

INVESTIGATION OF SIX GEOLOGICAL SAMPLES FROM WADY SITRA EASTERN DESERT - EGYPT USING k_0 NEUTRON ACTIVATION METHOD

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k_0 - Neutron Activation Analysis (k_0 -NAA) is applied to investigate six geological samples collected from Wady Sitra at Eastern desert of Egypt during the survey of gold presence in this area. The samples together with a group of standard (Mo, Fe, Sb and W) are irradiated for 3 hours in one of the inner irradiation site of the Second Egyptian Training and Research Reactor (ET-RR-2) operating at power of 19 MW. Mo, Fe and Sb are used to measure the neutron spectrum parameters α (epithermal non-ideality factor) and f (the thermal to epithermal flux ratio) while W is used to test the obtained results. The α and f parameters are measured using the so-called bare triple monitor method and the obtained results was found to be $f = 17.5 \pm 0.35$ and $\alpha = 0.03 \pm 0.002$. A Fortran computer program is designed and used to calculate the values of $Q_0(\alpha)$ for the analyzed elements . The concentration values of 25 elements in the present rock samples have been presented.

Keywords: *Neutron Activation Analysis, k_0 Standardization, Reactor Parameters, Activation Analysis ,Geological Samples.*

INTRODUCTION

Mining in Egypt has had a long history that goes back to pre-dynastic. Egypt has substantial mineral resources, including 48 million tons of tantalite (fourth largest in the world), 50 million tons of coal, and an estimated 6.7 million ounces of gold. The Eastern Desert of Egypt has more than 90 gold deposits and occurrences [1-2] are spread over the whole area covered by the basement rocks of Precambrian age, see the map Fig.1 [3]. Wadi Hammamat and Bir Umm Fawakhir along with Wady Sid, Wady

Abbed, Wady El-Hudi. Also copper mines at Eastern desert include Araba, Wady Sitra, the Hamash area. Gold is a less-common metal, so it is necessary to apply very susceptible methods having a high precision for Au concentration to be found in geological samples at searching, exploration and mining. As an example of a successful decision of this problem it can be pointed applying the neutron- activation techniques [4-6]. Also determination of rare earth elements (REEs) in geological samples are useful indicator of various surface geological processes and can assists geochemists in research on the processes involved in the formation of different rocks through the analysis of the rare earth elements (REEs) and other trace elements. In addition to modeling geochemical processes, other applications include location of ore deposits and tracking elements of environmental importance.

Experimental nuclear reactors are the most efficient neutron sources for high sensitivity activation analysis induced by thermal and epithermal neutrons and also for numerous scientific investigations. But because there are significant variations in the neutron flux levels and the spectra at different locations; therefore an accurate knowledge of spectral characteristics of the irradiation facility is crucial for the accuracy of the applied methods. The k_0 standardization method is used for precisely determination of elemental concentration [7-9] of the constituents. For a correct application of k_0 -standardized NAA, the knowledge of the thermal-to-epithermal flux ratio, f , and of the epithermal neutron flux shape parameter α , is of great importance [10-13]. In the present work, the parameters f and α are determined using the so called bare triple monitor method [14-15].

Single Comparator Method (k_0 -Standardization Method).

The k_0 - factors for the majority of the elements that can be determined as:

$$k_{0,Au}(a) = \frac{(N_p / wt_m SDC)_a}{(N_p / wt_m SDC)_{Au}} * \frac{\varepsilon_{p,Au}}{\varepsilon_{p,a}} * \frac{f + Q_{0,Au}(\alpha)}{f + Q_{0,a}(\alpha)} \quad (1)$$

Once the k_0 values have been determined, the concentration of an element σ_a can be obtained by co-irradiation of a sample and a suitable flux monitor (comparator) as Au-monitor

$$\rho_a = \frac{(N_p / Wt_m SDC)_a}{(N_p / wt_m SDC)_{Au}} * \frac{1}{k_{0,Au}(a)} * \frac{\varepsilon_{p,Au}}{\varepsilon_{p,a}} * \frac{f + Q_{0,Au}(\alpha)}{f + Q_{0,a}(\alpha)} * 10^6 \quad ppm \quad (2)$$

where the subscript "Au" refers to the co-irradiated gold monitor and N_p is the net number of counts under the full-energy gamma-ray peak measured in t live time, W

is the weight of the sample, w is the weight of the gold monitor, t_m is the measurement real time, $S = (1 - e^{-\lambda t_{irr}})$, λ is the decay constant, t_{irr} is the irradiation time, $D = e^{-\lambda t_d}$, t_d is the decay time, $C = \{(1 - e^{-\lambda t_m}) \lambda t_m\}$, f is the thermal to epithermal flux ratio, ε is the absolute full energy gamma-ray peak efficiency and $Q_0 = \frac{I_0}{\sigma_0}$ is the ratio between the resonance neutron integral and the thermal neutron cross-section at 2200 m/s, α is the deviation in the real epithermal neutron spectrum $(\frac{1}{E^{1+\alpha}})$ from the ideal $(\frac{1}{E})$ law.

EXPERIMENTAL

Sample Preparation

Wady Sitra occurrence in central Eastern desert of Egypt located at latitude of $(25^{\circ} 31' 0.01")$ and longitude $(34^{\circ} 25' 0.01")$ are chosen for our survey. Six serpentinites rocks samples from this area are collected and grounded into fine powder using an agate mortar and homogenized to search for gold and rare earth elements.

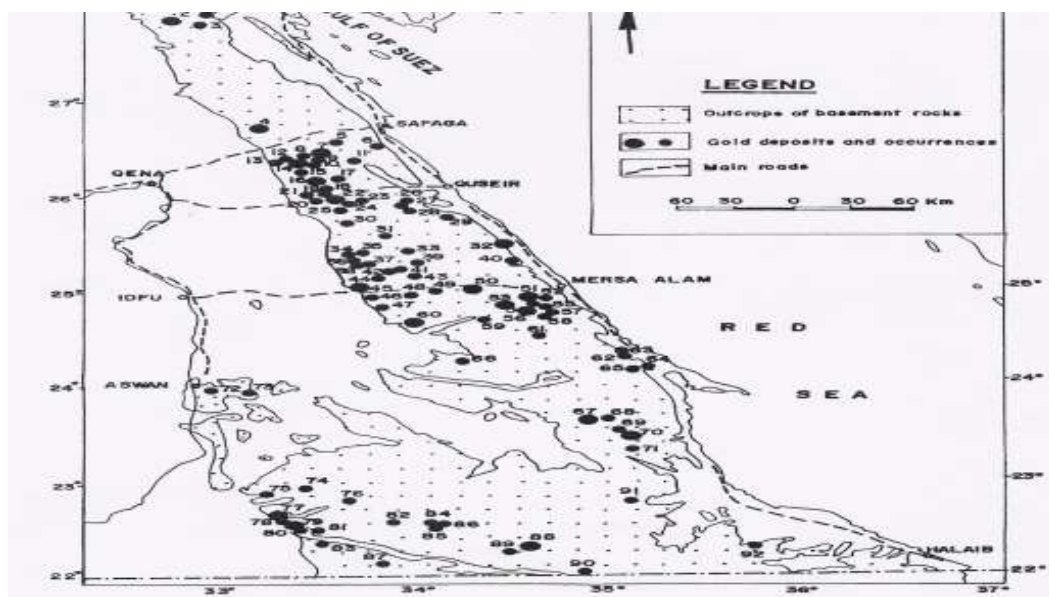


Figure 1. A map showing the position of gold mining in Eastern Desert

Irradiation and Gamma-Ray Measurements

Standard samples of Fe, Sb, Mo, W, Au and six geological samples from Wadi Sitra at Eastern Desert of Egypt are enveloped in thin aluminum foil. All samples and an empty weighted aluminum foil are placed in a can. The can is located inside an irradiation box at the periphery of the ET-RR-2 reactor core. The samples were irradiated for 3 hours power reactor. The standard samples Mo, Sb and Fe are used to calculate the neutron spectrum parameters α and f while W is used to test the method. All flux monitors are irradiated as thin or dilute wires to minimize the neutron self shielding effects. The gamma rays spectra collected using a Canberra vertical p-type HPGe detector with relative efficiency 40%, 1.9 keV FWHM at 1.332 MeV of ^{60}Co . The peak areas were determined interactively with the Genie spectroscopy software package of Canberra. The detector is calibrated using a certified calibration gamma sources, ^{137}Cs (662 keV), ^{60}Co (1173 and 1332.51 keV), ^{133}Ba (276.29, 302.71, 355.86 and 385.7 keV) and ^{22}Na (1274 keV).

To eliminate the subtraction of the background gamma-ray of thin aluminum foil, an empty foil is weighed and irradiated in the same container and analyzed separately as a back ground concentrations

Qualitative and Quantitative Analysis

A Fortran computer program is constructed to calculate the $Q_0(\alpha)$ and another Excel worksheet is constructed to calculate the concentration of 26 elements in the geological samples. To obtain more high precision results, the net peak areas for all isotopes are corrected for the dead time, measuring time and the decay of the radioisotopes during the gamma ray measurements.

To study the impact of the value of α on the values of concentrations, the elemental concentration of one sample is calculated by using two values of α ($\alpha = 0$, and $\alpha =$ the measured value -0.03).

RESULTS AND DISCUSSION

Due to the significant variations in the neutron flux levels and the neutron spectra at different locations in the reactor core, it is important to know the knowledge of spectral characteristics at the present irradiation position. α and f are the important irradiation parameters in the k_0 -NAA and are determined experimentally using the so-called bare triple monitor method [14-15] the reactor parameters can be determined from the intersection of the plots f versus α .

Table (1) gives the results for different combinations of pairs of three isotopes for the neutron spectrum parameters α (the deviation of the non-ideal $1/E^{1+\alpha}$ epithermal

neutron spectrum of irradiation position and f (the thermal to epithermal flux ratio) which plotted as shown in fig. (2).

Table 1. The results f and α determination for different combinations of pairs of three isotopes: Sb, Fe, Mo.

Suggested Values of α	Sb-Fe	Fe-Mo	Sb-Mo
-0.01	10	19.95	11.49
-0.02	13.25	18.98	14.1
-0.03	16.74	17.8	16.9
-0.04	20.49	16.4	19.88
-0.05	24.52	14.75	23.07
-0.06	28.85	12.82	26.47
-0.07	33.49	10.59	30.1
-0.08	38.48	8.017	33.96
-0.09	43.83	5.079	38.08

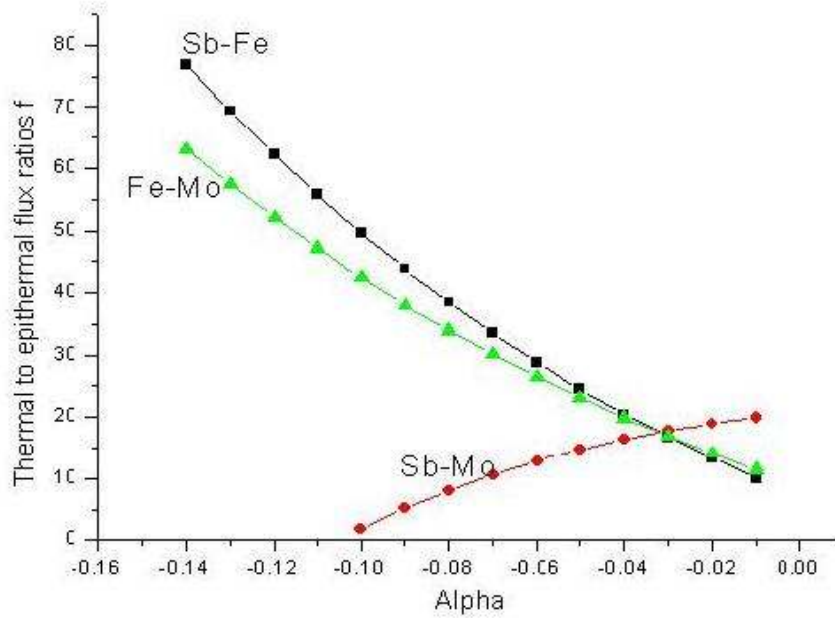


Figure 2. The deviation of the non-ideal epithermal neutron spectrum of irradiation position versus the thermal to epithermal flux ratio

As shown in fig. (2), the intersection of three curves gives a unique solution for $f = 17.5 \pm 0.35$ and $\alpha = 0.03 \pm 0.002$

The experimental value determination of α is used to calculate $Q_0(\alpha)$ for all isotopes which investigated in this work by using the following equation [16].

$$Q_0(\alpha) = \left(\frac{Q_0 - 0.429}{(E_r)^\alpha} + \frac{0.429}{(2\alpha + 1)(E_{cd})^\alpha} \right) (1eV)^\alpha \quad (3)$$

where E_{cd} is the effective Cd cut-off energy (=0.55 eV in standard conditions) and E_r is the effective resonance energy. Table (2) indicates the calculated values of $Q_0(\alpha)$ for all isotopes present in the samples under investigation compared by Q_0 values.

Table 2. List the values of the tabulated Q_0 and the calculated $Q_0(\alpha)$ for $\alpha = -0.03$

Product Isotope	($\alpha = -0.03$)	($\alpha = 0$)	Standard Deviation	Population Standard deviation
⁴⁶ Sc	63.50	62.01	1.054	0.745
⁵¹ Cr	695	693.12	1.329	0.940
⁵⁹ Fe	1200	1118	57.9	41
⁶⁰ Co	20	19.63	0.262	0.185
⁶⁵ Zn	826	822.86	2.220	1.570
⁷⁵ Se	21	21.3	0.212	0.150
⁸⁵ Sr	7297	7756	325.127	230
⁹⁵ Zr	6198	6439	170.554	121
¹⁰³ Ru	3809	3814	3.535	2.5
^{114m} In	150	151.17	0.827	0.585
¹²⁴ Sb	3.1	3.20	0.070	0.05
¹⁴⁰ La	71	69.74	0.89	0.63
¹⁴¹ Ce	34	33.7	0.212	0.15

¹⁴⁷ Nd	46.0	45.9	0.707	0.50
¹⁵³ Sm	33	33.01	1.054	0.745
¹⁵² Eu	1.08	1.06	0.014	0.01
¹⁶⁰ Tb	22.1	22.53	0.304	0.215
¹⁷⁵ Yb	28	27.35	0.460	0.325
¹⁷⁷ Lu	2	2.943	0.042	0.03
¹⁸¹ Hf	445	441	2.828	2
¹⁸⁶ Re	6799	6749	0.034	0.024
^{187m} Hg	77.5	75.72	1.259	0.89

For a 1/v thermal cross-section behavior such as (¹⁷⁶Lu (n,γ) ¹⁷⁷Lu , ¹⁵¹Eu (n,γ) ^{152m}Eu, ¹⁵¹Eu(n,γ) ¹⁵²Eu and ¹⁶⁸Yb (n,γ) ¹⁶⁹Yb) one has a good approximation $Q_0 = ((3.14)^{0.5}/2)(s_0+0.484)$ [16]. Where s_0 is the Westcott's factor. In order to test the applied techniques W (known its weight and purity) is used. Table (3) present the results of the calculated concentration using K_0 -NAA method ($\alpha = -0.03$) compared by its certificated values.

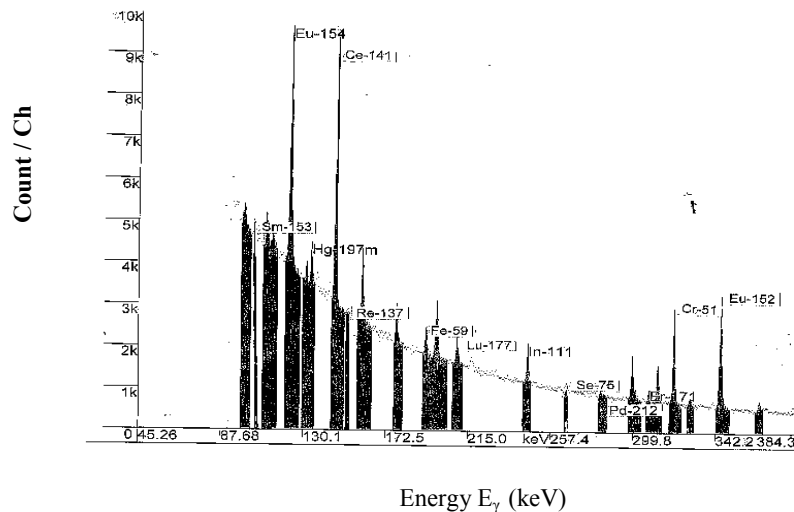


Figure 3-a. A portion of the gamma-ray spectrum of the sample code114a (in the energy range 45.26 to 384.3 keV)

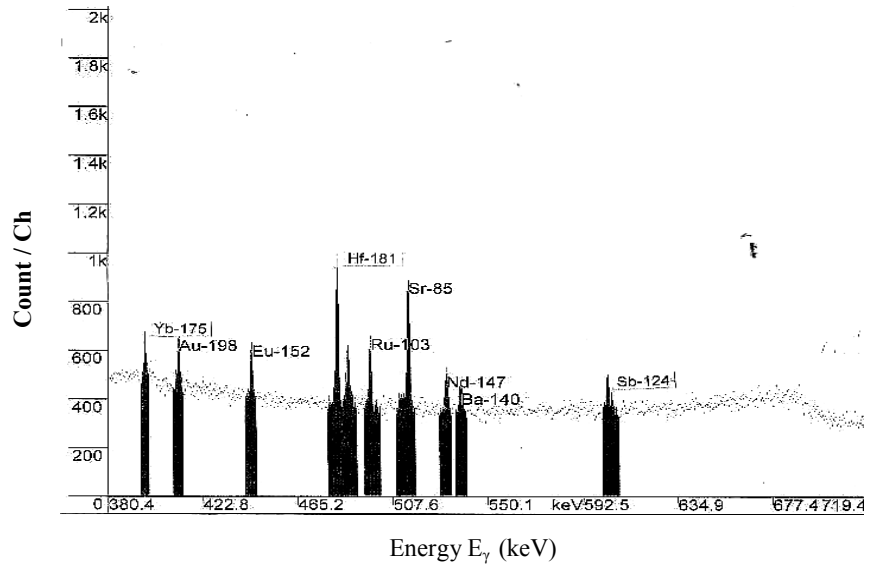


Figure 3-b. A continuity of gamma-ray spectrum (in the energy range 380.4 to 719.4 keV)

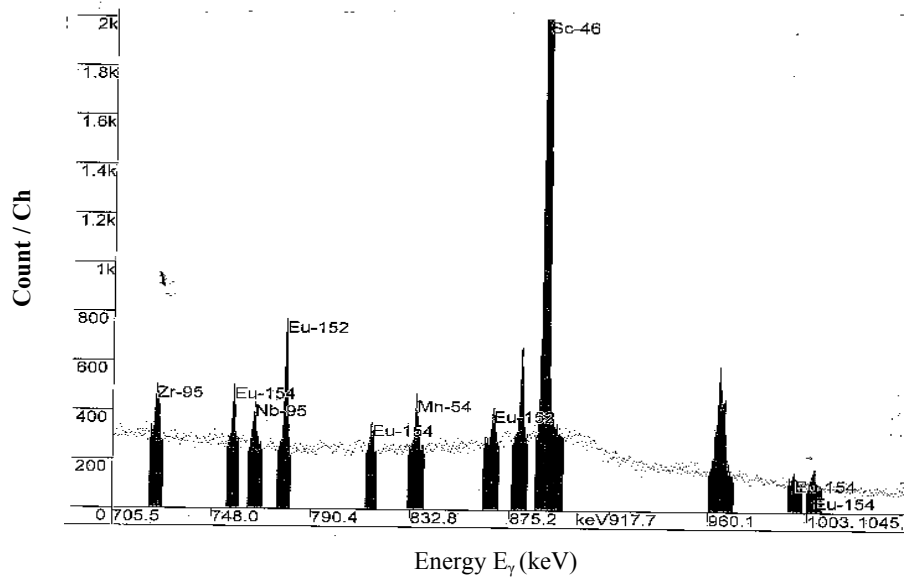


Figure 3-c. A continuity of gamma-ray spectrum (in the energy range 705.5 to 1045 keV)

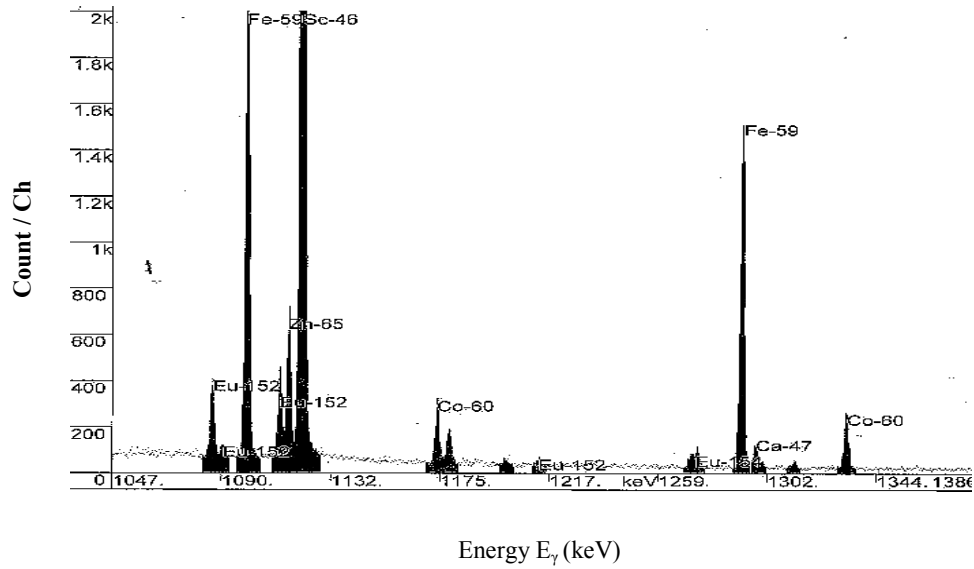


Figure 3-d. A continuity of gamma-ray spectrum (in the energy range 1047 to 1386 keV)

To study the effect of the epithermal non-ideality factor (α) on the concentration values, the elemental concentrations are calculated for two values of α ($\alpha = 0$, $\alpha = - 0.03$). Table (3) indicates the elemental concentration of sample 114a for two vales of α .

Table 3. List of elemental concentration of sample 114a using $\alpha = 0$ and $\alpha = -0.03$

Product Isotope	($\alpha = - 0.03$)	($\alpha = 0$)	Standard Deviation	Population Standard deviation
⁴⁶ Sc	63.50	62.01	1.054	0.745
⁵¹ Cr	695	693.12	1.329	0.940
⁵⁹ Fe	1200	1118	57.9	41
⁶⁰ Co	20	19.63	0.262	0.185
⁶⁵ Zn	826	822.86	2.220	1.570
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¹⁴⁰ La	71	69.74	0.89	0.63
¹⁴¹ Ce	34	33.7	0.212	0.15
¹⁴⁷ Nd	46.0	45.9	0.707	0.50
¹⁵³ Sm	33	33.01	1.054	0.745
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¹⁸¹ Hf	445	441	2.828	2
¹⁸⁶ Re	6799	6749	0.034	0.024
^{187m} Hg	77.5	75.72	1.259	0.89

Table (3) indicates that the position of irradiation, represented in the value of (α) will affect the value of the concentrations in certain degree depends on flux ratio.

With the introduction of high-resolution HPGe γ -ray detectors many trace elements, including the REE, could be determined without any chemical separations. A total of 25 elements were determined by k_0 -NAA. The identified radio-nuclides and their concentrations are shown in table (4).

The gamma line selected for a particular case was either the most abundant one or the line with good combination of peak area and freedom from spectral interferences. ¹⁵²Eu has photo peaks free from interference at 799 keV is, another peak at 122 keV cannot be resolved from the 124 keV line of ¹⁵⁴Eu are not used. Ytterbium can be determined using the 396.3 keV peak of ¹⁶⁹Yb and for Lutetium, the high abundance peak at 208.4 keV of ^{177m}Lu was used. In addition the results of Cr and Fe values are corrected due to the interference of ⁵⁰Cr(n, γ)⁵¹Cr and ⁵⁴Fe(n, γ)⁵⁵Fe reactions [17].

Table 4. List of the elemental concentration of the six investigated samples in ppm.

<i>Code106_c</i>	<i>Code 110 a</i>	<i>Code110c</i>	<i>Code110 f</i>	<i>Code114b</i>	<i>Code 114a</i>	<i>Identified gamma ray energy</i>	<i>Product Isotope</i>
15±0.23	11.00 ±0.17	12.5±0.2	12.00 ±0.12	62.50 ±0.01	63.5±0.63	889.3& 1120.5	⁴⁶ Sc
6101 ±79	2821±42	5746±86	N.D.	123.0±1.8	695±10	320.1	⁵¹ Cr
1100±15	800±13	8000±80	1100±15	1100±15	1200± 18	1099,1291, 192.3	⁵⁹ Fe
213±32	135.0±1.3	207±3	18.41 ±0.24	34.0±0.5	20.00 ±0.23	1173.23& 1332.5	⁶⁰ Co
244.0 ±2.6	290.0±4.7	323.0±4.8	223±6	276.0±4.4	826.0±8.3	1115.5	⁶⁵ Zn
N.D.	N.D.	N.D.	N.D.	N.D.	21.0±0.2	264.65	⁷⁵ Se
N.D.	N.D.	N.D.	N.D.	N.D.	7297±73	513.99	⁸⁵ Sr
N.D.	N.D.	N.D.	N.D.	N.D.	6198±62	724.18& 756.72	⁹⁵ Zr
N.D.	N.D.	N.D.	N.D.	915±12	3809±38	497.08	¹⁰³ Ru
N.D.	N.D.	N.D.	N.D.	N.D.	150.0±1.5	190.3	^{114m} In
9.50 ±0.09	1.10±0.01	1.40±0.01	0.140 ±0.001	N.D.	3.10±0.03	602.7	¹²⁴ Sb
N.D.	N.D.	N.D.	N.D.	N.D.	71.00 ±1.42	487	¹⁴⁰ La
28.00 ±0.36	2.70±0.04	45.00 ±0.54	0.04±0.00	28.00 ±0.33	34.0±1.1	145.4	¹⁴¹ Ce
N.D.	N.D.	N.D.	0.7	N.D.	46.0±4.5	531	¹⁴⁷ Nd
0.50 ±0.05	N.D.	23.00 ±0.32	N.D.	N.D.	33.0±0.4	103.2	¹⁵³ Sm
N.D.	N.D.	0.16±0.00	N.D.	N.D.	1.08±0.01	799	¹⁵² Eu
N.D.	N.D.	N.D.	N.D.	2.70±0.08	22.10 ±0.48	965.1	¹⁶⁰ Tb
N.D.	N.D.	N.D.	N.D.	4.70±0.05	28.00 ±0.61	396.3	¹⁷⁵ Yb
N.D.	N.D.	N.D.	N.D.	1.30±0.04	2.00±0.02	208.4	¹⁷⁷ Lu
49.0 ±0.5	41.00 ±0.04	2.70±0.03	0.50±0.01	25.6±0.4	445.0±4.4	482.16	¹⁸¹ Hf
102.4 ±1.1	74.0±1.1	N.D.	9.2±1.8	N.D.	N.D.	1121.3	¹⁸² Ta
N.D.	N.D.	N.D.	N.D.	N.D.	6799±68	155	¹⁸⁶ Re
N.D.	N.D.	N.D.	N.D.	N.D.	6.70±0.07	411.8	¹⁹⁸ AU
N.D.	N.D.	N.D.	N.D.	0.5	77.5±0.8	133.95	^{197m} Hg

CONCLUSION

The irradiation reactor parameter f and α at the irradiation position are calculated by bare triple monitor method and used to calculate the elemental concentrations of each samples. The standard samples W and is used to check the used technique. Using this method, concentrations of 25 elements including some REEs (La, Ce, Nd, Sm, Eu, Tb, Yb and Lu) are determined. The elemental concentration show that Au is found only in sample coded as 114 as traces. The presence of Sb suggest that these samples may be the upper level of an gold occurrence. The results obtained show that, mining activities releases heavy metal pollutant such as Cr. The study of REE concentrations gives an important source of scientific information, that helps in predicting the source and evolutionary history of the rocks. The results obtained indicate the viability of using the k_0 NAA for the determination of the elements La, Ce, Nd, Sm, Eu, Tb, Yb and Lu. The levels of the rare earth element differed may be depending on their origin and geochemistry. So k_0 NAA technique can be effectively used on routine analysis of a large number of samples and it represents an effective method for monitoring gold in geological samples.

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تقدير نسب تركيز العناصر المتواجدة في ست عينات جيولوجية من وادي سيترا بالصحراء الشرقية- مصر باستخدام طريقة k_0 بالتنشيط بالنيوترونات

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تم تطبيق أسلوب التنشيط بالنيوترونات باستخدام طريقة k_0 في تحليل ودراسة نسب تركيزات عناصر ستة عينات جيولوجية تم جمعها من وادي سيترا في الصحراء الشرقية المصرية بغرض الكشف عن مناجم الذهب. تم تشييع العينات مع مجموعه من العينات المعيارية لمدة 3 ساعات في قلب المفاعل المصرى البحثى الثانى عند قره 19 ميجاوات. استخدم الحديد و الانتيمون و المولوبديوم لحساب كل من نسبة الفيض النيوترونى الحرارى الى الفوق حرارى ($f=17.5$) و تعيين معامل حيود النيوترونات (α) عن القانون $1/E$. حيث وجد ان قيمة (-0.03 - α). و قد أمكن باستخدام عينة عيارية من النتجستين التأكد من دقة الحسابات. تم تصميم و استخدام برنامج فورتوران لحساب كل من α و Q_0 . و بالتحليل العناصرى للعينات تحت الدراسة تم التعرف على نسب تركيزات 25 عنصر فى العينات الجيولوجية.