CHAPTER 59

Absolute Activity Liquid Scintillation Counting: An Attractive Alternative to Quench-Corrected DPM for Higher Energy Isotopes

Michael J. Kessler, Ph.D.

The liquid scintillation efficiency tracing technique is a new and powerful method of quantitating radionuclides being analyzed in a liquid scintillation analyzer.[1-5] This technique has several advantages over conventional DPM analysis. First, no quench curve (quenched standard set) is required for each nuclide. Second, the technique can be used effectively for almost all pure beta and beta-gamma emitters. Third, only a single unquenched ^{14}C sample (same as that used to normalize the liquid scintillation analyzer) is required to calculate DPM results. Fourth, the efficiency tracing technique provides a simple method for DPM quantitation in the sample. Fifth, relatively small errors (105%) in the calculation of DPM can be achieved using this technique. Sixth, different radionuclides can be intermixed in the sample batch.

This efficiency tracing technique is based on a patented procedure which requires a liquid scintillation analyzer containing a multichannel analyzer, a sophisticated data reduction system, and a standard radionuclide (sealed calibration standard) of known absolute activity. The efficiency tracing procedure is accomplished in the following manner. First, the system is standardized (normalized) with an unquenched ^{14}C standard. The reference spectrum of this standard is analyzed, and the counting efficiency is determined in six separate regions simultaneously. Second, the % efficiency in each of the six regions is calculated and plotted against the actual number of counts in each region. Figure 1 shows a typical % efficiency vs CPM plot for the ^{14}C standard. Third, an extrapolated CPM value for the 100% efficiency is determined based on the plot of the resultant line analyzed by the method of least squares. Fourth, when an unknown sample is analyzed, its spectrum is analyzed in the same six regions, and the results are plotted using the same x-axis (% efficiency) generated by the ^{14}C standard. A line is then drawn through the resultant points. A curve is fitted (least square) through these six points and extrapolated to 100%

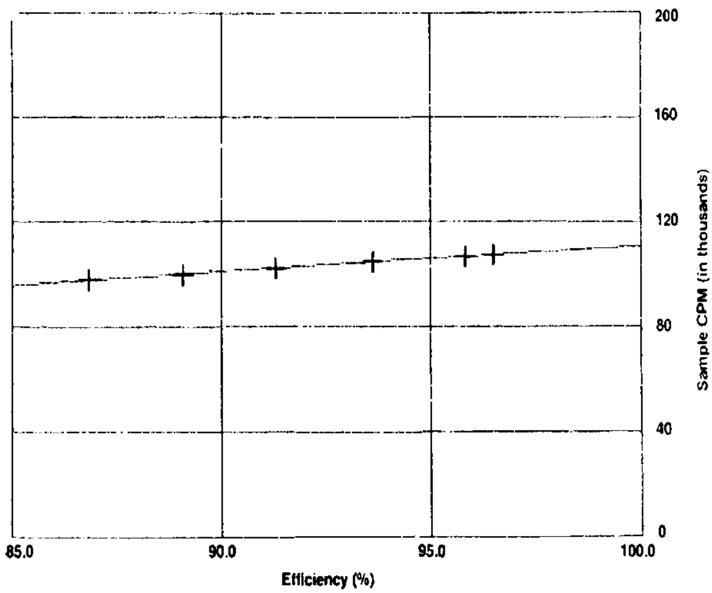


Figure 1. Typical efficiency tracing^{TM} plot for ^{14}C unquenched sample.

efficiency. The extrapolated CPM value at this point is equal to the number of DPM in the sample.

Over a dozen radionuclides (^{14}C, ^{32}P, ^{36}Cl, ^{46}Sc, ^{59}Fe, ^{60}Co, ^{63}Ni, ^{86}Rb, ^{90}Sr-Y, ^{131}I, ^{134}Cs, ^{147}Pm) have been assayed[6,7] using this technique with excellent statistical results (SD = 1.4%) between the DPM calculated by the efficiency tracing and the absolute activity (DPM) of the sample. Specific examples of the DPM results achieved using the efficiency tracing technique on the Packard LSA Model 2500TR for radionuclides ^{36}Cl, ^{59}Fe, ^{63}Ni, and ^{14}C are shown in Table 1.

Specific examples of the actual efficiency tracing plots of the data show excellent statistical results; % recovery, equal 100% ± 2% for ^{36}Cl, ^{59}Fe, and ^{63}Ni are shown in Figures 2 to 4.

As can be seen from the previous table and results, excellent DPM values can be obtained for radionuclides ranging from ^{63}Ni to ^{32}P over the energy range of 60 to 1700 keV.

In order to further assess the reliability and accuracy for determining DPM values, a series of samples with various types of counting vials, scintillation cocktails, sample volume, microvolume samples, and color or chemical quenching degree were quantitated using the efficiency tracing technique (Table 2).

The mean % recovery is 99.91% with a very small standard deviation of only 1.15. These results conclusively demonstrate that using the efficiency tracing technique produces DPM results which are independent of:

Table 1. Efficiency Tracing Results of Various Nuclides at Various Quench Levels

Radionuclide	SIS	tSIE	DPM (ET)	DPM (Actual)	% Rec.
^{36}Cl	973	919	117,381	118,414	99.1
^{36}Cl	580	537	117,607	118,414	99.3
^{36}Cl	188	174	116,606	118,414	98.5
^{36}Cl	117	109	117,154	118,414	99.0
^{59}Fe	241	460	2,231,719	2,310,000	97.0
^{59}Fe	223	428	2,204,944	2,310,000	95.4
^{59}Fe	151	315	2,283,158	2,310,000	98.8
^{59}Fe	115	305	2,306,399	2,310,000	99.9
^{63}Ni	30	583	200,826	200,000	100.4
^{63}Ni	29	535	197,909	200,000	99.0
^{63}Ni	16	235	195,066	200,000	97.0
^{63}Ni	15	206	185,546	200,000	93.0
^{14}C	173	1,000	111,280	111,700	99.6
^{14}C	86	505	112,603	111,700	100.8
^{14}C	39	209	116,373	111,700	104.1
^{14}C	24	120	125,131	111,700	112.0

1. cocktail density variation
2. different vial sizes
3. varying sample volume
4. color quenching
5. chemical quenching
6. vial composition

The independence of the DPM values on the chemical quench level of the sample can be further demonstrated using ^{63}Ni at various quench levels (629,

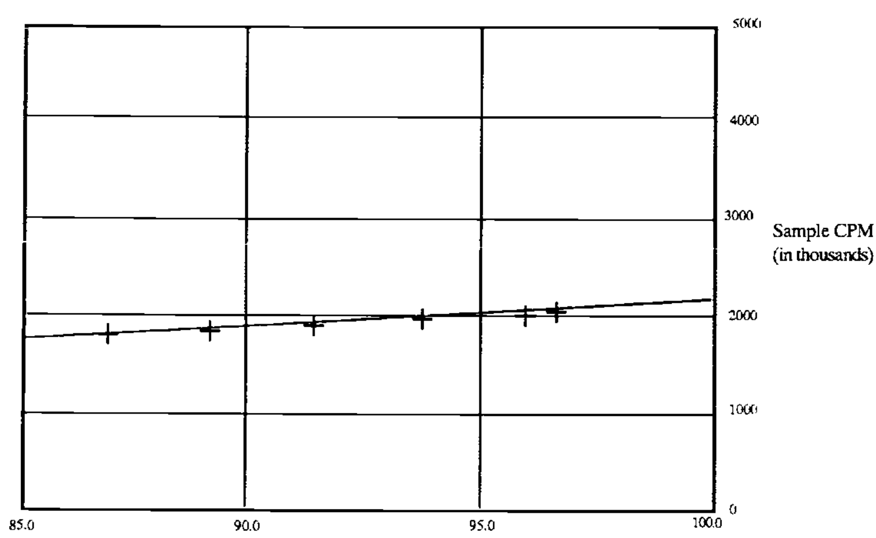


Figure 2. Efficiency Tracing Curve for ^{59}Fe at tSIE = 437 Quench Level.

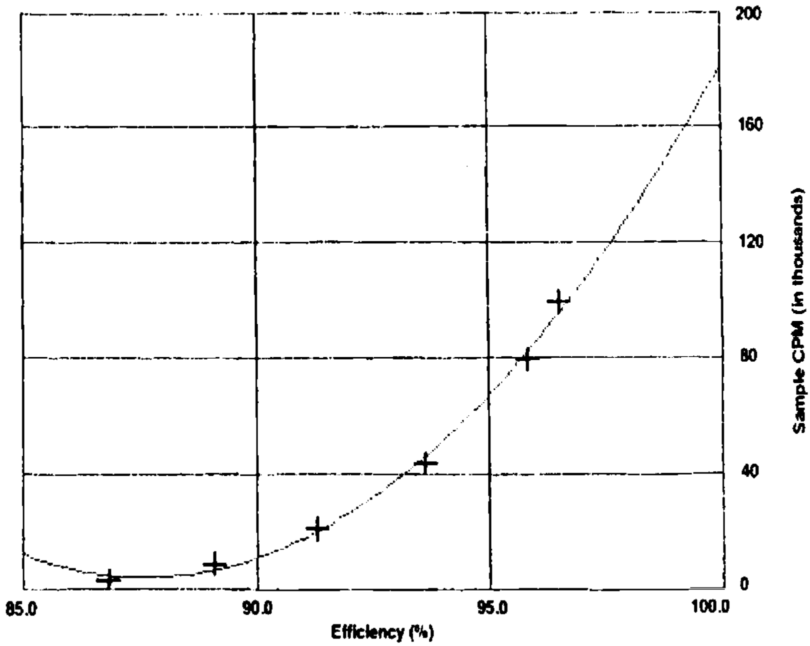


Figure 3. Efficiency tracing of ^{63}Ni at tSIE = 204 (quenched).

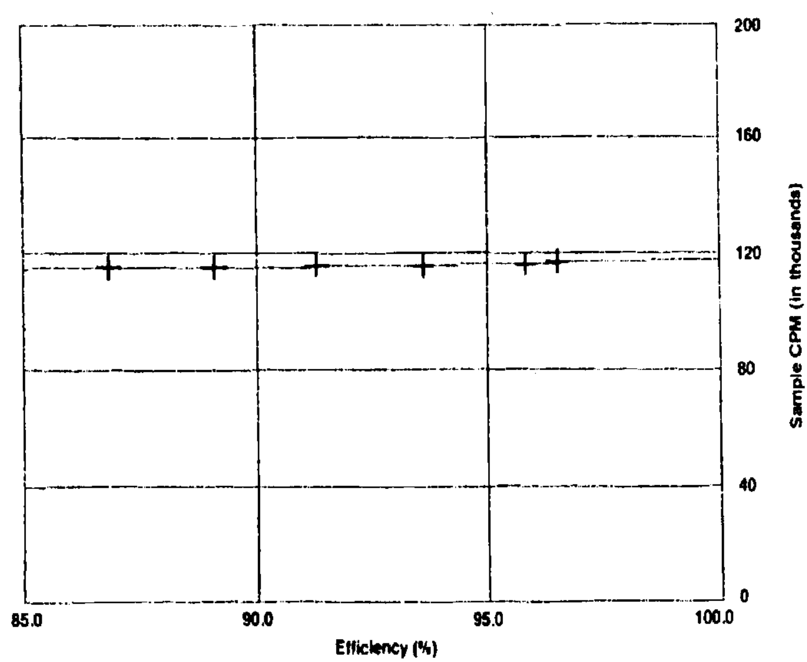


Figure 4. Efficiency tracing curve for ^{36}Cl at tSIE = 953 (unquenched).

Table 2. Efficiency Tracing Results for ^{14}C Using Various Conditions

Sample	tSIE	DPM (ET)	DPM (Actual)	% Recovery
1. Std. vial Color quenched	625	143,457	144,000	99.6
2. Mini vial Color quenched	644	144,020	144,000	100.0
3. Std. vial (2.0 mL)	576	128,536	129,500	99.3
4. Std. vial (8.0 mL)	967	127,808	129.500	98.7
5. Std. vial (15.0 mL)	959	128,432	129,500	99.2
6. Mini vial (0.5 mL)	608	128,608	129,500	99.3
7. Mini vial (2.0 mL)	776	128,099	129,500	98.9
8. Mini vial (5.0 mL)	862	128,768	129,500	99.4
9. Std. vial Chemical quench	690	142,734	144,000	99.1
10. Mini vial Chemical quench	653	143,072	144,000	99.4
11. Std. vial Bray's cocktail	207	119,173	120,800	98.7
12. Micro volume 400 μL	739	10,992	10,760	102.1
13. Micro volume 200 μL	693	11,209	11,080	101.1
14. Micro volume 100 μL	643	11,043	10,860	101.7
15. Micro volume 50 μL	590	10,747	10,650	100.9
16. Micro volume 25 μL	548	10,715	10,460	102.4
17. Std. Vial - No Chemical quench	989	110,129	111,700	98.6
18. Std. vial Chemical quench	785	111,312	111,700	99.7
19. Std. vial Chemical quench	281	112,279	111,700	100.5
20. Mini Vial - No Chemical quench	862	132,904	133,500	99.6

501, 381, 221) plotted in Figure 5. The plot indicates that each efficiency tracing plot (different tSIE values) has a different slope, but all intersect the 100% activity line at approximately 197,000 DPM. This clearly indicates that the final DPM results are independent of the color or chemical quench level of the sample.

In addition, if efficiency tracing is used for low level DPM samples, results similar to those shown in Figure 6 can be expected. A set of samples containing 100 DPM was analyzed by this technique, and found efficiency tracing 98.813 DPM with a coefficient of variation of 0.714. Similar results were obtained from Dr. Ishikawa in Japan, DPM as low as 22.53 ± 1.11 for a set of low level samples. These results indicate that the efficiency tracing method can be used accurately to determine DPM in low level samples.

This technique can be used for most pure beta and beta-gamma emitters. The one exception is tritium, whose efficiency tracing plot can result in large errors (up to 25%) for highly quenched samples. In addition, the efficiency tracing technique is not applicable to radionuclides which decay by isometric transitions and electron capture (EC). The reason for this is that radionuclides that decay by electron capture are followed by the emission of an X-ray or

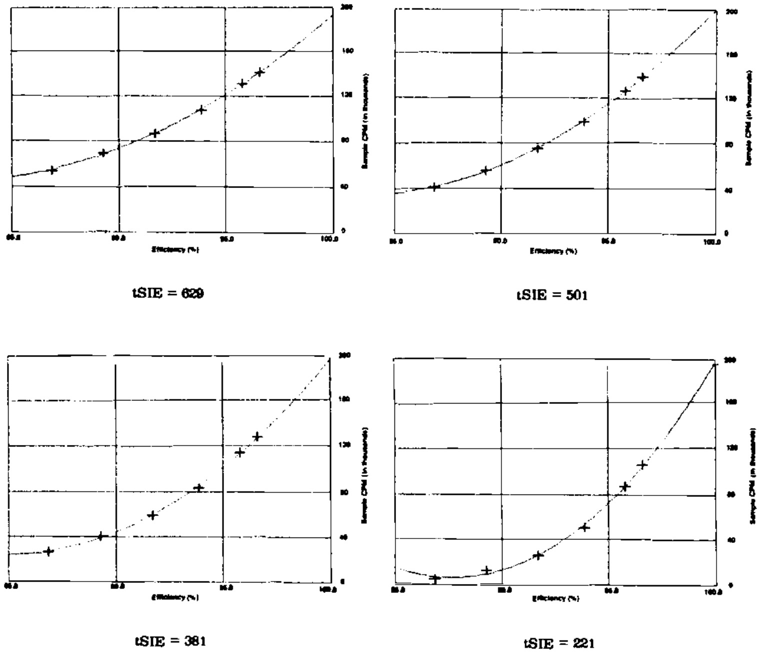
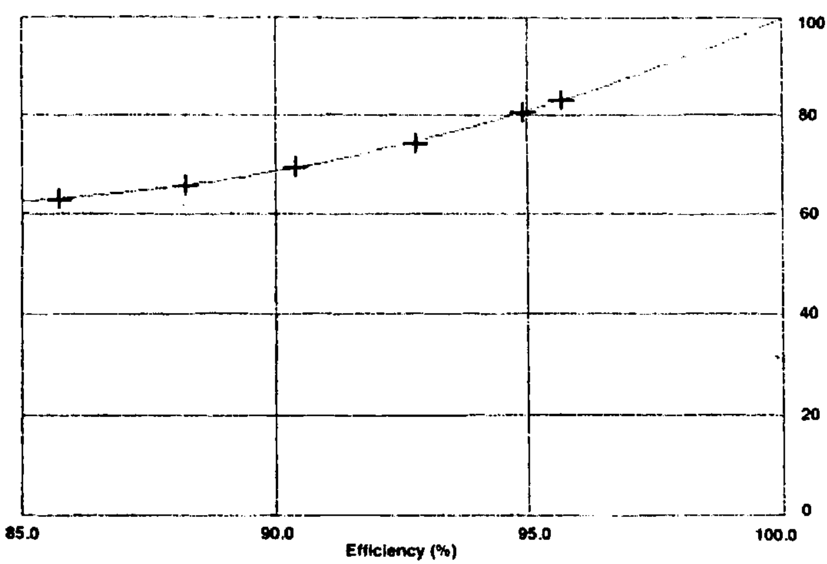


Figure 5. Efficiency tracing curves for ^{63}Ni at four quench levels.



Mean (4) = 98.813 DPM S.D. = 0.705 C.V. = 0.714

Figure 6. Efficiency tracing of ^{14}C sample containing 100 DPM and statistics for sample counted four times.

Auger electron. This makes it difficult to find the true absolute activity of these radionuclides using this technique.[6]

In summary, the efficiency tracing technique is an accurate and reliable method of calculating DPM (absolute activity) for most pure beta and pure beta-gamma emitters (minimum energy = 60 keV). This method only requires counting an unquenched ^{14}C standard of known activity and does not require the preparation of a quenched curve set for each individual radionuclide analyzed. The final DPM for each sample is calculated from the respective spectrum compared to the standard unquenched sample using six separate spectral regions. The data (DPM) are calculated using a special curve fitting routine and extrapolation technique to determine the absolute activity (DPM) of the sample. This efficiency tracing method is available on all Packard liquid scintillation analyzers.

REFERENCES

1. Grau Malonda, A., E. Garcıa-Toraño, and J.M. Los Arcos. *Int. J. Appl. Radiat. Isot.* 36(2).157 (1985).
2. Grau Malonda, A. and E. Garcıa-Toraño. *Int J. Appl. Radiat. Isot.* 33:249 (1982).
3. Grau Malonda, A. *Int. J. Appl. Radiat. Isot.* 33:371 (1982).
4. Coursey, B.M., J.A.B. Gibson, M.W. Gertzmann, and J.C. Leak. "Standardization of Technecium-99 by Liquid Scintillation Counting," *Int. J. Appl. Radiat. Isot.* 35(12):1103 (1984).
5. Yura, O. *Radioisotopes* 20:493 (1971).
6. Ishikawa, H.M. Takiue, and T. Aburai. *Int. J. Appl. Radiat. Isot.*35(6):463 (1984).
7. Ishikawa, H. and M. Takiue. *Nuc. Instrument Methods* 112:437 (1983).