Chapter 6

Prompt Gamma-Ray Activation Analysis

6.1 Introduction

A nucleus after absorbing neutron can get excited. This process can be used for the characterization of material composition. In the case of neutron activation analysis, the gamma-rays emitted by the radioisotope produced in the process are measured. However, the gamma-rays emitted by the excited nuclei decaying to ground state when measured, the method is called prompt gamma-ray activation analysis (PGAA) in principle the PGAA method can be used to detect almost all the elements in the periodic table from hydrogen to uranium. Prompt gamma-ray activation analysis is a non-destructive nuclear technique. In this method the sample under study is bombarded with thermal or fast neutrons. The gamma-rays, emitted through (n, γ) or (n, n'γ) type nuclear reactions, are measured during neutron irradiation.

The PGAA technique is very useful in detection of light elements such as hydrogen, boron, carbon, nitrogen, oxygen, silicon, phosphorous in a variety of samples [1-5]. This technique offers a unique advantage that the sample under test if kept in a cover or container can be analyzed. This is mainly because neutrons can penetrate the container, if made of medium or high Z element, without much attenuation in the intensity. As a result, the elements of the sample kept or hidden in a container can interact with the incident neutrons. The gamma-rays emitted from the elements have relatively high energies. These gamma-rays after existing from the container or cover can reach the detector without much attenuation in the intensity.

This PGAA technique is therefore useful in the analysis of multielement, without changing the shape, size and weight of the sample under interrogation. By using both thermal and fast neutrons, the PGAA technique has been successfully used for qualitative determination of a number of elements such as hydrogen, boron, carbon, nitrogen, oxygen, in explosive class materials. The landmine detection has therefore become

possible with this technique. Similarly, detection of (i) toxic elements such as cadmium, mercury (ii) rare earth elements such as samarium and gadolinium and (iii) archeological and geological samples, have been analyzed with PGAA technique which are difficult to analyze by chemical and other methods [6, 7].

6.2 Prompt Gamma-Ray Activation Analysis -: Thermal Neutrons

The main source of thermal neutrons is a nuclear reactor. A white beam of neutrons is incident on a suitable monochromator, and a beam of neutrons of energy in the range 1 meV to 100 meV can be obtained out of the reactor core for experiments. The flux of neutrons can be varied in the range of 10^5 to 10^7 n/cm^2-s. The other commonly used source for thermal neutrons is a radioactive Californium-252 source. In this source, the thermal neutrons are generated by slowing down of the fast neutrons of average energy ~2.3 MeV produced by spontaneous fission of Californium-252. These fast neutrons are thermalized in high density polyethylene blocks surrounding the source. The thermal neutrons from this Cf-252 can be used for variety of applications, including prompt gamma-ray analysis. However, the difficulty faced with the reactor neutrons is that the sample has to be brought in the reactor hall for the analysis. Even though in principle a Cf-252 can be transported to any place of interest, however, the heavy weight shielding around the source makes it difficult to take the source in a public places.

The thermal neutrons can be used for the analysis of elements such as hydrogen, nitrogen, iron, nickel, titanium, etc. The experimental set up has to be validated by analyzing standard samples with known weight of the elements. In principle, almost all the elements from hydrogen to uranium can be analyzed, however, inaccurate and incomplete data are significant hindrance in the qualitative analysis of a number of low and high Z elements. In general with neutron beam of flux 10^7 n/cm^2-s, the estimated amount of the elements can be reported with an accuracy of ~10%.

The Table 6.1 gives a few nuclear reactions which are induced by thermal neutrons, along with the energies of the prompt gamma-rays emitted by the excited nuclei produced in the reaction [8, 9]. As observed in Table 6.1 hydrogen and nitrogen on

interaction with thermal neutrons emit 2.223 MeV and 5.1 MeV gamma-rays respectively. It is observed from Table 6.1 that a number of elements have reasonable values of the reaction cross-sections for the emission of prompt gamma-rays. It is interesting to note that the elements namely samarium and gadolinium have very high cross-sections for the emission of prompt gamma-rays. However, the elements such as lithium, nitrogen, oxygen and carbon have very low values of cross-sections, and therefore use of the thermal neutrons can not be made for the analysis of such elements. The schematic of emission of prompt and delayed gamma radiations in neutron capture process is shown in Figure 6.1.

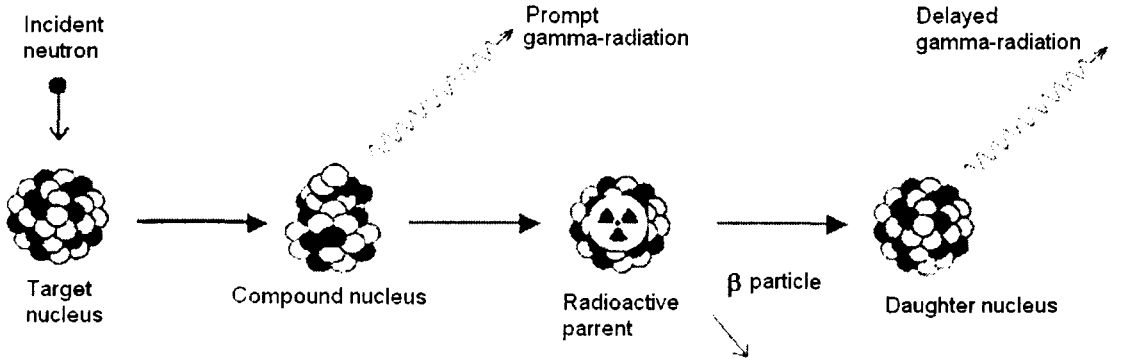


Figure-6.1: The induced gamma-radiations in the neutron capture process.

Table-6.1: Some of the nuclear reactions induced by thermal neutrons along with the energies of the prompt gamma-rays and reaction cross-section [8, 9].

Nuclear Reaction	Gamma-ray energy (MeV)	Cross-section (barns)
$^{1}H(n, \gamma)^{2}H$	2.223	0.3326
$^{7}Li(n, \gamma)^{8}Li$	2.032	0.038
$^{12}C(n, \gamma)^{13}C$	4.945	0.026
	1.261	0.0012
$^{14}N(n, \gamma)^{15}N$	1.884	0.014
	5.1	0.03
	10.83	0.075
$^{16}O(n, \gamma)^{17}O$	0.87	0.0001
$^{28}Si(n, \gamma)^{29}Si$	3.538	0.119
$^{31}P(n, \gamma)^{32}P$	0.636	0.031
$^{32}S(n, \gamma)^{33}S$	5.42	0.52
$^{35}Cl(n, \gamma)^{36}Cl$	1.951	6.51
	6.11	4.3
$^{50}Cr(n, \gamma)^{51}Cr$	0.834	1.38
$^{56}Fe(n, \gamma)^{57}Fe$	7.631	0.68
$^{113}Cd(n, \gamma)^{114}Cd$	0.558	1866
$^{149}Sm(n, \gamma)^{150}Sm$	0.334	4900
$^{157}Gd(n, \gamma)^{158}Gd$	0.182	7680

6.3 Prompt Gamma-Ray Analysis -: Fast Neutrons

Some of the elements such as carbon, nitrogen and oxygen which have low sensitivity for thermal neutrons can be analyzed with the fast neutrons, such as 14 MeV neutrons. The fast neutrons must have energies above the excitable nuclear levels of the interacting nuclei. The excited nuclei while decaying to lower energy levels emit gamma-rays which are used for the analysis. The fast neutron interrogation is carried out with neutrons of energies above 7 MeV.The cross-sections for the 14 MeV neutrons for the interaction with a few nuclei and energies of the gamma-rays are given in Table 6.2.

Table-6.2: Nuclear reactions induced by 14 MeV neutrons and the corresponding gamma-ray energies and reaction cross-sections [8-10].

Nuclear reaction	Gamma-ray (MeV)	Cross-section (barn)
$^{12}C(n, n'\gamma)^{12}C$	4.43	0.23
$^{14}N(n, n'\gamma)^{14}N$	1.6, 2.3 and 5.1	0.314
$^{16}O(n, n'\gamma)^{16}O$	6.13	0.75
$^{133}Ba(n, n'\gamma)^{133}Ba$	1.43	0.400

In general, the fast neutron induced reactions useful for such studies are limited. However, explosive class materials contain carbon, hydrogen, nitrogen and oxygen as prominent elements. As a result, the fast neutrons can be used for the analysis of explosive class materials. A 14 MeV neutron generator can be made in a laboratory to obtain neutron flux ~10^8 n/cm^2-s. Similarly, with the availability of portable 14 MeV neutron tubes, the analysis by measuring prompt gamma-rays has become convenient.

6.4 Prompt Gamma-Ray Analysis -: Pulsed Fast Neutrons

A pulsed fast neutron source can be used for the elemental analysis of bulk samples. Fast neutrons, namely 14 MeV energy neutrons, can be produced through D-T reaction [11, 12]. The deuteron beam falling on a tritium target can be pulsed, so that pulses of neutrons are obtained. The pulse duration ~ 1 to 5 μsec is normally used for such studies. The penetrating depth of 14 MeV neutrons is large in almost all the elements. The elemental concentration in the interrogated material, such as oil, coal, cement, mineral etc. can be determined conveniently. This method is also very effective in the analysis of explosive class materials [13, 14] and illicit drugs [15]. The main advantage of this method is that explosives and illicit drugs even if hidden in container can be detected. However, the required amount of the explosives and illicit drugs has to be more than 200 grams. In general, all the explosives and illicit drugs contain carbon, hydrogen oxygen and nitrogen as their constituent elements.

Fast neutrons can be converted into thermal neutrons using suitable moderator. In this manner, the advantage of both the fast neutrons and thermal neutrons can be utilized in the analysis of elements such as hydrogen, carbon, nitrogen and oxygen present in the explosives and drugs.

This method is also very effective in the detection of landmines. According to a report of United Nations, efforts are being made by all the countries around the world to remove the underground mines which might have been plotted during the world war. A rough estimate indicates that there are 110 million live landmines active in about seventy countries. These live landmines inflict casualties at a rate of about 2000 persons per month. Some of these are killed and the rest of them lose the vital body organs. Even today, a number of countries are still planting landmines in the region of International borders enemy.

Even today, not much attention has been paid towards the development of demining technology. As a result plows or rakes are employed to take out the underground mines to surface. The modern technology of manufacturing landmines aims at using plastic components and at the same time makes minimum use of metals. As a result in several cases the metal detectors are not useful for the detection of landmines.

The fast neutrons can play an important role in locating landmines, if a portable 14 MeV neutron generator is made and mounted on a vehicle. This system will be useful because the demining in the military area is required to be performed very quickly, with high success rate.

6.4.1 Pulsed Fast Neutron Activation Analysis

The pulsed fast neutron activation analysis can be used in two modes; prompt gamma-ray measurement and neutron time of flight. The deuterium ions are bombarded on the tritium target in pulses. The duration of the pulse and the time gap between the two consecutive pulses have typical values 10 μsec and 100 μsec respectively.

In general when 14 MeV neutrons fall on a sample, the nuclei of the elements get excited. The excited nuclei during de-excitation emit gamma-rays. These gamma-rays are the characteristics of the elements present in the sample. The fast neutrons ~10^4 n/pulse can interact with carbon and oxygen through (n, n'γ) reaction. For the detection of nitrogen, thermal neutrons are preferred. The prompt gamma-rays emitted by carbon and oxygen are of 4.44 MeV and 6.13 MeV respectively. The neutron pulse last for a period of ~10 μsec. After that next neutron pulse arrive after a period of ~100 μsec on the sample. In the gap of 100 μsec, the fast neutrons get thermalized after passing through the moderator and hence can interact with nitrogen nuclei of the sample through (n, γ) reaction. The nitrogen is determined through the 3.94 MeV and 5.1 MeV gamma-rays. The other gamma-rays emitted by nitrogen is of 10.83 MeV energy which is the highest energy gamma-ray produced in the neutron induced reaction with nitrogen.

In the case of time of flight method, the time required for the neutrons to travel from the point of creation to the point of interaction with the sample is precisely measured. As the time of flight depends on the energy of neutrons, the prompt gamma-rays emitted are due to monoenergetic neutrons.

The neutrons are bunched in the short pulse of typical duration ~5 μsec. The pulse duration is normally made smaller than the time of flight of the neutrons. A schematic diagram showing the timing for the pulsed neutron beam is shown in Figure 6.2. The time required for the neutrons to pass through the sample of size close to that of suitcase can

be around 20 to 30 nsec. The advantage of this method is that by rotating the sample, 3D multielemental mapping can be made. This method is very effective in the detection of landmine and other explosive class materials.

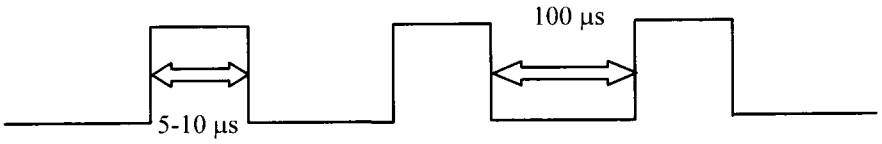


Figure-6.2: A schematic diagram showing timing for pulsed neutron beam

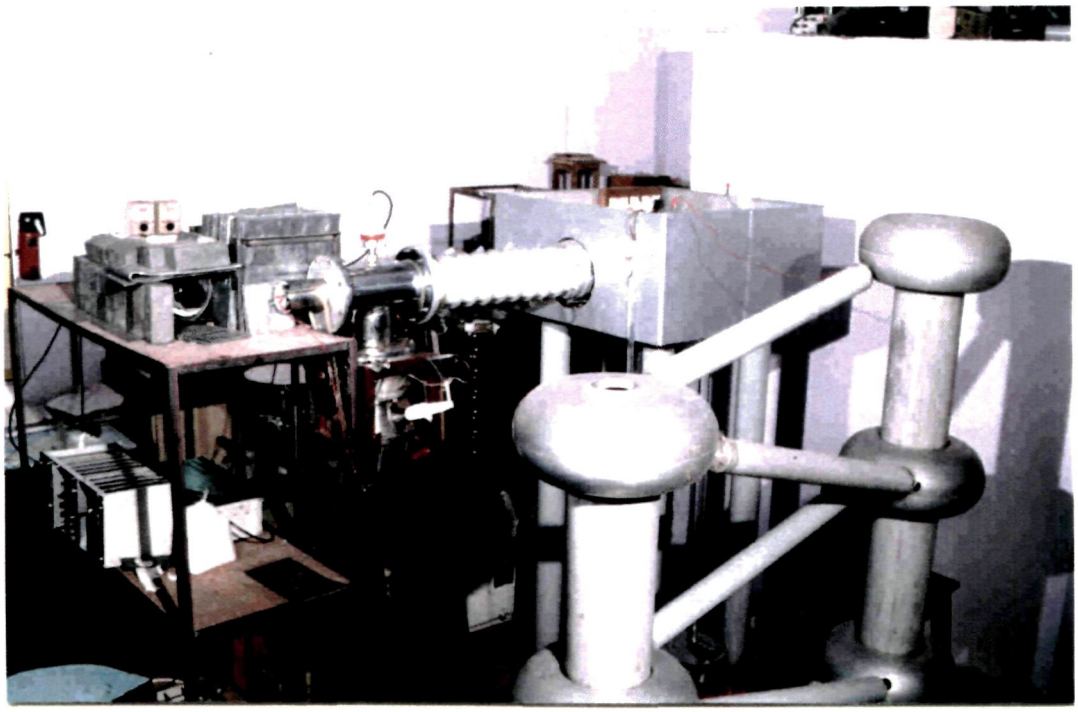
6.5 Experimental

In this laboratory of the Department of physics, University of Pune an effort has been made to analyze explosive class materials using 14 MeV neutrons. A feasibility study has been conducted using chemical compounds containing hydrogen, carbon, nitrogen and oxygen. These chemical compounds acts as explosive stimulants, having the same elemental composition as the explosives.

The 14 MeV neutron generator [11, 12] of the Department of Physics, University of Pune was employed for the present work. The 14 MeV neutrons were produced by impinging ~175 keV energy deuterium ions on the tritium target of 8 Curie activity. The neutron flux at the position of the sample was measured by neutron activation method and found to be ~10^7 n/cm^2-s. The 3"x 3" NaI(Tl) detector coupled to a 4096 channel analyzer, was used for the measurement of the prompt gamma-rays emitted by the sample. The experimental set up used in this experiment is shown in photograph 6.1.

In this method the sample under study was kept at a distance of 400 mm away from the tritium target. The NaI(Tl) detector was mounted perpendicular to the direction of the neutron incident on the sample. The weight of the sample taken for the experiment was 200 gm. The distance between the sample and the detector was typically 300 mm. The detector was housed in a case made of lead blocks. A 30 mm diameter hole was drilled in the lead blocks for providing passage to the gamma-rays from the sample. The

output of the NaI(Tl) detector was connected to 4096 channel MCA through amplifier and other electronic modules. The MCA was calibrated over 88 keV to 1332 keV gamma-ray energies using the Canberra make Multi Gamma Standard MGS-3 source. This radioactive source had Cd-109, Co-60, Cs-137, Mn-54 and Sn-113 isotopes.



Photograph-6.1: Experimental set-up used for the prompt gamma-ray activation analysis around the 14 MeV neutron generator.

The sample under study was kept in the line with the tritium target and irradiated with 14.77 MeV. The deuterium ion current on the tritium target was kept ~100 μA. The neutrons interact with the elements of the sample and thereby excite the nuclei of the elements. The elements were identified through characteristic gamma-rays emitted by the nuclei on interaction with the neutrons. The characteristic gamma-rays emitted by a few elements are given in Table 6.3. The intensities of the emitted gamma-rays were recorded by the NaI(Tl) detector coupled to 4096 channel analyzer. For each sample, the spectra of the prompt gamma-rays were recorded for a period of 2000 seconds. Initially, chemical compounds containing nitrogen, oxygen and hydrogen were irradiated with 14.77 MeV

neutrons and the prompt gamma-rays emitted by the sample were measured using the NaI(Tl) detector. The prompt gamma-rays emitted by nitrogen, oxygen and carbon were detected. A schematic diagram showing the experimental arrangement for the prompt gamma-ray analysis is shown in Figure 6.3. From the recorded spectra, the energy calibration of the NaI(Tl) detector was made. In other experiment about 200 gm of urea sample was irradiated with neutrons and the prompt gamma-rays emitted by the urea sample were recorded. The neutron irradiation and recording of gamma-rays were simultaneously carried out for a period of 2000 seconds. A typical prompt gamma-ray spectra recorded from urea sample is shown in Figure 6.4. Similar experiment was repeated using a sample of sugar. The prompt gamma-ray spectra recorded from sugar is shown in Figure 6.5 The characteristic gamma-rays emitted due to fast neutron interaction with the elements present in the chemical compositions were used to determine the elemental concentrations, by the following expression (6.1).

$$A_E = m \,.\, s.\, t \,.\, \theta \,.\, \sigma_0 \,.\, I_\gamma .\, \phi .\, \varepsilon \qquad (6.1)$$

Where, m-mass of the element, s-sensitivity defined as N_A/M, A_E-area under the photopeak, N_A-Avogadro's number, M-molar weight, θ-isotropic abundance, σ_0-neutron capture cross-section, I_γ-gamma yield, ϕ-neutron flux, ε-detector efficiency, t-counting period.

Table-6.3: Characteristic gamma-rays emitted by a few elements on interaction with thermal/fast neutrons [8, 9].

Element	Gamma-rays energy (MeV)
Lead	2.6
Carbon	4.43
Oxygen	6.1
Nitrogen	4.9, 5.1, 10.8
Hydrogen	2.2

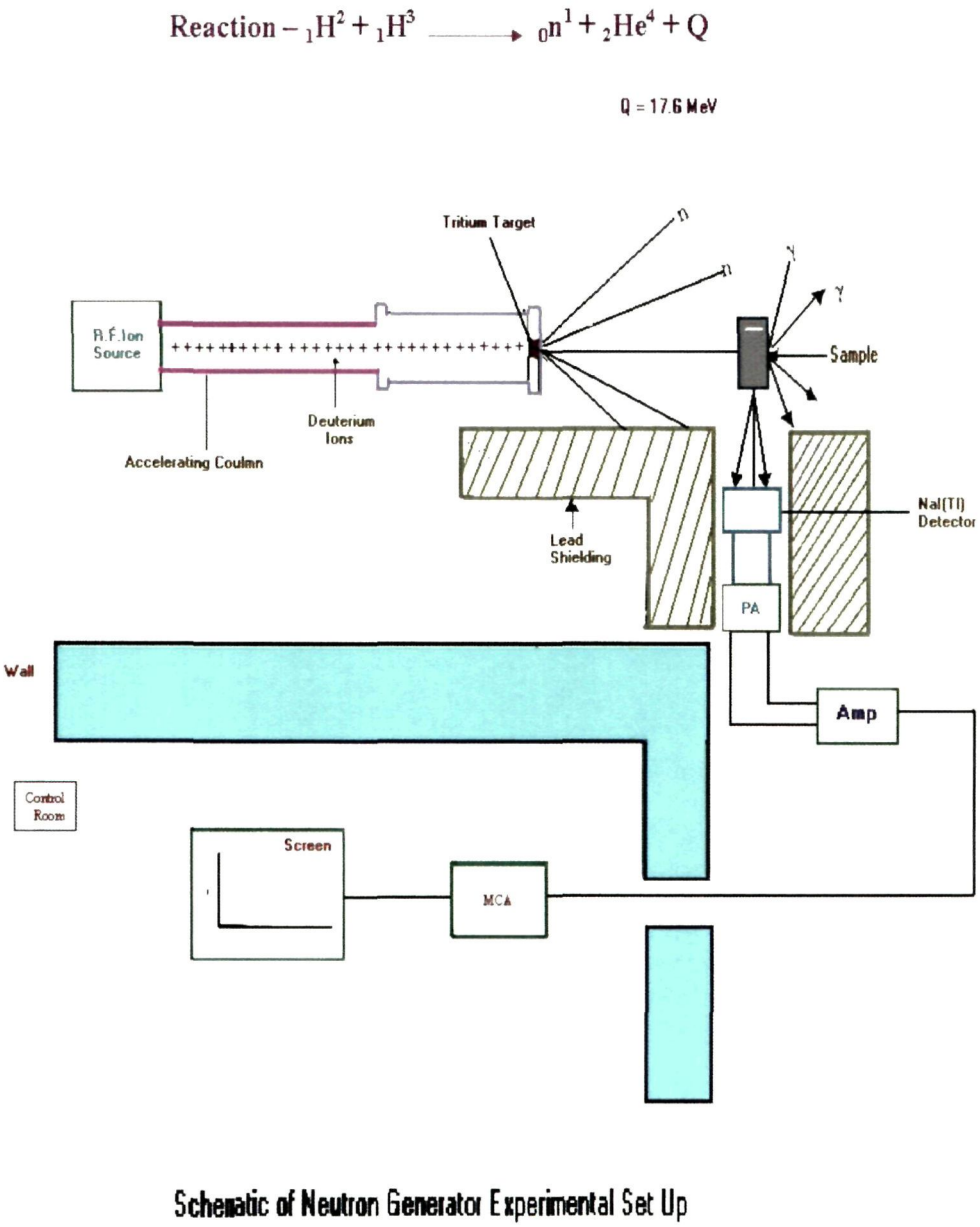


Figure-6.3: Schematic of 14 MeV neutron generator experimental set-up for prompt gamma-ray activation analysis.

6.6 Results and Discussion

A typical spectrum of the gamma-rays emitted from a chemical compounds urea and sugar during irradiation with 14.77 MeV neutrons is shown in Figure 6.4. and Figure 6.5 respectively. The characteristic gamma-rays of energies 6.1 MeV from Oxygen, 5.1 and 4.9 MeV from Nitrogen and 4.4 MeV from Carbon are well separated. The intensity of each photopeak is well above the background counts. The gamma-rays of 2.6 MeV energy are produced through the interaction of neutrons with the lead shielding. A computer program was used to fit the gamma-ray spectra and to estimate the area under each photopeak. The concentration of each element present in the sample was estimated from the area under the photopeak and using expression (6.1). The measured and the calculated values of the elemental concentrations and weight percentage of the elements in the urea and sugar are given in Table 6.4.

These results indicate that the prompt gamma-ray neutron activation analysis technique can be effectively used for the detection of carbon, nitrogen and oxygen in a sample. The elements such as nitrogen, carbon and oxygen are present in all the explosive class materials. The results of the present study therefore reveal that one can find out whether or not a given object under interrogation contains explosive class materials. The objective of the present work was to carry out a feasibility study to detect explosive class materials using the existing 14 MeV neutron generator. The detection of low Z elements such as carbon, nitrogen, oxygen etc. depends on the matrix of the sample under study, and therefore, the estimation of actual weight of the elements in the sample is possible with the pre-calibrated data only.

In the present experiment the main difficulty faced was the background gamma-rays emitted from the surrounding materials during neutron irradiation.

A lot of efforts was therefore required to design the neutron and gamma-ray shieldings between the neutron generator and the NaI(Tl) detector. Moreover, the required size of the sample is relative large to obtain a good signal to noise ratio.

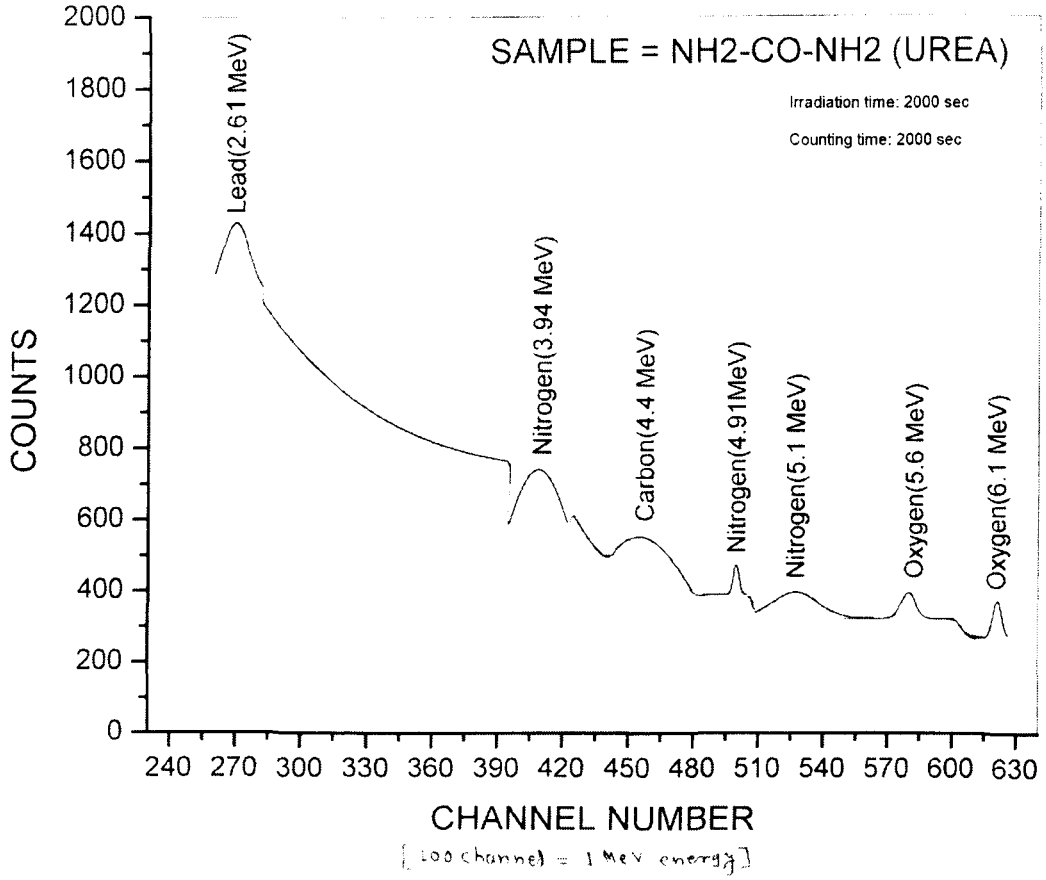


Figure-6.4: A typical recorded spectra of the prompt gamma-rays emitted from a chemical compound, urea, during neutron irradiation

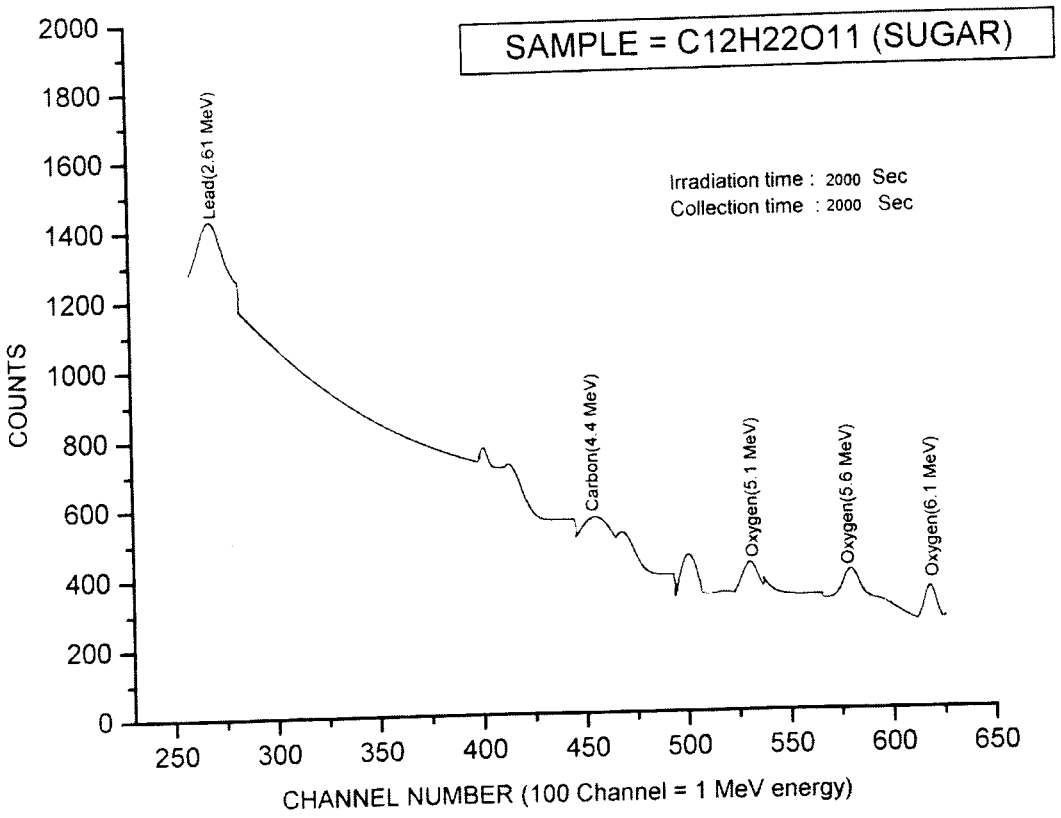


Figure-6.5: A typical recorded spectra of the prompt gamma-rays emitted from a chemical compound, sugar, during neutron irradiation.

Table-6.4: Concentration and weight percentage of the chemical composition.

Chemical Composition	Element present	Calculated Weight%	Measured Weight%	Measured Concentration (No. of atoms) $x10^{24}$
Urea	Carbon	19.99	22	2.2
	Nitrogen	46.64	42	3.6
	Oxygen	26.64	31	2.33
Sugar	Carbon	42.1	39	3.91
	Oxygen	51.41	57	4.29

6.7 Conclusion

The objective of the present study was to carry out a feasibility study to detect explosive class materials using prompt gamma-ray analysis technique. The results of this study indicate that 14 MeV neutrons with flux ~10^7 n/cm^2-s cab be used to detect elements such as carbon, nitrogen and oxygen in a sample of weight ~200 grams. These elements are presents in illicit drugs and explosives. It is therefore possible to detect hidden illicit drugs and explosives carried in passenger luggage at public places. A portable 14 MeV neutron generator can be made for this purpose. This facility will be also useful for military applications. Such facility can be used as a stand-alone monitor or can be utilized in conjection with an X-ray inspection system at places like airport, railway station, etc.

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