Volume 17, No 2 | International Journal of Radiation Research, April 2019

Characterization of Am-Be neutron source based PGNAA setup using aqueous solutions of Chlorine and Boron

D. Meena[1], S.K. Gupta[1*], H.S. Palsania[1], N. Jakhar[1], N. Chejara[1], P. Meena[2]

[1]Department of Physics, University of Rajasthan, 302004 Jaipur, India
[2]Department of Zoology, University of Rajasthan, 302004 Jaipur, India

► ***Original article***

Corresponding authors:
S.K. Gupta, Ph.D.,

E-mail: *shail2405@gmail.com*

Revised: September 2018
Accepted: October 2018

Int. J. Radiat. Res., April 2019; 17(2): 301-308

DOI: 10.18869/acadpub.ijrr.17.2.301

ABSTRACT

Background: A 5 Ci 241Am-Be radio isotopic neutron source-based prompt gamma neutron activation analysis (PGNAA) setup was designed for estimation of minimum detectable concentration (MDC) of elements in aqueous solutions. ***Materials and Methods***: Performance variables related to PGNAA setup (source to sample distance, sample to detector distance and volume of water) were optimized experimentally. Characterization of the setup was done by chlorine and boron elements using sodium chloride (NaCl) and boric acid (H_3BO_3) compounds respectively. ***Results:*** PGNAA setup was calibrated for different concentrations of Cl and B in aqueous solution. The chlorine concentration was varied over 1.2 g/L, 2.4 g/L, 3.6 g/L, 4.8 g/L, 6.0 g/L and 7.2 g/L in water samples while boron concentration was varied over 0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L and 0.5 g/L. The MDC of chlorine and boron were calculated for various characteristic prompt gamma energies. Estimated MDC of chlorine and boron are 175 ± 53 ppm at prompt gamma energy 6110 keV and 3 ± 0.95 ppm at prompt gamma energy 478 keV respectively. ***Conclusion:*** In the present work, PGNAA setup was developed and characterized for the in -situ analysis of aqueous solutions using a 5 Ci Am-Be neutron source. The chlorine concentration was varied from 1.2 to 7.2 g/L and boron concentrations were varied from 0.1 to 0.5 g/L. The setup shows linear response for both chlorine and boron for wide energy range. The obtained results were also compared with other previous published work. It shows good agreement with present results.

Keywords: *PGNAA, minimum detection concentration, ^{241}Am-Be neutron source, HPGe detector.*

INTRODUCTION

The PGNAA technique is well established non -destructive technique for detection of trace component of elements in the samples [1-2]. This technique allows fast, accurate and discrete sampling of elements irrespective of their physical and chemical form. In this technique, samples of the interest are irradiated by neutrons and characteristic prompt gamma emitted due to inelastic reaction (n,n',g) and neutron capture reactions (n,g) are detected. Reactor, neutron generator and radio isotopic sources can be used as a neutron source. The radio isotopic neutron sources are compact, transportable and relatively economical and have long half-life [2]. Radio isotropic neutron source based PGNAA setups are useful for in-situ and online multi-elemental analysis in wide variety of applications [3-7]. A wide range of radio isotopic source based PGNAA setup for analysis of bulk concrete sample [8], cement raw material [9], domestic waste water and industrial liquid effluent [3], detection of heavy metals in

sediments [5] saline water [10] and cancer tumor treatment [11] have been developed in recent years. PGNAA setup require a sophisticated geometric arrangement. Geometrical arrangement of this setup was optimized to achieve higher neutron flux at sample volume and better detection limit.

In the series of elements detection in aqueous solution by PGNAA technique, many set-up have been developed by different researcher. WenBao *et al*. developed a set-up includes a 300 mCi ^{241}Am-Be neutron source and a 4 - inch BGO detector. The Am-Be source placed inside the aqueous solution. The geometry of set-up is optimized by simulation code (MOCA) to enhance detection efficiency and decrease the measurement time [12].

In the present work, 5 Ci Am-Be neutron source (emitting ~ 1.25×10^7 n/sec [13] and HPGe detector based PGNAA setup for detection of elements in aqueous solution is proposed. In this study, the minimum detection limit of PGNAA setup is estimated. Parameter related to geometrical arrangements like volume of sample, and distance of sample from source and detector were optimized experimentally. Characterization of PGNAA setup was performed using various concentrations of sodium chloride (NaCl) and boric acid (H_3BO_3) aqueous solution. The minimum detectable concentration was obtained for Cl and B in aqueous solution. The setup will be used for detection of heavy metals, in soil and water, and salinity of water.

MATERIALS AND METHODS

Detector efficiency

A gamma spectroscopy system with a Canberra Coaxial HPGe (GC1518) semiconductor detector is used for the measurements. It is operated under a high voltage of 2.5 kV, equipped with a Canberra digital spectrum analyzer (DSA1000) with 8192 channels working with the acquisition software Genie 2000. These are supplied by Canberra Industries from USA. The performance parameters of Canberra HPGe detector are as follows: relative efficiency 16.4 %, FWHM 1.63 keV at 1332.5 keV (^{60}Co). The efficiency of the HPGe-detector is calculated using standard gamma sources. In order to eliminate the coincidence loss, all sources were individually placed at 25 cm far from the detector surface [14]. The fifth order polynomial of the form (equation 1) is used as fitting functions for absolute full energy peak efficiency and shown in figure 1.

$$\epsilon_\gamma = exp\left(\sum_{i=0}^{4} a_i \left(\ln E_\gamma\right)^i\right) \quad (1)$$

The PGNAA Setup

The PGNAA setup consist of 5Ci ^{241}Am-Be neutron source, a source tank, a coaxial HPGe detector and a plastic sample container. HPGe detector was placed such that the detector end cap face remains perpendicular to the sample container axis and midpoint of the sample container height matches with the center of detector face. The complete arrangement for experiment was done inside the lab which was shielded properly by concrete. Figure 2 shows a schematic diagram for the PGNAA setup.

Source background minimization and detector shielding

^{241}Am-Be neutron source background contains 4438 keV gammas from ^{9}Be(α,n)^{12}C reaction emitted directly from neutron source and 2223 keV gammas from ^{1}H(n,γ)^{2}H reaction (σ_{th}=0.333 b) due to thermal capture of neutron by the hydrogen present in paraffin. To enhance the signal to noise ratio and minimize gamma background from source, top of the source tank was covered with two lead disks (5 cm thick, 36 cm diameter), which almost eliminates 4443 keV gamma rays. Further, 2223 keV gammas were minimized by placing 12 cm thick lead bricks over lead disks.

Emitted neutrons from the source interact with shielding and surrounding materials and produce additional background. In order to prevent this additional background, detector active volume was covered with 2 cm thick lead (Pb) rings. For minimization of neutron induced radiation damage in the active volume of the detector, a 4cm thick container filled with natural lithium carbonate (Li_2CO_3) powder and

0.5 mm thick cadmium foil is placed in front of detector end cap. The ^{6}Li nuclide captures thermal neutrons by the $^{6}Li(n,^{3}H)^{4}He$ reaction and produces no gamma-rays. While, ^{113}Cd absorbs thermal neutrons by the absorption reaction $^{113}Cd\ (n,\gamma)^{114}Cd$ but produces 559 keV and 651 keV etc. prompt gamma-rays [15]. 4 cm thick natural lithium carbonate (Li_2CO_3) placed in front of the detector face absorbs about 23.4% neutrons and about 3.93% of neutrons are further absorbed by 0.5 mm thick cadmium foil. The scattered neutrons enter in the HPGe detector active volume which have fast and thermal neutron components. Fast neutron interacts inelastically with five natural isotopes of germanium while thermal neutrons captured by isotopes. In both cases, characteristics gamma rays emit from Ge crystal. These gammas can be used to characterize the neutron field entering in the detector volume [16].

Optimization of PGNAA setup performance variables

Important performance variables of PGNAA setup such as - aqueous sample volume, source to sample and sample to detector distance were optimized experimentally. Radio isotopic neutron source has relatively lower neutron flux compare to the reactor. Thus, maximizing average thermal neutron flux at sample volume and minimization of gamma rays which emits from neutron source itself and gamma ray emission from surrounding and shielding materials due to neutron activation. These were main focus of the optimization strategy.

Hydrogen peak counts (Eγ = 2223 keV) are used as indicator of neutron flux at sample volume. Cylindrical plastic containers of different sizes were used as sample container for 1L, 2L, 3L, 5L and 10L sample volume. Variation of hydrogen peak count rate with water volume is shown in figure 3. Hydrogen peak count rate is exponentially increasing function of water volume and saturate at about 10L.

Spectrums were recorded and analyzed for different sample to detector and sample to source distance combination with 5L water samples. 595.81 keV germanium gamma count rate (Figure 4(a)) and hydrogen peak count rate (figure 4(b)) decreases exponentially with source distance and attain constant value at source distance 25 cm.

Finally, Sample volume was fixed at 5L. The optimal sample to source and sample to detector distance were fixed at 25 cm and 10 cm respectively. Dead time of detector was about 5% at this geometrical arrangement.

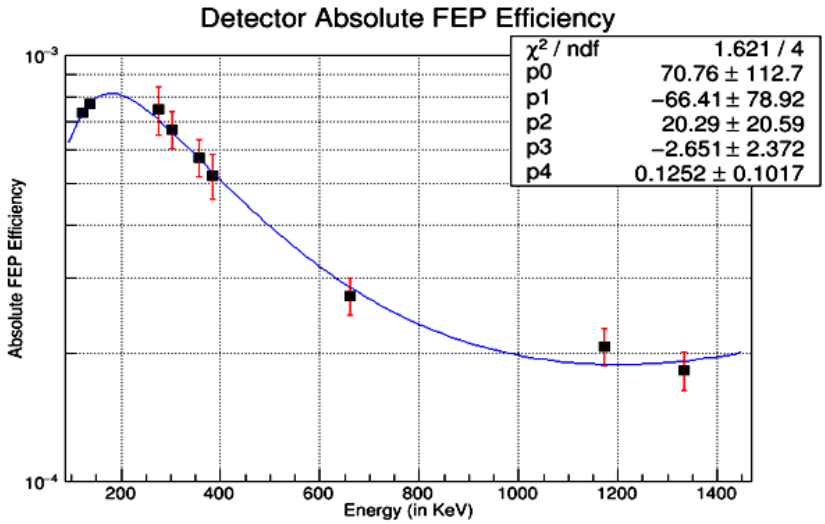

Figure 1. The absolute efficiency of HPGe detector with gamma ray energies (80 keV to 1332 keV).

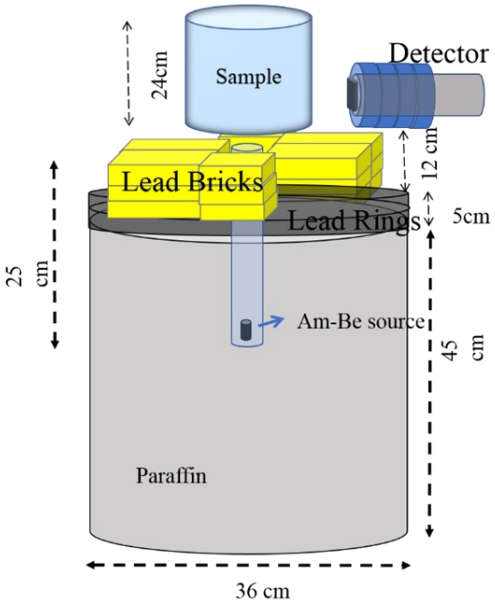

Figure 2. Schematic diagram of PGNAA setup in this work.

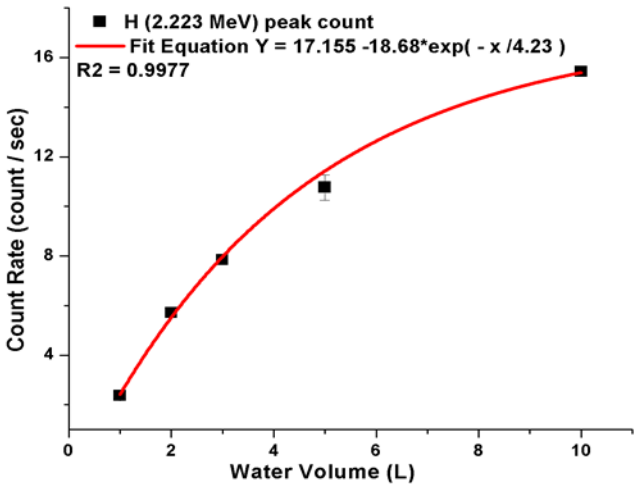

Figure 3. Variation hydrogen prompt gamma (2223 keV) ray peak count rate with volume of sample container.

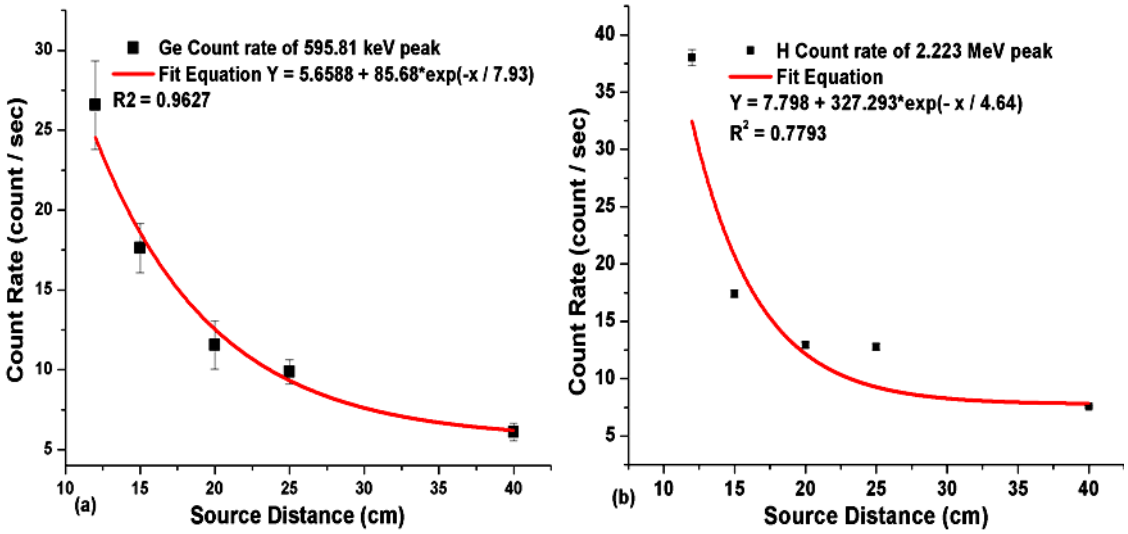

Figure 4. (a) Germanium 595.81 keV energy peak count rate with source distance from sample container **(b)** Hydrogen peak 2223 keV peak count rate with source distance from sample container.

Minimum detection concentration

The sensitivity of the PGNAA setup to chlorine and boron were calculated for different characteristic prompt gammas energies and presented in the form of a minimum detectable concentration (MDC). MDC for a peak is defined by setting threshold probability of a false detection and non-detection. The peak will be considered or not in spectrum, it will be defined by threshold. If the peak is truly present in the acquire spectrum, a chosen threshold indicates that there is a 95% probability of the counts exceeding from threshold [17]. Equations 2 and 3 were used for of the MDC and error calculation.

$$MDC = 4.653 \times \frac{C}{N_p} \times \sqrt{N_B} \quad (2)$$

$$\sigma_{MDC} = \frac{C}{N_P} \times \sqrt{2N_B} \quad (3)$$

where C is concentration of element of

interest in (g/L), N_P and N_B are the net count and background count of characteristic peak respectively. MDC for Cl and B for various prompt gamma peaks were calculated for sample irradiation time 14400 and 7200 seconds, which is summarized in table 1. We observed better MDC values for higher irradiation time in the comparison of lower irradiation time for same concentration.

Table 1. Prompt gamma-rays energy, microscopic absorption cross-section [18], MDC and MDC error of the chlorine and boron.

Element	Energy (keV)	Cross-section σγ(Eγ) b	MDC(ppm)	MDC(ppm)
			Irradiation Time =14400 sec	Irradiation Time =7200 sec
Cl	6110.842(18)	6.59(6)	175±53	280±85
B	477.595(3)	716(25)	3.0±0.95	4.6±1.4

RESULTS

Calibration of PGNAA setup using aqueous NaCl and H_3BO_3 solution

Five liter de-ionized water was used to prepare the sample of each element. Aqueous solutions were prepared by dissolving pure analytical product of chlorine (NaCl) and boron (H_3BO_3) in de-ionized water. Initially, in this work aqueous solution of chlorine and boron were used for the calibration of PGNAA setup.

Pulse height spectra of prompt gamma rays were recorded for 14400 sec live time for chlorine concentrations over 1.2g/L, 2.4 g/L, 3.6 g/L, 4.8 g/L, 6.0 g/L and 7.2 g/L in the sample. Figure 5 shows enlarged spectra of characteristic prompt gamma peak of chlorine at energies 6110 keV, 1951 keV, 1959 keV, 1165 keV and 517 keV for different concentration of chlorine.

0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L and 0.5 g/L concentrations of boron solutions were prepared with boric acid (H_3BO_3) in de-ionized water and spectra were recorded for 7200 seconds live time. Figure 6 shows enlarged spectra of characteristic prompt gamma peak (478 keV) energy for different concentration of boron in solution. It is clearly observed, peak area under the 478 keV gamma ray increases with increasing the concentration of boron in sample, whereas backgrounds peaks like 511 keV annihilation peak remains unchanged for all set of samples. It indicates that background remains constant for different concentration of boron as well as chlorine.

PGNAA setup was calibrated using characteristic prompt gamma rays of Cl and B with different concentration. The counts of different characteristic peak were obtained by integrating the channel counts in the peak area. Figure 7 (a) & (b) show linear relationship between characteristic peak count rate and sample concentration for various characteristic prompt gamma peaks of Cl and B respectively. Hydrogen amount remains unchanged during increasing concentrations of Cl and B in the aqueous solutions.

The various characteristic prompt gamma rays of chlorine were taken in overlap pulse height spectra (figure 5). Figure 7 (a) & (b) give the relationship (calibration curve) between the characteristic peak count rate and the concentration of Cl and B. In figure 7(a), calibration curve drawn only for 6110.84 keV and 1164.86 keV prompt gamma rays of Cl. As shown in figure 7(a), there is a poor linear relationship for the characteristic 1164.86 keV prompt gamma peak. The linear correlation coefficient for 6110.84 keV and 1164.86 keV are 0.9903 and 0.9384, respectively. So, minimum detectable concentration was obtained at 6110.84 KeV in case of chlorine. In figure 7(b), calibration curve drawn for 478 keV prompt gamma peak of B. It is show good response with linear correlation coefficient 0.9970.

Minimum detectable concentration and respective error calculated using equation (2) and (3) respectively for chlorine and boron.

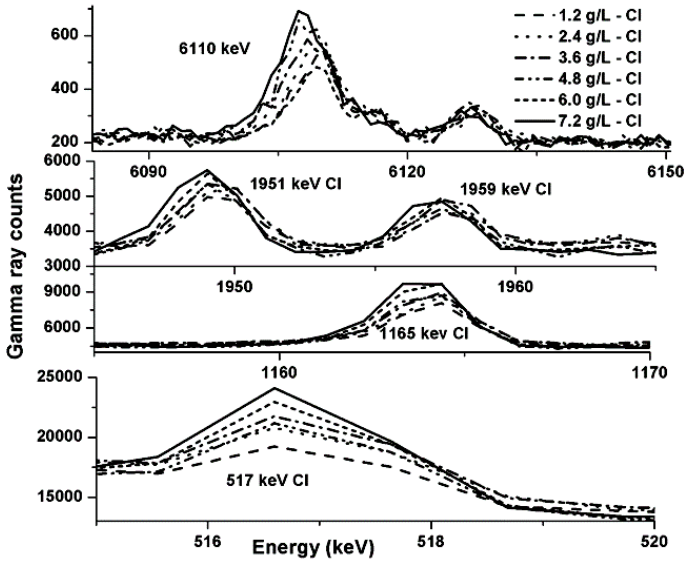

Figure 5. Enlarged spectra for chlorine different characteristic prompt gamma ray energies peak from water samples containing 1.2 g/L, 2.4 g/L, 3.6 g/L, 4.8 g/L, 6.0 g/L and 7.2 g/L chlorine.

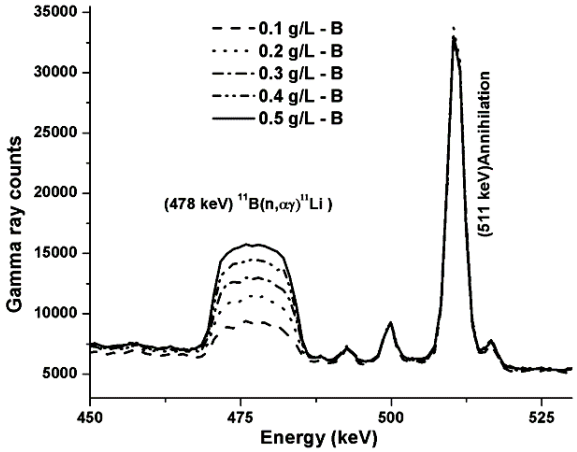

Figure 6. Enlarged spectra for boron characteristic prompt gamma ray (478 k.eV) peak from water samples containing 0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L and 0.5 g/L boron.

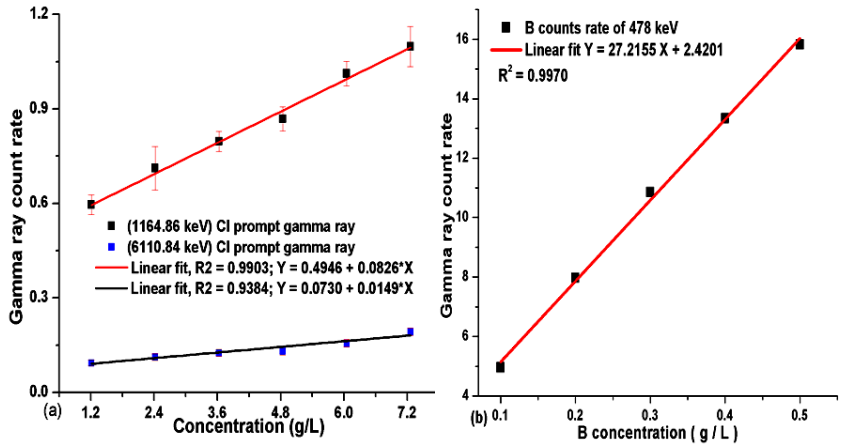

Figure 7. (a). Linear response for chlorine (calibration curve) **(b)** Boron with characteristic prompt gamma ray energies.

DISCUSSION

In order to evaluate the proposed facility, the results are compared (Table. 2) with other published studies which are mostly based on isotopic 5 Ci ^{241}Am-Be neutron source. Experiments have been done to obtain the MDC value of Cl and B using prompt gamma neutron activation method. In the present work, MDC for Cl (175.37±53.30 ppm at 6110 keV) and for B (3.13±0.95 ppm) are at 95% confidence level. MDC for Cl and B reported by other authors are summarized in table 2.

Table 2. Comparison of MDC values of different published work.

Reference	MDC_{Cl} (in ppm)	MDC_{B} (in ppm)
Idiri *et al.*[5]	180	1.6
Naqvi *et al.*[19-20]	260	104±32
R.Khelfi *et al.*[21]	22.3	-
L.Yougsheng *et al.*[4]	54	-
W. Jia *et al.*[22]	-	7.0

Minimum detectable concentration for chlorine (180 ppm at 6110 keV) and boron (1.6 ppm at 478 keV) reported by Idiri *et al.* [5] using 1 Ci Am-Be neutron source are not 95% confidence level. Because they have considered background at one sigma level. In the present study, it is carried out that PGNAA set-up gives better result for chlorine MDC value compare to Idiri *et al.* [5] and Naqvi *et al.* [19] while in case of boron better value compare to Naqvi *et al.* [20] and W. Jia *et al.* [22].

CONCLUSION

In the present work, 5 Ci Am-Be neutron source-based PGNAA set-up developed and characterized for the in-situ analysis of aqueous solutions. Characterization of the setup is done by chlorine and boron. The chlorine concentration were varied from 1.2 to 7.2 g/L and boron concentrations were varied from 0.1 to 0.5 g/L. The set-up shows linear response for both chlorine and boron for wide energy range. Estimated MDC values are 175±53 ppm at 1.2 g/L and 3±0.95 ppm at 0.1 g/L for Cl and B respectively. MDC values are obtained by present setup gives good agreement with some published work of researchers. The set-up could be used for detection of elements in heavily polluted water and detection of heavy metals, detection of elements in industrial soil and water, and salinity in drinking ground water.

ACKNOWLEDGEMENT

The authors would like to acknowledge the Department of Physics, University of Rajasthan, Jaipur and University Grant Commission, New Delhi for providing financial support for consumables and contingency under DRS Phase-II and CAS programme.

***Conflicts of interest*: Declared none.**

REFERENCES

1. Alfassi ZB and Chung C (1995) Prompt Gamma Neutron Activation Analysis, CRC Press, Boca Raton, Florida.
2. Greenberg R R, Bode P, Fernandes D N, Elisabete A (2011) Neutron activation analysis: A primary method of measurement. Spectrochimica Acta Part B : *Atomic Spectroscopy, **66 (3)**: 193-241*.
3. Da-Qian H, Wen-Bao J, Zhou J, Can C, Jia-Tong L, Hong-Tao W (2016) Heavy metals detection in sediments using PGNAA method. *Appl Radiat Isot, **112**: 50–54*.
4. Yongsheng L, Wenbao J, Daqian H, Qing S, Can C, Haojia Z, *et al.* (2015) A new Am–Be PGNAA setup for element determination in aqueous solution. *Appl Radiat Isot, **95**: 233–238*.
5. Idiri Z, Mazrou H, Amokrane A, Bedek S (2010) Characterization of an Am–Be PGNAA set-up developed for in situ liquid analysis: Application to domestic waste water and industrial liquid effluents analysis. *Nucl Instrum Meth Phys B, **268(2)**:213–218*.
6. Naqvi AA, Garwan MA, Nagadi MM, Maslehuddin M, Al-Amoudi OSB, Khateeb-ur-Rehman (2009) Non-destructive analysis of chlorine in fly ash cement concrete. *Nucl Instrum Meth Phys A, **607(2)**: 446–450*.
7. Révay Z and Molnár GL (2003) Standardization of the prompt gamma activation analysis method. *Radiochimica Acta, **91(6)**: 361–369*.

8. Khelfi R, Idiri Z, Omari L, Seghir M (1999) PGNAA of bulk concrete samples with an Am-Be neutron source. *Appl Radiat Iso,* ***51(1)****: 9-13*.
9. Panjeh H, Najafabadi R I, Ketter John (2013) Utilization of 241Am-Be neutron source in PGNAA setup used in cement raw material analysis. *Int J Sci Adv Tech,* ***3****: 25-33*.
10. Da-Qian H, Jiang Z, Jia W, Cheng C,Wang H, Li J (2016) The background influence of cadmium detection in saline water using PGNAA technique. *J Radioanal Nucl Chem,* ***310(1)****: 27-31*.
11. Fantidis JS (2015) Antoniadis A Optimization study for BNCT facility based on a DT neutron generator. *Int J Radiat Res* ***13(1)****: 13-27*.
12. Wen BJ, Da-Qian H, Can C, HaoJia Z , Qing S (2014) Optimization of PGNAA set-up for the elements detection in aqueous solution. *Sci China Tech Sci,* ***57(3)****:625-629*.
13. Radioactive Products – The Radiochemical Center – Amersham (1969/70):183.
14. IEEE Standard Association, IEEE Standard (325-1996) Standard Test Procedures for Germanium Gamma ray Detectors.
15. Turhan Ş (2007) Efficiency calibration of an HPGe detector in the 0.1–2.5 MeV energy range for Am-Be neutron source-based PGAA applications. *J Radioanal Nucl Chem,* ***273****: 443-450*.
16. Nikolov J, Medic Z, Jovancevic N, Hansman J, Todorovic N, Krmar M (2014) Applicability of the Ge(n,γ) reaction for estimating thermal neutron flux. *Physics Procedia,* ***59****: 71 – 77*.
17. Gedcke DA (2001) ORTEC Application Note AN59, How Counting Statistics Controls Detection Limits and Peak Precision. www.ortec-online.com.
18. International Atomic Energy Agency PGAA Database Files, https://www-nds.iaea.org/pgaa/databases.htm.
19. Naqvi AA, Maslehuddin M, Garwan MA, Nagadi MM, Al-Amoudi OSB, Raashid M, *et al.* (2010) Effect of silica fume addition on the PGNAA measurement of chlorine in concrete. *Applied Radiation and Isotopes* ***68****: 412–417*.
20. Naqvi AA, Al-Anezi MS, Kalakada Z, Matouq FAA, Maslehuddin M, Gondal AA, Isab AA, *et al.* (2012) Response tests of a LaCl3:Ce scintillation detector with low energy prompt gamma rays from boron and cadmium. *Applied Radiation and Isotopes,* ***70****: 882–887*.
21. Khelifi R, Amokrane A, Bode P (2007) Detection limits of pollutants in water for PGNAA using Am-Be source. *Nucl Instrum Phys Res,* ***262****: 329-332*.
22. Wen B J, Can C, Qing S, Daqian H, Yongsheng L, Yan Z, *et al.* (2015) Study on the elements detection and its correction in aqueous solution. *Nucl Instrum Phys Res,* ***342****: 240-243*.