ADDIS ABABA UNIVERSITY

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Dated: March, 2012

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Date: March, 2012

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Title: Measurement of Thermal Neutron Capture Cross Section in Antimony

Department: Physics

Degree: M.Sc. Convocation: March Year: 2012

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Abstract

The measurement of thermal neutron cross sections gives basic information about the internal structure of atomic nuclei and their structure. Measurement of Thermal Neutron cross-section of Sb -121 has been performed at the Addis Ababa University Nuclear physics Laboratory using the concept of neutron activation Analysis. Irradiations without Cadinium cover were performed and the neutron transmission through the cover was calculated. High resolution gamma ray spectrometry measurements was undertaken in order to check the consistency of the results. The thermal neutron cross section measured for Sb-121 using slow neutrons is ($5.45 \pm 0.23 b$). The results are discussed and compared with previous Measurement. In general there is good agreement between the observed and calculated value of thermal neutron capture cross section.

Keywords: Neutron sources, Neutron interaction, Thermal neutron cross section, Slowing down of Neutron, Neutron Activation analysis, HPGe

Acknowledgements

Above all, I would like to thank the Almighty God, for letting me accomplish this stage. I am deeply indebted Professor A.K.Chaubey, my advisor, for his many suggestions and constant support and friendly approach during this research. His tireless follow up and his consistent support will be in my memory forever. My strongest thank is addressed to my mammy Dunge Torban and my intimate Friends, who lived for myself. They are the hero of my success without their push and support, this stage is unthinkable. I have derived materials from many research journals and books, and am indebted to the authors of those publications and books. Finally, I wish to thank Ato Yihunie Hibstie and Hailu Geremew For their best friendship throughout my study period.

INTRODUCTION

The accurate knowledge of neutron capture cross sections induced in radioactivite nuclei has become important in recent years due to research and development of nuclear transmutations associated with waste disposal management. In the previous work the cross section for $^{121}Sb(n,\gamma)^{122}Sb$ reaction has been measured for 564KeV gamma energy which is equal to $5.84 \pm$ 0.2b [1]. In the present paper the cross-section of $^{121}Sb(n,\gamma)^{122}Sb$ reaction in our libratory has been investigated.

Neutrons together with protons are the constituents of atomic nuclei. The neutron was discovered after more than two decades of speculation that electrically neutral particles exist in atoms**.** Because the neutron is electrically neutral, it easily interacts with nuclei and does not interact directly with electrons. Since the nucleus of an atom is about one ten-thousandth the size of the electron cloud, the chance of neutrons interacting with a nucleus is very small, allowing them to travel long distances through matter.[2]

As a free particle, the neutron is an important and yet unique *tool* used for various applications: in medicine to initiate powerful nuclear interactions whose products can directly destroy cancer cells (neutron capture therapy for example), for research on physical and biological materials, for imaging through easy allocation of light atoms especially hydrogen, to investigate properties of magnetic materials (neutrons possess a magnetic moment and thus act as small magnets), to track atomic movement (thermal neutron energies almost directly coincide with the energies of atoms in motion), and to maintain the fission chain reaction in nuclear reactors. Free neutrons are unstable and break up in short time by β^- decay to a proton, electron and antineutrino. However, free neutrons will most likely interact with the surrounding matter and disappear through nuclear interactions long before they decay.[2]

Among heavy elements thermal and epithermal neutrons can cause **(**n,α) and (n, p) reactions, as well as neutron capture, depending on the energies for the various particles. Among heavier elements the neutron result primarily in capture (n, γ) and fission reactions, fast neutrons being required for particle emission reaction such as (n, 2n), (n, p), etc.

The probability of a neutron interacting with nucleus for a particular reaction is dependent upon not only the kind of nucleus involved, but also the energy of neutron. Accordingly, the absorption of thermal neutrons in most material is much more probable than the absorption of a fast neutron. Also, the probability of interaction will vary depending up on the type of interaction involved.

The probability of a particular interaction occurring between a neutron and a nucleus is called the microscopic cross section (σ) of the nucleus for the particular interaction. This cross section will vary with the energy of the neutron. The microscopic cross section may also be regarded as the effective area of the nucleus presented to the projectile. The larger the effective area, the larger the probability of interaction. Because the microscopic cross section has definition of an area, it is expressed in unit of area, or square centimeters. A square centimeter is large compared to the effective area of a nucleus; hence it is expressed in a smaller unit of area called a barn. One barn is 10^{-24} cm^2.[3]

Antimony is in period 5 of the nitrogen group (Group VA), and is variously classed as a nonmetal or a metalloid. It may have oxidation numbers of -3, +3, and +5 in its compounds. This has also two isotopes available in natural isotopes. Out of the most abundant is ^{121}Sb(53.7 %) and the other is ^{123}Sb(42.7%).The major use of antimony is in lead alloys - mainly for use in batteries - adding hardness and smoothness of finish. The higher the proportion of antimony in the alloy, the harder and more brittle it will be. Alloys made with antimony expand on cooling, retaining the finer details of molds. Antimony alloys are therefore used in making typefaces for clear, sharp printing. Babbitt metals, used for machinery bearings, are alloys of lead, tin, copper and antimony. These metals are hard but slippery and so ideal for use as bearings. Antimony is used in the semiconductor industry as an n-type dopant for silicon. Antimony trioxide is used as a flame retardant in adhesives, plastics, rubber and textiles.

Irradiating sample of antimony by a uniform neutron beam of known flux and measuring the induced radioactivity by counting gamma radiations using precalibrated detectors, HPGe thermal neutron capture cross section of the Sb-121 can be measured. To perform such an experiment one needs, standard sources for the calibration of the detector, thermal neutron

source to have beam of thermal neutrons for the activation of the sample of the isotope and the respective detector for the measuring of emitted radiations. In performing this experiment Am-Be neutron source of 2ci, High Purity Germanium detector and standard calibration sources are used in nuclear laboratory of AAU. When a natural Antimony foil is irradiated with neutron beam, thermal neutrons interact with antimony to produce compound nucleus of Sb-122 of easily measurable life time. While decay of Sb-122 ground state proceeds directly in to Te-122 ground state, the isomeric states of Sb-122 decay in to the excited states of Te-122. Several gamma rays are emitted in their de-excitation. Hence, by measuring the spectrum of the gamma ray emitted, the thermal neutrons capture cross section for Sb-121 can be found.

In the following Chapters, Discovery of neutron, Radioactive decay of Neutron, properties of neutron, classification of neutron(chapter 1), Neutron physics, neutron sources, neutron interaction, neutron slowing down, Neutron activation analysis(chapter 2), experiment(chapter 3), data and data analysis(chapter 4) and theoretical determination of thermal neutron capture cross section (chapter 5) will be dealt.

General objective of study

The objective of this thesis is:

- To study the capture reaction (n,γ) using the medium element isotope (^{121}Sb) and measure its thermal neutron capture cross section using high purity germanium detector(HPGe)
- To calculate the half-life of given sample.
- To compare the theoretical value of thermal neutron capture cross section with the measured value.

CHAPTER ONE

1. NEUTRON

1.1 Discovery of Neutron

From the time of Rutherford it has been known that the atomic mass number ***A*** of nuclei is more than twice the atomic number *Z* for most atoms and that almost all of the mass of the atom is concentrated in its center, *i.e.* at the nucleus. However, it was presumed that the only fundamental particles were protons and electrons. Rutherford had speculated that the nucleus was composed of protons and proton-electron pairs tightly bound together and the fact that an atom was neutral in charge required that somehow a number of electrons were bound in the nucleus to partially cancel the charge of the protons. Quantum mechanics, however, indicated that there was not enough energy available to contain electrons in the nucleus [2]

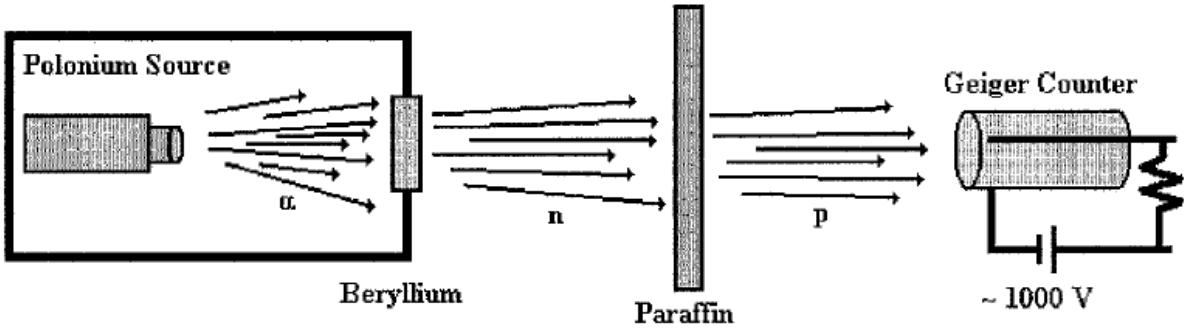


Figure 1.1 Experimental setup that led to the discovery of a neutron (1932)[2]

An experimental breakthrough came in 1930 when Bothe and Becker bombarded a beryllium target with α particles emitted from a radioactive source. The experiment produced neutral radiation that was observed to be highly penetrating but non-ionizing. In the following years Curie and Joliot showed that when paraffin (a material rich in protons) is bombarded with this neutral radiation it ejects protons with energy of about 5.3 MeV (see Fig 1.1). Bothe and Joliot - Curie each explained that the radiation was high energy gamma rays. This, however, proved to be inconsistent with what was known about gamma ray interactions with matter.[2]

In 1932 Chadwick performed a number of experiments using different target materials to discover that the emitted radiation was actually a stream of new particles that he named neutrons. The discovery proved that there is a neutral particle in the nucleus, but also that there are no free electrons in the nucleus as Rutherford had speculated. Amazingly, once free from the nucleus, neutrons are unstable and decay with a half life of about 15 minutes into a proton, an electron, and an antineutrino.[3]

The α-Be reaction in the experiment shown in Fig. 1.1 was explained by Chadwick:

$$He_2^4 + Be_4^9 \rightarrow n_0^1 + C_6^{12} \qquad 1.1$$

where He_2^4 represents the α particle. He argued that if a photon interacts with a proton in the paraffin target and transfers 100 MeV/c of recoil momentum, the photon itself must have had a momentum of nearly 50MeV/c, which corresponds to an energy of 50 MeV. As the energy of the α particles striking the beryllium target was only about 5 MeV, it was impossible that 50 MeV gammas were being emitted. Instead, Chadwick suggested a new particle with approximately the same mass as a proton, which solved the contradiction related to the energy of the assumed photons. In the collision of two particles of equal masses, the incident particle (neutron) can transfer all of its kinetic energy to the target particle (proton).

1.2 Radioactive Decay of Neutron

The first determination of the mass of the neutron by Chadwick and Goldhaber shows that, its mass was greater than that of proton. Depending on this fact they suggest that free neutrons should be unstable and they should decay into the proton, electron and antineutrino.

$$n \rightarrow p + e + \bar{\nu} \qquad 1.2$$

From the radioactive decay of neutron there is no physical law violated .In 1948, A.H Snell and his workers were able to detect protons and electrons coming out from a thermal neutron source with an estimated half life about 10 to 30 minute. J.M.Robinson was able to determine accurate half-life of neutron and its beta ray (electron) spectrum using intense thermal neutron source. [4] The end point energy spectrum of the experiment was 784kev[4],which was in excellent agreement with the mass difference of 784kev between a neutron and hydrogen atom. The value of half life of neutron was determined from the value of the density of neutron beam and the number of neutrons decaying per unit time per volume. The accepted value of the mean lifetime

of free neutron is 10.26 ± 0.04 minute; while neutrons bound in a nucleus apparently are stable. [5].

1.3 Classification of neutron

Depending on their energy neutrons can be classified into the following parts:

Neutron energy	Name
- 0.025 eV	Cold
0.025 eV	Thermal
0.025 eV - 0.4 eV	Epithermal
0.4 eV - 0.6 eV	Cadmium
0.6 eV - 1 eV	Epicadmium
leV-10eV	Slow
10 eV - 300 eV	Resonance
300 eV - 1 MeV	Intermediate
MeV - 20 MeV	Fast
> 20 MeV	Relativistic

Table 1.1 Classification of neutron energy [2]

1.4 Properties of Neutron

Neutrons, as mentioned previously, are non charged subatomic particles having a mass (*m*) of 1.0087 atomic mass units(1.675×10^{-27} kg), spin of 1⁄2, and a magnetic moment (μn) of -1.9132 nuclear magnetos . These properties of the neutron give rise to two principal modes of interaction which are different from those of X-rays as neutrons are zero charge particles, their interaction with matter, both nuclear and magnetic, is short ranged. As a result of this small interaction probability, neutrons can penetrate deep into condensed matter. Moreover, the interaction between the neutron and atomic nuclei involve complex nuclear interactions between

the nuclear spins and magnetic moments. For this reason, there is no general trend throughout the periodic table of an atom's ability to scatter neutrons. In addition, different isotopes of the same element may have very different abilities to scatter neutrons. This concept of a difference in scattering power, or *contrast*, between various components in a sample as a result of the different scattering properties of the various elements (particularly 1H and 2H) is the core principle of neutron scattering, and from which biology greatly benefits. The second mode of interaction is the magnetic dipole interaction between the magnetic moments associated with unpaired electron spins in magnetic samples and the nuclear magnetic moment of the neutron.[6]

Neutrons are composed of three quarks. The color assignment of individual quarks is not important, only that all three colors are present. The compositions of quarks are: 1 up and 2 down.

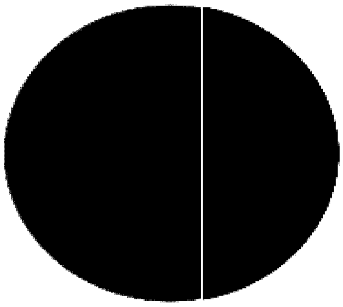


Figure 1.2 Three quarks of neutron.

Quarks carry fractional electric charges (+2/3, -1/3 and -1/3). Antiquarks carry opposite electric charges. There are different types of quarks distinguished by their *flavor*, i.e., *u, d, s, c, b* and *t* quarks. Their masses range from ~ 5 MeV/c^2 for the lightest quark *(u)* to ≈ 180 GeV/c^2 for the heaviest quark *(t).*[7]

CHAPTER 2

2. NEUTRON PHYSICS

Neutron physics is the study of the travel of neutrons through matter and the resulting reactions, most notably power generation via the fission of nuclei of heavy atoms. In general terms, it describes the interaction of elementary particles — neutrons — with the nuclei of atoms that form matter.[3]

The neutron is a subatomic hadron particle which has the symbol n or n^0 no net electric charge and a mass slightly larger than that of a proton. With the exception of hydrogen, nuclei of atoms consist of protons and neutrons, which are therefore collectively referred to as nucleons. The number of protons in a nucleus is the atomic number and defines the type of element the atom forms. The number of neutrons is the neutron number and determines the isotope of an element.

2.1. Neutron Sources

Neutron source is a general term referring to the Variety of device that emit neutrons. These devices are diverse in size, complexity and applications in areas of physics, engineering, medicine and research. Depending up on the mechanisms used to produce neutrons; neutron sources are divided in to two broad categories:

1. Radioactive sources
2. Accelerator based sources

2.1.1 Radioactive source

1. Spontaneous fission

Many of the transuranic heavy nuclides have an appreciable spontaneous fission decay probability to produce neutrons. Gamma rays and beta are also the other products of such fission process. The most common spontaneous fission source is Californium-252. Its half-life of 2.65 years is long enough to be reasonably convenient, and the isotope is one of the most widely produced of all the transuranics. For every 32 alpha emissions, there is a spontaneous fission

with an average 3.8 neutrons and half-life of 85.5 year. The neutron yield is $2.30\times10^{6} n/s$ microgram. Hence, a small-encapsulated source provide significant yield. The peak of relative number of neutrons lies between 0.5MeV and 1MeV with the higher energy neutrons being 8MeV to 10MeV. [8]

2. Radioisotope (α, n) sources

Because energetic alpha particles are available from the direct decay of a number of convenient radionuclides, it is possible fabricate a small self-contained neutron source by mixing an alpha-emitting isotope with a suitable target material. These neutron sources uses (α , n) reaction in order to produce neutron, those light nuclei like; Li, Be, B, N, O, F, are used as a targets and radio nuclide like; Pu, Ra, Am, Cm, Th, and U are used for sources .[9]

- **(α, n) Targets**

A number of light nuclei are good to undergoing (α, n) reaction relatively with low energy α − particle. This reaction has higher probability in light nuclei than heavy nuclei, because the threshold energy or coulomb barrier is smaller in light nuclei.

Some of the reactions are:

The Be(α, n)Reaction

The B(α, n)Reaction

The F(α, n)Reaction

The Li(α, n)Reaction

The nuclei are then taken to as targets in table(2.1) along with the reaction energy (Q-value), threshold energy and neutron energy for 5.5Mev.[9]

Target	Q (Mev)	Threshold energy (Mev)	Mean En for E_α =5.5Mev	Yield ($n/10^6\alpha$)
^{6}Li	- 3.975	6.620	NA	NA
^{7}Li ^{10}Be	-2.79 5.702	4.382 exothermic	0.5883 5.005	3.156 80.073
^{11}B	1.06	"	2.243	5.72
^{15}B	0.157	"	2.993	23.724
^{13}C	2.215	"	4.72	9.904
^{17}O	0.587	"	2.523	0.152
^{18}O	-0.697	0.852	2.374	0.333
^{19}F	-1.95	2.361	1.304	0.106

Table 2.1: Traits of common(α, n)source[9],[10]

As shown from table (2.1), the greatest yield achieved by a beryllium target for (α, n) reaction. Therefore, it has usually been used as a target material for a neutron source. The reaction is:

$$Be_4^9(\alpha,n)C_6^{12} \qquad 2.1$$

Even though, those nuclei in the table undergo (α, n) reaction, only those isotopes which can undergo exothermic or low energy threshold are considered to be useful neutron source. [9]

- **(α, n) Radionuclide**

Alpha emitting radionuclide are also very important components of (α, n) reaction neutron sources. Uranium and transuranic elements are usually alpha emitters since the probability of (α, n)reaction increases with alpha -particle energy and the number of alpha-particle depends on half-life; those alpha -emitters having high energy and long half life on the order of 1 up to 2500 years are preferable. Radioactive neutron sources mentioned in the fig (2.2) can be taken as smaller device neutron sources .^{226}Ra-Be neutron source has long half-life and high neutron yield, therefore it can be considered as a good neutron source.

Source	$T_{½}$	Most common E_α (Mev)	Yield (n /10^6 α)
^{239}Pu-Be	24110yr	5.156	57.2
^{210}Po-Be	138.38d	5.304	63.7
^{241}Am-Be	432.2yr	5.486	71.5
^{238}Pu-Be	87.7yr	5.499	72.1
^{238}Cm-Be	18.1yr	5.805	105.3
^{244}Cm-Be	162.8d	6.113	88.1
^{226}Ra-Be	1600yr	4.784,5.490,6.002 7.687, 5.304	736.8
^{227}Ac-Be	21.8yr	6.038,5.726, 6.819 7.386, 6.623	707.2
^{228}Ra-Be	5.75yr	5.4223,5.686,6.288 6.779,6.051(36%) 8.784	707
^{228}Th-Be	1.911yr	64% ^{228}Ra	707.2
^{232}U-Be	68.9yr	5.320 +^{228}Ra	755.5

Table 2.2 Characteristics of Be(α,n) Neutron sources[10]

3. Nuclear Reactors Sources

Nuclear fission which takes place within the reactors produce very large quantities of neutrons and can be used for a Variety of purposes including power generation and experiments. A nuclear reactor is a source of products of fission process, such as, energy, neutron and some useful radioactive isotopes. The fission reaction which takes place in the reactors using slow or thermal neutrons having energy 0.0253eV and velocity about 2200m/s [5].These neutrons react with natural uranium producing several prompt neutrons. The average number of neutrons produced per fission of uranium-235 by a thermal neutron is: ν=2.47 ±0.03 neutron/fission [4].

The energy of neutrons from thermal fission of ^{235}U is different value extending from about 0.04MeV to about 17MeV with a distribution maximum average value about 0.75MeV. In order to use these neutrons having different energy value for fission process in the reactor and for experiments they have to be slow down in the reactor by using a substance moderator. The energy released and the number of neutrons per fission of uranium-235 is given by;

$$n_0^1 + U_{92}^{235} \rightarrow \left(U_{92}^{236}\right)^* \rightarrow Ba_{56}^{141} + Kr_{36}^{92} + 3n + Q \qquad 2.2$$

Where Q represent the energy released in the reaction which is about 175MeV.Such types of nuclear reactors may produce a large number of thermal neutron flux at about 5.5 × 10^6n/m^2.s and ,that of fast neutron flux at about 7.1 × 10^6n/m^2.s [5]

4. Photoneutron sources

Some radioisotope gamma-ray emitters can also be used to produce neutrons when combined with an appropriate target material. The resulting photoneutron sources are based on supplying sufficient excitation energy to a target nucleus by absorption of gamma-ray photon to allow the emission of a free neutron. Only two target nuclei, ^{9}Be and ^{2}H, are of any practical significance for radioisotope photo neutron sources. The corresponding reactions can be written:

$$Be_4^9 + h\nu \rightarrow Be_4^8 + n_0^1, Q = -1.666MeV$$
$$H_1^2 + h\nu \rightarrow H_1^1 + n_0^1, Q = -2.226MeV \qquad 2.3$$

A gamma-ray photon with energy of at least the negative of the Q-value is required to make the reactions energetically possible, so that the only relatively high-energy gamma-rays can be applied. For gamma-ray energies that exceed this minimum, the corresponding neutron energy can be calculated from

$$E_n(\theta) \cong \frac{M(E_\gamma + Q)}{m+M} + \frac{E_\gamma\left[(2mM)(m+M)(E_\gamma+Q)\right]^{1/2}}{\left((m+M)^2\right)}\cos\theta \qquad 2.4$$

Where θ = angle between gamma photon and neutron direction

E_γ = gamma energy(assumed<<931MeV)

M = mass of the recoil nucleus $*c^2$

m = mass of neutron $*c^2$

The advantage of photoneutron sources is that if the gamma rays are monoenergetic, the neutrons are also nearly monoenergetic.[10]

Y Source	$T_{1/2}$	E_Y(Mev)	Target	E_n	Yield(n/10^{10}Bq)
^{24}Na	15hr	2.7541 2.7541	Be D	967 263	340000 330000
^{28}Al	2.24m	1.7787	Be	101	32600
^{38}Cl	37.3m	2.1676	Be	446	43100
^{56}Mn	2.58hr	1.8107 2.1131 2.9598 2.9598	Be Be Be D	129 98 1149 365	91500 91500 91500 162
^{72}Ga	14.1m	1.8611 2.2016 2.5077 2.5077	Be Be Be D	174 476 748 140	64900 64900 64900 25100
^{76}As	26.3hr	1.7877 2.0963	Be Be	109 383	3050 3050
^{88}Y	107d	1.8361 2.734 2.734	Be Be D	152 049 253	229000 22900 160
^{116m}In	54.1m	2.1121	Be	397	15600
^{124}Sb	60.2d	1.691	Be	23	210000
^{140}La	40.3hr	2.5217 2.5217	Be D	760 147	10200 6600
^{44}Pr	17.3m	2.1856	Be	462	690

Table 2.3: Photoneutron source properties[10]

In table (2.3), some of the photo neutron sources with their properties listed .Target materials which have large threshold energy needs to have gamma-radiation of several MeV. But these radiations are dangerous which requires larger radiation shielding.

2.1.2. Accelerator Source

Accelerator neutron sources are neutron sources /devices/ which used proton, tritium or deuteron as a projectile to hit a target material in order to emit neutrons. These systems vary in size and diversity, and they include large installations such as the Spallation neutron sources. Those particle accelerators have three basic parts in general. It consists of a source to generate positively charged ions, one or more structures to accelerate the ions in KeV up to MeV and a target material. Usually these accelerators used proton and deuteron as a projectile. The energy and intensity of projectiles can be controlled by the system in order to get the desired amount and energy of neutron. Those materials made from light atoms, such as, deuterium, tritium, lithium, and sodium, are taken as a target material. These devices are relatively effective in producing, approximately, mono energetic neutrons with a variety of intensity. Due to this reason, they have many industrial application.[11]

- **Neutron Producing Reaction**

No neutron source is perfectly mono energetic but certain reactions can be used to produce a neutron energy spectrum with a reasonably small energy spread. Because, the energy of neutrons depends on certain factors, such as, energy spread of accelerated particles, the reaction cross-section, exited level of the residual nucleus, etc.

Reaction	Q Value	Best Neutron Energy
$^{7}Li(p,n)^{7}Be$	-1.64Mev	0.1--------0.4Mev
$^{3}H(p,n)^{3}He$	-0.76Mev	0.1--------3Mev
$^{2}H(^{2}H,n)^{3}He$	3.27Mev	4----------10Mev
$^{3}H(^{2}H,n)^{4}He$	17.59Mev	13---------16Mev

Table 2.4: Neutron producing reactions with its best neutron energy. [12]

Unfortunately, there is no single reaction suitable for producing neutrons across the entire energy range. Table (2.4) list several reactions that could be used to make neutrons and energy range in which it works the best. Notice that all these reactions involve either beams of protons or deuterons with small energy range [11].Apart from their usage in terms of the energy of neutrons produced, these particle accelerators are very important for experiments and industrial applications due to their size, diversity, durability and their consistency in number of neutrons produced per second. Depending up on the energy of particles accelerated, target nucleus and the number of neutrons produced; particle accelerators can be divided in to two broad categories.

1. **Light -Ion Accelerators**

Compact light-ion accelerators with hydrogen (H), deuterium (D), or tritium (T)ion sources may be used to produce using targets of deuterium, tritium, lithium, beryllium and other low-z metals. Typically these accelerators operate with a voltage in the range of 1 MeV and above. The number of neutrons produced is smaller as compared to the larger ones.[11]

2. **High Energy Particle Accelerator**

A spallation source is one of the most high-flux neutron source .In the neutron spallation source, a high energy proton beam is used in order to bombard the heavy metal(like mercury) target to yield neutrons and sometimes used to generate heat. Spallation sources may generate an average neutron flux at about 5×10^{10}n/(cm^2.s) and above. [12]

2.2 NUETRON INTERACTION

Reactions, which are initiated by neutrons, are of primary concern in the study of nuclear reactors. When charged particles approach the nucleus, they must overcome the repulsive electrostatic force before getting close enough for the strong nuclear force to act. This requires that the incident particle possess a significant amount of energy in order for the reaction to occur. Neutrons have no charge and therefore are not subject to overcoming electrostatic repulsion in order to penetrate the nucleus. This means that neutrons need not possess large amounts of kinetic energy to initiate nuclear reactions. In fact, low energy neutrons initiate the reactions, which are of prime importance to power production (fission reactions).Neutron interactions can be described in three steps: the condition before the interaction when the neutron is approaching

the nucleus, an intermediate stage when the incident neutron forms a compound nucleus with the target, and the condition after the interaction.

One of the important and characteristic features of neutron interactions with matter that proceed through a compound nucleus formation is that cross sections exhibit maximum values at certain incident neutron energies. These maximum values are called the *resonances*. Nuclei have various excited states that correspond to different configurations of the nucleons within the nucleus. An incident neutron and a target nucleus are more likely to combine and form a compound nucleus if the energy of the incident neutron is such that the compound nucleus is produced in one of its excited states. These resonances appear in the cross section because it is necessary to form the compound nucleus before the interaction can proceed. The excitation energy of the compound nucleus is equal to the kinetic energy of the incident neutron plus the separation (binding) energy of the neutron in the compound nucleus. Neutrons interact by different mechanisms depending on the neutron energy and the material of the absorber [2]

2.2.1 Elastic Scattering (n, n)

There are two possible ways for a neutron to scatter elastically from a nucleus:

***Resonance or* Compound elastic scattering:** the neutron is absorbed by the target nucleus to form a compound nucleus followed by re-emission of a neutron, and

Potential elastic scattering: the neutron is scattered away from the nucleus by the short range nuclear force is schematically depicted in Fig. **2.1.** Potential scattering is the most common form of neutron elastic scattering. The more unusual of the two interactions is resonance elastic scattering which is highly dependent upon initial neutron kinetic energy.

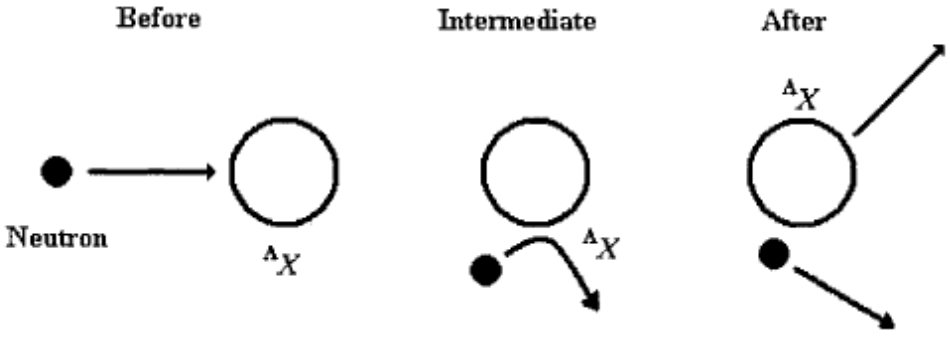


Figure 2.1 Potential Elastic Scattering

Potential scattering in which the neutron never actually touches the nucleus and a compound nucleus is not formed takes place with incident neutrons of energies up to about 1 MeV. Neutrons are scattered by the short range nuclear forces as they approach the nucleus. The cross section is expressed by the relation

σ_{el}(potential scattering) = $4\pi R^2$ (where *R* is the nuclear radius) [2]

An elastic scattering reaction Momentum and kinetic energy are, however, conserved and there is usually some transfer of kinetic energy from the neutron to the target nucleus. The target nucleus thus gains the amount of kinetic energy that the neutron loses and moves away at an increased speed. If the neutron collides with a massive nucleus it rebounds with almost the same speed and loses a negligible amount of energy. However, light nuclei will gain a significant amount of energy from such a collision and will therefore be more effective in slowing down neutrons. The largest energy transfer occurs for a head-on collision in which the neutron does not change its initial direction. Neutrons lose most of their incident energy when they interact elastically with light elements such as hydrogen. This is because the hydrogen nucleus has a mass (of one proton) nearly equal to that of the neutron. Materials with a large content of hydrogen, such as water or paraffin, are therefore very important in the slowing down of neutrons. For example, in the case of hydrogen, the energy of a head-on scattered neutron will be zero, which means that the neutron transferred all of its energy to the hydrogen nucleus.[2]

2.2.2 Inelastic Scattering

In order for a neutron to undergo inelastic scattering with a nucleus its incident energy must be sufficient to place the target nucleus in an excited state. As a result, the inelastic cross section exhibits threshold energy (and is zero up to that energy). In general, the energy levels of the excited states of a nucleus decrease with increasing mass number. Elements of high and moderate mass number usually have minimum excitation energy in the range of 0.1 MeV to 1 MeV. Elements of lower mass number have increased nuclear excitation energies. This is why neutron inelastic scattering is more probable for heavier nuclei and thus the inelastic cross section is non-zero over a large energy region for heavier nuclei.

Inelastic scattering proceeds in two steps . The interaction involves formation of a compound nucleus as an intermediate stage of the interaction process. The compound nucleus is formed in an excited state due to the energy imparted to it by the incident neutron. In the next step, a neutron of lower kinetic energy is expelled from the nucleus leaving the nucleus in a lower exited state. The nucleus then regains stability, usually by emitting the excess energy in the form γ rays.[2] The energy of the emitted γ rays is equal to the excess energy of the excited state of the target nucleus.

2.2.3 **Transmutation (n, p), (n, α)**

A nucleus may absorb a neutron forming a compound nucleus, which then de-energizes by emitting a charged particle, either a proton or an alpha particle. This produces a nucleus of a different element. Such a reaction is called a transmutation. Transmutation is the transformation of one element into another by a nuclear reaction.

1 Neutron-Proton Reaction (n, p)

Oxygen-16 captures a neutron and emits a proton to form nitrogen-16:

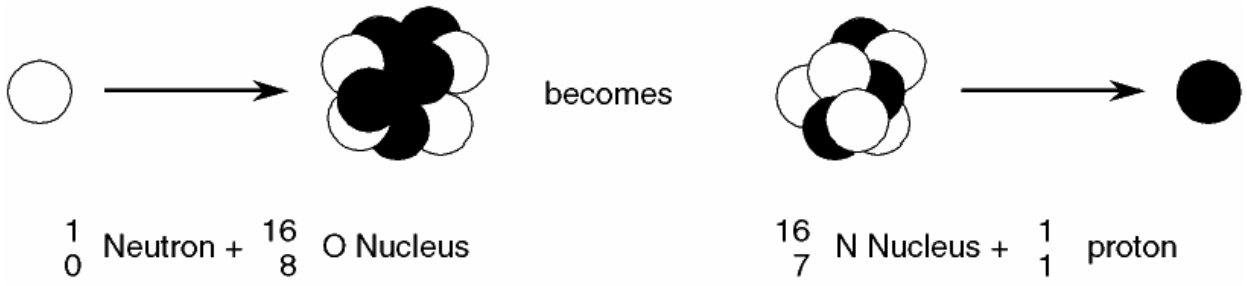


Figure 2.2 Neutron-Proton reaction

The product, nitrogen-16, is radioactive with a half-life of 7.1 seconds so this example is an activation reaction. N-16 is a beta emitter, but more important, it also emits very penetrating, high-energy gamma rays.

2 Neutron-Alpha Reaction (n, α)

Neutrons captured by boron-10 cause the following reaction:

$$n_0^1 + B_5^{10} \rightarrow Li_3^7 + \alpha \qquad 2.5$$

It can be observed that the cross section is very large at low neutron energies. For this reason, ^{10}B is used as an absorber material for unwanted low energy neutrons. As neutron energy increases, the cross section decreases following *1/v* dependence. The charged particles produced in this reaction are ejected in opposite directions with relatively high energies. They produce

considerable ionization along a short range and are capable of causing considerable damage to biological tissue. This reaction is the basic interaction upon which boron neutron capture therapy for the treatment of brain and skin cancers was developed. [2

2.2.4 Radioactive Capture (n, γ)

Neutron capture (absorption of a neutron) is often called radiative capture because γ rays are produced in the majority of these reactions. In this reaction neutrons form an isotope with mass number increased by one from the original nucleus as shown figure below. The newly formed nucleus can be radioactive and will therefore decay. The neutron capture reaction does not require any specific neutron energy and the reaction can occur at any neutron energy level. These reactions are almost always exothermic (positive Q-value) because the binding energy of the newly formed nucleus is larger than the sum of the binding energies of the neutron and the original nucleus. The simplest radiative capture occurs when hydrogen absorbs a neutron to produce deuterium (heavy Hydrogen); [2]

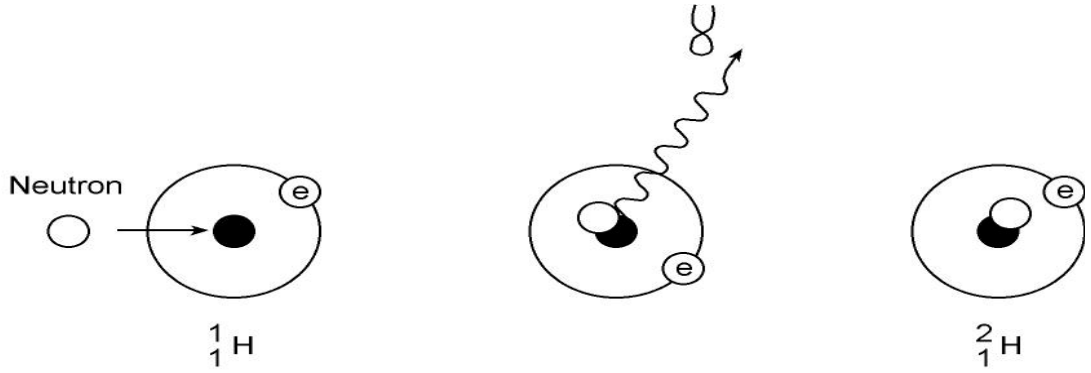


Figure 2.3 Radioactive Capture

The deuterium formed is a stable nuclide. However, many radiative capture products are radioactive and are beta-gamma emitters.

The radiative capture cross section is usually divided into three regions:

- In the low-energy region, for most nuclei, the radiative capture cross section varies as the inverse square root of incident neutron energy. Since the neutron speed is proportional to the square root of energy, the radiative cross section is said to vary as*1/v*. Since the cross sections are usually plotted on a log-log scale the *l/v* dependence appears as a straight line with a slope of -1/2. Nuclei that do not show *1/v* dependence are called non*1/v* absorbers.

- Above the *l/v* region is a resonance region in the same energy range as the resonance region for elastic scattering (because the nucleus formed in radiative capture is identical to the compound nucleus formed in elastic scattering). The radiative capture cross section in the resonance region may be expressed using the Breit-Wigner formula.
- Above the resonance region (ending around 1 keV in heavy nuclei and at higher energies in lighter nuclei) the radiative cross section drops rapidly and smoothly to very small values

2.2.5 FISSION

Fission represents a class of nuclear interactions in which the original target nucleus splits into smaller nuclei. Fission also represents the class of neutron interactions that produces neutrons and energy and as such is a basic principle of nuclear power generation. Fission can be a spontaneous process. For example, ^{240}Pu and ^{252}Cf decay by spontaneous fission; however, such nuclei are rare and the decay rate is very low. In the fission process, a neutron interacts with the target nucleus creating a compound nucleus that is unstable and splits into smaller nuclei releasing two or more neutrons and energy. The compound nucleus thus temporarily contains all of the charge and mass involved in the reaction and exists in an excited state. The excitation energy added to the compound nucleus is equal to the sum of the binding energy of the incident neutron and its kinetic energy. The smaller nuclei formed after the compound nucleus decays are called fission products or fission fragments. Not every interaction of a neutron with a nucleus leads to fission after forming the compound nucleus. Since the compound nucleus is in an excited state, it can reach stability by emitting γ rays. [2]

2.3 Maxwell-Boltzmann Distribution

In a medium in which neutrons are not absorbed and from which neutrons cannot escape, the only possible interaction is scattering with the nuclei of the atoms. The scattering interactions reduce the neutron energy. However, an endless slowing down process is not possible because of the thermal motion of the atoms. Due to that fact they cannot be assumed to be stationary, which is usual approximation in analyzing neutron interactions. When neutron energy becomes comparable to the energy of thermal motion of the atoms, the neutrons come to a thermal equilibrium. It means that the probability that a neutron will gain or lose energy in a collision

with the nuclei is equal. The average kinetic energy of thermal motion of the atoms (according to the kinetic theory of gases) is given by:

$$E = 3/2\ kT \qquad 2.6$$

where k is the Boltzmann constant (1.380662 **x 10^{-23}J**/K), and T is temperature of the medium (in Kelvin). Therefore, in a thermal equilibrium state, neutrons can gain or lose kinetic energy ($mv^2/2$), i.e. exchange their kinetic energy with the nuclei of atoms in the medium. In an ideal medium without absorption and leakage, the neutron energy distribution will be the same as that of the atoms in thermal motion. The thermal neutrons, even at a specific temperature, do not all have the same energy or velocity. Such spectrum is called a Maxwellian-Boltzmann distribution, or referred as a ***Maxwellian distribution.*** Although such conditions are not satisfied in a real reactor system, it is useful to assume that neutrons become thermalized to the extent that they follow the Maxwellian distribution

$$\frac{n(E)}{n} = \frac{2\pi}{(nkT)^{3/2}} e^{-E/kT} E^{1/2} \qquad 2.7$$

$$\frac{n(v)}{n} = \frac{4\pi v^2}{(2\pi kT/m)^{3/2}} e^{-mv^2/2KT} \qquad 2.8$$

where:

n = thermal neutron population per unit volume

m = neutron rest mass

T = temperature in K

n(E) and n(v) = Maxwellian energy (or velocity) distribution of neutrons per unit volume and unit energy (or velocity) interval. The first represents the fraction of neutrons having energies (or velocities) within a unit energy interval (or velocity interval)and the second represents the Maxwellian distribution. The most probable neutron velocity V_p is found by setting the derivative of n(v) with respect to velocity equal to zero

$$\frac{dn(v)}{dv} = \frac{8\pi v\ n}{(2\pi kT/m)^{3/2}} e^{-mv^2/2kT} - \frac{4\pi v^2}{(2\pi kT/m)^{3/2} 2kT} e^{-mv^2/2kT} = 0 \qquad 2.9$$

$$v_p = \sqrt{\frac{2kT}{m}} \qquad 2.10$$

The most probable energy can be obtained in the same way to give kT/2.The kinetic energy of thermal neutrons with most probable velocity is

$$E_p = \frac{m v_p^2}{2} = \frac{m2kT}{2m} = kT \qquad 2.11$$

The average neutron velocity is obtained from

$$v_{av} = \frac{\int_0^\infty n(v) v dv}{\int_0^\infty n(v) dv} = \frac{\int_0^\infty \frac{4\pi n v^3 e^{-mv^2/2kT}}{(2\pi kT/m)^{3/2}} dv}{\int_0^\infty \frac{4\pi v^2 e^{-mv^2/2kT}}{(2\pi kT/m)^{3/2}} dv} = \sqrt{\frac{8kT}{\pi m}} \qquad 2.12$$

The ratio of the average velocity to the most probable velocity of neutrons in the Maxwellian spectrum is

$$\frac{v_{av}}{v_p} = \frac{\sqrt{8kT/\pi m}}{\sqrt{2kT/m}} = \frac{2}{\sqrt{\pi}} = 1.128 \qquad 2.13$$

The cross section at these velocities changes accordingly and is shown as follows. The neutron flux for the Maxwellian distribution of neutrons is given by

$$\phi(E) = vn(E) = \frac{2\pi n}{(\pi kT)^{3/2}} e^{-E/kT} E \sqrt{2/m} \qquad 2.14$$

The average absorption cross section for this population of neutrons assuming 1/v dependence can be estimated. For the 1/v absorption cross section dependence, the following correlation between the cross sections holds

$$\sigma_a(E) = \sigma_a(E_p)\sqrt{\frac{E_p}{E}} \quad \text{where} E_p = kT \qquad 2.15$$

The average absorption cross section is then

$$\overline{\sigma_a}\left(E_p\right)_{avg} = \frac{\int_0^\infty \sigma_a(E)\phi(E)dE}{\int_0^\infty \phi(E)dE}$$

$$= \frac{\sigma_a\left(E_p\right)\int_0^\infty \left[\left(\sqrt{\frac{E_p}{E}}\right)\frac{2\pi n}{(\pi kT)^{3/2}}\left(\sqrt{\frac{2}{m}}\right)Ee^{-E/kT}\right]dE}{\int_0^\infty \frac{2\pi n}{(\pi kT)^{3/2}}\left(\sqrt{\frac{2}{m}}\right)Ee^{-E/kT}dE}$$

$$= \frac{\sigma_a\left(E_p\right)\int_0^\infty \left[\left(\sqrt{\frac{kT}{E}}\right)Ee^{-E/kT}\right]dE}{\int_0^\infty Ee^{-E/kT}dE}$$

$$= \frac{\sqrt{\pi}}{2}\sigma_a\left(E_p\right)$$

2.16

The values for microscopic absorption cross sections at a higher temperature are lower than the tabulated value (which is generally for the most probable neutron velocity at ambient temperature) and any cross sections which involve absorption (fission, capture) must be corrected for the existing temperature. The average absorption cross section at the average neutron velocity and temperature, *T*, higher than the ambient is given by

$$\overline{\sigma_a}\left(E_p,T\right) = \frac{\sqrt{\pi}}{2}\sigma_a\left(E_p,293k\right)\sqrt{\frac{293}{T}} \qquad 2.17$$

2.4 SLOWING DOWN OF NEUTRONS

Neutrons are slowed down in both elastic and inelastic scattering collisions with the nuclei of the atoms in a medium. In each collision, the neutron transfers a portion of its kinetic energy to the target nucleus in the form of kinetic energy if the collision is elastic or excitation energy if the collision is inelastic. Inelastic scattering is dominant with heavy nuclei, while elastic scattering is dominant with light nuclei. Moderator materials have low mass numbers and remove a large amount of energy from neutrons in a single collision and are also weak absorbers. The slowing

down of a neutron from fission energies to roughly 1 eV is called moderation and the slowing down below 1 eV is called the thermalization. [2]

2.4.1. Elastic Scattering in the Moderating Region

Elastic scattering in the moderating region is described by assuming that the colliding particles behave as elastic spheres, with the assumption that the target nuclei are stationary. In considering the scattering collision processes, two frames of references (Fig. below) are used

(a) The laboratory system (LS): scattering nucleus is at rest before the collision, and the neutron is moving toward the nucleus; after the collision, the neutron changes its direction of motion and velocity, and the nucleus moves from the rest position with some velocity. The viewpoint is that of a stationary external observer.[2]

(b) The center of mass system (COM): neutron and nucleus are stationary in the collision. The observer is located at the center of mass of neutron plus the nucleus (compound nucleus) and travels with the velocity of the compound nucleus. The center of mass is an imaginary point where the system is balanced.[2]

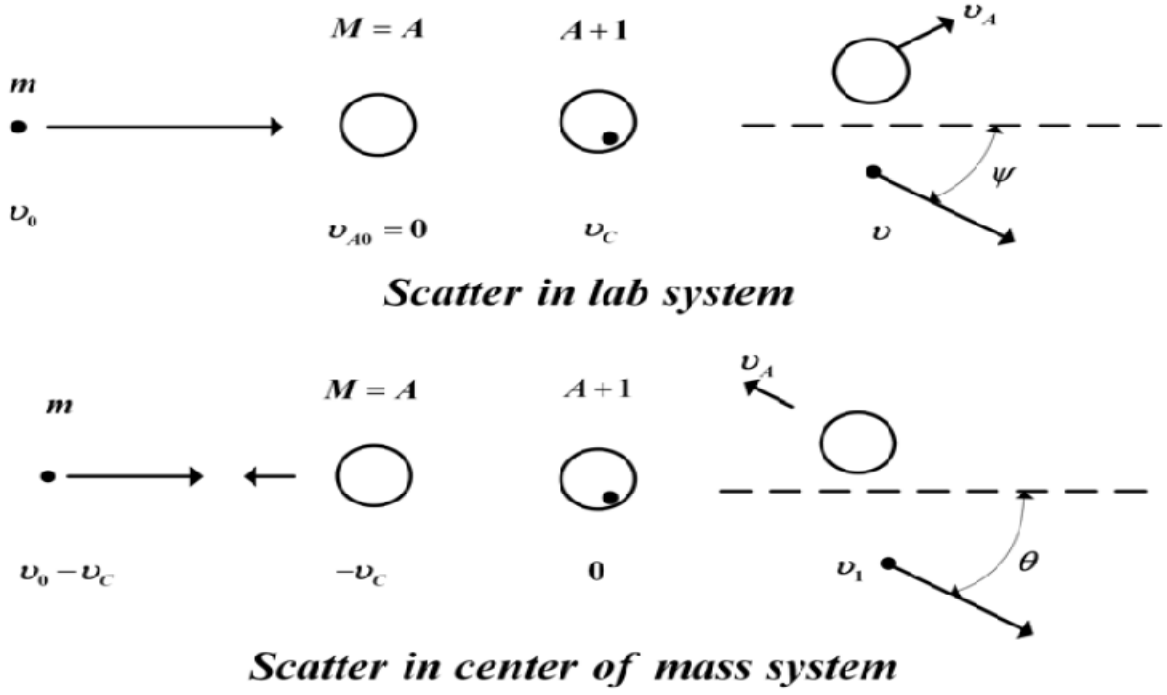


Figure 2.4 Scatter in lab and center of mass systems[2]

Where:

V_0: initial neutron velocity in *LS*

v_{A0}=0:nucleus velocity in *LS*

v_c: compound nucleus velocity in *LS*

v_A: recoil nucleus velocity in the *LS*

v : recoil nucleus velocity in the *LS*

ψ : neutron scattering angle in *LS* with respect to original neutron direction

v_{A1} : recoil nucleus velocity in the *COM* system

v_1: scatterd neutron velocity of in the *COM* system

θ : neutron scattering angle in the *COM* system

Laboratory system: Since the nucleus is stationary, its velocity is equal to zero and the momentum of a compound nucleus in *LS* is equal to the momentum of the incoming neutron

$$mv_0 + Mv_{A0} = (m+M)v_c \qquad 2.18$$

giving the velocity of the compound nucleus to be,

$$v_c = \frac{V_0}{(1+A)} \qquad 2.19$$

Center of mass system: In order to follow the splitting of the compound nucleus it is convenient to transfer to the center of mass system. In this system, the observer travels at the velocity and direction of the compound nucleus after the collision. Thus, the velocity of the neutron and nucleus before the collision must be reduced by the velocity of the compound nucleus *vc*. The velocity of the compound nucleus itself will become zero as it will appear stationary after the collision . Thus

- The velocity of incident neutron

$$V_0 - v_c = V_0 - \frac{V_0}{(1+A)} = \frac{Av_0}{(1+A)} \qquad 2.20$$

- The velocity of nucleus: -v_c

According to the conservation of energy law, the kinetic energy before the collision must equal the kinetic energy of the particles after the collision. The binding energy to form and break up the compound nucleus is the same and thus cancels out. The only energy to be conserved is,

therefore, kinetic energy. The kinetic energy before the collision and available to the compound nucleus is the sum of kinetic energy of neutron and nucleus:

$$T(COM)=\frac{1}{2}(v_0-v_c)^2+\frac{1}{2}A(-v_c)^2 \qquad 2.21$$

Eliminating the target nucleus velocity from the above equation gives

$$T(COM\ )=\frac{A{v_0}^2}{2(1+A)}=\frac{A}{1+A}T(LS)_0 \qquad 2.22$$

where T(LS)o represents kinetic energy of the incident neutron in LS. From the above equation it can be seen that the kinetic energy before the collision in the COM system for light nuclei is half of the incident neutron energy in LS. Thus, the difference between these two systems is more evident for light nuclei.

According to Fig.2.4 the kinetic energy in the COM system is shared between the scattered neutron and scattered nucleus flying away in opposite directions. Thus, the conservation energy law in *COM* gives

$$\frac{A{v_0}^2}{2(1+A)}=\frac{A{v_{A1}}^2}{2}+\frac{{v_1}^2}{2} \qquad 2.23$$

The conservation of momentum equation gives

$$v_1=Av_{A1} \qquad 2.24$$

By combining the last two equations it follows

$$\frac{A{v_0}^2}{2(1+A)}=\frac{A{v_{A1}}^2}{2}+\frac{A^2{v_1}^2}{2}$$

$$v_{A1}=\frac{v_0}{1+A} \qquad 2.25$$

$$v_1=\frac{Av_0}{1+A} \qquad 2.26$$

Laboratory system (LS): It is useful to now convert back to the *LS* in order to compare the kinetic energy of the scattered neutron with the kinetic energy of the incoming neutron. Conversion

from the *COM* system to the *LS* system is depicted in Fig 2.4 above and shows the transfer of velocities from one system to another using the Pythagorean Theorem

$$v^2 = (v_1 \sin\theta)^2 + (v_1 \cos\theta + v_c)^2$$

or

$$v^2 = \left(\frac{A v_0 \sin\theta}{1+A}\right)^2 + \left(\frac{A v_0 \cos\theta}{1+A} + \frac{v_0}{1+A}\right)^2$$

Which gives

$$v^2 = \frac{A^2 + 2A\cos\theta + 1}{(1+A)^2} v_0^2 \qquad 2.27$$

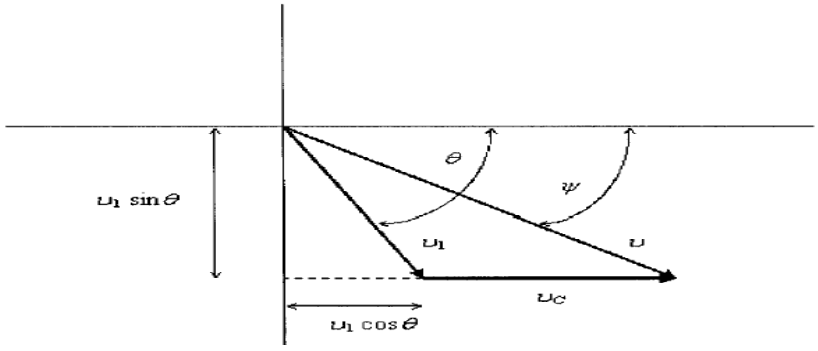


Figure 2. 7 Diagram of velocities for conversion from COM to Lab system

From this equation it is possible to obtain the ratio of kinetic energy of the neutron after collision to that before the collision

$$\frac{T(LS)}{T(LS)_0} = \frac{v^2/2}{v_0^2/2} = \frac{A^2 + 2A\cos\theta + 1}{(1+A)^2} \qquad 2.28$$

This equation leads to the following conclusions:

- This ratio reaches its maximum when $\theta = 0$, or a glancing collision.

Therefore, in forward scattering, neutron energy is not changed

$$\left(\frac{T(LS)}{T(LS)_0}\right)_{\max} = \frac{A^2 + 2A + 1}{(1+A)^2} = 1 \qquad 2.29$$

- The minimum ratio of energies is obtained for a head-on collision in which the neutron does not change its direction, or $\theta = \pi$

$$\left(\frac{T(LS)}{T(LS)_0}\right)_{\min} = \frac{A^2 + 2A(-1) + 1}{(1+A)^2} = \frac{(1-A)^2}{(1+A)^2} = \alpha \qquad 2.30$$

In the example of hydrogen **(A** = 1), the value of the defined parameter α becomes

$$\alpha_H = \frac{(1-A)^2}{(1+A)^2} = 0$$

indicating that in head-on collision with a hydrogen nucleus, the neutron energy after the collision will be zero. In other words, a hydrogen atom can cause a neutron to lose all of its energy in a single collision event.

2.4.2 Energy Distribution in Elastic Scattering Logarithmic Energy Decrement

The energy that a neutron loses in an elastic collision with the nuclei of a medium is a function of medium atomic number and the scattering angle. The logarithmic energy decrement is defined as the logarithm of neutron energy per collision

$$\xi = \ln E_0 - \ln E = -\ln \frac{E}{E_0} \qquad 2.31$$

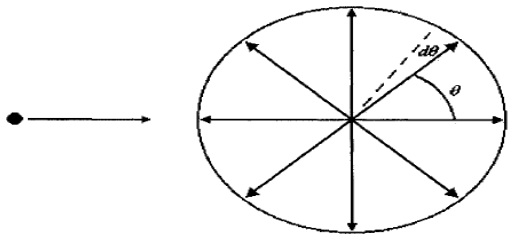
The probability that a neutron will scatter into an angle between θ and θ +dθ is the ratio of the area of the differential ring to the total area of the unit sphere

$$\frac{2\pi \sin\theta d\theta}{4\pi} = \frac{\sin\theta d\theta}{2}$$

2.32

The differential number of neutrons, dn, scattered into a differential angle is the product of the total number of neutrons, n, and the probability that neutrons will scatter into a differential angle between θ and θ+ dθ

$$dn = n\frac{\sin\theta d\theta}{2} \qquad 2.33$$



The differential number of neutrons, *dn*, multiplied by the logarithmic decrement

$$\xi dn = -\ln\frac{E}{E_0}dn = \frac{n}{2}\sin\theta d\theta\left[-\ln\frac{A^2+2A\cos\theta+1}{(A+1)^2}\right]$$

and integrated from 0 to π will give the total logarithmic decrement for all *n* neutrons of the system

$$\xi = -\ln\frac{E}{E_0} = \frac{1}{n}\int_0^{\pi}\frac{n}{2}\sin\theta d\theta\left[-\ln\frac{A^2+2A\cos\theta+1}{(A+1)^2}\right]$$

This integral can be solved introducing the following change of variables

$$x = \left[\frac{A^2+2A\cos\theta+1}{(A+1)^2}\right], dx = -\frac{2A\sin\theta d\theta}{(1+A)^2}$$

$$\xi = -\ln\frac{E}{E_0} = \int_0^{\alpha}\frac{\ln x}{2}\left[\frac{(A+1)^2}{2A}\right]dx = \frac{(A+1)^2}{4A}\int_1^{\alpha}\ln x dx$$

The constant term can be rearranged in the following form

$$\frac{(A+1)^2}{4A} = \frac{(A+1)^2}{(A+1)^2-(A-1)^2} = \frac{1}{1-\frac{(A\_1)^2}{(A+1)^2}} = \frac{1}{1-\alpha}$$

and the logarithmic decrement becomes

$$\xi = 1+\frac{\alpha}{1-\alpha}\ln\alpha \qquad 2.34$$

$$\xi = 1\frac{(A-1)^2}{2A}\ln\frac{(A-1)}{(A+1)}$$

The average logarithmic energy loss per collision is only a function of mass of the target nucleus and is not dependent on neutron energy; it is usually approximated with

$$\xi = \frac{2}{A + 2/3} \qquad 2.35$$

Since ξ represents the average logarithmic energy loss per collision, the total number of collisions necessary for a neutron to lose a given amount of energy may be determined by expanding into a difference of natural logarithms of the energy range in question

2.5 Neutron Cross Section

The quantitative description of nuclear interactions requires known neutron cross section data. A rate at which a particular neutron interaction with a given target material will occur depends on the neutron energy and speed, as well as the nature of the target nuclei. The cross section of a target material for any given reaction thus represents the probability of a particular interaction and is a property of the nucleus and incident neutron energy. [2] In order to introduce the concept of a neutron cross section, consider a parallel monoenergetic neutron beam falling on thin target of thickness x and area *A*, as shown in Fig 2.6 The intensity of the incident neutron beam is described with the number of neutrons per unit volume, *n*, and their velocity, v, as

$$I_0 = nv\left[\left(nuetrons/cm^3\right)\left(cm/s\right) = neutros/cm^2/s\right] \qquad 2.36$$

The total number of nuclei in the target of atomic density N is

Total number of nuclei in target = *Nax*

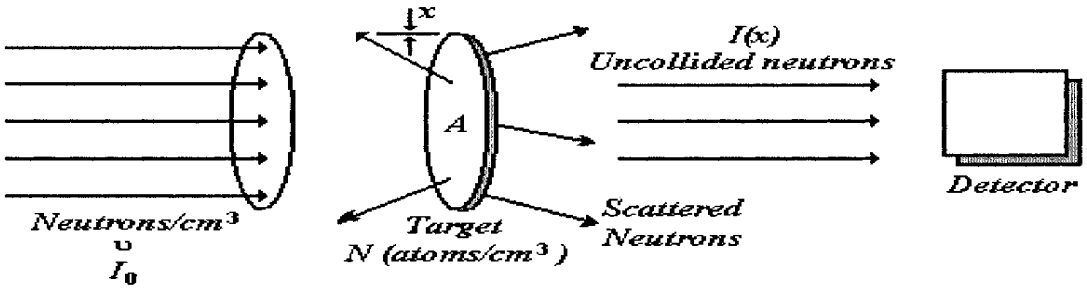


***Figure** 2.8* Concept of neutron cross section [2]

The number of neutrons that collide with the target nuclei is proportional to the neutron beam intensity and the total number of nuclei in the target. Number of neutron collisions per second in the whole target = $\sigma I_0 Nax$ 2.37

where σI_0 represents the number of neutron collisions with the single target's nuclei per unit time, and σ is referred to as the effective cross sectional area, frequently called the *microscopic cross section.* It follows σ = number of neutron collisions per unit time with one nucleus per unit intensity of the incident neutron beam.

The neutron microscopic cross section thus represents a visible area and for some interactions is closely equal to an actual area, πR^2. The accepted unit of microscopic cross sections is the barn (b), which is equal to 10^{-24}cm^2. All neutron cross sections are functions of neutron energy and the nature of the target nucleus. The probability of a neutron undergoing an interaction with all nuclei in the target as sketched in Fig. 2.6 is equal to the ratio of the reaction area to the total area

$$\sum x = \text{(reaction area) / (total area)} \qquad 2.38$$

The reaction area of the target (of volume Ax) is defined as the number of nuclei in the target material, *NAx,* multiplied by the area of each nucleus, σ

$$\sum x = \frac{N\sigma Ax}{A} = N\sigma x \qquad 2.39$$

Thus, the relation between the microscopic **(** σ **)** and macroscopic ($\sum$) cross section is

$$\sum = N\sigma \ \text{(cm}^{-1}\text{)} \qquad 2.40$$

The number of nuclei in a target material made of a single element (also called the number density), *N,* is obtained from

$$N = (N_a \times \rho)/A \qquad 2.41$$

where A is the atomic mass number and N_a, is Avogadro's number.

2.5.1 Probability of Neutron Interactions

Neutrons travel with constant direction and speed until they interact with the medium. Considering only a homogeneous medium, the probability of a neutron interacting is a function of the distance at which a neutron will interact, x. This probability can be expressed as a MacLaurin series of distance x as

$$P_{reaction}(x) = a + bx + cx^2 + dx^3 + \ldots \quad (2.42)$$

where *a, b, c, d, …* are the coefficients of expansion. Since the interaction of a neutron cannot occur at zero distance, the first term is equal to zero. For a sufficiently small distance the above equation reduces to

$$P_{reaction}(\delta x) = b\delta x \quad (2.43)$$

The probability that a neutron will not interact along the distance δx is

$$P_{non-reaction}(\delta x) = 1 - b\delta x \quad (2.44)$$

Since every interaction is independent of the previous interaction, the probability that a neutron will not interact along the distance $x + \delta x$ can be written as a product of two probabilities

$$\begin{aligned} P_{non-reaction}(x+\delta x) &= P_{non-reaction}(x) P_{non-reaction}(\delta x) \\ &= P_{non-reaction}(x)[1 - b\delta x] \end{aligned} \quad (2.45)$$

Rearranging terms it follows

$$\frac{P_{non-reaction}(x+\delta x) - P_{non-reaction}(x)}{\delta x} = -b\, P_{non-reaction}(x)$$

Taking the limit as $\delta x \to 0$ and replacing the constant *b* with $\sum$ (macroscopic cross section)

$$\frac{dP_{non-reaction}(x)}{dx} = -\sum P_{non-reaction}(x) \quad (2.46)$$

Integrating Eq.2.5.1.5 above gives the probability that a neutron does not interact and the probability that a neutron will interact along the distance *x*

$$P_{non-reaction}(x) = e^{-\sum x}$$
$$P_{reaction}(x) = 1 - e^{-\sum x} \qquad 2.47$$

The macroscopic cross section in the above equation is replaced with the linear attenuation coefficient (μ_l) in case of γ ray attenuation

2.6 **Neutron Activation Analysis**

Neutron Activation Analysis (NAA) is a quantitative and qualitative method of high efficiency for the precise determination of a number of main-components and trace elements in different types of samples. NAA, based on the nuclear reaction between neutrons and target nuclei, is a useful method for the simultaneous determination of about 25-30 major, minor and trace elements of geological, environmental, biological samples without or with chemical separation. NAA was discovered in 1936 by Hevesy and Levi, who found that samples containing certain rare earth elements became highly radioactive after exposure to a source of neutrons. [13]

In NAA, samples are activated by neutrons. During irradiation the naturally occurring stable isotopes of most elements that constitute the rock or mineral samples, biological materials are transformed into radioactive isotopes by neutron capture. Then the activated nucleus decays according to a characteristic half-life; some nuclide emit beta particles and gamma-quanta, too, with specific energies. As the irradiated samples contain radionuclide of different half-lives different isotopes can be determined at various time intervals.[14]

The advantage of the method is its sensitivity and accuracy especially in respect of some trace elements. The method is of a multielement character, i.e. it enables the simultaneous determination of many elements without chemical separation. In the case of instrumental determination, (INAA), the preparation of samples involves only the preparation of representative samples, and this reduces the danger of contamination to a minimum and accelerates the whole analytical process. If the determination of some special elements or groups

of elements can be carried out only through chemical separation, it is possible to carry out after irradiation. [13].

The development of the method has contributed to the elaboration of some very simple and accurate methods of standardization, which lead to a surpassingly accurate analysis.

2.6.1 Instrumental vs. Radiochemical NAA

With the use of good sample handling, prompt/Delayed gamma-ray measurement with solid-state detectors, (HPGe), and computerized data processing it is generally possible to simultaneously measure more than thirty elements in most sample types without chemical processing. The application of purely instrumental procedures is commonly called instrumental neutron activation analysis (INAA) and is one of NAA's most important advantages over other analytical techniques. If chemical separations are done to samples after irradiation to remove interferences or to concentrate the radioisotope of interest, the technique is called radiochemical neutronactivation analysis (RNAA). The latter can be performed infrequently.[14]

2.6.2 Prompt vs Delayed Neutron Activation Analysis

Depending up on the time of measurement, NAA can be divided in two categories.

(a) In-beam (Prompt gamma ray neutron activation analysis (PGNAA)): The measurements should be taking place during irradiation. The PGNAA technique is usually performed by using a beam of neutrons extracted through a reactor beam port.Fluxes on samples irradiated in beams are on the order of one million times lower than on samples inside a reactor, but detectors can be placed very close to the sample compensating for much of the loss in sensitivity due to flux. The PGNAA techniqueis most applicable to elements with extremely high neutron capture cross section elements which decay too rapidly to be measured by DGNAA; elements that produce only stable isotopes after the emission of prompt gamma ray; or elements with weak gamma ray intensities.[14]

(b) Off-beam (Delayed gamma ray neutron activation analysis(DGNAA)); The gamma ray measurement takes place after sample irradiation. This technique is used for the most majority of elements that produce radioactive nuclides. The DGNAA techniqueis flexible with respect to time such that the sensitivity for a Long-lived radionuclide that suffers from the shorter-lived radionuclide to decay.[14]

2.6.3 Fundamental Equations in NAA

Consider an isotope X; in the nuclear reaction, it absorbs a neutron (n).

$$X + n \rightarrow X^* + \gamma \qquad 2.48$$

Where γ is a prompt gamma ray, X^* designates excited radioactive nucleus. X^* nucleus decay via beta minus decay which converts a n (neutron) to a P (proton) there by creating a stable isotope of Y, a negative electron, an antineutrino *v* and gamma ray.

$$X^* \rightarrow Y + \beta^- + \bar{v} + \gamma \qquad 2.49$$

So in the nuclear reaction we form X^* and it decays. Hence, the number of X^* radioactive isotopes formed with time is given by an equation for;

Accumulation = production in reaction - disappearance by decay.

That is:

$$\frac{dN}{dt} = \phi\sigma N_0 - \lambda N \qquad 2.50$$

Where ϕ _ thermal neutron flux in neutrons/cm^2 sec

σ =capture cross section, i.e., probability of absorbing neutrons in cm^2

λ = decay constant

No = original number of X nuclei in the neutron source. This equation can be integrated to obtain

$$N = \phi\sigma N_0\left[1 - \exp(-\lambda t_{irr})\right] \qquad 2.51$$

where N is the number of nuclei activated at time t_{irr}.

The activity of the sample just after irradiation time, t_{irr}, can be written as,

$$A = \phi\sigma N_0\left[1 - \exp(-\lambda t_{irr})\right] \qquad 2.52$$

where $A = \lambda N$ number of decays per second at the end of irradiation, (Activity) and

$$N_0 = \frac{m \times \theta \times 6.23 \times 10^{23}}{w}$$

m = mass of the element in the irradiated sample

θ =isotopic abundance

w = Atomic weight of the element

After a delay of time t_d (the time needed to take the samples from the neutron source to the detector) ,the decay rate of the sample becomes;

$$A_0 = \phi\sigma N_0[1-\exp(-\lambda t_{irr})][\exp(-\lambda t_d)] \qquad 2.53$$

After counting time of t_c, the counts per second (n) accumulated in the given counter becomes;

$$n = \phi\sigma N_0 \varepsilon I_\gamma k[1-\exp(-\lambda t_{irr})][\exp(-\lambda t_d)][1-\exp(-\lambda t_c)$$
$$n = \varepsilon I_\gamma k A_0\left[1-\exp(-\lambda t_c)\right] \qquad 2.54$$

Where I_γ is the gamma ray abundance and ε the photo peak efficiency of a gamma ray and k is self absorption coefficient correction for gamma ray absorption in the sample. [15]. the neutron flux up on the target nuclei may be calculated from the above equation. i.e

$$\phi_1 = \frac{n(\exp\lambda t_d)}{\varepsilon I_\gamma k\sigma N_0[1-\exp(-\lambda t_{irr})][1-\exp(-\lambda t_c)} \qquad 2.55$$

where n is the activity of the sample in cps.

Where $k = \dfrac{\exp(\mu_m H)}{\mu_m H}$

where μ_m is mass absorption coefficient.

H=1/2 X_m

X_m=ratio of mass of the sample to area of the sample holder.

2.6.4 Principles of the Method

In the process of NAA the neutrons interact with the stable isotopes of the target element converting them to radioactive ones. The compound nucleus formed emits gamma rays promptly with extremely short half lives in the order of ms and these can be measured during irradiation through a technique called prompt gamma activation analysis (PGAA). In most cases, the radioactive isotopes decay and emit beta particles accompanied by gamma quanta of characteristic energies and half-life, $T_{1/2}$, and the radiation can be used both to identify and accurately determine the amount of elements of the sample. Subsequent to irradiation, the samples can be measured instrumentally by a high resolution semiconductor detector, or for better sensitivity, chemical separations can also be applied to reduce interferences.

The qualitative characteristics are: the energy of the emitted gamma quanta (E) and the half life of the nuclide (T1/2)[14].

The quantitative characteristic is: the intensity, which is the number of gamma quanta of energy E measured per unit time. Sample preparation means in most cases only pulverizing, homogenising, mass determination, packing, as well as the selection of the best analytical process and the preparation of the standards, if any. For irradiation one can choose from the various types of neutron sources according to need and availability. If there is no chemical treatment, the measurement is performed after a suitable cooling time. Measurement, evaluation and calculation involve taking the gamma spectra and the calculating trace element concentrations of the sample. The most widely used gamma spectrometers consist of germanium based semiconductor detectors connected to a computer used as a multichannel analyzer for spectra evaluation and calculation.

2.6.5 Irradiation Facilities

Neutron Sources;

Isotopic neutron sources; In the case of the most frequently used isotopic neutron sources an alpha emitting radioactive material is mixed with beryllium and (α,n) reaction generates the neutrons. The major advantage is that the isotopic neutron sources can be made portable and generate a stable neutron flux. But, as the neutron flux is rather low in comparison to a nuclear

reactor their use in NAA is limited to the determination of elements of high activation cross section which are present in major concentrations [11].

2.6.6 Irradiation Conditions

The incident flux of the irradiating particles directly affects the level of radioactivity produced, generally the neutron self absorption is negligible. In a nuclear reactor the neutron flux cannot be freely changed, but there are some possibilities to choose. The effective cross-section of a nucleus depends on the energy of the bombarding particles so; the use of a thermal neutron filter is an important option of selection. This type of analysis is called Epithermal Neutron Activation Analysis (ENAA),where Cd or B is used as a filter. Some elements can be determined with higher sensitivity by ENAA, e.g. As, Br, Rb, Sr, Mo, Sb, Ba, Ta and U[14].

Measurement of radioactivity generally, the activation product emits more than one gamma quanta, each of different energy and emission probability. To choose the proper analytical gamma line(s) the gamma abundance, the efficiency and the possible interferences must be considered, in exceptional cases self absorption can be the main problem. Sometimes, it is also possible to use more than one gamma line in order to improve the accuracy and the reliability of the measurement. The efficiency of the measurement depends on the solid angle presented by the detector to the sample. Therefore, the detection efficiency is highly affected by the shape of the sample and its distance from the detector. The counting rate can also be changed in this way. In the case of shortage of samples i.e. very low concentrations in samples the use of a well-type detector is desirable[13].

2.6.7 Absolute Method

Since the theory of NAA is well established, an ”absolute” standardization procedure can be applied. The quantitative measurement can be affected by determining the neutron flux and counting the absolute gamma/beta rays. The direct calculation of concentration is made by applying nuclear constants according to equations earlier in this chapter. According to Girardi’s investigation, systematic errors up to tens of percent may be the consequences of the uncertainties of nuclear data taken from literature, especially those on decay schemes and activation cross-section[13].

2.6.8 Relative Method

The method is based on the simultaneous irradiation of the sample with standards of known quantities of the elements in question in identical positions, followed by measuring the induced intensities of both the standard and the sample in a well known geometrical position. The calculation of the unknown quantity (Msample) is made according to equation given below[14].

$$\frac{A_{sample}}{A_{standard}} = \frac{M_{sample}}{M_{standard}} \qquad 2.59$$

The disadvantage of the classic relative method lies in the multielement application. The procedure of the standard preparation and counting is rather laborious, and this is coupled with the occasional loss of information if an unexpected element appears for which no standard has been irradiated. The use of home-made multielement standards can be an answer to these problems, but this raises the question of homogeneity and stability. There are commercial multielements Standards Reference Materials (SRM) available[13].

2.6.9 Advantage and Disadvantage of NAA [16]

1. Advantage of NAA

- Because of its detection sensitivity, the NAA method has found important applications in many fields, like, medicine, biology, geochemistry, industry, art, military, archaeology, environmental science and forensic chemistry[14]
- **High sensitivity**

This advantage is first connected with the fact that measurements are based on the detection of each individual event which constitutes a radioactive disintegration. This means that 80-90% of radioactive disintegration from one sample can be detected. This fact is also associated with the high probability (cross section) of many nuclear reactions, to the high available fluxes of particles.

- **Easy standardization**

As the radioactivity produced does not depend on the chemical state, standardization is facilitated. Any chemical forms of elements may used as standards. There is no error related to the chemical environment of the elements. This is also a great advantage contributing to the accuracy of the method.

- **Multi-element analysis**

Using y-spectrometry, multi-element analysis is possible. Generally, about 40 elements can be determined after a single irradiation with reactor neutrons.

- **Direct measurement of radioactivity**

Using y-spectrometry and choosing appropriate irradiation conditions, instrumental analysis without chemical separation is possible in many cases.

- **Low cost counting equipment**

Compared to optical or mass-spectrometry, y-spectrometry uses rather low cost systems.

2 Disadvantage of NAA

- **Availability of irradiation sources**

The main disadvantage is the necessity of having access to rather heavy irradiation sources. This aspect limits the number of laboratories able to use activation analysis.

- **Delay of answer**

This delay occurs mainly from the fact that the irradiation source is generally not situated in the laboratory using it for analysis, and also from the long measurement times used for low radioactivities.

- **Variation of detection limits from one element to another**

One characteristic of nuclear reactions is the variation of cross sections from one isotope to another,leading to a variation in detection limits. It is impossible to perform a comprehensive analysis using only one type of irradiation. As already pointed out, reactor neutron irradiation leads to a multi-element analysis, but some elements are excluded. Other means of irradiation have to be used in order to extend the analysis to these elements.

- **Safety problems**

Obtaining the necessary authorizations, the knowledge of handling radio-elements and the use of control equipment may be a constraint. However, the radioactivity levels used in activation analysis are generally low.

CHAPTER THREE

3. EXPERIMENT

3.1 INTRODUCTION

Nuclides exist in two main forms, stable and unstable. A nuclide is considered to be stable if there is no proof of its spontaneous transformation into another nuclide. The probability of transformation is characterized by the half-life, which is defined as the time needed for half of the starting amount of an unstable nuclide to transform. Radioactivity is defined as the spontaneous nuclear transformation of an unstable element resulting in the formation of a new one. In this experiment we will study how to stable Sb-121 is bombarded with themal neutron and transformed to unstable Sb-122 and how to identify a given sample of interest and its capture cross section in the case of radioactivity resulting from bombardment with thermal neutrons. ”thermal” means with energies comparable with the Boltzmann energy kT, i.e. around 0.025 eV.[2]

3.2 Experimental setup

3.2.1 Neutron sources

The neutrons are produced in an americium-beryllium source. In this source 241Am is mixed into beryllium powder.

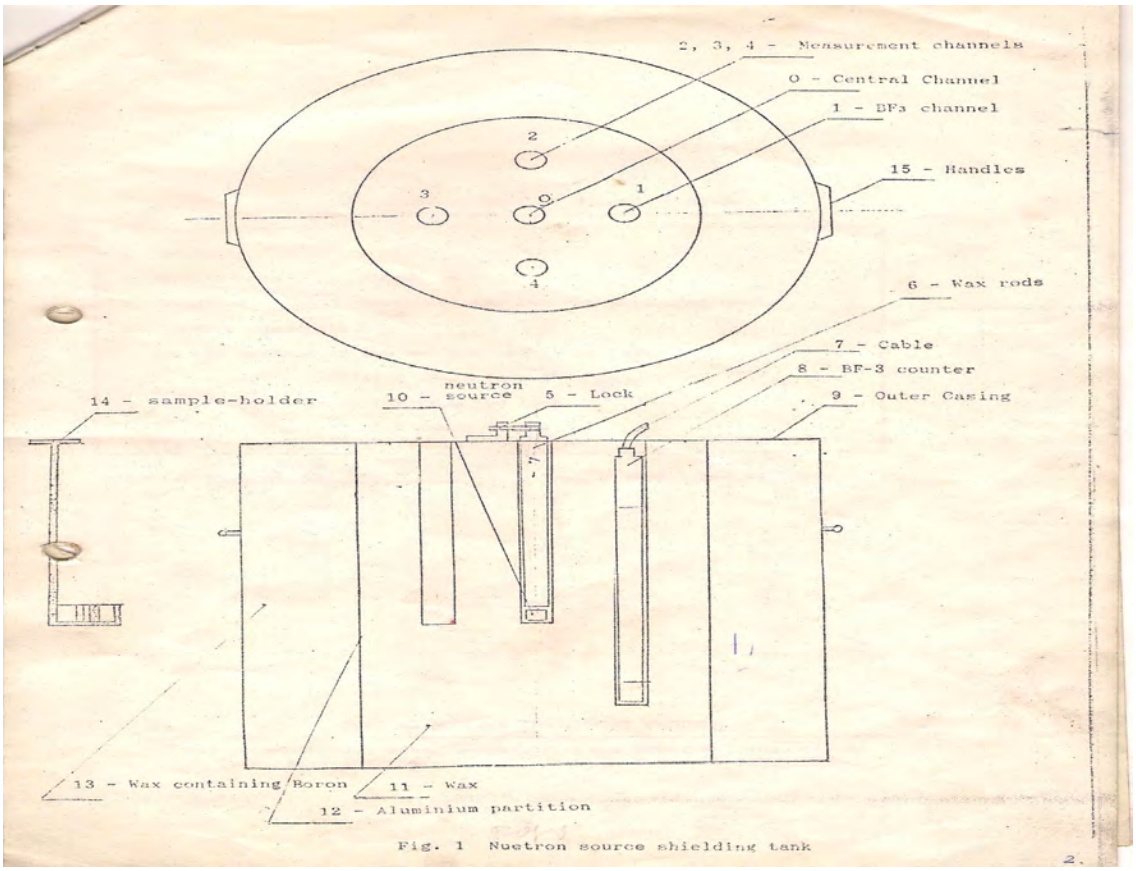


Figure 3.1 : schematic diagram of Am-Be neutron source

The neutron shielding tank is shown as fig.1, wher:

0- Central channel (for 241Am-Be neutron source, 30 × 30 mm 2 Ci)

1- BF3 channel, there is a red marked number 1 on the surface of the thank.

2, 3, 4- Measurement channels, there are wax rods inside. While any of channels is to be used, raise out the wax-rod and drop in a positioning sample-holder(14), on which the sample will be put. At one term of the holder there is a tong, sample of 10mm can be clipped or put on the top of tong for irradiation.

5- Lock, preventing neutron source missing

6- Wax rods, neutron protecting-aluminum cylinder filled with wax. Total four rods: three for measurement channels and one for BF3 channel.

7- Cables, connect BF3 tube to preamplifier FH1048.

8- BF3 counter SZJ-4 274 × 28 mm (active length 304 mm)

9- Outer Casing, 600 × 600 mm

10- 241Am-Be Neutron source 30×30 mm 2 Ci

11- pure wax filled

12- Aluminium partition

13-wax containing Boron

14- Positioning sample-holder(plexiglass), total three.

15- Handles

The following reaction is occurred in americium-beryllium source during bombardments of neutrons:

$$^{241}Am \rightarrow ^{237}Np + \alpha(2Curie, i'.e74GBq)$$
$$\alpha + ^{9}Be \rightarrow ^{12}C + n(5.75MeV) \qquad 3.1$$

Neutrons ejected with this energy can cause nuclear reactions but the cross-section is small. They can be slowed down by putting them into a medium containing light nuclei which does not contain nuclei that absorb them. Such a medium is called a moderator; the neutrons elastically scatter from the light nuclei, losing a portion of their energy at each collision. After a small number of collisions their energy becomes comparable with the thermal energy of the scattering nuclei and they are said to be thermalied. Suitable moderator nuclei are protons, deuterons, beryllium and all the stable nuclei of carbon, nitrogen and oxygen. All have very low cross-sections for neutron capture except the proton. This disadvantage is partly compensated for by the proton's small mass and by the fact that suitable hydrogen containing chemical compounds are freely available: water, plastics, wax, etc[17]. With a source of slow neutrons we can cause nuclear reactions to occur by processes such as

$$^{A}Z + n \rightarrow ^{A+1}Z + \gamma \qquad 3.2$$

where $^{A+1}$Z is radioactive with a "reasonable" half life and where the slow neutron capture cross-section is reasonably large. By "reasonable" we mean that the half life is much greater than the time required to take the irradiated sample from the source to the counter[18].

3.2.2 GAMMA RAY DETCTORS

In order for a gamma ray to be detected, it must interact with matter that interaction must be recorded. Fortunately, the electromagnetic nature of gamma-ray photons allows them to interact

strongly with the charged electrons in the atoms of all matter. The key process by which a gamma ray is detected is ionization, where it gives up part or all of its energy to an electron. The ionized electrons collide with other atoms and liberate many more electrons. The liberated charge is collected, either directly (as with a proportional counter or a solid-state semiconductor detector) or indirectly (as with a scintillation detector), in order to register the presence of the gamma ray and measure its energy. The final result is an electrical pulse whose voltage is proportional to the energy deposited in the detecting medium.[18]

1 Solid-State Detectors

In solid-state detectors, the charge produced by the photon interactions is collected directly. The gamma-ray energy resolution of these detectors is dramatically better than that of scintillation detectors. The sensitive volume is an electronically conditioned region (known as the ***depleted region)*** in a semiconductor material in which liberated electrons and holes move freely. Germanium possesses the most ideal electronic characteristics in this regard and is the most widely used semiconductor material in solid-state detectors. As the detector functions as a solid-state proportional counter, with the ionization charge swept directly to the electrodes by the high electric field in the semiconductor, produced by the bias voltage. The collected charge is converted to a voltage pulse by a preamplifier. The most popular early designs used lithium-drifted germanium [Ge(Li)] as the detection medium. The lithium served to inhibit trapping of charge at impurity sites in the crystal lattice during the charge collection process. In recent years, manufacturers have produced hyper pure germanium (HPGe) crystals, essentially eliminating the need for the lithium doping and simplifying operation of the detector.[18]
The purpose of an HPGe detector is to convert gamma rays into electrical impulses which can be used, with suitable signal processing, to determine their energy and intensity.

Due to its far superior resolution, *HPGe* is the only radiation detection technology that provides sufficient information to accurately and reliably identify radionuclides from their passive gamma ray emissions. *HPGe* detectors have 20-30 times better resolution than *NaI* detectors. Also, unlike *NaI* detectors, *HPGe* detectors are resistant to information (signal) degradation caused by changes in background radiation, shielding, multiple radionuclide interference, and temperature

variations. *HPGe* is also much more effective than *NaI* detectors in stand-off radionuclide identifications such as are encountered in vehicle or shipping container inspections.

In the quiescent state, the reverse-bias-diode configuration of a germanium solid state detector results in very low currents in the detector (usually in the pico- to nano ampere range). This leakage current can be further reduced from its room temperature value by cryogenic cooling of solid-state medium, typically to liquid nitrogen temperature (77 K). This cooling reduces the natural, thermally generated electrical noise in the crystal but constitutes the main disadvantage of such detectors: the detector package must include capacity for cooling, and this usually involves a dewar for containing the liquid coolant.[18].there are different kinds dewars therefore it can be selected.

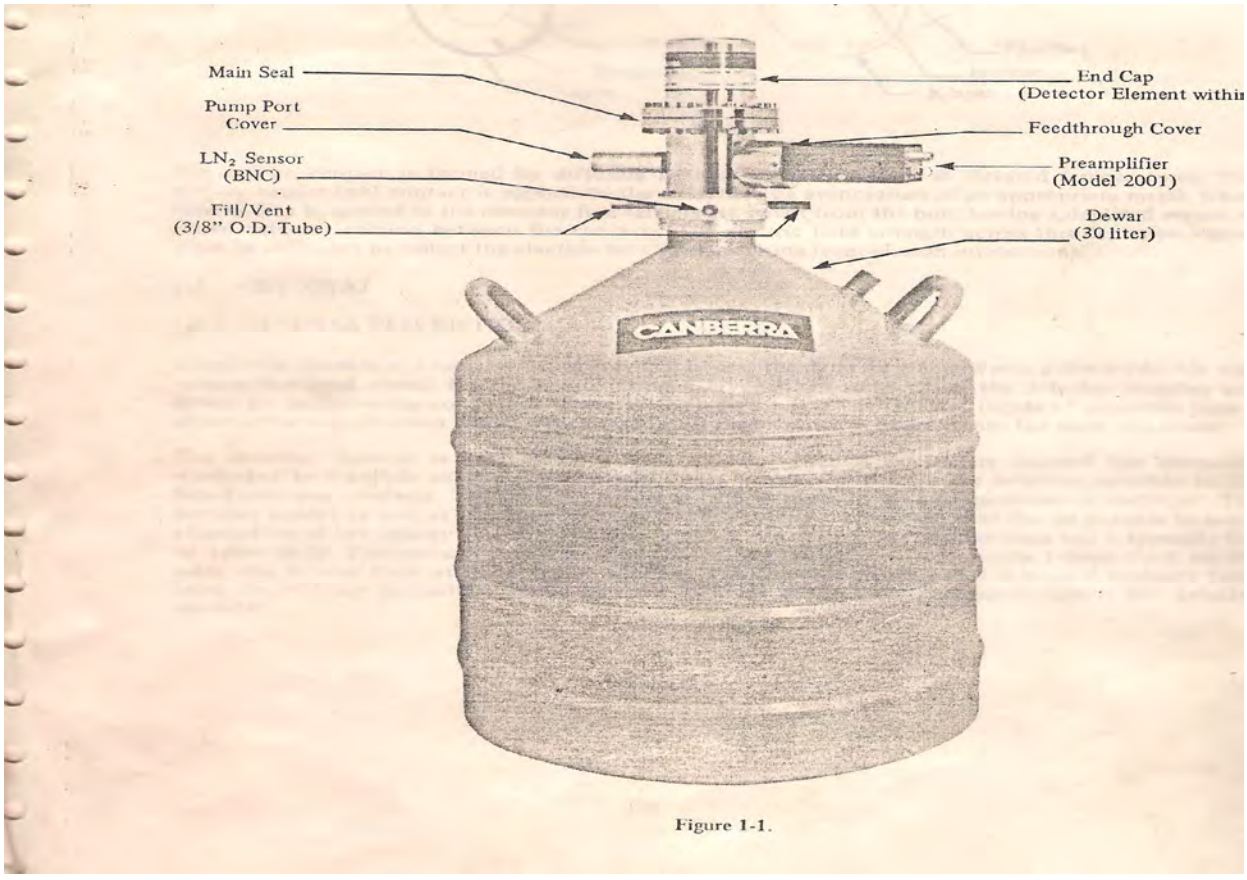


Figure 3.2 A schematic diagram *of* HPGe detection *sys*tem.

HPGe is Semiconductor gamma spectrometers consist of a semiconductor radiation detector assembly and signal processing electronics interfaced to a pulse height analyzer and a computer.

The detector is a semiconductor crystal between two conductor electrodes. A potential difference is established between the electrodes thereby producing an electric field in the semiconductor. When a γ-ray photon enters the semiconductor it produces free charge carriers in the crystal, the number of which is proportional to the energy lost by the γ-ray photon. The charge motion resulting from the influence of the electric field produces an induced current pulse in the external circuit. The integrated current pulse is proportional to the energy lost by the γ-ray photon. The pulses are to a multichannel pulse-height analyzer (MCA) where they are sorted and stored according to the amplitude distribution to produce a pulse-height graph that corresponds to the gamma energy spectrum. The MCA may be a dedicated instrument or an analog-to-digital converter (ADC) interfaced to a computer.

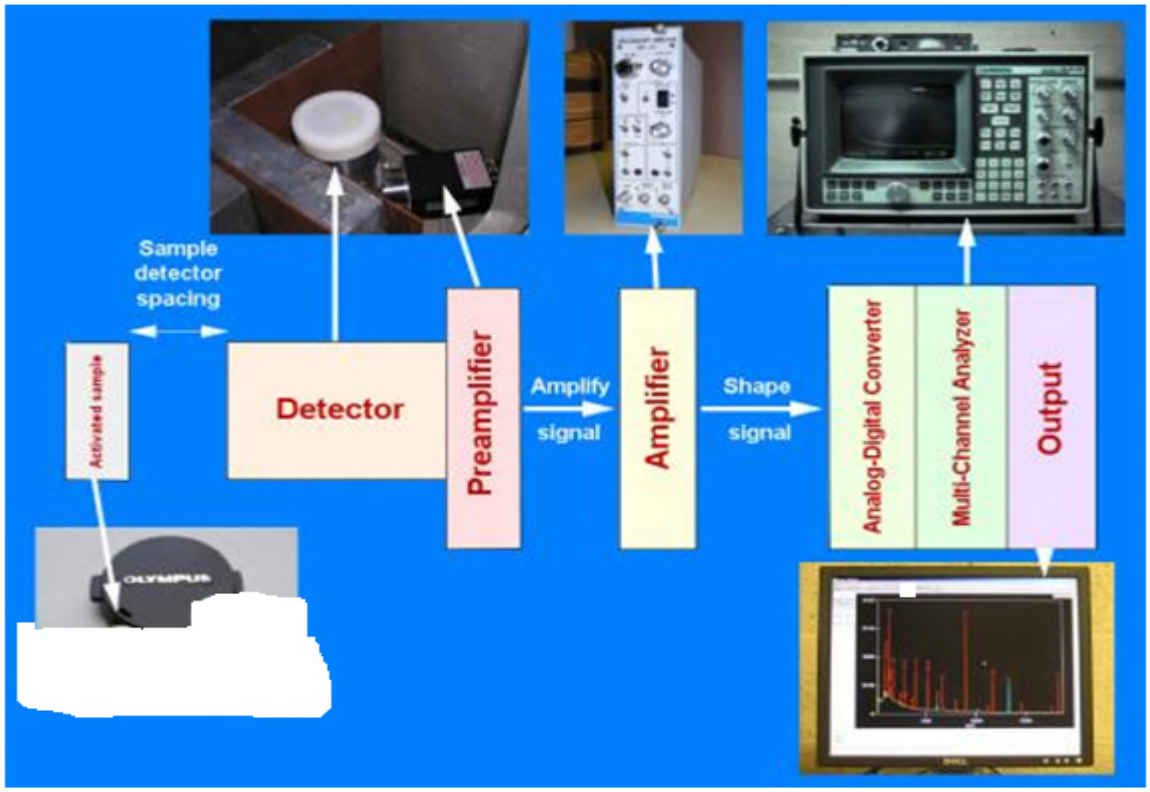


Fig.3.3 Blocks diagram of organization of experiment.

3.3 Background Measurements

Extraneous radiation called background radiation is always present. Gamma rays emitted by certain radioisotopes in the ground, in air, and from various building materials as well as cosmic radiation from outer space can all provide counts in a detector in addition to those from a sample being measured. The background in gamma ray detectors can be expected to increase roughly as the detector volume. Therefore, in critical situations where low background is at a premium, it is important to select a detector size that is not larger than necessary to give a reasonable counting efficiency for the sample to be counted. In our setup background radiations were measured and noticed to differentiate it from those radiations of sample.

3.4 Sampling and Irradiation

The chemical compounds or samples prepared for the experiment are;

Potassium Iodide (KI)

Antimony trioxide powder (Sb_3O_2)

3.4.1 Antimony Trioxide

Compounds of antimony were known to ancient cultures. They have been found, for example, in the colored glazes used on beads, vases, and other glassware. But these compounds were not widely used until the Middle Ages when they became popular among alchemists. They thought that antimony could be used to convert lead into gold. It was during this period that records about the properties of antimony begin to appear. The element was probably first named by Roman scholar Pliny (A.D. 23-79), who called it stibium. Muslim alchemist Abu Musa Jabir Ibn Hayyan (c. 721-c. 815) probably first called it antimony— *anti* ("not") and *monos* ("alone"). The name comes from the fact that antimony does not occur alone in nature. [19]

Antimony (Sb) is a chemical element with the atomic number 51. It is a brittle, fusible crystalline solid that exhibits poor electrical and heat conductivity properties. Antimony is commonly used in batteries as it increases the hardness and strength of lead. It is also used in flame proofing compounds and paint. Antimony and some of its alloys are unusual in that they expand on cooling. The major use of antimony is in lead alloys - mainly for use in

batteries - adding hardness and smoothness of finish. The higher the proportion of antimony in the alloy, the harder and more brittle it will be. Alloys made with antimony expand on cooling, retaining the finer details of molds.[20]

Antimony has 31 isotopes whose half-lives are known, mass numbers 104 to 136. Of these, two are stable and found naturally in the percentages shown: ^{121}Sb (57.36%) and ^{123}Sb(42.64%)

Antimony trioxide or Sb_2O_3, is the most widely produced compound of elemental antimony. The nations that produce the most antimony trioxide are China, South Africa, Bolivia, Russia, Tajikistan, and Kyrgyzstan.

Figure 3.4 Antimony Trioxide sample

Typical applications for antimony trioxide include flame retardant synergist for use in plastics, rubber, paints, paper, textiles, and electronics; polyethylene terephthalate polymerization catalyst; a clarifying agent for glass; an opacifier for porcelain and enamel; and a white pigment for paint. When used as a flame retardant, antimony trioxide is often used in combination with halogenated compounds. Antimony trioxide is used as a synergist to enhance the activity of the halogenated flame retardant. In the absence of antimony trioxide about twice as much halogenated compound would be needed to reach the same level of flame retardancy. Thus the

use of antimony trioxide in halogenated flame retardant applications reduces costs and often enables physical properties to be improved. In the minor applications it provides unique properties not easily obtained through other products. For example, friction modification, light stability of inorganic pigments.[21]

Disadvantage of antimony trioxide [22]

- Antimony trioxide can cause irritation of the respiratory tract.
- It can cause the heart to beat irregularly or stop.
- It can cause irritation, redness, and pain of eyes when in contact.
- Prolonged or repeated exposure may damage the liver and the heart muscle.
- There is an association between Sb_2O_3 production and an increased incidence of lung cancer.
- Persons with pre-existing skin disorders, impaired respiratory function, or heart disorders

These samples are borrowed from India by prof.A.K Chaubey in a powder form. Due to the volatility of pure iodine atom, the metal iodide salt is preferred for the determination of neutron flux 99 percent of the salt has I-127 than the rest isotopes of iodine and the natural abundance of Sb-121 is 57.3 percent. The samples has to be prepared suitably for irradiation in solid form by putting them in the ring and fasting both side by sticky tape, so that, we can have three samples having the following masses.

1. Potassium iodide(sample1) = 0.1178gm
2. Potassium iodide(sample2) = 0.4316gm
3. Antimony trioxide Sb_2O_3 (sample3) = 0.1775gm

The samples were made ready by sandwiching the antimony trioxide sample in between the two KI targets in order to have fixed geometry and by putting them in the sample holder. Irradiation process started when the samples are inserted in the Am-Be neutron source found in our

laboratory of AAU Science faculty nuclear physics department. The duration of irradiation was about 15 days.

3.5 Activation of KI

Thermal neutron reaction with the KI target results in (n, γ) reaction with that of I-127 element.

$$^{127}I + n \rightarrow ^{128}I + \gamma(prompt) \rightarrow ^{128}Xe + \beta^{-} + \gamma(delayed)$$

The excited iodine[^{128}I] de-excited to the ground level of Xe atom by emitting three types of beta particles having the following end point energy and branching ratio[1].

$$\beta_1 = 2.12MeV \rightarrow 80\%$$
$$\beta_2 = 1.676MeV \rightarrow 11.6\%$$
$$\beta_3 = 1.149MeV \rightarrow 1.52\%$$

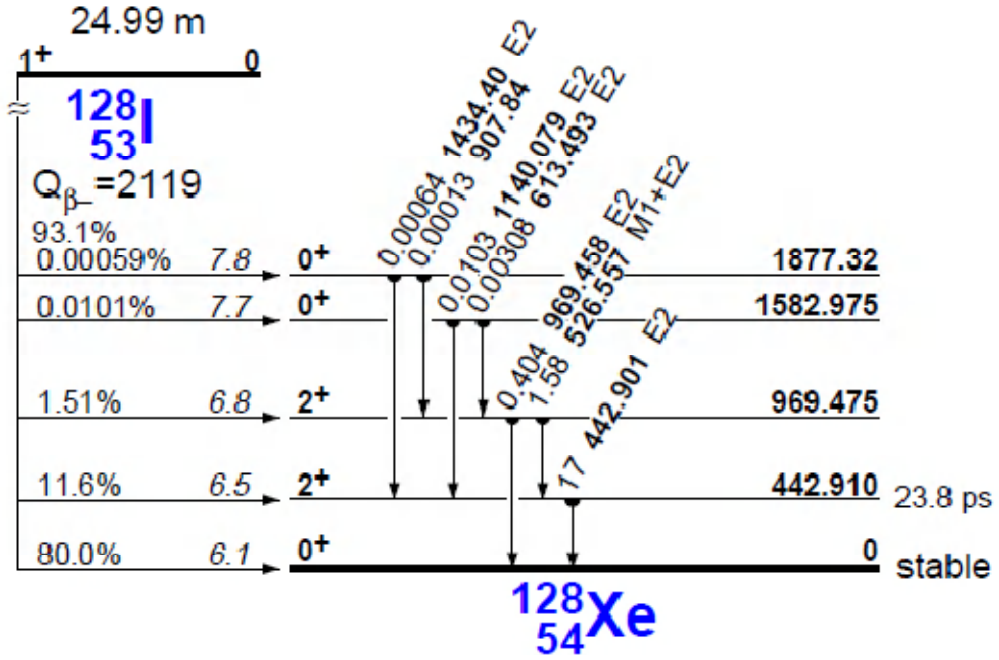
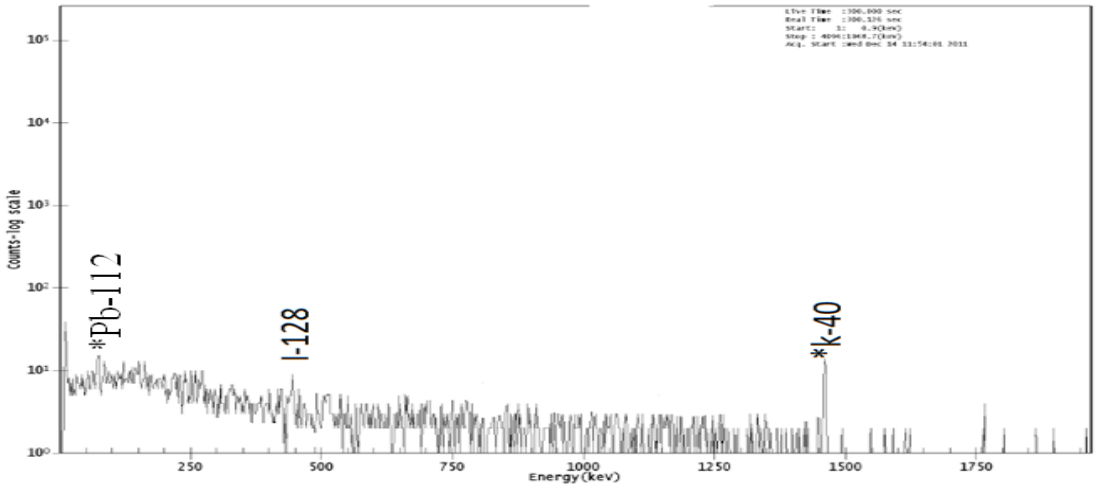
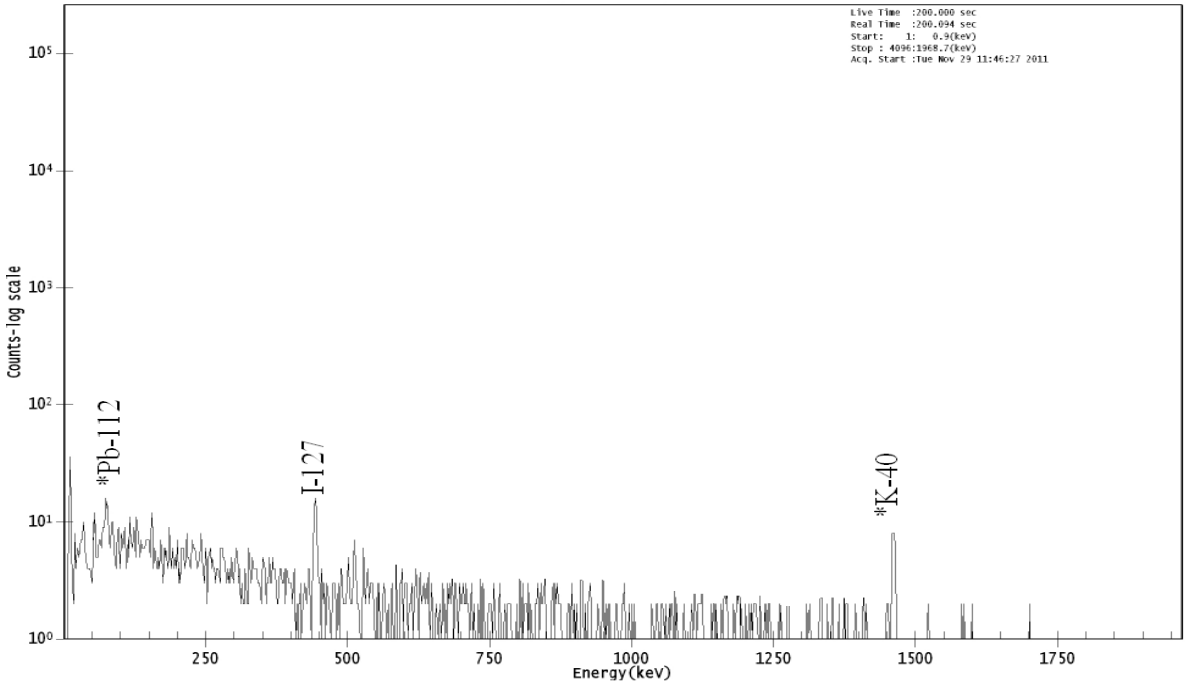


Figure 3.4: Decay scheme of I-128 [1]



Graph 3.1 Gamma spectrometer of front I target.

* Indicates that the elements are not present in Sample.



Graph3.2 Gamma spectrometer of back I target.

* Indicates that the elements are not present in Sample.

3.6 Activation of Antimony

During time of irradiation from antimony trioxide only antimony has been excited. Since oxygen is light element neutron activation analysis *cannot provide information.* Thermal neutron reaction with the antimony target results in (n, γ) reaction with that of Sb-121 Metalloid element.

$^{121}Sb + n \rightarrow ^{122}Sb + \gamma(prompt) \rightarrow ^{122}Te + \beta^{-} + \gamma(delayed)$ (E_{γ}=563.6KeV, E_{max}=1982KeV)

The excited antimony[^{122}Sb] de-excited to the ground level of Te-122 atom by emitting three main types of beta particles having the following end point energy and branching ratio[21].

$\beta_1 \rightarrow E_1 = 1.982 MeV \rightarrow 26.07\%$
$\beta_2 \rightarrow E_2 = 1.418 MeV \rightarrow 66.75\%$
$\beta_3 \rightarrow E_3 = 0.725 MeV \rightarrow 4.6\%$

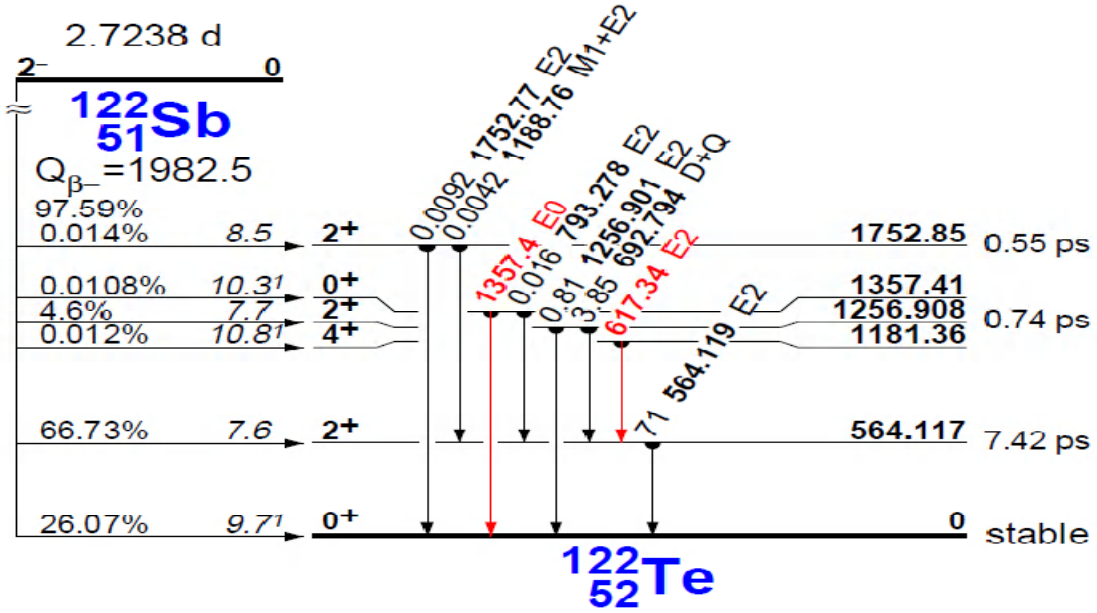
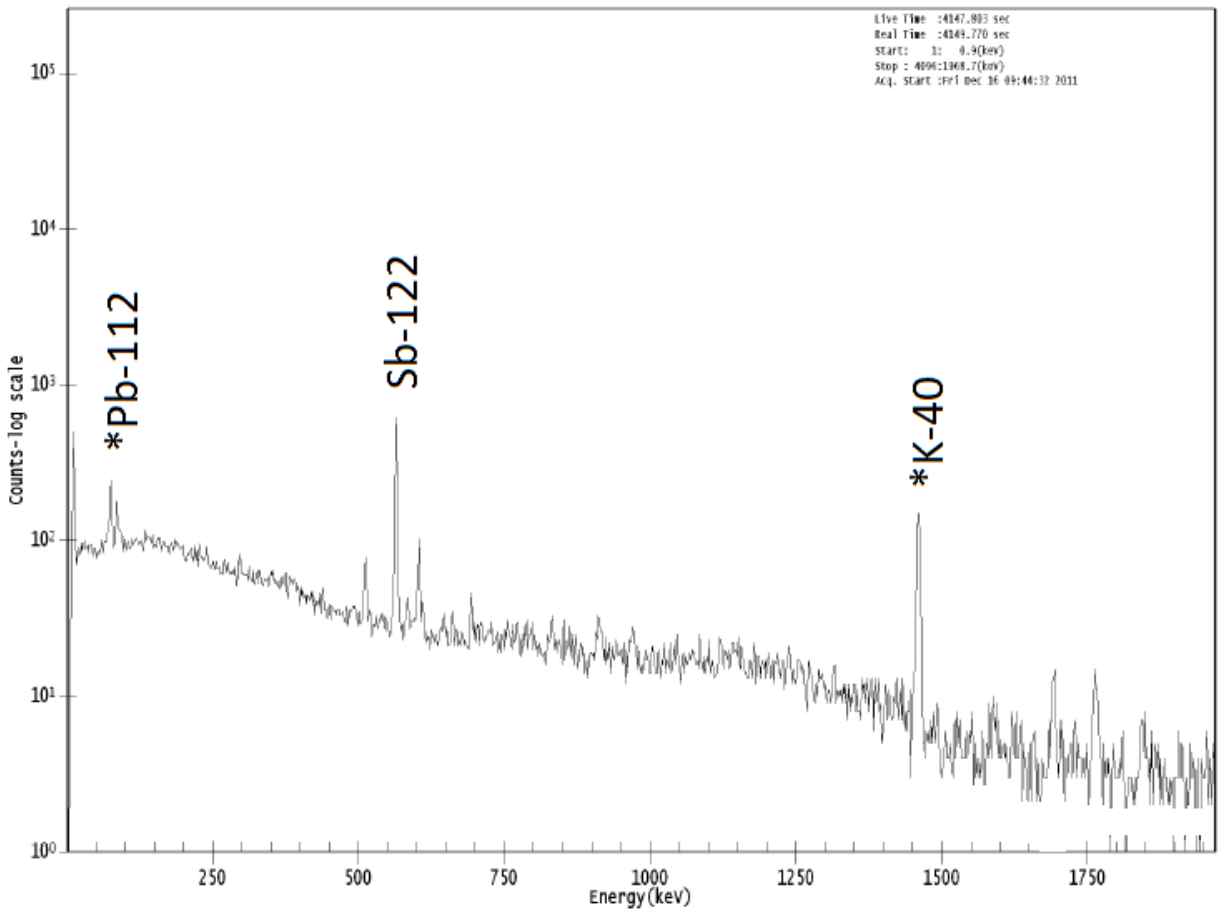


Figure 3.5 Decay Scheme of Sb-122 [1]



Graph 3.3 gamma spectrometer of Antimony target.

* Indicates that the elements are not present in Sample.

3.7 Measurement (Detection)

Before the measurement of the activity of the samples detection system or detector (HPGe and the associated electronics) has to be prepared in its best operating region and the background radiation had to be determined , then samples can be taken by noting down the time of irradiation stopped and taken to the detector as fast as possible in order to minimize the delay time(td).The activity of each samples measured consecutively by varying count rate time till the samples are decayed. The two KI samples counted for 2 hours and the long half-life Sb-122 was counted for 7 days .At each count the corresponding time is recorded. such measurement were done with samples and cross section for $^{121}Sb(n,)^{122}Sb$ reaction was calculated.

Chapter 4

4. Data and Data Analysis

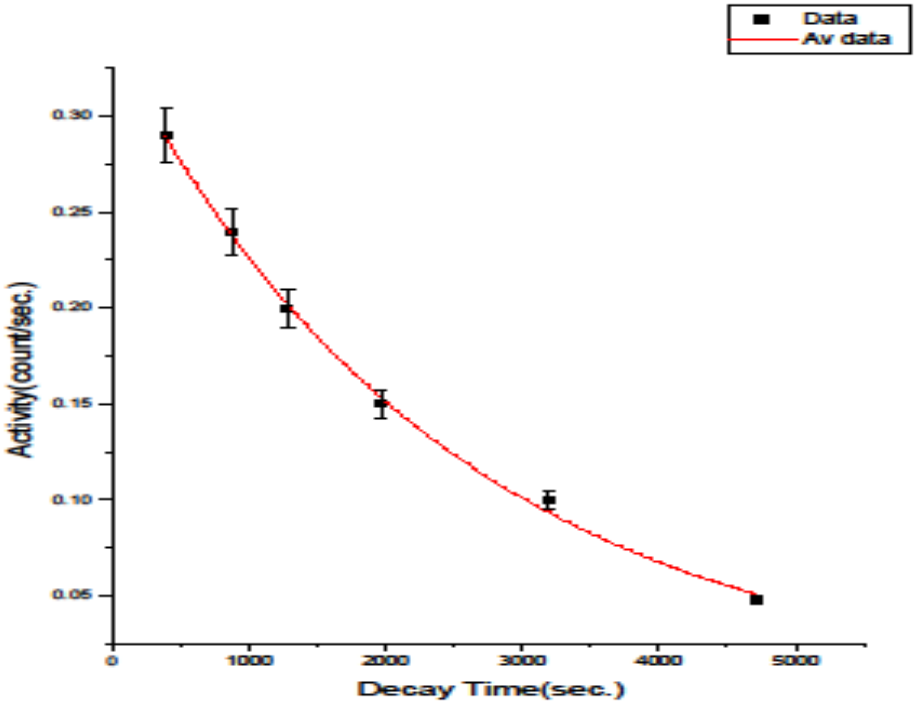
Using instrumental neutron activation analysis method, the reaction of thermal neutron with Sb-121 can be studied by taking two KI samples which are used to determine the thermal neutron flux. After sample was irradiated for sixteen days the decay of element by emitting gamma energy is measured by High purity Germanium detector.

4.1 Data From HPGe Detector

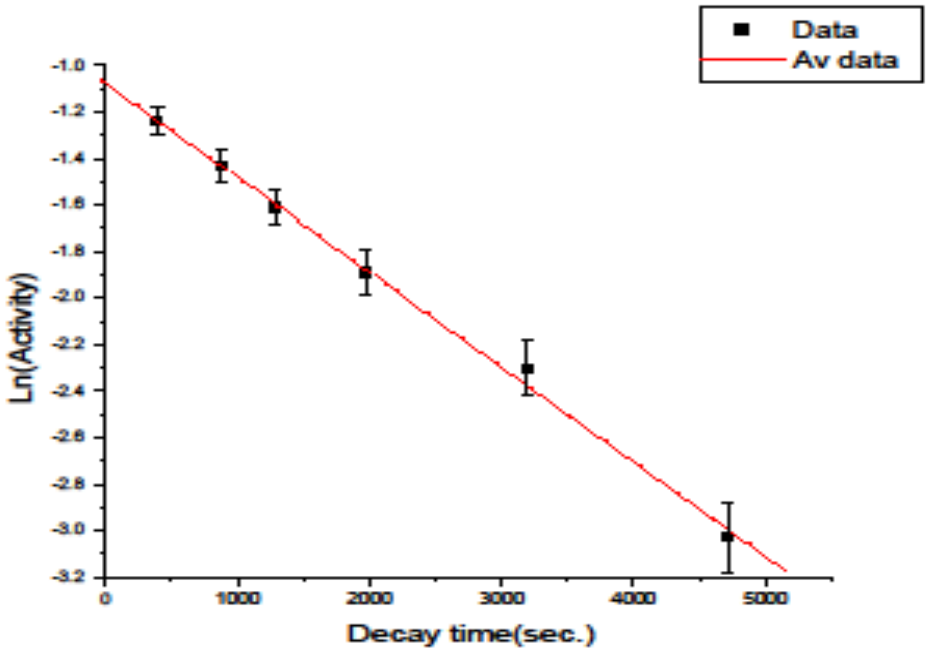
High Purity Germanium (HPGe) is the only radiation detection technology that provides sufficient information to accurately and reliably identify radionuclides from their passive gamma ray emissions.

Energy(KeV)	**Area**	**Count time(Second)**	**Decay time(second)**
443.5	29 ± 9.27%	100	396
443.5	24 ± 12%	100	876
443.5	40 ± 2%	200	1288
443.5	39 ± 2%	300	1972
443.5	56 ± 18%	400	3045
443.5	14 ± 5%	600	4715

Table 4.1 Decay Table of front I-127



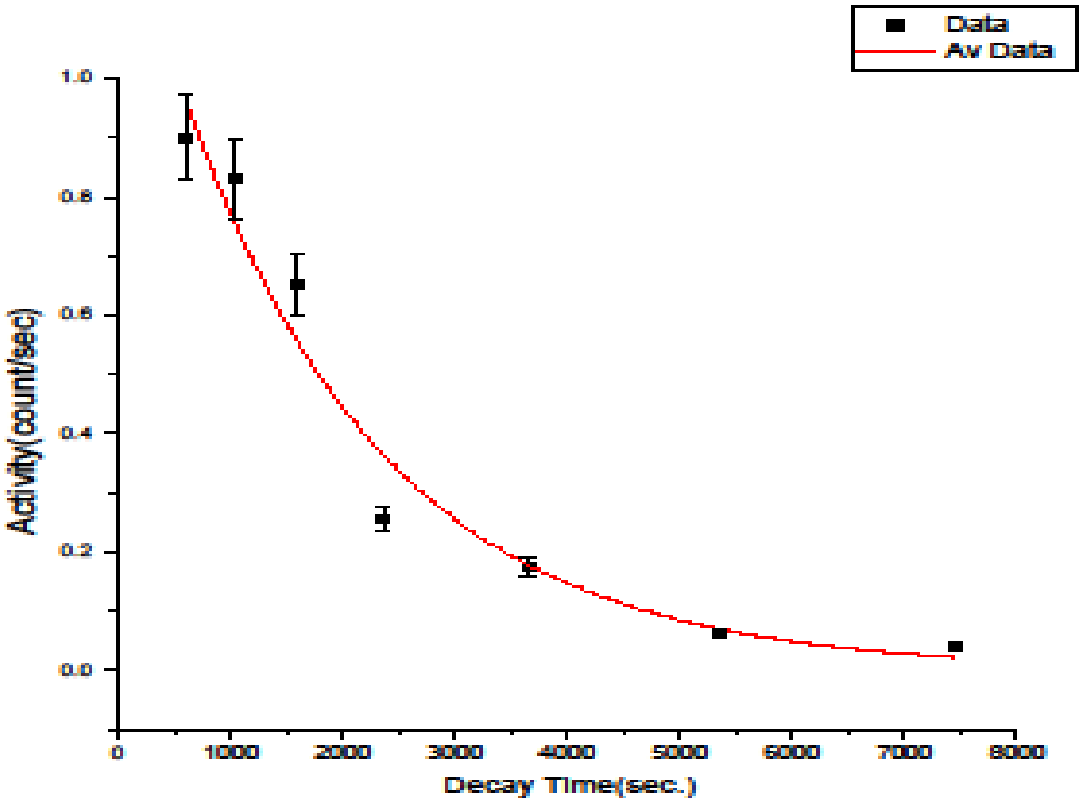
Graph 4.1: Exponential decay curve of front I target



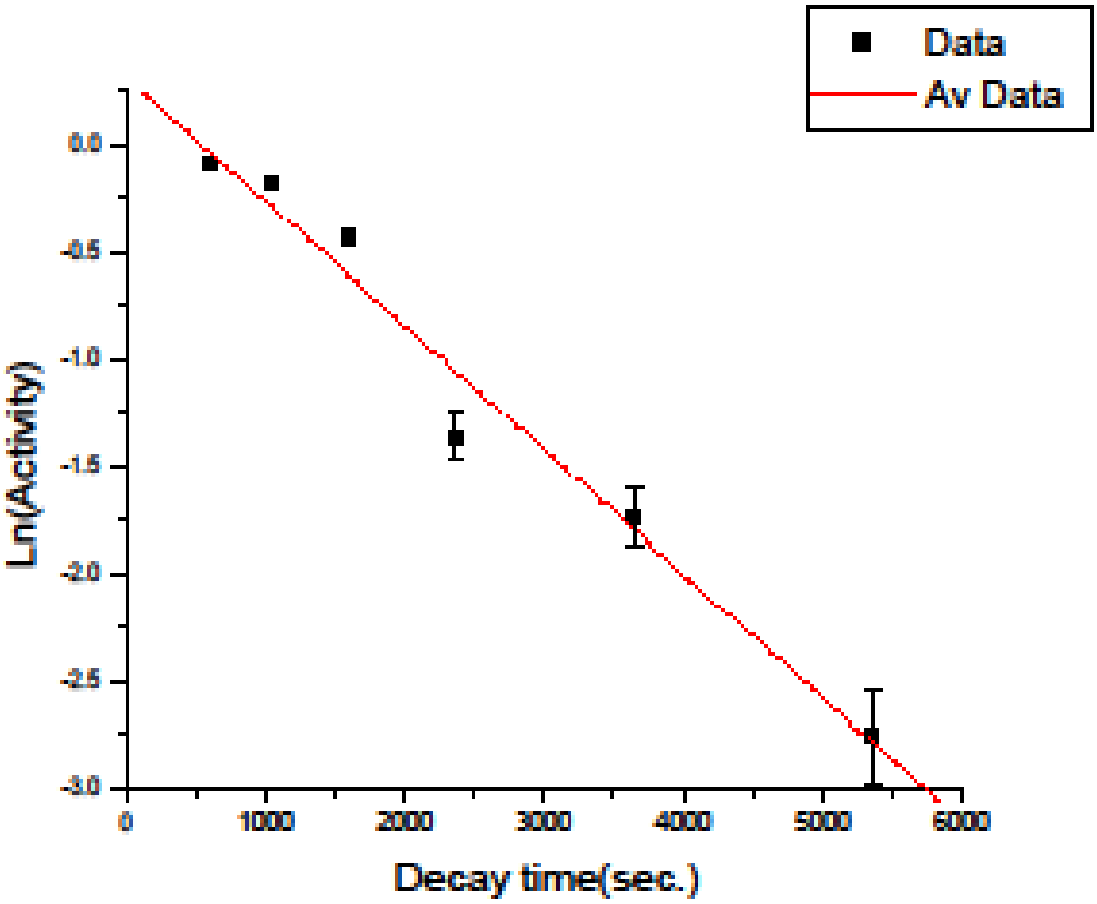
Graph 4.2 Logarithmic decay curve of front I target

Gamma energy (KeV)	Area	Count time (second)	Decay time(second)
443.5	90 ± 11.62%	100	607
443.5	83 ± 11.72%	100	992
443.5	130 ± 9.4%	200	1544
443.5	107 ± 11.94%	300	2260
443.5	70 ± 13.77%	400	3501
443.5	38 ± 3.5%	600	5164
443.5	31 ± 8.7%	800	7450

Table 4.2 Decay table of back I -127 target



Graph 4.3: Exponential decay curve of back I target

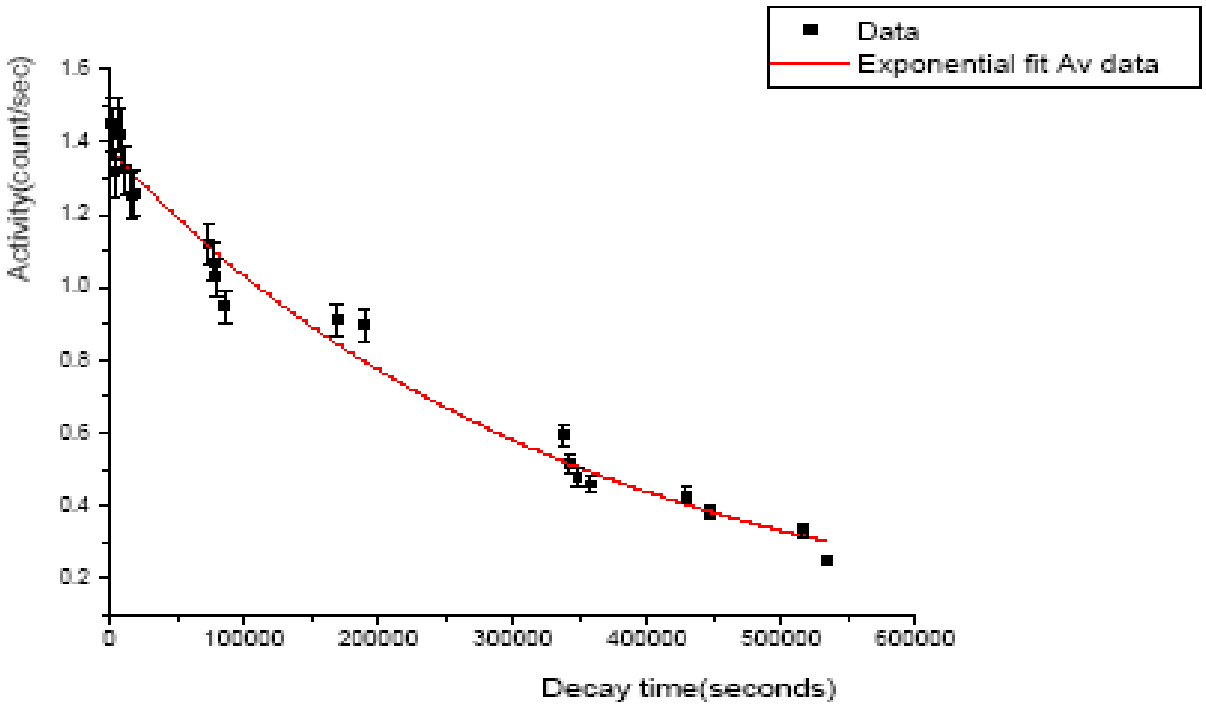


Graph 4.4 Logarithmic decay curve of back I target

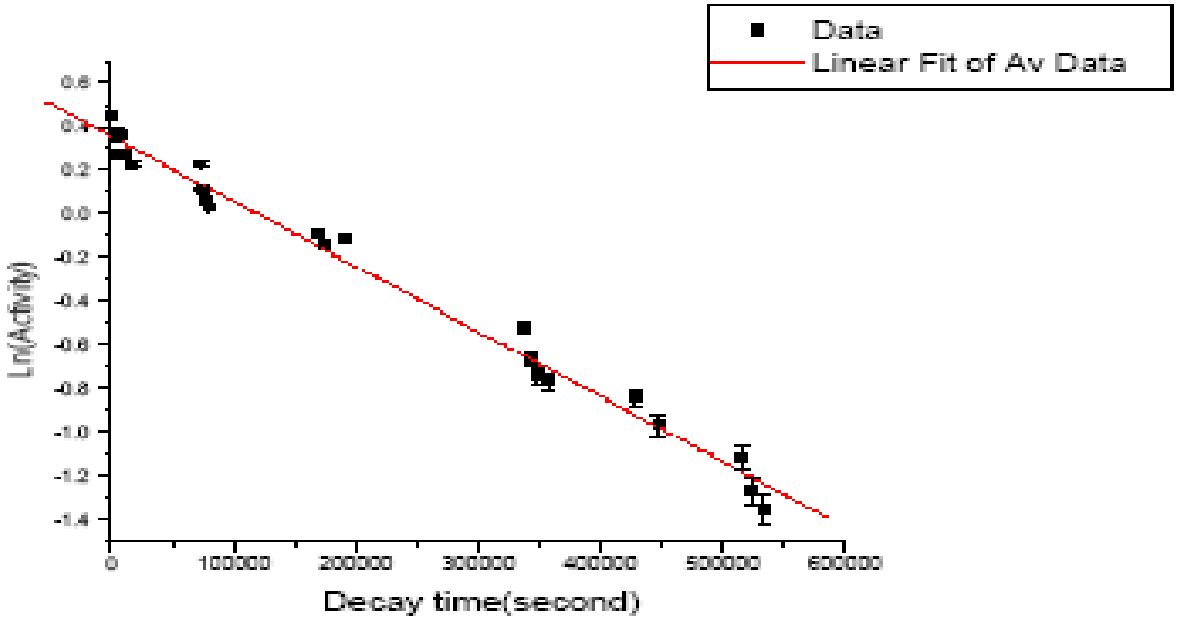
Gamma Energy(KeV)	Area ± error(%)	Count time (sec)	Decay time(second)
563.6	145 ± 9.34	100	230
563.6	427 ± 5.4	300	2610
563.6	526 ± 4.93	400	3920
563.6	875 ± 3.76	600	5790
563.6	1142 ± 3.33	800	8380
563.6	1325 ± 3.19	1000	11540
563.6	1877 ± 2.71	1500	16110

563.6	2529± 2.31	2000	17920
563.6	2525 ± 2.13	2000	73000
563.6	2799 ± 2.13	2500	73585
563.6	3211 ± 2.01	3000	77725
563.6	3090 ± 2.11	3000	79645
563.6	3314 ± 2.09	3500	86290
563.6	3802 ± 1.84	4174	168826
563.6	4695 ± 1.67	5400	174720
563.6	3227 ± 1.94	3600	190440
563.6	2138 ± 1.67	3600	337980
563.6	1847 ± 2.74	3600	342540
563.6	3445 ± 2.09	7200	348720
563.6	4139 ± 1.93	9000	357960
563.6	3102 ± 2.14	7200	428880
563.6	2808 ± 2.35	7200	434940
563.6	2078 ± 2.69	5400	447420
563.6	2389 ± 2.49	7200	516180
563.6	2522 ± 2.62	9000	524640
563.6	1871 ± 23	7200	533580

Figure 4.3: Decay table of Sb-122



Graph 4.5: Exponential decay curve of Sb-122

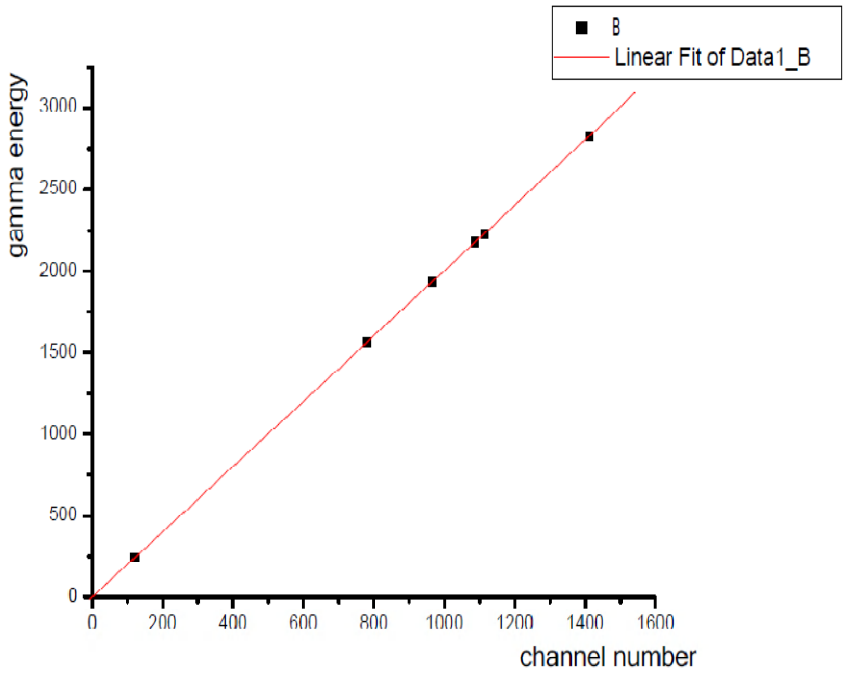


Graph 4.6: Logarithmic decay curve of Sb-122

Before the measurement has taken, calibration of the detector was done by Europium source such that the HPGe detector works properly.

Channel number	Gamma energy
244	121.8
1564	780.4
1966	966.1
2180	1087.9
2232	1113.9
2828	1411.4

Table 4.4 Energy Calibration Data



Graph 4.7 Energy Calibration.

4.2 Efficiency curve of the HPGe detector

This carve gives efficiencies of all energies of gamma detected by HPGe which emitted from the radioactive elements at zero distance from the detector which was measured by using Europium source. To plot it by the results of my colloquies;

Energy of gamma ray	Geometric eff. Of det.
122.8	0.123
244.7	0.086
344.3	0.081
444.0	0.052
778.9	0.035
867.4	0.032
964.0	0.031
1085.8	0.038
1112.1	0.035
1408.1	0.03

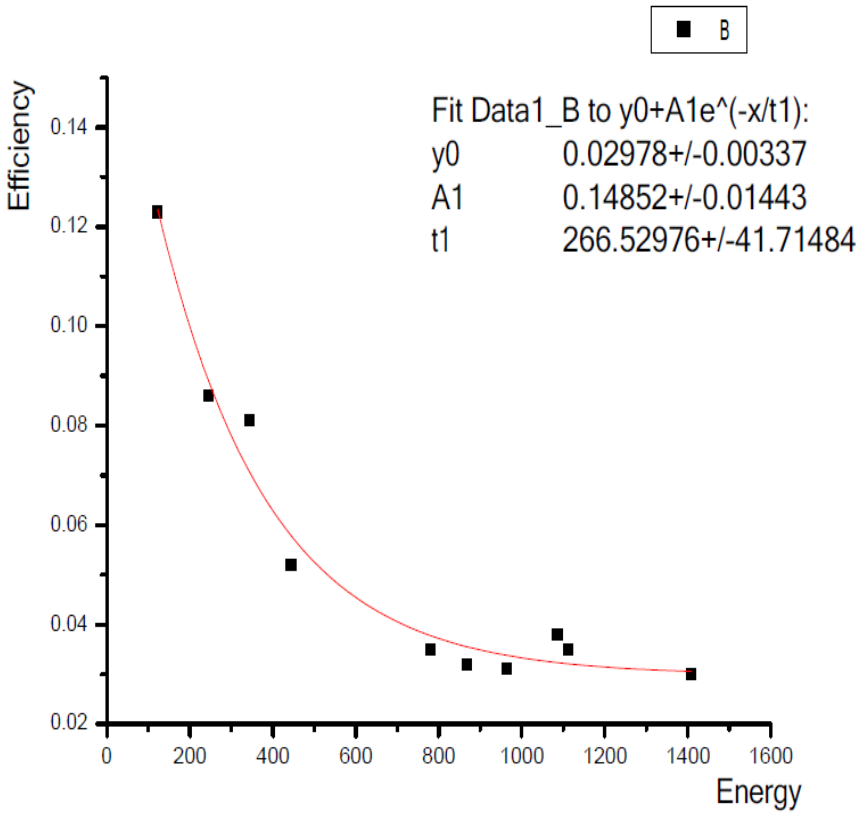


Table.4.5 Decay table of efficiency

Graph 4.8 Exponential decay curve of Efficiency.

4.3 Results and Discussions from HPGe detactor

4.3.1 Decay constant determination for the front and back I target for first experiment

From the logarithmic curve presented in graph(4.2) for the front KI target, the decay constant of I-127 can be determined by computer software which shows the logarithmic decay curve of iodine.

$\lambda_1 = 4.58\times10^{-4}s^{-1}$

From the logarithmic graph for the back I target, graph(4.4), the decay constant or the half-life of I-127 can be calculated by determined by computer software, which represent the logarithmic decay of its activity.

$$\lambda_2 = 4.62\times10^{-4}s^{-1}$$

4.3. 2. Determination of Half-life of Sample

From the logarithmic curve presented in fig(4.6) for the antimony target, the decay constant of Sb-122 can be determined by computer software which shows the logarithmic decay curve of antimony.

$\lambda_3 = 2.961\times10^{-6}s^{-1}$

Therefore, $T_{1/2} = \frac{\ln 2}{\lambda_3} = \frac{0.693}{2.961\times10^{-6}s^{-1}} = 2.71dys$ this is exactly conceded with the theoretical value.

4.3.3 Neutron flux determination

The incident neutron flux used to activate the target elements may be calculated from the equation 3.8

$$\phi_1 = \frac{n(\exp\lambda t_d)}{\varepsilon I_\gamma k\sigma N_0[1-\exp(-\lambda t_{irr})][1-\exp(-\lambda t_c)}$$

For the flux one given parameters are:

n = 0.049 count/sec.

t_d = 4715 sec

t_c = 600 sec

t_{irra} = 1296000 sec.

ε = 0.05 from gaph 4.8

$\lambda = 4.620 \times 10^{-4}\, s^{-1}$

$k = \dfrac{e^{-\mu_m H}}{\mu_m H} = 0.999996 \approx 1$

$k \approx 1$

$N_0 = \dfrac{mNf}{A}$ where m = measured mass of Iodine

N= Avogadro's Number

A = Atomic weight

f = percentage of gamma abundance

Therefore,

$$N_0 = \frac{mNf}{A} = \frac{127 gram/mole}{168 gram/mole} \times \frac{0.1178 gm}{127 gram/mole} \times 6.02 \times 10^{23} atom/mole = 4.22 \times 10^{20} atoms$$

$\sigma = 6.2 barn$

The value of neutron flux incident is found to be;

$$\phi_1 = \frac{0.049 c/s \times e^{4.60 \times 10^{-4} s^{-1} \times 4715 s}}{4.22 \times 10^{20} atoms \times 6.2 \times 10^{-24} cm^2 \times 0.05 \times 0.17 \times \left(1 - e^{-4.60 \times 10^{-4} \times 1296000}\right)\left(1 - e^{-4.62 \times 10^{-4} \times 600}\right)}$$

$$= 3.61 \times 10^4\, n/cm^2 s$$

The flux in the back I target can be found using the same procedure. Given parameters for flux two are:

n = 0.063 count/sec.

t_d = 5364 sec.

t_c = 600 sec.

ε = 0.05 from gaph 4.8

$$N_0 = \frac{mNf}{A} = \frac{127 gram/mole}{168 gram/mole} \times \frac{0.4316 gm}{127 gram/mole} \times 6.02 \times 10^{23} atom/mole = 1.55 \times 10^{21} atoms$$

Using the given values in the above, the neutron flux in the second I target becomes

$$\phi_2 = \frac{0.063 c/s \times e^{4.621 \times 10^{-4} s^{-1} \times 5364 s}}{1.55 \times 10^{21} atoms \times 6.2 \times 10^{-24} cm^2 \times 0.05 \times 0.17 \times \left(1 - e^{-4.60 \times 10^{-4} \times 1296000}\right)\left(1 - e^{-4.62 \times 10^{-4} \times 600}\right)}$$

$$= 3.58 \times 10^4\, n/cm^2 s$$

The average neutron flux captured by the two iodine samples calculated as;

$$\phi_{avg} = \frac{\phi_1 + \phi_2}{2}$$

Then, $\phi_{avg} = \frac{3.61\times10^4 + 3.58\times10^4}{2}$

$= 3.6\times10^4 n/cm^2 s$

4.3.4 Neutron Capture Cross-Section of ^{121}Sb

To evaluate the capture cross-section of Sb-121 by thermal neutron flux already determined, we use equation 3.8

$$\sigma = \frac{n(\exp \lambda t_d)}{\varepsilon I_\gamma k \phi N_0 [1-\exp(-\lambda t_{irr})][1-\exp(-\lambda t_c)}$$

Taking Activity at t_d=0 and Eliminating t_c because half life of the sample is large, in few seconds the nuclide may not decayed, thermal neutron capture cross section can be given by:

Where n = 1.49 c/s

$N_0 = \frac{mNf}{w}$ Where m = mass of target

N = Avogadro's number

f = natural abundance of target element

w = atomic weight

$\varepsilon = 0.05$ from gaph 4.8

$$N_0 = \frac{mNf}{w} = \frac{242g/mole \times 0.1775g}{290g/mole} \times 0.573 \times \frac{6.03\times10^{23} atom/mole}{242g/mole} = 2.11\times10^{20} atoms$$

$$\sigma = \frac{1.45c/s \exp(2.97\times10^{-6} s^{-1} \times 0s)}{2.11\times10^{20} \times 3.6\times10^4 \times 0.05 \times 0.72 \times \left[1-\exp\left(-2.97\times10^{-6} s^{-1} \times 129600s\right)\right]}$$

$= 5.45 \pm 0.23 barn$

Therefore, the experimental value of thermal neutron capture cross section of Sb-121 is 5.45 $\pm$ 0.23 barn which is less than the value presented in the literature which is 5.84 $\pm$ 0.2 barn.

Total error in experiment can be calculated as:

$$Error(\%) = \frac{previousvalue - experimentalvalue}{previousvalue} \times 100\%$$

$$= \frac{5.84 - 5.45}{5.84} \times 100\%$$

$$= 6.67\%$$

4.4 Sources of Error Estimation

The possible systematic and random source of errors for this are:

- ✓ The capture cross section of Sb-121 in this experiment is slightly less than the value found in the literature the reason may be the presence of epithermal neutrons around 4%, which is already observed by irradiating, samples one without cadmium cover the other with cadmium cover.[23]
- ✓ In the measurement of masses of samples
- ✓ The background radiation (the counter is not shielded)
- ✓ personal errors in the rounding of the calculated results
- ✓ In the libratory class there are also radiation emitting nuclei which can disturb the radiation emitted from sample.
- ✓ Contamination of sample before and after irradiation.

CHAPTER 5

5. Theoretical Estimates of Thermal Neutron Capture Cross Sections

5.1 Compound Nucleus

When considering the interactions of neutrons with matter, it is important to distinguish real reactions from potential scattering. In *potential scattering* there is no actual contact between the neutron and the target nucleus (i.e. nuclear forces are not brought into action); the wave associated with the neutron is scattered by the nuclear field. In a "real" reaction, on the other hand, the neutron penetrates the nucleus. In the energy range of interest here, the reaction can be described by the *compound-nucleus model*. This model has three stages:[2]

1/ The entrance channel: The target nucleus incorporates the incident neutron, producing the isotope of the next rank up, and giving this isotope an excitation energy equal to the sum of the *binding energy* of the extra neutron (work of the nuclear binding forces) and the *kinetic energy* contributed by the neutron,

2/ The actual life of ***the compound nucleus***, an isotope of the target nucleus: its lifetime is brief on a human scale — on the order of 10^{-14} s — but long on a nuclear scale, i.e. compared to the time required for the new nuclear bond to be created, which is on the order of 10^{-22} s. This means that the excitation energy can become "uniform" within the compound nucleus. In other words, it lives long enough to "forget" that it was created via the entrance channel. What happens next will be independent of the process that created the compound nucleus (absorption of a neutron, a proton, a gamma particle, etc.),

3. The *exit channel*: the excited compound nucleus will very quickly disintegrate by a radioactive-type process. With the energy acquired when the neutron was absorbed, there are several possible mechanisms which compete with each other.

5.2 Statistical Estimates of Thermal Neutron Capture Cross Sections

An estimate of the cross sections of nuclear reactions with thermal neutrons in terms of the average parameters of the target nucleus (the strength function, the average level spacing, and the

average reaction width) is obtained. At thermal energies, the cross section of a reaction is completely determined by the parameters of several low-lying resonances. These parameters are vary significantly from one nucleus to another and the exact value is unpredictable. [24]

These results from:

a. Variations of the strength functions from one nuclide to another and

b. random fluctuations of the positions and widths of the low-lying resonances that dominate the thermal cross section.

In particular, the expected value of the thermal capture cross section is expressed through the strength functions of s resonances- S_0 for neutrons and $S_{\gamma 0}$ for photons:

$$\sigma = \pi^3 \left(\frac{A+1}{A}\right)^2 \lambda\!\!\!^{-2}_T \left(\frac{E_T}{E_0}\right)^{1/2} S_0 S_{\gamma 0} \quad [13] \qquad 5.1$$

$$\sigma = 0.40 \times 10^8 \left(\frac{A+1}{A}\right)^2 S_0 S_{\gamma 0} \quad [13] \qquad 5.2$$

Here, A is the atomic weight of the target nucleus, E_T = 0.025 eV is the thermal energy, and E_0=1eV. In Eq. 5.2 we assume the following:[24]

1. All reaction widths are equal to the corresponding mean values.
2. The energy spacing between the resonances of the same spin are constant.
3. The resonances are located symmetrically with respect to the zero neutron energy point.

By using the above formula the thermal neutron capture cross section in antimony is calculated as following: According to the extreme compound, or black, nucleus model the strength function is constant for all nuclei, and for s-wave neutrons is given by

$$S_0 = \frac{\left\langle g\Gamma_n^0 \right\rangle}{\left\langle D_0 \right\rangle} \text{ Where, } \frac{\left\langle \Gamma_n^0 \right\rangle}{\left\langle D_0 \right\rangle} = \frac{2k_0}{\pi k} = 1 \times 10^{-4} \quad [25] \qquad 5.3$$

Where $\left\langle \Gamma_n^0 \right\rangle$ is the average S-wave reduced neutron width, D_0 is the average s-wave level spacing, K_0 is the wave number for a 1eV neutron and k is the wave number inside the nucleus.

$$g = \frac{2J+1}{(2s+1)(2I+1)} \quad [24] \qquad 5.4$$

$where J = I \pm 1/2$ I=5/2 spin of Sb- target

S= ½ spin of neutron

then, substituting the values of S and I one can obtain the value of g=7/12 or 5/12

$g_{avg}=0.5$

$$therefore S_0 = \frac{\langle g\Gamma_n^0 \rangle}{\langle D_0 \rangle} = 0.5\times10^{-4}$$

$$S_{\gamma 0} = \frac{\langle \gamma_{\gamma 0} \rangle}{D_0} \qquad [25] \qquad 5.5$$

And the value of $\xi = \frac{D}{2\pi\gamma_\gamma}$ was calculated [26]. From that paper one can drive for the

$\frac{D}{\gamma_\gamma} = 60\times 2\pi$ where $\xi = 60$ [26]

Therefore, $S_{\gamma 0} = \frac{\langle \gamma_{\gamma 0} \rangle}{D_0} = \frac{1}{60\times 2\pi} = 0.00265$

Using equation 5.2 thermal neutron capture cross sections for antimony target is:

$$\sigma = 0.40\times10^8\left(\frac{A+1}{A}\right)^2 S_0 S_{\gamma 0}$$

$$= 0.40\times10^8\left(\frac{121+1}{121}\right)^2 0.5\times10^{-4}\times 0.00265$$

$$= 5.32\pm 0.21 barn.$$

In this case of study, as the above calculation indicate the theoretical value of the thermal neutron capture cross section agreed with the measured value of thermal neutron capture cross section (with in error) which is obtained in our laboratory 5.45 ± 0.23barn.

5.3 Summary and Conclusion

Instrumental Neutron activation Analysis using Am-Be neutron source has profound effect in identifying the type of given element of sample. Here in this work, the half life and capture cross section of the sample Sb-122 is determined. The result of this work suggests that, using Instrumental Neutron Activation Analysis technique it is possible to perform environmental radio-analysis with an improved counter shielding and good precision of measurement. Additionally, these results show that the measured thermal neutron capture cross section have good agreement with statistical estimates of thermal neutron capture cross section. Neutron activation method, other than its wide application in the qualitative and quantitative elemental analysis, it is also be used in the determination of the neutron flux of a neutron source.

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Declaration

This thesis is my original work, has not been presented for a degree in any other University and that all the sources of material used for the thesis have been dully acknowledged.

Name: Getu Ferenji

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Place and time of submission: Addis Ababa University, March, 2012

This thesis has been submitted for examination with my approval as University advisor.

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Signature:

THE MEASUREMENT OF THERMAL NEUTRON CAPTURE CROSS SECTION IN ANTIMONY

By

Getu Ferenji Tadesse

SUBMITTED IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
MASTER OF SCIENCE IN PHYSICS
AT
ADDIS ABABA UNIVERSITY
ADDIS ABABA, ETHIOPIA
February 2012