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Factors Controlling Radionuclides Migration within Different Media

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Abstract: *This study aims to examine the mobility of uranium and its isotopes and daughters during the acid leaching. In the same context the study of the daughter / parent ratios in the three phases, original sample, leach liquor and residue will gave a complete picture about the behavior during weathering and alterations of these rocks. Five samples of different rock types were collected and prepared for various analyses. Wet chemical analyses were carried out for major oxides and trace elements. Natural radionuclides were measured by HPGe detector in original samples, pregnant solution and residuals. The results showed that the relation between the sum of activities of ^{238}U in both pregnant solutions and residuals with originals have two modes. The first mode is represented by the sum of ^{238}U activities in leachate and residual is nearly equal the activity in the original sample. In the second mode the sum is higher than the original, sometimes twice or more. The well noticed thing the samples of the second mode show high content of Pb (2060-4343ppm).*
The behavior of ^{226}Ra in the acidic leaching is completely different from ^{238}U and its isotopes. Selective leaching studies have shown that uranium isotopes are leached to the same extent but that is not observed in ^{234}U. The lowest leachability in all the samples is present in ^{226}Ra, ^{214}Pb and ^{214}Bi.

Keywords: *Radionuclides, HPGe detector, Sediment rock, Leaching.*

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I. Introduction

The ^{238}U, ^{235}U, and ^{232}Th are the parents of the three natural decay series, each of these series consists of many daughter products generated through successive decay of parent radionuclides. Natural uranium is a composite of the isotopes ^{238}U (99.28%), ^{235}U (0.72%) and ^{234}U (0.0057%) **[Powell et al., 2007].** These isotopes have the same geochemical behavior. While on a mass basis there is far more ^{238}U than ^{235}U in a natural sample, the activity ratio is approximately 21.7 **[Porcelli and Swarzenski, 2003].**

The radioactive series reaches secular equilibrium when the activity ratios of daughter/parent are equal to unity **[Wally El-Dine, 2008].** Radioactive disequilibrium arises in the natural radioactive series due to recent fractionation events (continuous or instantaneous processes) which lead to the loss or gain of radionuclides that are mobile in the ambient environmental conditions. The long-lived radionuclides in the natural radioactive series, are generally used to assess the state of radioactive disequilibrium **[Sam, 2000].**

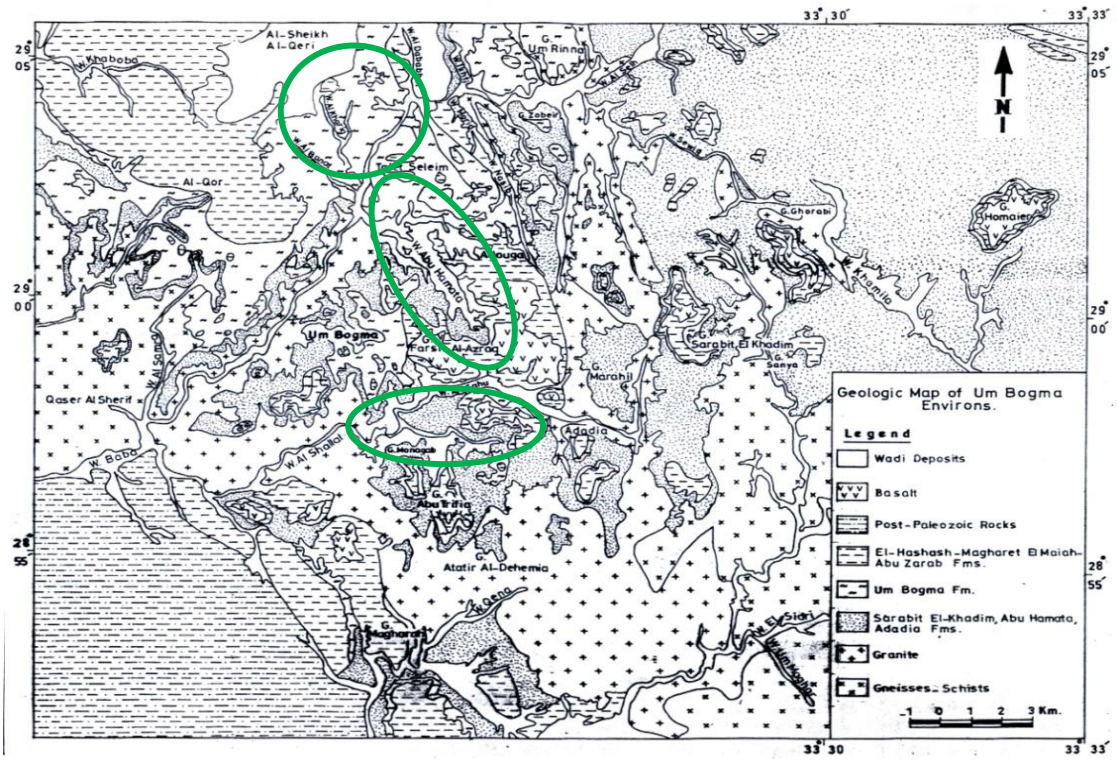
The physical and chemical processes used to extract uranium from ore, such as crushing and acid treatment, produce large amounts of mill tailings. In France today, approximately 50 million tons of uranium mill tailings (UMTs) are stored on the surface in specific areas. They contain 99% of the radium present in the original ore and have a much higher porosity and permeability than that of the rock from which they are derived. The average ^{226}Ra activity value for French UMTs is between 4000 and 60,000 Bq kg^{-1} **[Ferry et al., 2002].**

Acid leaching is known to be predominant process for uranium recovery from ores, usually with sulfuric acid because its relatively low cost **[Umanskii and Klyushnikov, 2012].**

II. Materials and methods

1. Study area

The study area **(Fig.1)** is located in the southwestern Sinai, Egypt. Where the samples collected from three localities; Wadi Nasab is at the intersection of longitude 33° 26' 20" and latitude 29° 2', Wadi Sad El-Banat is at the intersection of longitude 33° 24' and latitude 29° 2' 30" and Wadi Um Hamd is at the intersection of longitude 33° 25' 45" and latitude 28° 57'.



(Fig.1): Geologic map of the studied area in southwestern Sinai, Egypt **[After El Aassy et al., 1986].**

2. Sampling.

Five sedimentary rock samples were collected from three different locations **(Fig.1)** and different rock types; three samples from Wadi Nasab, one sample from Wadi Sad El Banat and one sample from Wadi Um Hamd. These samples were prepared for $\gamma - Ray$ spectrometric analyses by HPGe detector where the samples first dried, crushed and sieved through -60 mesh size. Weighted samples were placed in polyethylene bottles of 250 cm^3 volume. The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reached before measured by the gamma spectrometer. This step was necessary to ensure that radon gas is confined within the volume and the daughters still also remain in the sample. About 5 g of each sample was ground to -200 mesh for chemical analyses of major oxides and trace elements content, by the X-ray fluorescence technique (XRF).

3. Radioactivity measurements

High purity vertical germanium was coupled to a PC-computer with a special electronic card to make it equivalent to a multichannel analyzer. The system also contains the usual electronic components of preamplifier, amplifier and power supply. The detector was surrounded by a special heavy lead shield of about 10 cm thickness with inside dimension 28 cm diameter 40.5 cm height. 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 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 **[Pekala et al., 2010].** The primordial ^{238}U is the most abundant isotope of U (99.27 %) and the initial member of the ^{238}U-decay chain with a long half-life time (4.4683 Ga). It decays to ^{234}Th with the emission of the α - particle. Through two consecutive β-transitions, ^{234}Th decays to ^{234}Pa (half-lives of 24.10 days and 6.69 h, respectively) and to ^{234}U, with the half-life time of 245,250 years, which decays to ^{230}Th **[Pekala et al., 2010].** Uranium-238 activity was determined indirectly from the gamma rays emitted by its daughter products (^{234}Th and ^{234m}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 **[Technical Reports Series, 1989].** ^{226}Ra activity concentration was measured from 186.1 KeV (3.29 %) after the subtraction of the 185.7 KeV (54 %) of ^{235}U. The concentrations of ^{214}Pb and ^{214}Bi were measured from (295.1(18.7%), 351.9 (35.8%)) KeV and (609.3 (45%), 1120.3(14.9%) ,1238.1(5.96%), 1764.5(16.07%)) KeV. For the actinium series gamma energies of 143.8 KeV (10.5 %), 163.4 KeV (4.8 %), 185.7 KeV (54 %) and 205.3 KeV (4.7%) were taken to represent the ^{235}U concentrations. ^{40}K was

determined directly from the 1460 KeV (10.7%) peak energy. ^{234}U activity was determined directly from the gamma rays emitted from this nuclide at energies of 53.2 (0.123%) keV and 120.9 (0.034%) keV. For the measurement of the ^{230}Th activity, the γ-ray emission at 67.7 (0.37%) keV is used **[Technical Reports Series, 1989].**

4. Leaching Procedure

The γ −measured samples were poured from the containers and prepared for leaching experiments, using H_2SO_4 (Lab. grade) acid leaching on 150g sample weight under the conditions; solid/liquid ratio 1:3, acid concentration (110gm/L), stirring time two hours and at room temperature. Filtration was carried out to separate leachate from residual which is dried at room temperature. The residual and leachate have been weighted and packed will in polyethylene bottles of 250 ml volume and stored for more than a month for measuring by HPGe detector. The leachability of the radionuclides was calculated according to the following equation:

$$\text{Leachability}\ (\%) = \frac{\text{Activity concentration in leachate}}{\text{Activity concentration in original}} \times 100 \qquad (1)$$

III. Results and Discussion

3.1. Geochemical Characterization of Samples

Major oxides for the analyzed samples are shown in **table (1),** for samples K1 and K3, it is obviously that high level of the loss on ignition attaining about 30.5% and 34.5%, as both contains high carbonate content, while K3 is characterized also by high SO_3 (7.3%). Sample K14 is characterized by high alumina (19.1%) and iron (22.8%). Sample K15 is characterized by high SO_3 content (8.2%). Sample K21 is characterized by high Al_2O_3 (22.9%) and K_2O + Na_2O contents (6.4%).

Table (1): The concentration of the Major elements in (%) for the samples.

Locality / Major oxides	Wadi Nasab			Wadi Sad El-Banat	Wadi Um Hamd
	K1 (Calcareous shale)	**K3 (Calcareous shale)**	**K14 (Claystone)**	**K15 (Siltstone)**	**K21 (Siltstone)**
SiO_2	15.89	12.63	19.876	32.956	29.337
Al_2O_3	6.824	4.304	19.145	6.787	22.911
TiO_2	0.741	0.675	1.269	3.4	2.216
CaO	24.533	21.216	0.96	5.775	0.446
MgO	7.704	5.954	3.147	0.816	4.76
MnO	0.674	1.159	0.154	0.013	0.042
Fe_2O_3	9.005	9.867	22.848	22.335	18.575
Na_2O	0.182	0.068	3.966	2.567	2.825
K_2O	1.019	0.82	1.372	0.573	3.575
P_2O_5	0.031	0.03	0.852	0.297	0.258
SO_3	1.973	7.313	0.193	8.197	0.414
L.O.I	30.5	34.3	19	13.3	10.2

From **table (2)** It's clear that for Wadi Nasab location, sample K1 contain interesting values of Cu and U ; namely more than 3180 and 599 ppm respectively, Sample K3 contain interesting values of Cu, Ag and U ; namely more than 1735, 14.75 and 244 ppm respectively. The analyses of sample K14 have revealed the presence of interesting values of Zn, Pb, V, Sr, As, REEs and U; namely more than 26916, 4343, 3018, 1022, 183, 5187 and 101 ppm respectively. Sample K15 which is siltstone, represents Wadi Sad El-Banat showed the presence of interesting values of Cu, Zr, Sr, Pb, V, As, REEs and U ; namely more than 1101, 1531, 2619, 3851.2, 4748, 324, 827 and 72 ppm respectively. Sample K21 which is siltstone type, represent Wadi Um Hamd location, the analyses have revealed the presence of interesting values of V, Cr, Pb, Ba, Zn, Zr, REEs and U; namely more than 8360, 3604, 2060, 1557, 4231, 1287, 652 and 663 ppm respectively.

Table (2): The concentration of the trace elements in (ppm) for the selected samples for leaching process.

Locality	Wadi Nasab			Wadi Sad El-Banat	Wadi Um Hamd
Element	K1 (Calcareous shale)	K3 (Calcareous shale)	K14 (claystone)	K15 (siltstone)	K21 (siltstone)
Cr	91.88	107.69	244.34	574.56	3604.68
Co	331.12	292.19	344.42	102.26	102.41
Ni	161.13	136.60	816.4	-	219.8
Cu	3183.76	1735.52	981.54	1101.24	750.12
Zn	122.99	69.04	26916.56	248.93	4231.81
Zr	165.08	156.81	629	1531.8	1287.6
Rb	23.36	25.83	47.72	15.75	130.55
Nb	17.03	20.59	27.82	167.76	63.12
Ba	89.16	51.98	110.64	104.18	1557.3
Pb	76.88	74.29	4343.04	3851.2	2060.16
Sr	59.82	53.50	1022.45	2619.5	929.5
Ga	7.16	8.05	14.89	16.92	26.98
V	220.79	213.37	3018.4	4748.8	8360.8
As	63.72	72.08	183.08	324.37	120.45
Mo	18.68	20.1	67.24	486.18	59.91
Ag	11.65	14.75	8.82	8.75	8.85
Cd	6.05	6.18	21.86	1.3	8.71
I	18.3	18.69	27.75	35.65	25.34
Sc	14.71	23.54	12.98	18.58	16.22
Y	28.86	26.53	766.29	80.36	141.82
La	41.75	60.14	289.27	221.71	137.74
Ce	60.4	78.87	879.44	351.42	235.93
Nd	29.52	30.22	2338.25	131.73	100.26
Sm	18.03	12.28	838.64	22.36	20.56
Yb	0.74	-	62.51	1.36	-
Tl	0.67	6.16	7.91	6.24	9.2
Bi	1.49	2.47	5.90	5.13	5.49
Th	10.48	8.44	16.41	26.72	30.2
U	599.52	244.03	101.89	72.36	663.98

3.2. Radiometric Measuremrnts

The results of γ-detection of radionuclides in the original samples (Bq/Kg), residuals (Bq/Kg) and leachates (Bq/l) are collected in **table (3).** The activity concentration of ^{238}U in the original samples, residuals and pregnant solutions are higher than the typical world average value of 33 Bq/Kg **[UNSCEAR, 2010]**.

On the other hand the behavior of ^{226}Ra in the acidic leaching is completely different from ^{238}U and its isotopes. It is almost 7 times lower than the leachability of ^{238}U and ^{234}U under the same conditions in all samples. In this case most of the dissolved ^{226}Ra reacted with the available SO_4^{2-} to form $RaSO_4$ (precipitate) also due to solubility difference of the two elements **[Megumi and Mamuro, 1977].** So most of the ^{226}Ra remains in the leachate ore (residuals) **[Fernandes et al., 2006].** The authors (op.cit) point to the tailings dam, waste rock piles can constitute an important source of pollutants to the environment. The $RaSO_4$ will precipitate on the ore and accumulated on the residual to form an environmental problem. The behavior of each radionuclide in the samples shows that the ^{226}Ra, ^{214}Pb and ^{214}Bi activity concentrations are higher in the residual than that in the original for all samples. The activity concentration of ^{226}Ra in the original samples, residuals and pregnant solutions are higher than the typical world average value of 32 Bq/Kg, **[UNSCEAR, 2010]** except the pregnant solutions K14 and K15.

For ^{232}Th, all the original samples and the residuals are higher than the permissible level 45 Bq/Kg except residuals of K1 and K3. For ^{40}K, the original samples (K14and K21) and the residual (K14 and K21) are higher than the permissible level 412 Bq/Kg. The activity concentration of ^{232}Th and ^{40}K for all the pregnant solutions are lower than the typical world average.

3.3. Leaching Experiments

Leaching experiments were performed under conditions that approximated natural conditions. From **table (3),** it is clear that, there is a difference in γ-activity between the summation of activities of residual and leachates with the activity of the original sample. it can be noticed that the relations between the activities of radionuclides in original samples and the summation of them in the pregnant solutions and residuals are different from one sample to another especially in the nuclides of ^{238}U decay series. These differences can be categorized in two groups:

1. The summation of activities of the measured radionuclides before ^{226}Ra in the ^{238}U decay series in pregnant solution and residual is nearly around 100% (93% - 106%) except ^{234}U as in K1and K3 (shales), but from ^{226}Ra to ^{214}Bi are higher than the same radionuclides in the original sample.
2. The summation of activities of the measured radionuclides in the pregnant solution and residual is higher than the activities of the originals as in the samples K14 (claystone), K15 and K21 (siltstones). The variations are very high in the ^{226}Ra subseries.

From the previously mentioned results, it can be concluded that the geochemistry of each sample **(Table 2)** plays its role in these variations as in the following:

1. In the first case, the two samples K1 and K3 contain low concentration of Pb (76.88 and 74.29 ppm).
2. In the second case, the samples K14, K15 and K21 have high concentration of Pb (4343, 3851 and 2060 ppm respectively).

From these two notices, it can be concluded that Pb played its role as a good attenuated element for γ-radiations and this very clear in the measurements of original samples (K14, K15 and K21) and their leachates and residuals. The behavior of each radionuclide is noticed very well within the same sample during the leaching process.

The highest leachability (leaching efficiency %) present in sample K21 **(Table 3)**, where it is (89.2%) for ^{234}U, while the ^{235}U is 54.72% and ^{238}U is 55.6%. but ^{234}U, ^{235}U and ^{238}U have similar chemical behavior, and should therefore 33.6% of ^{234}U was transferred physically by the α-recoil effect. The lowest leachability presents in sample K15, where it is 5.14% and 5.23% for ^{238}U and ^{235}U, respectively, also 1.72% of ^{234}U transferred physically.

The lowest leachability in all the samples is present in ^{226}Ra, ^{214}Pb and ^{214}Bi. The lowest release (leachability) of ^{214}Pb and ^{214}Bi of the all samples is coinciding with the studies of **[White and Brantley, 2003]** which showed that the behavior of Pb isotopes is heavily affected by the early breakdown of U-rich minerals.

The more release of ^{234}U and the variability of ^{234}U/^{238}U was observed before by several authors whom mentioned that the release of excess ^{234}U may originate both from preferential release from damaged lattice sites **[Bourdon et al., 2009]** and direct α-recoil **[Kigoshi, 1971; Depaolo et al., 2006]**. However, it remains under which of the two mechanisms is most important **[Andersen et al., 2009]**.

Table (3): Activity concentrations of radionuclides in original samples, residuals and leachates (pregnant solutions).

Radionuclide	K1(Calcareous Shale) Wadi Nasab							
	Original (Bq/Kg)		Solution (Bq/l)		Leachability (%)	Residual (Bq/Kg)		Summation (Residual+solution) (%)
^{238}Useries								
^{234}Th	8067.56	±27.9	1636.48	±11.1	20.28	6924.72	±43.4	106.12
^{234m}Pa	8039.53	±186.4	1627.02	±52.5	20.24	6935.64	±265.2	106.51
Average	**8053.55**	±107.2	**1631.75**	±31.8	**20.26**	**6930.18**	±154.3	106.31
^{234}U	**6843.91**	**±1586**	**1694.5**	±152.1	**24.76**	**5945.26**	±164.5	111.63
^{230}Th	7239.35	±180	812.3	±42.9		7478.4	±752.4	114.52
^{226}Ra subseries								
^{226}Ra	**7637.09**	±21.9	**110.33**	±1.7	**1.44**	**13318.4**	±50.7	175.83
^{214}Pb	5184.2	±9.9	68.53	±1.2	1.32	10976.4	±23.1	213.05
^{214}Bi	5065.27	±25.6	69.33	±2.8	1.37	10149.8	±58.1	201.75
^{235}U	**371.21**	±7.3	**73.53**	±2.5	**20.0**	**318.36**	±11.1	105.57
^{232}Th series								
^{228}Ac	30.57	±4.5	2.22	±0.5	7.28	27.27	±3.4	96.49
^{208}Tl	29.66	±2.0	-	-	0	28.29	±3.6	95.38
Average	**30.12**	±3.3	**2.23**	±0.5	**7.4**	**27.78**	±3.5	99.63
^{40}K	**211.16**	±11.8	**80.76**	±3.6		**373.27**	±23.5	215.0
^{238}U/^{235}U	21.696		22.191			21.768		
^{234}U/^{238}U	0.849		1.038			0.858		
^{226}Ra/^{238}U	0.948		0.068			1.922		

Radionuclide	K3(Calcareous shale) Wadi Nasab							
	Original (Bq/Kg)		Solution (Bq/l)		Leachability (%)	Residual (Bq/Kg)		Summation (Residual+solution) (%)
^{238}Useries								
^{234}Th	3764.45	±33.6	1009.09	±9.5	26.81	2498.49	±30.5	93.18
^{234m}Pa	3700.58	±273.7	930.28	±83.4	25.14	2493.19	±226.1	92.51
Average	**3732.52**	±153.7	**969.69**	±46.5	**25.98**	**2495.84**	±128.3	92.85
^{234}U	**4747.17**	±402.5	**1982.55**	±80.9	**41.76**	**5037.53**	±151.3	147.88
^{230}Th	11544.3	±168.2	1985.04	±57.7		9361.24	±204.5	98.28
^{226}Ra subseries								
^{226}Ra	**12040.1**	±36.8	**252.84**	±9.4	**2.1**	**13377.6**	±53.1	113.21
^{214}Pb	6681.92	±12.0	120.27	±3.9	1.8	9286.23	±21.5	140.77
^{214}Bi	6443.97	±31.1	111.16	±9.6	1.73	8548.65	±54.0	134.38
^{235}U	**176.62**	±7.4	**44.78**	±2.4	**25.35**	**116.09**	±8.8	91.08
^{232}Th series								
^{228}Ac	29.59	±3.2	4.64	±0.4	15.68	24.77	±4.4	99.39
^{208}Tl	28.05	±2.3	4.91	±0.7	17.5	24.24	±3.2	103.92
Average	**28.82**	±2.7	**4.77**	±0.5	**16.55**	**24.51**	±3.8	101.59
^{40}K	**342.6**	±20.1	**122.15**	±6.0		**322.03**	±18.7	129.65
^{238}U/^{235}U	21.133		21.653			21.499		
^{234}U/^{238}U	1.272		2.045			2.018		
^{226}Ra/^{238}U	3.226		0.363			5.36		

Radionuclide	K14(claystone) Wadi Nasab							
	Original (Bq/Kg)		Solution (Bq/l)		Leachability (%)	Residual (Bq/Kg)		Summation (Residual+solution) (%)
^{238}Useries								
^{234}Th	1433.94	±16.5	129.74	±3.1	9.05	1923.79	±21.8	143.21
^{234m}Pa	1414.85	±80.4	129.29	±17.4	9.14	1725.52	±134.8	131.1
Average	**1424.39**	±48.4	**129.52**	±10.2	**9.09**	**1824.66**	±78.3	137.19
^{234}U	**1321.51**	±207.8	**143.16**	±40.7	**10.8**	**4732.56**	±184.1	368.95
^{230}Th	1456.42	±99.8	37.18	±15.9		2707.96	±159.2	188.49
^{226}Ra subseries								
^{226}Ra	**1989.93**	±15.9	**25.03**	±0.9	**1.26**	**2611.61**	±22.7	132.5
^{214}Pb	1313.74	±6.1	11.27	±0.6	0.86	1601.67	±8.9	122.77
^{214}Bi	1259.78	±16.1	14.79	±1.0	1.17	1477.85	±22.5	118.48
^{235}U	**66.025**	±3.5	**6.1**	±0.4	**9.24**	**81.27**	±5.1	132.33
^{232}Th series								
^{228}Ac	52.6	±2.7	-	-	-	70.69	±3.8	134.39
^{208}Tl	52.33	±2.1	5.05	±0.4	9.65	64.94	±2.7	133.75
^{212}Bi	50.003	±5.6	-	-	-	-	-	-
Average	**51.71**	±3.4	**5.05**	±0.4	**9.78**	**67.82**	±3.2	140.92
^{40}K	**469.76**	±13.7	**99.02**	±3.5		**667.76**	±20.0	163.23
^{238}U/^{235}U	21.57		21.225			22.452		
^{234}U/^{238}U	0.928		1.105			2.594		
^{226}Ra/^{238}U	1.397		0.193			1.431		

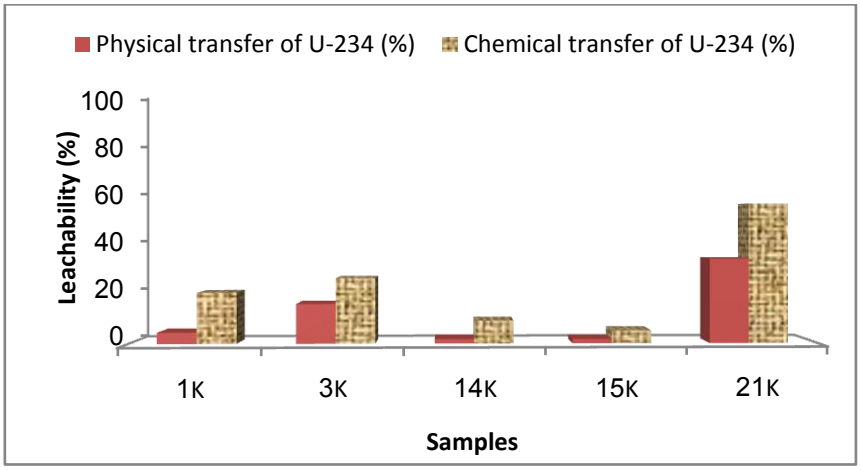
Radionuclide	K15 (siltstone) Wadi Sad El Banat							
	Original (Bq/Kg)		Solution (Bq/l)		Leachability (%)	Residual (Bq/Kg)		Summation (Residual+solution) (%)
^{238}Useries								
^{234Th}	1063.35	±13.9	55.07	±2.1	5.18	1663.13	±21.9	161.58
^{234m}Pa	1020.06	±61.9	52.09	±13.4	5.11	1408.52	±114.5	143.19
Average	**1041.71**	±37.9	**53.58**	±7.7	**5.14**	**1535.83**	±68.2	152.58
^{234}U	**1233.1**	±71.8	**84.55**	±27.4	**6.86**	**3077.48**	±142.3	256.43
^{230Th}	994.29	±82.7	27.5	±9.9		1692.13	±167.0	172.95
^{226}Ra subseries								
^{226}Ra	**1215.53**	±10.5	**13.31**	**±0.7**	**1.09**	**1718.54**	±18.7	142.48
^{214}Pb	1096.56	±4.9	9.47	±0.4	0.86	1521.1	±8.7	139.58
^{214}Bi	1078.27	±12.6	13.25	±0.8	1.23	1389.43	±21.5	130.09
^{235}U	**47.95**	±3.5	**2.509**	±0.3	**5.23**	**72.106**	±3.7	155.61
^{232}Th-series								
^{228}Ac	107.45	±3.5	4.1	±0.5	3.82	114.77	±4.5	110.63
^{208}Tl	103.35	±2.1	5.35	±0.3	5.18	115.28	±3.2	116.72
^{212}Bi	107.46	±6.3	-	-	-	-	-	-
Average	**106.09**	±2.8	**4.72**	±0.4	**4.45**	**115.03**	±3.9	112.88
^{40}K	**182.71**	±8.9	**81.65**	±2.8		**324.32**	±15.2	222.19
^{238}U/^{235}U	21.724		21.354			21.299		
^{234}U/^{238}U	1.184		1.578			2.004		
^{226}Ra/^{238}U	1.167		0.248			1.119		

Radionuclide	K21 (siltstone) Wadi Um Hamd							
	Original (Bq/Kg)		Solution (Bq/l)		Leachability (%)	Residual (Bq/Kg)		Summation (Residual+solution) (%)
^{238}Useries								
^{234Th}	8678.9	±30.6	4891.5	±17.6	56.36	7234.97	±36.7	139.72
^{234m}Pa	8582.77	±111.1	4706.7	±75.5	54.84	7270.68	±247.8	139.55
Average	**8630.84**	±70.8	**4799.1**	±46.5	**55.6**	**7252.83**	±142.3	139.64
^{234}U	**8550.92**	±342.9	**7627.4**	±303.5	**89.2**	**9975.87**	±1322	205.86
^{230Th}	8640.25	±155.7	3449.4	±65.0		12151.62	±311.2	180.56
^{226}Ra subseries								
^{226}Ra	**10020.2**	±20.5	**753.81**	±4.6	**7.52**	**20445.23**	±63.2	211.56
^{214}Pb	8920.13	±8.6	173.93	±1.6	1.95	19667.95	±26.5	222.44
^{214}Bi	8891.01	±23.0	173.35	±4.3	1.95	18145.29	±65.7	206.04
^{235}U	**403.26**	±5.6	**220.68**	±3.2	**54.72**	**332.92**	±12.6	137.28
^{232}Th-series								
^{228}Ac	63.09	±3.1	5.96	±0.7	9.45	68.04	±5.9	117.29
^{208}Tl	61.58	±1.8	5.32	±0.6	8.64	65.03	±4.4	114.24
Average	**62.34**	±2.4	**5.64**	±0.5	**9.05**	**66.54**	±5.2	115.78
^{40}K	**819.37**	±12.8	**95.32**	±4.0		**1196**	±25.4	157.6
^{238}U/^{235}U	21.403		21.75			21.79		
^{234}U/^{238}U	0.991		1.589			1.375		
^{226}Ra/^{238}U	1.161		0.157			2.819		

Selective leaching studies have shown that uranium isotopes are leached to the same extent but that is not observed in ^{234}U, hence the difference between ^{234}U and ^{238}U leaching percent could represent the ^{234}U percent that resulted from α-recoil as shown in **table (4)** and **(Fig.2)**. The difference in the resulting values may be related to the rock type **[El-Aassy et al., 2017].**

Table (4): Transferring of ^{234}U by chemical leaching and physical α-recoil.

Location	Rock Type	Sample	^{234}U Chemical transfer (%)	^{234}U Physical transfer (%)
Wadi Nasab	Calc. Shale	K1	20.26	4.5
	Calc. Shale	K3	25.98	15.78
	Claystone	K14	9.09	1.74
Wadi Sad El-Banat	Siltstone	K15	5.14	1.72
Wadi Um Hamd	Siltstone	K21	55.6	33.6



(Fig.2): Chemical and physical transfer of ^{234}U to leachate.

IV. Conclusions

It is noticed that, there is a difference in γ-activity between the summation of activities of residual and pregnant solution with the activity of the original sample. This phenomenon is varied in magnitude within the different radionuclides. The type of sample plays its role in these variations. Sometimes, there is an attenuation of the gamma activities during the leaching process, due to the presence of high Pb concentration.

The behavior of radionuclides (^{238}U, ^{235}U, ^{234}U, ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{232}Th and ^{40}K) was studied during the acid leaching process, the nuclides before ^{226}Ra in the ^{238}U decay series are easily released in the pregnant solutions

than the ^{226}Ra itself and the nuclides after it (^{214}Pb and ^{214}Bi), these are concentrated in the residuals. The ^{235}U has nearly the same leachability as ^{238}U, while ^{232}Th is immobile except in the presence of organic matters.
The uranium isotopes are leached to the same extent but that the same is not observed between ^{234}U and ^{238}U due to the effect of α-recoil phenomenon.

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