CHAPTER 21

A New Quench Curve Fitting Procedure: Fine Tuning of a Spectrum Library

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ABSTRACT

In liquid scintillation counting the counting efficiency and the spectrum of the sample depend on the quench level; quench calibration is needed to establish the true activity of the sample. The quench curve relates the counting efficiency to an appropriate quench indicating parameter. The quench curve is formed by measuring a number of reference samples with known activity and varying quench levels. Typically six to ten reference samples are required in quench calibration.

This presentation shows that the number of reference samples can be reduced by using model based quench curve fitting. The model uses a large set of reference samples, which form the spectrum library of the isotope. The fine tuning of the spectrum library is the model adjustment procedure in which the spectrum library data is modified to fit to the data of the new calibration.

The principles of the fine tuning are illustrated and a theoretical example is calculated. Measurements made by a liquid scintillation counter that uses the described principle, the Wallac 1410, are presented and a comparison with a conventional counter is made.

INTRODUCTION

The quench level of liquid scintillation counting samples is often variable. Quenching decreases the number of photons detected after one radioactive decay. In instruments the variation of quench level is seen as variations of the counting efficiency and shifts of the pulse height energy distribution from sample to sample of the same isotope. The reduction in the amount of photons shifts the pulse height energy distribution or the spectrum to the smaller pulses and decreases the counting efficiency. Figure 1 shows three ^{14}C spectra of different quench levels.

To relate the disintegration rate of a sample to the count rate it is necessary to determine the quench level of the sample and correlate it to the counting efficiency. There are different methods to estimate the quench level of a sample, but most of them rely on the effect of quenching on the position of the

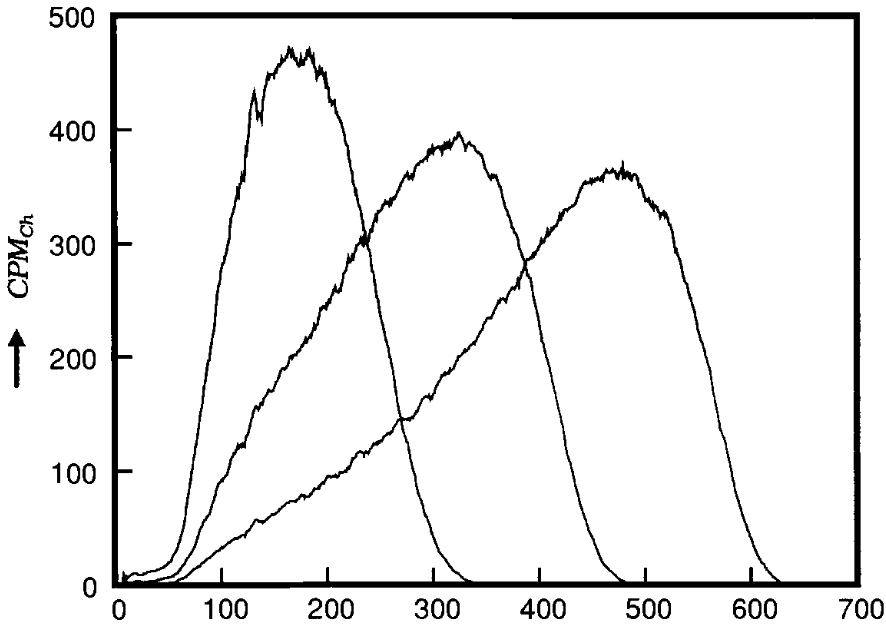

Figure 1. Three ^{14}C spectra of different quench levels.

spectrum on the amplitude scale. A common method uses an external gamma ray source or an external standard and measures the shift in pulse amplitudes of the spectrum resulting from Comptom electrons scattered by this external standard. The end point of an external standard spectrum (Sqp(E)) can be used as a quench indicating parameter.

The relationship between a quench indicating parameter and the counting efficiency of a radionuclide is established by measuring reference or standard samples of known activity and expressing the counting efficiency of these samples as a function of the quench indicating parameter. The whole procedure is called quench calibration and the resulting function, which correlates counting efficiency and the quench indicating parameter, is called a quench curve.

Figure 2 shows tritium quench curves for different scintillation solutions. Because typical quench curves are not linear but curved, normally six to ten reference samples have to be prepared and measured to make a quench calibration that covers the desired quench level range. Figure 2 shows that even if the quench curves differ from each other, the basic shape of the curves is the same. This is especially true for one radionuclide, but the same shape can be used as a good approximation for isotopes with energy of the same order of magnitude.

Traditionally quench curves have been formed by fitting a polynomial or a spline function to the quench calibration data or by making a linear interpola-

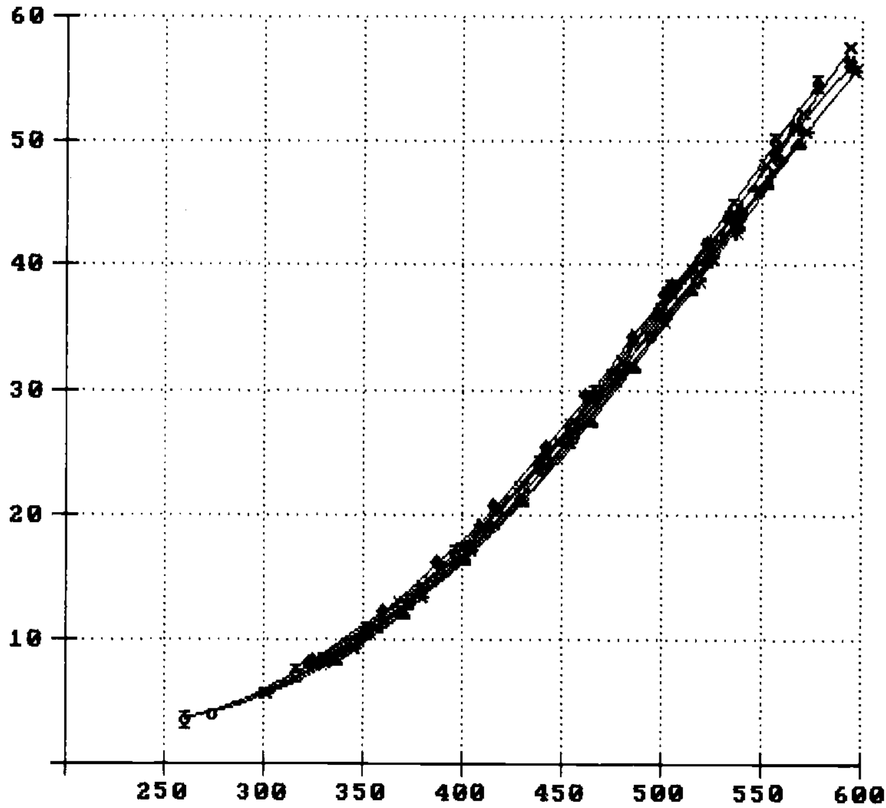

Figure 2. Tritium efficiency quench curves for different commercial scintillation solutions[1].

tion between the data points. In this presentation a quench curve producing method, which uses the known form of the quench curve is introduced; the method is called model based quench curve fitting.

This introduction is concerned with the efficiency quench curve, but the spectrum can be treated in an analogous way. The spectrum can be expressed as parametric form, and the parameter values can be expressed as a function of the quench indicating parameter. Another possible way to treat spectrum data is to normalize the spectrum and follow the variation of the spectrum proportion in each channel.

The preservation of the quench curve form can be realized in many ways. It is possible to develop a theoretical quench curve model with a few parameters and find the solution of these parameters for the particular quench data. This kind of procedure can preserve the right form of the curve if the number of free parameters is low enough. Another more straightforward possibility is to use an experimental model. The model adjustment should fulfill two demands: flexibility and stiffness. The procedure should be flexible enough to follow the natural variation and stiff enough to preserve the right shape of the curve. One possible solution is to make a transformation of the variables, make the fit on them, and then return to the real values by inverse transformation.

THEORETICAL EXAMPLE

As a theoretical example, we may consider a situation where the true efficiency quench curve is assumed to be a second degree polynomial. The quench curve model is then also a second degree polynomial as shown in Figure 3. Figure 3 also shows the measured counting efficiency and sample quench index value pairs for two reference samples, which are assumed to have 4 and 5% higher counting efficiency values than predicted from the model. Figure 4 shows the same model curve and reference data points after a transformation on the efficiency scale. Here the curve fitting is not made on efficiency values but on the relative deviations of efficiency from the model. A linear interpolation is used as a fitting method. Figure 5 shows the corresponding fit in untransformed coordinates; it also shows a normal linear interpolation without transformation. The comparison between model based fitting and normal fitting is shown in Figure 6, where the relative dpm errors of both systems are given as a function of counting efficiency. The error in normal fitting is three times greater than the one using model based estimation.

EXPERIMENTS

The Wallac 1410 counter uses a model based fitting principle for quench calibration. As a model, the 1410 has built in spectrum libraries for the most common isotopes. The spectrum library includes counting efficiencies, whole sample spectra and external standard spectra, external standard based total quench indication parameters, and external standard based color quench indication parameters of reference samples. The data is retrieved from the spectrum library using both quench parameter as keys. The quench calibration in the 1410 effects both counting efficiency and spectra, and it is called fine tuning of the spectrum library.

A tritium quench series of nine samples was made using Hisafe liquid scintillation cocktail and nitro methane as quenching agent. Using two samples of the series as references, a quench calibration has been made in the 1410 and a RackBeta 1219. In all measurements 0.5% one sigma counting statistics were used. RackBeta 1219 uses linear interpolation, as only two reference samples are used. Figure 7 shows the efficiency quench curve made in the 1410. Figure 8 shows the relative error of activities in the quench series samples, which have been measured as unknown samples. The systematic errors are one order of magnitude lower in System 1410 when using the fine tuning principle.

To illustrate the robustness of the 1410 fine tuning principle an experiment was conducted, where the spectrum library of another isotope is used as a

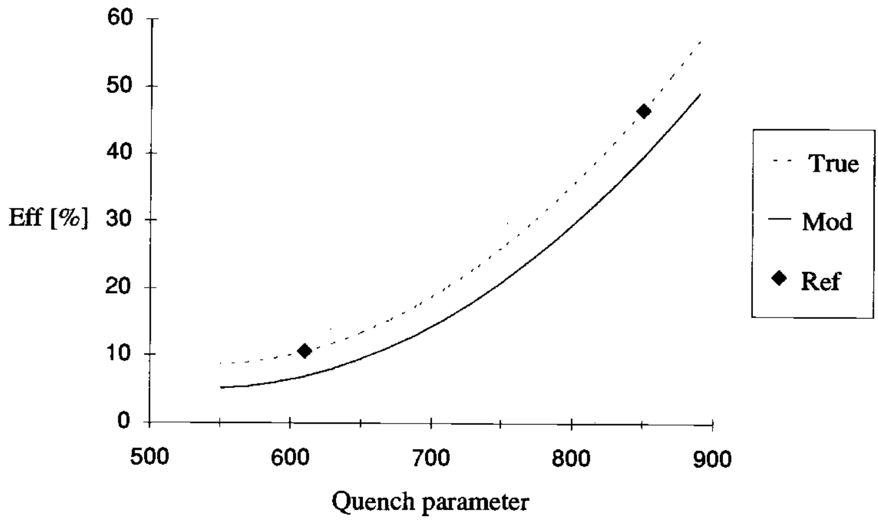

Figure 3. Mod is a tritium efficiency quench curve model, Ref is used for measured quench parameter—counting efficiency values of reference samples—and True is the real efficiency quench curve corresponding to the reference samples.

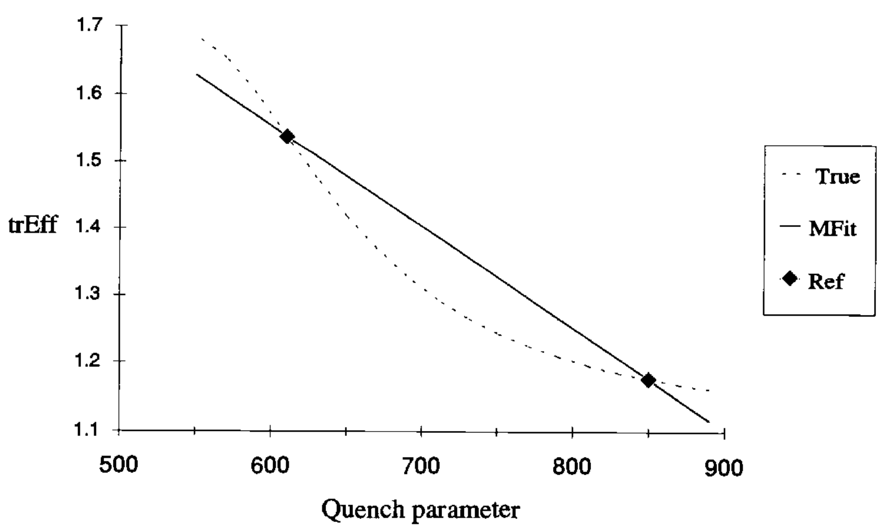

Figure 4. Transformed efficiency quench curves. The values of trEff-axes are calculated by dividing the efficiency with the efficiency of the model curve of the corresponding quench parameter value. MFit is the model based quench curve in this coordinate system. This quench curve was made using a linear interpolation of the efficiency quench curve in this coordinate system. Symbols True and Ref are defined in Figure 3.

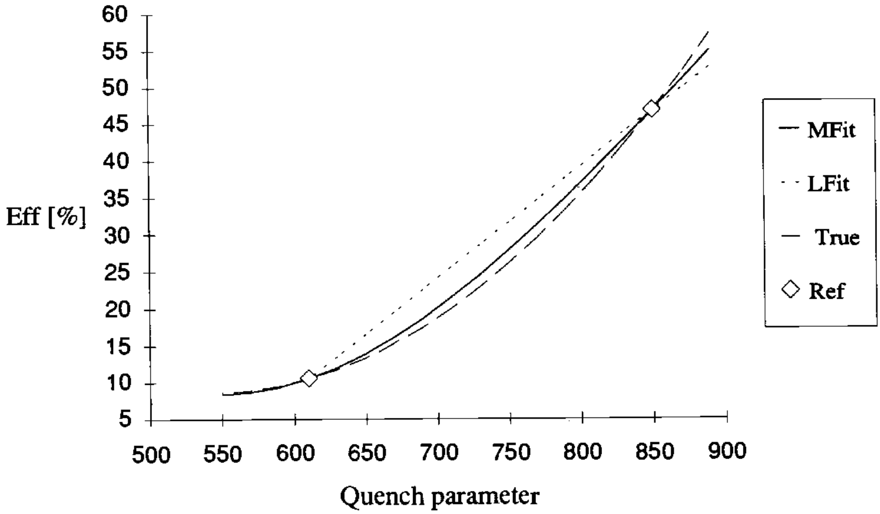

Figure 5. LFit is a quench curve made using a linear interpolation without the model. Symbols MFit, True and Ref are defined in Figures 3 and 4.

model in quench calibration. A quench series of nine ^{14}C samples was made using Hisafe cocktail and nitro methane as a quenching agent. Quench was calibrated in the 1410 using three ^{14}C samples, having the ^{45}Ca spectrum library as a model. Figure 9 shows the efficiency quench curve. All nine samples were measured as unknowns, and the dpm errors are shown as a function of sample

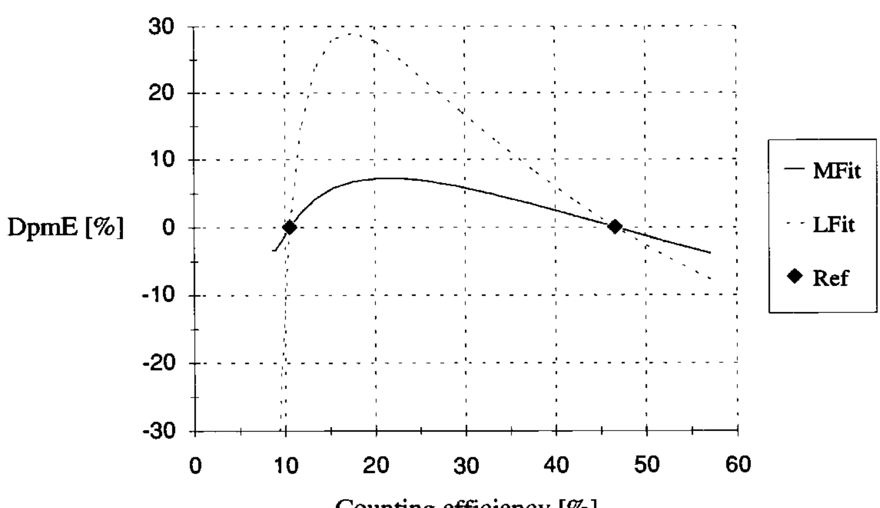

Figure 6. Relative DPM errors, DpmE as a function of counting efficiency for the model based quench curve and for the quench curve not based on a model.

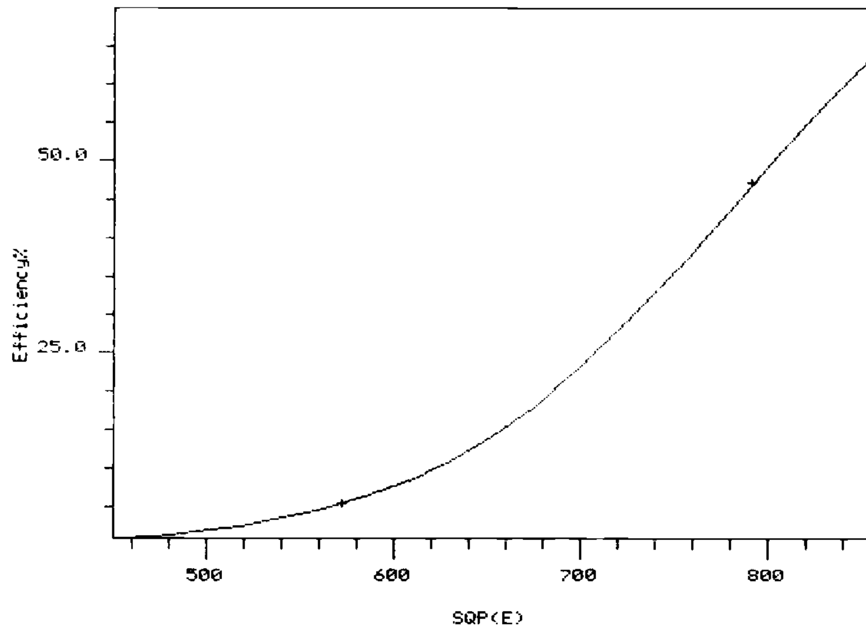

Figure 7. Tritium efficiency quench curve of the 1410 using two reference samples.

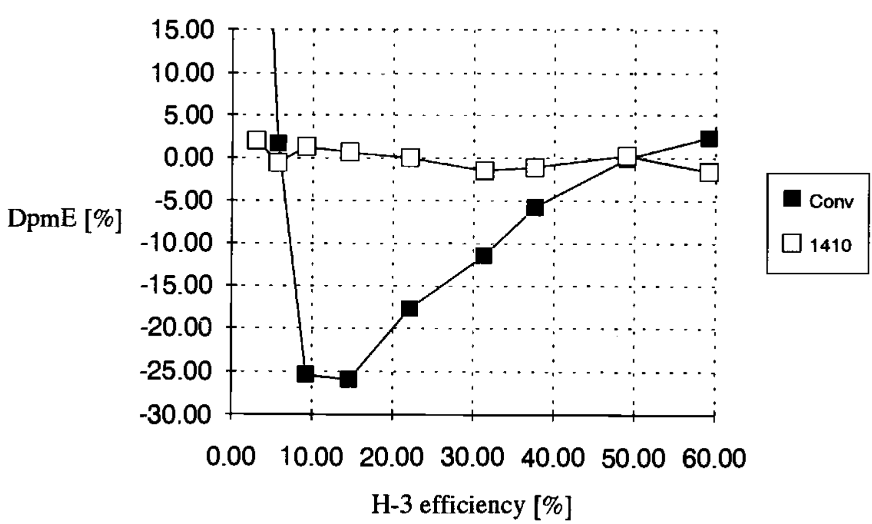

Figure 8. DPM error, DpmE for ^{3}H samples as a function of the counting efficiency of the samples when two reference samples were used in quench calibration. The counting efficiencies of the reference samples were 6.7% and 47.3%. *1410* is the 1410 and Conv is the 1219.

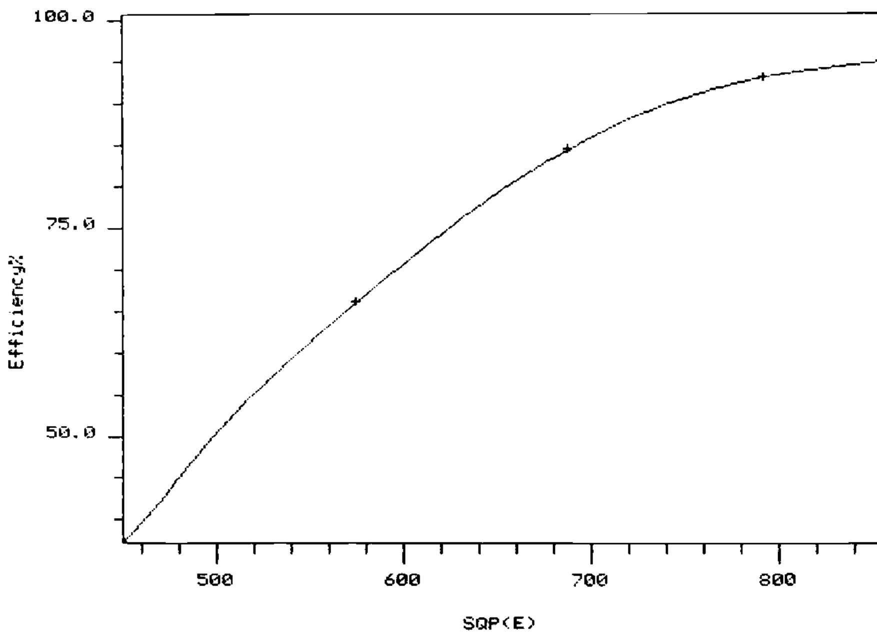

Figure 9. Carbon efficiency quench curve of the 1410 using three reference samples and ^{45}Ca spectrum library.

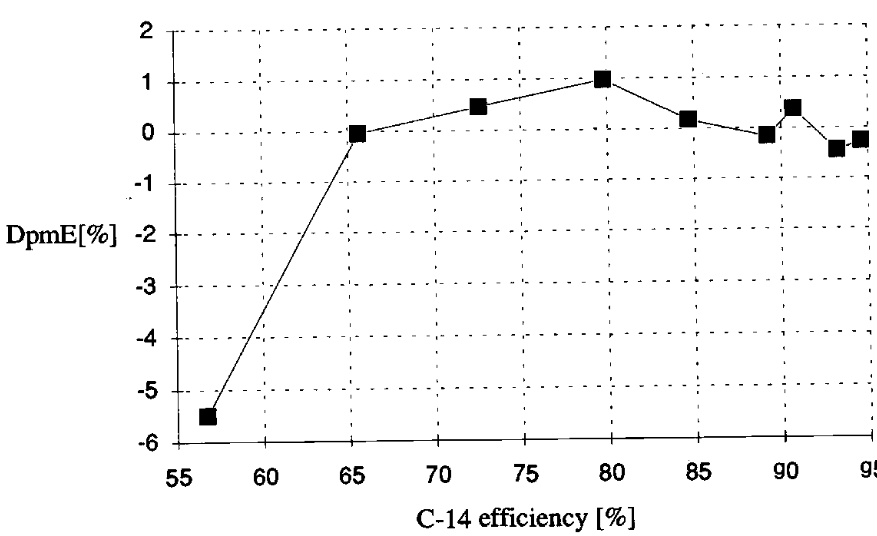

Figure 10. DPM error, DpmE for ^{14}C samples as a function of the counting efficiency of the samples when three reference samples and ^{45}Ca library were used in quench calibration. The counting efficiencies of the reference samples were 93%, 84%, and 65%.

quench level in Figure 10. In every case the interpolation error is under 2%, hence a model could also be selected from a spectrum library of some other isotope. Of course, the library of a radionuclide with a similar energy would be preferred.

CONCLUSIONS

Using the model based quench calibration or spectrum library fine tuning considerably increases the accuracy of the results when only a few reference samples are used.

REFERENCES

1. Rundt, K. "On the Determination and Compensation of Quench in Liquid Scintillation Counting," PhD Thesis, Åbo Akademi, Turku, Finland (1989).