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Determination of aluminium, silicon and magnesium in geological matrices by delayed neutron activation analysis based on k_0 instrumental neutron activation analysis

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HIGHLIGHTS

- In this work, concentrations of silicon, aluminium and magnesium in geological matrices were determined by INAA based on k_0-IAEA software.
- Silicon concentration is best estimated using ^{29}Si (n,p) ^{29}Al reaction within 5 min activation and 15–20 min delay before counting.
- The irradiation scheme overestimates magnesium concentration, so we propose a method able to account for quantitative contribution from other reactions.
- The method employed is quick and robust for estimating silicon, aluminium, magnesium and other elements using k_0-method.

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ABSTRACT

In this work, concentrations of silicon, aluminium and magnesium in geological matrices were determined by Neutron Activation Analysis based on k_0-IAEA software. The optimum activation and delay times were found to be 5 min and 15–20 min respectively for the determination of Si via ^{29}Si (n,p) ^{29}Al reaction. The adopted irradiation scheme did not work for the determination of magnesium. Each sample was irradiated under a thermal neutron flux density of 5.0×10^{11} n cm^{-2} s^{-1}. Cadmium covered activation indicated that a permanent epithermal irradiation site for research reactors would be very useful for routine determination of silicon in environmental samples.

1. Introduction

One of the advantages of Instrumental Neutron Activation Analysis (INAA) is its relative simplicity which allows less technical people to be trained on the job to be able to do elemental analysis. However determination of certain elements requires a (high) technical understanding of the activation (nuclear reaction) and/or decay mechanisms in order to obtain very good results. One such difficulty is the determination of aluminium (Al), silicon (Si) and magnesium (Mg). In many instances, the inability or difficulty to produce radioactive products by activation of low atomic number elements such as hydrogen (H), carbon (C), nitrogen (H), oxygen (O), and silicon (Si) had been considered as an advantage because it means less interference which translates to improved sensitivity (Robert et al., 2011). However, silica or silicates are major constituent of materials such as rocks, lavas, slags, refractories, ceramics, glasses, cements and ashes (samples for routine analysis by INAA). For most environmental and/or geological sample analysis, major elemental components like calcium, aluminium, magnesium as well as silicon are of great importance to the researcher. Therefore, there is the need to ensure high degree of accuracy in the determination of these elements by INAA.

There are two main kinds of interferences in the calculation of trace element concentration by INAA (Alfassi, 2000). The first is the formation of the same radionuclide from two different elements and the other is gamma interferences during spectrometry measurement

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due to gamma interactions with the detector medium. The former could be reduced by utilizing neutron interactions (reactions) other than the (n,γ) reaction, mostly produced by thermal neutron. Fortunately, typical neutron source(s) used for activation analysis (especially a research reactor) is constituted mainly of three energy groups. Thermal neutrons, epithermal neutrons and fast or fission neutrons, and their relative abundances depend on the reactor structure. Usually thermal neutrons of a typical research reactor constitute the majority of neutrons (up to about 90–95%). In the usual INAA, the whole reactor neutron energy spectrum is used for irradiation where each energy group may contribute to nuclear reaction of the target isotope in one or more of these interactions (n,γ), (n,n), (n,p), (n,2n) and or (n,α) reactions (Glascock, 2004). However, situations in which part of the neutron spectrum is used are also preferable due to the characteristic differences in the activation cross-sections for the desired and the interfering nuclides in the various parts of the energy spectrum or interfering gamma lines during spectrum acquisition.

It is however clear that an advantage can be taken of using the epithermal and fast neutrons in the case where the required element of interest is activated more strongly by them relative to the major elements or to the interfering element (branch activation analysis called Epithermal Neutron Activation Analysis – EPNAA). For example, Mg is normally determined through its less abundant isotope (^{26}Mg – 11.01%) by means of the (n,γ) reaction to form ^{27}Mg radionuclide [^{26}Mg (n,γ) ^{27}Mg]. However, the cross-section for this reaction is low (see Table 1 below), and makes the measurement not very sensitive.

More so, it suffers interference from the (n,p) reaction on ^{27}Al to form ^{27}Mg [^{27}Al (n,p) ^{27}Mg] which emits the same gamma energy (γ-line) as the (n,γ) reaction. Aluminium is also determined by the activity of its radionuclide ^{28}Al via the (n,γ) reaction of ^{27}Al [^{27}Al (n,γ) ^{28}Al]. It can also be produced by the (n,α) reaction on ^{31}P isotope [^{31}P (n,α) ^{28}Al] and by the (n,p) reaction on ^{28}Si [^{28}Si (n,p) ^{28}Al] (Alfassi and Lavi, 1984). In the case of Si determination, there are three stable isotopes involved: ^{28}Si (92.2%), ^{29}Si (4.7%) and ^{30}Si (3.1%), but the only radionuclide produced by the (n,γ) reaction is ^{31}Si. However, this radioisotope is barely a γ-ray emitter with an intensity of 0.07% at the 1266-keV γ-line. From Table 1, one can also observe that ^{31}Si has relatively high crossection and a longer half life (σ = 0.108 b, half life = 2.62 h) for which longer activation in an intense neutron source and longer decay time may prevent interference by the 1st escape peak of ^{28}Al γ-line to 1266 keV γ-line. This may allow the use of the (n,γ) reaction for the determination of Si (Kathryn, 1996). However, for low power research reactors, the activation of Si by higher energy neutrons (EPNAA) leads to the formation of ^{28}Al via both ^{31}P (n,α) ^{28}Al and ^{28}Si (n,p) ^{28}Al reactions and ^{29}Al by the ^{29}Si (n,p) ^{29}Al reaction. In cases where the concentration of Si is high enough (as in sediment, soil and rock matrices), Si can best be determined by the ^{29}Si (n,p) ^{29}Al reaction (Hancock, 1982) though the use of the ^{29}Al radioisotope may also reduce the accuracy of the measurement and limit the minimum amount (high detection limit) of Si that can be determined. The other problem associated with the measurement of ^{29}Al is that its main γ-line at 1273-keV is interfered by the 1st escape peak (1266 keV) from ^{28}Al of γ-line 1778 keV by virtue of pair production of γ-detector interactions.

In the Ghana Research Reactor-1 facility (GHARR-1), the interrelationship between the determination of Al, Si and Mg have not received much attention, as is the case in most of of the literature with few exceptions (Hancock, 1982; Kathryn, 1996; Alfassi and Lavi, 1984). This may be due to one or more of these reasons; GHARR-1 is a research reactor known for its high thermal neutron component (thermal to epithermal ratio (f)) of 18.8 (Akaho and Nyarko, 2002), and all elemental determination has been considered to be by means of thermal neutrons, the contribution from epithermal and fast neutrons are considered negligible except where conscious effort is made to irradiate the sample under a cadmium or boron shield. Hence the determination of Si is not entirely encouraged (not routine). Generally, most research reactors were designed without epithermal activation in mind and hence EPNAA would require conscious effort by the technician to use either boron or cadmium absorbers in order to achieve substantial (n,p) reactions (Hancock, 1982; Alfassi and Lavi, 1984). Also in facilities where the transfer systems are manually operated, this method (EPNAA) becomes radiologically inconvenient, time consuming and may not be employed for routine analysis except in a few cases when the need arises.

These seeming problems are compounded by the fact that the INAA quantification method employed in the GHARR-1 facility (and other INAA laboratories) for routine elemental analysis is the relative comparator method and in most cases multi-element analysis is of major interest and convenience. There is, however, no known reference standard which contains silicon that could give better counting statistics (if routine irradiation and counting is applied) in order to be used in quantitative analysis of silicon in samples, if at all, and the peak (1273-keV line) could be statistically appreciable for estimation. However, the current work is motivated by the fact that the k_0-method allows for a single comparator (gold monitor) element to be used in concentration evaluation of any identifiable element in the sample. As such the idea for this work would be to improve counting statistics for the 1273-keV γ-line of the sample and quantify the concentration based on a single comparator method (k_0 via k_0-IAEA software).

The flexibility, accuracy, precision and general acceptance of the k_0-INAA technique relative to classical NAA (relative comparator method), without the tedium for preparation of synthetic standards from pure substances, have been reported by Matthias and Blaauw (2006). With the k_0-Instrumental Neutron Activation, the analysis rather borrows on well characterized reactor neutrons (Baidoo et al., 2013) and constant in-situ thermal neutron flux monitoring (comparator monitor) during the irradiation of samples. Then, the comparator element can be used in routine measurements instead of needing separate standards for each element, based on equation (1) below: (Girardi et al., 1965):

$$\rho = \frac{((p_a/t_c)/SCDW)}{((p_{au}/t_c)/SCDw)} \frac{1}{k_0} \frac{[f + Q_0(\alpha)_{au}]}{[f + Q_0(\alpha)_a]} \frac{\varepsilon_{p_{au}}}{\varepsilon_p(E_y)} \quad (1)$$

Table 1
Relevant nuclear data for Mg, Al and Si [IAEA-TECDOC-564 (1990)].

Target isotope	σ (b)	RI	Isotopic abundance (%)	Reaction	Half-life	Energy (keV)	γ-Emission probability (%)
^{26}Mg	0.0372	0.024	0.1101	^{26}Mg (n,γ) ^{27}Mg	9.46 m	1014.43, 843.76	71.4
^{27}Al	0.226	0.16	1	^{27}Al (n,γ) ^{28}Al	2.24 m	1178.99	100
^{30}Si	0.108	0.106	0.031	^{30}Si (n,γ) ^{31}Si	2.62 h	1266.20	0.07
Crosssection averaged in a ^{235}U fission neutron spectrum [(n,p) reaction crosssection]							
Target isotope	$\bar{\sigma}$ (mb)	Isotopic abundance (%)		Reaction	Half-life (m)	Energy (keV)	γ-Emission probability (%)
^{28}Si	6.4	0.9223		^{28}Si (n,p) ^{28}Al	2.24	1178.99	100
^{29}Si	3.01	0.0467		^{29}Si (n,p) ^{29}Al	6.56	1273.36	91.30
^{27}Al	4.0	1		^{27}Al (n,p) ^{27}Mg	9.46	1014.43, 843.76	71.4