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Elemental Analysis for Geological Samples from Southern of Eastern Desert-Egypt Using Nuclear Non Destructive Techniques

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Abstract: Ten representative geological samples were collected from the Southern of Eastern Desert, Egypt, for analysis by instrumental neutron activation analysis as a sensitive nondestructive analytical tool for the determination of twenty of the major, minor and trace elements. The samples were properly prepared together with standard reference material and simultaneously irradiated in a neutron flux of 2×1011 n/cm2s in the Second Egyptian research reactor (ETRR-2) facility. The gamma spectra were collected by an HPGe detector and the analysis was done by means of a computerized multichannel analyzer. Twenty elements were determined from qualitative and quantitative analysis, which were Al, Na, Cl, K, Sc, Ca, Cr, V, Mn, Fe, Co, Ba, La, Ce, Tb, Yb, Lu, Hf, Pa and Np. Elemental concentrations were also checked using EDX technique.

The aim of this research is to use two analytical techniques named Neutron Activation Analysis (NAA) and Energy dispersive X-ray (EDX) to achieve an accurate knowledge about content and determination of the concentration for each element in selected geological samples from southern of eastern desert -Egypt.

Keywords: geological samples, NAA, EDX, rare earth elements, Eastern Desert-Egypt.

1 Introduction

The elements in the earth's crust are composed of various kinds of elements rarely found exclusively, but are usually combined with other elements to make various substances known as minerals. Neutron Activation Analysis (NAA) is a qualitative and quantitative method for the precise determination of a number of elements in different types of geological samples [1-3]. NAA with gamma-ray spectroscopy is a particularly useful analytical technique for geological investigations, because it can simultaneously provide data with respect to many elements, many of them are difficult to determine by other method [4,5].
NAA and EDX are the most commonly used techniques for measuring the concentration of radioactive isotopes in the geological materials. The radioactive emissions and radioactive decay paths for each isotope are well known. Using this information, it is possible to study spectra of the emissions of the radioactive sample, and determine the concentrations of the elements within it. The application of NAA technique ensure the production of quality base line data as the method is treated a referee method to check the accuracy of other analytical methods worldwide especially for solid sample analysis [6-10].

NAA conducts qualitative and quantitative analyses of the unknown samples by irradiating them with neutrons (n, γ) reaction and detecting the emitted γ-rays from the outgoing radioactive nuclides after irradiation. Qualitative analysis [11-13] can be achieved by analysis of γ-lines in the g spectrum detected and registered by (HPGe) detector and its associated electronic circuit.

In this work, NAA and EDX techniques were used to identify and quantify the major, minor and trace elements in geological samples from Southern of Eastern Desert, Egypt.

2 Experimental

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2.1 Sample Collection

In this study, the selected samples are collected from Hadarba area which is located in the southern extremity of the Eastern Desert of Egypt near the Sudan Frontier. It is easily reached through the asphaltic road Marsa Alam-Halaib along the Red Sea coast. Ten geological samples along the extending of the investigated area are take in equal distance to represent this area by quarting take sample from these selected samples and mixed well to investigate the geological samples [14].

NAA is a quantitative and qualitative method for the precise determination of a number of major, minor and trace elements in different these samples. The samples measured using NAA are activated by neutrons whereas during irradiation the naturally occurring stable isotopes of most elements are transformed into radioactive isotopes by neutron capture. Then the activated nucleus decays according to a characteristic half-life. Some nuclides emit β particles only, but most nuclides emit gamma- quanta, too, with specific quantity of radioactive nuclides is determined by measuring the intensity of the characteristic gamma-ray lines in the spectra. As the irradiated samples contain radionuclide of different half-life different isotopes can be determined at various time intervals [1].

2.2 Sample Irradiation

The collected samples, were placed in a polyethylene vials and irradiated in the thermal irradiation sites with thermal neutrons flux about 2×1011 n cm-2 s-1 at the ETRR-2. The irradiations were carried out by two sets of samples one for short and the second for long irradiation. After irradiation, samples is analyzed qualitatively and quantitatively. Qualitative analysis is used for identifying the appearance of elements in the sample regardless of their concentrations. It can be viewed as a "probing" process for the quick assay of the materials.
The quantitative analysis was used for exact determination of the element weight (g) in the samples.

The weight of an element was calculated according eq. (1) [10,15]

$$W = \frac{\lambda M A}{\varphi a \sigma N_a I_\gamma \zeta (1-e^{-\lambda t_i}) e^{-\lambda t_d} (1-e^{-\lambda t_c})} \qquad (1)$$

Where: W=Element weight (g), λ= Decay constant of the product radioisotope)S-1), M=Atomic weight of target nuclide, A=Area under the characteristic photo peak, Φ=Thermal neutron flux (n.cm–2.S–1), A=Isotopic abundance of a given isotope in the natural target element, which is used for characterization, σ =Thermal neutron absorption cross section (b), Na= Avogadro's number (6.022 x 1023 mole-1), Iγ=γ-ray emission probability per decay (absolute intensity), = ξDetector absolute efficiency at the energy of the characteristic photo peak. ti=Irradiation time (S), td =Cooling time (S), tc =Counting Time of γ-spectrum (S).

2.3 Gamma-Ray Measurements

Two gamma spectrometers based on high purity germanium detectors, with necessary electronics, were used for the measurements of the delayed gamma activity after short time about 30 sec and longtime about 1800 sec irradiation. Both systems were calibrated for energy and efficiency to increase the accuracy of the results. A set of standard point gamma sources were used such as 137Cs, ^{133}Ba, ^{60}Co and ^{152,154,155}Eu. For system No.1, HPGe detector model ORTEC, GEM-100210 with relative efficiency 100 % and energy resolution, full width at half maximum (FWHM), 2.1 keV for the 1332.5 keV gamma line of 60Co was used. For system No.2, HPGe detector model Canberra GC-6020 with relative efficiency 60% and energy resolution 1.9 keV, for the 1332.5 keV gamma line of 60Co, was used [15-20] .
Gamma spectra of the irradiated samples were measured after appropriate cooling time, repeatedly in a cylindrical lead shield at suitable distance from the detector to minimize pile-up and coincidence effects. Energy calibration was performed before measurements, the γ-energy lines of good resolution and free from any interference have been used in the characterization. The characteristic nuclear data used for the analysis of the target isotopes, taken from the NUDAT-2.6 data base [21], are tabulated in Table 1.

The elemental analysis of the samples was also performed, for comparison, using EDX. JEOL-JSM 5600 LV scanning electron microscope was used for this measurement.

2. 4 Sources of Uncertainties

The uncertainty measurement is a critical issue in any experiment. The sources of uncertainty considered are the contribution from the sample and standard mass measurements, and the contribution from counting statistics [1]. In the present measurements the uncertainty ranges of 12-16%.

3 Results and Discussion

In the present study, NAA and EDX techniques as sensitive analytical techniques are applied to study the essential elemental content in different geological samples.
The induced activities were counted by γ -ray spectrometry using High Purity Germanium (HPGe) detector to investigate the elements present in these samples. From their gamma ray spectra, twenty elements were identified from qualitative and quantitative analysis, which were Al, Na, Cl, K, Sc, Ca, Cr, V, Mn, Fe, Co, Ba,

La, Ce, Tb, Yb, Lu, Hf, Pa and Np.
The results of these elemental concentration of the activation analysis for the geological samples are presented in Figs1-5 and the numerical values are given in Tables 2,3 and 4.

Table 1: Nuclear data of the target isotopes.

Stable Isotope	Abundance (%)	Activation Product	Half-life	E_γ (keV)	I (%)
Na-23	100	Na- 24	14.95 h	1369.0 2754.0	100 99.94
Al-27	100	Al-28	2.24m	1778.85	100
Cl-37	24.23	Cl-38	37.24 m	1643 2167	31.91 42.41
K-41	6.73	K-42	12.35h	1525	18.08
Sc-45	100	Sc-46	83.81 d	889.27 1120.54	99.98 99.99
Ca-48	0.18	Ca-49	8.72m	3084.4	92.1
Cr-50	4.35	Cr-51	27.7d	320.08	10.08
V-51	99.75	V-52	3.75 m	1434.06	100
Mn-55	100	Mn-56	2.58 h	846.75 1810.72 2113.05	98.9 27.2 14.3
Fe-58	0.28	Fe-59	44.50 d	1099.25 1291.59	56.51 43.21
Co-59	100	Co-60	5.27 y	1173.23 1332.50	99.98 99.99
Ba-138	71.7	Ba-139	83.06	165.86	24.0
La-139	99.91	La-140	1.68 d	328.76 487.02 815.77 1596.21	20.6 44.30 22.90 95.40
Ce-140	88.43	Ce-141	32.50 d	145.44	48.20
Tb-159	100	Tb-160	72.30 d	879.38 966.17 1177.96	30.01 25.21 15.07
Yb-168	0.13	Yb-169	32.02 d	177.21 197.96 307.74	22.20 35.80 10.05
Yb-174	31.8	Yb-175	4.19 d	282.52 396.33	3.0 6.40
Lu-176	2.59	Lu-177	6.73 d	208.37	11.00
Hf-180	35.1	Hf-181	42.39 d	133.02 482.0	43.30 80.50
Th-232	100	Pa-233	26.96 d	312.17	38.60
U-238	99.27	Np-239	2.35 d	106.12 277.59	27.20 14.38

The concentration of trace elements: Al, Cl, K, Ca, Mn, Co,V and Ba were determined and shown in figures 1 and 2 and presented table 2. From Table 2, the elements Al, Cl, K, Ca, Mn, and Co have the concentration values range from 34.26 to 649.82 ppm from the geological samples, and the element Al is found to have higher concentration in all samples range varied from 445.97 to 649.82 ppm. While the other elements in the samples may be considered as minor elements and the lowest values for Ba and V for all samples.
The activation converts ^{238}U and ^{232}Th into ^{239}Np and ^{233}Pa, respectively, by INAA and successive β^--decay:
$^{238}U(n,\gamma)\,^{239}U(\beta^-)^{239}Np$ E_γ = 228&277 keV
$^{232}Th(n,\gamma)\,^{239}U(\beta^-)^{233}Pa$ E_γ = 312.17 keV
As shown in table 3The average concentrations were range from 34 to 90 ppm for Pa-233 and from 42 to 100 ppm for Np-239,and the element Ce is found to have higher concentration in all samples range varied from 91.1 to 570.67 ppm.
Iron has the highest concentration in all samples ranges from 170391 to 381850 ppm see table 4. The average value concentration of **Na- 24** with half life time (**15.03h)** were estimated due to 1368.0 and 2754.0 keV and ranged from 17937 to 101789 ppm as shown in figure 4.
Cr-51 produced from ^{50}Cr, isotopic abundance 4.345% ,by the ^{50}Cr (n,γ) ^{51}Cr reaction. The thermal neutron cross section for this reaction is 15.970.2 barns, which makes it suitable for the neutron activation analysis. ^{51}Cr has a half-live of 27.7days and disintegrates by electron capture emitting only one γ -ray of 320 keVwith9.83%abundance.
The average concentration of Chromium was calculated due to the detection of gamma transition 320.03 keV ranged from 796 to 3764 ppm, also Lu has relatively high concentration in all samples ranges from 1423 to 2388 ppm as shown in figure 5.
The concentrations of the elements from geological samples which were collected from Southern of Eastern Desert were quantitatively analyzed also using EDX technique and listed in Table 5.

Table2. Concentrations of the elements from geological samples which were collected from Southern of Eastern Desert.

Element	Product	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	average
Al	Al-28	570.7	649.82	451.2	514.63	627.87	643.59	445.97	500.16	478.23	539.36	542
Cl	Cl-38	68.95	87.26	52.65	73.3	61.63	74.43	69.8	61.1	58.35	65.22	67
K	K-42	213.93	278.1	116.18	188.89	203.27	294.66	231.75	208.94	195.06	258.91	219
Ca	Ca-49	192.28	125.65	218.56	248.18	159.96	97.08	154.64	144.37	209.67	198.54	175
Mn	Mn-56	69.84	80.93	82.52	98.19	79.78	41.81	139.44	40.54	55.21	97.82	78
Co	Co-60	45.08	63.22	49.09	69.34	74.92	46.8	34.26	48.33	53.39	64.76	55
V	V-52	0.96	0.94	1.19	1.63	1.16	0.26	1.38	0.52	0.79	1.02	1.0
Ba	Ba-139	3.71	4.98	4.15	1.84	5.96	4.64	3.87	4.94	5.03	4.28	4.0

Table 3: Concentrations of the elements from geological samples which were collected from Southern of Eastern Desert.

Element	Product	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	
Sc	Sc-46	64.3	57.5	60.3	90.3	68.1	84.0	44.8	51.5	67.1	82.5	67
La	La-140	54.5	78.1	69.5	60.1	178.8	93.8	145	93.8	102.4	98.0	97
Ce	Ce-141	179.6	427.2	91.1	151.8	570.6	471.1	229.1	229.1	250.3	235.7	283
Tb	Tb-160	9.13	6.1	6.7	5.9	7.7	5.8	7.5	6.1	9.5	7.62	7
Yb	Yb-169	30.3	18.9	21.9	23.7	331.8	17.6	24.8	16.9	24.1	23.7	53
Hf	Hf-181	80.7	93.5	51.7	144.1	75.5	79.5	52.8	71.8	81.8	79.5	81
Th	Pa-233	49.4	52.1	34.2	42.2	90.4	49.3	68.8	52.1	53.3	40.0	53
U	Np-239	56.5	78.1	59.0	69.3	86.1	95.1	100.1	42.2	85.1	82.0	75

Table 4: Concentrations of the elements from geological samples which were collected from Southern of Eastern Desert.

Element	Product	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	average
Na	Na-24	43656	18235	74859	17937	83779	53473	101789	44285	68677	69979	57487
Cr	Cr-51	1488	1331	3764	1643	1668	882	1760	796	1766	2448	1754
Fe	Fe-59	257145	241802	260327	381850	297380	247288	170391	195834	275265	358020	268530
Lu	Lu-177	2368	1423	1714	2046	2388	1735	2049	1424	1832	1924	1890

Table 5: EDX Concentration (%) for geological samples.

Element	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	average
O	58.1	50.8	44.5	51.7	55.8	49.2	47.7	59.3	50.4	48.3	52.0
Na	1.0	1.6	2.0	2.1	1.0	2.8	3.3	1.9	1.13	3.3	2.0
Mg	0.98	0.90	1.58	1.15	0.63	1.02	1.32	1.30	1.40	1.20	1.0
Al	3.15	6.66	7.63	6.10	2.65	6.56	11.32	5.91	5.39	7.65	6.0
Si	36.72	32.73	30.18	31.34	39.77	31.98	20.50	28.32	35.44	29.80	32.
S	-	0.29	0.18	0.29	0.23	0.25	0.24	0.29	0.23	0.24	0.25
Cl	-	0.79	2.61	1.71	-	1.91	1.73	0.71	1.21	2.67	1.7
K	-	3.40	4.10	2.49	-	3.21	2.92	2.15	1.59	1.63	2.7
Fe	-	2.67	7.16	2.49	-	2.93	7.53	-	3.19	4.20	4.3

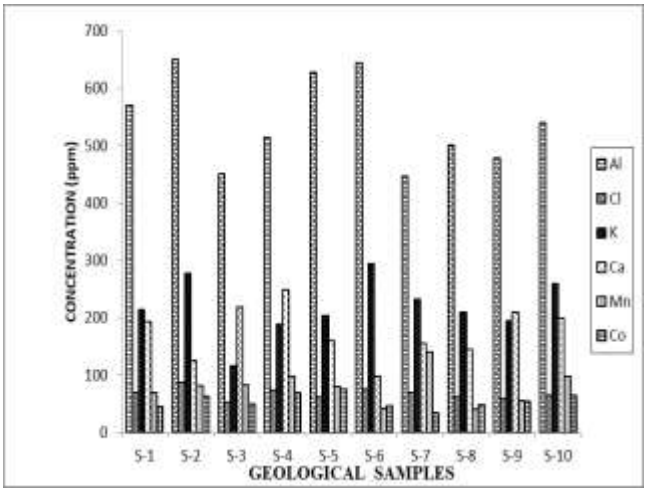

Fig.1: Concentrations for some elements from geological samples (group 1).

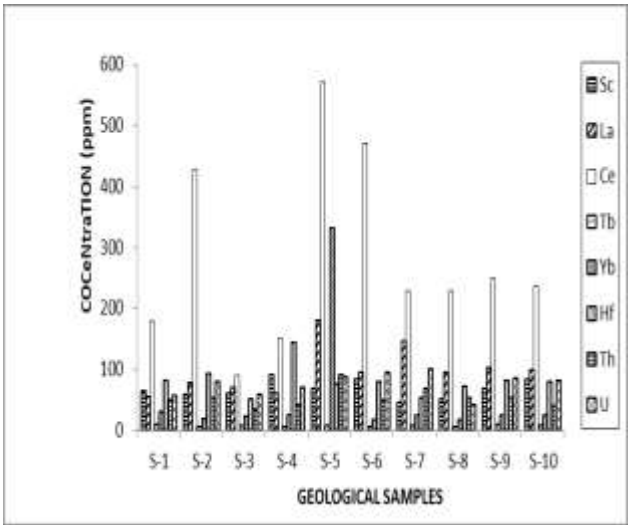

Fig.3: Concentrations for some elements from geological samples (group 3).

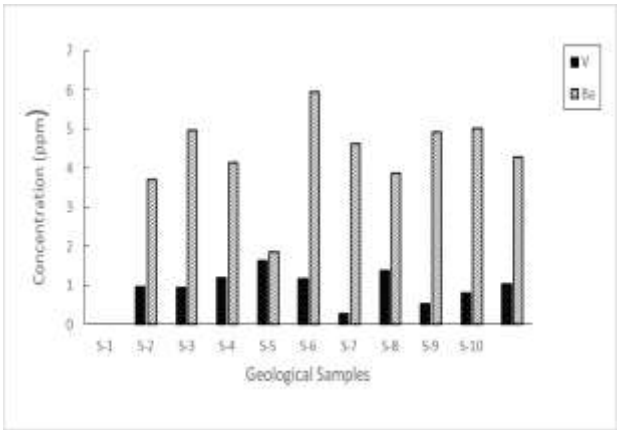

Fig.2: Concentrations for some elements from geological samples (group 2).

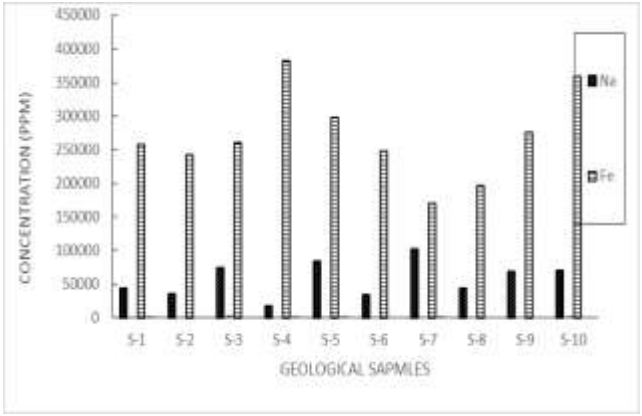

Fig.4: Concentrations for some elements from geological samples (group 4).

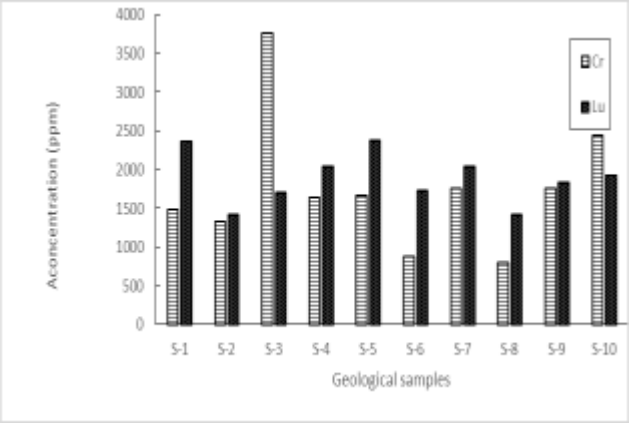

Fig.5: Concentrations for some elements from geological samples (group5).

Conclusions

Twenty elements were quantitatively determined for geological samples from southern of Eastern Desert – Egypt by Instrumental neutron activation analysis and EDX techniques. The elements Al, Na, Cl, K, Sc, Ca, Cr, V, Mn, Fe, Co, Ba, La, Ce, Tb, Yb, Lu, Hf, Pa and Np were determined. Elemental concentrations were also checked using EDX technique. From the obtained results, it is clear that INAA is an effective and useful tool to provide good data for geological samples with a precision and satisfying accuracy. The results revealed that the geological samples have valuable elements of nuclear and industrial interest.

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