Journal of Radioanalytical and Nuclear Chemistry, Vol. 262, No. 3 (2004) 661–664

Prompt gamma neutron activation analysis of boron with a ^{241}Am–Be neutron source

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(Received May 10, 2004)

A prompt gamma neutron activation analysis (PGAA) setup installed at ANRTC has been used to analyze boron. 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, the thermal neutron fluence rate measured was $2.36 \cdot 10^4$ 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. The boron content in boric acid prepared from Turkish borate ores is measured to be 15.91±0.46% wt.

Introduction

Neutron-induced PGAA is a well known, 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 with a large neutron capture cross sections (Cd, Sm and Gd) by irradiating them continuously with neutrons. In principle, PGNAA is based on detection of capture gamma-rays emitted within 10^{-14} to 10^{-12} seconds by the target material while it is 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 in the energy 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 with substantial time to PGAA activities. The general feature of these facilities consists of an extracted neutron beam from a 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 radioisotope neutron sources such as ^{252}Cf and ^{241}Am–Be have 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,αγ)^{7}Li* reaction is exceptionally large ($\sigma_{th} = 3840$ b)[22] boron is one of the most sensitive element 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 ($T_{1/2} = 1.05 \cdot 10^{-13}$ s). This reaction involves only ^{10}B, which has approximately an 19.8% abundance in naturally occurring B (i.e., $\sigma_{th} = 768$ b). The 478 keV photopeak, as a result of Doppler broadening, has a broad line shape width that is about 8–10 times larger than that of normal γ-peaks. Moreover, the line shape depends on the chemical and physical nature of the boron compound.[21]

In this study, the Cd-ratio and corresponding thermal neutron fluence rate at irradiation position was measured first using a gold monitor. Also, a Monte Carlo code of particle transport MCNP-4B was used to calculate the thermal neutron fluence rate. Then, 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(α,n)^{12}C from the neutron source and 2.223 MeV (100%) from ^{1}H(n,γ)^{2}H in moderator were analyzed. The sensitivity and the detection limit of the standard boric acid samples were determined and also the boron content in boric acid prepared from Turkish borate ores, as the first application of the ANRTC PGAA setup.

Experimental

PGAA setup

The PGAA setup 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) × 132 cm (L) × 103 cm (H). The neutron source is placed in a polypropylene tube, positioned in a 55 cm diameter cylindrical tank made of polypropylene as well. It is 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 in Fig. 1. Thus, the gamma-rays emitted directly from the source was reduced substantially by using the lead rings placed in the cylinder tank.

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0236–5731/2004/USD 20.00
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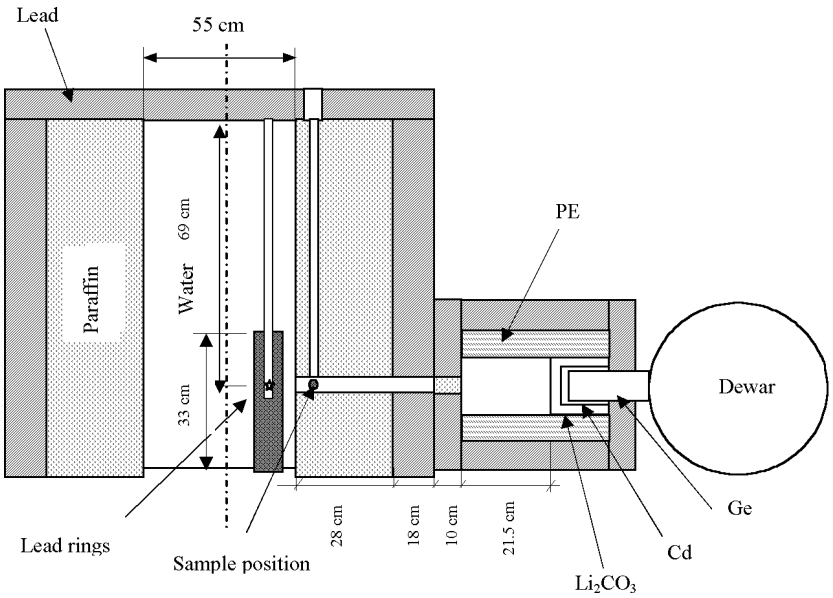


Fig. 1. Schematic diagram of the PGAA setup (PE: polyethylene)

The moderator tank was shielded additionally with paraffin in all sides to protect the detector sides from fast neutrons. The thickness of paraffin at the front side of the tank was 28 cm and other sides of it were 18 cm, respectively. Two surfaces of the neutron irradiation unit were also shielded by using chevron type lead bricks of a thickness of 18 cm, and the other surfaces were behind the concrete walls of a thickness of 1 to 2 m in a basement. The background-prominent gamma-rays, which are especially the 2.223 MeV ones from the $^{1}H(n,\gamma)^{2}H$ reaction ($\sigma_{th} = 0.333$ b for thermal neutrons) formed in hydrogenous materials used for neutron moderation, were reduced remarkably in view of the permissible gamma-dose for the overall irradiation room. The measured gamma-dose rate was less than 40 μR/h and the estimated neutron dose rate less than 1 mrem/h at the working area. The neutrons thermalized in the moderator travel through the hole with 6 cm diameter for sample irradiations. The detector was shielded with both a powder of natural lithium carbonate (Li_2CO_3) contained in a hollow wooden box (powder thickness: 5 cm at the front of detector window, 1.5 cm on the sides of the Al end-cap of the detector) and a 1 mm thick cadmium cap against thermal neutrons. The ^{6}Li nuclide (7.52% abundance in natural lithium) captures thermal neutrons by the $^{6}Li(n,^{3}H)^{4}He$ reaction and produces no gamma-rays. Cadmium is an efficient and economical absorber but thermal neutrons by the absorption $^{113}Cd(n,\gamma)^{114}Cd$ reaction produce additional prompt gamma-rays such as 0.559 MeV and 0.651 MeV, etc. and can become radioactive with time. Both the neutron shields encasing the detector end-cap are surrounded additionally by polyethylene bricks of 5 cm thickness to thermalize stray fast neutrons. These neutron shields are 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 the Ge detector. However, only the shield containing lithium carbonate is removed while boron is being analyzed. Finally, the detector assembly is surrounded by chevron lead bricks with a 10 cm thickness to reduce the 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. The 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.

Results and discussion

The efficiency calibration of a REGe detector over a wide range of energies from a few keV to about 10 MeV which can only be covered with data sets from different radioactive sources and from (n,γ) combined reactions (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 an ^{152}Eu standard ampoule source (5 ml) in the case of Li_2CO_3 case and Cd cap. The experimental data are fitted to the functions using a least square fitting method (Fig. 2). The relative deviations between the experimental efficiency and the calculated ones remained within 0.02–3%. The efficiency calibrations have not yet been completed for higher energy range up to 10 MeV.

The thermal fluence rate at the irradiation position was measured by activating ^{197}Au foils without and with cadmium cover and then counting the delayed gamma-rays of 0.4118 MeV energy with a coaxial p-type HpGe detector, calibrated for efficiency measurement using ^{152}Eu standard source. Thus, the thermal fluence rate and the cadmium ratio was measured as $2.36\cdot10^4$ $n\cdot cm^{-2}\cdot s^{-1}$ and 22, respectively. Also the thermal fluence rate at this position was calculated as $2.27\cdot10^4$ $n\cdot cm^{-2}\cdot s^{-1}$ by using MCNP code.[33]

An experiment was carried out using standard boric acid. The observed 478 keV peak was 3 times higher than that of the background of 0.77 cps for the ^{10}B(n,α)^{7}Li reaction. After subtracting the background from the 478 keV peak,[22] the 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 sensitivity, S, and counting time, t_c. Then the detection limit for boron element can be calculated and was about 43 mg for a counting time of 60,000 seconds by using the equation:[34]

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

Then an application was made to determine boron content in boric acid prepared from Turkish borate ore. The time for irradiation of boric acid samples at the present PGAA setup was chosen to be 24,300 seconds. The net count rates after substracting the 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 the thermal neutron fluence incident on the sample.

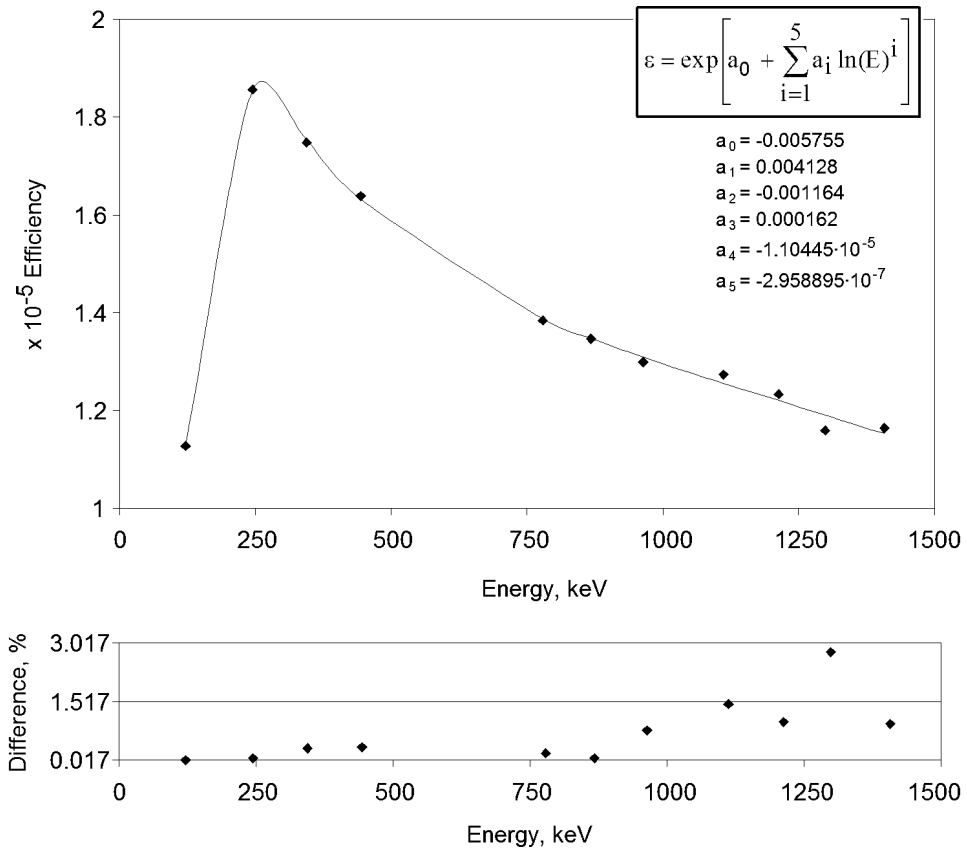


Fig. 2. Absolute full-energy peak detection efficiency curve of the REGe detector

The single 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 energy is not possible because of the lack of a built-in prompt gamma library. Therefore, the peak energies observed in the spectra are selected manually by the analyst on the display of the MCA. The most important drawback of the present technique is the high level of the background arising from various origins. The main background peaks are the followings:

Neutron captures in the elements constitutive of the experimental setup, essentially from H, C, Pb, Li and Cd and the 2.223 MeV gamma-rays from hydrogen, 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,γ)^{114}Cd reaction in the neutron shield produces additional prompt gamma-rays.

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 setup at ANRTC. Since the estimated thermal neutron fluence rate of about 10^4 n·cm^{-2}·s^{-1} is low, the expected sensitivity for the elements considered would be low. The detection limit for boron is about 43 mg for a counting time of 60,000 seconds using standard boric acid. It is possible to achieve a lower detection limit by extending the counting time and increasing the 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 the future, it is planned to built a digital spectrum analyzer based on digital signal processing technique (DSA) to enable a data acquisition system. This will allow an effective and fast data evaluation by using a gamma software. Isotopic neutron sources as ^{241}Am–Be are available and easily transportable. This technique may be developed for in situ application for borate ore exploitation and mining.

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