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Non-destructive aerial and terrestrial analysis of uranium isotopic activity by sensitive γ -spectrometer for El-Sella site

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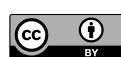
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Abstract. Any comprehensive radioactivity surveillance for a specific site should be carried out by a sensitive airborne system (e.g. carried on aircraft) together with terrestrial sampling and analysis. In the framework of ore, environmental and nuclear materials monitoring and investigation, the activity and isotopic compositions signature are considered crucial factors that must be considered, determined and evaluated. In this work and based on previous aerial monitoring, a very sensitive hyper pure germanium detector (Hp-Ge) of 50% efficiency with the Genie2000 software has been used for spectroscopic non-destructive assay (NDA) of radioactivity content for samples collected from the Egyptian eastern desert (El-Sella Site). The identification of specific signature isotopes through their characteristic gamma lines and a calculation of their specific activities, activity ratios of $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ and mathematically estimation of the natural enrichment percentage are very vital and nuclear forensic targets. The results of measurements, analysis and calculations show that the Site samples have high radioactivity, natural enrichment origin and high uranium concentration. The obtained results are given tabulated, depicted, discussed and compared with the recent nationally published work and the international levels and limits.

1. Introduction

Environmental sampling (ES) is considered a very important tool and one of the powerful technical verification measures presented for the International Atomic Energy Agency (IAEA) safeguards. Many safeguard verification measures aim to confirm the amount and type of nuclear material declared by a state, ES gives a strong means for identifying undeclared activities and nuclear material and helps the IAEA in drawing reliable conclusions [1]. The most common and ideal method for measurements is the non-destructive assay because of the importance of not losing the radioactive and nuclear material needed to the analysis itself. This technique is used by the nuclear industry, defense facilities, and safeguard inspectors to make rapid accurate measurements of sensitive nuclear materials [2].

The first analysis performed after receiving the material is the identification of the radioisotopes in the material, the risk assessment, uranium isotopic abundance, activity and enrichment. This kind of analysis performed by a high-resolution gamma spectrometer (HRGS); mostly through using high pure germanium (Hp-Ge) detector [3] which was used to determine the characteristics of nuclear and radioactive materials and identify its radioisotopes from its gamma lines[2]. Energy and efficiency calibration were performed at the detector to ensure accurate results. Additionally, from the uranium isotopes activity, the enrichment can be determined. It is well known that gamma rays emitted from the



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radioactive material have specific energies and intensities that differ from one material to another according to the radioisotopes that exist in the material and its activities[2]. ^{238}U can be identified from its daughter energy lines ^{234}Th (63.29 keV) and ^{234}Pa (1001.03 and 766.36 keV) and ^{235}U can be identified from its own gamma energy lines (143.7, 163.3, 185.7 and 205.3 keV) [4].

The enrichment of uranium in the environmental samples should be natural, 0.72%, except in locations near uranium enrichment facilities as the uranium separation technologies mostly use UF_6 gas. This leads to trace amounts of enriched uranium being released to the environment from those facilities[5]. The concentration of uranium varies according to the location where it is collected and the type of material that it is mixed with. For example, the uranium concentration approximately 4 parts of uranium per million when uranium mixed with granite which covers 60% of the earth crust [6]. High-grade uranium contains about 200,000 ppm ($\mu\text{g/g}$) of uranium, while low-grade uranium contains about 1000 ppm U. There are slightly uranium-rich locations in Egypt like Gattar mountain which contains uranium in the range from 1000 to 5000 ppm [7].

This work focuses on the analysis of high radioactive samples by performing a set of measurements to calculate specific activity, natural enrichment verification and uranium isotopic composition samples of the El-Sella site in the South Eastern desert.

2. Experimental Work

2.1. Sample collection and preparation

The sample was collected from the Egyptian eastern desert, the El-Sella area. The sample was prepared for measurements in the nuclear engineering department laboratory at the Military Technical College through a set of sequential steps. The sample was grounded, crushed, homogenized and sieved until passing through mesh number (200). Then the sample was dried by oven at 105 °C for 1 day and then was weighed to be 323 g to be the same as the reference material used for calibration. The sample was then sealed in a cylindrical plastic container for 28 days to attain secular equilibrium[8]. This step is very important to ensure that all the uranium daughters remain in the sample as the radon daughter Rn-222 is in a gaseous form so it may escape from the sample unless it is tightly sealed.

2.2. Gamma spectrometer setup

The non-destructive technique used in this work was based on using hyper pure germanium (Hp-Ge) detector that has a 50% efficiency and a resolution of 1.95 keV at 1332 keV of ^{60}Co source with its associated electronics such as the spectroscopic amplifier and high voltage supply. The detector high voltage (H.V.) is 3.3 kV is connected to a Canberra DSA-1000 multichannel analyzer (MCA). The Hp-Ge crystal was fully immersed in liquid nitrogen for cooling (-200 °C) to reduce noise and leakage current. The Hp-Ge detector was shielded by a 10 cm thick lead material to avoid background radioactivity. The system used for identifying the radioactive nuclides through their gamma energies and calculation of their activities by analyzing the spectrum obtained from the detector was the Genie-2000 software [4].

2.3. Gamma Spectrometer Energy Calibration

The energy calibration was performed by using standard IAEA sources (point sources) model RSS8 and MGS-1 which contain certain radioisotopes (^{133}Ba - ^{137}Cs - ^{60}Co - ^{22}Na - ^{152}Eu) which have known activities (Table 1).

Table 1. Radionuclides of standard sources used for energy calibration

Model	Radioisotope	Energy (keV)	T _{1/2} (day)	I _γ	Activity (Bq)	Reference date
RSS-8	Ba-133	81		0.342		
		276		0.071		
		303	3942	0.184	37000	
		356		0.622		
	Cs-137	384		0.089		
		662	11023	0.852	37000	
	Co-60	1173		0.999		
		1333	1923.55	1	37000	1/10/2014
	Na-22	1275	949	0.995	37000	
MGS-1	Eu-152	121.78		0.284		
		344.3		0.266		
		1408.01	4927.5	0.208		
		964.1		0.145	37000	
		1112.07		0.136		
		778.9		0.13		

The reference sources were counted for 1800 seconds in the (Hp-Ge) detector to obtain proper results[9]. With known energy readings of each radioisotope, a relationship can be obtained between the channel number and the energy which can be shown as follows in equation 2.1:

$$\text{Energy} = 0.1355 + 0.1758 \times \text{Channel number} \quad (2.1)$$

2.4. Gamma Spectrometer Efficiency Calibration

Efficiency calibration is considered to be an essential factor to attain accurate and valid measurements. The standard sources used for efficiency calibration should have the same geometry and density and should be measured at the same conditions as the unknown samples. The efficiency calibration was performed by using standard IAEA sources; **S-14** (²³⁴U, ²³⁵U, ²³⁸U) and **S-16** (Am-214, Cs-137, Co-60) for uranium samples which can be found in table 2.

Table 2. Radionuclides of standard sources used for efficiency calibration

Model	Radioisotope	Energy (keV)	T _{1/2} (day)	I _γ	Activity (Bq)	Reference date
S-14	U-235	143.1		0.105	588.1	
		163	2.57E+11	0.047	588.1	
		185.7		0.53	588.1	
	U-238	205		0.047	588.1	06/09/2010
		63	1.63E+12	0.038	12780	
		1001		0.007	12780	
	U-234	53		0.001	12520	
		120	8.97E+7	0.00034	12520	
S-16	Am-241	59.5	158000	0.357	5518.1	
	Cs-137	661.7	10980	0.852	2140.8	01/04/2011
	Co-60	1173.2	1925	0.999	3490.9	
		1332.5		1	3490.9	

The standard sources were counted in the detector for 86400 sec (24 hr) [10]. The absolute efficiency can be calculated by using the following equations.

$$\varepsilon_{abs} = \frac{C}{A_s I_\gamma T} \quad (2.2)$$

Where ϵ_{abs} is the absolute efficiency, C is the net counts of photopeak for a certain isotope in the reference material, A_s is the known specific activity for each isotope in the reference material, I_γ is the branching ratio, and T is the counting time. The two IAEA standard sources were used to cover a range of low and high gamma energies; usually single efficiency calibration is carried out for all energies but for this work, the calibration was divided into two categories, low and high, according to the energy. The low energy efficiency calibration was used for energies from 59.5 to 120.9 keV whilst the high energy efficiency calibration was used for energies higher than 120.9 keV. Figure 1 shows the relationship between efficiency and energy for the low energies and can be applied in equation 2.3 to calculate the efficiency by the known energy. Moreover, since Figure 2 shows the relationship between efficiency and energy for the high energies, the results can be applied in equation 2.4. This method is used to reduce the error in the measurements as much as possible.

$$Eff = -0.1787 + 0.1934 \cdot \ln E \quad (2.3)$$

$$Eff = 0.0191 - \frac{7.425}{E} + \frac{3352.26}{E^2} + \frac{236871.9}{E^3} \quad (2.4)$$

Where Eff is the absolute efficiency of the detector and E is the energy for each radioisotope in (keV)

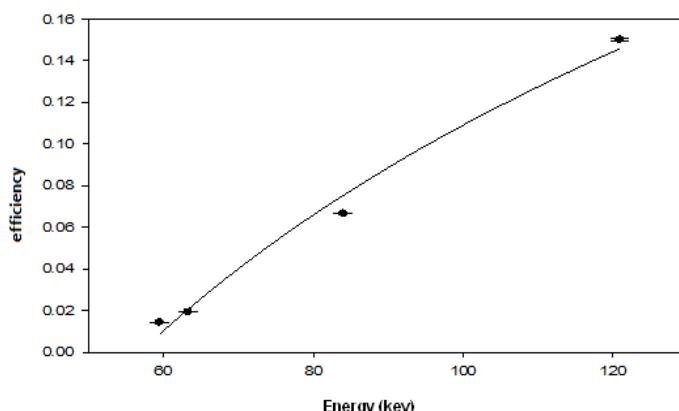


Figure 1. Efficiency calibration of the system for low energies

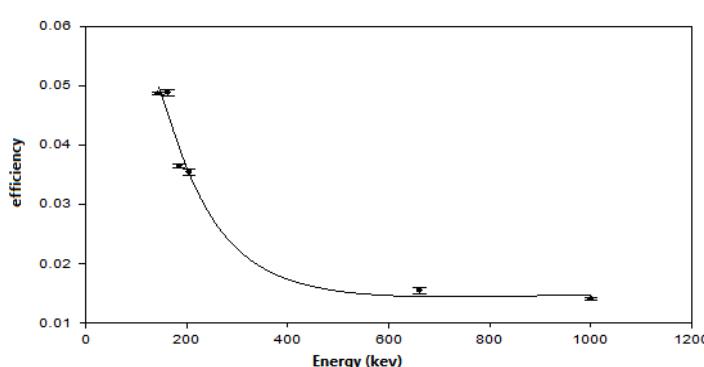


Figure 2. Efficiency calibration of the system for high energies

2.5. Spectrum analysis and activity calculation

Each uranium isotope emits gamma rays which have certain energies. These gamma rays when interacting with the detector are converted into signals and are processed to be shown as a spectrum.

Each peak in the spectrum indicates the gamma energy line and each energy line indicates the presence of a certain radioisotope. The sample was measured for 86400 sec. The photo-peaks which have high intensities were obvious in the spectrum which can be seen in Figure 3.

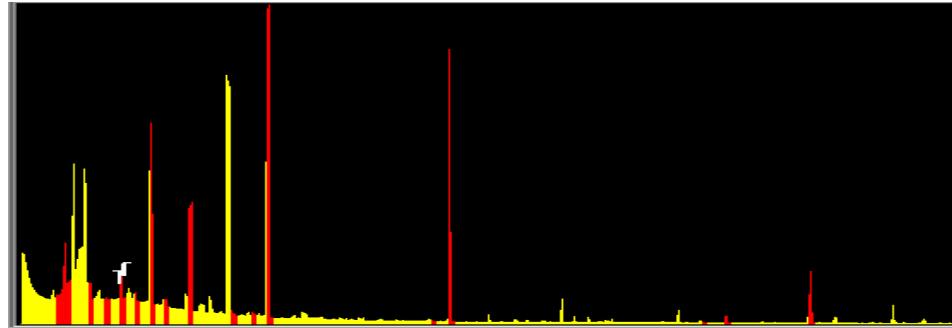


Figure 3. Typical spectrum for El-Sella sample using Genie2000 software

The activity can be calculated by estimating the counts from the radioactive sample spectrum which is counted three times to estimate the average and the standard deviation. Each line in the spectrum has a certain energy, efficiency and branching ratio. The uranium isotope ^{238}U itself is not emitting gamma lines so it can only be identified by the energy lines from its daughters, ^{234}Th (63.29 keV) and ^{234}Pa (1001.03 and 766.36 keV) which are considered to have the same activity of ^{238}U . The ^{235}U isotope can be identified by its energy lines (143.7, 163.3, 185.7 and 205.3 keV). On the other hand, ^{234}U can be identified from its two energy lines (53 and 120.9 keV). The sample activity can be calculated using the following equation:

$$A = \frac{C}{\varepsilon_{abs} \cdot I_\gamma T \cdot m} \quad (2.5)$$

Where, A is the specific activity of the radioisotope (in Bq/kg). C is the net count of the photo_peak after subtraction of the background counts. ε_{abs} is the absolute efficiency of the detector, I_γ is the branching ratio, m is the mass of the sample and T is the time of counting (sec).

3. Result and Discussion

3.1. Specific Activity and Activity Ratio results

The results of specific activity obtained by using equation (2.5) for each isotope were summarized in Table 3. It was found that the average value of specific activity for the ^{235}U isotope was 1268.675 ± 98 Bq/kg as the values of energy lines (143.7, 163.36, 185.72 and 205.23 keV) were 1167.4, 1353.3, 1201.2 and 1352.8 Bq/kg respectively. As for ^{238}U , the average specific activity was 27186.25 ± 376 Bq/kg as the values of energy lines (63.29 and 1001 keV) of daughters (^{234}Th and ^{234m}Pa) were 26919.77 and 27452.74 Bq/kg respectively. The average specific activity of ^{234}U was 22067.78 ± 82 Bq/kg as its two energy lines (53.24 and 120.9 keV) specific activity were 22067.78 and 22125.81 Bq/kg respectively. The activity ratio of $^{235}\text{U}/^{238}\text{U}$ was also calculated and found to be in line with the natural reference value (0.046) [4]. The remarkable result was the $^{234}\text{U}/^{238}\text{U}$ activity ratio as its value was 0.81, smaller than unity <1, which means that there is disequilibrium between the parent and daughter in the decay series. The reason for this is that a disturbance occurred due to uranium migration probably by the groundwater interaction with rocks [11]. The result of specific activity and activity ratios were then compared with another work at the same location (El-Sela) and the results were in a comparable range. The previous

work found that the specific activity for ^{238}U ranged from 500 to 80065 Bq/kg, for ^{235}U ranged from 10 to 3557 Bq/kg and the $^{234}\text{U}/^{238}\text{U}$ activity ratio ranged from 0.6 to 0.92 [12].

3.2. The Natural Enrichment Verification Results

The enrichment percentage of the sample can be estimated by the following atom ratio equation:

$$\%E = \left(\frac{N_{^{235}\text{U}}}{N_{^{234}\text{U}} + N_{^{235}\text{U}} + N_{^{238}\text{U}}} \right) \times 100 \quad (3.1)$$

Where, $N_{^{235}\text{U}}$, $N_{^{234}\text{U}}$ and $N_{^{238}\text{U}}$ are the number of radionuclide of ^{235}U , ^{234}U and ^{238}U respectively, which can be estimated from the decay constant and the activity by using the decay equations as follow:

$$N_{^{235}\text{U}} = \frac{A_{^{235}\text{U}}}{\lambda_{^{235}\text{U}}} = \frac{C}{\varepsilon I_\gamma T \lambda_{^{235}\text{U}}} \quad N_{^{234}\text{U}} = \frac{A_{^{234}\text{U}}}{\lambda_{^{234}\text{U}}} \quad N_{^{238}\text{U}} = \frac{A_{^{238}\text{U}}}{\lambda_{^{238}\text{U}}} \quad (3.2)$$

So, the enrichment percentage equation (3.1) can be written as follow:

$$\%E = \left(\frac{\frac{A_{^{235}\text{U}}}{\lambda_{^{235}\text{U}}}}{\frac{A_{^{234}\text{U}}}{\lambda_{^{234}\text{U}}} + \frac{A_{^{235}\text{U}}}{\lambda_{^{235}\text{U}}} + \frac{A_{^{238}\text{U}}}{\lambda_{^{238}\text{U}}}} \right) \times 100 = \left(\frac{A_{^{235}\text{U}} \cdot (\tau_{1/2})_{^{235}\text{U}}}{A_{^{234}\text{U}} \cdot (\tau_{1/2})_{^{234}\text{U}} + A_{^{235}\text{U}} \cdot (\tau_{1/2})_{^{235}\text{U}} + A_{^{238}\text{U}} \cdot (\tau_{1/2})_{^{238}\text{U}}} \right) \times 100 \quad (3.3)$$

Where, $\lambda_{^{234}\text{U}}$, $\lambda_{^{235}\text{U}}$, $\lambda_{^{238}\text{U}}$ are the decay constants and $(\tau_{1/2})_{^{234}\text{U}}$, $(\tau_{1/2})_{^{235}\text{U}}$ and $(\tau_{1/2})_{^{238}\text{U}}$ are the half-lives time of ^{234}U , ^{235}U and ^{238}U respectively.

The result of the sample natural enrichment was obtained from applying equation (3.3) by using the average specific activity of uranium isotopes and the half-life time of each isotope. The calculated results are shown in Table 3. The enrichment percentage of the verified sample is of natural origin (0.729% \pm 0.016) as expected for all the natural environmental samples.

Table 3. Activity results of uranium isotopes and natural enrichment percentage of the sample

Isotopes	Energy (keV)	$t_{1/2}$ (year)	Activity (Bq/kg)	S1	
				Av Activity \pm SD	Calculated Enrichment %
U-235	143.7	7.038E+8	1167.4	1268.675 \pm 98	0.729 \pm 0.016
	163.36		1353.3		
	185.72		1201.2		
	205.23		1352.8		
Th-234	63.29	4.468E+9	26919.77	27186.25 \pm 376	-
Pa-234m	1001		27452.74		
U-234	53.24	2.455E+5	22067.78	22067.78 \pm 82	-
	120.9		22125.81		

3.3. Uranium Isotopic Mass and Total Uranium Concentration Calculations

The uranium mass of certain sample can be calculated by knowing the activity and the decay constant for each uranium isotope through applying the following equations:

$$N_{^{238}U} = n_{^{238}U} N_A = \frac{m_{^{238}U}}{M_{^{238}U}} N_A = \frac{A_{^{238}U}}{\lambda_{^{238}U}} \quad (3.4)$$

Then the mass of uranium isotopes can be expressed as follows

$$m_{^{238}U} = \frac{A_{^{238}U}}{\lambda_{^{238}U}} \cdot \frac{M_{^{238}U}}{N_A}, \quad m_{^{235}U} = \frac{A_{^{235}U}}{\lambda_{^{235}U}} \cdot \frac{M_{^{235}U}}{N_A}, \quad m_{^{234}U} = \frac{A_{^{234}U}}{\lambda_{^{234}U}} \cdot \frac{M_{^{234}U}}{N_A} \quad (3.5)$$

Where, $m_{^{238}U}$, $m_{^{235}U}$ and $m_{^{234}U}$ are the uranium masses of ^{238}U , ^{235}U and ^{234}U respectively. $M_{^{238}U}$, $M_{^{235}U}$ and $M_{^{234}U}$ are the atomic weight of ^{238}U , ^{235}U and ^{234}U respectively. N_A is the Avogadro's number. The total uranium concentration in the sample can be calculated by using the ratio of the total masses of uranium isotopes and the total sample mass as finally calculated by the following Equation:

$$m_{U_t} = \frac{m_{^{238}U} + m_{^{235}U} + m_{^{234}U}}{m_{sample}} \quad (3.6)$$

The uranium isotopes masses of ^{234}U , ^{235}U and ^{238}U were calculated by using equation 3.5 which depends on the activity of the isotope in Bq, the decay constant in s^{-1} , molar mass in $g.mol^{-1}$ and Avogadro's number. The uranium isotopes masses results are found to be 0.0309 mg for ^{234}U , 5.112 mg for ^{235}U and 705.08 mg for ^{238}U and summarized in Table 4.

Table 4. The Uranium isotopes masses and total uranium concentration of the samples

Isotopes	Activity (Bq)	Decay constant $\lambda = \ln(2)/t_{1/2}$ (s^{-1})	Uranium isotopes masses (mg)	Uranium content in 323 g (mg)	Uranium concentration (ppm)
U-234	7127.64	8.94374 E-14	0.0309	710	2198.8
U-235	408.9	3.12161 E-17	5.112		
U-238	8781.07	4.9218 E-18	705.0835		

As shown from table 4. the total uranium concentration as calculated by using equation 3.6, which divides the summation masses of each uranium isotope over the mass of the sample, was found to be 710.2 mg in the sample weight of 323 g. Henceforth, the total uranium concentration was concluded to be 2198.8 ppm. The results compared with previous work and found to be relatively comparable with our results, where their calculated total uranium concentration ranged from 203 to 6147 ppm for this site (El-Sela) [12].

4. Conclusion

In this work, a non-destructive assay technique was applied based on a gamma spectrometer (Hp-Ge) detector, which was calibrated for energy and efficiency by a modified technique to reduce the errors in the results. The Hp-Ge detector was used to measure and analyze specific radioactive sample collected from the El-Sela area in the southeastern desert in Egypt. The sample was well precisely prepared for verification. The measured specific activity was then calculated and found to be very high; 1268.675 ± 98 Bq/kg for ^{235}U , 27186.25 ± 376 Bq/kg for ^{238}U and 22067.78 ± 82 for ^{234}U . The activity ratio of $^{235}U/^{238}U$ was calculated and found to be a natural signature value (0.046). The $^{234}U/^{238}U$ activity ratio was less than unity indicating the occurrence of disequilibrium. The enrichment percentage of the sample was calculated from a derived equation and found to be of natural origin; 0.72%. The uranium isotopes masses were estimated from an equation deduced from the decay equation for ^{234}U , ^{235}U and ^{238}U and

found to be 0.0309, 5.112 and 705 mg, respectively. The total uranium concentration in the sample was also calculated and found to have a high value of 2198.8 ppm ($\mu\text{g/g}$). The verified sample results were compared with previous work for the same Site.

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