# Emergency Preparedness

10

# Emergency Radio-bioassay Methodologies for First Responders & Public Approach

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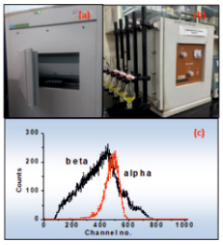


*(a) Ultra low background LSC (b) Automated solvent extraction equipment (c) Typical gross α/β spectrum obtained using PSA technique*

ABSTRACT

Following radiation emergency, first responders and the affected public are required to be assessed for internal contamination due to α/β emitting radionuclides. Timely assessment of internal contamination is necessary for triaging people for medical intervention. As a part of preparedness and response to any radiation emergency, rapid radio-bioassay methodologies are required. The present article highlights application of Liquid Scintillation Counter for screening of gross α/β activities and Solid Extraction Chromatography based radiochemical separation methods for sequential separation and estimation of radionuclides in bioassay samples at BARC.

KEYWORDS: *Radiation emergency, Internal contamination, Rapid techniques, Bioassay samples, First responders*

## Introduction

Major nuclear accidents like Chernobyl and Fukushima, necessitated efforts in developing better capabilities for emergency preparedness and response. This includes assessment of external and internal radionuclide exposures to first responders (e.g. radiation-safety, emergency medical, fire-protection, police personnel's etc.) and the affected population. In-vivo and in-vitro monitoring techniques are usually employed to estimate internal contamination due to radionuclides. Assessment of internal contamination by radionuclides emitting non-penetrating radiations like α and β is quite challenging. It requires collection of samples, like Nasal Swab (NS), excreta etc., as early as possible, in a controlled manner to avoid inadvertent contamination from external sources [1].

Urine is one of the most universally used matrices for internal contamination assessment due to its non-invasive sample collection, easy availability and adequate sensitivity to meet the requirements. In case of radiation emergency, large numbers of individuals need to be monitored in a short duration to expedite the decision for prompt medical intervention, if required. Hence, it is necessary to develop rapid and reliable bioassay methods.

### Clinical Decision Guide (CDG)

National Council on Radiation Protection and Measurements [1] has defined operational quantity, CDG i.e., intake corresponding to internal dose of 250 mSv, to assist physicians in making treatment decisions to reduce any long-term health consequences due to internal contamination.

Methodologies developed need to be sensitive enough to detect activity in biological samples corresponding to derived CDG values (Table 1). Derived CDG are values of activity levels in bioassay samples corresponding to 1 CDG intake.

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## Gross α/β measurements using Liquid Scintillation Counter

Ultra-Low Level Liquid Scintillation Counter (LSC, Model: Quantulus 1220) equipped with Pulse Shape Analysis (PSA) circuit was used for gross α/β measurements due to its high sensitivity, low detection limits and 100% counting efficiency for α emitting radionuclides [RNs]. Spectral Quench Parameter of External Standard [SQP(E)] is used to determine the counting efficiency for each sample through calibration curves. Optimization of α/β separation was carried out using $^{241}$Am and $^{90}$Sr/$^{90}$Y as α and β standards respectively. The optimum PSA setting was determined where there was equal and minimum spillover of α pulses into the β Multichannel Analyzer (MCA) and vice a versa (Fig.1a). Optimized PSA settings were plotted against corresponding SQP(E) values as shown in Fig.1b.

All subsequent measurements were performed using the defined optimum PSA settings. The sample to scintillator ratio was maintained at 1:10 for NS/tissue samples and 5:10 for urine samples. Optiphase HiSafe III scintillator and 20 mL

*Table 1: Derived CDG values in urine for a few selected radio RNs [1-2].*

| RN and absorption type | Derived CDG values in urine (Bq/100 mL) for time T (days) | | | | |
|---|---|---|---|---|---|
| | 1 d | 7 d | 10 d | 15 d | 30 d |
| $^{239}$Pu (M) | 1.1E-01 | 1.2E-02 | 7.3E-03 | 5.4E-03 | 4.6E-03 |
| U(nat.) (M) | 1.9E+02 | 5.4E+00 | 4.5E+00 | 3.6E+00 | 2.2E+00 |
| $^{241}$Am (M) | 1.0E+00 | 3.4E-02 | 2.8E-02 | 2.3E-02 | 1.5E-02 |
| $^{210}$Po (M) | 1.1E+00 | 2.0E+00 | 2.0E+00 | 1.8E+00 | 1.5E+00 |
| $^{90}$Sr (F) | 3.5E+04 | 3.3E+03 | 2.1E+03 | 1.4E+03 | 5.0E+02 |

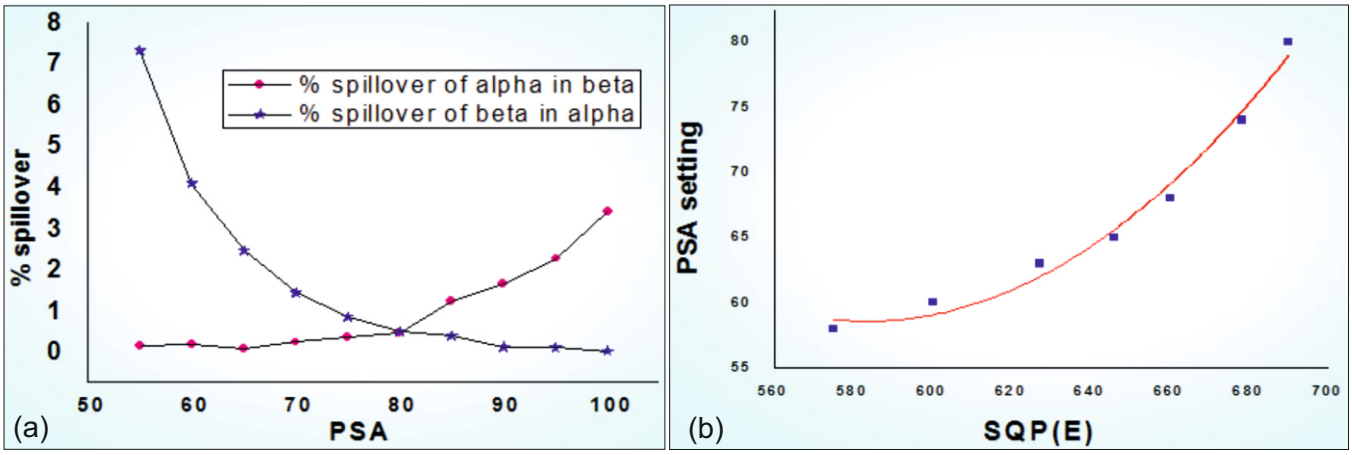


*Fig.1: (a) Spill over for determination of optimum PSA setting using $^{241}$Am,$^{90}$Sr/$^{90}$Y standards, (b) Variation of SQP(E) against optimized PSA setting.*

capacity polyethylene vials were used for standardization purpose. The gross count rate for α and β were determined and the corresponding activities were calculated using the following equations:

$$A_\alpha = \frac{Z_\alpha E_\beta - Z_\beta E_{\beta f}}{E_\alpha E_\beta - E_{\alpha f} E_{\beta f}} \quad (1)$$

$$A_\beta = \frac{Z_\beta E_\alpha - Z_\alpha E_{\alpha f}}{E_\alpha E_\beta - E_{\alpha f} E_{\beta f}} \quad (2)$$

Where, $E_\beta$, $E_\alpha$ and $E_{\beta f}$, $E_{\alpha f}$ are the counting and spillover efficiencies for β and α respectively, $Z_\alpha$ is the α count rate in α MCA and β spillover ($A_\beta E_{\beta f}$) in the same MCA. Similarly, $Z_\beta$ is β count rate that is a combination of both β disintegrations ($A_\beta E_\beta$) and α spillover ($A_\alpha E_{\alpha f}$) in β MCA.

In case of any radiation emergency, it may not be feasible to determine the SQP(E) for each and every sample and its corresponding optimal PSA value. To overcome this concern, an average SQP(E) corresponding to 'typical' urine was estimated by measuring SQP(E) in 100 individual urine samples and the quench distribution curve obtained is shown in Fig.2. The average SQP(E) was found to be 700 (SD = ± 27) with optimal PSA value of 82.

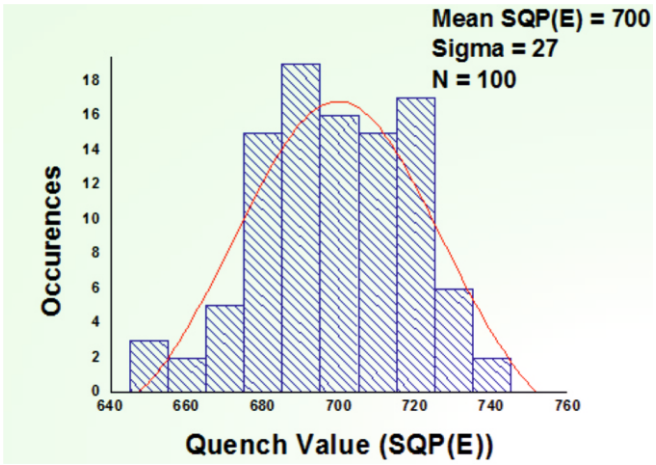


*Fig.2: Observed quench levels of urine samples and overlaid Gaussian distribution curve.*

*Table 2: MDA for gross α/β in bioassay & NS samples.*

| Matrix | CT (min) | MDA (Bq) | |
|---|---|---|---|
| | | Gross α | Gross β |
| NS/tissue | 2 | 1.5 | 16.0 |
| Urine (100 mL) | 5 | 0.7 | 3.0 |
| | 60 | 0.1 | 0.8 |

Background measurements were performed by collecting bioassay samples from the unexposed individuals to determine Minimum Detectable Activity (MDA) [3] based on optimized PSA. These are given in Table 2 for various counting times (CT).

The optimized CT using Quantulus 1220 LSC for screening gross α/β in NS/tissue/ fecal samples is 2 min. Most of the α emitters can be detected in direct urine with a CT of 5 min, except Pu(α) which requires a minimum CT of 1h to achieve the required sensitivity. Following the above procedure for calibration of LSC, methodologies have been developed at BARC, for estimation of gross α/β in NS, excised tissue and fecal samples.

NS is a best indicator of possible internal contamination by inhalation. Sawant et. al. reported the procedure for collection of NS [4]. The NS results are further confirmed by carrying out bioassay monitoring. The NS analysis is as follows: NS are wet digested using conc. $HNO_3$ and $H_2O_2$ and the solution is evaporated to dryness. The residue obtained is extracted using dilute HCl. 1 mL of this extracted sample was counted using LSC. MDA observed for gross α/β is shown in Table 2. The technique standardized is sensitive enough to detect gross α/β activity in NS samples significantly below derived CDG level (Fig.3).

Tissue samples excised from contaminated wound needs to be analyzed to know the radioactive contaminant(s) and their isotopic composition. This information is extremely essential for internal dose assessment as well as for decorporation treatment. Sawant et. al., reported the procedure for analysis of tissue samples [5] and gross α/β is estimated similar to that mentioned above for NS analysis.

Fecal samples collected from the internally contaminated individuals are dry ashed in muffle furnace at 450°C. Further wet digestion is done using conc. $HNO_3$ and

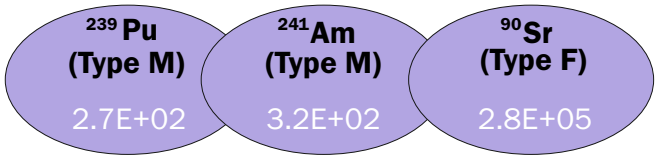


*Fig.3: Derived CDG values (Bq) in NS for a few selected RNs [1-2].*

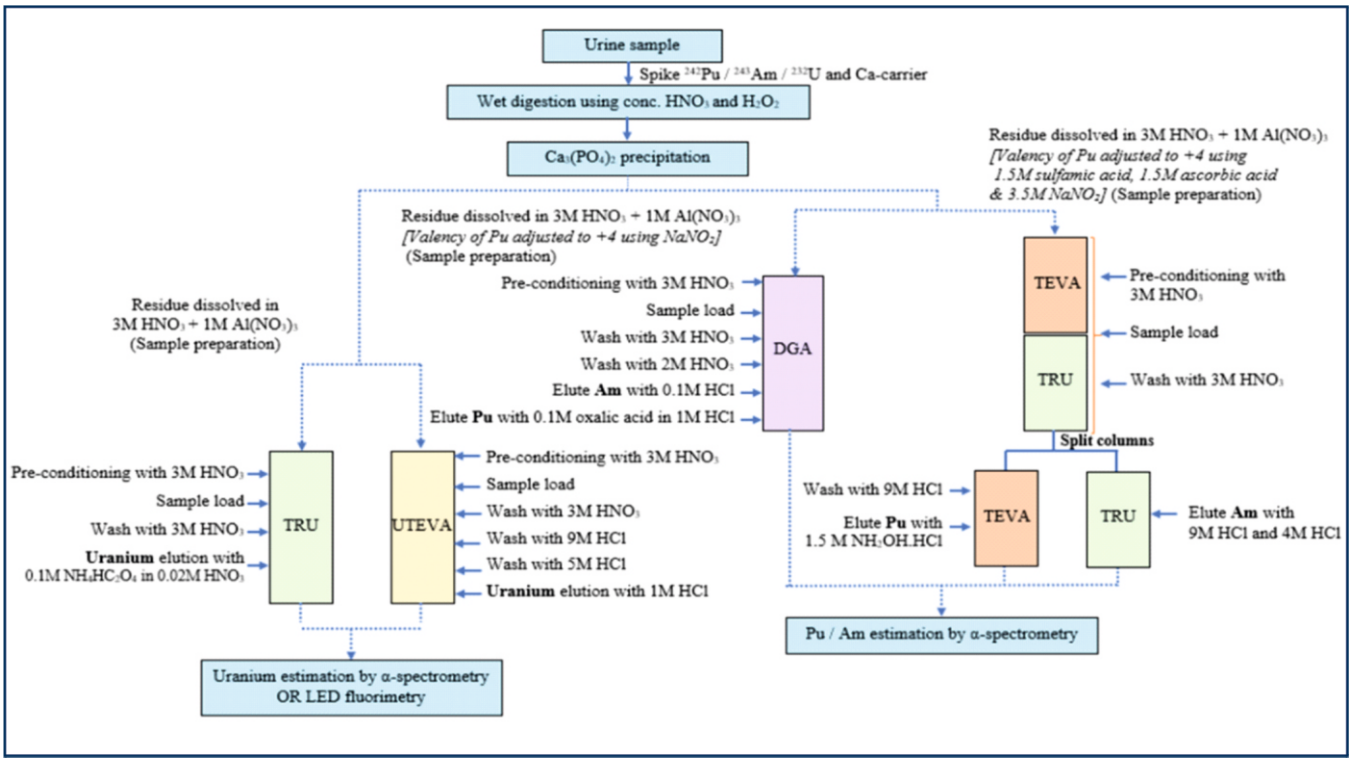


*Fig.4: Radiochemical procedure for estimation of actinides in bioassay samples [8-10].*

$H_2O_2$. The silica present in the sample is destroyed using HF and RNs present is extracted using dilute HCl. The procedure adopted for direct estimation of gross α/β in fecal samples is similar to that mentioned above for NS analysis.

### Sequential Separation of Radionuclides

A rapid technique is developed for sequential estimation of Pu and Am in NS using extractive LSC [6]. MDA obtained for Pu(α), Pu(β) and Am(α) is 45.6, 522 and 72 mBq respectively for 5 min CT. The developed method is sensitive enough to sequentially separate and estimate Pu isotopes and Am in NS below derived CDG value within 10 minutes (Fig.3).

For concentrating and isolating uranium in urine, liquid-liquid extraction technique is applied [7]. The method standardized requires ~ 1 h for complete analysis of a single sample. MDA obtained is 240 mBq/100 mL for 5 min CT. This method is sensitive enough to detect soluble compounds of U in urine below derived CDG value, even in samples collected one month after the exposure (Table 1).

For separation of actinides in bioassay samples efforts have been made to reduce the overall analysis time using extraction chromatography resins (UTEVA, TEVA, TRU and DGA). The individual radionuclides are estimated using LSC, LED Flourimetry (LF) for uranium and α-spectrometry techniques [8-10]. Further reduction in analysis time is achieved using micro-precipitation technique for source preparation for α-spectrometry [11]. The urine sample for analysis is prepared and loaded onto TRU / UTEVA columns for U estimation and DGA / stacked TEVA & TRU column for Pu and Am estimation as shown in steps described in Fig.4.

Activity of U, Pu and Am in eluted fractions are determined by α-spectrometry or LF in case of U. Estimation of U using LF is accomplished within 10 min for single sample analysis as against 24h required by α-spectrometry.

Radiochemical separation procedure is also standardized for estimation of Sr in urine samples using Sr-spec resin [12]. MDA obtained is 1.1Bq/100 mL for 5 min CT. The method standardized is sensitive enough to detect radioactive Sr in urine below derived CDG value collected one month post exposure (Table 1).

Few international incidences have shown potential of $^{210}$Po for malevolent purposes. Polonium in urine is usually estimated by chemical deposition onto a silver planchette followed by counting in α-spectrometry. In emergency situations, resourcing of silver planchette may be difficult and hence, a procedure is developed using SS planchette [13]. MDA of the technique is 12.8 mBq/100 mL for 1h CT. This method is sensitive enough to detect $^{210}$Po in urine below derived CDG value collected one month post exposure (Table 1).

### Quality Assurance (QA)

As part of QA, all the above methods are tested based on the performance criteria for radio bioassay according to ANSI N13.30 [14]. The methods developed and systems used are also validated by participation in inter-laboratory inter-comparison exercise for estimation of these RNs in emergency bioassay samples.

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