Study of the Performance of Efficiency Tracing Technique on a TriCarb 2100TR Liquid Scintillation Analyzer

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ABSTRACT

The liquid scintillation Efficiency Tracing (ET) technique is a practical method of quantifying radionuclides being analyzed in a liquid scintillation analyzer. This technique has several advantages over conventional liquid scintillation methods: no quench curve (quenched standard set) is required for each nuclide being analyzed; only a single unquenched $^{14}$C sample (same as that used to normalize the liquid scintillation analyzer) is required to calculate radionuclide activity (DPM) or concentration; the technique can be used effectively for almost all pure beta and beta-gamma emitters (minimum energy = 70 keV).

A Tri-Carb 2100TR Liquid Scintillation Analyzer from Packard was used to perform a series of experimental works to assess performances of ET method. In this paper are presented the results obtained for activity determination of $^{14}$C samples with various quench and activity levels. The experimental tests were made using both the ET method implemented on the analyzer by the producer and our ET method, using the reference spectrum of the unquenched $^{14}$C standard, used to normalize the liquid scintillation analyzer. The results indicate that the measured values using implicit ET method are in good agreement (uncertainty $\leq$ 3.5 %) with the reference values only for samples with a quenching level not very high (quenching indicating parameter tSIE $>$ 200 - 300). Otherwise, using our ET method, good results (percent recovery more than 96.5%) were obtained for any quenching level (tSIE $>$ 100) even at a count rate level one order of magnitude lower than it is recommended for this method.

1 INTRODUCTION

Compared to other methods for radioactivity measurement, the liquid scintillation techniques have remarkable advantage that radiations can be measured without self-absorption, external absorption and scattering under $4\pi$ geometry. This advantage leads to an accurate radiation measurement with a high counting efficiency. Nevertheless, other perturbation phenomena can arise. The most important of them is the quenching, which refers to any factor which results in the loss of photon emission from the sample to be measured. In liquid scintillation counting, the terms counting efficiency and quenching are synonymous and the counting efficiency of the counting system must be determined. In other words, to quantify the radioactivity in a sample the conversion from counts per minute (CPM) to disintegrations per minute (DPM) is necessary.

Different quench monitoring techniques were already developed by the suppliers of liquid scintillation analyzers. Packard Instrument Company (the supplier of the TriCarb 2100TR Liquid Scintillation Analyzer from the Institute for Nuclear Research – Pitesti) developed the tSIE (Transformed Spectral Index of the External Standard) quench monitoring
The method [1]. The tSIE quench indicating parameter is determined by evaluating the spectrum of the sample and Compton electron spectrum of a $^{133}$Ba external standard source with a Spectralyzer Multichannel Analyzer, using a mathematical spectral transform technique. The tSIE quench correction curve for a certain radionuclide is obtained measuring a set of standards from that radionuclide having the same activity and different quench levels. Representative results obtained for $^3$H and $^{14}$C samples in our laboratory were presented in [2].

Generally, the methods based on quench monitoring techniques are very accurately, but they need a set of $^3$H and $^{14}$C quenched standards, which are commercially available. Nevertheless, these quenching correction techniques are not very suitable for the routine measurement of nuclides other than $^3$H and $^{14}$C, because it is troublesome and time consuming to prepare individual quenched standards. In this case, more suitable seem to be those specific methods which do not require quench correction curves to assess $\beta$-radioactivity of a sample. Such a method is the Efficiency Tracing (ET) technique.

### 2 EXPERIMENTS

The principle of the ET technique in the liquid scintillation is based on the experimental results that, under a given condition in which a $\beta$-reference sample can be measured with 100% counting efficiency, the samples to be measured can also be measured with the same counting efficiency (100%) [3]. Liquid scintillation counting rates of the reference sample and the sample to be measured vary with the lower discriminator level of the counter. The counting rates of the sample to be measured are plotted against the counting efficiency of the reference sample at each discriminator level and the value extrapolated at 100% counting efficiency along the efficiency tracing curve provides the activity of the sample.

The radionuclide used as reference sample should be selected as follows:

1. it should have a long half-life;
2. the maximum counting efficiency achievable should be greater than 90%;
3. it should be a $\beta$-emitter, not giving rise to electron capture decay or isomeric transition.

On the basis of the foregoing aspects, apparently $^{14}$C is the most suitable reference sample.

In the ET technique, the most important fact is to select the appropriate amplifier gains, with which the counting rates of the reference sample and sample to be measured vary. The amplifier gains should be set so as to give a counting efficiency from 80 to 95% for the reference sample.

The differences in quenching effect, sample volume, radiation energy and kind of liquid scintillation between the reference sample and sample to be measured do not affect the final result in practice (total activity); these factors may vary the slope of the efficiency tracing curve, but the extrapolated value of the 100% counting efficiency converges, within the measurement uncertainty, to the same point which gives the total activity.

In the case where two or several radionuclides are present in a prepared sample, the extrapolated value obviously indicates the total activity.

The efficiency tracing technique implemented by the Packard Instrument Company on the TriCarb 2100TR Liquid Scintillation Analyzer from the Institute for Nuclear Research – Pitești is based on a patent-pending procedure which uses the unquenched $^{14}$C standard (used also to normalize the analyzer) [1]. The reference spectrum of this standard is analyzed and the counting efficiency is determined in six separate regions simultaneously: 0-2000, 2-2000, 4-2000, 6-2000, 8-2000, 10-2000 keV. When an unknown sample is measured, the function Data Mode is set to eff tracing dpm option and the analyzer provides directly the radionuclide
activity (DPM). The method is very simple, but has an important disadvantage: it is inappropriate for measuring low activity samples. This method has no background correction and for this reason Packard recommends its use for count rates greater than 1500 cpm.

To extend the domain of the applicability of the ET method to lower count rates, we implemented our ET technique, using the same procedure: the spectrum of the unquenched $^{14}$C standard with certified activity of $133100 \text{ dpm/vial} \pm 1.5\%$ at 25 Aug. 2000 was recorded and analyzed in the same six counting regions and the efficiency counting for each region was calculated. When an unknown sample is measured, its spectrum is also recorded and analyzed in the same six regions and the counting rates (CPM) are plotted against the counting efficiency of the reference sample at each discriminator level. A curve is fitted through these six points and extrapolated to 100% efficiency; the extrapolated CPM value at this point is equal to the number of DPM or sample activity. In this case, because all spectra are recorded and analyzed by the operator, background correction can be made, using adequate blank samples.

In order to assess the reliability and accuracy for determining DPM values using ET technique, a series of samples with various levels of quench and radioactivity were measured. All samples were contaminated with known concentrations of $^{14}$C ($E_{\text{max}} = 156 \text{ keV}$). Comparative, based on the same measurements, the activity of each sample was calculated using the more accurately tSIE method. It must be mention that owing to specific measurement conditions (the spectra were acquired and analyzed on the entire energy range, up to 2000 keV), the AEC (Automatic Efficiency Control) feature of the analyzer did not operate and so automatically optimum region of interest adjustment was not performed.

Two kinds of sample were used:

• to assess the influence of quenching over the entire tSIE range (100 – 1000) the set of ten quenched $^{14}$C standard from Packard (certified activity of $134500 \text{ dpm/vial} \pm 1.5\%$ at 5 June 2001) was used;

• to assess the performance depending of the level of activity experimental samples with different levels of $^{14}$C concentration were prepared using a standard solution ($^{14}$C Toluene Internal Standard with certified concentration of $4.99 \times 10^5 \pm 3\% \text{ dpm/g}$ at 5 Jun. 2001) from Packard. For dilutions free toluene and distilled water were used. All samples were prepared and measured in 20 ml polyethylene vials using Ultima Gold XR LS cocktail.

3 RESULTS AND DISCUSSION

The first experiment: influence of quench level

The first experiment was made to verify the accuracy of the ET technique for various levels of quenching. Ten measurements for each level quench, corresponding to quenched $^{14}$C standards were made. The counting time was selected to achieve a $\%2$ sigma standard deviation $\%(2s)$ of 0.5% or lower of the count rate. The obtained results (measured $^{14}$C activities) are expressed as $DPM ET$ for Packard’s ET method, $DPM ET^*$ for our ET method and as $DPM tSIE$ for the conventional tSIE method and are presented in Table 1 as relative discrepancy (%) between mean of the measured values and known reference values. The quench level for each standard is given by the corresponding tSIE parameter.
Table 1: Discrepancy between measured values and known reference values of the activity of the $^{14}$C quenched standard set (ten levels of quench)

<table>
<thead>
<tr>
<th>Sample</th>
<th>tSIE</th>
<th>Discrepancy (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>DPM ET</td>
</tr>
<tr>
<td>A</td>
<td>1055</td>
<td>0.5</td>
</tr>
<tr>
<td>B</td>
<td>909</td>
<td>0.6</td>
</tr>
<tr>
<td>C</td>
<td>691</td>
<td>1.1</td>
</tr>
<tr>
<td>D</td>
<td>518</td>
<td>0.8</td>
</tr>
<tr>
<td>E</td>
<td>404</td>
<td>1.8</td>
</tr>
<tr>
<td>F</td>
<td>302</td>
<td>2.7</td>
</tr>
<tr>
<td>G</td>
<td>229</td>
<td>3.5</td>
</tr>
<tr>
<td>H</td>
<td>182</td>
<td>5.8</td>
</tr>
<tr>
<td>I</td>
<td>144</td>
<td>9.0</td>
</tr>
<tr>
<td>J</td>
<td>106</td>
<td>14.2</td>
</tr>
</tbody>
</table>

The results in Table 1 show that the measured values of the activity using the ET technique implemented by Packard on the our TriCarb 2100 TR LSA to quantify β-radionuclides are in good agreement with the reference values (relative discrepancy ≤ 3.5%) only for samples with a quenching level not very high. In the case of the samples with pronounced quenching effect (values of the quench parameter tSIE lower than around 200) the accuracy is more affected (discrepancy of about 15% for the extreme value 100 attributed to the tSIE parameter).

The second experiment: influence of activity level

The second experiment was made to verify if our ET method (in which all β-spectra including the reference spectrum, are recorded on disk and transferred to a desktop computer for analysis, and which allows background correction) provides good results below the recommended activity level. As mention above, because the implicit ET method makes no background correction, it is recommended only for relatively high count rates (more than 1500 cpm) [3]. For a sample containing $^{14}$C, measured with a 90% efficiency counting, this is equivalent with an activity of 1667 dpm/sample.

Four series of counting vials were prepared containing varying amounts of $^{14}$C: 2088, 857, 418 and 189 dpm/sample (four or five replicates for each level of activity). Only the first level of activity was greater than the recommended limit, while the last level was lower approximately one magnitude order than this limit. For background contribution blanks were prepared with the same compositions like the samples, but without $^{14}$C. To reduce pipetting errors the aliquots of solutions used to prepare samples were weighed (uncertainty of 0.001g). All vials were counted for 60 min. each to achieve a %2 sigma standard deviation %($2\sigma$) of 2% or lower of the count rate in each case. The results are presented in Table 2 in the same manner like in the first experiment.

In according with the results in Table 2, for samples contaminated with $^{14}$C and measured with a counting efficiency of about 90%, implicit ET method makes good results (percent recovery more than about 95%) for samples with activity greater than 850 dpm/sample (∼750 cpm). For the last two series of samples, relative discrepancy raised to about 10% for activity level of 418 dpm and about 25% for activity level of 189 dpm. Obviously, because the implicit ET technique makes no background correction all discrepancies are positively, supra-estimating the real values of the activity.
Table 2: Discrepancy between measured activities and known reference activities of the $^{14}$C experimental samples (four levels of activity)

<table>
<thead>
<tr>
<th>Sample</th>
<th>tSIE</th>
<th>Discrepancy (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Discrepancy (%)</td>
</tr>
<tr>
<td></td>
<td>DPM ET</td>
<td>DPM ET*</td>
</tr>
<tr>
<td>C1</td>
<td>224</td>
<td></td>
</tr>
<tr>
<td>2088 dpm</td>
<td>4.9</td>
<td>2.8</td>
</tr>
<tr>
<td></td>
<td>5.5</td>
<td>3.4</td>
</tr>
<tr>
<td></td>
<td>4.0</td>
<td>1.9</td>
</tr>
<tr>
<td></td>
<td>3.2</td>
<td>1.1</td>
</tr>
<tr>
<td></td>
<td>3.7</td>
<td>1.6</td>
</tr>
<tr>
<td>C2</td>
<td>276</td>
<td></td>
</tr>
<tr>
<td>857 dpm</td>
<td>4.9</td>
<td>-0.2</td>
</tr>
<tr>
<td></td>
<td>4.8</td>
<td>-0.3</td>
</tr>
<tr>
<td></td>
<td>4.7</td>
<td>-0.4</td>
</tr>
<tr>
<td></td>
<td>5.2</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>5.8</td>
<td>0.6</td>
</tr>
<tr>
<td>C3</td>
<td>227</td>
<td></td>
</tr>
<tr>
<td>418 dpm</td>
<td>11.5</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>8.2</td>
<td>-2.3</td>
</tr>
<tr>
<td></td>
<td>9.0</td>
<td>-1.5</td>
</tr>
<tr>
<td></td>
<td>9.5</td>
<td>-1.0</td>
</tr>
<tr>
<td>C4</td>
<td>257</td>
<td></td>
</tr>
<tr>
<td>189 dpm</td>
<td>25.0</td>
<td>1.8</td>
</tr>
<tr>
<td></td>
<td>24.2</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>24.3</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>25.8</td>
<td>2.5</td>
</tr>
</tbody>
</table>

Very hopeful results were obtained using the ET technique implemented by us. Practically, all results are equally accurate as those provided by the tSIE method, which is known as a more sensitive method. Percent recovery more than 96.5% were obtained for any quenching level over the entire measuring domain of our liquid scintillation analyzer (tSIE = 100 – 1000) and for any considerate level of activity. Even at a count rate level lower about one magnitude order than the recommended limit for this method the results were also very accurate.

4 CONCLUSIONS

The efficiency tracing technique presented in this paper can be successfully applied to the radioassay of beta activity in samples contaminated with almost all pure beta and beta-gamma emitters (minimum energy = 70 keV), using only the individual analyzer for which the method were developed (in our case, a model of TriCarb 2100TR Liquid Scintillation Analyzer from Packard Instrument Company).

Compared to other methods using liquid scintillation spectrometry, the method presented here offers a simple technique, less labour intensive, and requiring only basic skills and basic equipment. No quench correction curves or quench correction algorithms are needed. Only a single unquenched $^{14}$C standard of known activity is required to calculate radionuclide activity even if the samples are strongly quenched.

In conclusion, the efficiency tracing method is very useful for routine radioassay and extremely useful for the standardization of radionuclides of short-life for which quenched standards are not available.
REFERENCES


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