Measurements of Radioactivity Concentrations in Granites and Sedimentary-Rocks and their Leaching Components in Egyptian Deserts

Salha D. Y. Alsaadi, Jemila Mussa Ali, Areej Hazawi and Asma Mohammed AL-abrdi.

Physics Department, Faculty of Science, Omar Al-Mukhtar University, El-Beda, Libya.

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Abstract: Concentrations of radionuclides in sediments and granite samples were determined by γ-ray spectrometer using High Pure Germanium Detector; HPGe, with a specially designed shield. Six different rock samples were collected from different sites: four samples of sediments from Um Bogma southwest Sinai, and two granite samples from Gabal Gattar, the northern part of the Eastern Desert of Egypt, where all samples were subject to investigation. Their samples were selected because the activity before being dissociated in sulfuric acid was less than after dissociation. The samples were dissolved in sulfuric acid with the same parameters of solid to liquid ratio; S/L, acid concentration, and leaching time. After the leaching process, the pregnant solution was separated from the residual, and the latter was dried. The two units, named the pregnant solutions and residuals, were also measured radiometrically using the HPGe detector to determine the activity concentrations (Bq/kg) of the different radionuclides of the six samples. The results showed that the relation between the sum of activities of both pregnant solutions and residuals with the originals have different categories. In the sediment samples; the activity of solution+ residual was 72.37% from the original of siltstone, the activity of solution + residual was 90.02% from the original claystone sample, the activity of solution + residual was 92.6% from the original of shale, the activity of solution + residual was 74.07% from the original claystone. In the granite samples, the activity of solution + residual was 130.39% from the original of the first granite sample and 142.3% from the original of the second granite sample. This phenomenon varied in magnitude due to the different radionuclides in each sample. These variations depend mainly on the nature of the grain surfaces in the different rock types and their Pb content. As for leachability analysis, leaching experiments have been performed using sulfuric acid. The leaching efficiency (%) of uranium is estimated by the measurements of the HPGe detector. The result showed almost constant values for leachability. The non-frequent appearance of attenuation of gamma activities during leaching processes indicates that the acid solutions may have led to clean the grain surfaces and thus permit gamma activities of the inner grains to be measured. The treatment of the samples before measurements may have been needed.

Keywords: Natural Radioactivity; HPGe Detector; Sediment; Granite; Leaching

INTRODUCTION

In the leaching process, oxidation potential, temperature, and pH of the solution are important parameters, and are often manipulated to optimize the dissolution of the desired metal component into the aqueous phase (Merritt, 1971). The radium-226 (226Ra) is the main radionuclide in the tailings after leaching process for uranium extraction and is highly active with its solid decay products. The main danger is not during the radioactive releases in the leaching processes only but remains for many years after the end of oper-
ations (Metzler, 2004). Sulfuric acid (\(\text{H}_2\text{SO}_4\)) as an acidic reagent is widely used for uranium leaching because of its availability and low cost. In addition, nitric and hydrochloric acids are not only effective in uranium dissolution but also produce undesirable impurities in the leach liquor ore. Nitric acid appears to be attractive as an oxidizing agent, being suitable for later recovery processes by solvent extraction. In addition, the acid may also be generated autogenously by treating ore that has appreciable sulfides content with air or oxygens under pressure before leaching (Mahdy et al., 1996).

The physical and chemical processes used to extract uranium from ore, such as crushing and acid treatment, produce large amounts of mill tailings. In France today, approximately 50 million tons of uranium mill tailings (UMT) are stored on the surface in specific areas. They contain 99% of the radium present in the original ore and have much higher porosity and permeability than that of the rock from which they were derived. The average activity of 226Ra for French UMTs lies between 4000 and 60,000 Bq/kg. (Ferry et al., 2002) studied the behavior of most of the different radionuclides in the uranium-238 (238U) series and thorium-232 (232Th) series during acidic leaching. They concluded that the nuclides preceding 226Ra in the 238U decay series are easily released in the pregnant solution, compared with the 226Ra itself and its solid daughters: lead (214Pb) and bismuth (214Bi) (El Aassy, et al., 2012).

**Aim of the work:** First to study the transfer of different radionuclides from a solid material (ore) to the liquid phase (leachate). Then to measure the activity concentrations and activity ratios of radionuclides in original, leachate, and residual using an HPGe detector.

### MATERIALS AND METHODS

**Samples Description and Preparation:** Four samples of sediments were collected from Um Bogma, southwest Sinai, Egypt (Fig.1), and two Granite samples were collected from Gabal Gattar in the northern parts of the Eastern Desert (Asma, 2018) in fig(2). The collected samples were crushed and then quartered and packed in 250 ml bottles. The bottles were completely sealed for one month to allow radioactive equilibrium to be reached. This step is necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample. Four samples (Table 1) of different varieties were collected from the sedimentary rock in Um Bogma, southwest Sinai, Egypt.

**Table (1):** Description of the collected of sedimentary samples

| Samples | Description |
|---------|-------------|
| 2S      | Shale from the middle member of the Um Bogma Formation yellowish-brown, soft, fissile ferruginous with 2m thickness 6900 cps. |
| 3S      | Siltstone from the middle member of the Um Bogma Formation and overlying the previous sample, violet with brown patches, and ferruginous 0.2 m, 4200 cps. |
| 7S      | Claystone from the middle member of the Um Bogma Formation (MUF), reddish to yellowish-brown, soft to medium hard, highly ferruginous, 3500 cps. |
| 17S     | Gibbsite – bearing Shale, from the middle member of the Um Bogma Formation, yellow to brown with black patches and concretions of carbonaceous organic matter, 0.5 m, 4000 cps. |

**Fig. (1):** Geologic map of the Um Bogma area (El Aassy et al.,1986; Omar., 2016).
Leaching experiments: The leaching process was carried out by using Sulfuric acid (H2SO4), acid leaching on 150 g sample weight under the conditions; solid/liquid ratio 1:3, acid concentration 30%, stirring time of one hour, and at room temperature. Filtration was carried out to separate leachate from residual that was then dried. The leachate was packed well in 250 ml Marinelli beakers. Also, the residual was left for one month to be measured by an HPGe-detector.

The vertically mounted HPGe detector was coupled with a multichannel analyzer card which is pre-installed on a PC. The radiation measuring system is also composed of the usual electronic components; a preamplifier, an amplifier, and power supply units. The detector has a resolution (FWHM) of 1.85 keV for the 1332.5 keV γ-ray line of cobalt (60Co). The γ-ray spectrometer energy calibration was performed using 60Co, 226Ra, and americium (241Am) point sources. The detector was surrounded by a special heavy lead shield of 10 cm thickness with an inside diameter of 28 cm and height of 40.5 cm. The absolute detection efficiency of the HPGe detector was determined by using three well-known reference materials obtained from the International Atomic Energy Agency for U, Th, and K (potassium) activity measurements: RGU-1, RGTh-1, and RGK-1 (IAEA., 1987; Anjos et al., 2005). The sample containers were placed on top of the detector for counting. The same geometry and size were used for both the samples and the reference materials (Turhan et al., 2008). The Uranium standard (RGU-1) is U-ore diluted with silica with 4940 Bq/kg of 238U, 228 Bq/kg of uranium-235 (235U), a negligible amount of 40K (less than 0.63 Bq/kg), and some traces of 232Th (less than 4 Bq/kg). The thorium standard (RGTh-1) is Th-ore diluted with silica having 3250 Bq/kg of 232 Th, but containing some 238U (78 Bq/kg) and 40K (6.3 Bq/kg).

The potassium calibration standard (RGK-1) is produced from high purity (99.8%) potassium sulfate with 14000 Bq/kg of potassium with uranium and thorium contents lower than 0.001 and 0.01 ppm (parts per million), respectively (Anjos et al., 2005).

The γ-ray transitions used to measure the concentration of the assigned nuclides in the series are as follows: 238U was determined from the gamma rays emitted by its daughter products (Sutherland and de Jong., 1990), 234Th and 234mPa activities determined from the 63.3 and 1001 keV photo peaks, respectively, 214Bi (609.3, 1120.3, 1238.1, 1377.7, and 1764.5 keV), 214Pb (295.1 and 352.0 keV). The specific activity of 226Ra was measured using the 186.1 keV from its own gamma-ray (after the subtraction of the 185.7 keV of 235U). The specific activity of 232Th was measured using the 338.4, 911.2, and 968.9 keV from 228Ac, and 583 keV from 208Tl and 40K was measured using 1460.8 keV for potassium.

In order to determine the background contribution due to naturally occurring radionuclides in the environment around the detector, an empty polyethylene beaker of the same 250 cm3 volume was counted with the same geometrical conditions as the sample. The measurement time for both activity and background set at 70,000 seconds. The background spectra were used to correct the net
gamma-ray peak areas for the studied isotopes.

X-Ray Fluorescence (XRF) Spectrometry: XRF is used to determine the chemical analysis of major and trace elements in the samples. Detection limits for major and trace elements are typically of the order of a percent and part per million (ppm). Six samples were chosen for processing this application within the Scale of the laboratory: Four sediment samples from Um Bogma (siltstone (3S), claystone (17S), shale (2S), and (7S) claystone), and two samples of granite from Gabal Gattar. The collected technological sample had intermixed with organic-rich earthy soil materials and were crushed to a size of 200 mesh. Proper quartering of the technological sample was performed after its grinding to less than 200 mesh size to obtain a representative sample which was subjected to a complete chemical analysis of both major and trace elements content which were analyzed at the laboratories of the National Research Center (NRC), Cairo, Egypt, by the XRF technique, using Philips Unique II unit fitted with an automatic sample changer PW 1510 (30position), connected to a computer system using X-40 controlling program for spectrometry. The detection limit of the measured elements by the XRF technique was estimated to be 5ppm.

RESULTS AND DISCUSSION

The results of gamma-detector of radionuclides in original samples (Bq/kg), leachates (Bq/l), and residuals (Bq/kg) are presented in table (2, 3, 4, 5, 6, and 7) and fig (3). From this table, it is noticed that there is a difference between the sum of activities of residuals and leachates with the activity of the original sample. This difference varied mostly with the variation of the lithological type and/or geochemical composition.

| Radionuclide | Original [Orig.] (Bq/Kg) | Residual [Resid.] (Bq/Kg) | Solution [Sol.] (Bq/l) |
|--------------|--------------------------|---------------------------|----------------------|
| **238U series** |                          |                           |                      |
| 234Th        | 6159.5±31.6              | 7754.0±60.3               | 1996.8±11.2          |
| 234mPa       | 6483.8±152.1             | 5735.7±283.8              | 1682.6±46.1          |
| Average      | 6321.7±91.9              | 6744.8±172.1              | 1839.7±28.6          |
| 234U         | 6127.6±674.2             | 6912.7±184.7              | 1327.4±124.5         |
| 230Th        | 4746.4±182               | 1042.6±736.7              | 591.4±42.9           |
| **226Ra subseries** |                          |                           |                      |
| 226Ra        | 4841.9±14.93             | 7711.9±34.0               | 25.9±0.8             |
| 214Pb        | 4279.4±9.8               | 5536.3±18.23              | 17.8±0.8             |
| 214Bi        | 4107.1±25.4              | 4945±45.3                 | 29.2±2.4             |
| Average      | 4409.4±16.7              | 6064.4±32.5               | 24.3±1.3             |
| 235U         | 328.8±7.8                | 288.8±13.1                | 95.8±2.0             |
| **232Th series** |                          |                           |                      |
| 228Ac        | 55.9±3.7                 | 91.9±8.4                  | 6.2±0.9              |
| 208Tl        | 60.7±2.3                 | 64.9±4.9                  | 6.7±0.6              |
| Average      | 58.3±3                   | 78.4±6.7                  | 6.5±0.7              |
| 40K          | 342.8±13.3               | 619.4±28.1                | 143.0±4.8            |

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Table (3): Gamma radioactivities for different radionuclides in pregnant solution and residual (in percentages) of Siltstone sample No. (3S)

| Sediment sample (3S) | Radionuclide | Original [Orig.] (Bq/Kg) | Residual [Resid.] (Bq/Kg) | Solution [Sol.] (Bq/l) |
|----------------------|--------------|--------------------------|----------------------------|------------------------|
| 238U series          | 234Th        | 2610.67±22.5             | 2094.3±102.4               | 759.1±4.4              |
|                      | 234mPa       | 2032.9±107.3             | 1817.7±527.6               | 568.1±21.6             |
|                      | Average      | 2321.7±64.9              | 1656.0±315                 | 663.6±13.0             |
|                      | 234U         | 3946.5±91.9              | 1346.2±401                 | 513.1±97.5             |
|                      | 230Th        | 2230.0±158.8             | 2972.2±218.4               | 92.3±7.6               |
| 226Ra subseries      | 226Ra        | 2385.8±12.2              | 2645.4±31.1                | 15.5±0.6               |
|                      | 214Pb        | 2099.3±7.2               | 2008.2±10.8                | 8.6±0.5                |
|                      | 214Bi        | 2001.7±18.6              | 1807.0±31.6                | 2.7±0.3                |
|                      | Average      | 2162.2±12.6              | 2153.5±24.5                | 8.9±0.5                |
|                      | 235U         | 114.8±5.1                | 94.6±9.4                   | 28.5±1.4               |
| 232Th series         | 228Ac        | 67.8±3.5                 | 116.1±7.8                  | 1.4±0.01               |
|                      | 208Tl        | 68.5±2.1                 | 81.6±4.5                   | 5.1±0.4                |
|                      | Average      | 68.2±2.7                 | 98.9±6.1                   | 3.2±0.2                |
|                      | 40K          | 562.4±13.5               | 906.3±28.5                 | 92.1±3.4               |

Table (4): Gamma radioactivities for different radionuclides in pregnant solution and residual (in percentages) of Claystone sample No. (7S)

| Sediment sample (7S) | Radionuclide | Original [Orig.] (Bq/Kg) | Residual [Resid.] (Bq/Kg) | Solution [Sol.] (Bq/l) |
|----------------------|--------------|--------------------------|----------------------------|------------------------|
| 238U series          | 234Th        | 2941.5±22.9              | 3097.4±61.8                | 865.9±5.9              |
|                      | 234mPa       | 3318.5±124.9             | 3345.2±28.3                | 518.3±25.8             |
|                      | Average      | 3310.0±73.8              | 3221.3±45.1                | 692.1±15.8             |
|                      | 234U         | 3333.2±79.7              | 1124.3±91.1                | 390.9±84.3             |
|                      | 230Th        | 2264.7±161               | 1565.7±118.1               | 133.5±10.7             |
| 226Ra subseries      | 226Ra        | 2448.7±11.3              | 3230.8±32.1                | 62.9±1.8               |
|                      | 214Pb        | 2081.9±6.94              | 2164.2±15.8                | 19.1±0.7               |
|                      | 214Bi        | 1983.2±17.8              | 1898.1±37.2                | 20.6±0.8               |
|                      | Average      | 2171.2±12.0              | 2431.0±28.6                | 34.3±1.1               |
|                      | 235U         | 155.8±5.3                | 70.9±8.2                   | 34.5±1.8               |
| 232Th series         | 228Ac        | 59.2±3.2                 | 54.1±6.8                   | 8.5±1.0                |
|                      | 208Tl        | 59.1±2.1                 | 45.1±4.7                   | 7.5±0.6                |
|                      | Average      | 59.1±2.6                 | 49.6±5.8                   | 8.0±0.8                |
|                      | 40K          | 352.5±11.3               | 609.4±31.2                 | 106.6±4.1              |
Table (5): Gamma radioactivities for different radionuclides in pregnant solution and residual (in percentages) of Gibbsite sample No. (17S)

| Radionuclide     | Original [Orig.] (Bq/Kg) | Residual [Resid.] (Bq/Kg) | Solution [Sol.] (Bq/l) |
|------------------|----------------------------|---------------------------|------------------------|
| $^{238}$U series |                            |                           |                        |
| $^{234}$Th       | 3724.3±30.2                | 3862.7±47.7               | 1212.3±9.5             |
| $^{234}$mPa      | 4482.5±150.5               | 2907.8±274.7              | 1304.7±44.6            |
| Average          | 4103.0±22.5                | 3385.3±161.1              | 1258.5±27.1            |
| $^{234}$U        | 4198.9±86.3                | 3276.0±121.2              | 760.6±107.3            |
| $^{230}$Th       | 3722.8±213.9               | 1631.1±119.9              | 250.6±19.22            |

| $^{238}$Ra subseries |                          |                           |                        |
|----------------------|--------------------------|---------------------------|                        |
| $^{226}$Ra          | 4102.9±14.3              | 6048.5±30.6               | 20.2±0.8               |
| $^{214}$Pb          | 3322.8±8.8               | 3860.7±16.1               | 36.2±1.1               |
| $^{214}$Bi          | 3232.6±22.9              | 3299.0±38.9               | 40.9±3.0               |
| Average             | 3552.7±15.3              | 4402.7±28.5               | 32.4±1.6               |
| $^{235}$U           | 190.0±6.9                | 161.9±13.7                | 54.7±2.3               |

| $^{230}$Th series   |                          |                           |                        |
|---------------------|--------------------------|---------------------------|                        |
| $^{228}$Ac         | 73.9±4.4                 | 18.5±5.3                  | 7.3±1.2                |
| $^{208}$Tl         | 57.5±2.3                 | 29.0±4.3                  | 5.2±0.6                |
| Average            | 65.7±3.4                 | 23.7±4.8                  | 6.3±0.9                |
| $^{40}$K           | 523.9±14.4               | 759.1±30.7                | 106.5±5.1              |

Table (6): Gamma radioactivities for different radionuclides in pregnant solution and residual (in percentages) of granite sample No. (2G)

| Radionuclide     | Original [Orig.] (Bq/Kg) | Residual [Resid.] (Bq/Kg) | Solution [Sol.] (Bq/l) |
|------------------|----------------------------|---------------------------|------------------------|
| $^{238}$U series |                            |                           |                        |
| $^{234}$Th       | 2589.5±21.8                | 1176.6±40.1               | 872.8±6.5              |
| $^{234}$mPa      | 2363.5±111.1               | 2043.2±23.8               | 727.2±30.1             |
| Average          | 2476.5±66.4                | 1609.9±31.9               | 800.8±8.3              |
| $^{234}$U        | 2397.1±54.8                | 8155.3±208.2              | 1336.3±105.5           |
| $^{230}$Th       | 2269.8±109.2               | 3870.3±85.9               | 483.7±35.2             |

| $^{232}$Ra subseries |                          |                           |                        |
|----------------------|--------------------------|---------------------------|                        |
| $^{226}$Ra          | 2258.9±9.0               | 4293.0±31.5               | 2.5±0.2                |
| $^{214}$Pb          | 1925.9±5.8               | 3936.7±17.2               | 17.9±0.7               |
| $^{214}$Bi          | 1901.6±15.1              | 3651.3±43.4               | 17.9±0.4               |
| Average             | 2028.8±9.9               | 3960.3±30.7               | 12.7±0.4               |
| $^{235}$U           | 115.1±5.5                | 54.5±6.3                  | 33.0±1.5               |

| $^{232}$Th series   |                          |                           |                        |
|---------------------|--------------------------|---------------------------|                        |
| $^{228}$Ac         | 88.7±3.8                 | 132.3±11.5                | 3.9±0.3                |
| $^{208}$Tl         | 74.5±2.1                 | 56.7±7.0                  | 12.7±0.6               |
| Average            | 81.6±2.9                 | 94.5±9.3                  | 8.3±0.5                |
| $^{40}$K           | 1195±14.6                | 2096.4±37.3               | 100.6±3.6              |
**Table (7):** Gamma radioactivities for different radionuclides in pregnant solution and residual (in percentages) of granite sample No. (5G)

| Radionuclide | Original [Orig.] (Bq/Kg) | Residual [Resid.] (Bq/Kg) | Solution [Sol.] (Bq/l) |
|--------------|--------------------------|---------------------------|------------------------|
| **234Th**    | 2705.3±19.4              | 1076.7±37.4               | 950.5±11.4             |
|             | 234mPa                   | 2660.0±118.5              | 1049.6±31.9            | 992.9±49.4          |
| Average      | 2682.6±68.9              | 1063.1±34.6               | 971.7±30.4             |
| **234U**     | 2958.0±64.8              | 7016.0±196.1              | 1330.1±150.2           |
| **230Th**    | 2549.7±180.5             | 3906.2±17.3               | 772.1±56.1             |
| **236Ra**    | 2316.7±9.7               | 4100.6±31.9               | 23.5±0.9               |
| **214Pb**    | 2020.1±6.3               | 3868.7±15.1               | 34.4±1.3               |
| **214Bi**    | 2016.2±16.5              | 3474.9±46.6               | 47.2±3.6               |
| Average      | 2117.6±10.8              | 3814.7±31.2               | 35.1±1.9               |
| **235U**     | 123.7±4.8                | 56.2±5.8                  | 42.4±2.5               |
| **228Ac**    | 95.1±4.1                 | 112.2±7.8                 | 13.4±1.5               |
| **208Tl**    | 83.1±2.2                 | 56.4±4.3                  | 25.3±1.1               |
| Average      | 89.1±3.2                 | 84.3±6.1                  | 19.3±1.3               |
| **40K**      | 1264±15.8                | 2092.5±37.5               | 189.4±6.3              |

**Fig. (3):** Activity concentration of 238U, 234U, 226Ra, 214Bi and 235U in the original samples (Bq/kg), leachate (solution) (Bq/l) and residual (Bq/kg)

The measurements of leachability (leaching efficiency %): The chemical behavior of each radionuclides clearly varies in the same sample. The measurements of leaching efficiency % by using the HPGe detector showed that leachability is the highest (55.7%) for 234U in sample (2G) table (8), while the 238U is 36.2% in (5G) and 235U is 34.3% in (5G).

The variation of leachability was also according to the type of sample, the increased release of 234U and the variability of 234U/238U was observed before by several authors who mentioned that the release of excess 234U could arise from preferential release from damaged lattice sites (Bourdon et al., 2009). It is worth noticing that the values of leachabilities for 235U and 238U are about the same for most samples, and they are almost identical for samples (2S and 7S).

234U and 238U have similar chemical behavior. However, in Table (8) it is noticed that the leachability % for 234U is more than 238U for samples (7S, 3S, 5G, 2G). So these samples have physical transfer alongside the chemical transfer of 234U. The physical transfer is due to α-recoil and the natural 238U decay.
Table (8): The leachability of the chosen samples measured by HPGe.

| Radionuclide Concentration | Original | Residual | Solution | Leachability | sum | % |
|----------------------------|----------|----------|----------|--------------|-----|---|
| $^{238}$U(ppm)              | 509.8±7.4| 543.9±13.8| 148.4±2.3| 29.1         | 692.3| 135.8|
| $^{234}$U(ppb)              | 26.5±2.6 | 29.9±0.1 | 5.7±0.5 | 21.6         | 35.6 | 134.5|
| $^{235}$U(ppm)              | 4.1±0.7  | 3.6±1.1  | 1.2±0.2 | 29.1         | 4.8 | 116.7|
| $^{238}$U(ppm)              | 252.4±5.9| 259.7±3.6| 55.8±1.3 | 22.1         | 305.6| 121.1|
| $^{234}$U(ppb)              | 14.4±0.3 | 4.8±0.4  | 1.7±0.4 | 11.7         | 6.5 | 45.4|
| $^{235}$U(ppm)              | 1.95±0.4 | 0.8±0.4  | 0.4±0.1 | 22.2         | 1.3 | 67.3|
| $^{238}$U(ppm)              | 187.2±1.8| 133.5±12.9| 53.5±2.2| 28.6         | 187.1| 100|
| $^{3}$S                     |          |          |          |              |     |    |
| $^{238}$U(ppm)              | 17.1±0.4 | 5.8±0.5  | 2.2±0.5 | 13.0         | 8.1 | 47.7|
| $^{234}$U(ppb)              | 1.4±0.6  | 1.2±1.1  | 0.4±0   | 24.9         | 1.5 | 107.6|
| $^{235}$U(ppm)              | 330.8±5.2| 273.0±25.4| 101.5±1.0| 30.6         | 374.5| 113.2|
| $^{17}$S                    |          |          |          |              |     |    |
| $^{234}$U(ppb)              | 18.2±0.4 | 14.2±1.7 | 3.3±0.3 | 18.1         | 17.5 | 96.1|
| $^{235}$U(ppm)              | 2.4±0.4  | 2.0±0.8  | 0.7±0.1 | 28.8         | 2.7 | 113.9|
| $^{238}$U(ppm)              | 190.6±5.3| 94.8±2.5 | 64.5±0.7| 33.85        | 159.4| 83.6|
| $^{234}$U(ppb)              | 10.4±0.2 | 35.3±0.9 | 5.7±0.5 | 55.74        | 41.1 | 396.1|
| $^{235}$U(ppm)              | 1.4±0.4  | 0.3±0.5  | 0.4±0.1 | 28.66        | 0.8 | 52.4|
| $^{238}$U(ppm)              | 216.3±5.6| 86.88±2.8| 78.4±2.4| 36.22        | 165.3| 76.4|
| $^{5}$G                     |          |          |          |              |     |    |
| $^{234}$U(ppb)              | 12.80±0.3| 30.37±0.8| 5.7±0.6 | 44.96        | 36.12| 282.2|
| $^{235}$U(ppm)              | 1.54±0.0 | 0.35±0.4 | 0.5±31 | 34.32        | 0.88 | 57.1|

ppm = part per million                                          ppb= part per billion

Table (9): Major elements in sediment and granite samples

| Major   | 3S %  | 17S %  | 2S %  | 7S %  | 2G %  | 5G %  |
|---------|-------|--------|-------|-------|-------|-------|
| SiO2    | 39.57 | 28.608 | 21.451| 20.998| 70.755| 70.45 |
| Al2O3   | 26.38 | 16.31  | 14.297| 12.66 | 13.041| 13.092|
| TiO2    | 0.199 | 0.927  | 0.529 | 0.454 | 0.115 | 0.107 |
| Fe2O3   | 4.063 | 23.195 | 41.024| 35.696| 3.536 | 3.654 |
| MnO     | 0.014 | 2.168  | 0.069 | 0.099 | 0.055 | 0.055 |
| CaO     | 1.963 | 0.246  | 0.247 | 0.612 | 1.458 | 1.504 |
| MgO     | 0.772 | 0.763  | 0.459 | 0.537 | 0.293 | 0.334 |
| Na2O    | 4.999 | 3.587  | 1.575 | 7.434 | 5.344 | 5.475 |
| K2O     | 0.132 | 2.004  | 0.859 | 0.765 | 5.094 | 4.973 |
| P2O5    | 0.132 | 0.069  | 0.062 | 0.06  | 0.032 | 0.034 |
| SO3     | 2.22  | 0.427  | 0.389 | 0.129 | 0.094 | 0.083 |
From Table (9), it is obvious that the sample of siltstone (3S) has a high level of loss on ignition. Silica, alumina, and iron oxide contents are 39.57, 26.38, and 4.06% respectively.

The claystone sample (17S) has a high level of loss on ignition. Silica, alumina, and high iron oxide contents are 28.60, 16.31, and 23.19% respectively. The shale sample (2S) has a high content of loss on ignition. Silica, alumina, and iron oxide contents are 21.45, 14.29, and 41.02% respectively. The claystone sample (7S) has a high loss on ignition, attaining silica, alumina, and high iron oxide contents of 20.998, 12.66, and 35.69% respectively.

The chemical analysis of the two granite ore samples (2G, 5G) shows they are mainly composed of SiO2 and Al2O3 as 70.7% and 13% respectively for 2G, and 70.35% and 13.09% respectively for sample 5G. Meanwhile, a total ignition loss of about 1% was obtained at 1000 °C for the two samples 2G and 5G.

Table (10) presents the trace element analysis in the samples. The more interesting detail in these results is that the sedimentary samples contain a high concentration of Cl, Cu, Zn, Ni, Pb, U, Co, Cr, and some REEs which affect the characterization of these samples and their behavior during the radioactivity measurements process as will be seen later.

Table (10): Trace elements in sediment and granite samples.

| Trace | 3S (ppm) | 17S (ppm) | 2S (ppm) | 7S (ppm) | 2G (ppm) | 5G (ppm) |
|-------|----------|-----------|----------|----------|----------|----------|
| Ni    | 416.0    | 1742.7    | 1138.2   | 7300.5   | 0        | 0        |
| Cu    | 127.6    | 718.2     | 0        | 287.2    | 63.8     | 0        |
| Zn    | 2192.1   | 25760.2   | 21777.3  | 16573.9  | 176.6    | 168.6    |
| Rb    | 91.4     | 164.5     | 0        | 0        | 329.0    | 319.9    |
| Sr    | 591.5    | 371.8     | 321.1    | 202.8    | 0        | 42.2     |
| Y     | 78.7     | 393.5     | 259.7    | 165.2    | 0        | 0        |
| Zr    | 651.2    | 984.2     | 466.2    | 310.8    | 266.4    | 259      |
| Nb    | 55.9     | 0         | 0        | 0        | 167.7    | 146.7    |
| Ba    | 143.2    | 349.0     | 0        | 0        | 0        | 0        |
| Ce    | 230.3    | 3684.9    | 0        | 0        | 0        | 0        |
| Pb    | 798.1    | 1688.9    | 1865.2   | 204.1    | 111.3    | 102.1    |
| CL    | 2887     | 22510     | 11570    | 33240    | 0        | 630      |
| U     | 250      | 1020      | 1050     | 300      | 280      | 280      |
| V     | 0        | 5488      | 5488     | 0        | 0        | 0        |
| Co    | 0        | 2130.0    | 0        | 550.2    | 0        | 0        |
| Rh    | 0        | 110       | 0        | 0        | 0        | 0        |
| Mo    | 0        | 0         | 226.4    | 159.8    | 0        | 0        |
| Nd    | 0        | 0         | 0        | 154.2    | 0        | 0        |
| As    | 0        | 0         | 0        | 332.0    | 0        | 0        |
| Cr    | 465.1    | 0         | 362.5    | 259.9    | 143.6    | 157.3    |
From Tables (2 and 5), the lithological type played its role as in sample 7S in which the sum of gamma activity is lower by 20% than the activity of the original sample. The geochemical composition is very clear in samples 2S, 3S, and 17S. In these samples, the lead (Pb) content is 1865.3 ppm, 798 ppm, and 1689 ppm respectively, as shown in Table (10). The sum of γ- activity in leachate and residual is higher than the original sample. The sum is higher by 26.3% than the original in the sample 2S, while it is higher by 11.6% in the sample 17S respectively. The relation between the gamma attenuation and (Pb) concentration is very clear in Fig (5), in which the gamma attenuation increases with the increase of the Pb concentration in the measured sample.

![Fig. (5): Correlation between γ – attenuation and Pb concentration on sediment samples.](image)

**CONCLUSION**

The results showed that the sum of activities of both pregnant solutions and residuals in four samples is higher than that of the originals. Two samples (siltstone sample 3S) and (granite 5G) showed lower summation than the original. This phenomenon is varied in magnitude within the different radionuclides. The type of samples plays its role in these variations. The none-frequent appearance of the gamma attenuation activities is inevitable during the leaching process. In other logical cases, no attenuation or other causes for altering the measured activities are observed. In the leaching process, using acid solutions may have led to clean the grain surface and permitted gamma activities of the inner grains to be measured, which means that the treatment of samples before measurements may have been needed.

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قياسات تركيزات النشاط الإشعاعي في الصخور الرسوبية والجرانيت ومكونات الترشيح الخاصة بها في الصحراء المصرية

صالحة ذاوود الساعدي
قسم الفيزياء، كلية العلوم، جامعة عمر المختار، مدينة البيضاء، ليبيا.

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المستخلص: أجريت عملية قياس تركيزات النواديد المشعة في عينات الرسوبات والجرانيت باستخدام كاشف الجرمانيوم عالي النقاوة مصمم خصيصًا لهذا القياسات. تم جمع ستة عينات صخرية مختلفة من مواقع مختلفة، وأربع عينات من الصخور الرسوبية جمعت من أم بوقما في جنوب غرب سيناء، وعينتين من الجرانيت من جبل قطر بالصحراء الشرقية. في مصر، تم اختيار عينات بانشاط قليل للدودان في حمض الكبريت، أقل من النشاط بعد الدودان. أُثبتت العينات السبعة من الصخور المختلفة في حمض الكبريت مع مراعة الظروف نفسها، من حيث نسبة العينة الصلبة إلى المذيب (S / L)، تركيز الحمض المتبقي، ومن الاهربي، ثم تم عملية ترشيحها. بعد عملية الترشيح، فصل المتحر عن المتبقي، وتم تجفيف الأخير، واستخراج النشاط الإشعاعي لهما باستخدام كاشف الجرمانيوم عالي النقاوة لتحديد تركيزات النشاط (بيكيل/كم) للنواديد المستخلصة لعينات الستة. أظهرت النتائج أن العلاقة بين مجموع أنشطة كل من المحاليل والمنشطات والأصل لها فئات مختلفة. في العينة الأولى لعينات الرسوب، كان نشاط المحلول والمتبقي 72.37% من الأصل للطمي المتجر، ونشاط المحلول والمتبقي 90.02% من عينة الحجر الطيني، ونشاط المحلول والمتبقي 92.6% من الأصل للحجر الصخري (الثأري)، ونشاط المحلول والمتبقي 74.07% من الأصل للعينة الطينية. أما في عينات الجرانيت، كان نشاط المحلول والمتبقي 130.39% من العينة الأصلية الأولى من الجرانيت و142.03% من العينة الأصلية الثانية من الجرانيت. خالفت هذه الظواهر من المبدئات بسبب النواديد المشعة المختلفة في كل عينة. تعددت هذه الاختلافات بشكل أساسي على طبيعة أساطير الحبيبات في أنواع الصخور المختلفة ومحارب من الزمرد. أما بالنسبة لتحليل الترسبات، فقد تمت إجراء تجارب الترشيح باستخدام حمض الكبريت، وتم تقدير كفاءة ترشيح البرونز (٪) من خلال قياسات جهاز الكشف HPG. أظهرت النتائج فيما ثابته ترشيق للوردي، الترشيح يثير الظروف غير المتكرر للتوحيد لأنشطة جاما أثناء عملية الترشيح إلى أن المحاليل الحمضية قد تؤدي إلى تظيف أسطح الحبيبات، وبالتالي تسمح بقياس أنواع جاما للحمض الداخلية. قد تكون هناك حاجة إلى معالجة العينات قبل القياسات.

الكلمات المفتاحية: نشاط الإشعاعي الطبيعي، كاشف HPGe، الرسوبات، الجرانيت، الترشيح.