

## URINE GROSS ALPHA/BETA ANALYSIS BY LIQUID SCINTILLATION COUNTING FOR TERRORISM PREPAREDNESS

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**ABSTRACT.** In the case of a radiological or nuclear terrorism event, a large segment of the population might be exposed to radionuclides, resulting in extensive expectations of the public health system to test for internal contamination of radionuclides in people. Valuable information could be obtained from liquid scintillation counting (LSC) techniques by fast screening of urine samples from the potentially exposed individuals. This work describes the optimization of the LSC parameters, such as type of cocktail, discrimination, quenching, volume, and count time, to develop a rapid method for analyzing urine in an emergency response situation.

### INTRODUCTION

Liquid scintillation counting (LSC) has been a popular technique for the detection and quantitative measurement of radioactivity since the early 1950s, after the discovery of scintillation in organic compounds under nuclear radiation. The technique of LSC involves placing the sample containing radioactivity into a scintillation vial and adding a special cocktail composed of solvent, scintillator, and surfactant. Solvents transfer the nuclear energy to the scintillator, which then releases it as a flash of light whose intensity is a function of the energy and the type of nuclear decay. The alpha decay process is monoenergetic and an alpha pulse is about 35–40 ns longer than a pulse produced by beta particles. The counting efficiency is ~100% for almost all alpha decays, whereas the counting efficiency for beta emissions depends on the beta decay energy. For most beta decay with energies higher than 100 keV, the counting efficiency is about 90–100%, but for low-energy beta decays, it is in the range of 10–60% (L'Annunziata 2003).

Modern LSC instruments are equipped with either a pulse-decay analyzer (PDA) or a pulse-shape analyzer (PSA), which allow simultaneous analysis of alpha and beta emissions and collection of the analytical results in different channels through multiple channel analyzer (MCA) technology. These results are affected by: 1) radionuclide energy, 2) PSA discrimination setting, 3) quenching, 4) type of cocktail, 5) sample volume, and 6) count duration.

### OBJECTIVE

To develop a urine gross alpha/beta screening method by LSC and validate the method using available National Institute of Standards and Technology (NIST) samples, quality control (QC) materials and urine spikes by comparison of observed results with the target values.

### EXPERIMENT

#### Instrumentation

The ultra-low level liquid scintillation spectrometer Quantulus 1220 (PerkinElmer) was used. The low background is achieved by the unique detector shielding, which consists of a passive and an active shield. Analytical results were determined using WinQ and Easy View software for the instrument and process control and for reviewing the results and spectra.

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### Sample Volume

All experiments were done with 5 mL of water or urine sample and 15 mL of cocktail in 20-mL plastic scintillation vials. This optimization was done by the Sandia National Laboratories (SNL) group (Preston et al. 2007) on the basis of minimal detectable activity (MDA) consideration and quenching effect of water volume. It was shown that in the range of 2 mL of water with 18 mL of cocktail to 8 mL of water with 12 mL of cocktail, the quench factor is not changed significantly and the MDA is a fairly flat function. Therefore, a 5-mL sample size was chosen as an optimal amount to use with 15 mL of LSC cocktail.

### Choice of Cocktail

Four commonly used cocktails were considered for optimization: Ultima Gold LLT, Ultima Gold XR, Ultima Gold AB, and OptiPhase HiSafe3. All other factors such as PSA discrimination settings and efficiency curves were built using these 4 cocktails. The final decision for the optimal cocktail was done on the basis of urine spikes and QC analysis using the efficiency curves for each cocktail.

### PSA Setting Optimization

Pulse-shape analysis (PSA) setting optimization was done according to standard procedures (see Salonen 2006). Alpha and beta sources were analyzed by LSC at different PSA settings. Figure 1 shows the spillover of alpha in beta and beta in alpha versus PSA setting. The lowest spillover gave the optimal PSA setting. We used  $^{90}\text{Sr}/^{90}\text{Y}$  ( $E_{\text{max}} = 0.55$  MeV from  $^{90}\text{Sr}$  and 2.3 MeV from  $^{90}\text{Y}$ ) as a beta source and  $^{241}\text{Am}$  ( $E_{\text{max}} = 5.5$  MeV) as an alpha source. The standards were prepared in the same manner as the “patient” samples: 5 mL of deionized water spiked with radionuclides and 15 mL of cocktail. For  $^{90}\text{Sr}/^{90}\text{Y}$  and  $^{241}\text{Am}$ , PSA setting optimization was done for each cocktail. The data are presented in Table 1.

Table 1 Optimal PSA settings for  $^{90}\text{Sr}/^{90}\text{Y}$  and  $^{241}\text{Am}$  in 4 different cocktails.

Cocktail	Optimal PSA setting
Ultima Gold LLT (UGLLT)	120–125
Ultima Gold XR (UGXR)	105–110
Ultima Gold AB (UGAB)	115–125
OptiPhase HiSafe3 (OPHS)	115–125

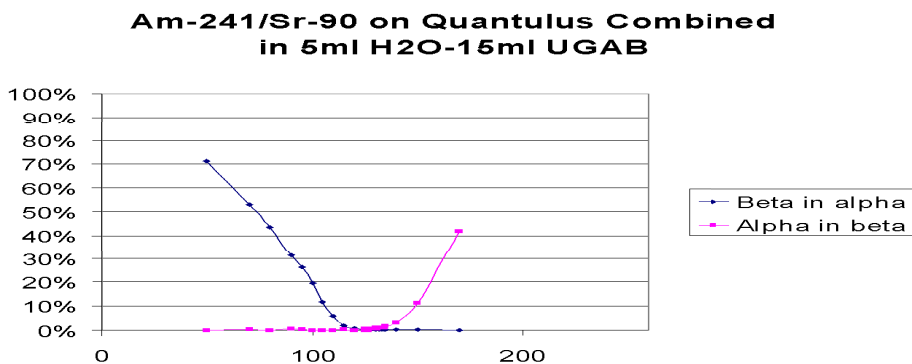


Figure 1 Typical curves for PSA optimization for  $^{90}\text{Sr}/^{90}\text{Y}$  and  $^{241}\text{Am}$  in Ultima Gold AB cocktail

## EFFICIENCY CURVES

To determine which cocktail and at which optimal setting results in the most accurate and precise data, efficiency curves were built. Quench or efficiency sets were created for each cocktail according to standard procedures (Preston et al. 2007): 17 vials with 20 mL of cocktail were prepared. One vial was left as is for background measurement. Eight vials were spiked with the same amount of  $^{90}\text{Sr}/^{90}\text{Y}$ , while 8 other vials were spiked with the same amount of  $^{241}\text{Am}$ . Both sets were then quenched with different amount of nitromethane: 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, and 0.7 mL. The prepared sets were analyzed for alpha and beta activity at the optimal PSA setting for each given cocktail, and the efficiency curves were built: the efficiency versus quench factor labeled SQP(E) and calculated by the instrument using an external gamma source ( $^{152}\text{Eu}$ ). The efficiency curves for the Quantulus normally are fit using third-order exponential equations. Typical efficiency curves built for  $^{241}\text{Am}$  and  $^{90}\text{Sr}/^{90}\text{Y}$  on the Quantulus 1220 using Ultima Gold AB cocktail at PSA 115 are presented in Figures 2 and 3.

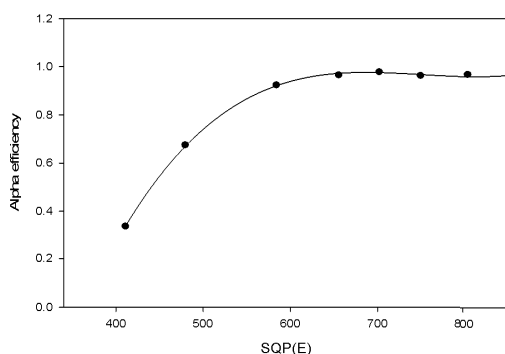


Figure 2 Typical efficiency curve for  $^{241}\text{Am}$  ( $Y = 1.8114E-8X^3 - 4.0731E-5X^2 + 0.03003X - 6.4986$ ;  $R^2 = 0.9995$ ).

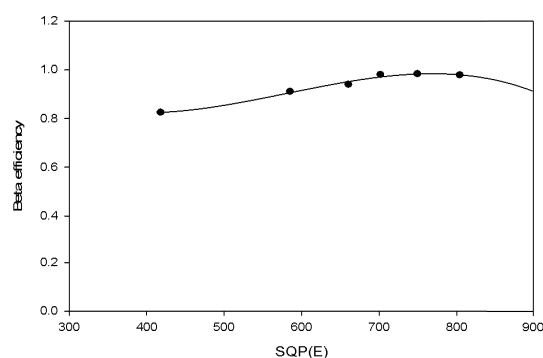


Figure 3 Typical efficiency curve for  $^{90}\text{Sr}/^{90}\text{Y}$  ( $Y = -6.1718E-9X^3 + 1.0768E-5X^2 - 0.0056X + 1.7421$ ;  $R^2 = 0.9866$ ).

$^{137}\text{Cs}$  and  $^{60}\text{Co}$  were also used as a beta source.  $^{137}\text{Cs}$  shows an efficiency of 110–115% in the SQP(E) range of 700–800, while  $^{60}\text{Co}$  efficiency is 60–65% for the same SQP(E) range. Therefore,  $^{90}\text{Sr}/^{90}\text{Y}$  (90–98% efficiency for each nuclide) as a beta standard for gross alpha/beta analysis looks reasonable, although it does not exclude the use of  $^{137}\text{Cs}$  or  $^{60}\text{Co}$  efficiency curves if the source of radiation is known to be either  $^{137}\text{Cs}$  or  $^{60}\text{Co}$ .

Most alpha-emitting radionuclides have an energy spectrum around 5000 KeV and an LSC efficiency of about 100%, therefore, the use of  $^{241}\text{Am}$  as a standard for gross alpha measurement is a reasonable choice.

## TYPICAL URINE

The urine from 40 different individuals not known to be contaminated with radionuclides was analyzed by LSC for gross alpha/beta to determine the background and quench factor SQP(E) for a “typical” urine. Five mL of urine was mixed with 15 mL of Ultima Gold AB cocktail and analyzed at an optimal PSA setting. The alpha results were found to be close to 0 Bq/L, while beta results were in the range of 20–100 Bq/L. This is probably due to the variable content of  $^{40}\text{K}$  in urine as the amount of potassium varies with the daily diet. The SQP(E) for a “typical” urine samples in Ultima Gold AB cocktail is normally in the range of 700–800.

**COCKTAIL OPTIMIZATION**

To determine the optimal cocktail, several urine spikes from different donors (differently quenched) were prepared and analyzed along with low and high quality control (QC) materials prepared by Eckert and Ziegler Analytics, Inc. Five mL of urine, spiked with  $^{241}\text{Am}$  and/or  $^{90}\text{Sr}/^{90}\text{Y}$ , and QC materials were mixed with 15 mL of each cocktail and analyzed by LSC at the optimal PSA setting for each given cocktail. The observed results were compared with the spiked amounts; the data are given in Table 2.

Table 2 Comparison of observed and spiked amounts for urine spikes and QC in different cocktails. All activity values in Bq/L.

Sample	SQP (E)	Cocktail	PSA	$^{241}\text{Am}$ added	$^{241}\text{Am}$ found	$^{90}\text{Sr}$ added	$^{90}\text{Sr}$ found	Alpha error	Beta error
Urine#13	761	OPHS	115	2069.4	2000	4196	4340	(3.35)	3.43
Urine#14	745	OPHS	115	2069.4	2010	4196	4150	(2.87)	(1.10)
Urine#15	718	OPHS	115	2069.4	1990	4196	4200	(3.84)	0.10
DIH20	793	OPHS	115	2069.4	2110	4196	4110	1.96	(2.05)
Urine#17	710	OPHS	115	2069.4	1870	4196	4320	(9.64)	2.96
Urine#18	677	OPHS	115	2069.4	1888	4196	4510	(8.77)	7.48
LU-077203	695	OPHS	115	1023	886	12207	12800	(13.39)	4.86
HU-077201	700	OPHS	115	2511	2250	30213	31500	(10.39)	4.26
Urine#13	741	UGXR	105	2069.4	2030	4196	4060	(1.90)	(3.24)
Urine#14	723	UGXR	105	2069.4	1930	4196	4130	(6.74)	(1.57)
Urine#15	686	UGXR	105	2069.4	1830	4196	4130	(11.57)	(1.57)
DIH20	767	UGXR	105	2069.4	2100	4196	4000	1.48	(4.67)
Urine#17	686	UGXR	105	2069.4	1680	4196	4300	(18.82)	2.48
Urine#18	664	UGXR	105	2069.4	1810	4196	4590	(12.54)	9.39
LU-077203	680	UGXR	105	1023	817	12207	12000	(20.14)	(1.70)
HU-077201	683	UGXR	105	2511	2040	30213	29800	(18.76)	(1.37)
Urine#13	797	UGAB	115	2069.4	2110	4196	4170	1.96	(0.62)
Urine#14	774	UGAB	115	2069.4	2100	4196	4210	1.48	0.33
Urine#15	742	UGAB	115	2069.4	2060	4196	4210	(0.45)	0.33
DIH20	831	UGAB	115	2069.4	2110	4196	4290	1.96	2.24
Urine#17	729	UGAB	115	2069.4	2040	4196	4170	(1.42)	(0.62)
Urine#18	701	UGAB	115	2069.4	2000	4196	4450	(3.35)	6.05
LU-077203	716	UGAB	115	1023	1002	12207	12113	(2.05)	(0.77)
HU-077201	729	UGAB	115	2511	2506	30213	29836	(0.20)	(1.25)
Urine#13	797	UGLLT	120	2069.4	2030	4196	4060	(1.90)	(3.24)
Urine#14	771	UGLLT	120	2069.4	1930	4196	4130	(6.74)	(1.57)
Urine#15	738	UGLLT	120	2069.4	1830	4196	4130	(11.57)	(1.57)
DIH20	825	UGLLT	120	2069.4	2100	4196	4000	1.48	(4.67)
Urine#17	731	UGLLT	120	2069.4	1680	4196	4300	(18.82)	2.48
Urine#18	694	UGLLT	120	2069.4	1810	4196	4590	(12.54)	9.39
LU-077203	712	UGLLT	120	1023	901	12207	12800	(11.93)	4.86
HU-077201	722	UGLLT	120	2511	2280	30213	31700	(9.20)	4.92

The best precision between observed and spiked amounts was seen for samples prepared in Ultima Gold AB cocktail.

**METHOD VALIDATION**

**NIST Samples Characterization**

To validate the method, the analysis of NIST samples, containing <sup>238</sup>U, <sup>234</sup>U, <sup>238</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Am, <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>65</sup>Zn, and <sup>54</sup>Mn, was used to compare the observed versus target values. NIST samples (5 mL) were mixed with 15 mL of UGAB cocktail and analyzed at PSA = 115. The analysis time was chosen to be 5 min for the sample and 5 min to analyze SQP(E). The comparison plots of 70 observed results versus target values for NIST samples gives the following equations: for alpha:  $Y = 1.1590X - 0.1097$ ;  $R^2 = 0.9825$ ; for beta:  $Y = 1.061X + 8.585$ ;  $R^2 = 0.9997$ .

The comparison demonstrates the method accuracy with a slope close to 1, low intercept, and  $R^2$  for the regression of 98–99% for both gross alpha and gross beta emissions. For gross beta nuclides, the higher intercept could be explained by the presence in NIST samples of <sup>137</sup>Cs whose LSC efficiency is higher than <sup>90</sup>Sr/<sup>90</sup>Y, which was used as a standard.

**Quality Control (QC) Characterization**

Low and high QC for the urine gross alpha/beta analytical method were characterized in our lab using a minimum of 20 independent runs. The results are presented in the Table 2. <sup>90</sup>Sr/<sup>90</sup>Y presents the beta source, while <sup>241</sup>Am presents the alpha source. Small errors between amounts, observed and reported by manufacturer, also confirm the method accuracy. The high (HU-077201) and low (LU-077203) QC data and spectra are presented in Table 3 and Figure 4, respectively.

Table 3 Characterization of low (LU-077203) and high (HU-077201) quality control (QC) materials.

QC sample	LU-077203		HU-077201	
	Alpha	Beta	Alpha	Beta
Average found (Bq/L)	1008	12,049	2569	30,359
SD	44	139	93	246
Target values	1023	12,267	2511	30,273
Error %	-1.47	-1.78	2.31	0.28

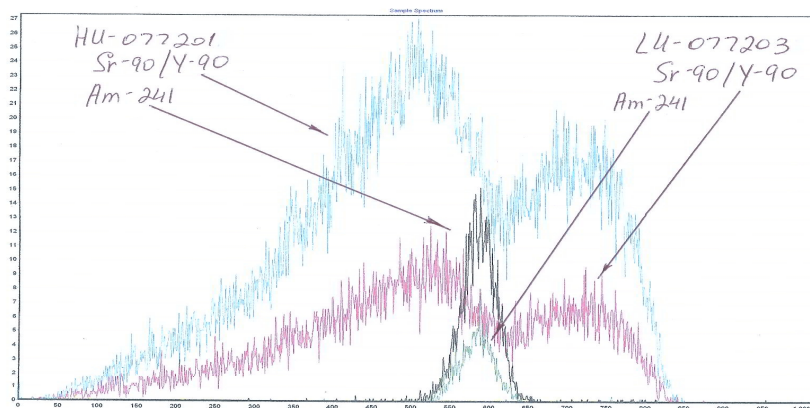


Figure 4 The high (HU-077201) and low (LU-077203) QC spectra

**MINIMAL DETECTABLE ACTIVITY (MDA)**

MDA, according to the formula, is proportional to the square root of the background and inversely proportional to the sample volume, efficiency, and square root of the counting time. As a result, 1) the lower the background is, the lower the MDA; 2) the larger the sample volume, the lower the MDA; 3) the higher the efficiency, the lower the MDA; and 4) the longer analysis time, the lower the MDA. The Quantulus 1220 has a background (5 mL of deionized water sample with 15 mL of Ultima Gold AB cocktail) alpha signal in the range of 0–1 Bq/L and beta signal in the range of 20–30 Bq/L. The optimal analysis time was chosen to be 5 min, but the time can be easily changed to fulfill the MDA requirements. Under chosen conditions, for a 5-mL sample size with 15 mL cocktail for 5 min analysis time, the MDA for gross alpha is 2.46 Bq/L while the gross beta is 28.41 Bq/L.

**CONCLUSIONS**

A fast and accurate method for urine gross alpha/beta analysis was developed and validated using quality control materials, urine spikes, and NIST NRIP PT samples. The method allows analyzing 60 samples on one instrument for 15 hr. Having 2 instruments, optimized and ready, the laboratory can screen 120 samples for 15 hr with MDA of about 2.46 Bq/L for gross alpha and 28.41 Bq/L for gross beta nuclides.

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