

MEASUREMENTS OF LOW-LEVEL RADIOACTIVITY BY GAMMA-RAY SPECTROMETRY

Heena

Lecturer in Physics, Dev Samaj College for women, Ferozepur City (India)

ABSTRACT

A low energy germanium detector (LEGe) detector shielded in a Pb-Sn housing has been established to measure the natural radioactivity in the various kinds of samples through direct γ -ray emission. Various aspects related to pulse shape processing in a gamma-ray spectrometer have also been discussed. The present work deals with experimental measurements of natural radioactivity present in di-ammonium phosphate (DAP) fertilizers, flyash samples and soil samples. DAP fertilizer samples were collected from different cities of Punjab, flyash sample was collected from Guru Nanak Dev Thermal Power Plant (GNDTP), Bathinda and soil samples were collected from Chandigarh and Ferozepur. Uranium content in all samples was determined from the intensity of the 63.3 keV γ -ray peak from ^{234}Th produced from the decay of ^{238}U . The fertilizer, flyash and the soil samples show the uranium content ranging 85-145 ppm, ≤ 17 ppm and ≤ 10 ppm, respectively.

Keywords- Bq, Ci, DAP, FWHM, LEGe

I. INTRODUCTION

Radioactive decay is the process by which a nucleus of an unstable atom loses energy by emitting alpha particles, beta particles, gamma rays and in rare cases there is neutron emission. Another kind of decay, which is called spontaneous fission, happens when a large unstable nucleus spontaneously splits into two (and occasionally three) smaller daughter nuclei, and generally immediately emits gamma rays, neutrons, or other particles as a consequence. Historically, radioactivity has been given in curie (Ci) units, defined as 3.7×10^{10} disintegrations/s. But now Becquerel (Bq), defined as one disintegration/s has become the standard unit of radioactivity. However, these units indicate nothing much about the amount of damage being done to human body. The amount of radiation (dose) that human body cells absorb is measured in grays (Gy). One gray is one Joule of energy absorbed per kg of body weight. To estimate the harm done to human body, ionisation capacity of the radiation needs to be taken into account. The α particles ionise very strongly, and cause 20 times more cell damage than the same dose of β particles, γ -rays or X-rays. The "dose equivalent" is measured in sieverts (Sv). A dose of 1 gray of β particles, γ -rays or X-rays will give a dose equivalent of 1 Sv. A dose of 1 gray of α particle will give a dose equivalent of 20 Sv.

Natural radioactivity is common in the rocks and soil that makes up our planet, in water and oceans, and in our building materials and homes. Radionuclides are found naturally in air, water and soil. Over 60 radionuclides (radioactive elements) can be found in nature, and they can be placed in three general categories:

- 1.1 Primordial - formed before the creation of the Earth
- 1.2 Cosmogenic - formed as a result of cosmic ray interactions
- 1.3 Human produced - enhanced or formed due to human action

1.1 Primordial radionuclides : Primordial radionuclides are left over from when the world and the universe were created. They are typically long lived, with half-lives often on the order of hundreds of millions of years. Radionuclides that exist for more than 30 half-lives are not measurable. Some nuclides like ^{232}Th have several members of its decay chain : $^{232}\text{Th} \rightarrow ^{228}\text{Ra} \rightarrow ^{228}\text{Ac} \rightarrow ^{228}\text{Th} \rightarrow ^{224}\text{Ra} \rightarrow ^{220}\text{Rn} \rightarrow ^{216}\text{Po} \rightarrow ^{212}\text{Pb} \rightarrow ^{212}\text{Bi} \rightarrow ^{212}\text{Po} \rightarrow ^{208}\text{Pb}$ (stable). The decay products are also in this category. Some other primordial radionuclides are ^{50}V , ^{87}Rb , ^{113}Cd , ^{115}In , ^{123}Te , ^{138}La , ^{142}Ce , ^{144}Nd , ^{147}Sm , ^{152}Gd , ^{174}Hf , ^{176}Lu , ^{187}Re , ^{190}Pt , ^{192}Pt , ^{209}Bi .

1.2 Cosmogenic radionuclides: Cosmic rays originate as primary cosmic rays, which are those originally produced in various astrophysical processes primarily outside of our solar system. Of primary cosmic rays, which originate outside of Earth's atmosphere, about 99% are the nuclei (stripped of their electron) of well-known atoms, and about 1% are solitary electrons (similar to beta particles). Of the nuclei, about 90% are simple protons, *i.e.*, hydrogen nuclei, 9% are alpha particles, and 1% are the nuclei of heavier elements [1]. Secondary cosmic rays, caused by a decay of primary cosmic rays as they interact with Earth's atmosphere, include neutrons, pions, positrons, and muons (Fig.1).

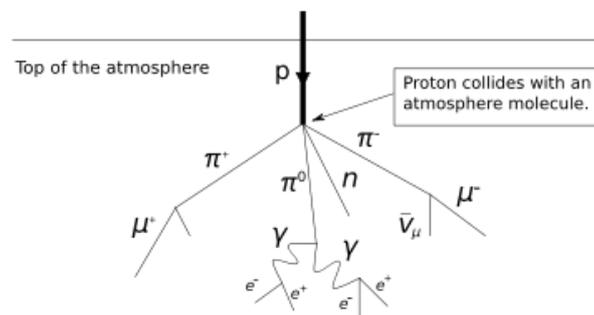


Fig.1: Production of secondary cosmic rays.

Cosmic rays are also responsible for the continuous production of a number of unstable isotopes in the Earth's atmosphere, such as ^{14}C . Cosmic rays kept the level of ^{14}C in the atmosphere roughly constant (70 tons) for at least the past 100,000 years, until the beginning of above-ground nuclear weapons testing in the early 1950's. This is an important fact used in radiocarbon dating used in archaeology. Measurements of many of these nuclides using ultra-sensitive techniques like Accelerator mass spectrometry (AMS) [2] have been exploited to determine geological dating.

1.3 Human produced radionuclides: Humans have used radioactivity for one hundred years, and through its use, added to the natural inventories. The amounts are small compared to the natural amounts discussed above, and due to the shorter half-lives of many of the nuclides [3]. The contributions have seen a marked decrease since the halting of above ground testing of nuclear weapons. A few human produced or enhanced nuclides are ^3H (12.3 y), ^{131}I (8.04 d), ^{129}I (1.57 x

10^7 y), ^{137}Cs (30.17 y), ^{90}Sr (28.78 y), ^{99}Tc (2.11×10^5 y) and ^{239}Pu (2.41×10^4 y). Most of these are produced in weapons testing and fission reactors. The present studies are mainly focused on the measurements of natural radioactivity in flyash from coal-fired thermal power plants, di-ammonium phosphate (DAP) fertilizer and soil samples. The samples collected from various locations were analyzed using low background germanium spectrometer due to higher efficiency of its detector.

II. LOW-LEVEL RADIOACTIVITY MEASUREMENT SYSTEM

The low-level radioactivity [4] is measured through the detection of α , β and γ emitted in the decay. The α and β particle detection is free from the problems of simultaneous detection of the background radiation. However, these measurements are discredited by large absorption corrections of α and β particles in the sample itself. Measurements involving γ -ray detection involve low and well-understood self-absorption corrections. The simultaneous detection of the background radiation poses limitation in these measurements.

A gamma spectrometry system consists of a Ge detector, associated electronics and data readout devices. The detector is often housed within a shield to reduce the background caused by sources other than the sample. The shield is constructed of a dense material (such as lead) that will absorb a large portion of background gamma rays. The shielding is usually crafted in such a way so as to minimize backscattering. The lead shielding material is usually graded with a two part thin metal shield such as tin and copper to reduce the effects of x-rays generated by the interaction of ambient photons with the lead. The sample is positioned within the shield at some distance from the detector. The distance will depend on a number of parameters, such as expected count rate and geometry of the sample container. As the counts accumulate, peaks develop that can be identified by energy and thus the nuclide identities from the spectrum are also identified. In general, the goal of the gamma spectroscopist is to derive nuclide-specific gamma emission rates of the sample from the spectral data.

In general, the spectral quality of a Ge detector will include:

- (a) Full Width at Half Maximum (FWHM) at the γ -ray energy of interest.
- (b) Peak to Compton ratio in the γ -ray energy region of interest and full energy peak efficiency.
- (c) Contribution from background sources

Despite the apparent simplicity with which gamma-ray measurements are made (little sample preparation is required), there are a number of correction factors to the raw counting data that must be considered:

- (a) The loss of pulses due to pulse pile up (at high count rates).
- (b) Coincidence summing (both random and cascading).
- (c) The decay of the source during counting.
- (d) The decay of the source from some previous reference time.
- (e) Attenuation of photons as a result of interactions with the sample.
- (f) Emission rate (or yield) of the specific photon energy.

III. BACKGROUND IN GAMMA-RAY SPECTRA

The importance of various components of the background changes greatly with the circumstances. In gamma-ray detectors without shielding, the cosmic-ray component is normally dominant. When significant shielding is

provided, both the cosmic flux and the background caused by ambient sources of gamma rays are decreased, and radioactive contamination of structural and shielding materials around the detector becomes an important fraction of the remainder. 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 necessary to select a detector size that is not larger than necessary to give a reasonable counting efficiency for the samples to be counted. A statistically based selection criterion is to choose a detector size that maximizes the ratio of S^2/B , where S is counting rate due to source alone and B is the counting rate due to background.

3.1 Detection limit

The minimum amount of an element that must be present in a sample in order to give a count rate that is greater than the uncertainty in the background spectrum is known as detection limit. The minimum peak counts must be larger than or equal to two times the square root of the background counts under the photo peak, then the probability that our assumption is correct is 95% ; if more certainty is required say 99.7% then minimum peak counts must be larger than or equal to three times the square root of the background counts under the photo peaks the conventional [5] criterion of statistical detect ability i.e., $N_p \geq 3\sqrt{N_B}$ where N_p is the number of counts under the photo peak , N_B is the number of counts in the background under in an interval having a width equal to the full width equal to the full width at half maximum (FWHM) of the peak or equal to double the FWHM. Detection limit (minimum analyzable limit) depends on various factors such as energy and intensity of the incident photons, measuring time, solid angle of detector and characteristic of X-ray spectrometer. Detection limit can be improved by increasing the peak to background ratio. To improve the detection limit, these emitted particles are detected in coincidence which eliminates the background. The detection limit in γ -ray measurements is also limited due to high Compton background in the spectrum. This can be improved upon by using an anti-Compton cylindrical detector (shielded from the radioactive sample) consisting of high efficiency scintillation material, which surrounds the high-resolution Ge detector. Any γ -ray Compton-scattered from the Ge detector will also be detected in the scintillation detector. The γ -ray event detected both in the Ge and scintillation detector is rejected from the Ge detector spectrum using coincidence electronics modules, which considerably suppresses the Compton background in the recorded γ -ray spectrum.

3.2 Shielding Materials

Lead is soft, malleable and ductile metal and is mostly used for gamma ray detector shielding. Because of high density and large atomic number, lead is the most widely used material for the construction of detector shields. Thickness of just a few centimetres of lead will provide a large reduction in the background of typical gamma rays detectors. The photoelectric absorption cross section predominates up to gamma ray energies as high as 0.5 MeV and even relatively hard gamma rays from external background sources can be absorbed efficiently. Lead is not effective against all types of radiations [6]. It is not effective absorber of neutron radiations. Lead is reasonably effective in removing many of cosmic-ray components of the background. Lead can be easily casted into solid shapes, although some care must be taken in the casting process to avoid porosity or voids in solidified shields. Ordinary lead normally contains a significant amount of natural activity caused by low-level contaminants, and therefore lead that is either specially refined or reclaimed from very old sources is preferred in the construction of shields for low background applications. The presence of ^{210}Pb is a source of background

primarily through the characteristic lead X-rays and bremsstrahlung created by high energy beta particles emitted by its decay product ^{210}Pb . Steel is a common material used for gamma ray shielding and is often used in situations where the size or configuration of the shield would make its construction from lead alone too expensive. In such circumstances, an outer layer of steel with an inner lead lining is often an effective compromise. Concrete is also often used in construction of large volume shields, because of its low cost. However, its activity is relatively high due to ^{40}K , U [7,8] and fallout products included in its composition. It is therefore used as the outer constituent of a shield with its own activity shielded by an inner layer of steel, lead or other shielding material of lower activity.

IV. EXPERIMENTAL SETUP FOR GAMMA-RAY MEASUREMENTS

A Low energy Ge (LEGe) detector having the horizontal configuration has been used to measure the γ -rays emitted from the ^{238}U radionuclide (Fig. 2). A plastic cap was mounted on the head of the detector to avoid any damage to the Be window of the detector. The detector was placed in the concentric cylinder of lead with inner diameter of 8.76 cm and outer diameter of 16.8 cm to shield the detector against the background surrounding the environ. A data collection time of 60,000s was sufficient to reduce the statistical error. Background spectra were also collected for same period of time. The net count rate of each sample was deduced by subtracting the corresponding background counts. Energy calibration is accomplished by measuring the spectra of ^{152}Eu and ^{241}Am . It is important that sources consist of known energy peaks that encompass the entire energy region over which the spectrometer is to be used

In the present experiment, the detector was kept in a lead shielding to reduce the background signals. The material used for shielding was lead because of its high density and atomic number. The shielding was made by melting the bricks of lead. These bricks were put in a strong container and then it was melted by putting the container having lead on a burner. The melting point of lead is 327°C . Once melted, it was put in a round symmetrical and cylindrical container of iron at the centre of which, a solid wooden block of diameter slightly greater than the diameter of cylindrical detector was fixed. The melted lead was poured slowly so that no pores between the material are left. Then it is allowed to cool for some time. After that it was extracted from the container and the central box is drilled out using milling machine to give it hollow cylindrical shape and to smoothen its surface. Concentric cylindrical lead shielding having the dimensions, inner diameter 8.8 cm, outer diameter of 16.8 cm, height 14 cm, constitutes the total volume of 703 cm^3 . It was used to shield the detector against the background from the surrounding environment (Fig. 3).

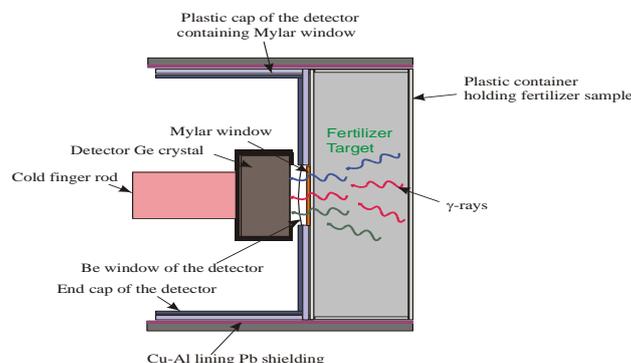


Fig.2: The experimental setup used for the detection of γ -rays emitted from the decay of radioisotopes present in DAP fertilizer, flyash and soils samples using LEGe detector



Fig. 3: Detector covered with lead shielding

4.1 General Characteristics of the detector setup

The “Low Energy Germanium detector” supplied by Canberra, US, (Model GUL0110P) consists of an N-type germanium crystal of 11.3 mm diameter with 100 mm² active area and 5 mm thickness. It is housed under vacuum in a stainless steel casing. The Be window is 0.008 mm thick and the crystal is situated at 5 mm from it. The detector is polarized under 500 V negative bias voltages. The nominative full width at half maximum (FWHM) is 338eV at 59.54keV using a 12 μ s amplifier shaping time constant (Fig. 4).

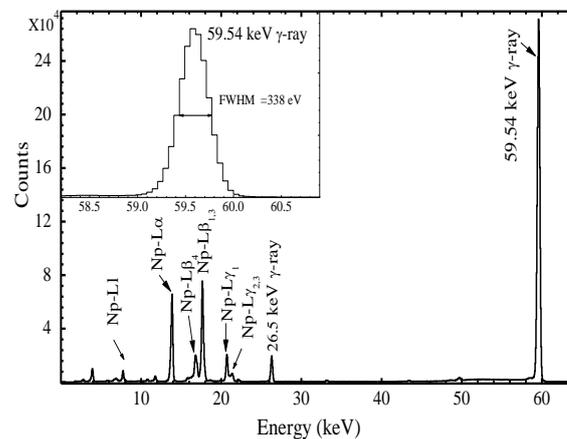


Fig.4. Direct spectrum taken using ²⁴¹Am annular source. Inset shows full width at half maximum (FWHM) is 338 eV at 59.54 keV.

4.2 Sample preparation for measurement of natural radioactivity

In order to measure the natural radioactivity in the DAP fertilizer, flyash and soil samples [9], all the samples were dried in oven at temperature 80°C to remove the moisture from the samples. These samples were ground with a pestle and mortar and the sieved through a mesh (1-2 mm size). A total of 90 g, 55.8 g and 92.6 g of DAP fertilizer, flyash and soil samples were weighed and then placed in plastic containers of uniform size. The plastic container (Fig. 2) was further covered with plastic wrap papers to eliminate the escape probability of decay product of Th and U, especially Rn isotopes. A time of 5 weeks was allowed after packing, which was sufficient time required to attain a state of secular radioactive equilibrium after their progeny [10-13]. A standard reference sample was also prepared by mixing uniformly 80 mg of uranyl acetate (80 mg) in 85 g of DAP sample for the efficiency calibration of detector. About 44.2 mg of uranium by weight was uniformly mixed in 85 mg of DAP. Similarly, a standard reference sample was also prepared by mixing uniformly 30 mg of uranyl acetate (30 mg) in 55.8 g of flyash sample for the efficiency calibration of detector. About 16.8 mg of uranium by weight was uniformly mixed in 55.8 g of flyash. In the similar way, the standard reference sample for soil was also prepared by mixing uniformly 21.16 mg of uranyl acetate (21.16 mg) in 92.6 g of soil sample for the

efficiency calibration of detector. About 11.88 mg of uranium by weight was uniformly mixed in 92.6 g of soil. All homogenized samples were kept identical to that of their respective standard reference sample regarding their geometrical shape, size and weight. The standard reference sample was also stored for 5 weeks to attain a stage of secular equilibrium.

V. RESULTS AND DISCUSSION

In the present work, the concentration of natural uranium present in DAP samples was determined from the direct gamma ray spectrum taken using LEGe detector for 20 hours. The decay schemes of the naturally occurring uranium are illustrated in (Fig. 5). The isotope of natural uranium (^{238}U) emits very weak gamma rays of 49.55 keV with an emission probability of 0.0064% and hence, the activity of natural U cannot be measured directly. However, the gamma ray emitted from the decay product of ^{238}U has been used to determine the concentrations of natural U present in the sample. In most environmental samples, the above presumption is valid only for the first member of the ^{238}U series, up to ^{226}Ra , ^{226}Ra decays into ^{222}Rn , an inert noble gas that does not form any chemical bonds and can escape into the atmosphere [14,15]. Fig. 6 shows the γ -ray background spectra taken without and with detector shielding. Typical γ -rays spectrum of all the samples along their reference standard sample is shown in Figs.7-9. The spectrum exhibit gamma ray peaks at 63.3, 92.6, 186.2, 238.6, 351.0 and 351.9 keV. The major peaks at 63.3 and 92.6 keV are due to decay of ^{234}Th into ^{234}Pa in ^{238}U decay series. Uranium content of the DAP samples was determined from intensity of 63.3 keV from ^{234}Th produced from decay of ^{238}U . The 92.6 keV gamma ray observed during the decay of ^{234}Th is the overlapped peak of two unresolved gamma rays with energy 92.3 keV and 92.8 keV and it also coincide with γ -rays from three natural decays. Hence, the information about activity and concentration of uranium in a sample containing high level of thorium cannot be predicted accurately. On the other hand, the 63.3 keV γ -ray peak is the result of contribution from 63.3 keV γ -ray emitted by deexciting ^{234}Th with emission probability of 3.9%, 63.9 keV γ -ray from ^{232}Th with emission probability of 0.255% and 63.9 keV γ -ray from ^{231}Th with emission probability of 0.023%. Hence, the contributions from ^{231}Th and ^{232}Th radionuclides can be neglected in the comparison with 63.3 keV γ -ray from ^{234}Th . The gamma ray of 63.3 keV is mostly used for the concentration measurements of ^{238}U in environmental samples than 92.6 keV, 766.4 keV and 1001.2 keV and used in the present measurements. The net area count after background corrections in each photo-peak was used in calculation of the activity concentration of each of the radionuclides using the expression

$$m_{\text{unknown}} = \frac{N_{\text{unknown}}}{N_{\text{known}}} m_{\text{known}} \quad (9)$$

where N_{unknown} is the count rate for γ -rays (63.3 keV or 92.6 keV) of ^{238}U which is related to unknown concentration (m_{known}) of ^{238}U in samples. N_{known} is the count rate for decay γ -rays (63.3 keV) of ^{238}U which is related to known concentration (m_{known}) in respective standard reference sample.

The elemental concentration of U in the DAP fertilizer, flyash and soil samples measured in the present work is given in Table 1. According to the United Nations Scientific Committee on the Effects of Atomic Radiation the normal concentration of uranium in soil is 300 $\mu\text{g}/\text{kg}$ to 11.7 mg/kg [16]. The concentration values of U in soil and flyash samples are found to be reasonable as compared to these values. Flyash produced at thermal power plants in Bathinda can be safely used for building materials like cement and bricks. In case of fertilizers it is recommended that the DAP fertilizers with the uranium extracted should be used in the water-logged areas in the Punjab state, so as to minimize

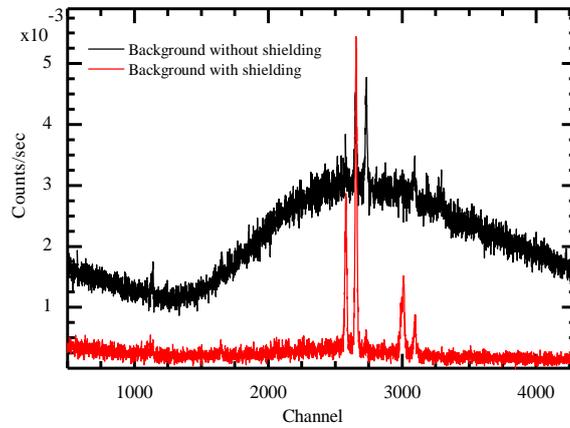


Fig. 6: Typical γ -rays background spectra without and with detector shielding.

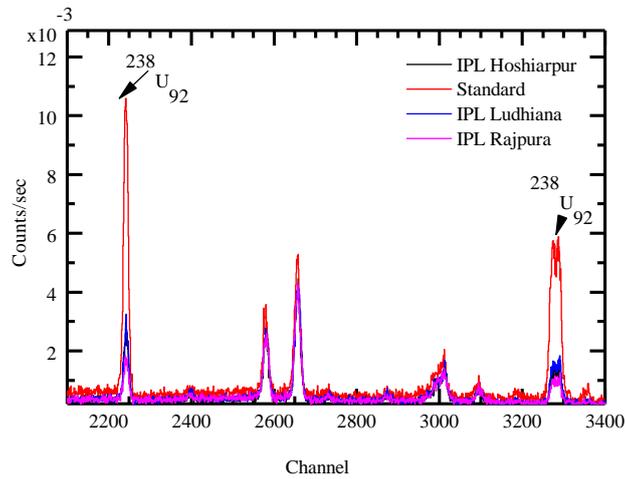


Fig. 7: Typical γ -rays spectra of prepared reference fertilizer sample and IPL samples.

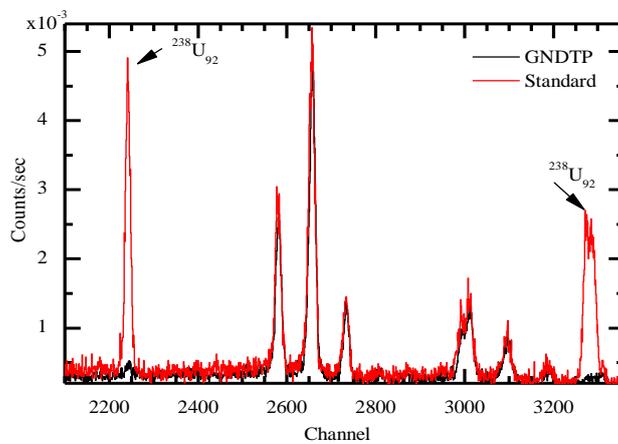


Fig. 8: Typical γ -rays spectra of prepared reference flyash and GNDTP flyash samples.

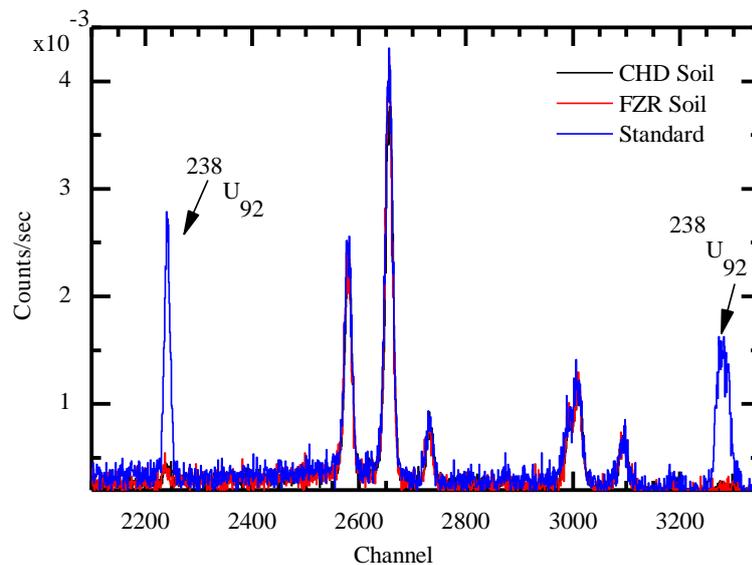


Fig. 9: Typical γ -rays spectra of reference soil sample and Chandigarh and Ferozpur soil samples.

VI. CONCLUSION

A low-background Gedetector based gamma ray spectrometer has been established to measure the natural radioactivity in the various kinds of samples. Various aspects related to pulse shape processing in the spectroscopy amplifier stage of a gamma-ray spectrometer have also been discussed. The natural uranium radioactivity measurements of DAP fertilizer samples, flyash sample collected from Guru Nanak Dev Thermal Power Plant (GNDTP) and soil samples collected from Chandigarh and Ferozpur city of Punjab, have been carried out. Uranium content in all samples was determined from the intensity of the 63.3 keV γ -ray peak from ^{234}Th produced from the decay of ^{238}U . The fertilizer, flyash and the soil samples show the uranium content ranging 85-145 ppm, ≤ 17 ppm and ≤ 10 ppm, respectively. The concentration values of U in soil and flyash samples are found to be reasonable as compared to the generally observed values in soils. Therefore, flyash produced at thermal power plants in Bathinda can be safely used for building materials like cement and bricks. It is recommended that the uranium extracted DAP fertilizers should be used in the water-logged areas in the Punjab state, so as to minimize ground water contamination.

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