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The effect of quench agent on urine bioassay for various radionuclides using Quantulus™1220 and Tri-Carb™3110

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Abstract

Following a radiological or nuclear incident, the National Response Plan has given the Department of Health and Human Services / Centers for Disease Control and Prevention the responsibility for assessing population's contamination with radionuclides. In the public health response to the incident, valuable information could be obtained in a timely and accurate manner by using liquid scintillation counting techniques to determine who has been contaminated above background for alpha and beta emitting radionuclides. The calibration plays a major role in this process therefore, knowing the effect of quench agents on calibration is essential.

Keywords

Liquid Scintillation Counting (LSC); Quantulus™ 1220; Tri-Carb™3110; quench curve; urine bioassay

Introduction

Normally the signal in counts per minute (CPM) is converted into activity units of becquerels per liter (Bq/L) by means of quench (efficiency) curve for a given radionuclide. The quench curve connects the quenching factor (SQP(E) - the Spectral Quench Parameter of the External standard or tSIE – transformed Spectral Index of the External standard, depending on the LSC instrument) with the instrument efficiency, that is the observed activity (CPM) divided by the added activity (DPM). The typical quench agents are nitromethane, nitric acid, carbon tetrachloride, and toluene [1,2]. Nitromethane is a good quench agent for high energy beta emitters as Sr-90 and alpha emitters as Am-241. However, in urine bioassay measurements for low energy radionuclides such as tritium (H-3), nitromethane is not the optimum choice. Tritium is more sensitive to quenching. Some

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researchers used carbon tetrachloride with yellow food dye [3] for urine tritium bioassay or tried to reduce the color quenching by ultraviolet photolysis [4]. Both approaches have limitations: carbon tetrachloride with yellow food dye is not the best match for urine while quenching decrease using ultraviolet photolysis creates an additional step in sample preparation as well as photoluminescence peak in spectra. For tritium bioassay we evaluated other possible quench agents such as black tea [5] by itself and black tea with addition of either urea or nitromethane. The choice and the amount of a quenching agent depends on the instrument: the Quantulus™1220 requires more quenching agent than Tri-Carb™3110. This work is devoted to the study of the effect of quench agent on the urine bioassay for such radionuclides as Am-241, Sr-90/Y-90, H-3, P-32 using Quantulus™1220 and Tri-Carb™3110.

Experimental

Reagents and materials

For gross alpha/beta analysis we used the Ultima Gold® AB cocktail (UGAB) from PerkinElmer Company; 99% Nitromethane from ACROS Organics; black tea solution (Lipton black tea from any grocery store, 1 regular tea bag steeped in 200 mL of boiling water for 10 min, the cooled solution was used for quenching); and 99% Urea from ACROS Organics. Deionized (DI) water was used for all solutions (≥18 MΩ·cm, from an Aqua Solutions Ultrapure Water System, Aqua Solutions, Inc.). “Base urine” was collected through anonymous human donations (according to Centers for Disease Control and Prevention Institutional Review Board protocol 3994) and acidified to 1% HNO_3. All radioactive source solutions were traceable to the National Institute for Standards and Technology (NIST) (Gaithersburg, MD, USA). Urine gross alpha/beta quality control (QC) materials U-GAB_Low_2015 and U-GAB_High_2015 were purchased from Eckert & Ziegler Analytics, Inc. They are base urine samples spiked with Am-241 and Sr-90/Y-90 at low and high levels (see Table 4). Reference Material (RM) and High Calibration Range material (HCR) were prepared in our laboratory by spiking base urine with NIST traceable reference solutions of Am-241 and Sr-90/Y-90 (for gross alpha/beta analysis), P-32 (for P-32 analysis) or H-3 (for tritium analysis). QC materials for tritium analysis (LU12318 and MU12319) were prepared in our laboratory by spiking base urine with H-3 NIST traceable reference solution at low and high levels (see Table 6). All reference solutions used for spiking were purchased from Eckert & Ziegler Analytics, Inc.

Instrumentation and labware

For this study we used two ultralow level liquid scintillation spectrometers Quantulus™1220 (#2 and #3) and two Tri-Carb™3110 (#1 and #2) (all from PerkinElmer Company) for Am-241, Sr-90/Y-90, H-3, and P-32 analysis in alpha/beta mode; 20-mL LSC plastic vials (PerkinElmer Company) for LSC analysis; a high precision analytical balance capable of accuracy weighing 0.0001 gm (Mettler-Toledo, LLC); 15-mL and 50-mL conical polypropylene tubes (Becton Dickinson Company) for solution preparation; a Brinkman bottletop dispenser with capacity from 5 mL to 25 mL (Brinkman Instruments, Inc.) for cocktail dispensing; and four electronic pipettes with total volume range from 5 μL to 5 mL (Eppendorf, Inc).

Sample preparation and LSC analysis

First, we determined the optimal PSA (pulse shape analysis) or PDD (pulse decay discriminator) settings for a urine matrix using base urine [6]. Next we built quench curves according to procedure [6] for each nuclide at optimal PSA or PDD settings on each instrument using quench agents such as nitromethane, DI water and nitromethane added, black tea and 10% urea mixture, and black tea and 5% nitromethane mixture as described in Tables 1, 2. For the Am-241 and Sr-90/Y-90 quench sets we used 20 mL of UGAB cocktail and 15 mL of UGAB cocktail for P-32 and H-3 quench sets. Then we optimized such parameters as sample analysis time, external standard analysis time, type of cocktail, and sample/cocktail volume for 20 mL vial geometry [6]. In addition, a region of interest (region in which the given nuclide will be counted) was optimized based on spectra for each nuclide from each instrument, since each instrument needs its own optimization depending on the nuclide of interest. Finally, for sample preparation we mixed 5 mL of urine sample with 15 mL of UGAB cocktail in 20-mL LSC plastic vials till a uniformed state was reached. Then LSC vials with solutions were placed on the LSC counter tray and LSC analysis was performed using parameters described in Table 3.

Results and Discussion

In this study we present the analytical results obtained using the optimal quench agents for each nuclide of interest (Tables 1 and 2). Black tea and urea were used as they are a better match for urine by color and chemical quenching. This is important for low energy nuclides which are more sensitive to quenching. The criteria for choosing the quench agent were activity results by LSC from known urine spikes.

Quench (efficiency) curves and LSC activities results of urine spikes

We chose Quantulus™1220 #2 and Tri-Carb™3110 #1 to show the examples of quench curves for each type of instrument.

Figures 1 and 2 represent quench curves collected from Quantulus™1220 instrument using 20 mL of UGAB cocktail and nitromethane (from 0 to 0.5 mL) as the quench agent. Both quench curves are fitted to polynomial equation third degree. Figures 3 and 4 represent quench curves collected from Tri-Carb™3110 using 20 mL of UGAB cocktail and nitromethane (from 0 to 0.3 mL) as a quench agent. The quench curve for Sr-90 is fitted to polynomial equation third degree while the quench curve for Am-241 is fitted to exponential equation.

Gross Alpha/Beta Quality Control Materials (QC), Reference Material (GAB-RM) and High Calibration Range Material (GAB-HCR) results were used to evaluate the gross alpha/beta analysis using the built quench curves. The results by Quantulus™1220 and Tri-CarbTM3110 are presented in Table 4. All activities are shown in Bq/L. The use of a nitromethane quench agent with Ultima Gold® AB cocktail for quench curve preparation gave the activity correlation in the range of ± 5% for gross alpha nuclides and ± 2% for gross beta nuclides when comparing the observed and target value data. The change of the

quench agent for others to include black tea and nitromethane or water and nitromethane did not give any benefit for both types of instruments.

However, for P-32 the best quench agent was nitromethane with 5 mL of DI water for Quantulus™1220. An example of a P-32 quench curve is presented on Figure 5. This quench curve was produced on Quantulus ™1220 using 15 mL of UGAB cocktail, DI water (5 mL – 4.7 mL), and nitromethane (0 mL – 0.3 mL) as a quench agent. For Tri-Carb™3110 the best quench agent for P-32 was black tea - 5% nitromethane mixture diluted with water at different ratios (total volume of 5 mL) mixed with 15 mL of UGAB cocktail. An example of a P-32 quench curve built on Tri-Carb™3110 is presented on Figure 6. Both quench curves for P-32 are fitted to polynomial equation third degree. Table 5 represents the results of P-32 urine spikes analysis using Quantulus and Tri-Carb instruments' quench curves with the optimal quench agents for each instrument. Base urine (BU) is not spiked and the solutions BU-P32–5K through BU-P32–1M are base urine spiked with P-32 in the range of 5 000 Bq/L – 1 000 000 Bq/L. The bias between found and target activity is in the range of ± 3% for all instruments. These results confirm the optimal choice of quench agents for P-32 urine bioassays for both types of instruments.

Figures 7 and 8 demonstrate the H-3 quench curves using the best quench agent: mixture of black tea - 10% urea diluted with DI water (total volume 5 mL), and UltimaGold®AB cocktail (15 mL) as shown in Tables 1 and 2. Both quench curves for H-3 are fitted to polynomial equation third degree. The Table 6 represents the characterization results for QC, RM, and HCR materials using optimal quench curves on both types of instruments. Both types of the instruments show the correlation between found and target activity in the range of ± 4%, which means the black tea with 10% urea imitates urine better than nitromethane with water or just black tea.

Conclusion

This work demonstrates the importance of the proper selection of appropriate quench agents used for urine bioassay with careful evaluation of the nature of nuclide and type of the instrument. We showed that for our gross alpha/beta urine screening, nitromethane in Ultima Gold® AB cocktail is optimal for radionuclides such as Sr-90/Y-90 and Am-241 while using either the Quantulus™ 1220 or Tri-Carb™3110 instrument. For the P-32 urine bioassay, DI water with nitromethane is the most appropriate quench agent (Table 1) while using the Quantulus™1220 whereas DI water, black tea, and nitromethane as a quench agent (Table 2) yields better results on the Tri-Carb™3110. For a tritium urine bioassay the best quench agents are DI water, black tea, and urea for both instrument types.

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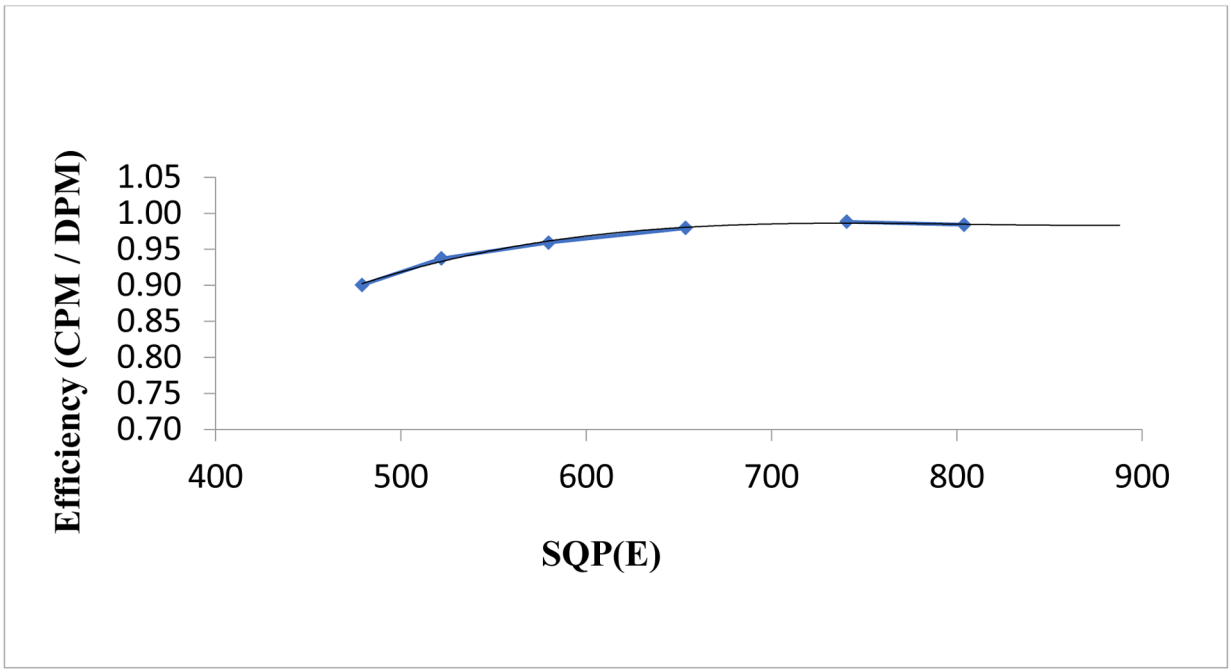

Fig.1.

Quench indicating parameter measurements in term of SQP(E) effect on Efficiency using nitromethane (0–0.5 mL) as a quench agent in the UGAB cocktail (20 mL) for Sr-90 on Quantulus™1220. Solid black line is the trendline

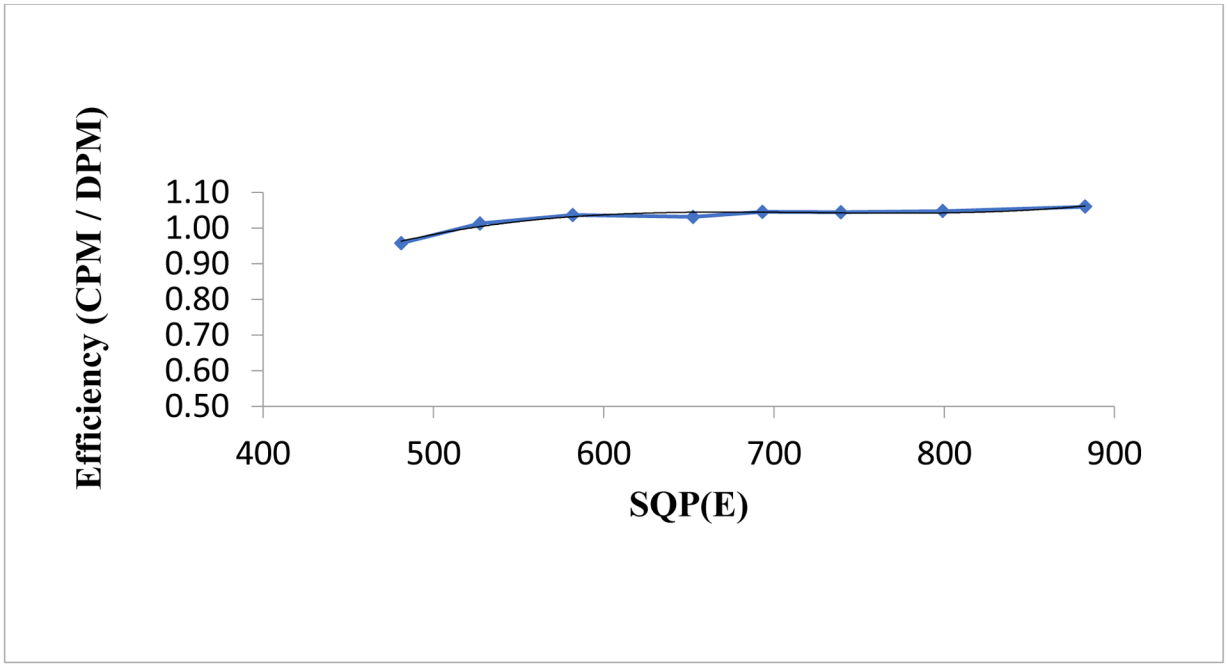

Fig.2.

Quench indicating parameter measurements in term of SQP(E) effect on Efficiency using nitromethane (0–0.5 mL) as a quench agent in the UGAB cocktail (20 mL) for Am-241 on Quantulus™1220. Solid black line is the trendline

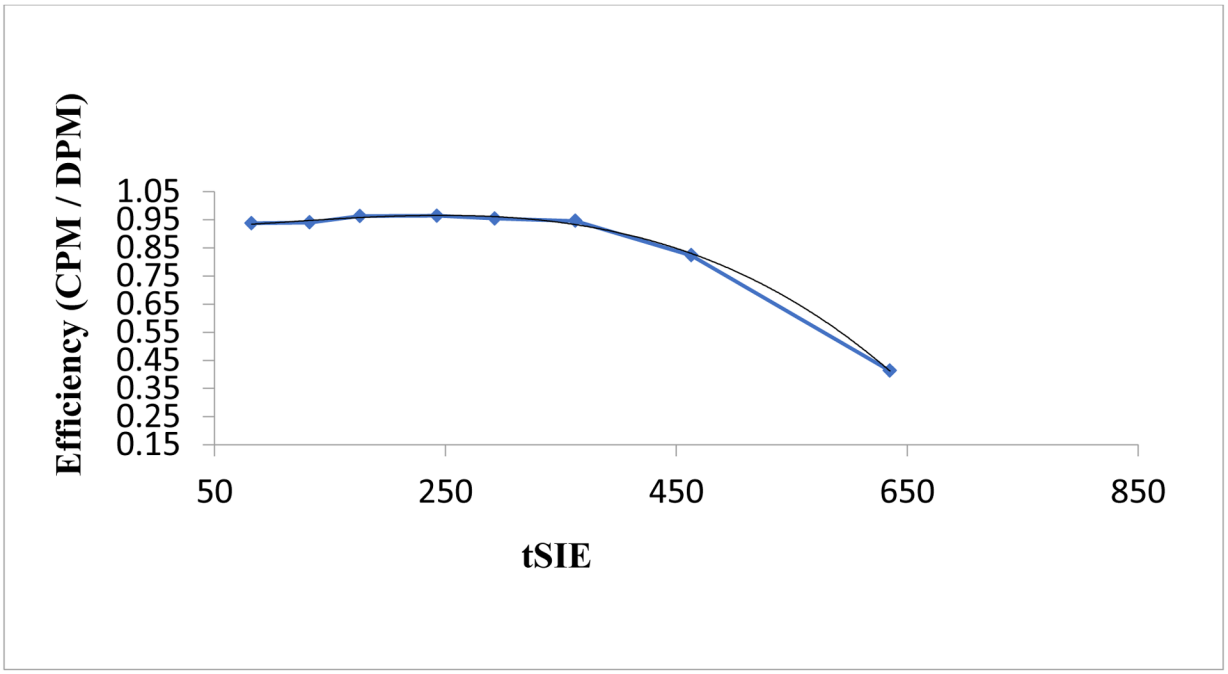

Fig.3.

Quench indicating parameter measurements in term of tSIE effect on Efficiency using nitromethane (0–0.3 mL) as a quench agent in the UGAB cocktail (20 mL) for Sr-90 on Tri-Carb™3110. Solid black line is the trendline

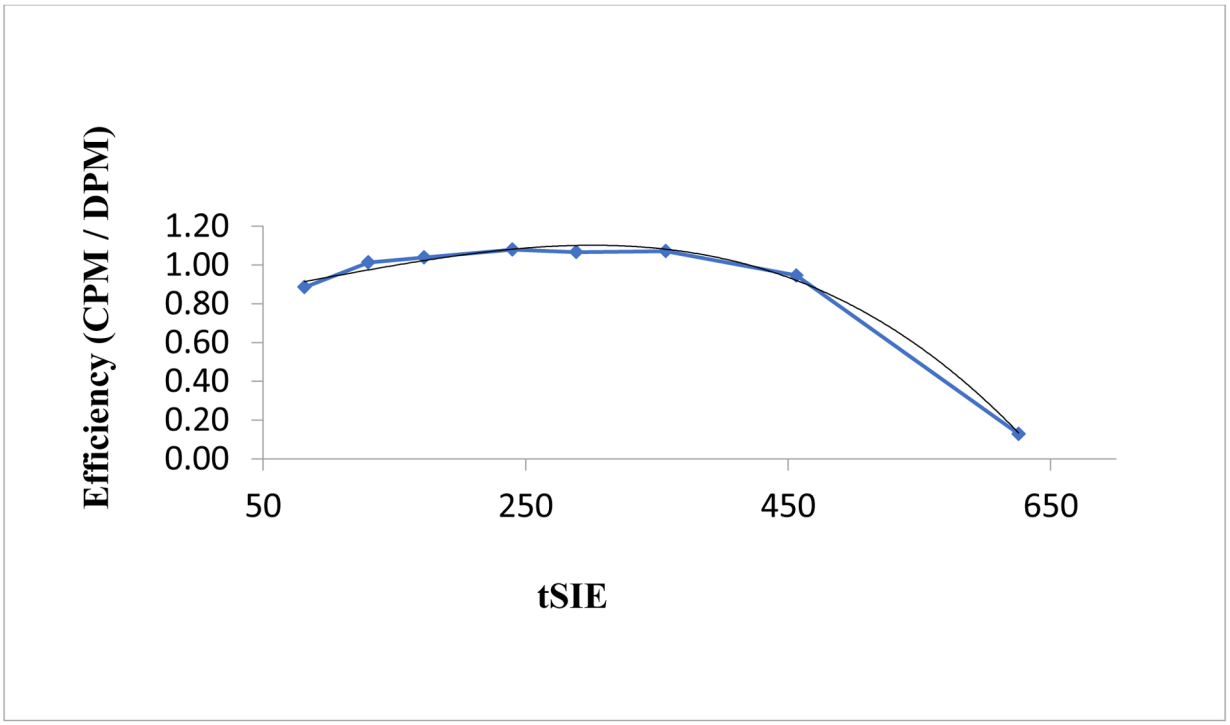

Fig.4.

Quench indicating parameter measurements in term of tSIE effect on Efficiency using nitromethane (0–0.3 mL) as a quench agent in the UGAB cocktail (20 mL) for Am-241 on Tri-Carb™3110. Solid black line is the trendline

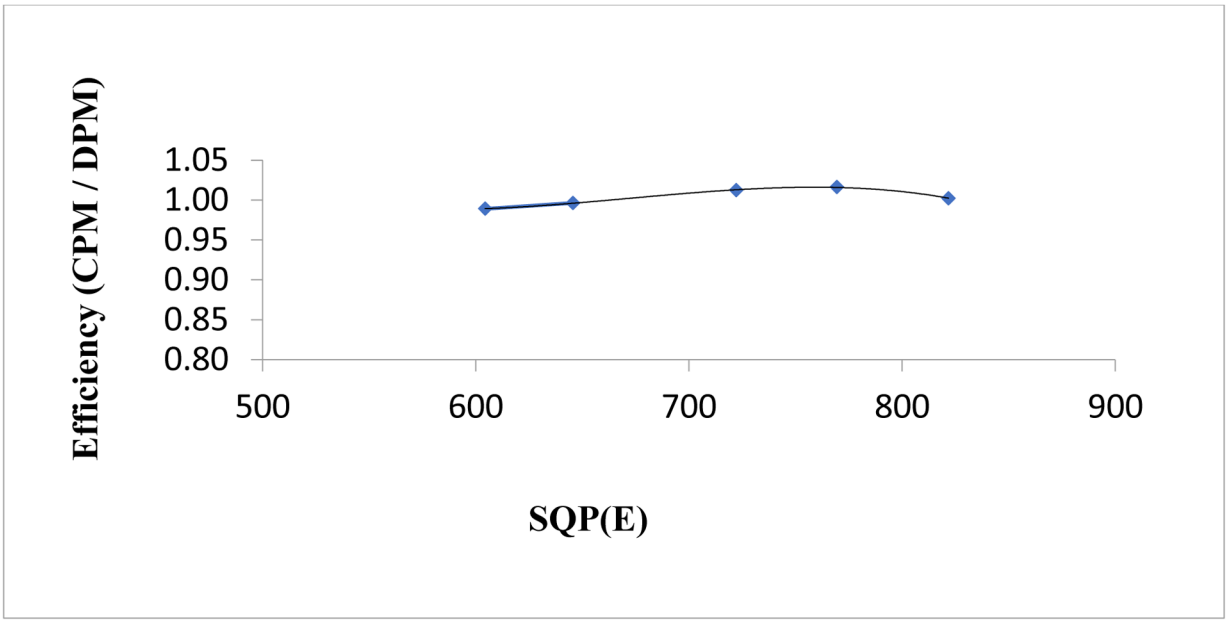

Fig.5.

Quench indicating parameter measurements in term of SQP(E) effect on Efficiency using DI water (5–4.7 mL) and nitromethane (0–0.3 mL) as a quench agent in the UGAB cocktail (15 mL) for P-32 on Quantulus™1220. Solid black line is the trendline

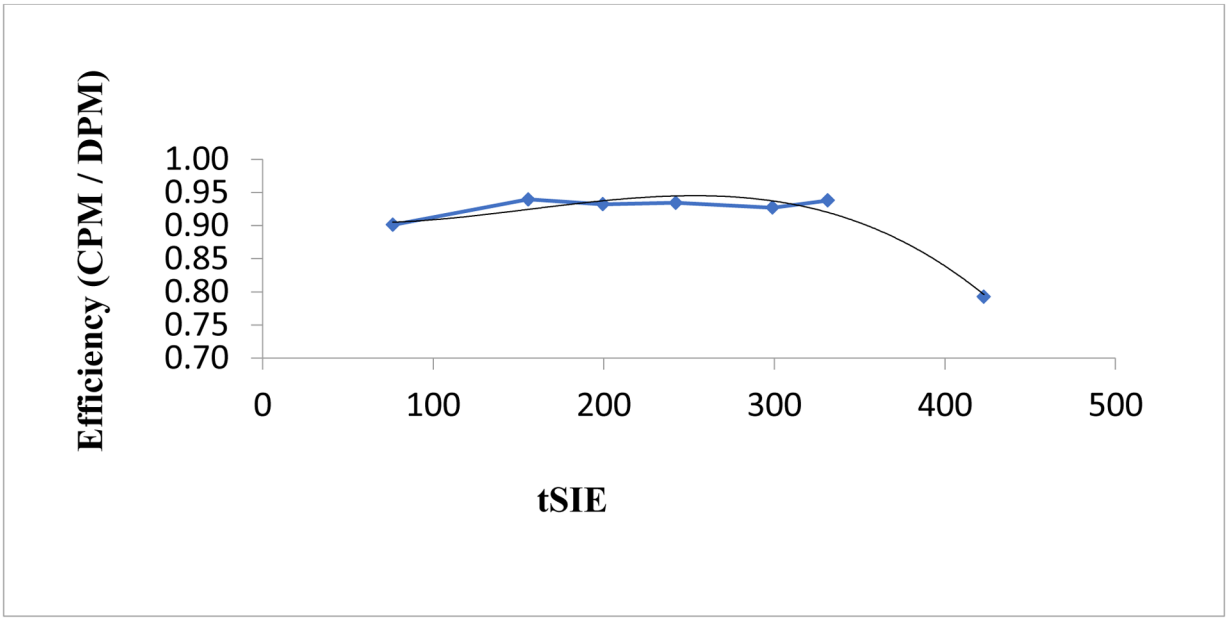

Fig.6.

Quench indicating parameter measurements in term of tSIE effect on Efficiency using DI water (5–2.5 mL) and Tea-5% nitromethane (0–2.5 mL) as a quench agent in the UGAB cocktail (15 mL) for P-32 on Tri-Carb™3110. Solid black line is the trendline

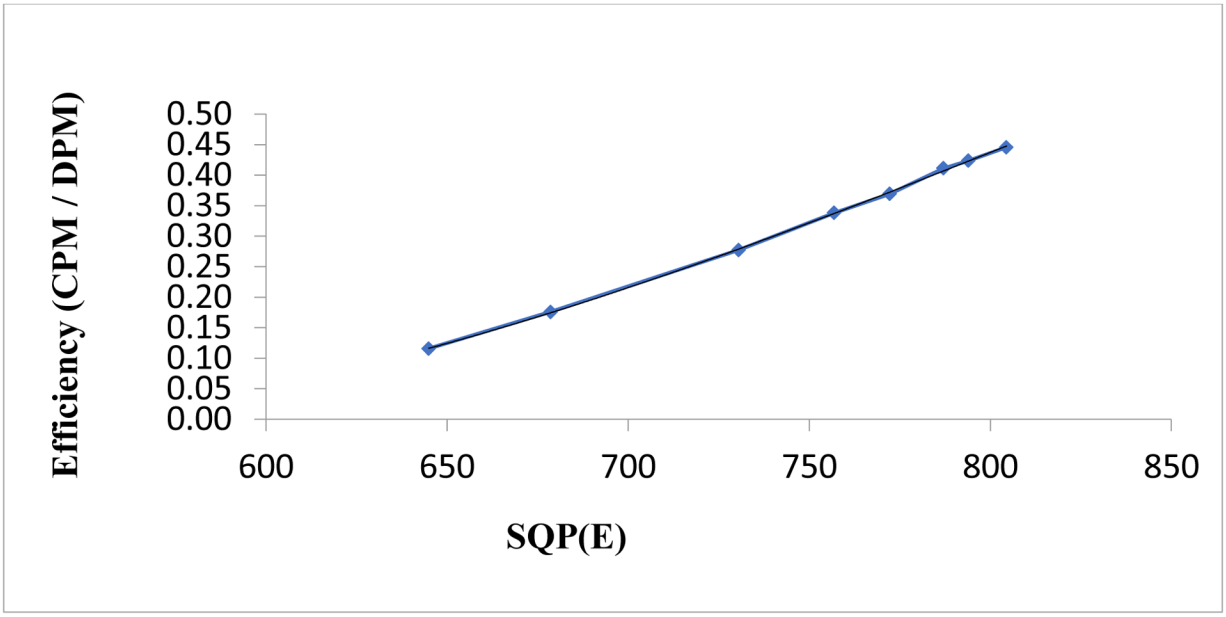

Fig.7.

Quench indicating parameter measurements in term of SQP(E) effect on Efficiency using DI water (5–0 mL) and black tea-10% urea (0–5 mL) as a quench agent in the UGAB cocktail (15 mL) for H-3 on Quantulus™1220. Solid black line is the trendline

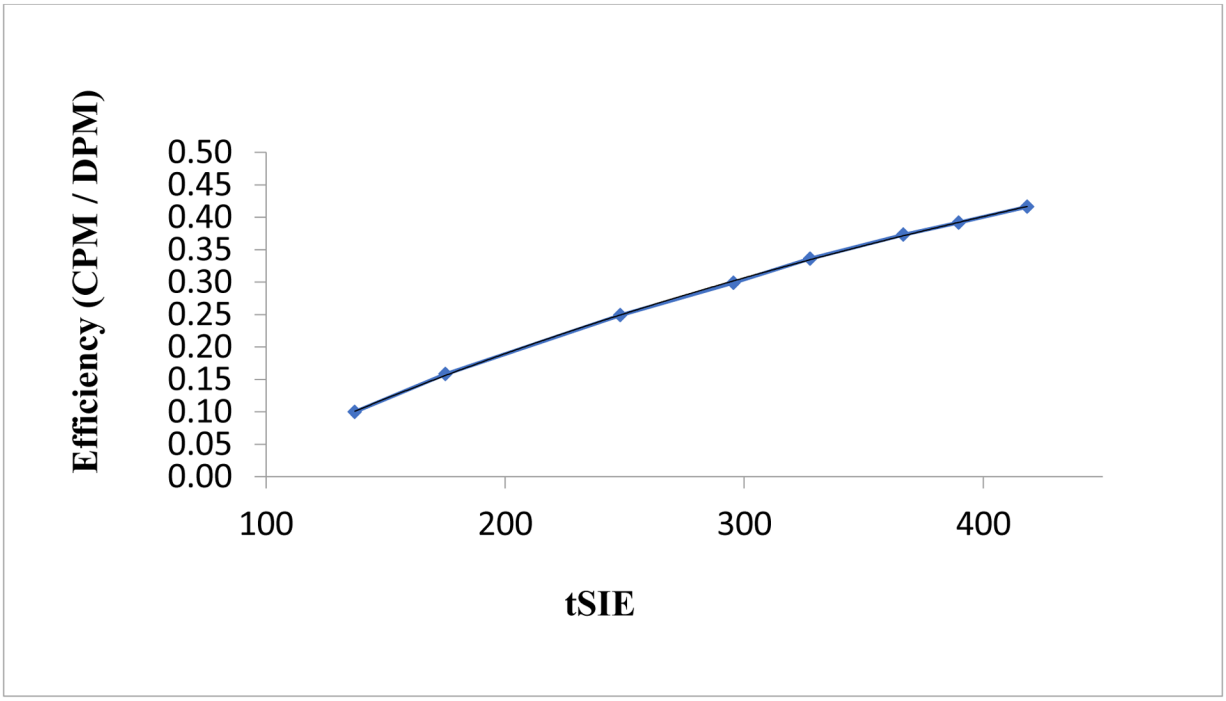

Fig.8.

Quench indicating parameter measurements in term of tSIE effect on Efficiency using DI water (5–1 mL) and black tea-10% urea (0–4 mL) as a quench agent in the UGAB cocktail (15 mL) for H-3 on Tri-Carb™3110. Solid black line is the trendline

Table 1

The optimal quench agents for urine bioassay of Am-241, Sr-90/Y-90, P-32, and H-3 on Quantulus™1220

Standards	Quench agent for Am-241, Sr-90/ Y-90	Quench agent for P-32		Quench agent for tritium	
	Nitromethane (mL)	DI water (mL)	Nitromethane (mL)	DI water (mL)	Black tea-10%urea (mL)
S1	0	5.0	0	5.0	0
S2	0.05	4.98	0.02	4.7	0.3
S3	0.1	4.96	0.04	4.3	0.7
S4	0.15	4.94	0.06	4.0	1.0
S5	0.25	4.9	0.1	3.0	2.0
S6	0.3	4.85	0.15	2.0	3.0
S7	0.4	4.8	0.2	1.0	4.0
S8	0.5	4.7	0.3	0	5.0

Table 2

The optimal quench agents for urine bioassay of Am-241, Sr-90/Y-90, P-32, and H-3 on Tri-Carb™3110

Standards	Quench agent for Am-241, Sr-90/Y-90	Quench agent for P-32		Quench agent for tritium	
	Nitromethane (mL)	DI water (mL)	Tea-5% Nitromethane (mL)	DI water (mL)	Black tea-10%urea (mL)
S1	0	5.0	0	5.0	0
S2	0.025	4.8	0.2	4.9	0.1
S3	0.05	4.7	0.3	4.7	0.3
S4	0.075	4.5	0.5	4.3	0.7
S5	0.1	4.3	0.7	4.0	1.0
S6	0.15	4.0	1.0	3.0	2.0
S7	0.2	3.5	1.5	2.0	3.0
S8	0.3	2.5	2.5	1.0	4.0

Table 3

LSC method parameters for different instruments

Parameter	Quantulus™1120 #2	Quantulus™1120 #3	Tri-Carb™3110 #1	Tri-Carb™3110 #2
PSA/PDD setting	90	80	125	165
Sample volume (mL)	5	5	5	5
Cocktail volume (mL)	15	15	15	15
Sample analysis time (min)	5	5	5	5
External Std analysis time	1 min	1 min	2 Ω (10–15 sec)	2 Ω (10–15 sec)
Alpha ROI	1–1024 channels	400–750 channels	0–300 KeV	0–200 KeV
High energy Beta ROI	1–1024 channels	1–1024 channels	0–2000 KeV	0–2000 KeV
Low energy Beta ROI	0–250 channels	0–250 channels	0–18.6 KeV	0–18.6 KeV

Table 4

Mean and Standard Deviation (SD) for QC, GAB-RM, and GAB-HCR using Quantulus™1220 (two instruments) and Tri-Carb™3110 (two instruments) determined by statistical analysis (SAS) during more than 20 runs performed on different days

QC and RM	Gross Alpha Mean	SD	Gross Alpha Target	Bias	Gross Beta Mean	SD	Gross Beta Target	Bias	Instrument ID
	(Bq/L)	(Bq/L)	(Bq/L)	(%)	(Bq/L)	(Bq/L)	(Bq/L)	(%)	
U-GAB_Low_2015	76.8	8.42	80	−4.0	1 770	47.0	1 740	1.7	Quantulus1220
U-GAB_High_2015	5 270	166	5 350	−1.5	105 100	1 880	106 000	−0.8	
GAB-RM	4 070	160	4 000	1.8	50 100	733	50 000	0.2	
GAB-HCR	15 100	450	15 000	0.7	153 000	2 490	150 000	2.0	
U-GAB_Low_2015	81.7	8.65	80	2.1	1 750	43.3	1 740	0.6	Tri-Carb3110
U-GAB_High_2015	5 290	303	5 350	−1.1	105 000	1 650	106 000	−0.9	
GAB-RM	4 190	189	4 000	4.8	49 500	1 150	50 000	−1.0	
GAB-HCR	14 700	340	15 000	−2.0	152 000	2 200	150 000	1.3	

Table 5

P-32 urine spikes analysis on Quantulus™1220 (two instruments average) and Tri-Carb™3110 (two instruments average) using P-32 quench curves built with optimal quench agents for each instrument type

Sample ID Number	P-32 Mean Activity (Bq/L)	P-32 Target Activity (Bq/L)	SD (Bq/L)	Average vs Target Bias (%)	Instrument ID
BU	45	N/A	3.6		Quantulus1220
BU-P32–5K	5 090	5 060	53	0.6	
BU-P32–10K	10 000	9 960	274	0.4	
BU-P32–25K	25 200	25 100	561	0.4	
BU-P32–50K	49 800	49 600	615	0.4	
BU-P32–100K	99 300	101 000	1 760	−1.7	
BU-P32–210K	210 000	211 000	1 900	−0.5	
BU-P32–420K	422 000	423 000	2 730	−0.2	
BU-P32–800K	810 000	809 000	4 613	0.1	
BU-P32–1M	1 010 000	1 000 000	16 851	1.0	
BU	49	N/A	1.0		Tri-Carb3110
BU-P32–5K	5 100	5 060	93	0.8	
BU-P32–10K	10 200	9 960	216	2.4	
BU-P32–25K	25 300	25 100	478	0.8	
BU-P32–50K	49 400	49 600	950	−0.4	
BU-P32–100K	99 200	101 000	1 830	−1.8	
BU-P32–210K	208 000	211 000	3 730	−1.4	
BU-P32–420K	418 000	423 000	7 510	−1.2	
BU-P32–800K	797 000	809 000	14 000	−1.5	
BU-P32–1M	987 000	1 000 000	20 200	−1.3	

Table 6

Mean and Standard Deviation (SD) for urine H-3 QC, H3-RM, and H3-HCR using Quantulus™1220 (two instruments) and Tri-Carb™3110 (two instruments) determined by statistical analysis (SAS) during more than 20 runs performed on different days

QC and RM	Mean	SD	Target	Bias	Instrument ID
	(Bq/L)	(Bq/L)	(Bq/L)	(%)	
LU12318 (Low QC)	7 930	379	8 000	−0.9	Quantulus1220
MU12319 (High QC)	969 000	32 000	1 000 000	−3.1	
H3-RM	99 400	1 840	100 000	−0.6	
H3-HCR	3 050 000	84 600	3 000 000	1.7	
LU12318 (Low QC)	8 090	469	8 000	1.1	Tri-Carb3110
MU12319 (High QC)	959 000	28 400	1 000 000	−4.1	
H3-RM	98 500	2 770	100 000	−1.5	
H3-HCR	3 070 000	76 300	3 000 000	2.3	