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Comparison of Epithermal and Instrumental Neutron Activation Analysis for the Determination of Uranium and Other Elements in Soil Samples Using BAEC TRIGA Reactor

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ABSTRACT
In this research, epithermal neutron activation analysis (ENAA) was implemented for the first time at 3 MW TRIGA research reactor of Bangladesh Atomic Energy Commission (BAEC) to determine Arsenic (As), Antimony (Sb), Samarium (Sm) and Uranium (U) in soil/sand samples and characteristically compared with instrumental neutron activation analysis (INAA) to find out advantageous features of ENAA over INAA. The ENAA detection limits (DL) were determined for the studied four elements (As, Sb, Sm and U) using standard reference materials (IAEA-Soil-7 and IAEA-SL-1) and compared with those of INAA. It is observed that DLs of As, Sb, Sm and U obtained by ENAA for SL-1 and Soil-7 are 17%, 42%, 28%, 25% and 29%, 25%, 31%, 35%, respectively lower than those obtained by INAA. The analytical sensitivities of those elements for SL-1 and Soil-7 are 31%, 9.0%, 75%, 28% and 25%, 11%, 15%, 48%, respectively higher than those obtained by INAA. Moreover, ENAA is the most effective if the Q_0 values of the interested nuclides are greater than 10. The calculated Q_0 values of As, Sb, Sm and U are 13.7, 20.3, 15.2 and 102, respectively. The findings from this research indicate that ENAA technique is better than INAA for the determination of trace amounts of As, Sb, Sm and U in soil samples using BAEC TRIGA Mark II research reactor. ENAA technique will be used for the determination of these elements, especially, U in soil/sand samples to find out potential U-mining areas in Bangladesh.

Keywords: Epithermal neutron activation analysis, detection limit, analytical sensitivity and Q_0 values.

1. Introduction

Neutron Activation Analysis (NAA) is a qualitative and quantitative elemental analytical method. NAA, based on the nuclear reaction between neutrons and target nuclei, is a useful method for the simultaneous determination of about 25–30 major, minor and trace elements in different sample matrices. An epithermal neutron activation analysis (ENAA) is a form of NAA with neutron energies ranging from 0.5 eV to 10 KeV. Nuclides with high epithermal neutron capture cross-sections are preferentially activated when irradiated under a cadmium filter. Therefore, elements with nuclides having high resonance integrals can potentially be determined by ENAA. Since the threshold energy of epithermal neutrons for Cadmium (Cd) is 0.5 eV, a thin foil of Cd (0.5-1 mm thick) strongly absorbs all neutron energies below this threshold energy [1]. For ENAA, a sample is irradiated in a cadmium box or in a borated capsule. This technique is used for the determination of trace amounts of elements in samples [2, 3]. In instrumental neutron activation analysis (INAA), thermal neutrons can activate major matrix elements, such as Aluminum (Al), Manganese (Mn), Sodium (Na), Chromium (Cr), etc. in geological samples because of their high thermal neutron capture cross-sections, whereas ENAA can largely reduce their radioactivities and enhance the sensitivity for some trace elements determination such as arsenic (As), Antimony (Sb), Samarium (Sm) and Uranium (U), etc. [4,5]. Therefore, ENAA can be a potential method for the determination of some elements, especially U in geological samples.

The 3 MW TRIGA Mark II research reactor of Bangladesh Atomic Energy Commission (BAEC) has been used for multiple purposes like training, education, radioisotope production and various R & D activities in the field of nuclear science and technology since 1986 [6,7]. Since its inception, BAEC TRIGA Reactor was used for the determination of major, minor and trace elements including Th and U in different sample matrices using INAA [8-10]. However, ENAA was not implemented at BAEC TRIGA Research Reactor (BTRR) due to the absence of a Cd-screened channel at the reactor. ENAA can also be implemented in the usual irradiation channel of a research reactor when samples along with standards are put into a Cd/B covered sample box.

The Rooppur nuclear power plant (2 x1200 MW) at Pabna, the first nuclear power plant in Bangladesh, is under construction. The uranium fuels for the plant will be supplied by Russia. However, it is very important to know the concentration level of U in soils and sands of Bangladesh to find out possible U-mining areas. Since ENAA can be used to determine a very low level of some elements like U in soil/sand samples, the characterization of ENAA at BAEC TRIGA reactor is needed to implement this technique. The main objective of this research is to compare ENAA and INAA at TRIGA Mark II research reactor of BAEC to determine

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U and some other potential elements in soil/sand samples.

2. Experimental setup

2.1 Sample collection and preparation

To characterize ENAA at TRIGA Mark II research reactor of BAEC, three reference standard materials: Certified reference material (CRM)-Soil-7 and SL-1 (lake sediment) from IAEA and standard reference material (SRM)-1633b from NIST, USA was used. Two sets of reference materials-one set in Cd-covered box and another set in bare were prepared for neutron irradiation.

About 50 mg of each reference material's powder sample was weighed in a polyethylene bag and double heat-sealed for neutron irradiation in the reactor.

2.2 Irradiation and measurement

Two sets of standards were put into polyethylene irradiation vial for neutron irradiation at the reactor. Three neutron flux monitors, Au-Al alloy (0.1% Au) at the bottom, middle and top of the stack of the samples were set to monitor neutron flux variation during irradiation. Samples were irradiated at dry central thimble (DCT) of the research reactor (figure 1) for 60 minutes at 500 KW reactor power. The thermal and epithermal neutron flux at DCT were 3.01 x 10^{13} and 1.3 x10^{12} $cm^{-2}s^{-1}$, respectively.

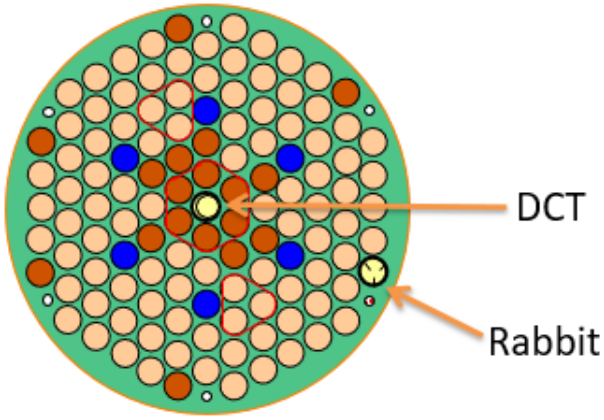

Fig. 1: Reactor core configuration and irradiation channels at BAEC TRIGA research reactor.

Three days later the irradiated samples were carried out from the reactor core to reduce activity induced in the Cd filter as well as to decay short-lived radionuclides in the standard samples. After irradiation, the activities of irradiated samples were counted using a high-resolution HPGe detector system. First and second gamma-ray counting was performed for 20 min. and 1h after a decay time of 3 days and 7 days, while the third counting was performed for 2h after a decay time of 30 days.

3. Results and Discussion

To compare the advantages of ENAA over INAA, three parameters such as detection limit (DL), analytical sensitivity and Q_0 values can be used. In this study, the above three parameters were examined for the determination of As, Sb, Sm and U. The relevant nuclear data for the determination of these parameters are given in Table 1.

Table 1: Relevant nuclear data for the four elements.

Elements	Product nuclide	Half-life	Energy (keV)	Ref.
As	^{76}As	26.5 h	559	4, 13
Sb	^{122}Sb	2.70 d	564	1, 13
Sm	^{153}Sm	46.8 h	103	1, 13
U	^{239}Np	2.36 d	278	1, 13

3.1 Detection limit

The detection limit represents the ability of a given analytical procedure to determine the minimum amounts of an element reliably. In this research, spectra collected for ENAA and INAA under the same experimental conditions are compared in Fig. 2. It is observed that ENAA spectrum has lower background than the INAA spectrum. In INAA, samples are activated by both thermal and epithermal neutrons and hence produce high activity. On the other hand, elements having nuclides with high epithermal neutron capture cross sections are preferentially activated in ENAA. Therefore, ENAA produces low background count relative to INAA. In NAA, the DL mainly depends on the irradiation, the decay and the counting conditions. The DL calculated using Currie's formula for the studied elements are tabulated in Table 2.

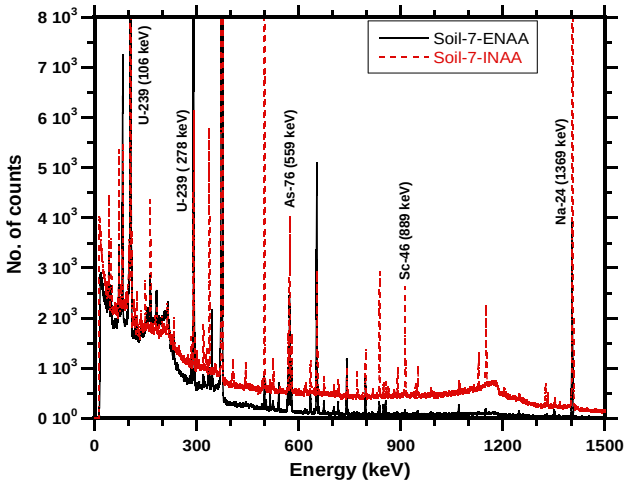

Fig. 2: Comparison of ENAA and INAA spectra for IAEA-Soil-7 under the same experimental conditions.

From Table 2, it is observed that the DL of As, Sb, Sm and U for SL-1 obtained by ENAA are 17%, 42%, 28% and 25%, respectively lower than those obtained by INAA. For Soil-7, the values for As, Sb, Sm and U obtained by ENAA are 29%, 25%, 31% and 35%, respectively lower than those obtained by INAA.

Table 2: Comparison of detection limit (mg/kg) for ENAA and INAA for IAEA-SL-1 and IAEA-Soil-7.

Elements	IAEA-SL-1			IAEA-Soil-7		
	ENAA	INAA	Ratio*	ENAA	INAA	Ratio*
As	0.29	0.35	0.83	0.27	0.38	0.71
Sb	0.070	0.120	0.58	0.090	0.120	0.75
Sm	1.00	1.39	0.72	0.88	1.28	0.69
U	0.40	0.53	0.75	0.34	0.52	0.65

Ratio*= DL of ENAA/DL of INAA.

Lower detection limits obtained by ENAA indicate better improvement of ENAA to determine studied elemental concentrations than those of INAA. Because lower detection limit means the ability of a technique to determine less amounts of an element in that sample and hence improvement in detection by that technique.

3.2 Comparison of Q values of ENAA with INAA

The ENAA is practically applicable if the nuclide of interest has higher Q_0 ($Q_0 > 10$) values [11-13]. The Q_0 values for ^{76}As, ^{122}Sb, ^{153}Sm and ^{239}Np (from ^{239}U) are calculated to 13.7, 20.3, 15.2 and 102, respectively (Table 3). The use of epithermal neutron activation is helpful because all four isotopes have a high ratio (Q_0 values). So, activation products will make the gamma-ray spectra simpler because of the reduction in activities in ENAA.

Table 3: Q_0 values for four elements (As, Sb, Sm and U).

Elements	σ_0 (b)	I_0 (b)	Q_0	Ref
As	4.60	63.0	13.7	13
Sb	6.25	127	20.3	1,13
Sm	206	3141	15.2	1, 13
U	2.70	275	102	1, 12, 13

Q_0 = Epithermal/thermal capture cross section

3.3 Analytical sensitivity

The analytical sensitivity (count per sec./unit mass) data using two methods are tabulated in Table 4. From Table 4, it is observed that for SL-1, the analytical sensitivities (cps/g) of As, Sb, Sm and U determined by ENAA are 31%, 9.0%, 75% and 28%, respectively higher than those obtained by INAA.

Table 4: Comparison of analytical sensitivity (cps/g) for ENAA and INAA in the case of SL-1 and Soil-7.

Elements	SL-1			Soil-7		
	ENAA	INAA	Ratio	ENAA	INAA	Ratio
As	9.86E7	7.52E7	1.31	8.44E7	6.78E7	1.25
Sb	2.06E9	1.89E9	1.09	2.09E9	1.89E9	1.11
Sm	0.30E9	0.17E9	1.75	0.21E9	0.18E9	1.15
U	0.50E9	0.39E9	1.28	0.52E9	0.35E9	1.48

Ratio = Analytical sensitivity of ENAA/INAA

For Soil-7, the values of the analytical sensitivities of As, Sb, Sm and U determined by ENAA are 25%, 11%, 15% and 48%, respectively higher than those obtained by INAA. In terms of analytical sensitivities, it can be concluded that ENAA is better than INAA for the determination of As, Sb, Sm and U in soil/sand samples.

4. Conclusions

In the present study, an ENAA method using a cadmium filter was characterized at BAEC TRIGA research reactor to determine trace levels of As, Sb, Sm and U without any chemical dissolution and separation, and compared with INAA. It is confirmed that ENAA is more preferable to INAA for the determination of As, Sb, Sm and U in soil/sand samples. This ENAA technique will be used for the determination of these elements, especially, U in soil/sand samples to find out potential U-mining areas in Bangladesh.

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