

Arab Journal of Nuclear Sciences and Applications

Web site: ajnsa.journals.ekb.eg



# **Relations Between Radionuclides Activities and Pb Concentration on Different Rock Types**

# E. S. Abd EL-Halim<sup>1</sup> and Asma Mohammed AL-abrdi<sup>2</sup>

<sup>(1)</sup> Faculty of Women for Arts, Science and Education, Physics Department, Ain Shams University, Egypt. <sup>(2)</sup> physics department, College of science Omar AL-mokhtar University, Libya.

ARTICLE INFO	ABSTRACT
<i>Article history:</i> Received: 30 <sup>th</sup> Nov. 2020 Accepted: 1 <sup>st</sup> Apr. 2021	The level of natural radioactivity in eight sediment rock samples collected from western south Sinai from three different locations Um Bogma, Wadi Um Hamd and wadi Sad El-Banat, were measured. Also, two granite samples, collected from Gabal Gattar,
Keywords: Natural radioactivity, HPGe detector, Sediment rocks, Granite; Leaching.	north eastern desert in Egypt was investigated. The gamma-ray spectrometry system with hyper pure germanium (HPGe) detector was been used to determined radioactivity concentrations. Samples were subjected to sulphuric acid leaching processes with the same parameters of solid 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 samples.
	The results showed that the relation between the sum of activities of <sup>238</sup> U in both solutions and residuals with originals have two modes. The first mode is represented by the sum of <sup>238</sup> U activities in leachate and residual is nearly equal the activity in the original sample. While the second mode, the sum is higher than the original. These variations depend mainly on the grain surfaces in the different rock types and its content high Pb concentration. This phenomenon varied in magnitude within the different radionuclides in each sample. Also, the leachability of U by the chemical
	analysis using several acidic leaching experiments for uranium have been performed using sulfuric acid.

# **INTRODUCTION**

Natural radionuclides are present in all rock materials in varying amounts depending on their concentration levels in sources rock materials. The three naturally occurring radioactive decay chains include the <sup>238</sup>U, <sup>235</sup>U and <sup>232</sup>Th, which decay through a series of radioactive elements up to stable Pb isotopes. These series are the main sources of gamma radiation in rocks, soils and water. Gamma ray from these radioisotopes represents one of the main external sources of population exposure [1]. The knowledge of distribution of the primordial radionuclides is an important role for peoples avoid long exposure.

In the leaching process, oxidation potential, temperature, and pH of the solution are important parameters, and are often manipulated to optimize aqueous phase [2]. The <sup>226</sup>Ra 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 only during the radioactive releases during leaching processes, but also for many years after cessation of operations [3]. Sulfuric acid as an acidic reagent is widely used for uranium leaching because of its availability and its low cost [4]. The physical and chemical processes used to extract uranium from ore, such as crushing and acid treatment produce large amounts of mill tailings. Studies of the behavior of most of different radionuclides in the 238U and 232Th series during the acidic leaching concluded that the nuclides before <sup>226</sup>Ra in the <sup>238</sup>U decay series are easily released in the pregnant solution than the <sup>226</sup>Ra itself and its solid daughters (<sup>214</sup>Pb and <sup>214</sup>Bi) [5][6].

dissolution of the desired metal component into the

### **Sample Description and Preparation:**

Sediment samples were collected from south western Sinai, Egypt, four samples from middle member of Um Bogma formation, 2S shale, 3S siltstone, 7S claystone and 17S Gibbsite-bearing shale. Two samples from wadi Um Hamd (A1, A2), two samples from wadi Sad El-Banat (A3, A4) and two granite samples (2G, 5G) were collected from Gabal Gattar, north eastern desert. The samples were collected from a depth of 10 cm. Each sample was collected from an area corresponding to one m<sup>2</sup> and was homogenized in situ, and this sand mixture, weighing approximately one kg.

The collected samples were prepared for  $\gamma$ -ray spectrometric analysis by HPGe detector. The samples were first dried, crushed and sieved to about 200 meshes. The samples were placed in polyethylene bottles and weighted. 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.

# **Experimental Technique**

### **Leaching Experiments**

The leaching process was carried out by using H<sub>2</sub>SO<sub>4</sub>, acid (150 g sediments and granite sample weight under the conditions; solid/liquid ratio 1:3, acid concentration 30%, stirring time one hour and at room temperature). Filtration was carried out to separate the leachate from residual which is dried. The leachate was packed well in 250ml marinelli and also the residual was left for one month for measurements by HPGe. High purity vertical germanium was coupled to a PCcomputer with a special electronic card to make it equivalent to a multichannel analyzer. The system also components contains the usual electronic of preamplifier, amplifier and power supply. The detector has resolution (FWHM) of 1.85 keV for the 1332.5 keV  $\gamma$ -ray line of <sup>60</sup>Co. The  $\gamma$ -ray spectrometer energy calibration was performed using 60Co, 226Ra and 241Am point sources. The detector was surrounded by a special heavy lead shield of 10 cm thickness with inside dimension 28 cm diameter 40.5cm height. To achieve a high efficiency of the detection, it is indispensable to assure that the highest possible number of particles emitted from the source enters into the effective volume of the detector. This can be achieved by the choice of the maximum possible measurement geometry, by exclusion Sample itself (self-absorption) and by exclusion or limitation of the kinetic energy losses of the detected

particles between the sample and the effective volume of the detector [6]. For efficiency calibration, it is essential that nuclides are used for which the gamma ray emission probabilities are known accurately and that a source of known activity is used. Whenever possible, sources that have been certified as to their radioactive content should be used [7]. The absolute detection efficiency of the HPGe detector was determined by using three wellknown reference materials obtained from the International Atomic Energy Agency for U, Th and K activity measurements: RGU-1, RGTh-1 and RGK-1 [8] [9]. 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 [10]. The uranium reference material (RGU-1) is U-ore diluted with silica with 4940 Bq.Kg<sup>-1</sup> of <sup>238</sup>U, 228 Bq.Kg<sup>-1</sup> of <sup>235</sup>U, a negligible amount of <sup>40</sup>K ( less than 0.63 Bq.Kg<sup>-1</sup>) and some traces of <sup>232</sup>Th (less than 4 Bq.Kg<sup>-1</sup>). The thorium reference material (RGTh-1) is Th-ore diluted with silica having 3250 Bq.Kg<sup>-1</sup> of <sup>232</sup>Th, but containing some <sup>238</sup>U (78 Bq.Kg<sup>-1</sup>) and <sup>40</sup>K (6.3 Bq.Kg<sup>-1</sup>). The potassium calibration reference material (RGK-1) is produced from high purity (99.8%) potassium sulphate with 14000 Bq.Kg<sup>-1</sup> of potassium with uranium and thorium contents lower than 0.001 and 0.01 ppm, respectively [9]. Uranium-238 activity was determined indirectly from the gamma rays emitted by its daughter products (<sup>234</sup>Th and <sup>234m</sup>Pa) whose activities are determined from the 63.3 (3.9%) and 1001(0.7%)keV photopeaks, respectively.

The gamma-ray transitions of  $^{228}$ Ac (338.4 (12.3%), 911 (29%)) KeV,  $^{212}$ Bi (727.3 KeV (7%)) and  $^{208}$ Tl (583.1 KeV (30%)) were used to evaluate the specific activity of  $^{232}$ Th. The activity concentration of  $^{226}$ Ra was measured from 186.1 KeV (3.29 %).  $^{40}$ K was determined directly from the 1460 KeV (10.7%) peak energy [11].

The background contribution due to naturally occurring radionuclides in the environment around the detector is determined; an empty polyethylene beaker of the same 250 cm<sup>3</sup> volume was counted with the same geometrical conditions as the sample. The measurement time for both activity and background measurement was 70000 seconds. The background spectra were used to correct the net- gamma- ray peak areas for the studied isotopes.

The Leachability of the radionuclides was calculated according to the following equation:

 $Leachability (\%) = \frac{Activity \ concentration \ in \ leachate}{Activity \ concentration \ in \ original} x \ 100$ 

#### X – Ray fluorescence (XRF) Spectrometry

XRF was used to determine the chemical analysis of major and trace elements. In this study, the concentration of trace elements was determined to focus light from the presence of high concentration for lead in most samples.

In this study, the trace elements is measured at the laboratories of the National Research Center (NRC), Egypt, by the X-ray fluorescence technique (XRF) using Philips Unique II unit fitted with an sample changer PW 1510 (30 position). The instrument is connected to a computer system using X-40 program for spectrometry. The detection limit of the measured elements by XRF technique was estimated at 2ppm for Rb, Nb, Ga, Y and S, at 8 ppm for pb and Cu and 5 ppm for other measured trace elements [12].

## **RESULTS AND DISCUSSION**

#### 1-Geochimical analyses:

The concentration of the trace elements in (ppm) for the samples under investigation is shown in figure (1) which represent high concentration in Ni, Zn, V and Co, samples 2S and 17S show high concentration in Pb (1065 and 4088.96 ppm), respectively. Samples A1and A4 has high concentration of Zn, Sr, Zr, Ba, Pb, U, V and Cr. While the samples 2G and 5G have high concentration in Rb, Zr, Cr.



Arab J. Nucl. Sci. Appl., Vol. 54, 3, (2021)





Fig. (1): Trace elements content (ppm) in the studied samples

#### 2- Radiometric Measurements

The distribution of radionuclides during samples processing includes, distributions in original samples, leachate (pregnant solution) and residual as shown in Tables (1-10), which are analyzed by gamma ray- detector. This difference in gamma activity is sometimes +ve (the sum is greater than the original) and in other time-ve (the sum is lower than the original). This difference is varied mostly with the variation of geochemical composition of the rock samples.

The activity concentration of <sup>238</sup>U in the original samples, residuals and pregnant solutions are higher than the typical world average value of 33Bq/kg [13].

The behavior of each radionuclide in the samples shows that the  $^{226}$ Ra Activity concentration is higher in the residual than that in the original for all samples. Also, the activity concentration of  $^{226}$ Ra in the original, residual and solution are higher than the typical world average value of 32Bq/kg [13].

For <sup>232</sup>Th all the original samples are higher than the permissible value of 45 Bq/kg. Also, the residuals samples are higher than the permissible level 45 except sample 17S. For <sup>40</sup>K the original samples are higher than the permissible level 412 Bq/kg except samples 2S, 7S, A3 and A4, while the residual samples are higher than the original in all samples [13].

The summation of the activities of the measured radionuclides of <sup>238</sup>U in solution and residual is less than original as shown in samples 3S and7S. From figure (1), it can be concluded that these samples contain low concentration of Pb (798.08 and 204.16 ppm) respectively. While the summation of the activities of the measured radionuclides of <sup>238</sup>U in samples 2S, 17S, A1, A2, A3, A4, 2G and 5G are higher than the original. In these samples the lead (Pb) content is 1065, 4088.96, 3961.13, 3965.91, 3852.12 3854.85, 3858.36 and 1102.08 ppm respectively.

Table (1): Activity concentrations of radionuclides in original<br/>samples, residuals and leachates (pregnant<br/>solutions) for sample No. 7S (claystone).

Radionuclide	Original	Solution	Leachability	Residual	Summation (Residual+Solution)/		
	(Bq/Kg)	( <b>Bq/l</b> )	(%)	(Bq/Kg)	Original (%)		
<sup>238</sup> Useries							
<sup>234</sup> Th	2941.5	865.9	29.437	3097.4	134.737		
<sup>234m</sup> Pa	3318.5	518.3	15.618	3045.2	107.382		
Average	3130.0	692.15	22.113	3071.3	120.238		
<sup>234</sup> U	333.2	390.95	11.728	1124.3	45.459		
<sup>230</sup> Th	2264.7	1330.55	5.897	1565.7	75.032		
<sup>226</sup> Ra subseries							
<sup>226</sup> Ra	2448.7	62.96	2.5711	3230.8	134.510		
<sup>214</sup> Pb	2081.9	19.18	0.921	2164.21	104.874		
<sup>214</sup> Bi	1983.2	20.61	1.039	1898.1	96.748		
<sup>235</sup> U	155.8	34.5	22.143	70.95	67.682		
<sup>232</sup> Th series							
<sup>228</sup> Ac	59.2	8.53	14.408	54.17	105.912		
<sup>208</sup> Tl	59.14	7.51	12.698	45.12	88.992		
Average	59.19	8.02	13.549	49.64	97.415		
$^{40}$ K	352.5	106.6	30.241	609.46	203.137		

<b>Table (2):</b>	Activity c	oncentration	is of ra	ndionuclides	in original
	samples,	residuals	and	leachates	(pregnant
	solutions)				

Table (4) :	Activity concentrations of radionuclides in original
	samples, residuals and leachates (pregnant
	solutions) for sample 3S (Siltstone).

			-		
Radionuclide	Original	Solution	Leachability	Residual	Summation (Residual+Solution)/
	(Bq/Kg)	(Bq/l)	(%)	(Bq/Kg)	Original (%)
<sup>238</sup> Useries					
<sup>234</sup> Th	3724.3	1212.32	32.551	3862.79	136.270
<sup>234m</sup> Pa	4103.02	1304.78	31.860	4907.85	151.416
Average	4103.02	1258.55	30.673	4385.33	137.554
<sup>234</sup> U	4198.98	760.619	18.114	3276.07	96.134
<sup>230</sup> Th	3722.82	250.653	6.732	1631.01	50.548
<sup>226</sup> Ra subseries					
<sup>226</sup> Ra	4102.93	20.252	0.493	6048.52	147.913
<sup>214</sup> Pb	3322.8	36.215	1.089	3860.75	117.279
<sup>214</sup> Bi	3232.6	40.930	1.266	3299.06	103.322
<sup>235</sup> U	190.01	54.755	28.816	161.95	114.052
<sup>232</sup> Th series					
<sup>228</sup> Ac	73.94	7.366	9.962	18.54	35.036
<sup>208</sup> Tl	57.54	5.259	9.139	29.047	59.621
Average	65.74	6.313	9.602	23.795	45.798
$^{40}$ K	523.9	106.509	20.330	759.155	165.234

Та

	105.254	157.155	20.550	100.507	525.7	IX I			
Table	Table (3) :Activity concentrations of radionuclides in originalsamples, residuals and leachates (pregnantsolutions) for sample No. 2S (Shale).								
Radionu	Summation (Residual+Solution/	Residual	Leachability	Solution	Original	Radionuclide			
	Original (%)	(Bq/Kg)	(%)	( <b>Bq/l</b> )	(Bq/Kg)				
<sup>238</sup> User						<sup>238</sup> Useries			
<sup>234</sup> Tl	158.305	7754.0	32.418	1996.8	6159.5	<sup>234</sup> Th			
<sup>234m</sup> P	114.412	5735.7	25.950	1682.6	6483.8	<sup>234m</sup> Pa			
Avera	135.794	6744.8	29.101	1839.7	6321.7	Average			
<sup>234</sup> U	134.548	6917.2	21.662	1327.4	6127.6	<sup>234</sup> U			
<sup>230</sup> Tl	232.121	10426	12.459	591.4	4746.4	<sup>230</sup> Th			
<sup>226</sup> R subser						<sup>226</sup> Ra subseries			
<sup>226</sup> R	159.809	7711.9	0.534	25.9	4841.9	<sup>226</sup> Ra			
<sup>214</sup> Pl	129.786	5536.3	0.4159	17.8	4279.4	<sup>214</sup> Pb			
<sup>214</sup> B	121.112	4945	0.7109	29.20	4107.1	<sup>214</sup> Bi			
<sup>235</sup> U	116.976	288.82	29.136	95.8	328.8	<sup>235</sup> U			
<sup>232</sup> Th se						<sup>232</sup> Th series			
<sup>228</sup> A	175.370	91.99	11.073	6.2	55.99	<sup>228</sup> Ac			
<sup>208</sup> T	118.115	64.93	11.182	6.79	60.72	<sup>208</sup> Tl			
Avera	145.604	78.46	11.139	6.50	58.35	Average			
<sup>40</sup> K	222.429	619.42	41.735	143.07	342.8	<sup>40</sup> K			

Radionuclide	Original	Solution	n Leachability Residu		Summation (Residual+Solution)/			
	(Bq/Kg)	(Bq/l)	(%)	(Bq/Kg)	Original (%)			
<sup>238</sup> Useries								
<sup>234</sup> Th	2610.67	759.19	29.080	2394.32	120.793			
<sup>234m</sup> Pa	2032.92	568.12	27.946	1656.06	109.408			
Average	2321.79	663.6	28.581	1656.06	99.908			
<sup>234</sup> U	3946.52	513.14	13.002	1346.25	47.1146			
<sup>230</sup> Th	2230.04	92.3	4.138	2972.27	137.422			
<sup>226</sup> Ra subseries								
<sup>226</sup> Ra	2385.84	15.5	0.649	2645.42	111.529			
<sup>214</sup> Pb	2099.3	8.6	0.409	2008.26	96.073			
<sup>214</sup> Bi	2001.7	2.79	0.139	1807.02	90.413			
<sup>235</sup> U	114.81	28.58	24.893	94.68	107.359			
<sup>232</sup> Th series								
<sup>228</sup> Ac	67.86	1.42	2.092	116.16	173.269			
<sup>208</sup> Tl	68.55	5.12	7.469	81.67	126.608			
Average	68.2	3.27	4.794	98.91	149.83			
$^{40}$ K	562.4	92.12	16.379	906.37	177.540			

(5): Activity concentrations of radionuclides in original samples, residuals and leachates (pregnant solutions) for Wadi Um Hamd sample No. A1.

Radionuclide	Original (Bq/Kg)	Solution (Bq/l)	Leachability (%)	Residual (Bq/Kg)	Summation (Residual+Solution)/ Original (%)
<sup>238</sup> Useries					
<sup>234</sup> Th	8670.9	4880.5	56.28	8630.8	155.823
<sup>234m</sup> Pa	8580.8	4700.8	54.78	8650.9	155.599
Average	8625.85	4790.65	55.53	8640.85	155.712
<sup>234</sup> U	8540.71	7620.8	89.22	9962.7	205.87
<sup>230</sup> Th	8625.89	3441.9	39.90	12140.72	180.64
<sup>226</sup> Ra subseries					
<sup>226</sup> Ra	10015.5	750.7	7.49	20440.9	211.58
<sup>214</sup> Pb	8910.9	165.91	1.86	19655.8	222.44
<sup>214</sup> Bi	8890.1	160.82	1.80	18140.7	205.86
<sup>235</sup> U	400.10	210.8	52.68	330.9	135.39
<sup>232</sup> Th series					
<sup>228</sup> Ac	60.7	6.1	10.05	65.4	117.79
<sup>208</sup> Tl	58.9	5.9	10.016	60.1	112.05
Average	59.8	6	10.033	62.75	114.92
<sup>40</sup> K	815.7	90.9	11.14	1194.2	157.54

Arab J. Nucl. Sci. Appl., Vol. 54, 3, (2021)

Radionuclide	Original (Bq/Kg)	Solution (Bq/l)	Leachability (%)	Residual (Bq/Kg)	Summation (Residual+Solution)/ Original (%)
<sup>238</sup> Useries					
<sup>234</sup> Th	8690.9	4885.9	56.22	8840.12	157.935
<sup>234m</sup> Pa	8595.8	4710.8	54.80	8885.55	158.174
Average	8643.35	4798.35	55.51	8862.83	158.054
<sup>234</sup> U	8570.12	7635.9	89.09	9980.80	205.55
<sup>230</sup> Th	8655.52	3455.1	39.91	12160.32	180.406
<sup>226</sup> Ra subseries					
<sup>226</sup> Ra	10050.52	775.32	7.714	20455.82	211.244
<sup>214</sup> Pb	8950.18	185.21	2.069	19675.21	221.899
<sup>214</sup> Bi	8899.13	185.81	2.087	18165.15	206.209
<sup>235</sup> U	410.58	230.12	56.047	340.21	138.90
<sup>232</sup> Th-series					
<sup>228</sup> Ac	62.91	6.12	9.728	70.22	121.34
<sup>208</sup> Tl	60.89	5.51	9.049	66.81	118.771
Average	61.9	5.81	9.388	68.515	120.055
$^{40}$ K	825.12	98.35	11.919	1210.1	158.576

Table (6):	Activity	concentration	s of ra	adionuclides	in original
	samples,	residuals	and	leachates	(pregnant
	solutions	) for Wadi Uı	n Han	nd sample N	o. A2.

Table (8): Activity concentrations of radionuclides in original<br/>samples, residuals and leachates (pregnant<br/>solutions) Wadi Sad El Banat Sample No.A4.

Radionuclide	Original	iginal Solution Leachability Resid		Residual	Summation (Residual+Solution)/
	( <b>Bq/Kg</b> ) (		(%)	(Bq/Kg)	Original (%)
<sup>238</sup> Useries					
<sup>234</sup> Th	1070.51	60.21	5.62	1670.81	161.70
<sup>234m</sup> Pa	1030.10	61.35	5.95	1410.25	142.85
Average	1050.30	60.78	5.78	1540.53	152.27
<sup>234</sup> U	1238.22	88.12	7.11	3080.53	255.9
<sup>230</sup> Th	998.85	37.10	3.71	1697.25	173.63
<sup>226</sup> Ra subseries					
<sup>226</sup> Ra	1220.15	19.18	1.57	1725.18	142.96
<sup>214</sup> Pb	1098.12	12.15	1.10	1530.21	140.45
<sup>214</sup> Bi	1085.28	15.81	1.45	1395.55	130.04
<sup>235</sup> U	48.9	3.5	7.15	75.82	162.20
<sup>232</sup> Th series					
<sup>228</sup> Ac	112.54	5.21	4.62	118.71	110.1
<sup>208</sup> Tl	105.92	5.42	5.09	116.82	115.3
Average	109.23	5.31	4.85	117.76	112.7
$^{40}$ K	187.58	85.55		328.85	220.9

 Table (7): Activity concentrations of radionuclides in original samples, residuals and leachates (pregnant solutions) for Wadi Sad El Banat Sample No. A3.

Table (9): Activity concentrations of radionuclides in original samples, residuals and leachates (pregnant solutions) for granite sample No. 2G.

Radionuclide	Original	Solution	Leachability	Residual	Summation (Residual+Solution)/	Radionuclide	Original	Solution	Leachability	Residual	Summation (Residual+solution)/
	(Bq/Kg)	(Bq/l)	(%)	(Bq/Kg)	Original (%)		(Bq/Kg)	( <b>Bq/l</b> )	(%)	(Bq/Kg)	Original (%)
<sup>238</sup> Useries						<sup>238</sup> Useries					
<sup>234</sup> Th	1060.8	51.4	4.84	1660.3	161.35	<sup>234</sup> Th	2989.57	872.83	29.196	3076.61	132.107
<sup>234m</sup> Pa	1015.7	50.8	5.00	1401.7	143.00	<sup>234m</sup> Pa	2363.53	727.29	30.771	2543.2	138.373
Average	1038.25	51.1	4.92	1531	152.17	Average	2676.55	800.05	29.891	2809.9	134.87
<sup>234</sup> U	1230.8	82.5	6.70	3060.9	255.40	<sup>234</sup> U	2397.13	1336.35	55.747	8155.31	395.959
<sup>230</sup> Th	991.5	25.4	2.56	1681.81	172.18	<sup>230</sup> Th	2269.87	483.74	21.311	3870.3	191.819
<sup>226</sup> Ra subseries						<sup>226</sup> Ra subseries					
<sup>226</sup> Ra	1210.61	10.12	0.835	1715.81	172.18	<sup>226</sup> Ra	2258.98	2.502	0.1107	4293.01	190.153
<sup>214</sup> Pb	1092.6	8.70	0.796	1518.2	139.74	<sup>214</sup> Pb	1925.9	17.90	0.926	3936.71	205.338
<sup>214</sup> Bi	1078.2	10.89	1.00	1382.7	129.24	<sup>214</sup> Bi	1901.6	17.92	0.942	3651.32	192.955
<sup>235</sup> U	45.73	2.4	5.24	70.8	166.42	<sup>235</sup> U	115.14	33.00	28.664	54.52	76.021
<sup>232</sup> Th series						<sup>232</sup> Th series					
<sup>228</sup> Ac	104.71	4.20	4.01	112.81	111.74	<sup>228</sup> Ac	88.74	3.90	4.398	132.39	153.596
<sup>208</sup> Tl	103.30	5.10	4.93	114.8	116.06	<sup>208</sup> Tl	74.58	12.71	17.043	56.72	93.106
Average	104.00	4.65	4.47	113.80	113.90	Average	801.66	8.30	10.172	94.56	125.971
<sup>40</sup> K	181.21	80.82	44.60	322.31	222.96	$^{40}$ K	1195	100.65	8.422	2096.44	183.857

Arab J. Nucl. Sci. Appl., Vol. 54, 3, (2021)

Table (10): Activity concentrations of radionuclides in original samples, residuals and leachates (pregnant solutions) for granite sample No. 5G.

Brunte bumple 10. 50.								
Radionuclide	Original	Solution	Leachability	Residual	Summation (Residual±solution)/			
	(Bq/Kg)	( <b>Bq/l</b> )	(%)	(Bq/Kg)	Original (%)			
<sup>238</sup> Useries								
<sup>234</sup> Th	2705.37	950.55	35.135	3076.77	148.863			
<sup>234m</sup> Pa	2660.02	992.98	37.329	2749.6	140.697			
Average	2682.69	971.76	36.223	2913.18	144.815			
<sup>234</sup> U	2958.02	1330.17	44.968	7016.07	282.156			
<sup>230</sup> Th	2549.74	772.15	30.283	3906.2	183.483			
<sup>226</sup> Ra subseries								
<sup>226</sup> Ra	2316.73	23.569	1.017	4100.64	178.018			
<sup>214</sup> Pb	2020.1	34.486	1.707	3868.76	193.220			
<sup>214</sup> Bi	2016.2	47.268	2.344	3474.90	174.693			
<sup>235</sup> U	123.7	42.455	34.320	56.20	79.754			
<sup>232</sup> Th series								
<sup>228</sup> Ac	95.14	13.411	14.096	112.24	132.072			
<sup>208</sup> Tl	83.1	25.317	30.465	56.48	98.432			
Average	89.12	19.364	21.728	84.361	116.388			
$^{40}$ K	1264	189.480	14.990	189.48	180.540			

Table (11) represents the relation between the gamma attenuation and (Pb) concentration. From the previously results, it can be concluded that Pb played its role as a good attenuated element for gamma radiation. The  $\gamma$ -attenuation increases with the increase in the Pb-concentration in the measured sample.

 Table (11): The relation between the gamma attenuation and (pb) concentration.

Sample No.	Original- <sup>238</sup> U	Residual- <sup>238</sup> U	Solution- <sup>238</sup> U	Summation- Original	Attenuation Factor (%)	Pb Concentration
	(Bq/Kg)	(Bq/Kg)	( <b>Bq/l</b> )	Ū		(ppm)
2S	509.8	543.9	148.36	182.46	35.79	1865.28
17S	230.88	273.0	101.49	143.61	62.2	1688.96
A1	695.63	696.84	386.34	387.55	55.71	2061.13
A2	697.04	714.74	386.96	404.66	58.05	2060.91
A3	83.73	123.47	4.12	43.86	52.38	3854.12
A4	84.70	124.24	4.9	44.44	52.47	3852.85
2G	190.60	226.6	64.52	100.52	52.74	1111.36
5G	216.35	234.93	78.37	96.95	44.81	1102.08

The high leaching efficiency % present in sample A1 where leachability for  $^{234}$ U (89.22%) and for  $^{238}$ U (55.53%).  $^{238}$ U and  $^{234}$ U have the same chemical behavior, So 33.69% of  $^{234}$ U was transferred physically by  $\alpha$ - recoil effect [14].

Also, in sample A2  $^{234}$ U have leachability (89.09%) and  $^{238}$ U have (55.51%). So (33.58%) of  $^{234}$ U was transferred physically. The low leachability in samples A3 and A4 for  $^{238}$ U 4.92% and 5.78% respectively. For  $^{234}$ U 6.7% and 7.11% respectively. So A3 has physically transferred 1.78% and A4 has 1.33% physically transferred.

In 2G  $^{234}$ U has leachability 55.74% and  $^{238}$ U has 29.89%, so the physically transferred 22%. In Sample 5G the leachability of  $^{234}$ U 44.96% and for  $^{238}$ U 36.22, so the transferred physically by  $\alpha$ - recoil 8.74%.

From leaching studies have shown that isotopes are leached to the same extent but that is not observed in  $^{238}$ U, hence the difference between  $^{234}$ U and  $^{238}$ U leaching could represent the  $^{234}$ U  $\alpha$ - recoil as shown in table (12).

 Table (12): Chemical and physical transfer for <sup>234</sup>U for studied sample.

Sample No.	Chemical Transfer (%)	Physical Transfer (%)	
A1	55.53	33.69	
A2	55.51	33.58	
A3	4.92	1.78	
A4	5.78	1.33	
2G	33.74	22	
5G	36.22	8.74	

### CONCLUSION

It is noticed that, the activities of the residual are higher than the activity of the solution. There is difference in  $\gamma$ - activity between the summation of activities of residual and solution with the activity of the original sample. The difference in  $\gamma$ - activity is sometimes –ve and in other time +ve, that its means summation is greater than the original. This phenomenon is due to the attenuation of the gamma activities during the leaching processes, for existence a high concentration of lead (Pb). The leaching processes by using acid solution may have lead to clean the grain surface and permit the gamma activities of the inner grains to be measured. Also, the residual have high concentration of  $^{226}$ Ra in all samples. The  $^{235}$ U has the same leachability as  $^{238}$ U but in leaching process find that  $^{234}$ U has leachability higher than  $^{238}$ U due to  $\alpha$ -recoil. So the leachability of  $^{234}$ U is divided into chemical transfer (%) physical transfer (%) due to  $\alpha$ -recoil.

# REFERENCES

- E. Svoukis, and H. Testertos, "Indoor and outdoor in situ high -resolution gamma radiation measurements in urban areas of Cyprus, Radiat. Prot.Dosim., Vol.123, pp. 384-390, (2006).
- [2] E. M. Ibrahim, "Behavior of Radionuclides in U-Series During Processing of Uranium Bearing Rock Materials", Ph.D. Thesis, Faculty of Women for Art, Science and Education, Physics Department, Egypt, (2011).
- [3] D.R. Metzler, (2004), Uranium Mining: Environmental Impact, Encyclopedia of Energy, Vol. 6, pp. 299-315, (2004).
- [4] I.E. El Aassy, M.G. El Feky, M.A. El Kasaby, E. M. Ibrahim, S. Sewefi and R. M. Attia, "Behavior of Radionuclides during Acidic Leaching Processes of Different Rock Materials, Allouga Locality, Southwestern Sinai, Egypt", International Journal of Scientific & Engineering Research, V. 8, Issue 1, ISSN 2229-5518, (2017).
- [5] I.E El Aassy, A.A. Nada, M.M. El Galy, M.G. El Feky, T.M. Abd El Maksoud, S.M. Talaat and E.M. Ibrahim, "Behavior and Environmental Impacts of Radionuclides during the Hydrometallurgy of Calcareous and Argillaceous Rocks, Southwestern Sinai, Egypt", Applied Radiation and Isotopes, Vol. 70, pp. 1024-1033, (2012).
- [6] R. Tykva and D. Berg, "Man-Made and natural radioactivity in environmental pollution and radio

chronology". Klwer Academic, P.O. Box 17, 3300 AA Dordecht, Netherlands, (2004).

- [7] G. Gilmore and J.D. Hemingway, "Practical Gamma-Ray Spectrometry". John Wiley & Sons, Chichester, (2008).
- [8] IAEA, Preparation and certification of IAEA gamma spectrometry reference materials, RGU-1, RGTh-1 and RGK-1, Report-IAEA/RL/148, International Atomic Energy Agency, (1987).
- [9] R.M. Anjos, R. Veiga, T. Soares, A.M.A. Santos, J.G. Aguia, Frasca' MHBO, D. Brage JAP, Uze'da, L. Mangia L., A. Facure, B. Mosquera, C. Carvalho , Gomes PRS, "Natural radionuclide distribution in Brazilian commercial granites", Radiat Meas Vol. 39, pp. 245–253, (1987).
- [10] S. Turhan and L. Gu"ndu"z, "Determination of specific activity of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K for assessment of radiation hazards from Turkish pumice samples", J. Environ Radioact Vol. 99, pp. 332–342, (2008).
- [11] R.N. Nair, F. Sunny and S.T. Manikandan, "Modelling of Decay Chain Transport in Groundwater from Uranium Tailings Ponds", Applied Mathematical Modelling, Vol. 34, pp. 2300-2311, (2010).
- [12] K. Gravani, C. Papachristodoulou, A. Oikonomouand K. Ioannidies, "A study of Ancient Pottery by Means of X-ray Fluorescence, Multivariate Statics and Mineralogical Analysis", Analytic Chemical Acta, pp. 347-353, (2006).
- [13] UNSCEAR, United Nations Scientific Committee on the Effect of Atomic Radiation, "Sources and Effects of Ionizing Radiation". Report to General Assembly with Scientific Annexes A and B, E. 10XI. 3., United Nations, New York, (2010).
- [14] W. Jinlong, Du. Jinzhou and Bi. Qianqian, "Natural Radioactivity Assessment of Surface Sediments in the Yangtze Estuary", Marine Pollution Bulletin, Vol. 114, pp. 602–608, (2017).