A Comparison between Neutron Activation Analysis and X-Ray Fluorescence Methods in Analyzing the Granite Samples

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Abstract: This study focused on the performance of where elements analysing techniques were used to detect the elements in granite stones. These techniques are NAA (neutron activation analysis) and XRF (X-ray fluorescence). They were applied to detect the elements in samples which had been chosen from different areas of Pulua Penang in Malaysia collected by geophysics group which helped to describe and identify the elements found in the granite stone that were used in the study procedures to control the analytical results. The integration of both methods has enabled the researcher to determine 40 elements in the samples. The numbers of elements detected by XRF analysis method are 12 elements (Ar, K, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu and Zn); while, the elements detected by NAA method have three folds of elements with XRF analysis method were 35 elements (Na, Al, Si, K, Ca, Sc,Ti, Mn, Fe, Co, Ga, Ce, As, Br, Rb, Zr, Sb, I, Cs, Ba, La, Nd, Sm, Eu,Tb, Dy, Yb, Lu, Hf, Ta, W, Au, Pa and Np). Seven common elements were detected in both techniques: K, Sc, Ti, V, Mn, Fe and Co. Si has a higher concentration in NAA technique which is 331.8 ppm. Sc has a lower concentration in XRF technique which is 0.25 ppm. Nd has a lower concentration in NAA technique which is 3.09×10^{-5} ppm. Finally, it is found that the NAA is better to detect the elements than XRF.

Key words: Radiation science, radiation physics, NAA, XRF, analyzing methods.

1. Introduction

NAA (neutron activation analysis) is a very useful and sensitive technique for performing both qualitative and quantitative multi-element analysis of major, minor and trace elements in samples from various fields of scientific or technical interest [1]. XRF (X-ray fluorescence) spectroscopy is widely used for the qualitative and quantitative elemental analysis of environmental, geological, biological, industrial and other samples [2]. Compared to other analytical techniques, such as AAS (atomic absorption spectroscopy), ICPS (inductively coupled plasma spectroscopy) and NAA, XRF has the advantage of being non-destructive, multi-elemental, fast and

cost-effective [3]. Furthermore, it provides a fairly uniform detection limit across a large portion of the periodic table and is applicable to a wide range of concentrations from a 100% to few parts per million (ppm). Its main disadvantage is that analyses are generally restricted to elements heavier than fluorine [4]. In comparison to well-developed atomic analytical methods like XRF, this nuclear analytical method, NAA is still preserving its role as a workhorse for the vast amount of analytical work because it is non-destructive (sample loses almost all radioactivity after some time) and instantaneous [5]. In this study, authors have used two different methods to analyse 12 samples of granite's stone collected from different regions in Pulau Penang, Malaysia. These methods were 10 MeV-NAA at the reactor of Malaysian nuclear agency and XRF at the biophysics lab in

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school of physics. The experimental procedures were tested on several certified reference materials: Five reference materials to granite's stone were determined in the analysed samples using these two techniques. The detailed examination of the results allowed authors to study the complementarity of the two methods and to choose the most suitable technique for each element.

2. Experiments

2.1 Collection and Preparation of the Samples

There are methods to analysis the elements in the materials. In this study, two methods were used for this purpose, which are XRF and NAA methods. The accuracy of the materials as samples is depending on granite stones (12 samples) with different locations from Penang Island, Malaysia collected by geophysics group. Table 1 shows the samples as a function of their locations and thicknesses.

2.2 NAA Technique

The stones are crushed by using the mortar and pestle then check the grain size of powder in Table 1, when it is dry and the powder is put in the vial and should be from plastic, because it has less attenuation and not reaction with neutron beam as shown in Fig. 1. The weights of vials are taken before and after putting the powder using electronic balance. The weight range of the powder were around 0.2015-0.2095 g and

marked each vial. Table 1 shows the weight of the samples and the thickness. Weight, solder marked and dry of each sample are checked before irradiation and the vials are sealed tightly using the soldering iron. The irradiation step was used the RR (rotary rack) tool to put the samples inside the core of reactor as shown in Fig. 2. Then, the samples were irradiated for 6 h and average of neutron flux was equal to 3×10^{12} n·cm^{-2}·s^{-1} where the neutron source is Am-241 (americium-241), Ba-133 (barium-133) with energy up to 10 MeV. After that, the samples was get out from the core of reactor and the samples were kept to 4-5 days to reduce the activity of the samples as shown in Table 2 which is indicated the activity of the samples directly and after 4-5 days by Geiger-Muller counter to use count of activity. The equipment of account is including of HPGe detector connected with nimbin system which contains power supply card and amplifier card, then nimbin is connected with PC computer including gamma vision software of analysis. The sample is put on distance 8 cm from the detector for 1 h then record the data and analysis it using Plot Fit and Peak Fit.

2.3 XRF Technique

Set up bench of the work and accurately the position of the source and the sample according to for the detector are shown in Fig. 3. The angle between the sample and detector is 0°. Table 3 indicates the

Table 1　The samples as a function of their locations and thicknesses collected from different area in Pulua Penang, Malaysia.

| No.	Location of sample	Weight (g) of powder	Weight (g) of solid	Thickness (mm)
1	Bukit jambul	0.2025	10.5	3
2	Pasir panjang	0.2054	13.34	3
3	Titi kerawang	0.2055	9.4	3
4	Bukit genting	0.2023	8.9	3
5	Ayer itam	0.2035	9.2	3
6	Telok bahang	0.2017	11.9	3
7	Air terjun bayan lepas	0.206	8.7	3
8	Titi serong	0.2059	11.3	3
9	Pantai acheh	0.2019	11.06	3
10	Telok tempoyak	0.2032	11.17	3
11	Relau	0.2038	12.7	3
12	Kampung epakat, Balik pulau	0.2027	9.5	3

Fig. 1 Vials.
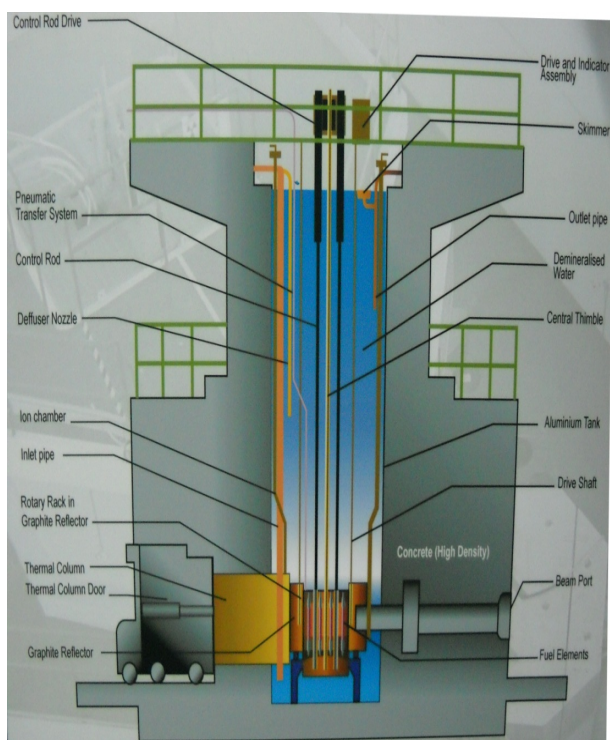
Fig. 2 The core of reactor.

Table 2 Activity of sample directly after irradiate and after 4-5 days.

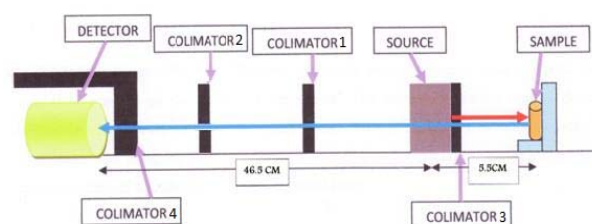| No.	Activity (mR) dirctly	Activity (mR) after 4-5 days
1	100	2
2	60	2
3	30	0.7
4	60	1.4
5	60	1.5
6	30	1.5
7	50	1
8	40	2
9	30	1
10	60	2
11	50	2
12	50	2

Fig. 3 Schematic diagram of the set-up, coming out from the sample indicates XRF path ray and coming out from the source indicates the path ray of gamma ray.

Table 3 The distance between the settings of XRF.

The setting	Distance (cm)
Sample & source	5.5
Source & collimator (1)	17.5
Collimator & collimator (2)	7.5
Collimator (2) & collimator of detector	5.5
Detector & sample	46.5

Fig. 4 Shape of effect of radiation on film of X-ray.

distance between the setting detector, source and sample place. To observe that the source, collimator and sample are placed at straight line, therefore, X-ray of film behind the collimator (1) and observe the shape of radiation beam geometry after 10 min to exposure the film of radiation. As shown in Fig. 4, the detector (Lege) is provided with the liquid nitrogen cryostat and connect it with nimbin system, which contains power supply card and amplifier card. Then, connect them with PC computer, which include maestro software. Calibration for the software and detector is done. To detect the X-ray's fluorescence from the sample a Lege detector, cooled with liquid nitrogen temperature is using to detect the characteristic X-rays. The detector has a high resolution. The pulses from the detector are processed by a shaping amplifier and converted into pulse height

Table 4　Comparison of the results obtained between NAA and XRF.

| Atomic number Z | Element | Concentration | |
		NAA	XRF
11	Na	3.013	-
13	Al	37.73	-
14	Si	331.8	-
18	Ar	-	0.74
19	K	19.94	0.68
20	Ca	11.14	-
21	Sc	0.26	0.25
22	Ti	1.01	1
23	V	0.033	1
24	Cr	-	0.92
25	Mn	0.105	0.65
26	Fe	26.08	0.8
27	Co	0.0116	1
28	Ni	-	1
29	Cu	-	1
30	Zn	-	0.99
31	Ga	0.011	-
32	Ce	0.02	-
33	As	0.01023	-
35	Br	0.104	-
37	Rb	0.047	-
40	Zr	0.24	-
51	Sb	0.019	-
53	I	0.0074	-
55	Cs	0.004	-
56	Ba	0.115	-
57	La	0.016	-
60	Nd	3.09E-05	-
62	Sm	0.0076	-
63	Eu	0.0102	-
65	Tb	0.00046	-
66	Dy	0.088	-
70	Yb	0.00313	-
71	Lu	0.00019	-
72	Hf	0.0099	-
73	Ta	0.00618	-
74	W	0.038	-
79	Au	0.0036	-
91	Pa	0.23	-
93	Np	0.033	-

by the ADC (analog-to-digital converter) of the MCA (multichannel analyzer). After checking, everything each sample is exposed of 24 h for the source. Then, the data were recorded and analysed.

3. Results and Discussion

The two methods have detected 40 elements in the samples as shown in Table 4. This shows that NAA method could be used to detected 35 elements, while 12 elements were detected by XRF. The reason of this different could be that the two methods have a different energy source. The NAA method was used neutron source with energy 10 MeV and HPGe detector which may able to detect the high energy and low energy, while the XRF method was used X-ray source with energy 59.53 KeV and Lege detector where this type of detector could able to detected low energy.

4. Conclusions

NAA technique has an excellent technique to detect the elements in the samples and XRF technique is a very good method to detect some elements, but unfortunately the XRF technique is limited (excitation energy of source is limited) and difficult to detect the other elements since the scattering from the source and the sample was presented. Some elements are missing, because it has short or long half life time and cooling time 4-5 days is the best time that it has the average range between the long and short half-life of elements which could detect many elements in the sample. Ti, V, Co, Ni and Cu have a higher concentration in XRF technique which is 1 ppm in each element. Si has a higher concentration in NAA technique which is 331.8 ppm. Sc has a lower concentration in XRF technique which is 0.25 ppm. Nd has a lower concentration in NAA technique which is 3.09×10^{-5} ppm.

Acknowledgments

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