

THERMAL AND EPITHERMAL ACTIVATION ANALYSIS USING K_0 -METHOD

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الخلاصة :

في هذه الدراسة تم استخدام تقنية جديدة في التحليل التثبيطي للنيوترونات الحرارية وفوق الحرارية وهي طريقة K_0 العيارية حيث تم تحليل عينات جيولوجية جهزت بخلط كمية من مسحوق العينات مع مسحوق خاص للشمع لتقليل الخطأ الناتج من الامتصاص الذاتي بحيث يكون الخلط متجانسا .

وحرصاً على دقة النتائج فقد تمّ تعيين معامل انحراف فيض النيوترونات فوق الحرارية عن القانون $1/E$ ، وتأثيرات الموافقة الحقيقية لأشعة جاما المتابعة ، ومعامل نفاذية للنيوترونات فوق الكادميوم وكذلك تغير كفاءة العداد مع تغير الشكل والوضع الهندسي للعينة بالنسبة للعداد وتصحيح النتائج بناءً على هذه المعالجات .

لقد أمكن في هذه الدراسة تعيين (٢٤) عنصراً في عينات جيولوجية عيارية وهي G-1, AGV-1, BCR-1 . وفي كلتا الحالتين الحرارية وفوق الحرارية اظهرت النتائج اتفاقاً جيداً مع النتائج المنشورة .

ABSTRACT

An analytical method was used for the determination of 24 elements in a variety of geological materials based upon a sequence of two irradiations and two counting periods of 50-100 mg of USGS samples. In this method, the deviation factor α , representing deviation of reactor epithermal neutron flux from $1/E$ law, true coincidence effects of cascade γ -rays, and the efficiency of the Ge(Li) detector were taken into consideration.

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1. INTRODUCTION

When using the K_0 -method [1–3] for standardization in epithermal neutron activation analysis (ENAA) and reactor neutron activation analysis (RNAA), the concentration of an element in a sample (weight W gram), determined through counting (on a Ge(Li)) of a gamma-line with energy E_γ , emitted by the formed isotope i , is calculated from:

ENAA:

$$\rho_i(\text{ppm}) = \frac{\left[\frac{(N_p/t_m)}{(\text{SDC})} \right]_i}{\left[\frac{(N_p/t_m)}{(\text{SDC})} \right]_{\text{comp}}} \times \frac{w_{\text{comp}}}{W} \times \frac{1}{K_{0,\text{comp}(i)}} \times \frac{f + Q_{0,\text{comp}}(\alpha)}{f + Q_{0,i}(\alpha)} \times \frac{\epsilon_{p,\text{comp}}}{\epsilon_{p,i}} \quad (1)$$

$$\rho_i(\text{ppm}) = \frac{\left[\frac{(N_p)_{\text{Cd}}/t_m}{(\text{SDC})} \right]_i}{\left[\frac{(N_p)_{\text{Cd}}/t_m}{(\text{SDC})} \right]_{\text{comp}}} \times \frac{w_{\text{comp}}}{W} \times \frac{1}{K_{0,\text{comp}(i)}} \times \frac{F_{\text{Cd,comp}} Q_{0,\text{comp}}(\alpha)}{F_{\text{Cd,i}} Q_{0,i}(\alpha)} \times \frac{\epsilon_{p,\text{comp}}}{\epsilon_{p,i}} \quad (2)$$

where “comp” = coirradiated single-comparator (power monitor) e.g. ^{197}Au ; $E_\gamma = 411.8$ keV with weight w (in μg), and W = sample weight (in g).

Detailed explanation of other symbols and notations can be found in References [1–3]. Some essential definitions follow.

N_p = peak area. Correction procedures and formula were worked out for losses due to true coincidence (residual uncertainty, for sources on top of the detector, $\cong 1.5\%$), and burn-up; corrections for spectral interferences and ^{235}U -fission can be performed with increased accuracy by applying K_0 -factors [4].

$S = 1 - \exp(-\lambda t_{\text{irr}})$, t_{irr} = irradiation time and $T = (\ln 2)/\lambda$ = half-life; $D = \exp(-\lambda t_d)$, t_d = decay time; $C = 1 - \exp(-\lambda t_m)/\lambda t_m$, t_m = measuring time. T -values were critically selected from literature [5, 6].

In case of complicated reaction/decay mechanisms (mother–daughter etc.), the term $(N_p/t_m)/\text{SDC}$ has to be replaced by adequate expressions [7].

$K_{0,\text{comp}}(i)$ = a composite nuclear constant, containing atomic weights, isotopic abundances, thermal cross-sections and gamma-intensities for isotopes “ i ” and “comp” [5, 6]. These absolute nuclear data being often unreliable, experimental K_0 -determination, with an accuracy of $\pm 1\%$, was performed for some 32 isotopes [8].

f = thermal to epithermal flux ratio. Accurate, “instantaneous” f -monitoring can be performed by coirradiating and counting a Zr-foil (^{95}Zr – ^{97}Zr) [9].

$Q_0(\alpha) = (Q_0 - 0.429) (\bar{E}_r)^{-\alpha} + 0.429 / [(2\alpha + 1) (0.55)^\alpha]$, with Q_0 = resonance integral to thermal cross-section ratio, \bar{E}_r = effective resonance energy, and α = measure for the deviation of the epithermal flux spectrum ($\sim 1/E^{1+\alpha}$) from the $1/E$ law. Values of Q_0 with an accuracy of $\sim 3\%$, were experimentally determined or evaluated [10]. \bar{E}_r values were calculated for 90 isotopes [11, 12]. “Instantaneous” α -monitoring was shown to be possible by coirradiating a Zr-foil and a Au–Al wire, followed by counting on a detector with known efficiency (^{95}Zr – ^{97}Zr – ^{198}Au) [9].

ϵ_p = detection efficiency. A procedure was developed to calculate ϵ_p -values for extended sources at any detector distance, taking into account gamma-attenuation; the residual uncertainty (for sources on top of the detector) is $\cong 2\%$ [4].

F_{Cd} = cadmium transmission factor for epithermal neutron [13]. Any element can be used as a comparator if its nuclear data relevant to activation and gamma spectrometry are known with good accuracy. The nuclear data involved in the present study are shown in Table 1 [8, 10–14].

2. EXPERIMENTAL

USGS samples we analyzed were: Basalt BCR-1, Andesite AGV-1, and Granite G-1. Sample preparation was carried out in a clean-room. About 50–100 mg of a sample was weighed in a small polycarbonate vial (for short irradiation) or were separately wrapped in thin aluminum foil (for long irradiation). The samples were irradiated for 20 seconds in the pneumatic irradiation facility in the Egyptian reactor. The radionuclides of Na, Al, Ca, Ti, V, Mn, K, Ba, Sr, and Dy were measured for 200 seconds within 6 minutes after irradiation. The measurement system is composed of an ORTEC 40 cm^3 Ge(Li) detector and 4000 channel pulse

Table 1. Nuclear Data Involved in the Present Study

Element	Isotope Produced	Q_0 [10]	\bar{E}_r (eV) [11, 12]	$T_{1/2}$	E_γ (keV)	K_0 [8, 14]
Na	^{24}Na	0.59	3130	14.96 h	1368.6	4.68×10^{-2}
					2753.8	4.62×10^{-2}
Al	^{28}Al	0.73	8240	2.2405 m	1779.0	1.75×10^{-2}
Ce	^{141}Ce	0.83	11 400	32.5 d	145.4	3.66×10^{-3}
K	^{42}K	0.97	9040	12.36 h	1524.7	9.20×10^{-4}
Ca	^{49}Ca	0.45	—	8.72 m	3084.2	1.01×10
Sc	^{46}Sc	0.44	4110	83.8 d	889.3	1.21
					1120.5	1.21
Ti	^{51}Ti	0.67	49 900	5.76 m	320.1	3.74×10^{-4}
					928.6	2.65×10^{-5}
V	^{52}V	0.55	5960	3.75 m	1434.0	1.96×10^{-1}
Cr	^{51}Cr	0.53	5940	27.70 d	320.1	2.65×10^{-3}
Mn	^{56}Mn	1.07	412	2.577 h	846.8	4.96×10^{-1}
					1810.7	1.35×10^{-1}
					2113.0	7.17×10^{-2}
Fe	^{59}Fe	0.96	325	44.50 d	1099.2	7.52×10^{-5}
					1291.6	5.76×10^{-5}
Co	^{60}Co	2.03	133	5.272 y	1173.2	1.31
					1332.5	1.32
Rb	^{86}Rb	14.8	694	18.65 d	1076.8	7.21×10^{-4}
Sr	$^{87\text{m}}\text{Sr}$	4.11	672	2.806 h	388.5	1.49×10^{-3}
Zr	^{95}Zr	5.88	4520	64.03 d	724.2	9.14×10^{-5}
					756.7	1.11×10^{-4}
La	^{140}La	1.24	76.8	40.28 h	1596.2	1.34×10^{-1}
Dy	^{165}Dy	0.2	—	2.334 h	94.7	3.50×10^{-1}
Sb	^{124}Sb	28.8	24.9	60.20 d	1690.9	1.43×10^{-2}
Cs	^{134}Cs	18.4	8.68	2.062 y	604.7	4.33×10^{-1}
Ba	^{139}Ba	0.75	20 400	83.1 m	165.9	1.05×10^{-3}
Sm	^{153}Sm	14.4	8.33	46.7 h	103.2	2.31×10^{-1}
Lu	^{177}Lu	0.32	—	6.7 d	208.4	7.11×10^{-2}
		[15]				
Th	^{233}Pa	12.0	—	27.0 d	312.0	2.52×10^{-2}
U	^{239}Np	102.3	15.8	2.355 d	277.6	3.48×10^{-3}

height analyzer. Other radionuclides such as those of Sc, Cr, Fe, Co, Zn, Zr, Sb, Cs, Tb, Lu, Hf, were measured for 1 ks with enough cooling after 48 hours irradiation. The decay during the measurements and the dead time losses in the multichannel analyzer were corrected for. A survey of the procedure followed is presented in Table 2.

3. RESULTS AND DISCUSSION

Table 3 shows the analysis results for some of the elements with good counting statistics (and thus low

uncertainties). The good correspondence proves the accuracy of the above outlined correction procedures (α , coincidence, efficiency, etc.). We have also the reported values on USGS samples by Falanagan for direct comparison. When considering BCR-1, the elements Ce, Co, Fe, La, Na, Sc, Sm, Th, and U for which the literature data show no large scattering, the concentrations found in this work are in excellent agreement with the reported values. For Cr, Cs, and Sb large variations between the literature data exist, thus no definite conclusions can be drawn, while

Table 2. Experimental Procedure

Samples:	50–100 mg; mixed with wax; Pellets ~7 mm dia × 4 mm; monitors: Zr-foil, 0.1% Au–Al wire
Irradiation:	WWR-M; various channels ($F = 21-67$; $\alpha = 0.012-0.085$); 3 samples per channel ($\times 2$)
Counting:	Ge(Li); ≥ 3 measure/sample (from short to long t_d); distance 20–2 cm (corrected for coincidence and geometry); low count rate (pulse pile-up negligible); dead-time stabilizer.
Calculation:	Peak area, detector efficiency; concentrations (coincidence correction considered).
Final Results:	Correction for threshold reaction, fission, blank, average with uncertainty.

Table 3. Elemental Abundances in Geological USGS Samples

Element	ppm BCR-1			ppm AGV-1			ppm G-1		
	RNAA This work	ENAA This work	Reported [16]	RNAA This work	ENAA This work	Reported [16]	RNAA This work	ENAA This work	Reported [16]
Al(%)	7.31	7.40	7.21	9.60	9.38	9.14	8.00	7.81	7.43
Ba	670.00	663.00	1275.00	1218.00	1230.00	1210.00	1205.00	1221.00	1200.00
Ca(%)	4.90	4.83	4.95	3.58	3.80	3.50	0.97	1.03	0.99
Ce	53.70	52.10	54.00	66.00	71.00	63.00	168.00	161.00	170.00
Co	36.00	38.20	38.00	15.60	14.40	14.10	2.21	2.39	2.40
Cr	16.10	13.90	17.00	12.70	13.30	12.00	22.00	25.00	20.00
Cs	1.06	0.92	0.95	1.20	1.38	1.40	1.39	1.52	1.50
Dy	6.30	6.00	6.30	3.60	3.80	3.50	2.36	2.54	2.40
Fe(%)	9.20	9.00	9.38	4.79	4.96	4.73	0.75	0.80	0.75
K(%)	1.42	1.44	1.41	2.38	2.29	2.40	4.52	4.48	4.55
La	25.10	25.90	26.00	38.00	37.00	35.00	105.00	104.00	101.00
Lu	0.55	0.58	0.55	0.29	0.34	0.28	0.19	0.20	0.19
Mn(%)	0.14	0.143	0.14	0.079	0.073	0.075	0.025	0.024	0.023
Na(%)	2.56	2.49	2.42	3.34	3.38	3.16	2.44	2.44	2.46
Rb	52.10	48.00	46.00	68.00	71.50	67.00	221.00	217.00	220.00
Sb	0.61	0.68	0.69	4.80	4.40	4.50	0.28	0.31	0.31
Sc	34.00	36.00	33.00	13.40	14.90	13.40	2.89	2.99	2.90
Sm	6.00	6.50	6.60	6.00	6.60	5.90	8.10	8.30	8.30
Sr	317.00	329.00	330.00	654.00	644.00	657.00	255.00	248.00	250.00
Ti(%)	1.34	1.36	1.32	0.63	0.66	0.62	—	—	—
Th	6.40	6.02	6.00	6.40	6.90	6.41	46.00	50.00	50.00
U	1.56	1.72	1.74	2.10	1.90	1.92	3.50	3.35	3.40
V	403.00	391.00	400.00	120.00	128.00	120.00	0.40	0.42	0.40
Zr	170.00	194.00	190.00	237.00	218.00	220.00	200.00	211.00	210.00

AGV-1 and G-1 results agree quite well with the reported values. The main criteria for the choice of one or other of the two procedures are summarized as follows.

RNAA

The procedure is suitable for elements with relatively high thermal cross-sections compared to the resonance integrals such as Ca, Ce, Cr, Fe, and Lu. The advantages of RNAA are the lower costs and higher sensitivity for these elements. The disadvantage is difficulty in using the K_0 approach with RNAA, due to the contribution of two neutron components (thermal and epithermal) in the activation process.

ENAA

This procedure is advantageous with elements having relatively high resonance integrals, such as Co, Cs, Rb, Sb, Sm, Sr, Th, Zr, and U. Additional advantages of ENAA are the possibility of using the K_0 approach materials, as well as the higher accuracy with some elements due to the suppression of the activity from matrix elements. The disadvantages of ENAA are higher costs and difficulties associated with the irradiation and handling of cadmium.

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