Energy Calibration of Liquid Scintillation Counter to allow Semi-Qualitative Nuclide Identification in Water Samples

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Abstract. An energy and quench related calculation algorithm applicable for liquid scintillation counting (LSC) to identify NORM-nuclides in environmental water samples is presented. To identify α-β emitters with spectrometric methods an energy calibration is necessary to ensure the proper conversion of channel numbers into the equivalent energy of a nuclide in a sample. Unlike in γ-spectrometry where there is a straightforward energy-to-channel number relation, a different approach was used in this study for the identification of α-active radionuclides, as the energy-to-channel number calibration is quenching dependent. Therefore prior to acquisition of data, energy calibrations under various quenching conditions at an optimized pulse shape analyzer (PSA) level was performed as part of the α-nuclide identification procedure. Quenching dependent energy calibration equations were derived. Validation of the method was done through certified radionuclide calibration solutions. The research has demonstrated that this analytical technique is able to produce quality and reliable analytical result for the identification of nuclides in water samples. Although the β-active nuclides cannot be readily identified they support the identification of the α-active nuclides. The method developed offers an attractive and cost-effective alternative to nuclide specific analysis through element separation followed by α-spectrometry.

KEYWORDS: LSC, NORMs, Quench, α/β-energy Calibration, PSA

1 INTRODUCTION

Naturally occurring radioactive materials (NORMs) in environmental samples are of crucial importance in the case of radiological impact studies in any environmental compartment. This information can be obtained from liquid scintillation counting (LSC) by fast screening of NORM-contaminated water samples. However, direct determination of these nuclides via LSC is far from straight forward since LSC can only quantitatively determine the total α/β-activity. In addition, identification of α-β emitters with the LSC method requires an exact determination of the peak position or end-point energy [1]. This requires energy calibration under various quenching conditions. The α-energy calibration is a correlation of α-energy and the channel peak position of the spectrum. The appearance of quenching in the sample affects not only the counting efficiency but the α-β discrimination [2] as well. LSC allows the measurement of both α- and β-activities and in some cases an indication can be obtained of the α-energy, although the resolution of α-spectra is much poorer than that attained by semi-conductors.

To identify α-β emitters with spectrometric methods an energy calibration is necessary to ensure the proper conversion of channel numbers into the equivalent energy of a nuclide in a sample. Therefore, prior to acquisition of data, an energy calibration under various quenching conditions has to be performed as part of the nuclide identification procedure.

In this investigation we evaluated the potential of a low-background liquid scintillation system with advanced spectrometry capabilities, the Quantulus 1220^{TM}, to be used directly for the identification of the most likely nuclides that contribute to the activity of NORM-nuclides in environmental water.

2 THEORITICAL OVERVIEW

The precise determination of the α-energy of each radionuclide requires exact determination of the peak position or endpoint energy. Theoretically [3], one can use either a linear or quadratic calibration relation between the peak position (channel number) and the energy of the particle of interest,

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$$E = A \times C + B \qquad \text{(Linear)} \qquad (1)$$

or

$$E = A \times C\hat{}2 + B \times C + D \qquad \text{(Quadratic)} \qquad (2)$$

where:

E	the energy in MeV.
A, B, D	constants.
C	the channel position (number).

A two-point energy calibration is normally acceptable from a general measurement point of view. In this study a somewhat different approach was used, as the energy-to-channel-number calibration is quenching dependent.

2.1 EXPERIMENTAL WORK

The experiments were carried out with the Quantulus 1220 ^{TM} (Perkin Elmer) equipped with an α/β-discrimination device. The PSA was optimized to achieve the best α/β separation. For all measurements, polyethylene vials (Wallac) and the ultima gold AB cocktail were used. Eleven (11) samples were prepared from the available certified ^{226}Ra standard reference solution (ACS/DC/48/02) to perform quench dependent energy-to-channel-number calibration for NORM nuclides. All samples were spiked with a small amount (100 μℓ) of the ^{226}Ra standard solution. To simulate various degrees of quenching, distilled water was used in 1-mℓ fractions between 0 and 10 mℓ and the volumes were adjusted to 20 mℓ with scintillation liquid. The samples were counted for 5 hours to ensure good statistics. For each of the samples the peak positions of the four peaks were determined as well as the quench parameter. The following data were gathered from the spectra (Table 1),

Table 1: Samples prepared from ^{226}Ra standard reference solution.

		^{214}Po	^{218}Po	^{222}Rn	^{226}Ra
	Energy (MeV)	7.687	6.002	5.49	4.774
ml	Quench parameter	Channel number	Channel number	Channel number	Channel number
10	779.11	707	643	622	593
9	787.40	722	652	635	604
8	806.02	734	666	647	615
7	817.00	746	673	661	627
6	820.73	756	690	668	634
5	828.96	762	696	675	639
4	834.95	772	704	685	649
3	848.98	781	715	695	660
2	862.28	793	728	706	671
1	867.89	807	743	719	685
0	890.57	837	772	748	714

These data are graphically represented in Figure 1.

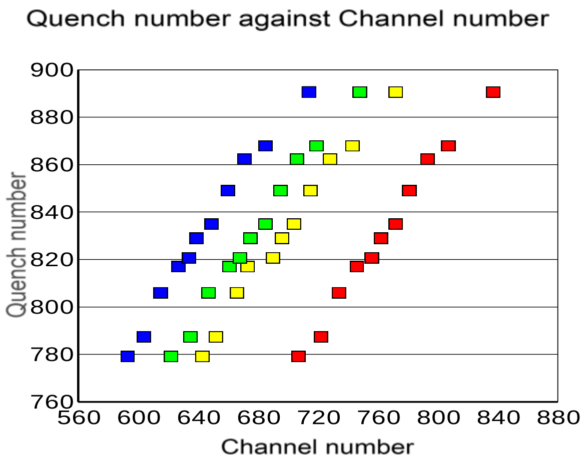

Figure 1: Graphic representation of samples prepared from ^{226}Ra standard reference solution.

For each of the eleven quench parameters a linear regression was done against the four energies, according to

$$E_\alpha = Ch.A_i + B_i \tag{3}$$

Where,

E_α = Energy of the alpha particle (MeV)
Ch = Channel number of the respective peak in the spectrum
A_i = Quench parameter dependent coefficient, and
B_i = Quench parameter dependent constant

3 RESULTS AND DISCUSSION

The measured samples used for quench dependent energy-to-channel-number calibration from the ^{226}Ra standard reference solution resulted in the following coefficients and constants given in Table 2, together with their respective uncertainties. Accordingly, it can be seen that the typical uncertainty in the energy calculation is between 2% and 5%, which will lead to an uncertainty of 100-200 keV.

Table 2: Results from a linear regression of the eleven quench parameters

Quench Parameter	A_i	Uncertainty in A_i	B_i	Uncertainty in B_i
779.11	0.025013	0.000510	-10.00472	0.04379
787.40	0.024914	0.000495	-10.16792	0.04267
806.02	0.024689	0.000526	-10.52628	0.04575
817.00	0.024554	0.000579	-10.73234	0.05060
820.73	0.024509	0.000601	-10.80146	0.05262
828.96	0.024407	0.000655	-10.95239	0.05759
834.95	0.024333	0.000698	-11.06089	0.06157
848.98	0.024158	0.000808	-11.31057	0.07168
862.28	0.023992	0.000917	-11.54156	0.08194
867.89	0.023922	0.000964	-11.63735	0.08638
890.57	0.023635	0.001156	-12.01478	0.10475

Thereafter, linear regression was applied to the values of the Quench Parameter against the Coefficient and Constant respectively, according to,

$$A_i = QP.X + Y \tag{4}$$

where,

A_i = Quench parameter dependent coefficient,
QP = Quench parameter – SQP(E)-value for the specific sample,
X = Quench parameter dependent coefficient, and
Y = Quench parameter dependent constant.

and

$$B_i = QP.R + S \tag{5}$$

where,

B_i = Quench parameter dependent constant,
QP = Quench parameter – SQP(E)-value for the specific sample,
R = Quench parameter dependent coefficient, and
S = Quench parameter dependent constant.

This gave the following results shown in Tables 3a and 3b,

Table 1a: Results of Quench Parameter against the Coefficient A_i

A_i	Regression Output:	
Y		3.4649E-02
Std Err of Y		4.0848E-06
R Squared		9.9992E-01
No. of Observations		11
Degrees of Freedom		9
X	-1.2360E-05	
Std Err of X	3.7685E-08	

Table 3b: Results of Quench Parameter against the Constant B_i

B_i	Regression Output:	
S		4.0525E+00
Std Err of S		1.9766E-02
R Squared		9.9909E-01
No. of Observations		11
Degrees of Freedom		9
R	-1.8081E-02	
Std Err of R	1.8235E-04	

This resulted in the following channel number -to- energy conversion Quench Parameter equation,

$$\text{Energy (MeV)} = \text{Channel Number}.\ \{[SQP(E)]\ .\ X + Y\} + \{[\ SQP(E)]\ .\ R + S\} \qquad (6)$$

This equation was verified with the 11 samples prepared from the ^{226}Ra standard reference solution and the results are shown in Tables 4a and 4b.

Table 4a: Results from verifying the equation from the ^{226}Ra standard reference solution

		^{214}Po			^{218}Po	
Energy (MeV)		7.687			6.002	
Quench parameter	Channel number	Energy (MeV)	% Deviation	Channel number	Energy (MeV)	% Deviation
779.11	707.2	7.661	-0.34%	638.7	5.947	-0.92%
787.40	716.5	7.668	-0.25%	648.2	5.968	-0.57%
806.02	737.1	7.677	-0.13%	669.5	6.007	0.09%
817.00	749.3	7.678	-0.12%	682.1	6.026	0.40%
820.73	753.5	7.677	-0.12%	686.3	6.032	0.50%
828.96	762.6	7.675	-0.16%	695.7	6.043	0.68%
834.95	769.3	7.672	-0.19%	702.6	6.049	0.79%
848.98	784.9	7.662	-0.33%	718.6	6.061	0.99%
862.28	799.6	7.647	-0.53%	733.8	6.067	1.09%
867.89	805.9	7.639	-0.63%	740.2	6.069	1.11%
890.57	831.1	7.599	-1.15%	766.2	6.064	1.03%
Averages		7.659	-0.36%		6.030	0.47%

Table 4b: Results from verifying the equation from the ^{226}Ra standard reference solution

		^{222}Rn			^{226}Ra	
Energy (MeV)		5.490			4.774	
Quench parameter	Channel number	Energy (MeV)	% Deviation	Channel number	Energy (MeV)	% Deviation
779.11	621.5	5.515	0.46%	590.0	4.728	-0.95%
787.40	630.5	5.526	0.66%	598.7	4.734	-0.83%
806.02	650.8	5.545	1.01%	618.2	4.741	-0.69%
817.00	662.7	5.552	1.13%	629.7	4.741	-0.70%
820.73	666.8	5.553	1.15%	633.6	4.740	-0.71%
828.96	675.8	5.555	1.19%	642.2	4.737	-0.78%
834.95	682.3	5.556	1.20%	648.5	4.734	-0.85%
848.98	697.6	5.553	1.14%	663.2	4.722	-1.08%
862.28	712.0	5.545	1.00%	677.1	4.707	-1.41%
867.89	718.2	5.540	0.92%	683.0	4.699	-1.57%
890.57	742.8	5.513	0.42%	706.7	4.659	-2.42%
Averages		5.541	0.93%		4.722	-1.09%

The relevance of the derived equation to other standard solutions was checked on the following reference solutions available; ^{238}U, ^{226}Ra, ^{210}Pb (^{210}Po), ^{209}Po, ^{229}Th, ^{232}Th and ^{232}U. The results obtained are shown in Table 5. These results seem to be in reasonable agreement taking the relatively poor resolution of the α-spectra into consideration. An example of the identification of NORM-nuclides in an unknown sample can be seen in Figure 2.

Table 5: Results from verifying equation (6) on other standard reference solutions.

Expected Energy (MeV)	Test solutions	Nuclides	Quench parameter	Channel number	Calculated Energy (MeV)	% Deviation
4.188	^{238}U	^{238}U	799.1	597	4.212	0.57%
4.756		^{234}U	799.1	623	4.827	1.49%
4.774	^{226}Ra	^{226}Ra	805.0	633	4.903	2.70%
5.490		^{222}Rn	805.0	659	5.517	0.50%
6.002		^{218}Po	805.0	684	6.108	1.76%
7.687		^{214}Po	805.0	758	7.856	2.20%
4.883	^{209}Po	^{209}Po	801.3	633	5.003	2.46%
5.304	^{210}Pb	^{210}Po	800.96	652	5.439	2.54%
3.998	^{232}Th	^{232}Th	799.5	588	3.997	-0.02%
5.400		^{228}Th	799.5	648	5.416	0.30%
5.673		^{224}Ra	799.5	666	5.842	2.97%
6.288		^{220}Rn	799.5	682	6.220	-1.08%
6.778		^{216}Po	799.5	710	6.882	1.53%
8.785		^{212}Po	799.5	799	8.987	2.30%
5.302	^{232}U	^{232}U	754.8	599	5.462	3.01%
5.400		^{228}Th	754.8	599	5.462	1.14%
5.673		^{224}Ra	754.8	605	5.605	-1.20%
6.288		^{220}Rn	754.8	630	6.201	-1.38%
6.778		^{216}Po	754.8	639	6.416	-5.34%
8.785		^{212}Po	754.8	725	8.467	-3.61%

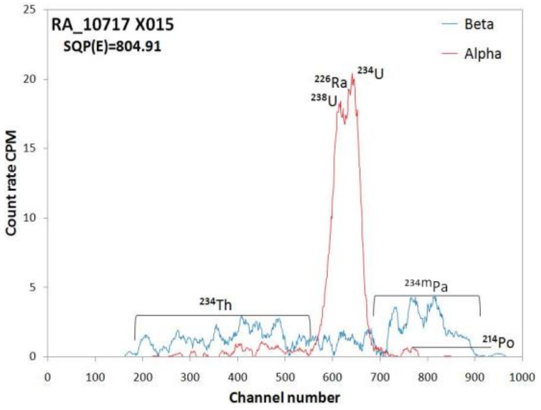

Figure 2: Spectra of the Quantulus 1220^{TM} from the unknown environmental water samples

4 CONCLUSION

From the results obtained, LSC together with special software for spectrum analysis can be used for the identification of α-active radionuclides in environmental water samples. The aim of this study was to establish a mathematical function that identifies α-active radionuclides in environmental water samples. This approach will be a convenient and cost-effective technique for a first order identification of α-active radionuclides in water samples, without going through elaborate and costly nuclide specific analysis.

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