STUDY OF THE PERFORMANCE OF EFFICIENCY TRACING TECHNIQUE ON A TRICARB 2100TR LIQUID SCINTILLATION ANALYZER

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The liquid scintillation Efficiency Tracing (ET) technique is a practical method of quantify radionuclides being analyzed in a liquid scintillation analyzer. This technique has several advantages over conventional liquid scintillation methods: i) no quench curve (quenched standard set) is required for each nuclide being analyzed; ii) the technique can be used effectively for almost all pure beta and beta-gamma emitters (minimum energy = 70 keV); iii) only a single unquenched ¹⁴C sample (same as that used to normalize the liquid scintillation analyzer) is required to calculate radionuclide activity (DPM) or concentration; iv) the efficiency tracing techniques provides a simple method for quantify of DPM in the sample; v) relatively small errors (1–5%) in the calculation of DPM are reported in literature; vi) different radionuclides can be intermixed in the sample batch. The most important disadvantages are: i) is not applicable to tritium and to radionuclides which decay by isomeric transitions and electron capture; ii) for samples of low count rates (< 1500 cpm), it is recommended that one use a conventional method, which utilizes quench correction curves. Radiation Protection and Environmental Protection Laboratory from the Institute for Nuclear Research – Pitesti is endowed with a Tri-Carb Liquid Scintillation Analyzer, model 2100TR. A series of experimental works were performed to assess performances of Efficiency Tracing method. In order to verify the usefulness of the technique, prepared samples containing known amounts of ¹⁴C activities were tested. 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 ¹⁴C standard, used to normalize the liquid scintillation analyzer. The results indicate that the measured values using implicit ET method are in good agreement (uncertainty ≤ 3%) 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 95%) were obtained for any quenching level (tSIE > 100) even at a count rate level lower one magnitude order than that is recommended for this method.

Key words: liquid scintillation, reference spectrum, efficiency tracing curve.


1. INTRODUCTION

The principle of the efficiency tracing technique in the liquid scintillation is based on the experimental results that, under a given condition in which a β-reference sample can be measured with 100% counting efficiency, the samples to be measured are also possibly measured with the same counting efficiency (100%) [1]. 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 reference sample should be selected from the following points of view [2]:

(1) it has long life, being accurately radio assayed;
(2) it can be measured with the maximum counting efficiency of more than 90%;
(3) it is a β-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 efficiency tracing 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 [2].

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.

2. EXPERIMENTAL

The efficiency tracing technique implemented by the Packard Instrument Company on the TriCarb 2100TR Liquid Scintillation Analyzer from the Institute for Nuclear Research – Pitesti is based on a patent-pending procedure which uses the unquenched $^{14}$C standard (used also to normalize the analyzer).
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 the disadvantage that the background correction is not possible and in conclusion for samples of low count rates the results will be strong affected.

Concomitantly we implemented our efficiency tracing technique, using the same procedure: the spectrum of the unquenched 14C standard with certified activity of 133100 dpm ± 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 an adequate blank sample.

In order to assess the reliability and accuracy for determining DPM values, a series of samples with various degrees of quenching and levels of radioactivity were quantitated using both efficiency tracing techniques. All samples were contaminated with known concentrations of 14C (E_{max} = 156 keV).

Two kinds of sample were used:

i) the set of quenched standard from Packard: Carbon-14 Toluene Standard (set of ten) 134500 dpm/vial ±1.5%, at 5 June 2001. These samples were used in the experiment where the influence of quenching was examined;

ii) experimental samples with different levels of 14C concentration, prepared using a standard solutions from Packard (14C Toluene Internal Standard with certified concentration of 4.99 × 10^5 ± 3% dpm/g at 5 Jun. 2001). For dilutions were used free toluene and distilled water. All samples were prepared and measured in 20 ml polyethylene vials and using Ultima Gold XR LS cocktail.

3. RESULTS

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 14C standards were made using both efficiency
tracing technique implemented by the supplier of the TriCarb 2100 TR LSA and efficiency tracing technique implemented by our laboratory. The counting time was selected to obtain a statistical uncertainty (±2σ) of the count rate lower than 0.5% in each case. The obtained results (measured $^{14}$C activities) are expressed as $DPM \ ET$ for the first method and as $DPM\ \ ET^*$ for the second 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 (Transformed Spectral Index of the External Standard).

**Table 1**

Discrepancy between measured values and known reference values of the activity of the $^{14}$C quenched standard set (ten levels of quench), using two efficiency tracing techniques

<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>

Because the implicit ET method (the method implemented by the supplier – Packard) does not background correction, it is recommended only for relative 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. The second experiment was made to verify if our ET method (in which all $\beta$-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. 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 0.001 g). All vials were
counted for 60 min. each, the statistical uncertainty (%2s) of the count rate being lower than 2% in each case. The results are presented in Table 2, in the same manner like in the first experiment.

Table 2
Discrepancy between measured activities and known reference activities of the 14C experimental samples (four levels of activity), using two efficiency tracing techniques

<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>C1 2088 dpm</td>
<td>224</td>
<td>4.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.7</td>
</tr>
<tr>
<td>C2 857 dpm</td>
<td>276</td>
<td>4.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5.8</td>
</tr>
<tr>
<td>C3 418 dpm</td>
<td>227</td>
<td>11.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>8.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.5</td>
</tr>
<tr>
<td>C4 189 dpm</td>
<td>257</td>
<td>25.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>24.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>24.3</td>
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<tr>
<td></td>
<td></td>
<td>25.8</td>
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</tbody>
</table>

4. DISCUSSION

The results in Table 1 show that the measured values of the activity using the efficiency tracing technique implemented by Packard on the our TriCarb 2100TR LSA to quantify β-radiouclides 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). On the other hand, because the implicit ET technique does not background correction, it provides good results (percent recovery more than about 95%) only for activities not very low. In according with the results in
Table 2, for samples contaminated with \(^{14}\text{C}\) and measured with a counting efficiency of about 90\%, good results were obtained for samples 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 418 dpm. Obviously, all discrepancies are positively, supra-estimating the real values of the activity.

Very good results were obtained using the ET technique implemented by us on the our analyzer. Percent recovery more than 96.5\% were obtained for any quenching level (tSIE > 100) and any level of activity. Even at a count rate level lower about one magnitude order than the recommended limit for this method the results were very accurate.

5. 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, the TriCarb 2100TR Liquid Scintillation Analyzer from Packard Instrument Company, Model No. A210001, Instrument Serial No. 425675).

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 \(^{14}\text{C}\) standard of known activity (same as that used to normalize the liquid scintillation analyzer) is required to calculate radionuclide activity even if the samples are strongly quenched (tSIE equal to 100).

REFERENCES