

DETERMINATION OF IRON AND NICKEL IN GEOLOGICAL SAMPLES BY ACTIVATION ANALYSIS WITH REACTOR FAST NEUTRONS

Abd El-Ghany El Abd

**Reactor Physics Department, Nuclear Research Centre,
Atomic Energy Authority,
Inchass, P.O: 13759, Egypt.*

Abstract

Threshold reactions induced by reactor fast neutrons are well recognized. The concentration of Fe and Ni were determined in nine geological samples by activation analysis with reactor fast neutrons using the threshold reactions $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ respectively. The fast neutron flux was determined using the reactions $^{92}\text{Mo}(n,2n)^{92m}\text{Nb}$ and $^{95}\text{Mo}(n,p)^{95}\text{Nb}$.

The determined concentration of Fe and Ni in the samples were checked by determining them in the GSJ JB-1 reference material using the same (n,p) reactions. There are a good agreement between the measured and recommended values.

The concentrations of Fe were also determined by the (n,γ) capture reactions in the geological samples and the JB-1 reference material using the $k_0 - \text{NAA}$ method. There are good agreements between the determined concentrations from the (n,p) and the (n,γ) reactions.

Key words: threshold reactions; neutron activation analysis; fast neutron flux; concentration.

Corresponding author: abdo_e@yahoo.com

1. Introduction

In reactor neutron activation analysis, threshold reactions of the types (n,p) , (n,n') , (n,α) and $(n,2n)$ induced by the fast component of the neutron spectrum of reactors (Yule et al., 1965) as well as the (n,γ) capture reactions produced by thermal and epithermal neutrons (De Corte, 1987) are well recognized. Activation analysis with reactor fast neutrons using threshold reactions are not widely used, because the low flux density of fast neutrons in reactors, the small cross sections of many threshold reactions and the relatively high threshold energy of neutrons. Consequently, very low activities are produced by these reactions which may further be obscured by those from the (n,γ) reaction products. However, threshold reaction can be used to determine the content of some elements in samples on the conditions; the density of the fast neutrons is high enough to compensate the smallness of the cross section, and the elemental contents in the sample under investigation are relatively high (for example in range of mg/g).

Threshold reactions provide useful alternative reactions for the determination of certain elements by NAA if the (n,γ) reaction products are formed with unfavorable radioactive properties. As an example, determining Ni using the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ reaction ($t_{1/2}$ of ^{58}Co is 70.82 days) is favored instead of using neutron capture reactions ($^{64}\text{Ni}(n,\gamma)^{65}\text{Ni}$) which needs Pneumatic irradiation rabbit system due to the short half live time of ^{65}Ni .

Fe and Ni were determined in blue-green algae using the reactions $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ (Mosulishvili et al., 2002). The iron isotopic abundances were determined in iron materials using threshold reactions (Burgio et al., 1995). Fast determination of Fe by instrumental neutron activation analysis through the formation of ^{56}Mn from both $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ and $^{56}\text{Fe}(n,p)^{56}\text{Mn}$ by using both reactor neutrons and epithermal neutrons irradiation was described and evaluated (Alfassi et al., 1984). ^{58}Co production from natural nickel in nuclear reactor was described with particular consideration of interfering nuclear reaction (Lincke, 1984).

The reaction $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ was used as a tool for neutron activation analysis of titanium in geological samples (Garcia et al., 1984). The $^{29}\text{Si}(n,p)^{29}\text{Al}$ reaction was used for determining silicon in both iron reference standard materials and steel alloys (Tomura et al., 2001; 2000). Thallium was determined in environmental samples using fast reactor neutrons (de Ruck, 1985) using the reaction $^{203}\text{Tl}(n,2n)^{202}\text{Tl}$.

Fast neutron reactions may also be chosen for the analysis in cases where the (n,γ) reaction product is also produced by an interfering reaction. For instance, the determination of aluminum in the presence of silicon, it may be better to use the $^{27}\text{Al}(n,p)^{27}\text{Mg}$ or $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ rather than $^{27}\text{Al}(n,\gamma)^{28}\text{Al}$, which may be seriously interfered with by the $^{28}\text{Si}(n,p)^{28}\text{Al}$ reaction (Tomura et al., 2000; Jonah et al., 2004; Arribère et al., 2000) depending on the relative concentrations of aluminum and silicon in the sample.

Aim of this work is the determination of the concentrations of Fe and Ni in geological samples and the GSJ Jb-1 reference material using threshold and neutron capture reactions.

2. Experimental

Nine geological samples representing some gold bearing rocks collected from eastern desert – Egypt (further details on the preparation of these samples and their k_0 – NAA will be reported in a forthcoming paper) and the GSJ-Jb1 reference material, altogether with the standard samples consisting of Au, Mo, Sn and Zr were co-irradiated in the inner irradiation site #2 near the core periphery of the second Egyptian research reactor (ETRR-2) for 2.5 hour. Before irradiation the samples were wrapped with clean aluminum foils. After a proper cooling time (4 -7 days), the Al-foils were removed, and the samples were transferred to clean Polyethylene vials. Further experimental details can be found in our pervious work (El Abd et al., 2007).

The fast neutron flux φ_f was determined using the threshold reactions $^{92}\text{Mo}(n,2n)^{92m}\text{Nb}$ and $^{95}\text{Mo}(n,p)^{95}\text{Nb}$. These reactions are characterized with two different threshold energies (see table 1).

3. Results and discussion

The results of the fast neutron flux determination using the reactions $^{92}\text{Mo}(n,2n)^{92m}\text{Nb}$ and $^{95}\text{Mo}(n,p)^{95}\text{Nb}$ are found; $\varphi_f = 1.45 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ and $\varphi_f = 1.3 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ respectively. At this irradiation position, the thermal and epithermal neutron fluxes are $\varphi_{th} = 7.7 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ and $\varphi_{ep} = 3.85 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ respectively. The neutron spectrum parameters α and f , needed to perform $k_0 - \text{NAA}$ were found (El Abd et al., 2007) to be: $\alpha = -0.01$ and $f = 20$. The value of the fast neutron flux determined via the reaction $^{92}\text{Mo}(n,2n)^{92m}\text{Nb}$ was used only in calculating the concentrations of Fe and Ni in the investigated samples since the gamma-line 934.5 KeV of ^{92m}Nb has better counting statistics than the gamma-line 765.8 KeV of ^{95}Nb ($^{95}\text{Mo}(n,p)^{95}\text{Nb}$). Moreover, the value of the cross section of the reaction $^{95}\text{Mo}(n,p)^{95}\text{Nb}$ is not accurately known in literature (0.267 mb and 0.35 mb) – in this work the first value is used to calculate the neutron flux.

Guevara et al. (2002) determined the fast neutron flux by using threshold reactions having neutron threshold energies range from 2.6 MeV to 13.4 MeV. They found the fast neutron flux does not depend on the neutron threshold energies. This can be checked in the #1 irradiation position characterized by the neutron spectrum parameters $\alpha = -0.028$, $f = 49$, $\varphi_{th} = 5.3 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ and $\varphi_{ep} = 1.08 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ (El Abd et al., 2007). The fast neutron flux was determined using the reactions $^{92}\text{Mo}(n,2n)^{92m}\text{Nb}$, $^{95}\text{Mo}(n,p)^{95}\text{Nb}$ and $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and are found to be $\varphi_f = 7.86 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$, $7.3 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ and $7.9 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$ respectively.

The concentration of Fe and Ni were determined in the nine geological samples using threshold reactions $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ respectively and the results are shown in table 2. They are the average values of four measurements together with their standard deviations. To credit our results, the concentration of Fe and Ni were determined in Jb-1 reference material using the same threshold reactions and the results are found to be $67 \pm 4 \text{ mg/g}$ and $133.4 \pm 13 \text{ }\mu\text{g/g}$. These results agree very well with recommended concentrations of 66.6 mg/g and $133 \text{ }\mu\text{g/g}$ for Fe and Ni respectively. Moreover, the Fe content of the JB-1 reference material determined in this work validates our previous results reported in (El Abd et al. 2007) ($66.2 \pm 8.6 \text{ mg/g}$) which was determined by neutron capture reaction using $k_0 - \text{NAA}$.

The $k_0 - \text{NAA}$ method was used to determine the Fe content in the geological samples. The results are shown in Table 2. As one can see, the Fe content determined by both nuclear reactions $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ agree very well. This agreement proves that the interference of Co via the threshold reaction $^{59}\text{Co}(n,p)^{59}\text{Fe}$ in

Fe determination via the neutron capture reaction $^{58}\text{Fe}(n, \gamma)^{59}\text{Fe}$ is negligibly small. The interference depends on the Co/Fe ratios in these samples. These ratios are less than 0.1%. The concentrations of Co and other elements in these geological samples using neutron capture reactions ($k_0 - \text{NAA}$) will be reported in a forthcoming paper.

The contribution of Mn via the reaction $^{55}\text{Mn}(n, 2n)^{54}\text{Mn}$ in Fe determination via the reaction $^{54}\text{Fe}(n, p)^{54}\text{Mn}$ is expected to be very small, because the high neutron threshold energy (11.6 MeV) and the small neutron cross section (0.258 mb).

Table 1 Nuclear data for some threshold reactions ; data were taken from Dorval et al. (2006)

Reaction	Threshold Energy (MeV)	σ (mb)	θ (%)	$t_{1/2}$	E_γ (KeV)	I (%)
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	2.6	111	68.077	70.86 d	810.76	99.45
$^{58}\text{Ni}(n, 2n)^{57}\text{Ni}$	13.4	0.00472	68.077	35.6 d	1377.63	81.7
$^{54}\text{Fe}(n, p)^{54}\text{Mn}$	2.8	81.7	5.845	312.12 d	834.8	99.97
$^{55}\text{Mn}(n, 2n)^{54}\text{Mn}$	11.6	0.258	100	312.12 d	834.8	99.97
$^{92}\text{Mo}(n, 2n)^{92m}\text{Nb}$	6	7.3	14.84	10.15 d	934.44	99.07
$^{95}\text{Mo}(n, p)^{95}\text{Nb}$	0.1	0.267	15.9	35.15 d	765.8	100

Table 2 elemental concentration of Fe and Ni by fast neutron activation analysis (FNAA) and $k_0 - \text{NAA}$

Sample code	Fe (mg/g)		Ni (ppm)
	FNAA	$k_0 - \text{NAA}$	FNAA
Sd#1	40.3 ± 3.4	40.7 ± 1.7	45.7 ± 3.8
Sd#2	41 ± 2.6	40 ± 1.2	181 ± 8
Sd#3	37.4 ± 1.7	37.8 ± 1.7	55.4 ± 2.3
Sd#4	64.2 ± 4.7	60.1 ± 3	208 ± 9
Sd#5	59 ± 1.7	60 ± 0.9	70 ± 5
Sd#6	42 ± 2	36 ± 0.6	6 ± 0.08
Sd#7	11.8 ± 0.5	11 ± 0.6	12 ± 0.5
Sd#8	54 ± 4	49 ± 1.7	137 ± 14
Sd#9	50.5 ± 1.6	48 ± 1.2	2595 ± 209

4. Conclusions

The concentrations Fe and Ni were determined in nine geological samples and the GSJ Jb-1 reference material by activation analysis with fast reactor neutrons using the

threshold reactions $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ respectively. The determined concentration of the samples and the GSJ Jb-1 reference material agree very well. The $k_0 - \text{NAA}$ method was used to determine the Fe content in the geological samples. The results are in good agreement with those determined using the threshold reactions.

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