2.10$$

This is the path length between two successive interactions by a neutron.

2.5 APPLICATIONS AND USES OF NEUTRONS.

Neutrons are one of the most versatile probes of modern physics (Morrison, 1951). Advantage is taken of all their properties as well as their mode of interactions with matter in their applications and uses. For example, the fact that neutron experiences no electrostatic repulsion means they can interact with nuclei even when they are very slow. Also, the experimental demonstration of neutron diffraction has not only demonstrated the reality of the wave-particle duality concept of matter, but it has also led to an

experimental technique which has contributed immense / to the knowledge of the properties of matter (Bacon, 1969). The use of thermal (low energy, long wavelength) neutrons in the study of condensed matters arises because the wavelength of thermal neutrons, (which is about 1.12×10^{-10}m) is comparable to the separation of the atomic nuclei in crystals. For example, the atomic nuclei separation is 2.56×10^{-10}m for metallic copper, and between 1.26×10^{10} --> 1.54×10^{-10}m for carbon (Bacon, 1969). Similarly, intermediate energy neutrons are also used to study the nature of the atomic nucleus, while high energy neutrons are used to probe the structure of the nucleons themselves (Marshall and Lovesey, 1971).

One of the important applications of neutrons is the Neutron Activation Analysis (NAA). This is a valuable analytical technique which is mainly applied to the determination of trace elements in a variety of matrices, ranging from biological specimen to geological samples. NAA has also been applied in Industry (Ciechanowski et al, 1988), Agriculture Tiwari el al, 1971), Medicine (Spyrou and Matthews, 1981), etc. Apart from high sensitivity, NAA has the added advantage of being a purely nuclear method which implies that the results are not influenced by the chemical state of the elements under investigation (Op de Beeck, 1986). Further discussion on NAA is given in

the later chapters, since it is the analytical technique employed in this work.

The magnetic property of a neutron, which also makes its interaction with atoms of some ferromagnetic substances possible, has also led to an important application of neutron in the solid-state investigation. This branch of neutron application, called Nuclear Magnetic Resonance (NMR), provides information about the dynamical nature of the interactions among the constituents of matter.

CHAPTER THREE

NEUTRON SOURCES

3.1 INTRODUCTION

Any freely existing neutron is either absorbed quickly by nuclei, or it decays on its own accord within a relatively short time. Thus, free neutrons are hard to find in nature. Neutrons can, however, be produced, artificially, by separating them from nuclei, particular those in which they are loosely bound (Beckurts and Wirtz, 1964) . This could be achieved through a variety of nuclear reactions and fission processes.

The nuclear reactions which lead to neutron production could arise from collisions between projectiles (which could be alpha particles, proton, deuterons, tritium, gamma radiations, or heavy ions like Li and Be,) and target nuclei. The collision processes could either be induced by natural radioactivity where the projectiles are from natural radioisotopes, as in the case of radioactive neutron sources, where an emitter of a suitable projectile is mixed with a suitable target material, or it could be artificially induced by arranging a suitable target material in the path of an accelerated projectile, as its the case in the neutron generators. Also the fission processes could either be spontaneous, as it

occurs in some transuranic elements of which Californium - 252 is the most common, or it could be artificially induced, as it is the case in Nuclear reactors.

Also, a neutron producing reaction can either proceed directly, or lead to the formation of excited compound nucleus, in which case if the excitation energy is higher than the binding energy of the "last neutron" in the compound nucleus then a neutron is very likely to be emitted (Beckurts and Wirtz, 1964). The remaining excitation energy (less the binding energy) is then distributed as kinetic energy between the neutron and the residual nucleus.

3.2 TYPES OF NEUTRON SOURCES

There are different types of neutron sources and, although they are all primarily based on nuclear reactions for their neutron production, they vary widely in physical sizes and complexity as do the intensities of the neutron beams they produce (Bacon, 1969). These variations result from the fact that there are different types of neutron producing nuclear reactions and fission processes. Most neutron producing devices can be classified under one of the following:

(i) Isotopic Neutron Sources

(ii) Neutron Generators

(iii) Nuclear Reactors

(i) ISOTOPIC NEUTRON SOURCES

Neutron production by these sources is based on natural radioactivity. Such sources are usually small in size (approximating to point sources) and they produce strong, almost isotropic beam of neutrons which is also nearly constant in time.

Isotopic or radioactive neutron sources are in three categories depending on the type of reaction invloved:

(a) Radioactive alpha-neutron sources

(b) Radioactive photo-neutron sources, and

(c) Spontaneous fission sources.

(a) Radioactive alpha -neutron Sources

A number of transuranic elements disintegrate by alpha decay. Such elements are used for neutron production through the (α ,n) reaction. The (α ,n) sources are made by mixing suitable target materials with a strong alpha emitter in a suitable proportion. The most yielding target material is beryllium because it has a low (α ,n) reaction threshold. Others like boron, flourine, and lithium have relatively less yields. For activation analytical purposes Be is mostly considered, the other targets are usually used for special purposes, such as stimulating a fission process (Leo, 1987).

When bombarded with alphas, beryllium undergoes a number of reactions which lead to the production of free neutrons. An excited compound nucleus may be formed which then decays through a variety of modes depending on the excitation energy (Beckuntz and Wirtz, 1964):

$$^{4}He + {}^{9}Be \longrightarrow {}^{13}C^{*} \longrightarrow \begin{cases} {}^{12}C^{*} + n \\ {}^{8}Be + \alpha + n \\ 3\alpha + n. \end{cases} \quad .... \ 3.1$$

The reaction may as well proceed directly without compound nucleus formation as in the following examples (Bekuntz and Wirtz, 1964):

$$\alpha + {}^{9}Be \longrightarrow {}^{12}C^{*} + n + 5.704 \text{ Mev}$$

$$\alpha + {}^{11}B \longrightarrow {}^{14}N + n + 0.158 \text{ Mev}$$

$$\alpha + {}^{7}Li \longrightarrow {}^{10}B + n - 2.790 \text{ Mev} \quad ... \ 3.2$$

The general characteristics of each (α, n) source type, which makes it desirable or otherwise when considering the choice of a radioactivce (α,n) source, are largely determined by the properties of the alpha-emitter involved. Some of them are summarized in table 3.1.

Table 3.1 Properties of the α-emitters of radioactive Be (α,n) sources (IAEA Isotopic, 1988).

Nuclide	Half life	Alpha Energy (MeV)	Yield per 10^6 alphas (n.s-1)	Yield (n.s^{-1}.Ci)	Yield n.s^{-1}.g^{-1}	Average Neutron Energy (MeV)	γ-Dose at 1m for 10^6n/s in Gy/n.
^{238}Pu	89y	5.50	-	2.8×10^6	4.5×10^7	4.0	< 0.01
^{239}Pu	24.360y	5.14	65	1.6×10^6	1.2×10^5	4.59	< 0.01
^{210}Po	138d	5.30	73	2.5×10^6	1.1×10^{10}	4.54	< 0.001
^{241}Am	458y	5.48	82	2.2×10^6	6.5×10^8	4.46	0.01
^{244}Cm	18.1y	5.79	100	3.0×10^6	2.5×10^8	4.31	< 0.01
^{242}Cm	163d	6.10	118	4.0×10^4	10^{10}	4.16	< 0.01
^{226}Ra	1620y	7.69-4.77	502	1.1×10^7	15×10^7	3.94	0.5
^{227}Ac	22y	7.36-5.65	702	1.5×10^7	1.7×10^9	3.87	0.07

(b) <u>Radioactive Photoneutron Sources</u>

When a nucleus absorbs gamma radiation, neutron production through a (γ,n) reaction takes place if the incident gamma energy exceeds the binding energy of a neutron in the nucleus. A photo neutron source is an assembly of a gamma emitter and a suitable target material. Most nuclei have neutron binding energies above 8 Mev (Bekurts and Wirtz, 1984, and IAEA, 1988), and, since the gamma energy of radioactive nuclei (gamma sources) rarely exceeds 3 Mev, only beryllium and deuterium whose neutron binding energies are 1.665 MeV and 2.225 MeV respectively are considered as suitable target materials (IAEA Guideline, 1988; Beckurts and Wirtz, 1964; Leo, 1987). The reactions are;

$$^{9}Be + \gamma \longrightarrow {}^{8}Be + n - 1.665 \text{ Mev}$$

$$^{2}H + \gamma \longrightarrow {}^{1}H + n - 2.225 \text{ Mev} \quad \ldots\ldots 3.3$$

Several natural and artificial radioactive isotopes can serve as the gamma emitters e.g ^{24}Na, ^{72}Ga, ^{88}Y, ^{140}La, etc , but in practice only ^{124}Sb is used as a gamma source (Beckurts and Wirtz, 1964). This could be as a result of its relatively convenient half-life of 60.2 days, its high gamma dose rate of about 10m Gy/h at one meter and the ease of its production by neutron irradiation of natural antimony in a nuclear reactor (IAEA Isotopic, 1988).

One of the disadvantages of photoneutron sources is the low cross section of the (γ,n) reaction which result into very low neutron yields per gamma(1 - 2 orders of magnitude lower than that of the alpha-type sources). Also, the non reacting gammas are not absorbed as easily as alpha particles, so the (γ,n) sources are accompanied by a large background of gamma radiation which poses serious handling problems. However, unlike (α,n) sources, which emit continuous spectrum of neutrons, photo neutron sources produce nearly monoenergetic neutrons by using monochromatic gamma rays, this is becaue gammas are not slowed down as is the case with alphas (Leo, 1987 and IAEA Isotopic, 1988).

(c) **Radioactive Spontaneous Fission Neutron Sources**

A number of transuranic elements also decay by spontaneous fission releasing several neutrons in the process. Such heavy nuclei are used as neutron sources. Among them, only ^{252}Cf is widely applied because all the other nuclei are strong alpha emitters and their neutron spectra show too much contribution from neutrons which are produced by (α,n) reactions (Gang and Batra, 1986).

In the real sense of it, these heavy nuclides are the true isotopic neutron sources (Garg and Batra, 1986), this is because in the other so called isotopic sources, neutron are only produced after an interaction with Be or other target materials. The spontaneous fission of ^{252}Cf can be represented as follows:

$$^{252}Cf \longrightarrow {}^{140}Xe + {}^{108}Ru + 4n + Q$$
$$^{252}Cf \longrightarrow {}^{140}Cs + {}^{109}Tc + 3n + Q \qquad \ldots 3.4$$

Another potential nuclide is 254Cf which decays primarily by spontaneous fission and gives a specific yield of about 10^{16} n. s^{-1}. g^{-1}. However, its very short half life of 56 days makes it not to be economically useful (Garg and Batra, 1986)

(ii) NEUTRON PRODUCTION WITH ARTIFICIALLY ACCELERATED PARTICLES (NEUTRON GENERATORS).

Any source of fast particles or gamma rays with enough energy directed against a target made of light element will set free neutrons (Morrison , 1951). As neutron sources, accelerators have advantages over those simple radiaoctive sources. These include their neutron output (Sometimes up to 10^{14}n/s) which lies many orders of magnitude above that of the radioactive sources, the possibility to produce monoenergetic neutrons of practically any desired energy with good resolution, and the fact that the source can be pulsed so that time - of - flight measurement can be carried out (Beckurts and Wirtz, 1964).

Several nuclear reactions are being used in such machines (Neutron Generators). For examples, monoenergetic neutrons can be produced in (p,n) reaction with accelerated protons e.g

$$^{7}Li + p \longrightarrow {}^{7}Be + n - 1.646 \text{ Mev}$$

$$^{3}H + P \longrightarrow {}^{3}He + n - 0.764 \text{ Mev} \qquad \ldots 3.5$$

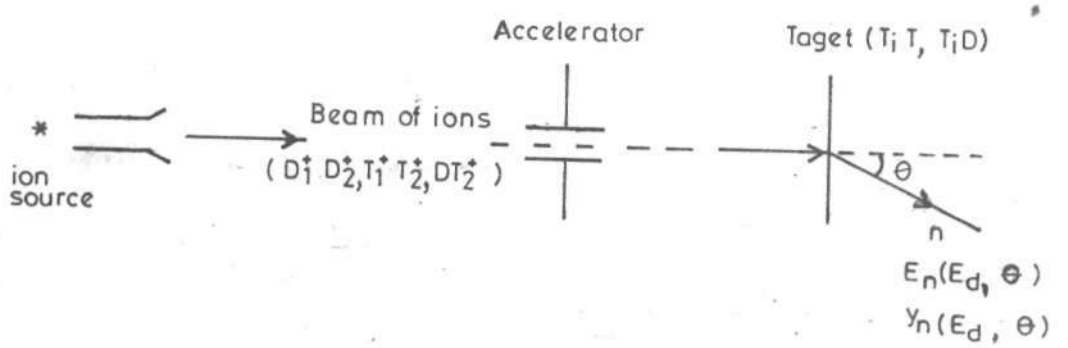
Also, if accelerated electrons of energy between 50 Mev and 100MeV strike a target they give rise to intense Bremstrahlung. A large part of this Bremstrahlung quanta exceeds the binding energy of neutron in the target substance (Beckunts and Wirtz, 1964), and neutrons can be produced in this way through the (r,n) reactions.

One of the most used reactions in the (d,n) reaction, in which a beam of deuteron nuclei bombard a target of deuterium, tritium, lithium or beryllium. With deuterium target, the reaction ^{2}H (d,n) He^{3} is suitable for the production of neutrons in the energy range 2 --- 10 Mev. While with tritium target, the reaction $^{3}H(d,n)^{4}He$ yields neutrons of 12 to over 20 Mev. (Beckurts and Wirtz, 1964).

$$^{2}H + {}^{2}H \longrightarrow {}^{3}He + n + 3.265 \text{ Mev}$$

$$^{2}H + {}^{3}H \longrightarrow {}^{4}He + n + 17.5 \text{ Mev} \qquad \ldots\ldots 3.6$$

The above reactions give a maximum yield of neutron (up to 10^{11}n. s^{-1}) when the initial deuteron particles are accelerated through a potential of about 150 Kev. Then, the energy distribution between the product (α-particle) and the neutron is such that the neutron leaves the target with an energy of about 14 Mev (Csikai, 1987). The acceleration potential is usually provided by a small electrostatic generator. The deuterons are accelerated in a vacuum tube until they collide with a thin or thick target of Zirconium or Titanium into which tritium or deuterium gas has been adsorbed (fig. 3.1). Tritium is a radioactive hydrogen isotope with a half-life of about 12.5 years (Csikai, 1987). Therefore suitable tritum target capable of lasting up to a few hundred hours of machine operation are commercially available.



FIG·3.1; The schematic diagram of neutron production in generators. D_2^+, T_2^+ and DT_2^+ are the singly ionized molecular ions, while D_1^+ and T_1^+ are the atomic ions. $E_n(E_d, \theta)$ and $y_n(E_d, \theta)$ are the neutron energy and yield respectively, both are dependent on the projectile (e.g deuterium ion) energy and the neutron scattering (emission) angle θ. (Csikai).

In addition to the "Big - 4" neutron sour reactions as the above (p,n) and (d,n) reactions are often called (Csikai, 1987), there are other possible fast neutron producing reactions, for example; at higher deuteron energies, Lithium and Beryllium metals can be used as targets:

$^{9}Be + {}^{2}H \longrightarrow {}^{10}B + n + 4.362$ MeV

$^{7}Li + {}^{2}H \longrightarrow {}^{8}Be + n + 15.028$ MeV

$^{7}Li + {}^{2}H \longrightarrow 2He^{4} + n + 15.122$ MeV

Apart from all these hydrogen reaction mentioned above, neutron production through inverse (p,n) and (d,n) reaction have also been considered (Drosg, 1987), following the development of the production of high intensity beam of heavy ions like Tritium (t), Li, and Be ions. Example of neutron producing inverse reactions are; $^{1}H(t,n)^{3}He$, $^{1}H(^{7}Li,n)^{7}Be$, $^{1}H(^{11}B,n)^{11}C$, $^{2}H(^{13}C,n)^{14}N$, $^{2}H(^{9}Be,n)\ ^{10}B$, etc. Neutron production by inverse reactions has the advantage of giving higher neutron energies, so that the energy gap between primary neutron and the secondary neutron from break-up reaction is appreciable. It also has a wider range of monenergetic neutron production, and a containment of neutron emission into a forward cone, which implies less background effect and simplified shielding requirement (Drosg, 1987).

(iii) NEUTRON PRODUCTION IN NUCLEAR REACTORS

Nuclear reactors are the most intense sources of neutrons. Their operation is based on the process of nuclear fission, in which a fissile nucleus (mostly ^{235}U) splits when struck by a single initial neutron, producing fission products as well as two or three further neutrons (Bacon, 1969). A typical fission reaction with ^{235}U is as follows:

$$n + {}^{235}U \longrightarrow {}^{236}U^* \longrightarrow {}^{141}Ba + {}^{92}Kr + 3n + Q$$

Since about 3×10^{17} fission occur per sec, and about 2.5 neutrons are produced (averagely) per fission, (Beckurts and Wirtz, 1964) hence, for a typical research reactor about 7.5×10^{17} neutrons are produced per sec. Though only a very small fraction of this is usually available for neutron experiment.

Spontaneous fission occurs only in rare cases (e.g. with U-238 in a process with half-life of about 10^{16} years) (Thewlis, 1961), thus fission is generally induced by supplying energy e.g. by the absorption of a photon (photofission), or by the absorption of light particles, such as a proton, or an alpha. Fission is, however, most readily induced by bombardment with neutrons since they experience no coulombic barrier and can thus travel relatively slowly (higher interaction probability). According to Bohr and Wheeler (1939), the energy of the compound nucleus formed by neutron

capture is first stored in the motion of individual nucleons like the thermal motion in liquids. Then this energy is converted into potential energy of deformation, and the nucleus becomes sufficiently distorted from spherical shape, it may then oscillate violently and split into two fragments. The initial fission fragments generally lie above the stability curve (i.e. they are unstable), having excess neutrons, they try to attain stability by emitting the excess neutrons.

The fission chain Reaction

If the neutrons evolved from a fission reaction could be used to induce further fission, a self-sustaining process or chain reaction could be feasible. For this to take place in a steady rate, one neutron from a fission must on the average, produce a further fission i.e. the ratio of the number of neutrons in successive generations (multiplication factor) must be UNITY. For multiplication factor less than one, a chain reaction cannot proceed (subcritical); and if the multiplication factor is greater than one (supercritical), an explosion may result considering the fact that an energy of about 200 MeV is released in the fission of one uranium - 235 nucleus (Beckurts and Wirtz, 1964). So, fission chain reaction is made to proceed at a controlled rate, and the essential

elements necessary to ensure this are the fuel elements, the moderator, and the control rods.

The fuel element can be natural uranium which consists of its different isotopes ^{238}U (99.3%) ^{235}U(0.7%) and ^{233}U(0.005%). Each of these isotopes have different cross sections for fission (n, f) and capture (n, γ) reactions when they are bombarded with neutrons.

The principal fission process in uranium involves the thermal neutrons with uranium-235, whereas, the probability of fission with uranium - 238 is very small and it is only with fast neutrons. Capture of many neutrons in the energy range between 5 eV and 2 KeV, by uranium-238, also occur without any fission, this causes a serious depletion in neutron population especially since there are neutrons of various energies in a reactor (from as high as 17 MeV to as low as 0.025 eV, with a most probable energy of 0.72 eV and an average of 2 MeV) (Thewlis, 1961)). Therefore, the simplest way to ensure chain reaction is to slow the neutrons down as quickly as possible, this is called MODERATION. Another means of ensuring chain reaction is to increase the proportion of the uranium - 235 isotope in the fuel element, this is called ENRICHMENT.

The role of control rods as a safety measure in nuclear reactors should also be mentioned, even though

they work against the primary function of the reactor, which is neutron production. These rods are usually made from materials of very high neutron absorption cross section like Boron (σ =3837 barn). If the critical state of neutron production in the core is being exceeded, which may lead to explosion, the rods are inserted inside the core and the neutrons are being absorbed until the critical stage is restored.

The beam of neutrons produced in the reactor are usually extracted out of the reactor in a variety of ways using facilities such as beam holes and thermal columns. A beam hole is, usually, a cylindrical aluminium pipe reaching outside from the core, while a thermal column is a prism of highly purified graphite, it produces a pure thermal neutron field from the impure field of neutrons leaking out of the core (Becon, 1969).

3.3 SELECTING AN APPROPRIATE SOURCE

When choosing a neutron source, certain general factors have been identified (Garg and Batra, 1986 and IAEA Isotopic, 1988) for consideration, these include; cost, available flux, toxicity and shielding requirements, availability, etc. There are other special consideration peculiar to specific applications, like mobility.

Generally, for analytical purposes, nuclear reactors and neutron generators are the most desired neutron sources because of their high intensity neutron output and, consequently, high sensitivity in analytical results. However, the high purchasing and maintenance costs of these high intensity neutron sources, coupled with their complicated shielding requirements often put them out of reach of most Universities and Research Centres, especially in the developing countries.

Isotopic neutron sources on the other hand, offer a consolation in the absence of Nuclear Reactors and Neutron Generators. Some of their intrinsic characteristics include: extremely small size (which makes them portable); relatively low purchase, installation, and maintenance cost; ideal short and long - term stability of neutron flux output; possibility of having either fast or thermalized neutron fluxes or both; their low strength which poses limited health hazard, etc. The low neutron outputs of isotopic sources result into an intrinsic insensitivity (IAEA Isotopic, 1988 and Op de Beeck, 1986) when compared to what is obtainable with Nuclear Reactors. This limit the extent of their application, since only samples with relatively simple composition, such as valuable minerals, concentrates, and alloys, can then be accurately analysed with Isotopic neutron sources.

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However, their short and long-term neutron output stability which makes possible the attainment of excellent precision (Op de Beeck, 1986) is an advantage.

In situation where nuclear reactor or/and neutron generators are available, isotopic neutron sources often play complimentary roles, for example, they can be used for preparatory experiments, and for training students. However, their intrisic characteritics could sometimes make them the preferred major activation analysis facilities even when nuclear reactors and neutron generators are already available (Op de Beeck, 1986, and Garg and Batra, 1986). One of such unique applications of isotopic neutron sources is the on-stream anaysis of materials flowing in pipes (Ceichanowski et al, 1988), or passing on conveyor belts in the industry, where a bulk sample gets irradiated at one end of the conveyor belt while the counting is done on the other end using a precalibrated detector - counting assembly. Other areas of application include well - logging and mineral exploration as well as other similar areas which require in - the field, or on - the spot evaluation methods, e.g. determination of percentage of bitumen in asphalt at construction sites with isotopic neutron sources (Csikai, 1987). Such methods are fast, although usually semiquantitative and, according to Op

de Beeck (1986), they are primarily monitoring methods rather than means of accurate analytical determination.

Comparing Isotopic Neutron Sources

Among the isotopic neutron sources, the short comings of photoneutron sources have been identified (IAEA Isotopic, 1988; Bacon, 1969; Beckurts and Wirtz, 1964; and Leo, 1987) as that of low neutron intensity, and very - high gamma - radiation background which requires expensive shielding. These make photoneutron sources the least used, except for some specific purposes, for example to reduce fission process in reactors.

Spontaneous fission sources, mainly ^{252}Cf, are noted for some of their advantages over the alpha neutron sources. According to Garg and Batra, (1986), the advantages of ^{252}Cf source include its relatively higher available neutron flux, its lower gamma-ray dose rates, and the similarity of its spectrum to that of a normal uranium fission in a reactor, this makes it (the spectrum) well known.

The suitability of alpha neutron sources over the spontaneous sources has also been emphasized (Spyrou and Matthews, 1981 and IAEA Isotopic, 1988). The longer half-life of Am/Be source (458 years) compared to the 2.7 years of ^{252}Cf, as well as the higher mean neutron energy of the alpha neutron sources which would

allow the investigation of reactions for which their threshold energies precluded the use of lower energy neutrons, and the fact that the degree of uniformity of activating flux achieved within an irradiated BULK material would be greater with neutrons of higher energy (Spyrou and Matthews, 1981), are some of the factors usually considered.

Among various alpha neutron sources, ^{241}Am is the most widely used. In the views of experts (IAEA Isotopic, 1988; and Garg and Batra, 1986) (see Table 3.1), both ^{210}Po and ^{242}Cm are too short lived to be of practical value. ^{239}Pu and ^{238}Pu are too toxic and too dangerous to be accepted in many countries. Radium (^{226}Ra) has a very long half life but its high level of gamma rays makes it difficult to shield. ^{227}Ac is an excellent source, but it is not available since its production from the irradiation of ^{226}Ra has been stopped. Similarly, ^{244}Cm is not readily available, therefore, one is left with only ^{241}Am which combines long half life with reasonably high neutron yield and low gamma dose. It is also commercially available often with minimal legal restriction (Op de Beeck, 1986). These probably explain the reason behind the procurement of the 5 Ci ^{241}Am-Be neutron source at the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria. A detailed description of this source is given in chapter four.

CHAPTER 4

CHARACTERIZATION OF THE ^{241}AM/BE NEUTRON SOURCE

4.1 NEUTRON MODERATION (THE HOWITZER)

For Neutron Activation Analysis, especially by radioactive capture reaction, a thermal neutron flux is usually required, although, epithermal and fast neutrons are also being used (Garg and Batra, 1986, and IAEA Isotopic, 1988). The cross section for the radioactive capture (n, γ) reaction is usually largest at thermal energies and, with few exceptions, no other reactions occur at these energies (Tiwari, 1971).

A 5-Curie ^{241}Am/Be Isotopic neutron source is primarily a fast neutron source with an average neutron energy of about 4.46 MeV (IAEA Isotopic, 1988), and there is only a very small percentage of slow energy neutrons (Rieppo, 1984) in its primary neutron spectrum. In order to enhance the intensity of this low energy neutrons, the source is often installed in a moderating material, usually hydrogenous materials. It was reported (Dunning et al, 1935), that Enrico Fermi and his co-workers observed that neutrons from an ordinary source, when surrounded by a substance containing hydrogen show much increased interaction with atomic nuclei, as well as enhanced production of radioactive atoms. Dunning et al (1935) have also quantitatively, shown that moderators such as H_2O and

D_2O have the effect of enhancing the efficiency of neutrons in the production of artificial radioactivity in silver. Their result showed that the enhancing effect of H_2O is about 5.5 times that of D_2O.

Fast neutrons from the source repeatedly collide with the atoms of the moderating medium as they travel through the medium. Loss of energy occur and the neutrons are slowed down gradually by these successive collisions (elastic or inelastic), until the neutrons come into thermal equilbrium with the molecules (or atoms) of the medium through which they are diffusing. At about 293K the mean thermal energy is about 0.0253 eV, corresponding to a Maxwellian velocity of 2200 m/s.

Most good moderating materials are made up of light atomic elements like hydrogen, deuterium, beryllium, and carbon, with the hydrogen being the obviously most efficient neutron moderator through elastic scattering process. For example a neutron can lose nearly all its energy and, on the average, will lose about half its energy in one impact with a (proton) hydrogen atom (Fink, 1936).

The Howitzer

The Neutron Howitzer is a general name given to a special geometrical arrangement of the moderator around a neutron source, such that neutrons are moderated and then concentrated towards a particular direction where

the samples are placed for irradiation (Fig. 4.1). Various such geometrical arrangements have been studied in the past (Fink, 1936) and the effect of varying their parameters (i.e of the Howitzers) on the magnitude and distribution of the thermal neutron flux obtainable have also been studied (Cox et al, 1968) using parraffin wax as the moderator. A detailed description of the Howitzer used in the present work is given under instrumentation.

4.2 NEUTRON DETECTION

As a result of its charge neutrality, the neutron is not readily detectable by the usual detectors employed in the ionizing radiation detection. However, when neutrons react with nuclei (through nuclear reactions, scattering, or fission processes) secondary charged particles are often produced, and in some cases delayed or prompt radioactivity may be induced. Neutrons are therefore indirectly detected through the detection of these secondary charged particles, or induced radiation.

4.2.1 Neutron Detection Through Nuclear Reaction

Typical reactions are: $^{10}B(n, \alpha)^{7}Li$, and $^{6}Li(n,\alpha)^{3}H$. Operation of the promptly indicating neutron detectors are based on these reactions, for example, the Boron Counter usually has the form of a

proportional counter filled with boron III flouride (BF_3) gas. An irradiated BF_3 counter (in a thermal neutron field) will lead to alpha and lithium particles, either or both of which can ionise the gas and give detectable pulses.

4.2.2 Neutron Detection by Recoil Proton Method.

Some neutron detectors use the effect of the recoiling protons from the n-p elastic scattering. Such detectors comprise of a chamber filled with hydrogen gas or paraffin, so that when a (fast) neutron enters the chamber, a recoil proton is produced which can be detected by some standard techniques using ionization chamber, nuclear emulsion, organic scintillation detectors, etc.

4.2.3 Neutron detection by Fission Processes

Neutron detection in the presence of a high gamma radiation background, as it is the case in the Nuclear Reactors, is possible by using the effects of the fission fragments produced when neutrons react with some fissionable (heavy) nuclei. Some of these fission fragments have total kinetic energy of the order of 160 MeV (Beckurts and Wirtz, 1964), and they can easily cause detectable ionization in some materials in which the effects of any gamma rays may not be noticed. It is therefore possible to make extremely gamma insensitive fission chambers for neutron detection.

4.2.4 Neutron Detection by Neutron Activation Method

Neutrons' interaction with some nuclei could lead to the production of radioactivity (beta or gamma) which can be quantitatively measured using beta or gamma-ray spectrometer, even "long" after such interaction has taken place (delayed radioactivity). Materials with large cross section for such reactions are usually called radioactive indicators or probes, e.g. indium foil. They are particularly useful for measurements in a neutron field (Buckurtz and Wirtz, 1964). Since the induced radioactivity is directly proportional to the available neutron flux, measurement of the induced activity is therefore, a measure of the quantity of neutrons in the neutron field. This is the neutron detection method employed in the present work.

4.3 PRINCIPLE OF NEUTRON ACTIVATION ANALYSIS

Neutron Activation Analysis is an analytical technique whereby a sample is irradiated in a neutron field, for a period of time, after which the induced radioactivity is then measured using some standard techniques. Such measurement can lead to both the qualitative and quantitative characterisation of the sample under investigation.

If a sample is irradiated in a neutron field, the number of activated nuclei per second N is directly

proportional to both the number of target nuclei, n, in the irradiated sample, as well as the neutron flux, ϕ ($n.cm^{-2}.S^{-1}$) at the irradiation position, i.e.;

$$N \propto n\phi \qquad \ldots\ldots\ldots 4.3.1$$

and

$$N = \sigma\, n\phi \qquad \ldots\ldots\ldots 4.3.2$$

where the proportionality constant, σ, is the probability of interaction (absorption, scattering, fission, etc) with the nuclei, and it is called the cross section. It is measured in barns (b), ($1b = 10^{-24}\ cm^{+2}$).

The radioactive nuclei formed begin to decay as soon as they are being formed during the irradiation, following the normal decay law ($dN/dt = N.\exp(-\lambda t)$). Therefore the number of radioactive nuclei present at any time t is given by;

$$N(t) = Ne^{-\lambda t} = \sigma n \phi e^{-\lambda t} \qquad \ldots\ldots 4.3.3$$

where λ is the decay constant of the radioisotope of interest and $e^{-\lambda t}$ is the fraction of the decayed radioactive nuclei. The number of radioactive nuclei formed and which are present at the end of irradiation (between t = O and $t = t_i$) is then given by:

$$N(t_i) = \int_0^{t_i} n\phi\sigma e^{-\lambda t} dt \qquad \ldots\ldots\ldots 4.3.4$$

and since n, ϕ, and σ are assumed constant with time;

$$N(t_i) = n \phi \sigma \ (1 - e^{-\lambda t_i})/\lambda \qquad \ldots\ldots 4.3.5$$

The term; $(1-e^{-\lambda t_i})$ in equation 4.3.5 is called the saturation factor. It accounts for decay during irradiation.

If the counting does not commence immediately, this value again reduces by a factor $e^{-\lambda t_w}$ where t_w is the waiting time or cooling time between the end of irradiation and the beginning of counting. Therefore the number of radioactive nuclei in the sample at the beginning of counting is;

$$N(t_w) = N(t_i)\ e^{-\lambda t_w} \qquad \ldots\ldots\ldots 4.3.6$$

The number of radioactive nuclei present at the end of the counting exercise (i.e. $t_w + t_c$ after the end of irradiation) is; $N(t_w + t_c) = N(t_i)\ e^{-\lambda (t_w + t_c)}$

$$\ldots\ldots 4.3.7$$

where t_c is the counting time. Therefore the number of radiation emitted within the counting period is

$$N(t_c) = N(t_w) - N(t_w + t_c)$$
$$= N(t_i)\ e^{-\lambda t_w} (1 - e^{-\lambda t_c})$$

$$N(t_c) = n \frac{\phi \sigma}{\lambda} \ (1-e^{-\lambda t_i}).e^{-\lambda t_w}.(1-e^{-\lambda t_c}) \qquad \ldots\ldots\ldots 4.3.8$$

But only a fraction of this number is often detected and recorded by spectrometers. The number of

radiation detected (or the Detector response) is related to the actual rate of emission $N(t_c)$ by the expression;

$$N_{det} = K.f.I.\varepsilon N(t_c)$$

$$= \frac{mN_A K.f I \varepsilon \phi.\sigma(1-e^{-\lambda t_i}).\ e^{-\lambda t_w}.\ (1-e^{-\lambda t_c})}{\lambda.M} \quad \ldots\ldots 4.3.9$$

where $n = mN_A/M$

and

N_A = Avogadro's Number

m = the mass of the sample material (compound)

M = Atomic mass number of the isotope of interest.

K = the mass fraction of the isotope in the target material

f = fractional intensity of gamma-ray of interest

ε = Detector efficiency for the photon

I = isotopic abundance (%) of the isotope

The activation procedure described by equation 4.3.9 is called delayed neutron activation analysis (DNAA). Some radionuclides decay within a fraction of a second after their formation, to account for such decays, counting is often carried out simultaneously with the irradiation exercise. This is referred to as the Prompt Neutron Activation Analysis (PNAA). The Detector response in this case is given (Spyrou and

Matthews, 1981) by;

$$N = \frac{y\varepsilon\phi\sigma t_i m N_A}{M} \quad \ldots\ldots 4.3.10$$

where y is called the prompt photon yield, it is the number of photons emitted per neutron captured for the gamma ray being measured, and all other terms remain as previously defined.

Again, instead of a single chain of irradiation, waiting, and counting times as described earlier under the DNAA, the sample could be allowed to cool after the first counting exercise and then the whole process of irradiation, waiting, counting and cooling can be repeated for a number of cycles. This is the Cyclic Activation Analysis (CAA).

The period for a cycle $T = t_i + t_w + t_c + t_d$. where all terms have their usual meaning and t_d is the time between the end and of counting and the beginning of the next irradiation.

The Detector response N_1 for the first counting period is given by equation 4.3.9.

And for the second counting period;

$$N_2 = N_1 + N_1 e^{-\lambda T} = N_1(1+e^{-\lambda T}) \quad \ldots\ldots 4.3.11$$

For the nth counting period;

$$N_n = N_1(1+e^{-\lambda T} + e^{-2\lambda T} + \ldots\ldots + e^{-(n-1)\lambda T})$$

$$N_n = N_1 \frac{(1 - e^{-n\lambda T})}{(1 - e^{-\lambda T})}$$

..... 4.3.12

Equation 4.3.12 is, however, assuming a non-varying neutron flux (Spyrou and Matthews, 1981; Op de Beeck, 1986, and IAEA Isotopic, 1988) as obtainable in the case of isotopic neutron sources. A treatment of the case of varying flux can be found in the work of Ortovaoli et al, 1981.

The summed up detected radiation for n cycles i.e. the cummulative response is now given by

$$N_c = \sum_{i=1}^{n} N_i$$

$$= N_1 \{1 + (1 + e^{-\lambda T}) + \ldots + \frac{1-e^{-n\lambda T}}{1-e^{-\lambda T}} \}$$

$$N_c = N_1 \left\{ \frac{n}{(1-e^{-\lambda T})} - \frac{e^{-\lambda T}(1-e^{-n\lambda T})}{(1-e^{-\lambda T})^2} \right\}$$

..... 4.3.13.

Some of the advantages of cyclic Activation Analsysis over the conventional activation method, as noted by many authors (Op de Beeck, 1986; IAEA Isotopic, 1988, and Spyrou and Matthews, 1981), include a reduction of counting statistical error and the enhancement of the signal - to - noise ratio for short-

lived isotopes in the presence of a background of long-lived ones. This is because, while the activities of those long-lived isotopes increase only slowly with increasing number of cycles, the short-lived activities can be saturated during each cycle.

ABSOLUTE NAA METHOD

Absolute NAA entails the absolute determination of the amounts of certain elements in a sample using NAA. In order to achieve this the absolute value of activity induced in the sample as well as the absolute neutron flux value must be known. The neutron flux can be absolutely determined only if we can absolutely count the activity induced in some suitable monitors of well known decay schemes such as gold foil. This method is not practical enough (Garg and Batra, 1987) since it is difficult to measure all the required nuclear parameters with high accuracy. Therefore there are usually many uncertainties (IAEA Isotopic, 1988). Problems of absolute activation is further highlightened under neutron field characterization in section 4.4 below.

Relative NAA Methods

In this activation method, activity of an unknown sample is compared to a standard whose concentration for the elements of interest are known to be close to

that of the unknown sample. The standard must be similar in matrix to the sample in order to control errors arising from self absorption of neutron and flux gradient during irradiation. The standard sample can be irradiated simultaneously along with the unknown samples, both standards and samples can be counted with the same detecting system simultaneously, or one after the other with decay correction. And, in the case of isotopic neutron sources, if the irradiation geometry is kept unchanged, simultaneous irradiation of samples and standards may not be necessary since flux variation (with time) do not occur (IAEA Isotopic, 1988 and Op de Beeck, 1986), this is particularly true of the long lived sources like ^{241}Am/Be source. The activation equations of sample and standards can be combined to give the following equation (Garg and Batra, 1987);

$$\frac{R_{std}}{R_{samp}} = \frac{W_{std}}{W_{samp}}$$

where the Rs refer to the experimental specific countrates, and w refers to the corresponding mass for standard and samples assuming identical composition and geometry. Both absolute and relative activation methods are however, limited by the effects of flux variation in the neutron field.

4.4 NEUTRON FIELD CHARACTERIZATION

4.4.1 Flux Gradient and flux perturbation

As neutrons from the source diffuse through the surrounding (moderating medium), they are attenuated in various ways, and this normally results in neutron flux gradient within the medium. Again, due to the usually modest available flux from isotopic neutron sources, large samples (and standards) are often required to give reasonable induced activity. Large materials, on the other hand, lead to increased flux perturbation. Additional perturbations also arise from the finite size (and the composition) of the source itself (Beckurts and Wirtz, 1964); since neutrons can be absorbed or inelastically scattered in the source too, thereby perturbing the neutron flux within its vicinity.

The total pertubation is usually made up (IAEA Isotopic, 1988) of a self-shielding effect in the interior of the sample, plus a flux depression in its vicinity, as well as a neutron scattering effect by the sample and its container. The error due to the flux gradient resulting from general attenuation and the total perturbation within the irradiating medium can be minimized by irradiating standard and samples of equal sizes and geometry as well as similar composition. It can also be partly accounted for by using attenuation correcting factors such as the following (IAEA, 1988);

$$F(\text{foil}) = 1 - 0.5d\,\Sigma_a(0.9226 + \log 1/\Sigma_a)$$
$$F(\text{cylinder}) = 1 - 4R/3\Sigma_a \quad ; \quad R\Sigma_a << 1$$
$$F(\text{sphere}) = 1 - R\Sigma_a \quad ; \quad R\Sigma_a << 1 \qquad \ldots\ 4.4.1$$

where d is the foil thickness, R is the radius of an infinitely long cylinder or the sphere diameter, Σ_a is the macroscopic neutron cross section of the material and F is the self-shielding factor.

On the other hand, the neutron field distribution within the medium (moderator) could be characterised by determining the relative neutron flux values at different distances from the source. From such measurement, the average relative neutron flux within the volume of an irradiated sample or standard of any geometry could be estimated. This latter consideration (measurement) is the subject of the present work. The intrinsic axial and radial flux gradient within the irradiation cavity above the ^{241}Am/Be source in the howitzer was investigated using foil activation method. A theoretical estimation of the flux distribution was also carried out using neutron diffusion theory.

4.4.2 **Instrumentation and source installation**

The 5 Curie ^{241}Am/Be Isotopic neutron source has a neutron emission rate of 1.29 x 10^7 $n.s^{-1}$ +/-10 per cent. Its active zone consists of a compacted mixture of Americium oxide and beryllium metal, doubly

encapsulated in stainless steel and sealed by argon arc welding. It is cylindrical with an outside dimension of 30 mm diameter x 48mm overall length (manufacturers specification) (See figure 4.2))

The moderation of the original fast neutrons is effected by a nearly 4πimmersion of the source in its transportation container which is a solid cube of side 510mm made of paraffin wax. It has a cylindrical cavity in it thus behaving like a Howitzer (Fig. 4.1). Some of the parameters of this, so called, Howtizer could not be varied, however, where necessary, modifications were made to the specification of Cox et al, (1968), and also based on prior optimization measurements carried out in this work. Such modifications were carried out with candle wax and perspex and these can be observed by comparing figures 4.1 and 4.3

The cylindrical source was installed inside a annular perspex having the same inner dimensions as the source, and an outer diameter slightly less than 90mm so as to fit into the steel sleeve (Figure 4.3). This arrangement keeps the source at a fixed position and consequently ensures reproducible irradiation geometry. Prior to being installed in the annular perspex, loops of thread were tied around the source, for easy and rapid transfer whenever the need arises. The source -

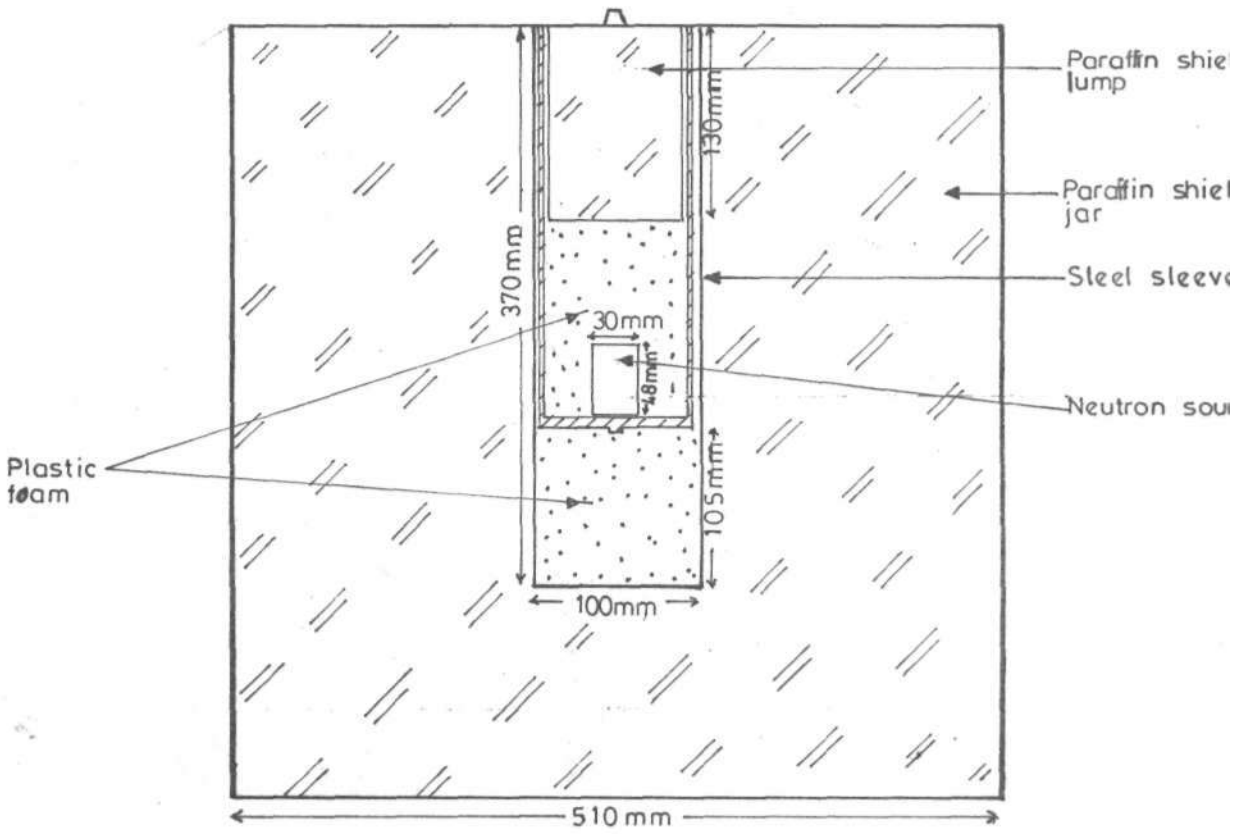


FIG. 4.1: THE TRANSPORTATION CONTAINER

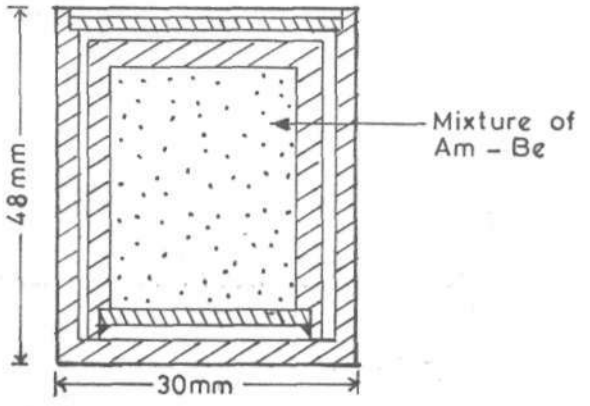


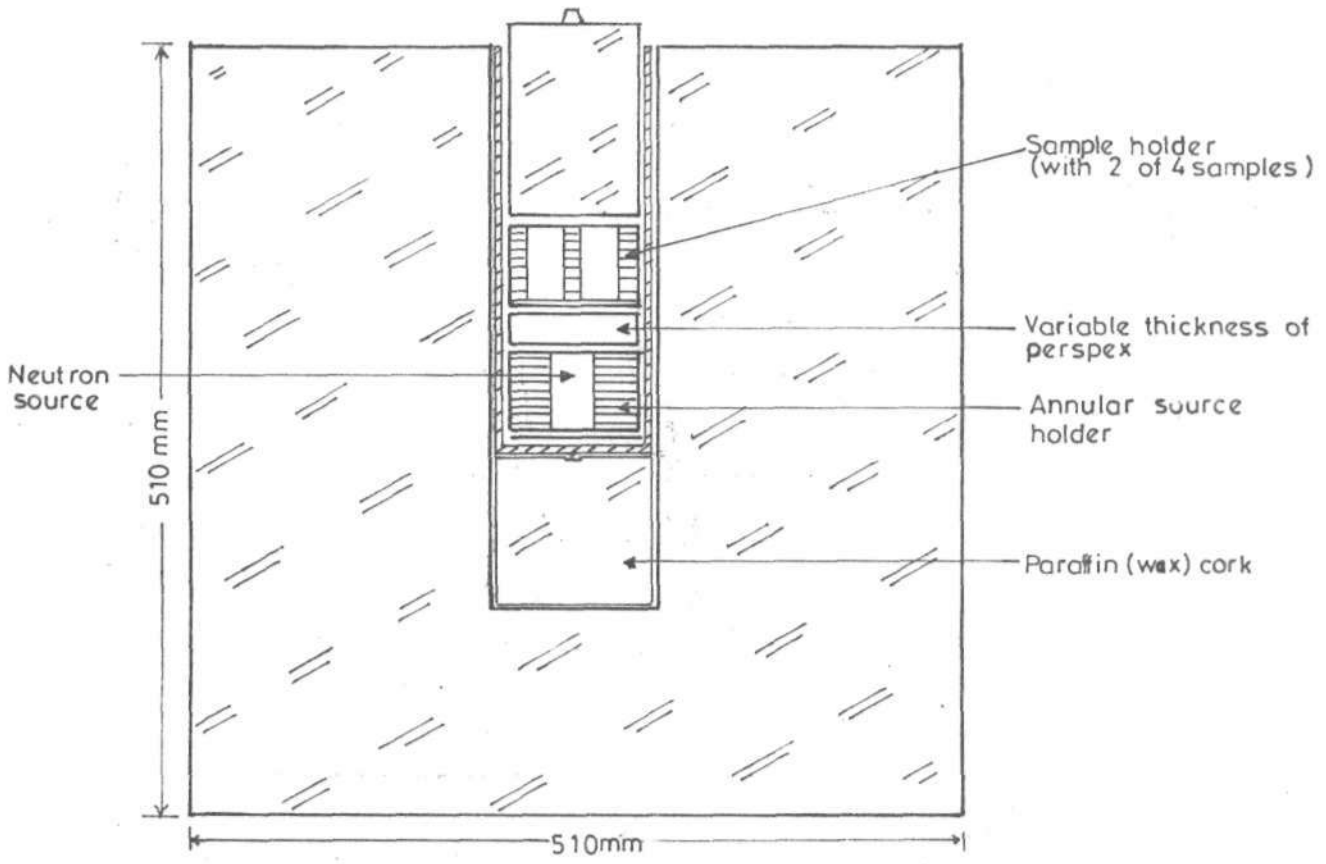
FIG. 4.2: CROSS SECTION THROUGH THE SOURCE

annular perspex arrangement rests on a 10mm thic perspex sheet at the bottom of the steel sleeve, this perspex sheet, together with the 105mm thick paraffin wax on which the sleeve rests, provide the backscattering effects in accordance with the findings of Cox et al, (1968).

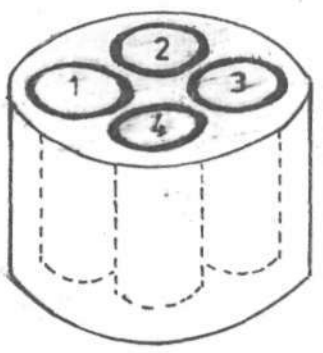
Sample Holder

A sample holder was also constructed with perspex (See figure 4.4). It has an external diameter of about 85mm and a height of 50mm. It contains four sample holes of equal dimensions and of about 25 cubic cm each. Four samples, or two samples and two standards, each of as much as 25 cubic cm can be irradiated simultaneously, and, to ensure reproducibility, the holes are labelled. Different marks are also made on the edge of the cylindrical cavity corresponding to each of the labelled holes for reproducibility during replacement. A loop of string is tied to the sample holder for lowering it into and removing it from the irradiation cavity.

The Instrumentation for the data acquisition and analysis was made up of the EG & G ORTEC (maestro) ADCAM 100 MCA work station comprising of a Hyper-germanium Detector, modular electronics consisting of Pre-Amplifier, Amplifier, High-Voltage Power supply, etc, and the 918A ADCAM multichannel Buffer (MCB) which



FIG·4·3 :THE HOWITZER (MODIFIED TRANSPORTATION CONTAINER)



FIG·4·4 :THE SAMPLE HOLDER

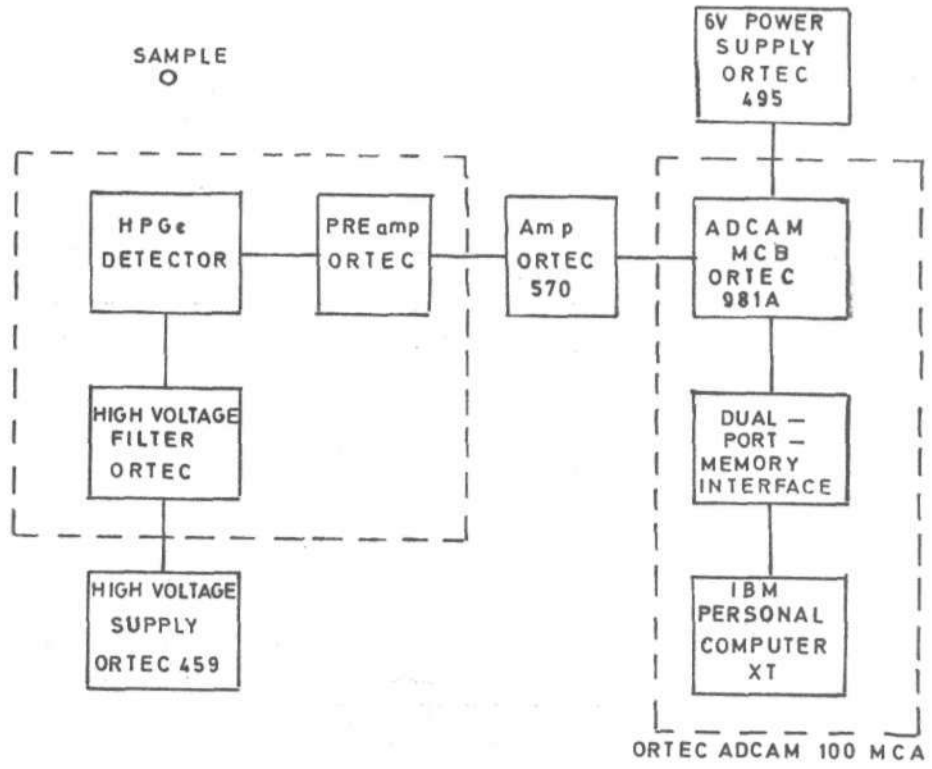
is connected to an IBM PC through a dual Port memory interface (Maestro ADCAM 100 MCA Manual, and Kabiru, 1990). See figure 4.5.

4.4.3 Relative Neutron Flux Distribution

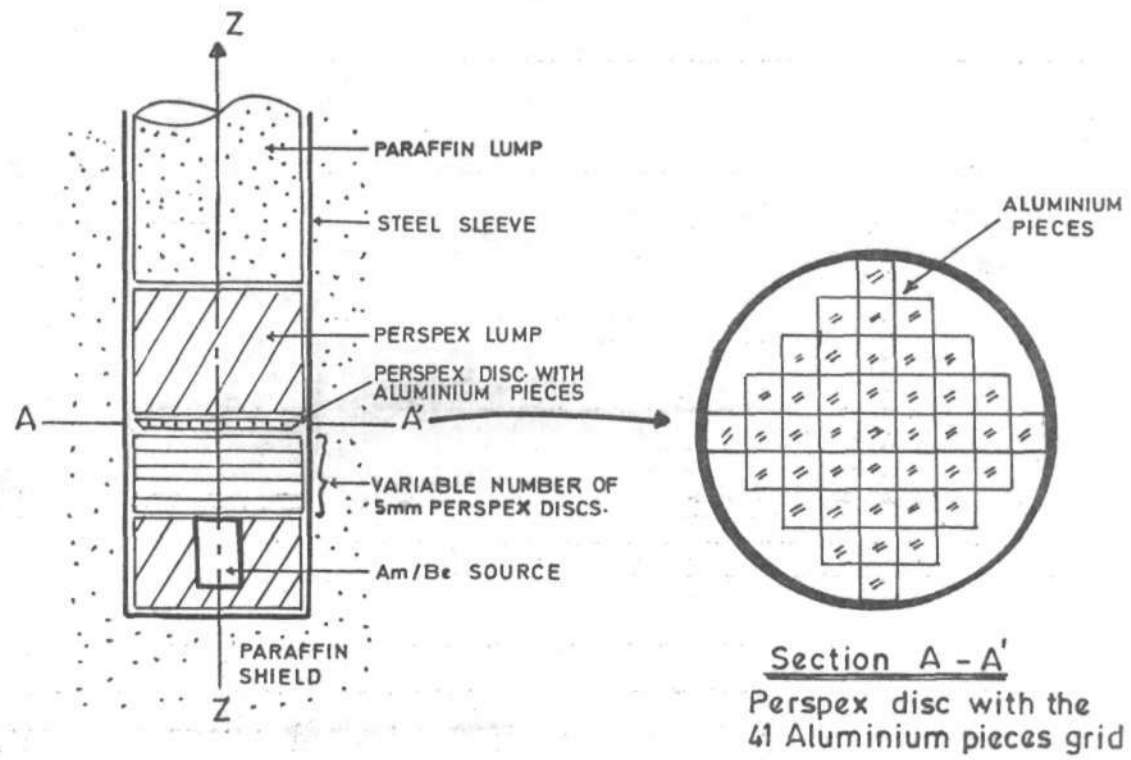
The radial and axial neutron flux distributions in the cylindrical cavity above the source were investigated by varying the thicknesses of the moderating perspex between the source and the monitor(s). Variation by 5mm was possible because the perspex was cut into discs of 5mm thick each. The lateral flux distribution at different heights from the source was studied by irradiating a grid of forty-one samples of about 10mm x 10mm aluminium chips cut from the same aluminium sheet (2mm thick) for 20 minutes. The samples were arranged in a grid on a perspex disc and fixed to the base of the sample holder, described earlier on, and then irradiated with the planes of the sample perpendicular to the axis of the source. A typical configuration of the aluminium pieces arrangement (grid) at irradiation position is shown in figure 4.6. The grid (of forty-one samples) is irradiated at every 5mm from the top of the source upwards. This arrangement, though very tedious, is expected to offer more detailed information about the lateral flux gradient than could the irradiation of circular foils which are then cut into sectors as suggested by another author (IAEA Isotopic, 1988).

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The samples were removed one at a time and counted for ten minutes each. The first sample was always removed after twenty minutes of irradiation, the remaining samples were returned to the same irradiation position. This removal and replacement is repeated until the last sample has been counted. The differences in irradiation time as well as decays during the removal and replacements of the remaining samples were accounted for in the calcuclations using equation 4.3.9, and a computer programme written to evaluate the large data generated. It is pertinent, however, to note that error due to the non-uniformity in irradiation times could be insignificant (even if unaccounted for) especially for the thermal reaction of aluminium of half-life 2.3 minutes which has already reached saturation even before the first sample was removed. To ensure that each aluminium sample is replaced back to their original irradiation position, the initial orientation was always noted with marks on both the sample holders and the cylindrical cavity as described earlier on. Such marks were always made to correspond during replacement. With this arrangement a replacement error of less than 1mm is guaranteed (i.e. <10 % replacement error). A set of forty-one samples were irradiated at every 5mm vertical distance increment from the top of the source in order to investigate the persistence, or otherwise, of any observable feature in the neutron flux distribution



FIG·4·5: BLOCK DIAGRAM OF THE DATA ACQUISITION AND ANALYSIS INSTRUMENTION AT C·E·R·T· ABU (Kabiru, 1990)



FIG·4.6: TYPICAL CONFIGURATION FOR THE ALUMINIUM PIECES GRID IRRADIATION FOR LATERAL NEUTRON FLUX MORNITORING

within the irradiation cavity, increasing distance from the source. Since we were interested, mainly, in the relative neutron distribution, the aluminium samples used were of technical grade rather than analytical grade (99.99%). Aluminium was used for this measurement for two main reasons; first, aluminium has been reported (Beckurts and Wirtz, 1964 and IAEA Isotopic, 1988) to have been used both as thermal probes and as threshold detectors, due to its relatively appreciable cross section values for thermal and fast neutrons. The reactions monitored are; $^{27}Al(n,\gamma)^{28}Al$ for thermal reaction and $^{27}Al(n,p)^{27}Mg$ for the threshold reaction. Therefore a single irradiation provides information about the two fluxes and makes the determination of their ratio, at every point, straight forward. Secondly, aluminium is relatively cheap, therefore, the large number of pieces required for the monitoring exercise could be easily afforded.

Table 4.1 Nuclear Data for the activation reaction of aluminium and indium.

Reaction	Isotopic Absorbance (%)	Cross section (mb)	Half life	Gamma Energy (ReV) (% Abundance)	Detector Efficiency(%)
$^{115}In(n,\gamma)^{116}In$	95.7	$(162.3 \pm 0.7) \times 10^{3}$	54.12m	1097.3 (56)	0.86
				1293.5 (84)	0.70
$^{27}Al(n,\gamma)^{28}Al$	100.0	23.2×10^{1}	2.25m	1779.0 (100)	0.47
$^{27}Al(n,p)^{27}Mg$	100.0	22.0	9.45m	843.0 (71)	1.20

The axial thermal neutron flux distribution was further investigated using high purity 1cm x 1.5cm indium foils. The indium foils were irradiated for 2 hours and counted for 20 minutes each. The nuclear data used in the analysis of both the indium and the aluminium monitors are as shown in table 4.1. The flux values were obtained by solving for ϕ in equation 4.3.9. The calculated values of the axial neutron flux are tabulated (Table 4.2).

Table 4.2 Axial neutron flux distribution

Distance r (cm)	Thermal flux ϕ_{th} ($n.cm^{-2}.s^{-1}$)		Fast Flux ϕ_f ($n.cm^{-2}s^{-2}$)	Ratio ϕ_f/ϕ_{th}	Ratio $\phi_f/(\phi_f+\phi_{th})$
	with indium (*)	with aluminium	with aluminium		
0	1.185×10^4	3.709×10^4 +/- 13%	4.870×10^5 +/- 6%	13.130	0.920
0.5	1.299×10^4	3.792×10^4 +/- 13%	3.772×10^5 +/- 8%	9.947	0.909
1.0	1.414×10^4	3.914×10^4 +/- 13%	3.682×10^5 +/- 15%	9.407	0.904
1.5	1.460×10^4	4.085×10^4 +/- 11%	3.135×10^5 +/- 14%	7.674	0.885
2.0	1.489×10^4	3.997×10^4 +/- 13%	3.067×10^5 +/- 17%	7.673	0.885
2.5	1.517×10^4	4.438×10^4 +/- 12%	2.736×10^5 +/- 19%	6.165	0.860
3.0	1.443×10^4	4.123×10^4 +/- 12%	2.432×10^5 +/- 22%	5.899	0.855
3.5	1.382×10^4	3.792×10^4 +/- 12%	1.933×10^5 +/- 28%	5.098	0.836
4.0	1.352×10^4	4.168×10^4 +/- 12%	2.045×10^5 +/- 30%	4.906	0.831
4.5	1.274×10^4	3.958×10^4 +/- 13%	2.191×10^5 +/- 30%	5.536	0.847
5.0	1.189×10^4	3.754×10^4 +/- 13%	1.705×10^5 +/- 34%	4.542	0.820
5.5	1.123×10^4	3.897×10^4 +/- 12%	1.440×10^5 +/- 36%	3.695	0.787
6.0	1.053×10^4	3.533×10^4 +/- 12%	1.510×10^5 +/- 38%	4.274	0.810
6.5	0.971×10^4	3.251×10^4 +/- 14%	1.487×10^5 (**)	4.574	0.821
8.0	0.749×10^4	2.705×10^4 +/- 16%	1.091×10^5 (**)	4.574	0.801
8.5	0.690×10^4	1.860×10^4 +/- 18%	0.956×10^5 (**)	5.140	0.837
10.5	0.410×10^4	1.750×10^4 +/- 19%	0.693×10^5 (**)	3.960	0.798

* Error insignificant (less than 0.8%)

** Errors are greater than 50%

The estimated maximum values of the fast and thermal flux values are 4.87×10^5 $n.cm^{-2}.s^{-1}$ and 4.44×10^4 $n.cm^{-2}.s^{-1}$ respectively with aluminium monitors, while a maximum thermal neutron flux value of 1.52×10^4 $n.cm^{-2}.s^{-1}$ was obtained from the analysis of indium monitors. The difference in the two values of thermal neutron fluxes can be explained as follows; neither of the two samples (indium and aluminium) is a pure thermal neutron absorber (Beckurts and Wirtz, 1964), and in the case of aluminium, the total induced activity could have included contributions from interactions with neutrons other than thermal neutrons

through (interferring) reactions like $^{28}Si(n,p)^{28}Al$ and $^{31}P(n,\alpha)^{28}Al$. This is quite possible considering the fact that the aluminium material used is not an analytical grade but a general purpose type.

The relative thermal neutron flux distribution as a function of the thickness of the perspex layer between the source and the monitors showed maximum values at about 2.5 cm moderator thickness with both types of monitors (See figure 4.7).

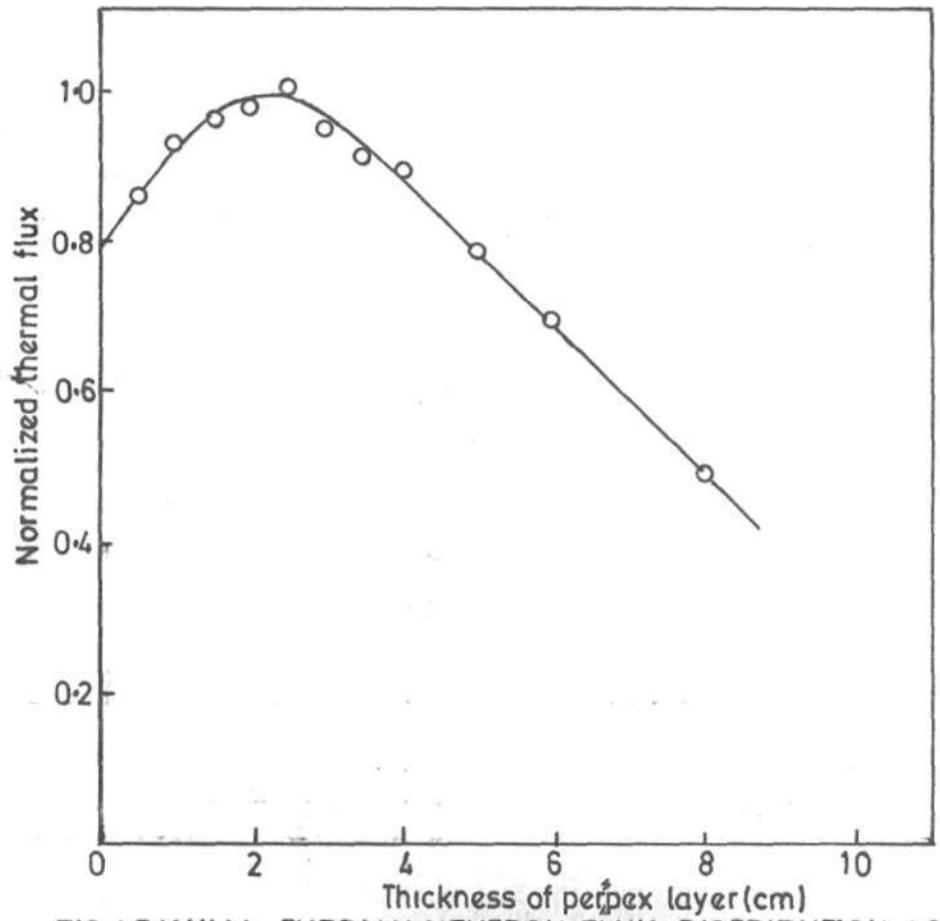
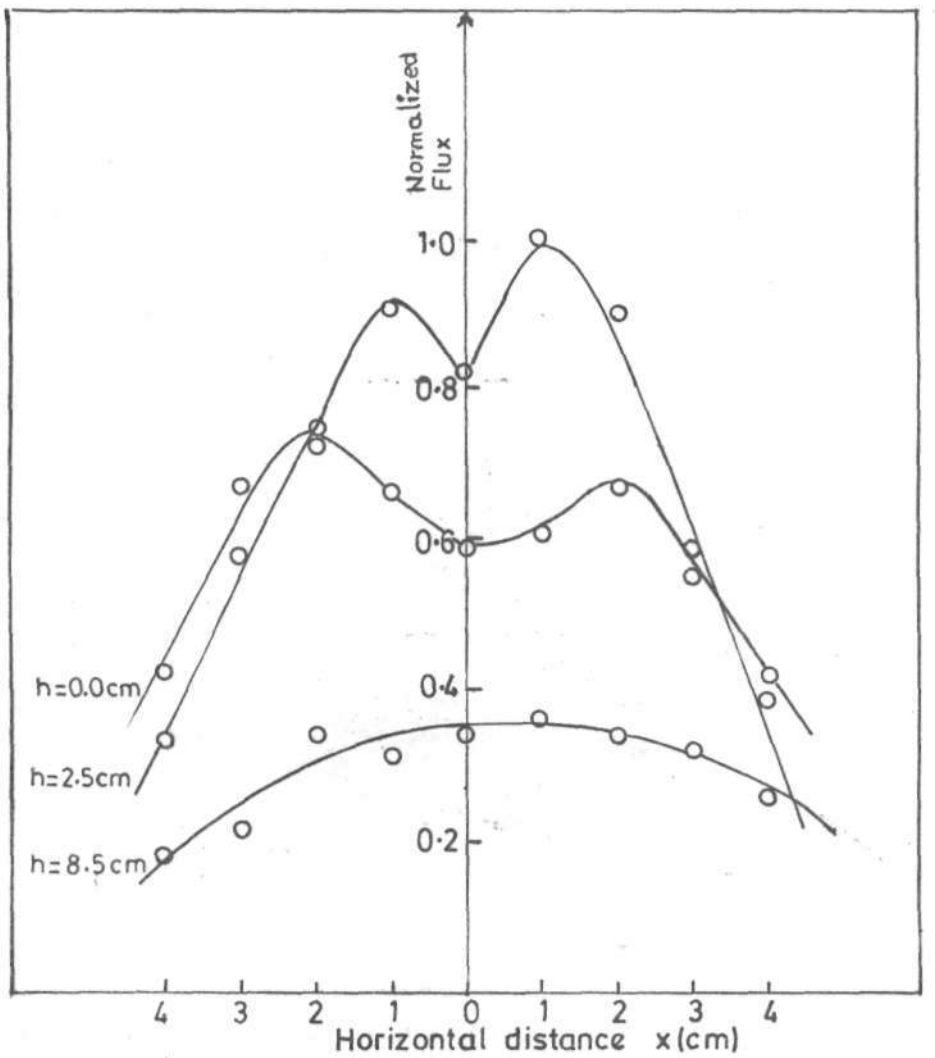


FIG-4.7: AXIAL THERMAL NEUTRON FLUX DISTRIBUTION AS A FUNCTION OF THE THICKNESS OF THE PERSPEX LAYER.
Normalization value = 1.52 X 10^4 n. $cm^{-2}.S^{-1}$ (with indium foil)

Table 4.3 Normalized lateral thermal neutron flux distribution at various vertical distances H from the source. The normalization value is 4.4 x 10^4 n.cm^{-2}.S^{-1}, corresponding to H=2.5 cm

	Normalized flux values 0 (x,H) with % error								
Lateral Distance x (cm)	-4	-3	-2	-1	0	1	2	3	4
Vertical Distance H (cm)									
0.0	0.42	0.66	0.73	0.66	0.59	0.61	0.67	0.59	0.42
	19%	13%	14%	13%	15%	15%	13%	13%	17%
0.5	0.51	0.60	0.84	0.88	0.54	0.86	0.69	0.59	0.40
	16%	14%	12%	11%	13%	13%	12%	15%	17%
1.0	0.34	0.69	0.66	0.73	0.79	0.81	0.71	0.49	0.41
	17%	14%	13%	12%	12%	12%	13%	15%	18%
1.5	0.40	0.68	0.77	0.86	0.83	0.84	0.74	0.73	0.36
	17%	12%	11%	11%	13%	10%	11%	14%	18%
2.0	0.35	0.59	0.50	0.84	0.74	0.83	0.72	0.61	0.47
	19%	14%	16%	11%	13%	11%	13%	13%	15%
2.5	0.32	0.57	0.72	0.91	0.82	1.00	0.80	0.55	0.39
	21%	13%	13%	10%	11%	10%	12%	13%	15%
3.0	0.30	0.60	0.55	0.76	0.73	0.85	0.75	0.74	0.40
	19%	13%	15%	12%	13%	13%	12%	15%	19%
3.5	0.29	0.37	0.83	0.83	0.76	0.86	0.72	0.59	0.42
	24%	15%	12%	12%	12%	11%	12%	14%	14%
4.5	0.33	0.49	0.63	0.66	0.63	0.65	0.72	0.43	0.21
	20%	16%	12%	13%	13%	13%	13%	16%	24%
5.5	0.25	0.36	0.50	0.,63	0.47	0.68	0.71	0.46	0.42
	23%	19%	14%	14%	15%	12%	12%	16%	18%
6.5	0.37	0.43	0.43	0.56	0.56	0.50	0.60	0.31	0.29
	18%	17%	16%	12%	15%	16%	14%	22%	24%
8.5	0.18	0.21	0.34	0.31	0.34	0.36	0.34	0.32	0.26
	28%	23%	18%	20%	18%	16%	18%	20%	24%
10.5	0.13	0.23	0.28	0.27	0.33	0.32	0.32	0.29	0.17
	38%	21%	22%	22%	19%	19%	19%	18%	28%

The lateral neutron flux distribution at different heights showed a symmetric distribution about the central axis of the source (See figure 4.8). A flux depression along the axis of the source was also observed. Similar features have been reported by Cox et al, (1968).



FIG·4·8 : LATERAL THERMAL NEUTRON FLUX DISTRIBUTION AT DIFFERENT VERTICAL DISTANCES h.
Normalization value = 4·4 X 10^{4} n.cm^{-2}. S^{-1}

4.4.4 Determination of Neutron flux using diffusion theory

The slow (thermal) neutron flux at the optimal irradiation position (r = 2.5cm) was calculated theoretically, for comparison, using the two-group diffusion theory for an isotopic point source of fast neutrons in an infinite medium. The group theory thermal neutron flux distribution (Beckurts and Wirtz, 1964) at a distance r from the source is given by;

$$\phi_{th}(r) = \frac{QL^2}{4\pi Dr}\left\{\frac{e^{-r/L}}{L^2-\tau_{th}} - \frac{e^{-r/\sqrt{\tau_{th}}}}{L^2-\tau_{th}}\right\} \quad \ldots\ldots \quad 4.4.2$$

where Q is the source's neutron intensity, L is the neutron diffusion length, D is the diffusion coefficient, and τ_{th} is the Fermi age of the neutrons. It is one-sixth of the mean squared slowing-down distance, $\langle r^2 \rangle$, which in turn is defined as the mean of the square of the distances between the source of fast nuetrons and the points at which the neutrons have slowed down to thermal energy.

The diffusion parameters; L, D, and are characteristics of the moderating medium. A set of the values of L (= 2.8 cm) and D(= 0.136) for plexiglass were quoted (Beckurts and Wirtz, 1964). However, the value of τ_{th} was not quoted and it has been estimated from the calculated value of the mean squared slowing-

down distance $\langle r^2 \rangle$ (Appendix iii).

The flux value of 3.2×10^5 $n.cm^{-2}.s^{-1}$ was obtained when the calculated value of $\tau_{th} = 21.1$ cm^2 (Appendix 4), and the quoted values of D and L, were substituted in equation 4.4.2.

The quoted values of L and D could, however, be in error since they are for plexiglass, and not for perspex which is the material used in this work.

CHAPTER FIVE

APPLICATION OF ISOTOPIC NEUTRON SOURCES IN QUALITY CONTROL

5.1 DETERMINATION OF ELEMENTAL CONCENTRATION BY NEUTRON METHODS.

In Industries and other related fields, determination of elemental constituents of (manufactured) products is important for process and quality control purposes. Most conventional chemical and instrumental methods of analysis are tedious and are no longer economic (Garg and Batra, 1986). The trend has been towards nuclear methods especially neutron methods which are rapid and non-destructive. The use of these methods in this regard have been widely reported (Semel and Helf, 1969; Garg and Batra, 1986; and Ciechanowski et al, 1988).

A detailed account of the potential of isotopic neutron sources and the principle of NAA in process and quality control has already been given by Garg and Batra, (1986) while describing the use of various isotopic neutron sources for the determination of elemental concentration in ores, alloys and other industrial samples. Their reports included the development of an on-stream Analyzer for the analysis of certain minerals during extraction and production processes. According to the report, in such analyzer, which are available commercially, a bulk sample is allowed to pass on conveyor belt where it gets

irradiated at one end using an isotopic source, while the counting is performed on the other end using a detector and counting assembly which may be precalibrated to give percentage abundance directly.

Isotopic neutron sources have also been used in the determination of hydrogen and moisture (water) content of certain liquid and solid materials (Semel and Helf, 1969), and in the determination of hydrogen concentration in liquids flowing through pipes (Ceichanowski et al, 1988).

Some of the neutron methods so far employed for hydrogen (moisture) determination include the fast neutron moderation and thermal neutron transmission techniques. The latter is less advantageous (Semel and Helf, 1969) because it requires high intensity of neutrons, like those obtainable from nuclear reactors, to obtain the same precision as obtainable with neutron moderation technique using a moderate isotopic neutron source. Besides these two, neutron reflection method (Csikai, 1987) and epithermal neutron moderation techniques have also been used.

5.2 DETERMINATION OF THE HYDROGEN CONCENTRATION IN LIQUIDS BY NEUTRON THERMALIZATION

5.2.1 Methodology

The fast neutron moderation technique for moisture and hydrogen determination is based on the phenomenon that fast neutrons, on passing through materials of high scattering cross section and stopping power, are slowed down to thermal energies. And, since it has been estblished (Fink, 1936; Amaldi and Fermi, 1936 ; Beckurts and Wirtz, 1964; etc) that the fractional loss in energy when a neutron collides with an atom is greatest for the hydrogen atom. Therefore by allowing a beam of fast neutrons to pass through a series of materials of (known) varying hydrogen densities, a relation could be established between the hydrogen content of these materials and the (corresponding) measured thermal intensities.

The usual experimental approach (Semel and Helf, 1969 and Ciechanowski et al, 1988) is to prepare a standard calibration curve with samples of known hydrogen content and to relate the measured thermal neutron intensities of an unknown sample to this curve. The method of measuring the neutron intensity employed in this work is the foil activation method, using thin foils of indium. However, specific applications requiring faster result may require special measuring devices, for example, in the determination of liquids

flowing through pipe line, Ciechnowski et al, (1988) used a miniature BF_3 filled proportional counter encased in a specially designed housing. Also, Semel and Helf, (1969) had earlier reported the use of BF_3 and lithium iodide Scintillator detector assembly.

INSTRUMENTATION

The major components of the apparatus used include the 5 Curie $^{241}Am/Be$ neutron source. This source strength according to Semel and Helf, (1969), is suitable for laboratory or in-plant quality control purposes. Although relatively smaller isotopic neutron sources have been used, but only for field measurment where large quantity of samples are usually available for assay. The apparatus also include a 400 ml cylindrical plastic container with lid, its diameter (8.0 cm) is slightly less than that of the cavity in the howitzer along which the fast neutrons are directed (See figure 5.1). Other components are; indium foils as thermal neutron detectors, and the gamma-ray spectrometer for the analysis of the radioactivity of the irradiated indium foils. The spectrometer comprises of a 2" x 2" sodium iodide (Thalium activated) detector, coupled with a multichannel Analyzer (MCA), through pre-Amplifier and standard Amplifier. The choice of these components bears in mind the need for mobility and compactness.

CALIBRATION

Equal volumes (400 ml) of four different standard hydrogenous compounds (water (H_2O), ethanol (C_2H_5OH), acetone (CH_3COCH_3), and toluene ($C_6H_5CH_3$)) were poured, one at a time, into the plastic container and then placed in the cavity right on top of the $^{241}Am/Be$ source. An indium foil is fastened to the lid of the container such that it is separated from the fast neutron source by the column of the hydrogenous substance (figure 5.1.) After two hours of irradiation the indium foils were counted for ten minutes and the count rates were obtained (Table 5.1).

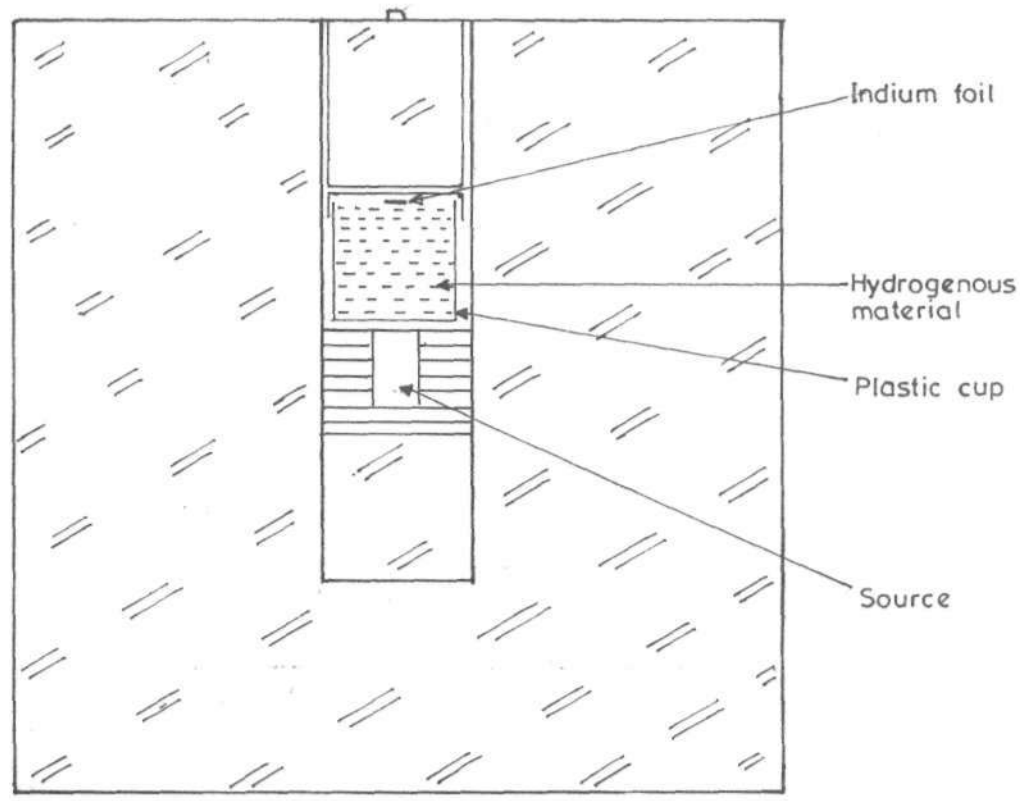
Calibration curves of count rate against hydrogen density (gm cm^{-3}),and count rate against H/C ratio were drawn (figures 5.2 and 5.3). The calibration graphs appear linear for the range of hydrogenous samples used.

Table 5.1 Thermal activation analysis of indium samples due to neutron thermalization by materials of known hydrogen concentration.

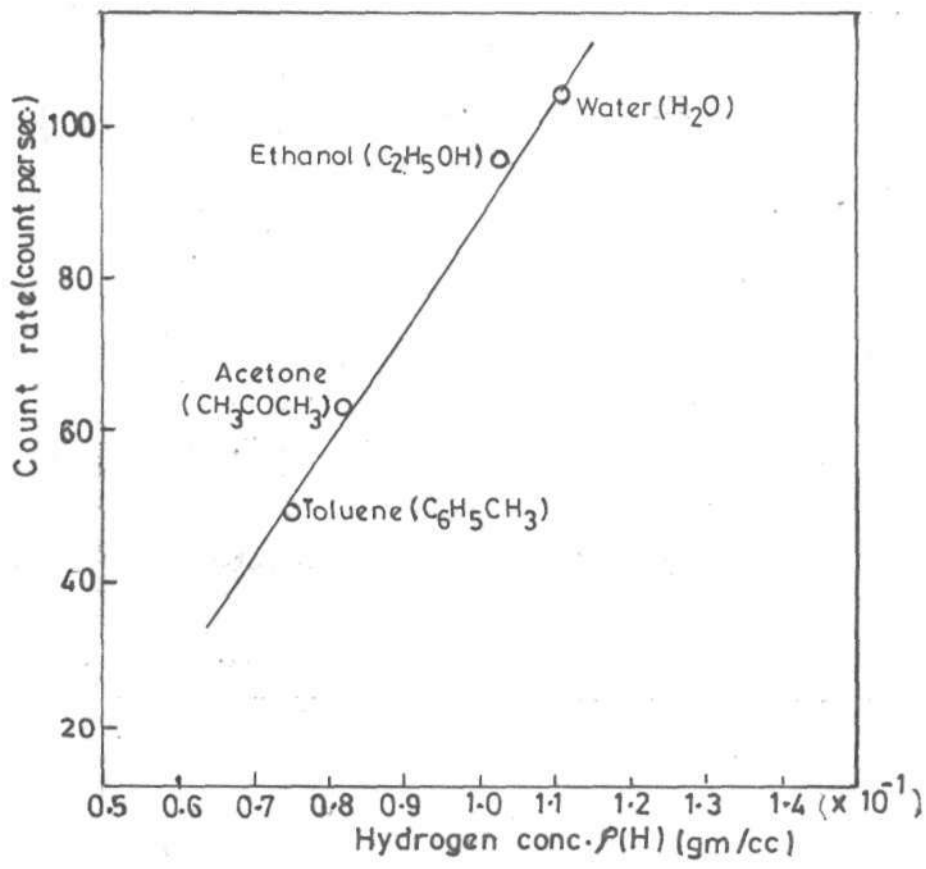
Hydrogenous material	Density (gm/cc)	Hydrogen Density (gm/cc)	H/C ratio (by wt.)	Count rate (count/sec.)
Water (H_2O)	1.0000	1.1111×10^{-1}	-	104.338
Ethanol (C_2H_5OH)	0.7897	1.0300×10^{-1}	0.2500	96.346
Acetone (CH_3COCH_3)	0.7920	0.8193×10^{-1}	0.1667	63.464
Toluene ($C_6H_5CH_3$)	0.8650	0.7522×10^{-1}	0.0952	49.343

APPLICATION

The neutron moderation technique as a method of determining hydrogen and moisture content of materials for quality control processes have been adapted in various ways for monitoring concentrations of various compounds whose concentrations are directly proportional to their hydrogen contents. The feasibility of its application in the industries for monitoring certain (compounds and) elemental constituents of manufactured products at various stages of their production is being envisaged. One of the most important advantages of this method is that it offers on the spot assessment without distrupting the normal (continuous) process flow. Equally important is its non-destructive effect on the material being analysed. It should also be noted that specific



FIG·5·1: THE HYDROGEN CONCENTRATION DETERMINATION EXPERIMENTAL SET UP



FIG·5·2: CALIBRATION GRAPH

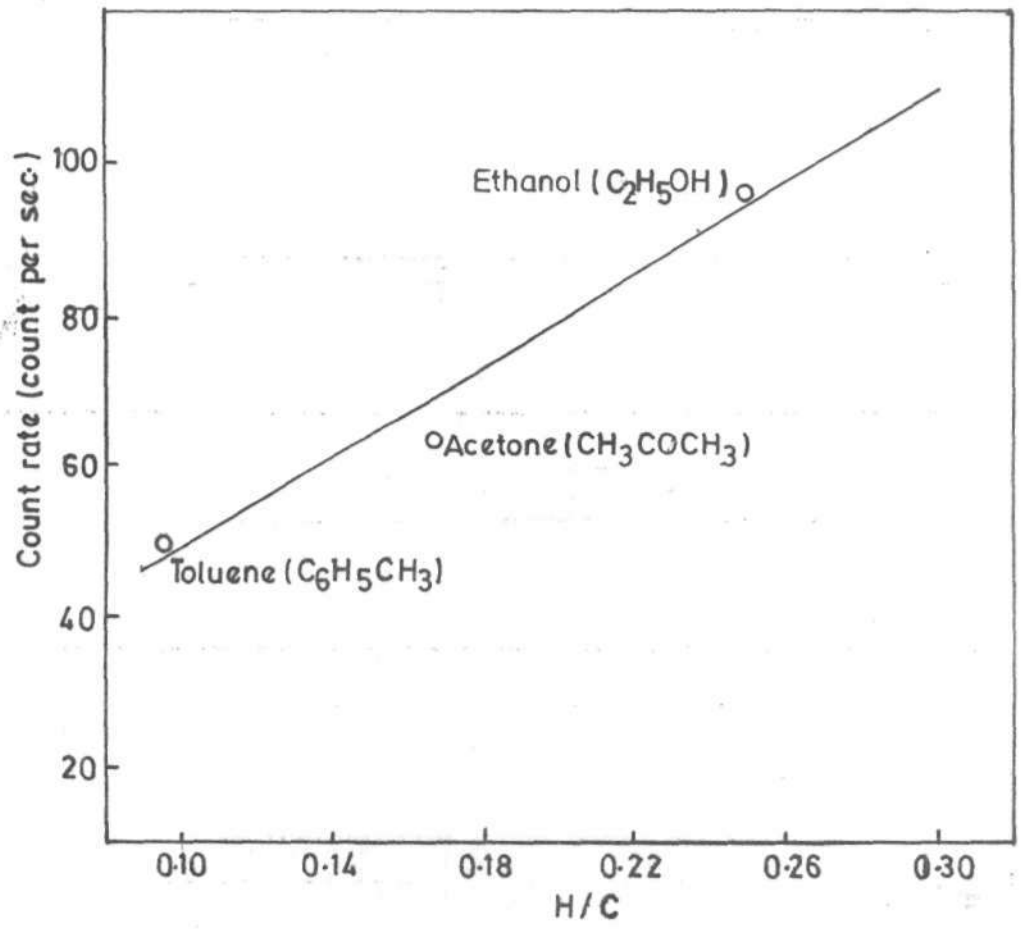


FIG. 5·3 : CALIBRATION GRAPH OF COUNTRATE AGAINST H/C RATIO.

applications often require unique designs and experimental set up as well as calibration procedure. However, all designs must provide adequate shielding measures, in addition to any other efficiency enhancement measures.

For future work, an important application of the above method is in the measurement of the hydrogen content of petroleum products. The calibration graph (figure 5.2 and 5.3) can be used to determine the hydrogen content of samples of petroleum products such as petrol, kerosine, diessel, and low pour fuel, etc, by taking equal volumes (400ml) of each of these samples and obtaining the count rates in the activated indium foils. The calibration graph of count rate against H/C ratio can be used to obtain the H/C ratio of these petroleum products. These two investigations (i.e. Hydrogen concentration and H/C ratio determinations) could reveal information which could be used in estimating the calorific value of each of these products (Hydrocarbons), and consequently the efficiency of the refinery processes by comparing such results to standard values.

This test could also be used to investigate and check the degree of adulteration of the products. It is often being alleged, for example, that petrol mixed with diessel are sold as pure petrol,(similarly for the

remaining products). Therefore by mixing petrol and diessel in various proportions and obtaining the count rate for each mixture, a calibration graph of count rate versus petrol percentage can be drawn. Similarly for the other products. By using these graph, the degree of adultration, or otherwise, of these products from any source (filling stations, etc) could be determined qualitatively.

This method could also be employed in identifying soils containing oil, or soils close to an oil zone. This is done by extracting the organic content of the soil and doing the NAA of the extract.

Finally it should be noted that there is the possibility for scattered thermal neutrons whose origin are not from the sample striking the detector. This is taken care of by carrying out background measurement, with the cylindrical container empty and subtracting this value from each gross measurement with sample. Also, when two miscible liquids are mixed, error could occur due to "volume Effect" (Semel and Helf, 1969). The volumes under consideration in this analysis are, however, relatively small and error resulting from volume effect could be ignored. A more serious limitation could arise if the samples being analysed contain other varying elements, other than hydrogen, which could have resulted to the observed

neutron moderation. In this case the neutron moderation technique may not supply sufficient accuracy (Semel and Helf, 1969), and further analysis may be required.

CHAPTER SIX

RESULTS, DISCUSSION AND CONCLUSION

6.1 Results and Discussion

The results obtained from the lateral neutron distrbution monitoring revealed that for a particular vertical distance value, the lateral neutron distribution is nearly symmetrical about the central axis of the source. This could imply a uniform distribution of the materials within the active zone of the source. With this observation many samples (or samples and standard) can be activated simultaneously in similar neutron field by just placing them at positions equidistant from the central axis of the source.

A sample holder was constructed, that can accomodate four samples of up to $25cm^3$ each, although, it can be modified to accomodate more so that larger quantities of samples can be irradiated and consequently the sensitivity of the analysis would be enhanced.

The lateral thermal neutron distribution showed a depression along the axis of the source, this feature has been attributed to various perturbations, especially to that due to the source itself.

The axial neutron flux distribution was monitored using two different probes; Aluminium and indium foils.

The neutron distribution curves obtained with the two foils showed that the optimum thermal neutron flux is obtained when about 2.5cm thickness of the moderating material (perspex) is separating the foils from the source. The difference between these two maxima values (4.4×10^4 $n.cm^{-2}.S^{-1}$ with the $^{27}Al(n,\gamma)^{28}Al$ reaction; and $1.5 \times 10^4 n.cm^{-2}s^{-1}$ with the $^{115}In(n,\gamma)^{116}In$ reaction) has been partly attributed to the impure state of the aluminium samples used (Technical grade samples), which could have led to over estimation due to contributions from interfering reactions like $^{28}Si(n,p)^{28}Al$ and $^{31}P(n,\alpha)^{28}Al$. Inspite of this shortcoming there is still an appreciable agreement between the two results in their pattern of distribution, this goes a long way to confirm the suitability of aluminium and indium detectors for neutron distribution measurement. The experimental results on one hand can be comparable to the one obtained theoretically using the diffusion theory (equation 4. 4.2) on the other hand, if one considers the uncertainties in the data, particularly the +/-10% uncertainty in the source intensity (Q) which is used in the calculation.

Explanations on the observed axial neutron Distribution

It was observed that after the initial sharp drop in the fast neutron flux, very close to the source, subsequently at relatively far distances from

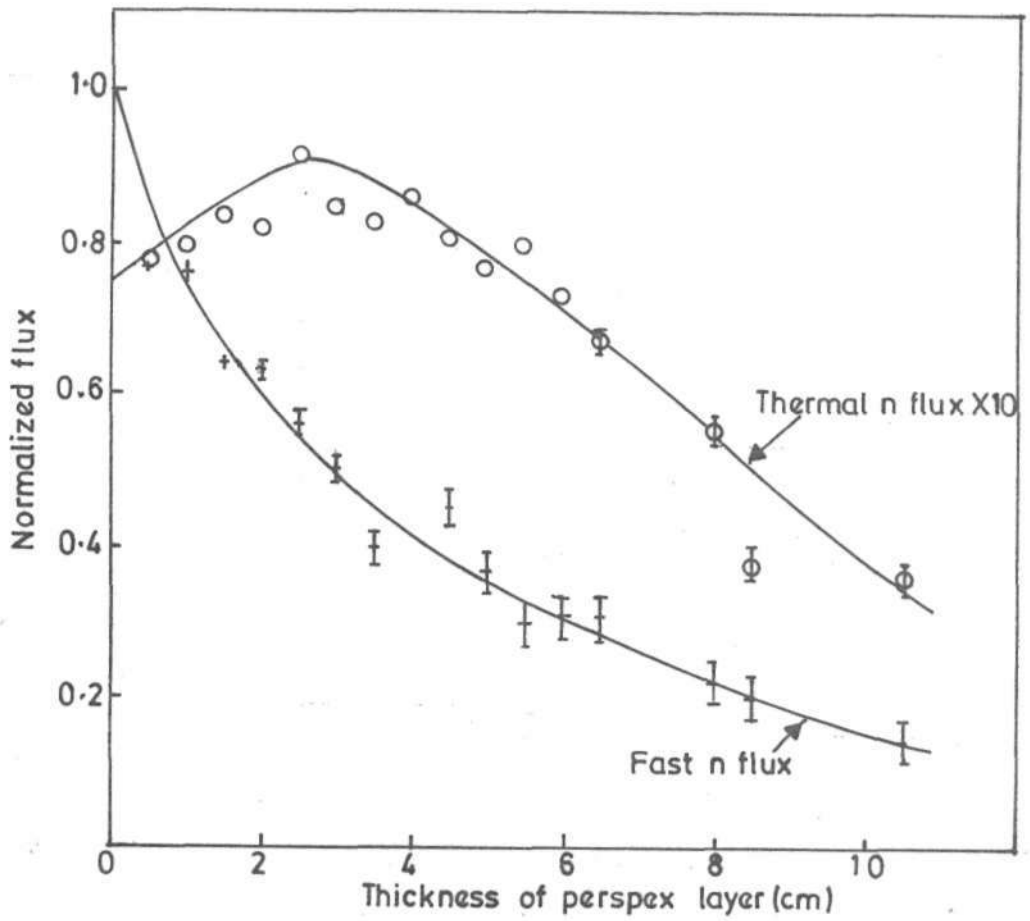


FIG.6.1: FAST AND THERMAL NEUTRON FLUX DISTRIBUTIONS AS FUNCTIONS OF THE THICKNESS OF THE PERSPEX LAYER. NORMALIZATION VALUE = 4.87 X 10^5 n.cm^{-2} s^{-1} (WITH ALUMINIUM FOIL)

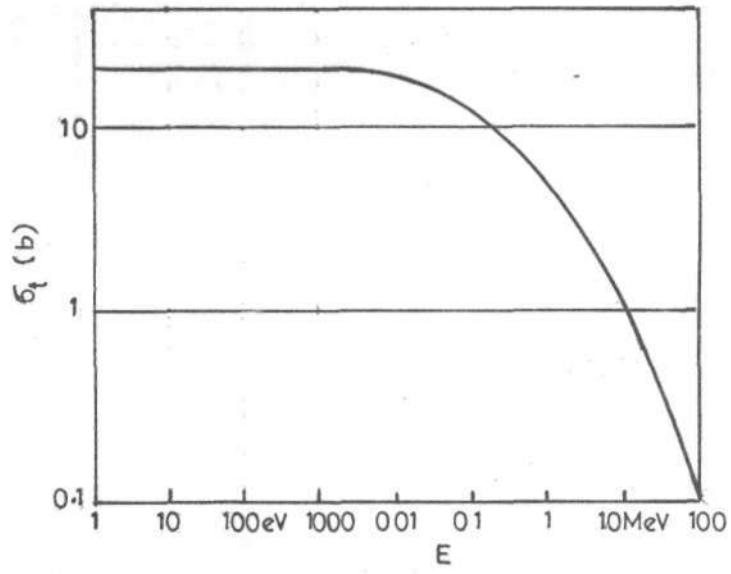


FIG.6.2: TOTAL CROSS SECTION OF HYDROGEN AS A FUNCTION OF NEUTRON ENERGY (Beckurts et al, 1964)

the source one could still detect a slowly varying and appreciable amount of fast neutrons. Whereas the slow (thermal) neutron flux curve is almost a linearly decreasing one at the corresponding distances (see figure 6.1). This behaviour, of fast neutrons, is often called the "neutron hardening" effect (Beckurts and Wirtz, 1964 and Amaldi and Fermi, 1936), it has been explained on the basis that the scattering cross section of hydrogen falls off very sharply with neutron energy (Figure 6.2). Hence, if a neutron from the source once collides and experiences a large energy loss, the cross section for additional collision is large. Thus the probability of such neutrons being moderated and finally absorbed near the site of its first collision is large. The slow (thermal neutron flux should therefore have a maximum value at a distance relatively close to the source when the latter is installed in a hydrogenous material, as observed in this work and earlier works (Rieppo, 1988; Csikai, 1987; Op De Beek, 1986).

Also, since the cross section for the first collision increases with decreasing energy, the more energetic neutrons in the primary spectrum are attenuated less. And with increasing distance from the source, these energetic neutrons become more and more important in determining the spreading out of the neutrons. So at large distances the neutron flux is

predominantly due to neutrons that have made no collisions or at most a few small angle collisions that produce only small energy loss (Beckurtz and Wirtz, 1964). This energy dependent scattering cross section (phenomenon) explains the optimum thermal flux value at about 2.5cm away from the source, and a subsequent fall in themal flux values with increasing distance. It also justifies the persistence of a fast neutron field at large distances from the source as observed (fig. 6.3 and 6.4).

Calculating the Average Neutron Flux within a Sample

The axial thermal neutron flux distribution within the irradiation cavity was fitted to a third order polynomial (appendix iii) given by

$$\phi(x) = 11871.40 + 3088.32x - 909.063x^2 + 58.339x^3 \quad \ldots\ldots\ldots\ldots 6.1$$

where x is the distance between the source and the sample, measured along the axis of the source.

The average neutron flux within a sample of thickness h (measured along the direction of x) can then be estimated/calculated using the following relation;

$$\bar{\phi} = (1/h)\int_0^h \phi(x)\,dx \quad \ldots\ldots\ldots\ldots\ldots 6.2$$

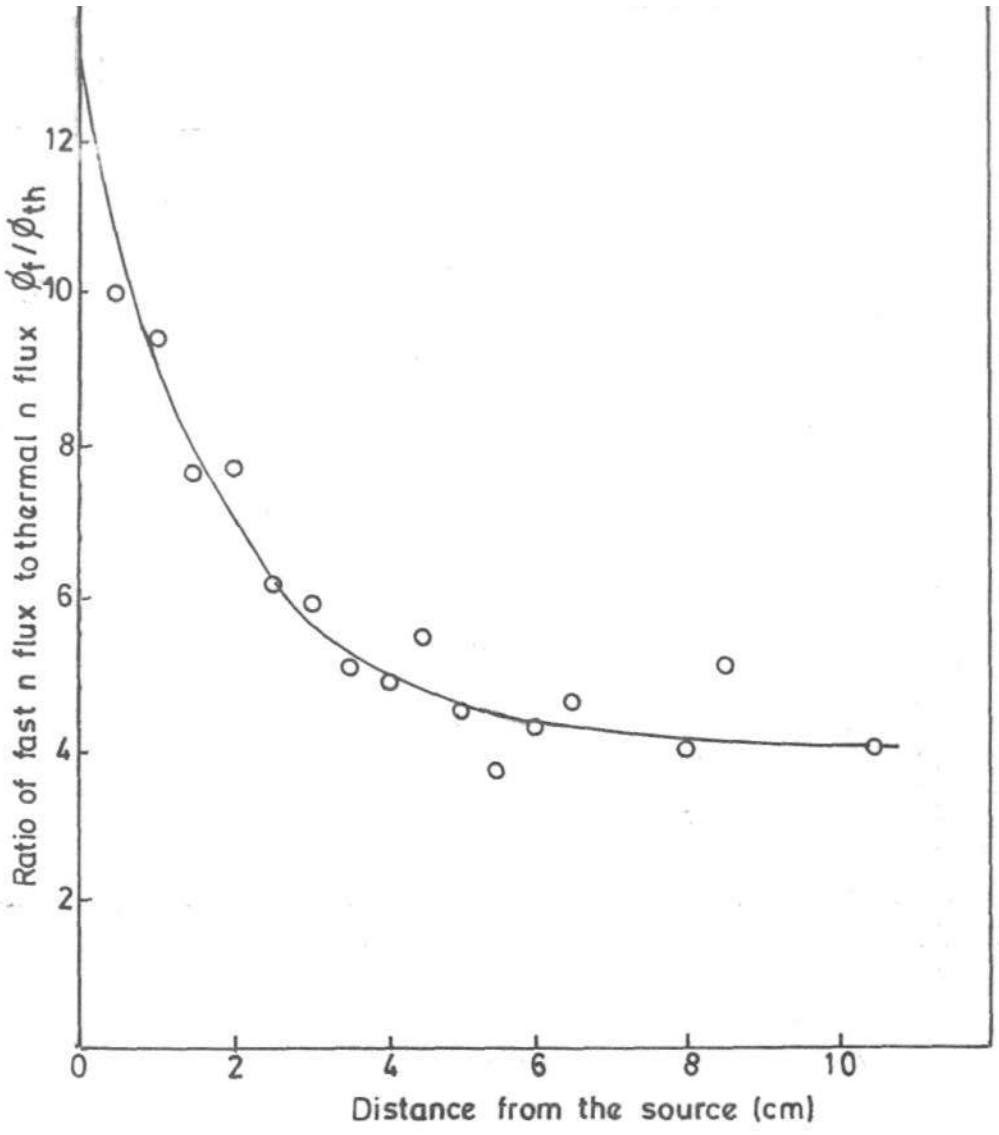


FIG.6.3: RATIO OF FAST n FLUX DISTRIBUTION TO THERMAL n AS A FUNCTION OF THE VERTICAL DISTANCE FROM THE SOURCE

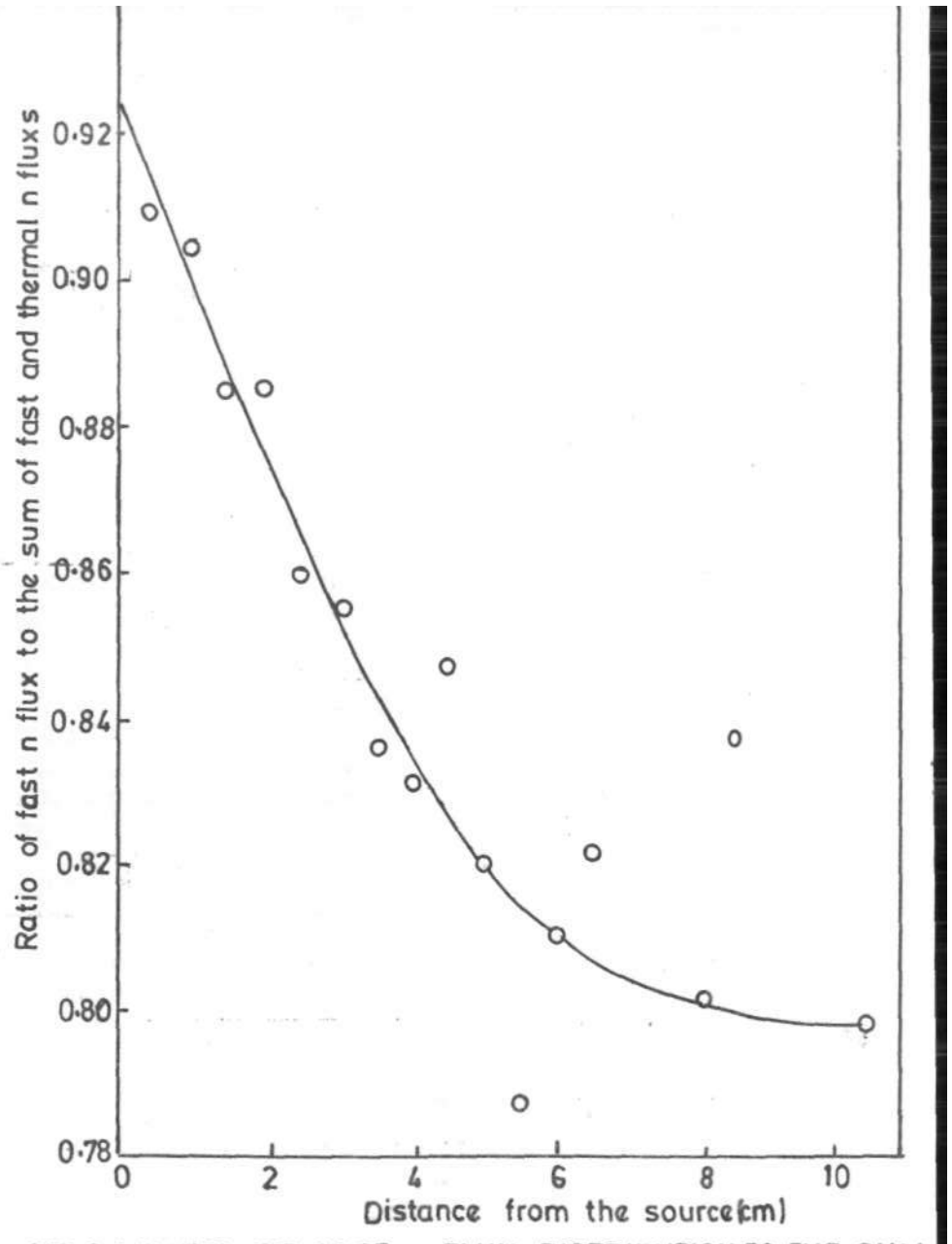


FIG.6.4: RATIO OF FAST n FLUX DISTRIBUTION TO THE SUM OF FAST AND THERMAL n FLUXES AS A FUNCTION OF THE VERTICAL DISTANCE FROM THE SOURCE

Equation 6.2 is a simplified representation of the complex spatial neutron distribution within the cavity. The real neutron field can only be adequately described by an equation which, in addition to being position dependent, must also be directional as well as energy dependent (i.e $\phi(r,\Omega,E)$). Such a relation should also consider the randomness of the neutron diffusion within the field, rather than assuming a uniform and parallel beam of neutrons.However, the assumption of energy independence could be justified within a moderated neutron field if the sample is irradiated under a cadmium cover, in which case, the sample then experiences a homogenous (thermal) neutron field, equivalent to a constant energy neutron field. It could also be argued, that while some neutrons which are originally within the parallel beam from the source, are being scattered off, so also are others outside the original beam, being scattered in, so that at the end very little is lost by our assumption of a parallel beam as against the randomly diffused one.

The lateral flux variation across the samples' cross sectional area is almost negligible over a few centimeters. Thus, a one dimensional neutron distribution can safely be assumed, especially when samples of small cross sectional areas are being irradiated.

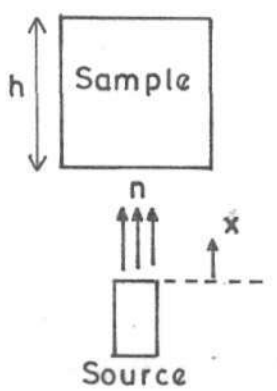


FIG. 6.5: SOURCE - SAMPLE ARRANGEMENT IN THE IRRADIATION CAVITY

Thus, inspite of its simplicity, equation 6.2 is still useful, particularly in the comparative neutron activation analysis. It can, however, be modified to approach a more realistic representation of the real neutron flux distribution by taking some of the mentioned factors into considerations.

6.2 CONCLUSION AND RECOMMENDATION

The characterization of the neutron field in the irradiation cavity of a five - Curie $^{241}Am/Be$ isotopic neutron source installed in a 'howitzer' has been carried out. The nearly symmetric lateral neutron distribution at various heights is attributed to a possibly uniform distribution of the active materials within the source. The advantage of this neutron behaviour has also been discussed, namely possibility to irradiate large quantity of samples and consequently, enhanced sensitivity in results of analysis.

The lateral flux distribution curves also showed depressions along the axis of the soure, the intensity of this depression reduces with increasing distance from the source. This feature has been attributed to pertubation due to the source itself, among others.

A characteristics optimum value of the thermal neutron flux was at about 2.5 cm from the source with the two detectors: aluminium (using the ^{27}Al (n,Y

)^{28}Al reaction) and indium (using the ^{115}In(n,γ)^{116}In reaction). The flux values obtained from the analysis of the two detectors showed appreciable agreement (4.4 x 10^4 n.cm^{-2}.s^{-1} with aluminium and 1.5 x 10^4 n.cm^{-2}.s^{-1} with indium) despite various limitations imposed, especially, by the impure form of the aluminium monitors used.

The experimentally obtained flux values are of the order of 10^4 while the calculated value is of the order of 10^5. This discrepancy has been largerly attributed to the uncertainties in the various data used, especially the +/-10% uncertainty in the value of the intensity of the source which was used in the calculation.

The possibility of coming up with a means of calculating the average neutron flux value within an irradiated bulk sample has been the major drive in this work. This has been achieved, to a large extent, through the fitting of the axial flux distribution curve to a third order polynomial; $\phi(x) = 11871.40 + 3088.32x - 909.063x^2 + 58.339x^3$, which can be substituted into the relation for calculating the average flux within a sample that is being irradiated (equation 6.2).

The axial neutron flux distribution (fig. 4.7) shows that the flux value at 0 cm from the source is

about 80% of the optimum value obtained at about 2.5 cm from the source, and at about 5 cm from the source the flux value again decreased back to about 80% of the maximum value. Thus, this region (from 0 cm to 5 cm) can be referred to as the region of maxima flux values. It is therefore recommended that samples should be irradiated within these region, and preferrably, with the centres of the samples coinciding with the 2.5 cm optimum (flux value) position.

The measurement of hydrogen concentration and the ratio of hydrogen to carbon in certain (hydrocarbons) hydrogenous materials, using the fast neutron moderation technique was also demonstrated with the $^{241}Am/Be$ Isotopic source.The applicability of this technique in continuous industrial process and quality control, as well as in other fields noted for requiring rapid or on-the-spot assessment techniques, particularly in the on-stream monitoring of the various stages of production in the oil refinery and other chemical industries, was also discussed.

Based on the 10^4 thermal neutron flux, the facility is now ready for the analysis of soil and geological samples for elements with large cross section for thermal neutron such as Mn (others are as listed in table 6.1).

Table 6.1

Neutron activation analysis of ores, alloys and industrial samples using isotopic sources
[Garg and Bartra, 1986]

Element determined	Nuclear reaction	Half-life of the product nucleus		Type of ore or sample analyzed
Thermal neutrons				
Aluminium	$^{27}Al(n, \gamma)^{28}Al$	2.3	m	Bauxites, aluminosilicates, sedimentary, rocks, coal ash and dressing products of ores, various alloys, chrome ores
Copper	$^{63}Cu(n, \gamma)^{64}Cu$	12.8	h	Copper ores, copper bearing drill core and other products
	$^{65}Cu(n, \gamma)^{66}Cu$	5.1	m	Copper ores
Manganese	$^{55}Mn(n, \gamma)^{56}Mn$	155	m	Pyrolusites, rhodenite and other manganese ores, sedimentary rocks and soils, multimetallic ores, ferro-manganese production, samples from ore dressing
Vanadium	$^{51}V(n, \gamma)^{52}V$	3.75	m	Titanium-magnetite ore, carnotite limonite, ferrovanadium
Selenium	$^{76}Se(n, \gamma)^{77m}Se$	17.5	s	Industrial samples, Cu-Zn ore
Rhodium	$^{103}Rh(n, \gamma)^{104m}Rh$	4.3	m	Metal concentrate
Silver	$^{107}Ag(n, \gamma)^{108}Ag$	145	s	Silver ore, lead deposits
	$^{109}Ag(n, \gamma)^{110}Ag$	24.4	s	Phosphorite ore, concentrates
Indium	$^{115}In(n, \gamma)^{116m}In$	53.7	m	Multimetallic ores and concentrates, sphalerite, intermetallic fusion
Dysprosium	$^{164}Dy(n, \gamma)^{165}Dy$	139	m	Phosphorite ore, apatite
Hafnium	$^{178}Hf(n, \gamma)^{179m}Hf$	18	s	Zircons, zircalloy
Tungsten	$^{186}W(n, \gamma)^{187}W$	23.9	h	Wolframite, hibnerite, tungsten concentrates, scheelite and fluorite
Rhenium	$^{187}Re(n, \gamma)^{188}Re$	16.8	h	Molybdenite concentrate
Irridium	$^{193}Ir(n, \gamma)^{194}Ir$	17.4	h	Metal concentrate
Gold	$^{197}Au(n, \gamma)^{198}Au$	2.7	d	Gold ores, metal concentrate
Fast neutrons				
Aluminium	$^{27}Al(n, p)^{27}Mg$	9.46	m	Bauxites, chrome ores and concentrates, coal
Fluorine	$^{19}F(n, \alpha)^{16}N$	7.14	s	Fluorite ores, concentrates, apatite fluorspar, cryolite and plastics, filters, glasses
Silicon	$^{28}Si(n, p)^{28}Al$	2.3	m	Chromeores, bauxites, coal ash, iron ore, concentrates and slags, smelter slag, taconite ore, coal
Chromium	$^{52}Cr(n, p)^{52}V$	3.75	m	Chrome ores, slag concentrates and tailings
Iron	$^{56}Fe(n, p)^{56}Mn$	155	m	Taconite ore and other iron ore
	$^{56}Fe(n, n'\gamma)$	Inelastic neutron scattering		Iron ores, concentrates and filings
Barium	$^{137}Ba(n, n'\gamma)$	Inelastic neutron scattering		Pure materials

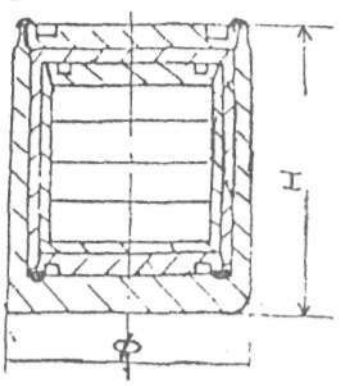
Table 6.1 (cont'd)

Element determined	Nuclear rection	Half-life of the product nucleus		Type of ore or sample analyzed
Uranium	$^{238}U(n, \gamma)^{239}U$	23.5	m	Uranium ore
Combination of thermal and fast neutrons				
Aluminium and	$^{27}Al(n, \gamma)^{28}Al$	2.3	m	Bauxite, nepheline, kaoline, manganesian rocks, coal
Silicon	$^{28}Si(n, p)^{28}Al$	2.3	m	
Prompt gamma counting				
Copper	$^{65}Cu(n, \gamma)$	–		Porphyrin copper deposits, mining and mill analysis
Iron	$^{56}Fe(n, \gamma)$	–		Iron ores, concentrate and filings
Nickel	$Ni(n, \gamma)$	–		Low grade nickel ore, nickel laterites, pentlandite
Gold	$^{197}Au(n, \gamma)$	–		Gold ores
Resonance neutrons				
Silver	$^{107}Ag(n, \gamma)^{108}Ag$	2.5	m	Ore dressing products
	$^{109}Ag(n, \gamma)^{110}Ag$	24.4	s	Ore dressing products

Appendix I

Specification
of Am-241/Be Neutron Source
I. A. E. of China

1. Serial number: AB-88/2031
2. Nuclide: Am-241 Half-life 433Y
   Nominal activity: 185 GBq (5Ci)
   Target material: Be
   Form of active zone: Compacted mixture of americium oxide with beryllium metal.
3. neutron emission: 1.29 $\times 10^7$ n/s $\pm$ 10%
   Measurement date: 14 April, 1988
4. Encapsulation
   Outside dimension: $\phi 30 \times 48$ (mm)
   Material: Stainless steel (1Cr18N:9Ti)
   Encapsulation: Doubly encapsulated in stainless steel sealed by argon arc welding (See Figure)
5. Quality Control:
   Classification ISO 2919/C56535
   Wipe test A
   Bubble test D
   Immersion test L
6. Transportation container:
   U. S. A /9163/B(U)
   Refrence No: 86RHXE-099/GN-710
7. Attention:

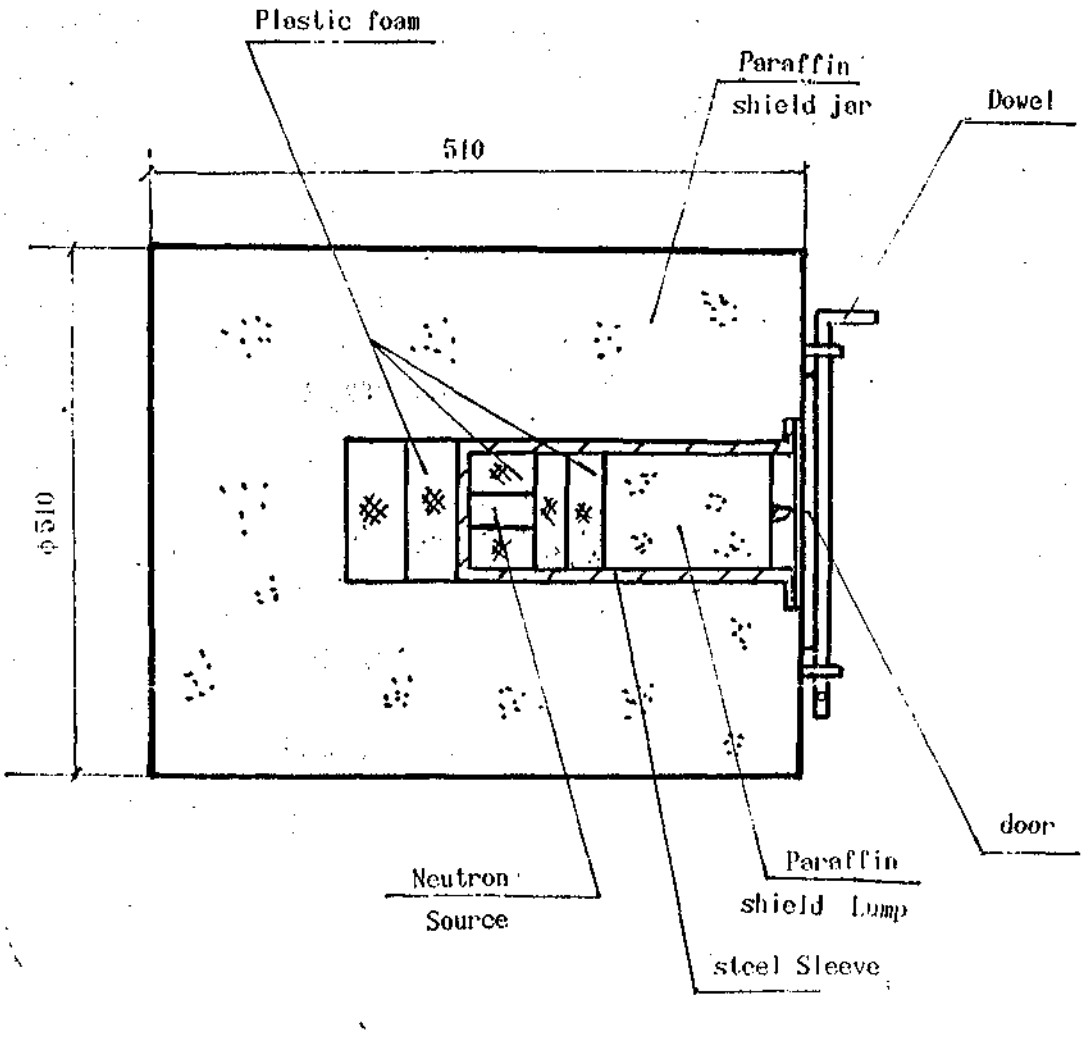


The source is strictly forbidden to be opened. If there is any qustion,please contact with I. A. E of China

Transportation container instruction

* Transportation container must be locked during transportation.

** When neutron source is taken out from steel sleeve, a long crucible tongs should be used.



放射性物质剂量证明书

Certification of Radioactive Package Dose

同位素名称 Radioisotope	镅—铍中子源 ^{241}Am—Be neutron Sources	强度（活度） Activity	1.2×10^7（中子/秒） 1.2×10^7（n/sec）
状态 State	固体　液体　气体 Solid, Liquid, Gas		
射线类型 Radiation Type	γ射线、中子 ✓ γ-ray 、neutron		
剂量率 Dose Rate	货包表面剂量 On the Surfare of Package		25mrem/h
	距一米处剂量 At the Distance of 1 Meter		
编号 Number			
件数 No. of Package	1	等级 Type of Packaging	Ⅲ

此产品经辐射安全检查合格，符合国际原子能机构1984年修订的《放射性物质安全运输规程》有关要求。

These packages have been checked for its radiation safety according to " Regulations for the Safe Transport of Radioactive Materials " revised by IAEA in 1984. It meets all requirements.

检查机关（盖章）
Inspection Institution

检查人员
Signature　Gan Qi Lun

签发日期
Date　1989.2.18

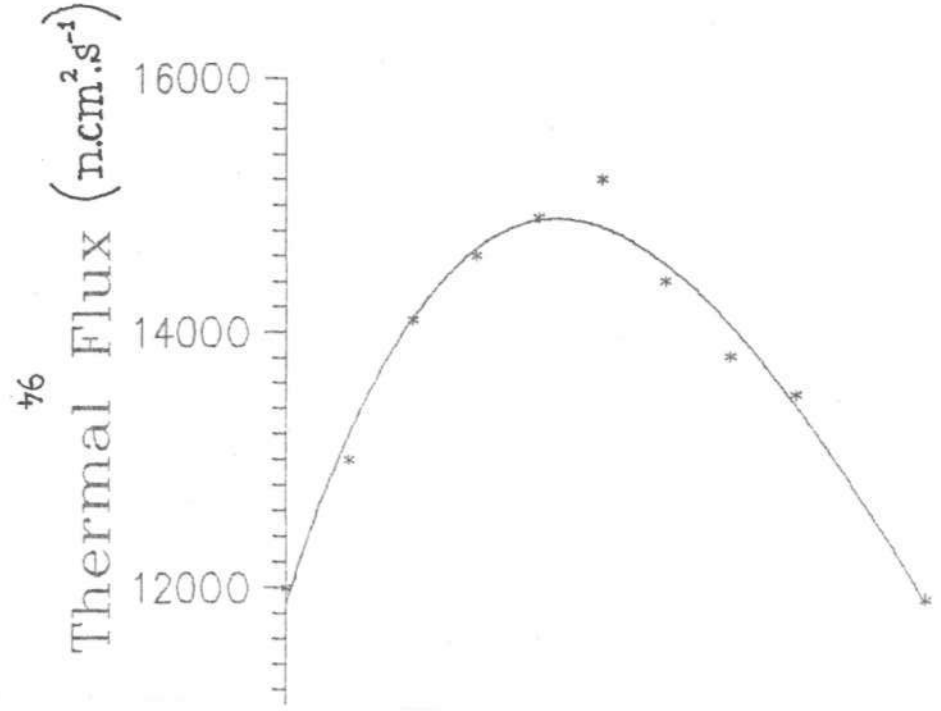


Table A2.1 Axial thermal neutron flux distribution curve fitting.

Menu 1.3.6.6 Polynomial Fit Statistics
GRAPHER Copyright (C) 1988 Golden Software, Inc.

Total points = 10 Current data file: 1 Fitting Interval Limits
Points in fit interval = 10 AMIDU 0 to 5

Degree	Sums of Squares of Residuals	PerCent of Residuals about Mean Explained	Polynomial Coefficients
0	1.1804E+007	0	11371.4
1	1.17816E+007	0.19	3088.32
2	611136	95	-909.063
[illegible]	[illegible]	98	58.339

Appendix III

The mean squared slowing down distance calculation

If one considers a point source of fast neutrons in an infinite (moderating) medium which produces a thermal neutron flux, $\phi_{th}(r)$, then the mean squared slowing down distance, $\langle r^2 \rangle$, from the source at which the thermal neutron is absorbed is given by relation

$$\langle r^2 \rangle = \int_0^\infty r^2 \Sigma_a \phi_{th}(r) 4\pi r^2 dr \Big/ \left(\int_0^\infty \Sigma_a \phi_{th}(r) 4\pi r^2 dr \right) \quad \ldots\ldots \text{A3.1}$$

where a is the total absorption cross section. This equation can be re-written (Amaldi and Fermi; 1936; Rush, 1948, and Beckurts and Wirtz, 1964) as:

$$\langle r^2 \rangle = \frac{\int_0^\infty A(r).r^4 dr}{\int_0^\infty A(r).r^2 dr}$$

where A(r) is the activity of a foil irradiated at a distance r from the source.

The activities A(r) of thin foils were measured in the neighbourhood of the source, at large distances, however, the activity values were obtained by extrapolation (Wade, 1958), by assuming that the flux $\phi(r)$ is directly proportional to $(e^{-r/\lambda})/r$, which also implies that $A(r).r^2$ is proportional to $e^{-r/\lambda}$.

$$\text{i.e} \quad A(r).r^2 = Ce^{-r/\lambda} \qquad \ldots \text{A3.2}$$

The mean free path λ (5.4 cm) was determined from the slope of the curve of In $A(r).r^2$ against r at a large distance from the source (figure A3.1). Using the value of λ and, A(r) at (r = 14cm), the constant C was also evalauted (see fig. A3.1). The values of and C are then used to obtained various values of $A(r).r^2$ and $A(r).r^4$ at other large values of r. (Table A3.2).

The extrapolated graphs of $A(r).r^2$ vs. r and $A(r).r^4$ vs.r were plotted using both tables (figures A3.2 and A3.3). The areas under the curves were calculated by numerical integrations using the simpson and tripezoidal rules, and the quotient of the integrals gave the value of the mean squared slowing down distance ($\langle r^2 \rangle$ = 173.7 cm^2).

The mean squared slowing down distance $\langle r^2 \rangle$ is related to the thermal Fermi age (t_{th}) by:

$$\langle r^2 \rangle = 6M^2 = 6(t_{th} + L^2) \quad .....A3.3.$$

where M is called the Migration length and L is the diffusion length.

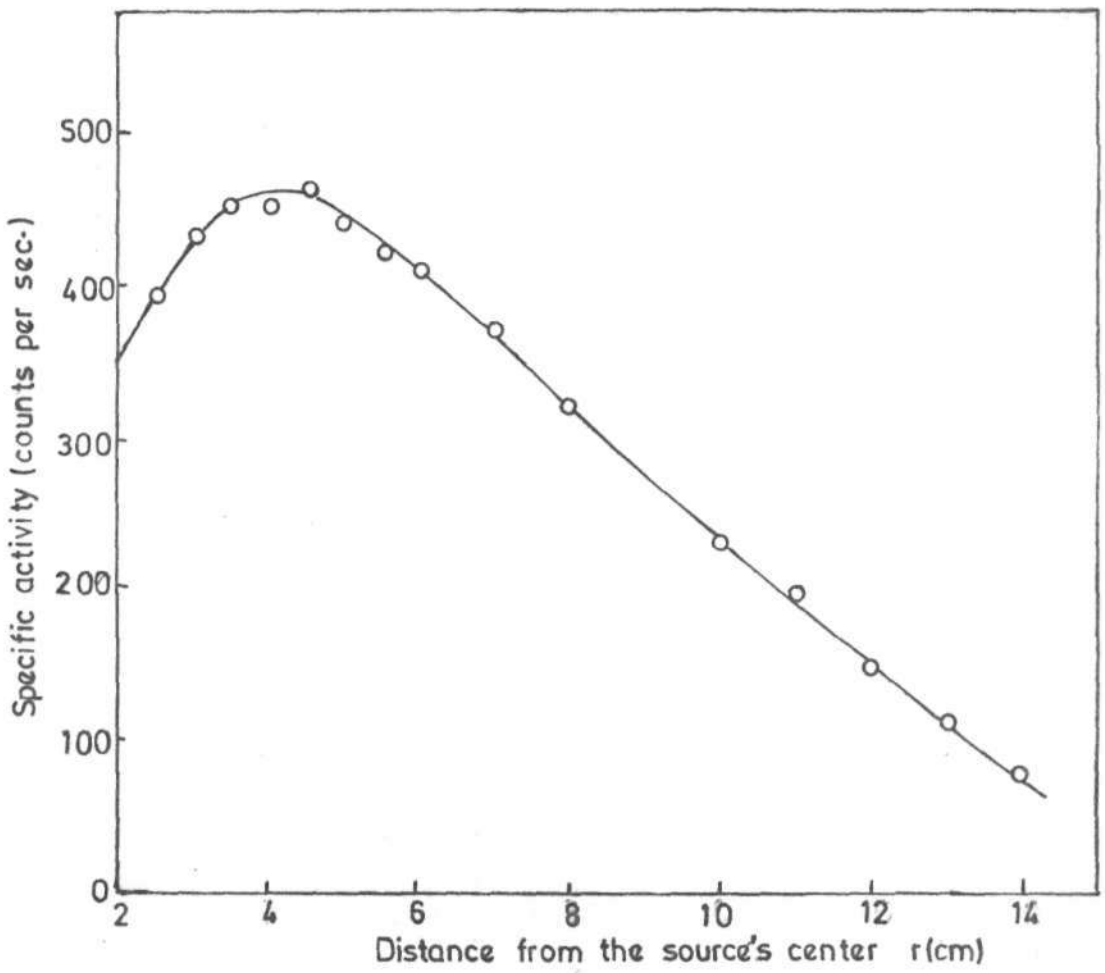
From equation A3.3, the Fermi age (T_{th}) was obtained to be 21.1 cm^2 using the quoted (Beckurts and Wirtz, 1964) value of L = 2.8 cm for plexiglass.

Table A3.1 Measured Activity at various distances in the neighbourhood of the source.

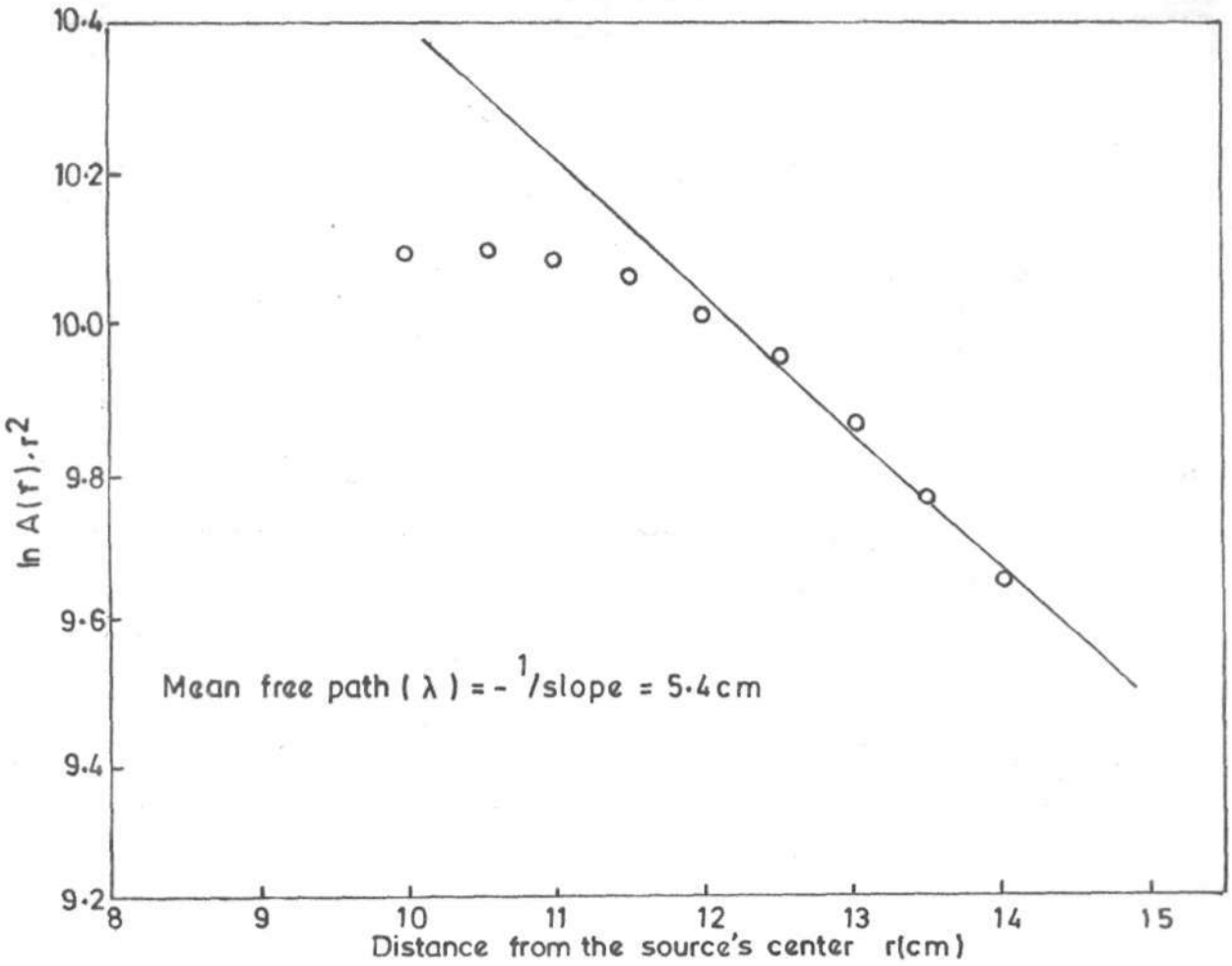
Distance (cm)	Specific Activity A(r) (counts/sec)
2.0	359.36
2.5	395.12
3.0	430.37
3.5	450.01
4.0	448.72
4.5	461.27
5.0	437.54
5.5	421.35
6.0	412.98
6.5	385.19
7.0	366.89
7.5	347.11
8.0	322.48
8.5	298.22
9.0	277.55
9.5	256.00
10.0	232.68
10.5	213.15
11.0	192.12
11.5	171.75
12.0	150.35
12.5	131.52
13.0	110.00
13.5	90.74
14.0	75.35
14.5	55.25

Table A3.2 Extrapolated values of activities normalized with the measured activities

Distance r(cm)	R2.A(r) (arbitray unit)	r4.A(r) (arbitra unit)	Distance	r2.A(r) (arbitray unit)	r4.A(r) (arbitray unit)
2.0	1.08	0.04	14.5	13.29	27.95
2.5	2.17	0.14	15.0	13.13	29.54
3.0	3.61	0.33	15.5	11.97	28.76
3.5	5.35	0.66	16.0	10.92	27.94
4.0	7.18	1.49	16.5	9.95	27.10
4.5	9.45	1.91	17.0	9.08	26.23
5.0	11.19	2.80	17.5	8.28	25.35
5.5	13.03	3.94	18.0	7.55	24.45
6.0	15.17	5.46	18.5	6.88	23.56
6.5	16.61	7.02	19.0	6.28	22.66
7.0	18.61	9.12	19.5	5.72	21.76
7.5	20.20	11.37	20.0	5.22	20.87
8.0	21.36	13.67	21.0	4.34	19.14
8.5	22.07	15.95	22.0	3.61	17.46
9.0	23.04	18.66	23.0	3.00	15.87
9.5	23.69	21.38	24.0	2.49	14.37
10.0	23.89	23.89	25.0	2.07	12.97
10.5	24.15	26.96	26.0	1.72	11.66
11.0	23.43	28.96	27.0	1.43	10.46
11.5	23.43	30.98	28.0	1.19	9.35
12.0	22.56	32.49	29.0	0.99	8.34
12.5	21.02	32.85	30.0	0.82	7.42
13.0	19.14	32.34			
13.5	17.28	31.51			
14.0	15.65	30.68			



FIG·A3·1:GRAPH OF SPECIFIC ACTIVITY A(r) AGAINST DISTANCE r·



FIG·A3·2 :GRAPH OF ln A(r)·r^2 VERSUS r.

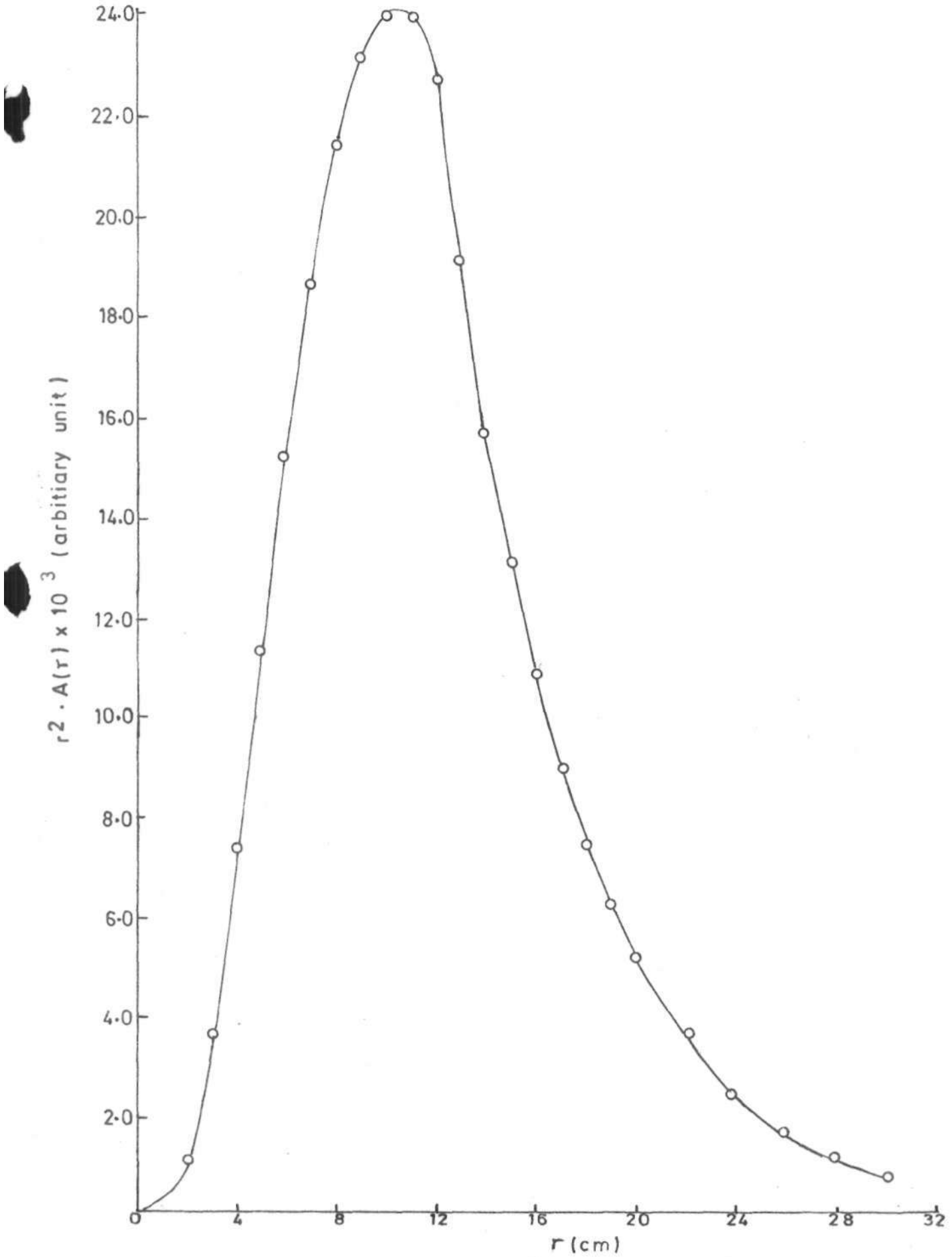


FIG. A3·3 : GRAPH OF $A \cdot r^2$ VERSUS r

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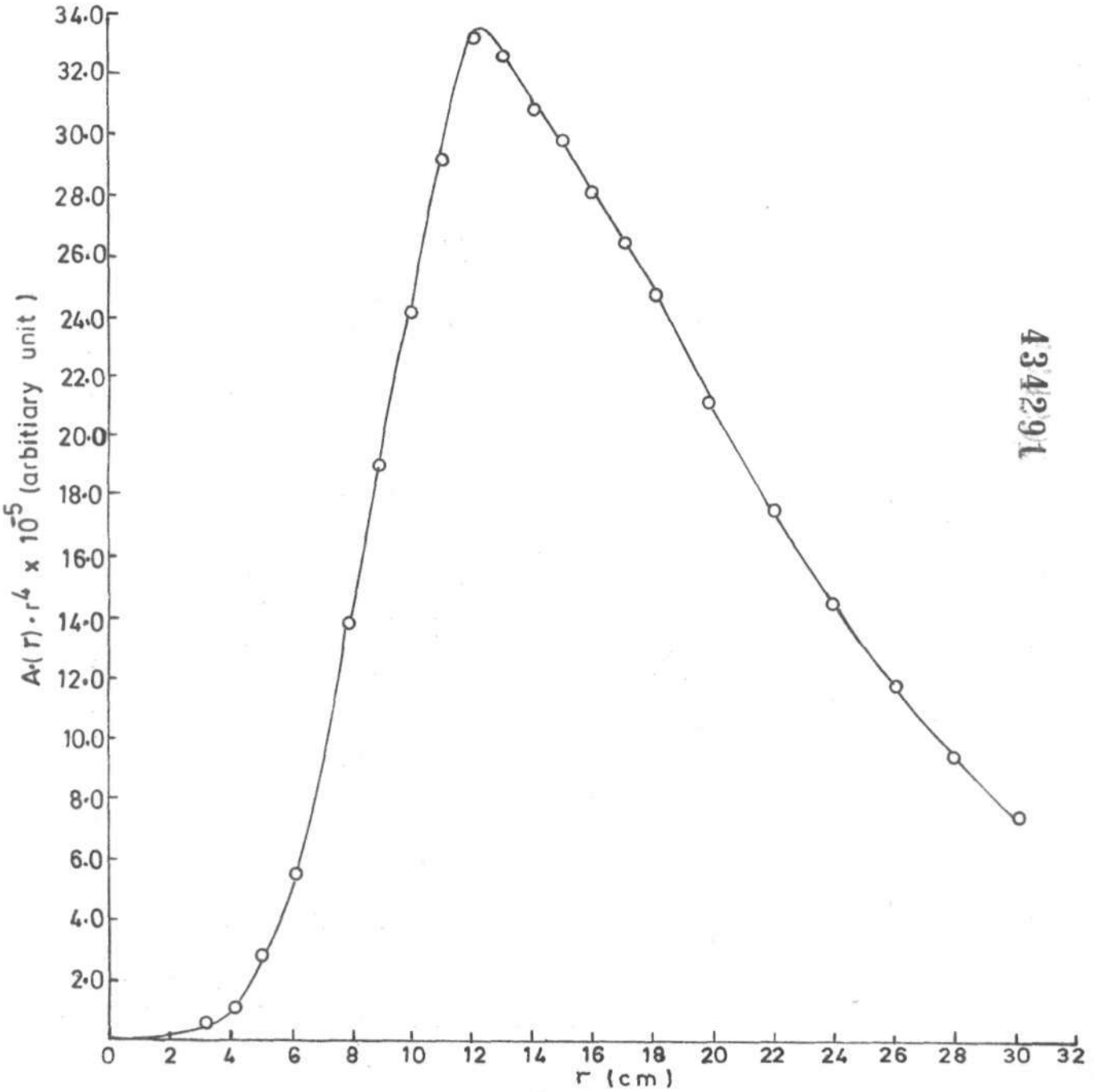


Fig. A3·4 : GRAPH $A \cdot r^4$ VERSUS r

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