PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS OF BORON IN SAMPLES WITH A ^{241}Am-Be NEUTRON SOURCE

Şeref Turhan*, Haluk Yücel, Ahmet Demirbaş
Ankara Nuclear Research and Training Center (ANRTC)
06100 Beşevler, Ankara, Turkey

Abstract

A prompt gamma neutron activation analysis (PGAA) set-up installed at ANRTC has been used to analyze boron in samples. It consists of a 22.6% REGe detector and a 740 GBq ^{241}Am-Be neutron source moderated with water and paraffin. At the sample irradiation position, thermal neutron fluence rate measured was 2.36×10^4 n.m^{-2}.s^{-1} and the corresponding Cd-ratio was 22 for gold monitor. The absolute detection efficiency in the range of 120-1500 keV was determined using ^{152}Eu standard solution. The sensitivity and detection limit for standard boric acid samples has been determined and also the boron content in boric acid prepared from Turkish borate ores is measured to be 15.91 ±0.46% wt. by this PGAA set-up.

1. Introduction

Neutron-induced PGAA is well known, a rapid, non-destructive and instrumental technique that complements conventional neutron activation analysis (INAA). The technique is most useful for determining light elements (H, B, C, N, Si, P, S and Cl) and the elements having with a large neutron capture cross-sections (Cd, Sm and Gd) in samples by irradiating them continuously with neutrons. In principle, PGNAA is based on detection of capture gamma rays emitted within 10^{-14} to 10^{-12} s by target material while it is being irradiated with neutrons. Nuclei formed in neutron-capture reactions have excitation energies equal to the binding energy of the added neutron. The excitation energy is released by emission of prompt gamma rays of which the energies range from about 100 keV to 10 MeV. Thus, the elemental concentration is retrieved for the identified elements in any matrix[1-4].

About 40 PGAA facilities at many research institutes have been reported in the literature[3,5-8]. Some of these facilities have devoted substantial time to PGAA activities and other ones have demonstrated the ability. The general trend in these facilities consists of an extracted neutron beam from reactor core that impinges on a sample target to produce the neutron-capture gamma reactions. However, PGAA facilities using accelerator based neutron generators such as D-T and radioisotopic neutron sources such as ^{252}Cf and ^{241}Am-Be has been widely tested for on-line and in-situ elemental analysis in various fields such as industrial, mineral and oil exploration, bulk detection of illicit drugs, explosives and other contraband materials[9-21].

Since the cross-section for the $^{10}B(n,\alpha\gamma)^{7}Li^{*}$ reaction is exceptionally large (σ_{th} = 3840 b)[22] boron is one of the sensitive elements in PGAA and a number of analyses have been reported for biological, geological and food samples[23-30]. This determination is usually made by detecting the prompt 478 keV γ-ray emitted by the recoiling $^{7}Li^{*}$ nucleus ($t_{1/2}$=1,05x10^{-13} s). This reaction involves only the ^{10}B, which has approximately 19.8% abundance in naturally occurring B (i.e. σ_{th} = 768 b). The 478 keV photopeak has a broad line shape width that is about 8-10 times larger than that of normal γ-peaks as a result of Doppler broadening. Moreover, the line shape depends on the chemical and physical nature of the boron compounds[21].

In this study, firstly the Cd-ratio and corresponding thermal neutron fluence rate at irradiation position was measured using a gold monitor. Also, Monte Carlo code of particle transport MCNP-4B was used to calculate thermal neutron fluence rate. Secondly, the high level of gamma ray background arising from various origins such as 0.0596 MeV (35.9%) from ^{241}Am, 4.433 MeV (0,6 γ /n) from the excited state of $^{13}C^{*}$ via $^{9}Be(\alpha,n)^{12}C$ from the

* Corresponding author. E-mail address: seref.turhan@taek.gov.tr (Seref TURHAN)

neutron source and 2.223 MeV (100%) from $^{1}H(n,\gamma)^{2}H$ in moderator were analyzed. Then the sensitivity and detection limit of the standard boric acid samples and boron content in boric acid prepared from Turkish borate ores were determined as the first application of the ANRTC PGAA set-up.

2. Experimental

PGAA set-up

The PGAA set-up with a 740 GBq ^{241}Am-Be neutron source, installed in a room at ANRTC, is shown in Fig.1. The physical dimensions of the irradiation unit are 132 cm(W) x 132 cm(L) x 103 cm(H). In the present irradiation unit, the neutron source placed in a polypropylene tube was positioned in a 55 cm diameter cylinder tank made of polypropylene as well. It was coaxially positioned in the geometrical center of lead rings with the thickness of 6 cm (internal diameter: 9 cm, outer diameter: 21 cm and total height: 33 cm) as shown Fig. 1. Thus, the gamma rays emitted directly from the source itself was reduced substantially by using the lead rings placed in the cylinder tank. The moderator tank was additionally shielded with paraffin in all sides to prevent the detector sides from fast neutrons. The thickness of paraffin at the front side of the tank is 28 cm and other sides of it are 18 cm, respectively. Two surfaces of the neutron irradiation unit was also shielded by using chevron type lead bricks in total thickness of 18 cm, and the other surfaces are back the concrete walls in the thickness of 1 to 2 m in a basement. The background-prominent gamma rays, which is especially the 2.223 MeV gamma ray from the $^{1}H(n,\gamma)^{2}H$ reaction (σ_{th} = 0.333 b for thermal neutron) formed in hydrogenous materials used for neutron moderation was reduced remarkably in view of the permissible gamma dose for overall irradiation room. The measured gamma dose rate is less than 40 μR/h and the estimated neutron dose rate is less than 1 mrem/h at working area. The neutrons thermalized in moderator travel through the hole with 6

cm diameter for sample irradiations. The detector was shielded with both the powder of natural lithium carbonate (Li_2CO_3) contained in a hollow wooden box (powder thickness: 5 cm at front of detector window, 1.5 cm on the sides of the Al end-cap of the detector) and 1 mm thick cadmium cap against thermal neutrons. The ^{6}Li nuclide (7.52% abundance in natural lithium) captures thermal neutrons by the reaction $^{6}Li(n,^{3}H)^{4}He$ and produces no gamma rays. Cadmium is efficient and economical absorbers but thermal neutrons by the absorption reaction $^{113}Cd(n,\gamma)^{114}Cd$ produces additional prompt gamma such as 0.559 MeV and 0.651 MeV, etc. and it can become radioactive in time. Both the neutron shields encasing the detector end-cap are surrounded additionally by polyethylene bricks in 5 cm thickness to thermalize stray fast neutrons. These neutron shields can be removable from the end-cap. In case of Cd analysis, the metal-cylindrical Cd shield case with thickness of 1 mm can be removed from the end cap of Ge detector. However, only shield containing lithium carbonate is removed while boron is being analyzing. Finally, the detector assembly is surrounded by the chevron lead bricks with a 10 cm thick to reduce gamma-ray background.

Gamma-ray detection system

The basic gamma-ray detection system consists of a REGe detector with its own preamplifier with a side-looking type LN_2 Dewar, a spectroscopy amplifier, a 8K ADC and a conventional Canberra 35^{+} type multichannel analyzer (MCA). The REGe detector has a relative efficiency of 22.6%, an energy resolution of 1.80 KeV at 1332.5 keV (^{60}Co) and of 0.97 keV at 122 keV (^{57}Co), and a peak-to-Compton ratio of 53.5:1. Data acquisition system allows the collection of only one type of the spectrum at a time because at present a single-input MCA is used.

3. Results and discussion

The efficiency calibration of a REGe detector over a wide range of energy from a few keV to about 10 MeV which can only be covered with data sets from different radioactive sources and from (n,γ) reaction combined (such as $^{14}N(n,\gamma)$, $^{35}Cl(n,\gamma)$, $^{59}Co(n,\gamma)$, etc.)[31-32] can be determined. In the present work, the absolute efficiency at a 77,5 cm source-to-detector distance which eliminates the coincidence summing losses and energies in the range 120-1500 keV was measured by using ^{152}Eu standard ampoule sources (5 ml) in the case of Li_2CO_3 case and Cd cap. The experimental data are fitted to the functions using least square fitting method (Fig. 2). The relative deviations between the experimental efficiency and the calculated ones remained within 0.017- 3%. The efficiency calibrations have not yet completed for higher energy range up to 10 MeV.

Thermal fluence rate at the irradiation position was measured by activating ^{197}Au foils without and with the cadmium cover and then counting the delayed gamma-rays of 0.4118 MeV energy on the coaxial p-type HpGe detector for which calibrated for efficiency measured using ^{152}Eu standard source. Thus, thermal fluence rate and cadmium ratio was measured $2.36x10^4 n.cm^{-2}.s^{-1}$ and 22, respectively. Also thermal fluence rate at this position was calculated as $2.27x10^4 n.cm^{-2}.s^{-1}$ by using MCNP code[33].

An experiment was carried out using the standard boric acid. The observed 478 keV peak is 3 times higher than that of background of 0.77 cps for $^{10}B(n,\alpha)^7Li$ reaction. After subtracting the background from the 478 keV peak[22], detection sensitivity for standard boric acid is estimated to be 0.27 cps/g-B. The detection limit in the weight dimension is proportional to the square root of the background region of interest, N_{Bqd}, and inversely proportional to both the sensitivity, S, and counting time, t_c. Then the detection limit for boron element can be calculated as about 43 mg level for a counting time of 60,000 s by using following equation[34],

$L_D = 3.29.(N_{Bqd})^{1/2} / t_c.S$

Then an application was made to determine boron content in the boric acid samples prepared from Turkish borate ore. The time for irradiation of boric acid samples at present PGAA set-up was chosen to be 24,300 s. The net count rates after substracting background from sample counts for 478 keV peak has been calculated for standard and Turkish boric acid samples. From the net count rates, the boron content in Turkish boric acid has been determined to be 15.91 ±0.46% wt. The lower detection limit can be achieved by extending the counting time and increasing thermal neutron fluence incident on the sample.

The singles gamma ray spectra from neutron capture are extremely complex. Hence, the analysis of prompt gamma ray spectra requires a high quality fitting computer code for data analysis. However, at present, automatic element identification on the basis of peak energies is not possible because of not available a built-in prompt gamma library. Therefore, the peak energies observed in the spectra are selected manually on the display of MCA by the analyst. Since the most important drawback of the present technique is the high level of the background arising from various origins; main background peaks are the following:

- Neutron captures in the elements constitutive of the experimental set-up, essentially from H, C, Pb, Li and Cd. The 2.223 MeV gamma ray from hydrogen is observed as the most important peak in the spectra,
- The recoiling ^{7}Li nucleus, initially in an excited state, decays with 94% by Doppler broadened 0.478 MeV prompt gamma ray, and the $^{113}Cd(n,\gamma)^{114}Cd$ reaction in the neutron shield produces additionally the prompt gamma rays, and

- The 4.433 MeV Doppler broadened gamma ray from the neutron source together with it single escape (3.922 MeV) and double escape (3.411 MeV) peaks, and the high Compton scattering background due to the large number of gamma rays.

Conclusions

The present work describes the first application of PGAA set-up at ANRTC. Since the estimated thermal neutron fluence rate of about order of 10^4 $n.cm^{-2}.s^{-1}$ is low, the expected sensitivity for the elements considered would be low. The detection limit for boron element is about 43 mg level for a counting time of 60,000 s using standard boric acid. It is possible to achieve a lower detection by extending the counting time and increasing thermal neutron fluence rate incident on the sample. Therefore, the necessary modifications in the source-detector geometry will be made resulting in an increase in the thermal neutron fluence rate. In future, it is planned to built to enable a digital spectrum analyzer based on digital signal processing technique (DSA) or a data acquisition system. This enables us an effective and fast data evaluation by using a gamma software. Isotopic neutron source as ^{241}Am-Be are available and easily transportable. This technique may be developed for in situ application for borate ores exploitation and mining.

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FIGURE CAPTIONS

Fig. 1. A schematic diagram of PGAA set-up (PE: Polyethylene)

Fig. 2. Absolute full-energy paek detection efficiency curve of the REGe detector

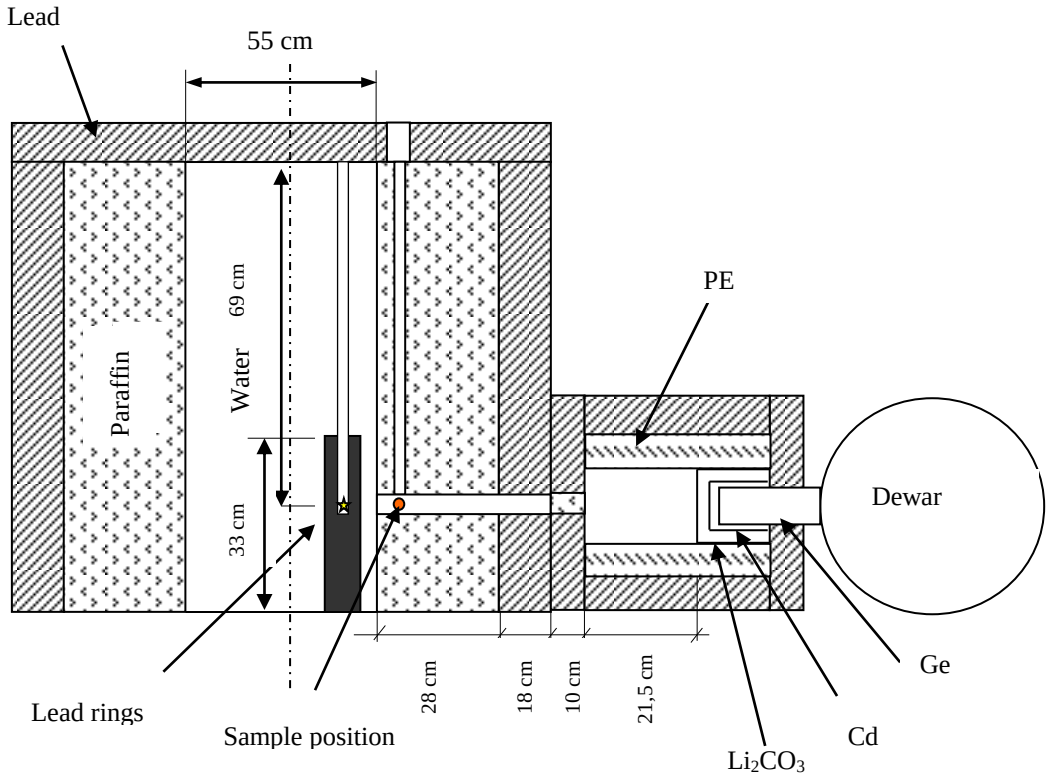
Lead
55 cm
69 cm
Water
Paraffin
33 cm
PE
Dewar
Ge
Lead rings
Sample position
28 cm
18 cm
10 cm
21,5 cm
Cd
Li_2CO_3

Fig. 1

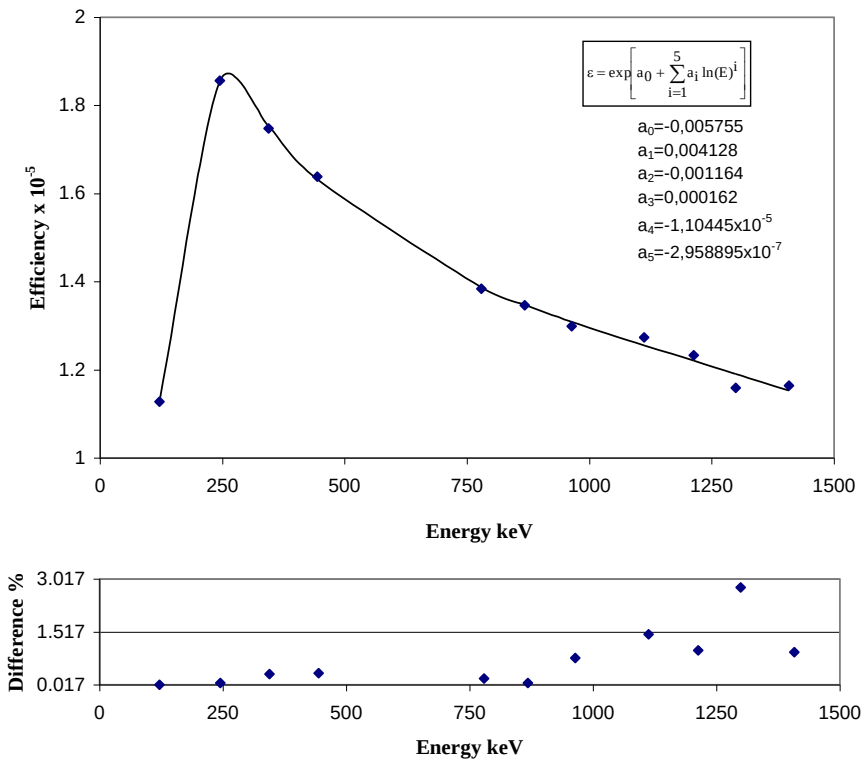
$\varepsilon = \exp\left[a_0 + \sum_{i=1}^{5} a_i \ln(E)^i\right]$
a_0=-0,005755
a_1=0,004128
a_2=-0,001164
a_3=0,000162
a_4=-1,10445x10^{-5}
a_5=-2,958895x10^{-7}
Efficiency x 10^{-5}
2
1.8
1.6
1.4
1.2
1
0
250
500
750
1000
1250
1500
Energy keV
Difference %
3.017
1.517
0.017
0
250
500
750
1000
1250
1500
Energy keV

Fig. 2