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# Optimization of parameters of alpha spectrometry with silicon detector for low level measurements of actinides in environmental samples

D.D. Rao [a,*], V. Sudheendran [b], P.K. Sarkar [a]

[a] Bhabha Atomic Research Centre, Health Physics Division, Trombay, Mumbai 400094, India
[b] Environmental Survey Laboratory, Health Physics Division TAPS Colony, TAPP Post Thane District 401504, India

## HIGHLIGHTS

- Several parameters relevant to low level alpha spectrometry have been investigated and appropriately optimized.
- The most important parameter has been the influence of chamber pressure on resolution when the chamber is in hold mode while the vacuum pump is electrically switched off for more than 40 h.
- Samples were counted for about 4 day for low levels of detection. Efficiency, tail length, detector size and other parameters were evaluated.

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## ABSTRACT

Determination of actinides in environmental and biological samples is an important activity of radiation protection program at nuclear energy facilities. High resolution alpha spectrometry with passivated ion implanted Silicon detectors is widely used for the determination of actinides concentration. Low levels of activity concentrations in these samples often require long counting duration of a few days to obtain accurate and statistically significant data for further impact assessment. In alpha spectrometry, the chamber in which Si detector operated is a critical component and maintained at a desired vacuum for minimizing the alpha particle attenuation. Experimental evaluation of variations in energy resolution and tailing of alpha spectra was investigated under different chamber air pressures from about 6.7 Pa to more than 2700 Pa under the chamber hold mode and pump electrically switched off conditions. As part of validation, data collected on an IAEA inter-comparison exercise sample are presented under short and long counting durations with pump operating and switched off conditions respectively. It has been observed that the FWHM values do not significantly degrade, to impact the low and medium level concentration alpha spectra, for variations in vacuum chamber pressures from about 6.7 Pa to 2700 Pa.



## 1. Introduction

Alpha spectrometry using passivated ion implanted planar silicon (PIPS) detectors is a sensitive and widely used detection and analysis system for the quantitative determination of isotopes of Pu, U, Am, Po etc., in several environmental and biological samples. Environmental samples such as soil, water, weeds, marine fish and sediments around nuclear power generation facilities and biological materials (urine and feaces) of personnel involved in nuclear fuel handling facilities are analyzed for the assessment of alpha emitting nuclides following a standard radiochemical separation as a part of radiation protection program (Manickam et al., 2008; Sansone et al., 2008; Shinohara and Kunihiko, 2004). These matrices generally have activity concentration of $^{239}$Pu, $^{238}$Pu, $^{240}$Pu, $^{241}$Am, $^{238}$U and $^{234}$U at very low levels of near the detection limit of measuring systems. The PIPS detector alpha spectrometry is also used for the evaluation of specific activities of Pu, U, Cm, Am and other isotopes at significantly high concentration in isotopes separation and characterization activities (Aggarwal et al., 1992). The challenges encountered in both situations of low level and high specific activity measurements are often extremely different from one another. The parameters such as energy resolution, detection efficiency and time of counting have contrasting importance in these types of measurements. Usually, small area detectors, low efficiency and high energy resolving power and short counting time of a few thousand seconds are required for high specific activity measurements. On the other hand, high efficiency, large area and long counting time and relatively less resolving power (can be tolerated) are the needs of low level environmental concentration measurements.

* Corresponding Author. Tel.: +91 22 2559 8288; fax: +91 22 2550 5151.
*E-mail address:* ddrao217@gmail.com (D.D. Rao).

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The Silicon detector in an alpha spectrometry system is housed in a vacuum chamber and maintained at a certain vacuum pressure during operation. The vacuum pump used for evacuating the chamber is generally kept on all the time, for the entire counting duration (few days) to maintain the vacuum pressure. The spectroscopy systems commonly have a valve to operate the chamber in 'Hold' and 'Pump' mode. Pump mode for continuously evacuating the chamber and hold mode for holding the vacuum pressure when the pump is in off condition. However, in hold mode there are always certain leakages (or air entry paths) and pressure increases as the hold time increases. Martin and Hancock (2004) investigated the degradation of energy resolution with increase in the nitrogen pressure in the chamber. For counting of environmental samples, for a few days duration, to achieve significantly lower levels of detection, either the vacuum pump shall be kept on continuously for all the time to maintain constant chamber pressure or operate in hold mode. However, un-manned operation of pump during non-working hours of the day ($\sim$ 16 h $d^{-1}$; in our laboratory 17:00–9:00 h) is not recommended due to safety concerns of vacuum pump. In view of this, we investigated the changes in energy resolution and other parameters of alpha spectra as the pressure in vacuum chamber increases for more than 30 h by operating the chamber valve in hold mode. In this paper, we present the influence of vacuum chamber pressure on the energy resolution, tailing effect by operating the chamber in hold mode (Vacuum pump off ) for more than 16 h, the source to detector distance on efficiency of detector, the surface area of the PIPS detector and the source diameter. The paper also presents, as an exemplification, the results of International Atomic Energy Agency (IAEA) inter-comparison exercise for $^{239}$Pu, $^{238}$Pu, $^{234}$U and $^{238}$U in spinach powder with radio tracers of $^{236}$Pu and $^{232}$U for short and long counting duration.

## 2. Instrumentation and materials

In the present measurements, a bench top eight chamber alpha spectrometer (model Octete plus) with Mastero basic alpha spectrometry software and a vacuum pump (Edward model RV8) were used. Each chamber has 6 level height adjustable source to detector distances of $\sim$5 mm each. In house prepared electroplated alpha standard sources of $^{239}$Pu and $^{241}$Am traceable to NPL standard $^{236}$Pu (Standard Reference no. E05090391/01) and an AMR mixed source of $^{241}$Am, $^{239}$Pu and $^{244}$Cm (AMR.43 No.9682A of Amersham Intl. Ltd.) were used for optimizing the parameters. The source strengths ranging from 5.4 Bq to 5550 Bq were used for checking the influence of varying concentrations. PIPS detectors of surface area 450 $mm^2$ and 600 $mm^2$ each with 100 μm depth were used. The laboratory prepared sources and also the samples have a plating diameter of 20 mm while that of AMR standard has a diameter of 5 mm. The energy resolution of an alpha peak was expressed as full width at half maximum (FWHM) and in the present measurements, it was evaluated using Mastero software and verified manually. The FWHM of PIPS detector is generally quoted in the range of 18 to 21 keV at 5486 keV of $^{241}$Am under ideal conditions. However, it strongly depends on the condition of various parameters such as source to detector distance (STDD), source thickness, source diameter and the vacuum chamber pressure. Measurements for absolute efficiency and energy resolution were performed for such a counting duration so that each measurement had about 10% uncertainty. The chamber pressure was measured and displayed in the units of mTorr by the software (later in the manuscript SI unit, Pa will be used) and the sensors have a range of 10 m Torr to 25,000 m Torr, over which it shows overflow.

## 3. Results and discussions

The measurements of energy resolution, expressed as FWHM, were performed with three sources: an AMR mixed source ($^{241}$Am, $^{239}$Pu and $^{244}$Cm) of diameter 5 mm on a 25 mm stainless steel (SS) planchet (5550 Bq total), a laboratory prepared source of $^{241}$Am (40.72 Bq (diameter 20 mm on a 25 mm SS planchet) and another laboratory prepared mixed source of $^{239}$Pu (12.07 Bq) and $^{241}$Am (5.84 Bq) of diameter 20 mm on a 25 mm SS planchet. For the purpose of completeness of alpha spectrometry, Table 1 gives the variations of resolution and absolute efficiency against STDDs for $^{241}$Am source at a chamber pressure of 6.7 Pa. The FWHM was found to decrease from about 65 keV (5 mm STDD) to about 38 keV (30 mm STDD) on a 450 $mm^2$ detector. The minimum STDD that can be utilized on the system is 5 mm. It is a standard practice that detector's resolution is measured at a STDD of 15 mm or higher. A difference of about 5–10 keV resolution can also be seen between the PIPS detectors of 450 $mm^2$ and 600 $mm^2$. The FWHM values at 20 mm STDD are found to be 42 and 47 respectively for 450 $mm^2$ and 600 $mm^2$ detector. These values are nearly 20 keV more than the technically specified FWHM and can be attributed primarily to source thickness. Vajda and Kim (2009) have also observed that FWHM in the range of 20–60 keV are possible depending on the quality of source, source to detector distance and the type of detector.

The changes in absolute efficiency for different STDDs are given in Table 1 for two PIPS detectors of 450 $mm^2$ and 600 $mm^2$ surface area. The maximum efficiency of 22.5% and 26% can be observed at 5 mm STDD for 20 mm source diameter and for 450 $mm^2$ and 600 $mm^2$ detectors respectively. A 5 mm increase in STDD results in a significant reduction in absolute efficiency which lowers the detection limit. A simplified solid angle expression for absolute efficiency ($E$) as defined by Jaffey (1954) in terms of STDD, source diameter and detector active area is given below for comparing the experimental efficiencies.

$$E = 0.5\left(1-\frac{h}{\sqrt{h^2+r^2}}\right)-\frac{3}{16}\left(\frac{a\times r}{h^2}\right)^2\left(1-\frac{h}{\sqrt{h^2+r^2}}\right)^5$$

where, $a$: radius of disk source (mm), $h$: source to detector distance (mm), $r$: radius of detector (mm).

The computed value of $E$ for 450 $mm^2$ detector ($r$=12 mm), STDD of 5 mm ($h$=5 mm), and disk source of radius 10 mm ($a$=10 mm), is 0.27 (27%). The value from Table 1 is 22.5% and the difference is primarily due to the difficulty in measuring the accurate STDD. A STDD of 6 mm results in 23% efficiency which is close to the experimental value. Pollanen et al. (2011) had also observed that an error in STDD determination of about 0.3 mm can vary the efficiency by about 5%. The experimentally observed efficiencies agree well with the theoretical values for STDDs of 10 mm and above. They are 14.9% and 15.1% and 17.5% and 18.2% for 10 mm STDD for a 450 $mm^2$ detector and 600 $mm^2$ detectors respectively.

**Table 1**
Variations in experimental efficiency and energy resolution with respect source to detector distance ($^{241}$Am source, 40.7 Bq).

| STDD (mm) | Efficiency (%) | | Resolution–FWHM: (keV) | |
|---|---|---|---|---|
| | 450 $mm^2$ | 600 $mm^2$ | 450 $mm^2$ | 600 $mm^2$ |
| 5 | 22.5 | 25.6 | 64.4 | 71.6 |
| 10 | 14.9 | 17.5 | 54.5 | 61.9 |
| 15 | 9.7 | 12.1 | 48.2 | 52.8 |
| 20 | 6.9 | 8.8 | 41.6 | 47.0 |
| 25 | 5.0 | 6.6 | 40.7 | 46.1 |
| 30 | 3.9 | 5.1 | 38.5 | 45.0 |