Gamma Ray Spectrometry for Analysis of Radio Elements with Applications in Uranium Estimation

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***Abstract*— Radiometric techniques have acquired immense importance among all the analytical analysis techniques. They have the edge over other analytical techniques due to the advantages of being adequately accurate, rapid and cost effective with minimum need on sample preparation. Among the various radiometric techniques, Gamma ray spectrometry is one of the most widely applied techniques for identification and quantitative estimation of the radio elements in a variety of matrices. The wide range of applications of gamma ray spectrometry in the field of Radiation monitoring in nuclear facilities, National Security, Geochemical investigation, Health physics, Nuclear medicine, Material research and mineral exploration have made the technique globally popular. The underlying principle is that the energy of gamma photons emitted from radio isotopes is characteristic of individual isotopes. This requires appropriate radiation detector, pulse processing, sorting of pulses with amplitude, display of pulse distribution and interpretation, all included in a single set-up called as gamma ray spectrometer. The technique began with the use of single channel analyzers for sorting pulses and has evolved with the advancement in electronics and detector technology. Today portable gamma ray spectrometers which are very handy to carry for field use with powerful software to directly display the spectrum and also to analyze radio isotopes are quite common. The technique was applied for estimation of Uranium, Thorium and Potassium concentration in the soil and rock samples collected from the area around Mody University, Lakshmangarh.**

I. INTRODUCTION

Naturally occurring radioactive elements are K^{40}, U^{238} and Th^{232}. Availability of uranium is more than many other metals in the soil. Uranium is a radioactive element, only its radioactivity helps in its discovery. Uranium is abundant in many types of rocks and it is low in water and soil. Uranium is used in the production of nuclear power in the nuclear reactor and in the defense sector, and radiation is used in agriculture, medicine and various industries. Due to being radioactive, alpha (α), beta (β) and γ (γ) radiations are emitted from it. The presence of these rays is detected by its measurement, which is used to measure different types of techniques. Radiometric techniques have acquired immense importance among all the analytical analysis techniques. They have the edge over other analytical techniques due to the advantages of being adequately accurate, rapid and cost effective with minimum need on sample preparation. Among the various radiometric techniques, Gamma ray spectrometry is one of the most widely applied technique for identification and quantitative estimation of the radio elements in a variety of matrices. the technique was applied for estimation of Uranium, Thorium and Potassium concentration in the soil and rock samples collected from the area around Mody University, Lakshmangarh.

II. SODIUM IODIDE SCINTILLATION DETECTOR

The NaI(Tl) scintillation detector consists of a thallium activated sodium iodide crystal called scintillator and an electronic device called photo-multiplier tube. A part of the gamma ray energy is absorbed in the crystal as gamma (γ) rays pass through this crystal resulting in scintillations of intensity proportional to the energy of the incoming radiation. The scintillations are incident on the photo-cathode of the photo multiplier tube and eject photoelectrons. There will be multiplication of the photoelectrons as electrons are accelerated to dynodes present between cathode and anode maintained at progressively high voltage as higher the voltage, the greater the multiplication resulting to the greater pulse size formed at the anode usually in the order of tens of mill volts. A linear or spectroscopic amplifier is required for proper operation of the counter operating in the range of 0 to 10 volts with size of the output pulse proportional to the input pulse size.

III. GAMMA SPECTROMETRY

Most radioactive sources produce gamma rays of various energies and intensities. The energy emitted is characteristic of individual isotopes and isotope identification is possible simply by measuring the energy. A set-up which can focus on individual emitted energies is called the gamma ray spectrometer. A gamma ray spectrum can be obtained when these emissions are detected and analyzed with a spectroscopy system which is performed through the process of counting and measuring the energy of individual photons emitted from elements.

IV. EXPERIMENTAL APPARATUS

The spectrometry system consisted of a 5 inch x 4 inch NaI(Tl) crystal integrated with the photomultiplier tube, pre- amplifier, high voltage unit, linear spectrometric amplifier, analog to digital converter, multichannel analyzer and display unit.

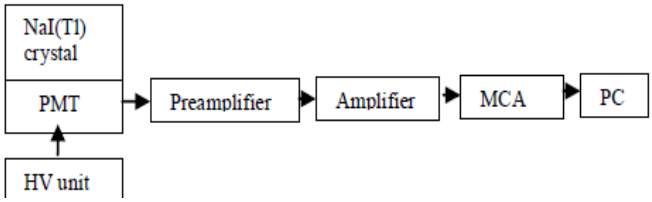


Figure 1. Block diagram of Spectrometry assembly

The output pulse from the PMT enters the preamplifier. The preamplifier provides a match between the high impedance of the detector and the low impedance of coaxial cables to the amplifier after collecting the charge from the PMT output. The amplifier shapes and amplifies the pulse increasing the pulse height and eliminating electronic noise problems. The multichannel analyzer (MCA) consists of an analog-to-digital converter (ADC), control logic, memory and display unit. The analog-to-digital conversion module (ADC) converts the analog information from the pulse train into a digital format which is then reshaped into a Gaussian shape. A computer receives the output of multichannel analyzer where data is stored, displayed and analyzed. Different software types are available by manufacturers including spectrum analysis tools such as energy calibration, peak area, net area calculation and resolution calculation.

V. Energy Caliberation

Energy calibration is the process to establish the relationship between energy and channel. For acquiring the linearity the values of A and B should be near to zero in the equation $y= Ax^3 + Bx^2 + Cx + D$ where A, B, C and D are the coefficients, x is channel and y is the energy. Acquiring the spectrum by the use of 662KeV energy Cs137 source and Co60 having three energies as 1175KeV, 1330KeV and 2508KeV for 500seconds provide peaks at channel 331 for KeV, channel 576 for 1175.3 KeV, channel 652 for 1330.6 KeV and channel 1228 for 2508.2 KeV. As we have known energies for four channels the values of the coefficients calculated are: A= 2.6×10^{-7}, B = -6.4×10^{-4}, C= 2.5 and D= -1.2×10^{2} .

VI. Regions Of Interest

Using known standards of K, U and Th a total four regions of interests (ROIs) were chosen among which first region of 200KeV centered at 1460KeV energy for K, second region also of 200KeV with centre at 1760KeV for U, third region of 400KeV with 2620KeV energy as centre for Th and fourth region from 400KeV to 3MeV for gross gamma measurement.

VII. Resolution Determination

Spectrum for Cs^{137} source with energy 662KeV was acquired. With the peak energy of 662 KeV, the photo peak was analyzed using the PHA software. In the given set-up, using NaI(Tl) detector full width half maximum (FWHM) was determined to be 52KeV. Hence, R = 52/662 = 7.9 %.

VIII. Chi-square Test

Chi-Square Test is used to determine whether there is a significant difference between expected frequencies and observed frequencies in one or more categories.

To check for any malfunctioning in the experimental set-up following steps are followed:

- Take 30 counts reading with a standard.
- Calculate χ^2 value using above mentioned formula.
- See for degree of freedom (n-1).

TABLE I. URANIUM COUNTS IN URANIUM CHANNEL

Sr. No.	Counts (x)	$(x-x')$	$(x-x')^2$
1	9786	-65.97	4352.0409
2	9981	129.03	16648.7409
3	9805	-46.97	2206.1809
4	9725	-126.97	16121.3809
5	9952	100.03	10006.0009
6	9815	-36.97	1366.7809
7	9827	-24.97	623.5009
8	9881	29.03	842.7409
9	9721	-130.97	17153.1409
10	9709	-142.97	20440.4209
11	9928	76.03	5780.5609
12	9972	120.03	14407.2009
13	9842	-9.97	99.4009
14	9951	99.03	9806.9409
15	9795	-56.97	3245.5809
16	9920	68.03	4628.0809
17	9816	-35.97	1293.8409
18	9871	19.03	362.1409
19	9805	-46.97	2206.1809
20	9945	93.03	8654.5809
21	10085	233.03	54302.9809
22	9924	72.03	5188.3209
23	9819	-32.97	1087.0209
24	9754	-97.97	9598.1209
25	9777	-74.97	5620.5009
26	9772	-79.97	6395.2009
27	9827	-24.97	623.5009
28	9882	30.03	901.8009
29	9874	22.03	485.3209
30	9798	-53.97	2912.7609

Where, Mean= $x' = 9851.97$ and $\Sigma(x-x')^2 = 227360.97$.

Hence, $\chi^2 = \frac{\Sigma(x-x')^2}{x'} = 23.08$

P value at 23.08 with 29 as d.f = 0.77 which is acceptable.

IX. Background Counting

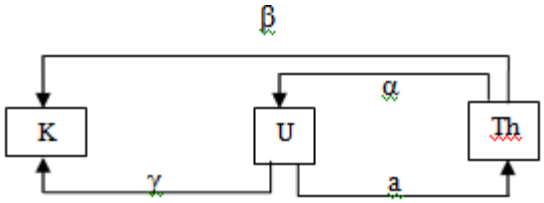
The spectrum in the absence of any source activity was acquired to identify the background radiations as some radiations are present everywhere. The background radiations then were subtracted from the actual measurements. The counts in all the four windows were determined.

X. Sensitivity Caliberation

The spectrum was acquired for uranium, thorium and potassium using standards of known concentrations. After subtracting the background counts in the respective channels, sensitivity factors were determined for uranium, thorium, potassium and total activity represented as eU3O8. Known standard for uranium consisted of uranium ore in equilibrium with the concentration of 1100ppm U3O8, for thorium 4400ppm of ThO2 in monazite ore and 26% of potassium in potassium dichromate salt.

XI. Determination of Stripping Factors

The stripping factors were calculated as α =0.35, β =0.50, γ =0.90 and a=0.04.



$$\alpha = \frac{\text{Net counts in U channel due to Thorium standard}}{\text{Net counts in Th channel due to Thorium standard}}$$

$$\beta = \frac{\text{Net counts in K channel due to Thorium standard}}{\text{Net counts in Th channel due to Thorium standard}}$$

$$\gamma = \frac{\text{Net counts in K channel due to Uranium standard}}{\text{Net counts in U channel due to Uranium standard}}$$

$$a = \frac{\text{Net counts in Th channel due to Uranium standard}}{\text{Net counts in U channel due to Uranium standard}}$$

XII. Spectrum for Samples

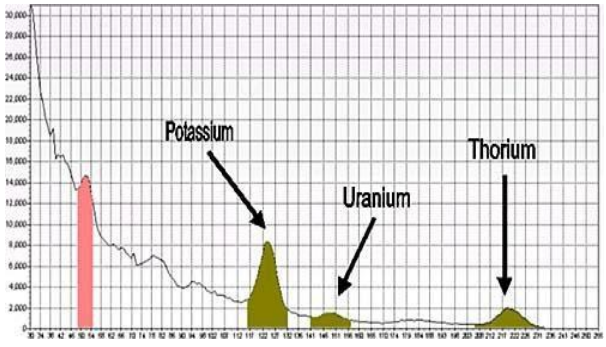


Figure 2. Sample spectrum

This spectrum has peaks corresponding to the presence of potassium, thorium and uranium. The heights of the peaks related to the number of gamma rays recorded (in counts per second), and the horizontal axis relates to the energy of the gamma rays (high energy at left and low energy at right). Concentrations of uranium, thorium and potassium are calculated using formulas as mentioned below:

$$\text{Uranium equivalent in ppm} = \frac{N_2^{\,U}}{S_U} = N_2 - \alpha N_3/S_U(1-\alpha a)$$

$$ThO_2\,(\text{ppm}) = \frac{N_3^{\,Th}}{S_{Th}} = N_3 - aN_2/S_{Th}(1-a\alpha)$$

$$\%K = \frac{N_1^{\,K}}{S_K} = [(N_1(1-a\alpha) - N_2(\gamma - a\beta) - N_3(\beta - \gamma\alpha))/S_K * (1-a\alpha)]$$

Where N_1, N_2, N_3 are net counts in potassium, uranium and thorium channel; S_K, S_U, S_{Th} are sensitivity constants for potassium, uranium and thorium.

TABLE II. URANIUM,THORIUM AND POTASSIUM CONTENT IN SOIL SAMPLES COLLECTED FROM MODY UNIVERSITY, LAKSHMANGARH:

Sample No.	Sample Name	U_3O_8 (Raeq) (ppm)	ThO_2 (ppm)	K (%)	eU_3O_8 (gross) (ppm)
S1	MT/MODY/W-1/2017	3	14	1.4	11
S2	MT/MODY/W-2/2017	3	13	1.4	11
S3	MT/MODY/W-3/2017	2	10	1.2	10
S4	MT/MODY/E-1/2017	2	11	1.4	11
S5	MT/MODY/E-2/2017	2	13	1.5	12
S6	MT/MODY/E-3/2017	2	10	1.4	11
S7	MT/MODY/S-1/2017	3	11	1.3	11
S8	MT/MODY/S-2/2017	3	12	1.3	12
S9	MT/MODY/S-3/2017	3	14	1.5	13
S10	MT/MODY/N-1/2017	2	11	1.5	10
S11	MT/MODY/N-2/2017	3	11	1.4	11
S12	MT/MODY/N-3/2017	3	12	1.6	12

TABLE III. URANIUM THORIUM AND POTASSIUM CONTENT IN ROCK SAMPLES COLLECTED FROM LAKSHMANGARH FORT:

Sample No.	Sample Name	U_3O_8 (Ra eq) (ppm)	ThO_2 (ppm)	K (%)	eU_3O_8 (gross) (ppm)
S13	MT/MODY/F-1/2017	5	30	3.8	28
S14	MT/MODY/F-2/2017	8	39	4.4	36
S15	MT/MODY/F-3/2017	5	53	4.3	39

XIII. Result and Interpretation

The soil samples from S1 to S12 collected from the area

outside the Mody University campus indicate Uranium concentration ranging between 2 to 4 ppm assuming that the Uranium is in secular equilibrium with its daughters, Thorium concentration between 10 to 14 ppm and Potassium concentration between 1.2 to 1.6 %. The average content of uranium, thorium and potassium is found to be 2.6ppm, 11.8ppm and 1.4%. The average Thorium content is slightly higher than the world's average content of thorium which is about 7ppm. These concentrations of the radio elements may vary depending upon the rock types from which the soil has been derived.

The total Equivalent Uranium concentration in the soil around University campus is measured to be between 10 to 13 ppm

The sample number S13 to S15 were collected from three different locations at Lakshmangarh Fort which indicated relatively higher radiation field measured through radiation survey meter. The uranium concentration in these samples varies between 5 to 8 ppm, Thorium concentration between 30 to 53 ppm and Potassium concentration between 3.8 to 4.3 %.

XIV. Conclusion

In the experimental work, application of Gamma Spectrometry in estimation of naturally occurring radio elements has been illustrated by analyzing 12 samples from the area around the Mody university campus, Lakshmangarh, Dist Sikar Rajasthan. The results of these samples indicate an average equivalent Uranium concentration in the soil around the Mody University campus is 11.25ppm eU3O8, with average Uranium, Thorium and Potassium concentrations of 2.6ppm, 11.8ppm and 1.4% respectively. This level of concentration does not pose any health hazard to the residents. The estimation of naturally occurring radio nuclides in the geological rock and soil samples is one of the measure applications of the gamma ray spectroscopy. Other application includes the identification and estimation of a wide range of other radio nuclides at the same time.

XV. Future scope of work

Gamma ray spectrometric technique is very helpful in a variety of spheres dealing in identification and estimation of radio elements. The present work illustrates its application in estimation of naturally occurring radio elements and has a wide scope for taking up similar work on a wider scale. It is possible to generate a distribution map of radio elements in the area and identification of locations with anomalous concentrations. Another important application of this distribution can be to generate an approximate map of the radiation levels in the area through use of standard dose conversion factors. In addition to the above, technique of Gamma ray spectrometry as studied above can also be employed for various applications in the field of medicine, agriculture and Research and Development.

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