Comparison of efficiency tracing and zero detection threshold techniques with CIEMAT/NIST standardization method under different quench conditions with Liquid Scintillation Spectrometer

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Efficiency tracing with ^{14}C and zero detection threshold techniques with ^{3}H as tracers are practical and simple methods which have been applied to determine the activity (DPM) of various beta-emitting radionuclides using Liquid Scintillation Spectrometer. Beta emitting radio-nuclides like ^{14}C (E_{max}, 156 keV), ^{36}Cl (E_{max}, 714 keV) and ^{204}Tl(E_{max}, 766 keV) were used for this study to determine the activity (DPM) with various quench levels. The results obtained using these methods are compared with CIEMAT/NIST standardization technique and were found to agree within ± 3.3% of the beta activity.

Keywords: CIEMAT/NIST, efficiency tracing, quenched samples, LSS, zero detection threshold.

CONVENTIONAL techniques such as sample channel ratio and external channel ratio are used for quench correction of the radionuclide being analysed using Liquid Scintillation Spectrometer (LSS). These techniques require a set of quench standards of the same radionuclide whose activity is to be determined. In the case of internal standard method, a standard of the same nuclide under assay is required, which may not be readily available in the laboratory. The results obtained using these methods provide an accuracy of ± 5%. All the above practical difficulties are overcome using efficiency tracing, zero detection threshold and CIEMAT/NIST techniques. Efficiency tracing and zero detection threshold methods do not require any quench correction curve of the particular radionuclide, which is an advantage. They require only a single unquenched ^{14}C and ^{3}H standard respectively. These three methods are simple and convenient to use. They are also useful for the determination of activities of short-lived radionuclides for which standards are not normally available. An attempt has been made to study the effect of quenching on the activity of the radionuclides using the above techniques and the results obtained are compared with CIEMAT/NIST standardization method.

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Packard Tricarb Model 2900TR Liquid Scintillation Spectrometer was used for the measurement that has the provision for measuring beta particle energy up to a maximum of 2000 keV. The scintillator used was Optiphase Hisafe-3 supplied by LKB Wallac, Finland. The unquenched ^{14}C and ^{3}H standards supplied along with the instrument were used as tracer for efficiency tracing and zero detection threshold methods respectively. Board of Radiation Isotope Technology supplied ^{36}Cl activity in liquid form, which was diluted to the required activity level. ^{204}Tl liquid solution used was supplied by Bureau International des Poids et Mesures (BIPM) France. Samples were prepared using 1ml of the activity in 15 ml of the Optiphase Hisafe-3 liquid scintillator in low potassium 25-ml glass vials.

CIEMAT/NIST method developed by Centro de Investigaciones Energeticas Medioambientales y Tecnologicas (CIEMAT) and the National Institute of Standards and Technology (NIST) was used for standardization of radionuclides with LSS[1,2]. Visual Basic CN2004 program was used for calculating the counting efficiency of the radionuclide to be assayed, where tritium is used as a tracer.

A set of quenched tritium standards (279500 dpm ± 1.6% – Standard Reference Material (SRM) 4947C supplied by NIST) supplied along with Packard Tricarb Model 2900TR was counted. A quench correction curve of tritium counting efficiency vs the quench indicating parameter (tSIE) was plotted (Figure 1 *a*). The tSIE value was calculated using the Ba-133 source by the instrument itself[3]. The theoretical counting efficiency of ^{3}H as a function of Figure of Merit (FOM) was calculated and plotted (Figure 1 *b*). From these two curves, the universal curve of FOM as a function of tSIE was plotted (Figure 1 *c*), which is independent of the radionuclide. The theoretical counting efficiency of the nuclides under study as a function of FOM was calculated and plotted (Figure 1 *d*). The nuclides under study were counted and tSIE values obtained were used to find the corresponding efficiencies using Figure 1 *c*, *d*. This method was used to find the activity in DPM of ^{14}C, ^{36}Cl and ^{204}Tl. The values obtained are shown in Table 1.

The theoretical counting efficiency for the radionuclide is calculated using the formula:

$$\varepsilon = \int_0^{Em} N(E)\{1-\exp(-(E*X(E))/2M\}^2 \mathrm{d}E,$$

where $N(E)$ is the number of particles of energy E (for beta-emitting radionuclide using Fermi theory of beta decay); $X(E)$ is the ionization quench factor = $1/E \int_0 \mathrm{d}E/1 + kB(\mathrm{d}E/\mathrm{d}X)$; $kB = 0.0075$ cm/MeV also called kB factor; $\mathrm{d}E/\mathrm{d}X$ is the linear stopping power for electron in MeV/cm; E is the energy of the particle; M is the FOM. (It is defined as the amount of energy required to produce one photoelectron at the photo cathode of PMT. The square term in the equation is due to two PMTs in coincidence being used.

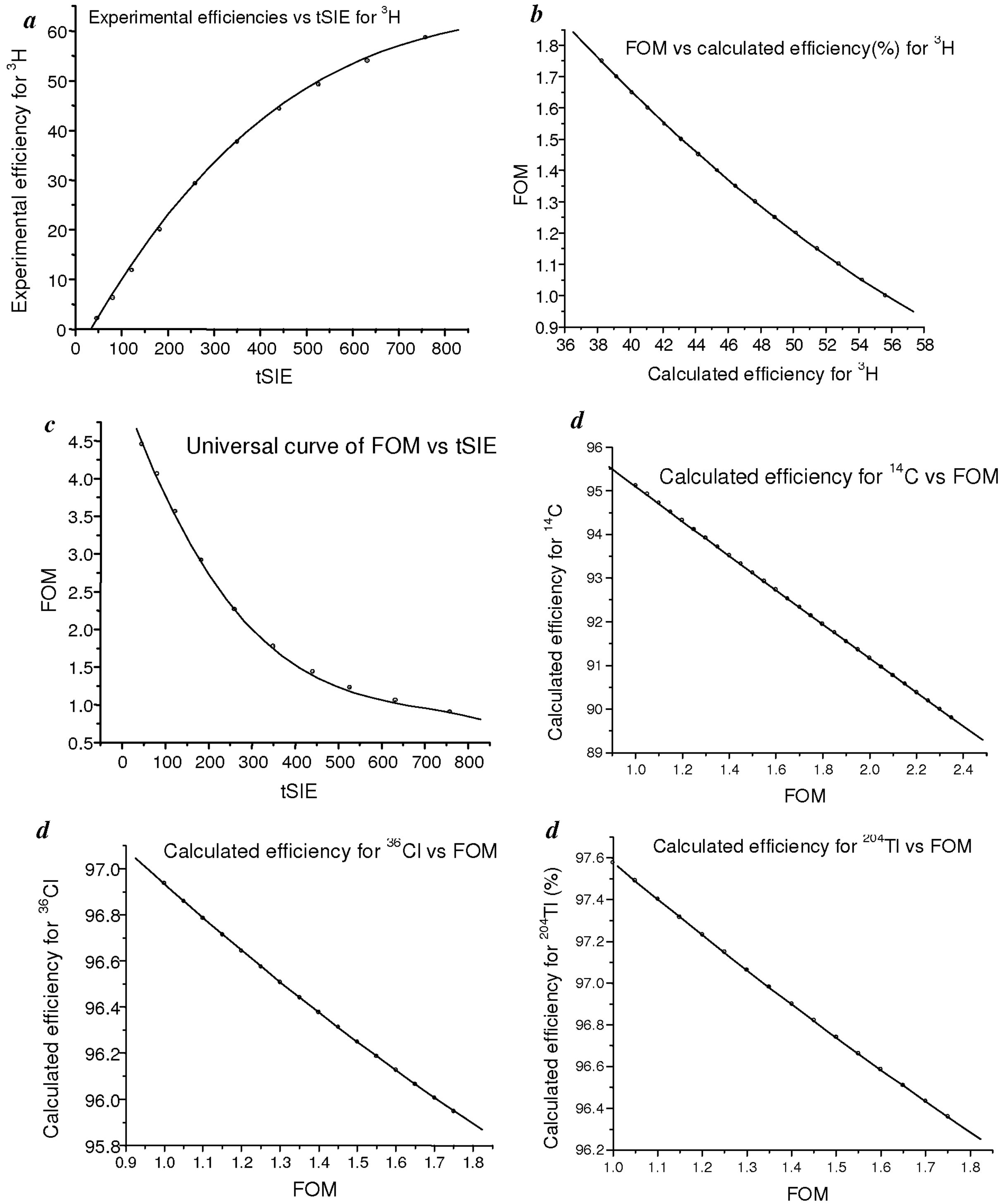

Figure 1. Plots for CIEMIT/NIST method.

The efficiency tracing technique[4] has been applied to determine the activity of nuclides using the counting efficiency of reference standard of unquenched ^{14}C. An unquenched ^{14}C standard was used as a tracer, because of its long half-life and counting efficiency more than 90% that can be accurately determined. It is a beta-emitter and standards are easily available.

The reference standard ^{14}C (124100 ± 1.3%) DPM used as tracer was procured from Packard Bioscience Company.

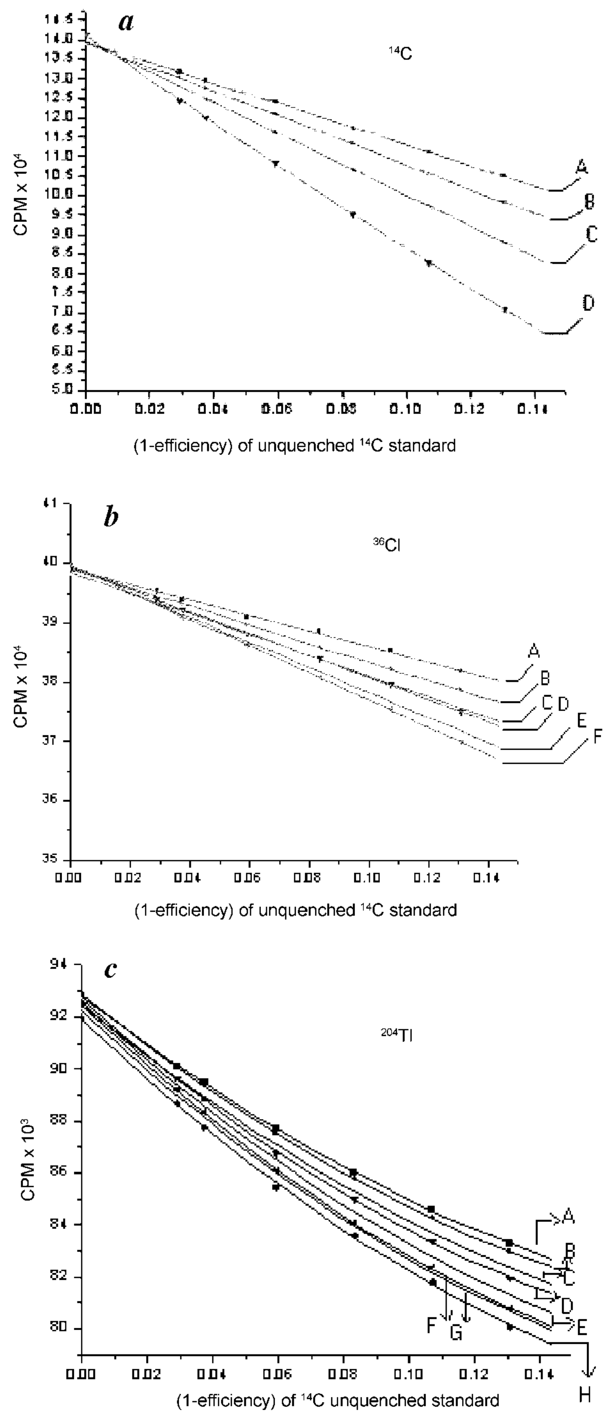

Figure 2. Efficiency tracing for ^{14}C (***a***), ^{36}Cl (***b***) and ^{204}Tl (***c***) under different quench conditions.

In this method unquenched ^{14}C standard was used as a tracer to determine the activity of ^{36}Cl and ^{204}Tl. Unquenched ^{14}C standard was counted in the counting regions 0–2000, 2–2000, 4–2000, 6–2000, 8–2000 and 10–2000 keV, where 0, 2, 4, 6, 8, 10 keV were the lower limit regions (LL) and 2000 keV was upper limit region (UL) on keV scale. Efficiency of the unquenched ^{14}C standard was determined in these counting regions. A set of quenched ^{14}C standards that was supplied along with the instrument was also counted in the same regions as that of the tracer to determine their activity. A set of quenched ^{36}Cl and ^{204}Tl samples was prepared by adding 1 ml of the sample to 15 ml of the Optiphase Hisafe-3 scintillator and increasing amount of carbon tetrachloride (CCl_4) as a quenching agent. These quenched samples were also counted in the same regions. The count rates obtained were plotted against the efficiencies of unquenched ^{14}C standard and extrapolated to (1-efficiency%) of unquenched ^{14}C standard to give the activity in DPM of these quenched samples Figure 2 *a*–*c*. Table 1 indicates the activity to 100% counting efficiency of quenched samples.

The zero detection threshold technique reported[5,6] has been applied to determine the activity of a quenched set of radionuclides. The term zero detection threshold refers to the determination of pulse height, where all beta particles of ^{3}H are taken into account. An unquenched ^{3}H standard was used as a tracer because it has long half-life and standards are easily available. It is a low-energy beta-emitter and hence is the most suitable standard for this technique. The reference standard ^{3}H (287100 ± 1.6%) DPM used as tracer was procured from Packard Bioscience Company.

In this method unquenched ^{3}H standard has been counted in the counting regions 0–2000, 2–200, 4–2000, 6–2000, 8–2000, 10–2000 keV, where 0, 2, 4, 6, 8, 10 keV denote the LL and 2000 keV the UL of the pulse height (keV) of the instrument. The counts obtained in these levels were plotted at several pulse heights (keV) of the instrument. The graph obtained was extrapolated to the count rate, which is equivalent to the activity in DPM of the ^{3}H standard. The keV value at this count rate represents the 'zero detection threshold' of the LSS instrument shown in Figure 3 *a*. Thus the zero detection threshold of our LSS is set at 5 keV on *X* negative scale. This zero detection threshold is used to find the activity of the radionuclides under study. A set of quenched ^{14}C, ^{36}Cl and ^{204}Tl samples was also counted in the same regions as that of the reference standard. The count rates obtained were plotted against the LL and extrapolated to zero detection threshold to give the activity in DPM of these quenched samples (Figure 3 *b*–*d*). Table 2 indicates the activity.

The slopes of the efficiency tracing curves and zero detection threshold curves vary with increasing amount of quencher. This is because quenching effect reduces pulse height. The extrapolated value to 100% counting efficiency in the case of efficiency tracing technique and zero detection threshold in the case of zero detection threshold technique converges to the same standardized DPM with some error. It is observed that in the case of ^{14}C whose energy is only 156 keV, if the tSIE value of the sample is

Table 1. Activity of ^{14}C, ^{36}Cl and ^{204}Tl under different quench conditions using efficiency tracing method

Radio-nuclide	Activity in DPM/ml ± % (as indicated in the sample)	Activity obtained by CIEMAT/NIST in DPM/ml	Experimental DPM/ml ± %	Per cent deviation obtained by CIEMAT/NIST in DPM/ml ± %	tSIE
^{14}C					
A	139500 ± 1.6	139944.38 ± 0.24	139157.8 ± 0.91	0.38	632.74
B		139841.52 ± 0.24	139407.7 ± 0.92	0.54	523.00
C		139659.21 ± 0.24	139091.1 ± 0.92	0.70	426.90
D		139540.71 ± 0.24	139562.3 ± 0.93	1.20	338.97
E		138975.8 ± 0.24	143424.2 ± 0.93	3.20	247.20
^{36}Cl					
A	400000 ± 10	392813.22 ± 0.96	398999.9 ± 1.10	1.57	338.17
B		393117.54 ± 0.96	399244.5 ± 1.10	1.55	317.74
C		392250.84 ± 0.96	398434.4 ± 1.10	1.57	295.48
D		394498.44 ± 0.96	399462.8 ± 1.10	1.26	219.53
E		394665.00 ± 0.96	399689.2 ± 1.10	1.27	212.76
F		395060.46 ± 0.96	399764.8 ± 1.10	1.19	194.53
^{204}Tl					
A	91284.4 ± 0.75	93461.54 ± 0.94	92762.47 ± 1.07	0.75	459.94
B		93536.80 ± 0.94	92871.63 ± 1.07	0.71	422.83
C		93196.47 ± 0.94	92476.52 ± 1.07	0.77	411.92
D		93319.02 ± 0.94	92654.50 ± 1.70	0.74	355.01
E		93189.47 ± 0.94	92631.96 ± 1.70	0.71	341.42
F		92924.95 ± 0.94	92528.90 ± 1.70	0.71	312.60
G		93083.60 ± 0.94	92261.70 ± 1.70	0.89	278.07

Table 2. Activity of ^{14}C, ^{36}Cl and ^{204}Tl under different quench conditions using zero detection threshold

Radio-nuclide	Activity in DPM/ml ± % (as indicated in the sample)	Activity obtained by CIEMAT/NIST in DPM/ml	Experimental DPM/ml ± %	Per cent deviation obtained by CIEMAT/NIST in DPM/ml ± %	tSIE
^{14}C					
A	139500 ± 1.3%	139944.38 ± 0.28	138663.9 ± 1.25	0.91	632.74
B		139841.52 ± 0.28	138775.4 ± 1.26	0.76	523.00
C		139659.21 ± 0.28	138320.0 ± 1.26	0.96	426.90
D		139540.71 ± 0.28	138667.1 ± 1.26	0.63	338.97
E		138975.80 ± 0.28	142365.64 ± 0.24	2.44	247.20
^{36}Cl					
A	400000 ± 10%	392813.22 ± 0.96	399091.5 ± 1.17	1.60	338.17
B		393117.54 ± 0.96	400647.7 ± 1.17	1.90	308.75
C		392250.84 ± 0.96	398345.1 ± 1.17	1.55	295.48
D		394498.44 ± 0.96	399350.9 ± 1.17	1.23	219.53
E		394665.00 ± 0.96	399148.3 ± 1.17	1.14	212.76
F		395060.46 ± 0.96	399092.4 ± 1.17	1.02	194.53
G		395232.30 ± 0.96	399232.1 ± 1.17	1.01	175.29
^{204}Tl					
A	90550.164 ± 0.75	93461.54 ± 0.94	94082.8 ± 1.14	0.66	459.94
B		93536.80 ± 0.94	94143.2 ± 1.14	0.65	422.83
C		93196.47 ± 0.94	93882.6 ± 1.14	0.74	411.92
D		93319.02 ± 0.94	93990.2 ± 1.14	0.81	355.01
E		93189.47 ± 0.94	94072.2 ± 1.14	0.85	341.42
F		92924.95 ± 0.94	93983.8 ± 1.14	0.92	312.60
G		93083.60 ± 0.94	93779.7 ± 1.14	0.48	278.07

less than 339, the error is less than 3.5% in determination of the activity taking into account all the methods, whereas in the case of high energy betas such ^{36}Cl and ^{204}Tl the error is 1.6 and 0.90 respectively. This is due to difference in the energy of the radionuclides under study.

The results obtained under study are within ± 3.3%. It has been found that the efficiency tracing technique cannot be adopted to the radionuclides with energy less than 156 keV, whereas zero detection threshold technique is used for low-energy beta-emitting radionuclides. Even

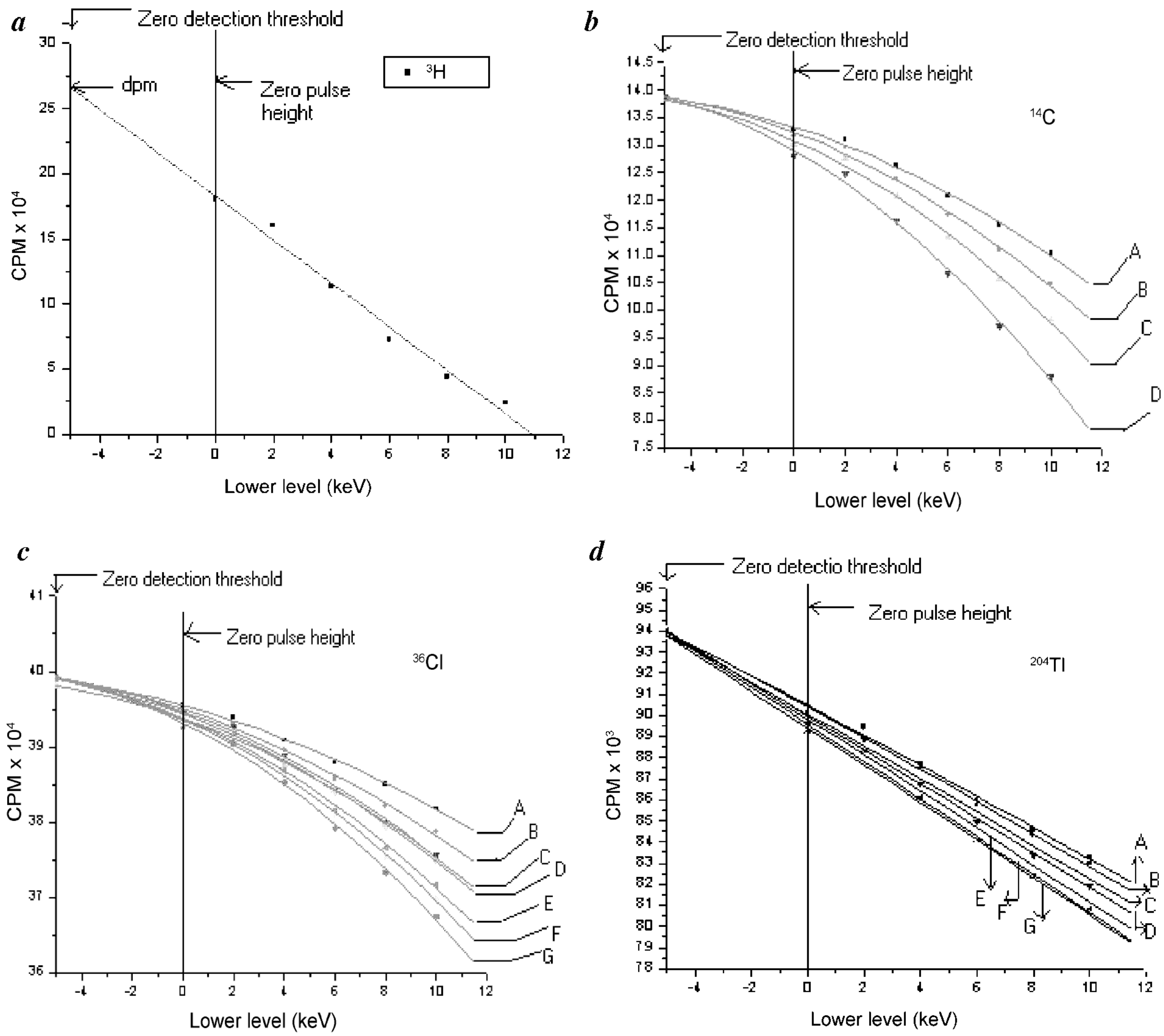

Figure 3. ***a***, Determination of zero detection threshold using ^{3}H standard. ***b–d***, Extrapolation of zero pulse height to zero detection threshold to get the DPM of ^{14}C (***b***), ^{36}Cl (***c***) and ^{204}Tl (***d***) under different quench conditions.

though specific radionuclides have been selected for this study, it is concluded that the proposed methods can be applied to all beta-emitters with the available LSS without any special arrangement.

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