Journal of RADIATION INDUSTRY
Vol. 18, No. 2, pp. 101-107, 2024
https://doi.org/10.23042/radin.2024.18.2.101

Original article

Efficacy Evaluation of Alpha/Beta Radioactivity Screening in Urine Samples using Liquid Scintillation Counting

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ABSTRACT Rapid screening for internal contamination by alpha- and beta-emitting radionuclides is essential in situations involving radiation workers or radiation accidents. This study focused on the use of urine samples and liquid scintillation counting to quickly and accurately assess contamination. Calibration of the alpha and beta detection areas ensured precise measurement results. The major radionuclides recommended for surveillance during accidents were also considered. This study evaluated the effectiveness of the method by examining various parameters, including the limit of detection, linearity, sensitivity, selectivity, accuracy, ruggedness, and blind test sample analysis. The liquid scintillation counting method is an effective tool for screening urinary samples to detect alpha- and beta-emitting radionuclides, particularly during radiation emergencies, despite some limitations in precision.

Key words: Radioactivity screening, Liquid scintillation counting, Radiation exposure

1. Introduction

Radiation workers or people injured due to radiation accidents must undergo a screening not only for external exposure but also for exposure due to internal contamination from alpha- and beta-emitting radionuclides (Table 1). The advantage of the gross alpha/beta screening analysis of urine samples is its ability to rapidly determine the absence or presence of contamination by radionuclides released by accident due to the inhalation and ingestion of radionuclides through the respiratory and digestive systems during an accident. This involves collecting and analyzing biological samples from the victims. Among human biological materials, urine samples are generally used for their ease of collection. Liquid scintillation counting is used to convert the decay energy of decaying alpha- and beta-emitting radionuclides in the sample into light for counting. To perform such measurements, the alpha and beta detection areas must be calibrated to effectively distinguish each energy and detection area [1]. If properly calibrated, accurate alpha and beta radioactivity measurement results can be obtained.

Gross alpha/beta screening analysis for several kinds of samples were developed and used in previous research. Li X *et al*. established a method for simultaneous determination of gross alpha and gross beta activity concentrations in water in environmental monitoring [2]. Ho PL *et al*. and Ross IK used gross alpha and beta activity screening techniques for the screening radionuclides in environment [3,4]. K Norlin *et al*. established screening method to measure gross alpha and gross beta activity in drinking water in emergency preparedness [5]. Marina S.M. *et al*. present three-stage protocol for gross alpha and gross beta evaluation in water samples in emergency response [6]. Piraner O *et al*. and Chen X *et al*. developed urine gross alpha/beta bioassay method using liquid scintillation counting techniques [7,8].

Each research team is developing and applying gross alpha/beta screening methods for rapid sample analysis. It is judged that the gross alpha/beta screening method can be used as a major test to establish a mass analysis system to meet numerous testing needs in preparation for terrorism or accidents related to nuclear facilities or radioactive materials. In particular, a systematic screening analysis method to detect internal contamination caused by gross alpha- and beta-emitting radionuclides will become an important means of radiation protection [9]. Therefore, this study explores the procedure for evaluating the efficacy of the gross alpha and beta radioactivity analysis method using urine samples, which can quickly detect internal

ISSN 1976-2402 **http://www.ksri.kr/**

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Received 1 February 2024 **Revised** 13 June 2024 **Accepted** 17 June 2024

Table 1. Alpha- and beta-emitting radionuclides for the internal contamination screening method

Alpha-emitting radionuclides			Beta-emitting radionuclides		
	Half-life (y)	Decay energy (MeV)		Half-life (y)	Decay energy (MeV)
^{238}U	4.47×10^{9}	4.270	^{14}C	5730	0.156
^{235}U	7.04×10^{8}	4.679	^{90}Sr	28.8	0.546
^{230}Th	7.54×10^{4}	4.770	^{32}P	14.3 d	1.711
^{238}Pu	87.7	5.593			
^{239}Pu	2.41×10^{4}	5.245			
^{241}Am	432	5.638			
^{243}Am	7370	5.438			

contamination after work for radiation workers in normal times or radiation emergencies, and describes the results based on the actual measurement data. To evaluate the efficacy of the screening, this study evaluated the effectiveness of the method by examining various parameters, including the limit of detection, linearity, sensitivity, selectivity, accuracy, ruggedness, and blind test sample analysis [10].

2. Materials and Methods

For the screening measurement of alpha and beta radioactivity in urine samples, a liquid scintillation counter (1220 Quantulus, PerkinElmer) and a liquid scintillation reagent (Ultima-Gold, Perkin Elmer) were used, respectively. For the manufacture of reference materials, ^{241}Am, ^{243}Am, and ^{90}Sr reference sources, prepared by primarily diluting certified reference materials with ensured traceability, were used. The mixing ratio of the reference source and liquid scintillation reagent was set at 5:15. The urine samples to which the reference sources are added were produced using actual urine samples collected in real settings, depending on the test items.

Urine samples derived from the human body show widely varying aspects depending on the sample provider and collection time. To prevent errors in the validity evaluation results arising from such differences, we used synthetic urine samples composed of similar components of real urine samples for some of the assessment items. They were provided by the National Institute of Standards and Technology (NIST, USA). Table 2 lists the components of the synthetic urine samples [11].

2.1. Limit of detection and limit of quantification

The limit of detection (LOD) and limit of quantification (LOQ) in the measurement of gross alpha and beta radioactivity in urine samples were evaluated. The measurement samples, urine samples collected from three individuals, were analyzed in triplicates to evaluate the effects that could be caused by the components of the actual urine samples. A small amount of reference sources for alpha and beta emitters was added to each sample (^{243}Am: 8.19 Bq/L, ^{90}Sr: 175 Bq/L), enabling the implementation of a low level, and doubling the usual LOQ level. Additionally, six actual urine samples were prepared and used as base samples without adding sources, and the average value of three measurements was used.

LOD was evaluated as Eq. (1). The standard deviations of the gross alpha and beta equipment response values from a total of nine measurements were substituted into s_0 for calculation. In addition, the equipment response values in the gross alpha and beta regions from six blank samples were calculated as the average value (b) of the blank samples. The LOQ estimation was evaluated as three times the LOD.

$$LOD = b + 3s_0 \quad (1)$$

2.2. Linearity and range

For the test, six sample sets were prepared for the measurement of gross alpha/beta radioactivity. The synthetic urine samples with reference sources were produced in a range of 50 to 175%, based on the typical measurement concentrations of 94.8 Bq/L (gross alpha) and 350 Bq/L (gross beta). The prepared test sample sets were measured in triplicates, of which the average value was used for each sample.

2.3. Sensitivity

The sensitivity of an analytical method is defined as the ratio of the change in measured signal value to the change in concentration of the analysis. In this study, alpha/beta reference sources were added to six synthetic urine samples at steadily increasing concentrations. The additive samples thus prepared were measured in triplicate for 30 min each time, and the regression

Table 2. Chemical components of synthetic urine samples

Reagent	Weight percent (%)
Oxalic acid	0.002
Pepsin	0.003
Lactic acid (liquid)	0.009
Magnesium sulphate	0.044
Glucose (dextrose)	0.046
Citric acid	0.051
Calcium chloride	0.060
Hippuric acid	0.060
Sodium silicate	0.007
Ammonium chloride	0.101
Creatine	0.104
Sodium chloride	0.220
Sodium dihydrogen phosphate	0.259
Potassium chloride	0.325
Sodium sulfate	0.409
Urea	1.517
Concentrated nitric acid	6.701
Water	90.08

line of equipment response values of each sample concentration was estimated. Sensitivity was evaluated by the relative standard deviation of the slope.

2.4. Selectivity

The selectivity of the measurement due to the presence of interfering substances was evaluated. Although many factors can affect the gross alpha/beta radioactivity concentration in urine samples, it is impossible to reproduce all situations. Therefore, the alpha/beta reference sources were added to actual urine samples collected from six individuals exhibiting low and high concentrations to create the measurement samples. There were no restrictions on personal information, such as gender and age in selecting the six sample providers.

2.5. Accuracy

To evaluate the accuracy of the analytical method, typically expected concentrations of gross alpha/beta radioactivity were selected. These were added to the synthetic urine samples to create seven samples with similar concentrations (gross alpha: 79.2-89.2 Bq/L, gross beta: 325-334 Bq/L). Each prepared sample was measured for 30 min. The assessment of the measured results was divided into trueness and precision; for these assessments, N42.22 and N13.30 standards from the American National Standards Institute (ANSI) were used, respectively. These standards are widely used for evaluating radioactivity concentration in bio-samples [12,13].

In the trueness evaluation, the difference between the calculated and measured values was compared with the synthetic standard uncertainty Eq. (2). For the precision evaluation, the relative bias and precision of the calculated and measured values were comprehensively assessed, and the root mean square error of the overall results was determined based on a 0.25 criterion Eq. (3).

$$|V_M - V_C| < 3 \times \sqrt{u_C^2(C) + u_C^2(M)} \quad (2)$$

V_C: Calculated value
V_M: Measured value
u_c(C): Combined uncertainty of calculated value
u_c(M): Combined uncertainty of measured value

$$\sqrt{B_r^2 + S_B^2} \leq 0.25 \quad (3)$$

B_r: Relative bias of measured value (N)
S_B: Relative precision of measured value (N-1)

2.6. Ruggedness

The analytical method of gross alpha/beta radioactivity in urine samples does not involve any significant external factors that may affect the analysis, since its procedures for pretreatment and measurement are relatively simple. Therefore, the equipment response over time after urine sample collection was evaluated when the quantity analyzable in the laboratory was exceeded. The samples were prepared by adding ^{241}Am and ^{90}Sr to synthetic urine samples as gross alpha and beta sources, respectively, creating seven samples with similar concentrations. These were measured thrice over three days from the production date. In addition, to assess the impact of the laboratory temperature and humidity changes, they were recorded simultaneously at 30-min intervals over three days.

2.7. Analysis of blind test samples

Radioactivity measurements were conducted by analyzing an NIST cross-analysis sample, which was combined with 13 alpha-emitting and 12 beta-emitting nuclides in synthetic urine samples. Each of the five independently produced samples was mixed with a liquid scintillator, followed by a dark adaptation period of over 7 h. Subsequently, the measurements were compared with NIST-certified values.

3. Results and Discussion

3.1. Limit of detection and limit of quantification

For gross alpha, standard deviations of the gross alpha and beta equipment response values from a total of nine measurements were 0.69. equipment response values in the gross alpha and beta regions from six blank samples' average value was 0.27. Therefore, LOD is estimated as 2.35 and LOQ is estimated as 7.04 (Table 3). For gross beta, standard deviations of the gross alpha and beta equipment response values from a total of nine measurements were 2.54. equipment response values in the gross alpha and beta regions from six blank samples' average value was 16.8. Therefore, LOD is estimated as 24.4 and LOQ is estimated as 73.2 (Table 3). Estimated LOD and LOQ of gross beta were around 10 times larger than gross alpha. This means that gross alpha analysis can detect lower levels of radioactivity.

Table 3. LOD and LOQ values of gross counting results

Sample ID	Gross alpha counts	Gross beta counts
	Response (min^{-1})	Response (min^{-1})
1-(1)	3.57	54.0
2-(1)	2.31	52.9
3-(1)	3.09	58.8
1-(2)	4.49	54.0
2-(2)	4.46	52.0
3-(2)	4.08	49.7
1-(3)	3.40	53.1
2-(3)	3.30	51.5
3-(3)	3.78	51.7
Blank sample (b)	0.27	16.8
Standard deviation (s_0)	0.69	2.54
LOD	2.35	24.4
LOQ	7.04	73.2

3.2. Linearity and range

A linear regression analysis was performed for the evaluation. The linearity of the equipment response values for each concentration showed a very high correlation, with both gross alpha and beta exhibiting a coefficient of determination (R^2) of 0.99 or higher. In addition, the y-intercept was close to 0 (Table 4). Fig. 1 is a graph evaluating the linearity of each analysis method. The International Organization for Standardization (ISO) guidelines define the application range of the test method as a concentration range with an appropriate level of uncertainty [14]. Accordingly, the concentration range can be set above the LOQ, the quantification limit of the test method performed in this analysis method.

3.3. Sensitivity

The relative standard deviations of the slopes of the regression lines of equipment response values according to sample concentration for gross alpha and gross beta, from three repeated measurements, were 9.6% and 0.89% respectively, both well matching within 10%. In the gross alpha evaluation, a higher standard deviation was observed, as the total radioactivity concentration was lower compared to the gross beta. The regression lines from the three repeated analyses were graphically represented for each analytical method (Fig. 2). Both the gross alpha and beta data showed well-matching results within a certain range, which is determined as an indication of the consistency of equipment sensitivity across different concentrations.

Table 4. Evaluation results of linearity

Sample ID	Ratio (%)	Gross alpha counts		Gross beta counts	
		241Am (Bq/L)	Response (min^{-1})	90Sr (Bq/L)	Response (min^{-1})
1	50	47	10.0	175	48.8
2	75	71	15.5	263	72.3
3	100	95	20.4	350	96.9
4	125	119	26.3	438	118.7
5	150	142	30.8	526	145.6
6	175	166	35.5	613	169.4
Regression model		y=a+bx			
Slope (b)		0.22		0.28	
y-intercept (a)		-0.02		0.05	
Coefficient of determination (R^2)		0.99		0.99	

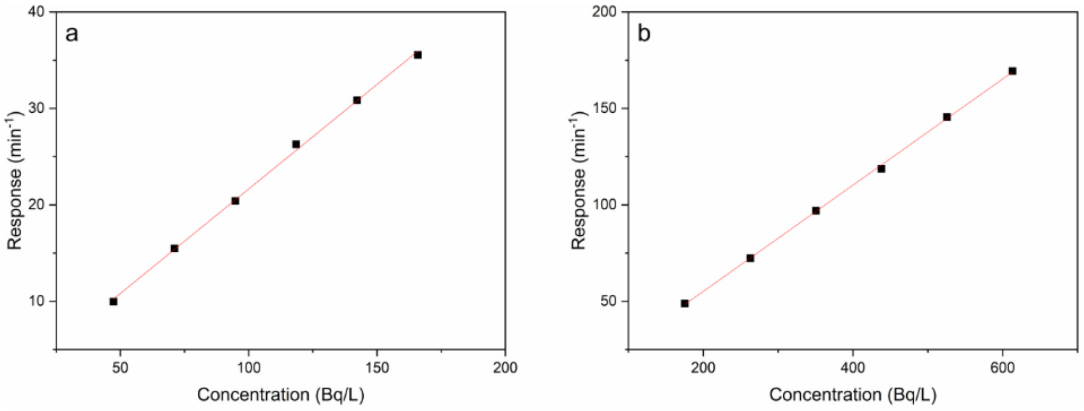

Fig. 1. Evaluation of linearity for (a) gross alpha and (b) gross beta counts.

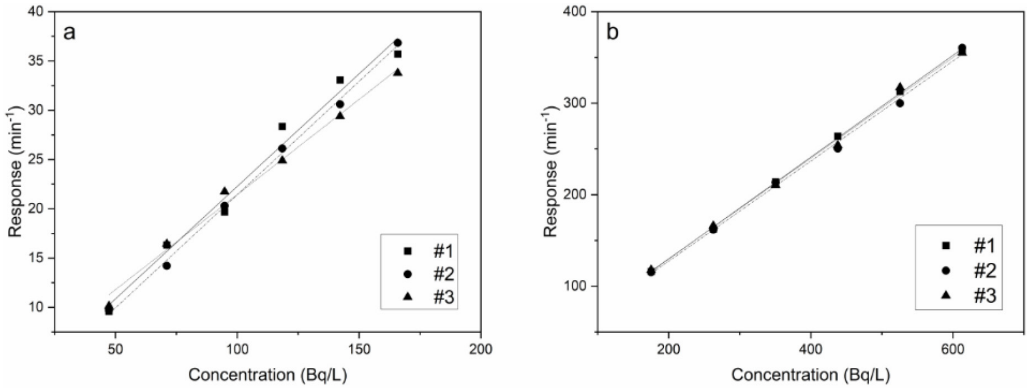

Fig. 2. Regression results of measured values of (a) gross alpha and (b) gross beta counts.

3.4. Selectivity

For the evaluation, the range of relative standard deviations for the average gross alpha/beta at low/high concentrations was set at 15% [15]. This evaluated the selective equipment response to the added reference sources in actual samples with different components collected from six individuals. This method was determined to efficiently evaluate the selectivity of the analytical method in a certain region from low to high concentrations. As a result of the evaluation, the relative standard deviations of both gross alpha and beta matched well within 15% (Table 5).

3.5. Accuracy

For gross alpha counting, Relative bias was −10.7% and Relative precision was 3.5%. Root mean square error (RMSE) was evaluated as 0.11. For gross beta counting, Relative bias was −5.9% and Relative precision was 1.0%. Root mean square error (RMSE) was evaluated as 0.06. The evaluations confirmed

Table 5. Analysis results of selectivity

Sample ID	Gross alpha counts			Gross beta counts		
	^{243}Am (Bq/L)	Response (min^{-1})	RSD*	^{90}Sr (Bq/L)	Response (min^{-1})	RSD*
A		3.06			54.0	
B	8.2	2.55	13.0 %	175	52.9	5.66 %
C		2.40			58.8	
D		17.6			178	
E	49	14.0	11.4 %	613	166	5.43 %
F		16.0			160	

*RSD: relative standard deviation

Table 6. Evaluation results of trueness and precision for gross alpha counting

Sample ID	Trueness		Precision		
	Absolute value	Traceability Limit	Relative bias	Relative precision	RMSE*
1	5.57	24.7			
2	12.4	22.9			
3	15.5	22.2			
4	7.61	24.1	-10.7 %	3.5 %	0.11
5	10.0	23.5			
6	8.45	23.9			
7	11.6	23.1			

*RMSE: Root mean square error

Table 7. Evaluation results of trueness and precision for gross beta counting

Sample ID	Trueness		Precision		
	Absolute value	Traceability Limit	Relative bias	Relative precision	RMSE*
1	25.2	86.3			
2	22.3	86.6			
3	25.2	85.9			
4	20.3	87.1	-5.9%	1.0%	0.06
5	16.2	88.1			
6	19.3	87.4			
7	17.1	87.9			

*RMSE: Root mean square error

that all seven samples met the standards for both trueness and precision (Tables 6 and 7).

3.6. Ruggedness

Table 8 displays the measurement results and relative standard deviation of the seven samples over three days. The evaluation showed very stable results, with a relative standard deviation within 5% in all test results.

3.7. Analysis of blind test samples

The gross alpha measurements across five samples showed a difference of –27.8%. In contrast, the gross beta measurements

Table 8. Evaluation results of ruggedness

Sample ID	Response of gross alpha counting (min^{-1})			Response of gross alpha counting (min^{-1})		
	#1	#2	#3	#1	#2	#3
1	21.5	21.4	21.7	206	206	206
2	19.8	21.7	23.1	209	209	212
3	19.1	20.2	21.4	211	211	212
4	21.0	21.0	23.0	213	213	215
5	20.4	20.4	22.1	216	216	213
6	20.8	20.9	21.1	210	210	211
7	20.0	19.2	20.6	213	213	212
Average		21.0			211	
SD		1.04			2.97	
RSD		4.96%			1.41%	

*RSD: relative standard deviation; SD: standard deviation

were more accurate, showing a difference below −1.2%. The significant discrepancy in the gross alpha measurement results is attributed to the oversight of Rn-222 and its progeny effects during the quenching correction. This underlines the necessity of considering these elements in future quantitative analyses for measuring bio-sample radioactivity stemming from natural-origin nuclides. Conversely, the liquid scintillation counting method is deemed effective for gross beta radioactivity measurement, with its high measurement efficiency. Complying with the ANSI bioassay standards, which advocate for precision within ±25% for bio-sample radioactivity analysis, this method is affirmed to be efficient based on its measurement outcomes.

5. Conclusion

The liquid scintillation counting method is a technique in which a sample is directly mixed with a scintillator to transfer the radioactive energy, and the purity of the sample is of crucial importance. Generally, pretreatment is performed to increase the purity by filtering impurities from a liquid sample and extracting the target nuclides. However, there is a time limit to sample pretreatment for the liquid scintillation counting method when screening in a radiation emergency. At such times, it is crucial to determine the presence or absence of radionuclides in the sample. Bypassing the process of enhancing the purity of the sample impairs the measurement efficiency that can be obtained from high-purity samples. Nevertheless, the high measurement efficiency of the liquid scintillation counting method is expected to be effectively applied to screening whether the sample contains alpha or beta emitters. Although it may not be evaluated as a precise analysis technique, it is deemed effective for screening many samples in a short time, as in a radiation emergency. This study verified the efficacy of the liquid scintillation counting method for screening urinary samples in detecting alpha- and beta-emitting radionuclides.

Acknowledgments

This study was supported by a grant of the Korea Institute of Radiological and Medical Sciences (KIRAMS), funded by Nuclear Safety and Security Commission, Republic of Korea (No. 50091-2024).

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