"Efficiencies" of phosphor screens used in Thick Source Alpha particle Counting

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(Received 14 July 1995; in final form 12 September 1995)

Introduction

Thick Source Alpha Counting (TSAC) has formed the basis for measuring background environmental concentrations of thorium and uranium for many years - for example, Aitken (1985), Huntley and Wintle (1978, 1981), Huntley *et al.* (1986), Jensen and Prescott (1983). The procedure involves placing the sample on a scintillator screen (usually zinc sulphide) viewed by a photomultiplier, with counters recording total alpha counts, 'slow' (within 400 ms) pair counts and sometimes 'fast' pair counts (within 20 ms). The counting system may be made up of discrete modules (eg: NIM), or purchased as dedicated alpha counting equipment (as in the case of the *Daybreak* model 582 alpha counter and printer/controller unit 584); both types are used in this department. Both counters have a screen area of 13.8 cm^2. Other systems, eg: ELSEC, are available but are not in use in this group.

One can measure a "screen efficiency" by counting a standard of known concentration and comparing the count rate observed with that calculated on the basis of the concentration of the standard. In what follows, the ratio of measured to calculated count rates will be referred to as the "efficiency".

Like Bowman (1976), Jensen (1982) and Jensen & Prescott (1983) found that the scintillator screens were less than 100% efficient and this view has been supported by Akber (1986, personal communication). Consequently, the Adelaide group has always used a "measured" efficiency less than 100% for calculating thorium and uranium contents. Huntley and Wintle (1981), Huntley *et al.* (1986), Aitken (1985) and the *Daybreak* manual seem to suggest that 100% efficiency is assumed. Examination of published counting procedures suggested that other workers assume 100% efficiency, and this procedural discrepancy prompted the investigation reported herein.

Method

Finely divided and diluted uranium or thorium ore in radioactive equilibrium with its daughters is placed in a cell on a scintillator screen. A "calibration curve" is then obtained by plotting count rate vs discriminator setting which is then extrapolated to zero discriminator setting. A linear extrapolation is commonly used but we find that a quadratic better represents the data set, as measured by the correlation coefficient.

Plotting a calibration curve for a known standard allows the determination of the appropriate discriminator setting for that screen. As is conventional, the discriminator is set to a level such that the count rate is 82% (uranium standard) or 85% (thorium standard) of the extrapolated intercept value. An example of such a calibration curve is shown in figure 1. The measured count rate at this point is compared with that calculated from the known uranium and thorium content of the standard and the conversion factors of Aitken (op. cit) or Huntley *et al.* (1986). This ratio is what is defined in the following tables under the heading "efficiency".

A selection of screens from different batches purchased from Wm. B. Johnson[1] since 1979 was tested against a variety of different standards. Within this text, "screen 5" refers to screens from batch 5. Extensive tests were carried out on the most recent screen batch purchased in 1993 ("batch 11"), and one purchased in 1980 ("batch 5"). The certified reference standards used were all from the US Department of Energy, New Brunswick Laboratory, Argonne IL 60439, USA.

[1] Wm. B. Johnson and Associates, P.O. Box 472, Ronceverte, WV 24970, USA

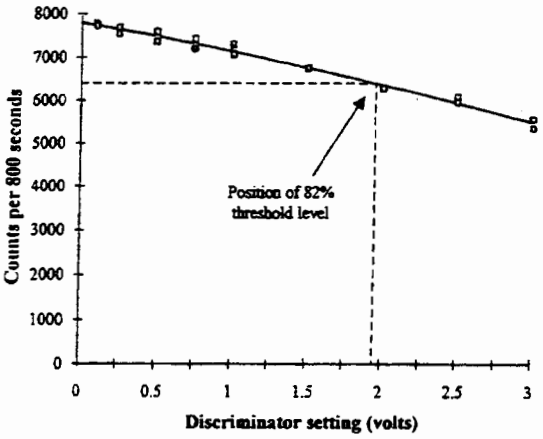

Figure 1

Calibration curve for screen 5 obtained on the Daybreak *counter using NBL 42.4 standard. The counting errors lie within the boxes.*

Direct measurement of the efficiency

The efficiencies obtained are shown in table 1 with the standard used to obtain them and the nature of the measurement. For those labelled "fitted calib curve", the fitted calibration curve was used to obtain the count rate measured at the appropriate discriminator setting. For those labelled "counted data", it was assumed that the fitted calibration curve measured on another screen from the same batch using the same standard was applicable, and the discriminator was set appropriately; at least 10^4 counts would be accumulated at this setting. In both cases, the resulting measured count rate and that expected from the given standard were then used to calculate the efficiency. The conversion factors of Huntley *et al.* (1986) were used.

As NBL 109, NBL 105 and NBL 104 have very low uranium and thorium concentrations, a complete calibration curve was impractical for these standards. To obtain count rate data for these, the discriminator was set as for NBL 106, which was observed to be of the same colour (and thus have the same reflectivity) as the others.

For each measurement a new disc from the appropriate batch was used and sufficient time was allowed for radon emission to reach equilibrium in the sealed cell, although there was little evidence that this made any difference. Measurements were made on both *Daybreak* and University of Adelaide NIM systems. The quoted errors are statistical counting errors only. These results give weighted mean efficiencies of 89.3 ± 2.8% for screen 5 and 86.5 ± 1.6% for screen 11 (± sample standard deviations).

The NBL 101 result for screen 5 is well outside the interval defined by Chauvenet's Criterion for rejection of outliers (Taylor 1982). We were unable to explain this result. Omitting this result gives a weighted mean efficiency for screen 5 of 88.9 ± 1.8%.

The above results clearly show that the properties of screens can vary significantly from batch to batch. Moreover, by studying the curves produced by standards of different colours, it was found that the discriminator setting for 82% uranium / 85% thorium threshold could vary by up to 25% between grey and white standards. This confirms results obtained by Huntley (1978) and Jensen (1982) concerning sample colour and reflectivity.

It is also evident that it is not possible to reproduce efficiency measurements to better than about ± 3%. This could be due to small variations within each batch which may be very difficult to control on a mass-produced scale, or to differences in filling the cell.

Indirect measurement of the absolute efficiency

Differences among different screens were also assessed by a more rapid but less rigorous method. This uses a permanent standard ("Hanstand"), comprising an approximately 2% natural uranium source encapsulated in a silicone plastic. The source rests on a thin ring just above the scintillator screen to avoid contamination. This provides a convenient way of transferring the standard between screens and for repeating measurements on the same screen. The standard allows easy determination of the relative efficiencies of screens with respect to a chosen reference; it cannot be used to find the efficiency directly since its concentration is not accurately known, and the standard is not in direct contact with the screen. However, the absolute efficiencies can be calculated by relating the relative measurements to the measured absolute efficiency of a reference screen as detailed previously. This indirect method gave similar results to those of the direct method.

Discussion

It may be questioned whether the "efficiency" described here is an "actual" or an "apparent" efficiency and whether it is really associated with the zinc sulphide screens or with

some other feature of the method. There seems to be no doubt that it is at least partly screen-related because application of the same procedures to different batches leads to different results. That screens can vary considerably is clear from figure 2 which shows that screens can differ for two different reasons, as evidenced by the differences in both slope and intercept.

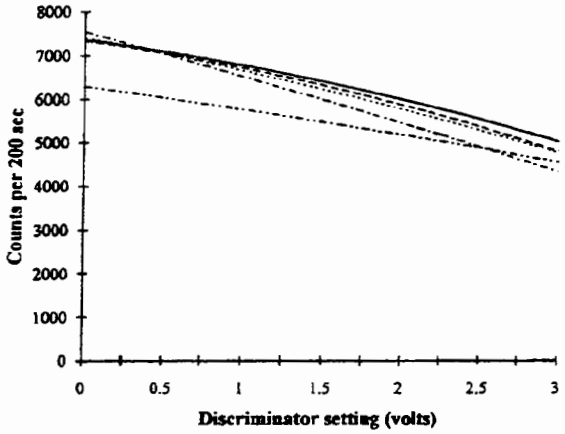

Figure 2

Calibration curves for selected screens on the Daybreak counter using the Hanstand standard. Experimental points are omitted for clarity. They are better than ±2% throughout.

-·-·- *Screen 4 with Hanstand,*
– – – – *Screen 5 (early sheet) with Hanstand,*
····· *Screen 5 (last sheet) with Hanstand,*
——— *Screen 6 with Hanstand,*
-·-·-· *Screen 11 with Hanstand.*

Table 2 shows results of measurements of efficiency carried out over the years on ten (out of our twelve) batches. Differences can be observed, although in recent years these have been small, with the exception of batch 11 which exhibits a markedly different slope. D.J. Huntley kindly made independent measurements on screens 6 and 11. In relative terms we get identical results, although we differ on the question of the derived efficiency.

Another possibility is that the factors given by Aitken (1985) and independently by Huntley *et al.* (1986) for converting alpha counts and pairs counts to concentrations are in error. Huntley's concentration-to-alpha-count-rate factor for uranium is 3% larger than Aitken's, his thorium-to-alpha factor is 2.6% larger, and for thorium-to-pairs his factor is 3.0% larger. These differences are consistent with Huntley's use of more recent alpha particle data. For consistency, we have used Huntley *et al.* (1986) factors throughout, including measurements made on the Daybreak system. These are not the same as those in the built-in Daybreak software, which appear to be the same as those of Aitken (1985).

Huntley *et al.* (1986) give count rate data for several standards: NBL-106, NBL-108, BL-2 and BL-3, and two measurements of DL-la, all incorporated into a lithium borate glass. For the first, the observed count rate exceeded the expected rate by 4% while for the others the observed count rate was about 3% lower than expected. Huntley *et al.* quite reasonably conclude that the observed and expected count rates are not significantly different. Huntley and Wintle (1981) found that for four of their standards, DL-1, BL-3, NBL-106 and NBL 108, the observed counting rate fell below the expected counting rate whereas for two samples, DL-2 and BL-2 the discrepancy was in the other sense. Those measurements were with finely powdered samples (as in the present work). The Huntley and Wintle (1981) conversion factors were superseded by those of Huntley *et al.* (1986). Correcting for the difference leaves the high count high and the low counts low. In the latter case the deficiency is smaller than the one we observe. Unfortunately, the only one of these samples common with ours is NBL-106, for which we obtain a count rate that is low. Huntley notes that thorium pairs count rates come out consistently low and we return to this point below.

Jensen (1982) found the same screen efficiencies for both finely powdered samples and those incorporated into lithium borate glass, provided the calibration curves were measured independently for each method of sample preparation.

A third possibility is that the counting apparatus does not measure what we think it measures. Perhaps the best answer to this is that we get the same value for the efficiency for the same batch on three different counters. Over the years we believe we have come to understand our apparatus well. However, at the urging of Huntley we rechecked our discriminator thresholds with a precision pulse generator. The zero offsets, 0.2% of full scale, were well within the manufacturer's specification of <0.5% of full scale, which implies a systematic error of the order of 0.2% (depending on the slope of the discriminator curve) in threshold estimation and hence in efficiency. No counts are being lost through exceeding discriminator upper levels. At the suggestion of Victor Bortolot we remeasured the screen area and the point to point uniformity of one of our photomultiplier cathodes. These are not the source of the discrepancy.

***Table 1** - Directly determined efficiencies*

Screen batch	Standard used	Source	Count rate (cm^{-2} ks^{-1})	Efficiency
5	NBL 42.4	fitted calib curve	594.6	89.0 ± 0.6%
	NBL 42.4	fitted calib curve	579.1	86.7 ± 1.0%
	NBL 42.4	counted data	601.4	90.0 ± 1.1%
	NBL 42.4	counted data	610.8	91.3 ± 1.2%
	NBL 42.4	fitted calib curve	586.6	87.8 ± 1.5%
	NBL 42.4	counted data	606.9	90.8 ± 1.2%
	NBL 42.4	fitted calib curve	588.0	88.0 ± 1.5%
	NBL 105	counted data	1.175	89.7 ± 2.1%
	NBL 105	counted data	1.213	92.5 ± 2.1%
	NBL 101	fitted calib curve	1298	101.0 ± 2.0%
	NBL 106	fitted calib curve	357.2	85.4 ± 1.7%
	NBL 109	counted data	3.730	85.6 ± 2.6%
	NBL 109	counted pairs data	0.141	87.6 ± 2.5%
11	NBL 42.4	fitted calib curve	569.5	85.2 ± 0.7%
	NBL 42.4	counted data	557.9	83.5 ± 2.0%
	NBL 42.4	fitted calib curve	565.6	84.7 ± 1.4%
	NBL 42.4	counted data	553.8	82.9 ± 2.0%
	NBL 42.4	fitted calib curve	587.1	87.9 ± 1.0%
	NBL 42.4	fitted calib curve	603.3	90.3 ± 1.0%
	NBL 42.4	fitted calib curve	590.0	88.4 ± 1.1%
	NBL 42.4	fitted calib curve	586.7	85.9 ± 0.8%
	NBL 42.4	fitted calib curve	578.5	86.6 ± 0.5%
	NBL 104	counted data	11.37	85.9 ± 1.5%
	NBL 104	counted data	11.37	85.9 ± 1.7%
	NBL 104	counted data	11.22	84.8 ± 2.9%
	NBL 104	counted data	11.22	84.8 ± 3.0%

Concentration of standards used:
(all standards in silica unless otherwise stated)

NBL 42.4: 0.52 ± 0.006% U in dunite
NBL 101: 1.00 ± 0.01% U
NBL 104: 0.0103 ± 0.0004% U
NBL 105: 0.00102 ± 0.00001% U
NBL 106: 1.00 ± 0.01% Th
0.035% U (nominal)
NBL 109: 0.0104 ± 0.0003% Th
0.00037% U (nominal)

***Table 2** - Efficiency factors of screens as used by the Adelaide group previously*

Batch	Date Acquired	Efficiency factor
1	Sept 1977	95 ± 2%
2	Nov 1977	81 ± 2%
3	Oct 1978	79 ± 2%
4	June 1979	79 ± 2%
5	Dec 1980	89 ± 2%
6	May 1982	92 ± 2%
7	unknown	90 ± 2%
8	Apr 1986	92 ± 2%
9	Aug 1987	89 ± 2%
10	Aug 1988	89 ± 2%
11	1993	85.9 ± 2%

The usual prescriptions for calibration suggest measuring discriminator curves, with a linear extrapolation to zero to find the intercept. Over many observations we have found that a quadratic fit is better and we always use it, as we have here. However, for the present paper we recalculated four of our calibration curves using a linear extrapolation, covering discriminator settings to about twice the 82% threshold. The extrapolated value would be increased by a percent or two but this is not enough to account for the discrepancy, although it reduces it. The effect of colour (or more strictly reflectivity) has been extensively discussed by Jensen (1982), Huntley (1978) and by Huntley and Wintle (1983). In the present case all calibrations were carried out directly or, in a few cases where counting rate was low, on screens of similar colour.

Most TSAC systems provide a pairs counting facility (see, e.g. Aitken 1985, Huntley and Wintle 1981):. The measurement of "slow" pairs, (i.e. coincidences between succesive alpha particles from ^{220}Rn and its daughter ^{216}Po in the thorium chain), allows an estimate of the concentration of thorium. Although the small number of coincidences, relative to the total alpha count, means that the statistical accuracy in the estimation of thorium is small, it is better than no estimate at all. It is well known that the contribution of uranium and thorium to the dose rate is determined essentially by the total alpha count rate, and is very insensitive to the relative contribution of uranium and thorium to that count rate. "Accidental" pairs arise from the random coincidence of two alpha particles within the resolving time of the apparatus; and the rate of these is proportional to the square of the total count rate.

The use of a uranium source of sufficiently low concentration allows the accidental pairs rate calculation to be checked, since all slow pairs can be attributed to random coincidences. Our checks of this gave results consistent with the known resolving time. In addition, for a thorium source of low enough concentration the pairs rate, after allowing for accidentals, gives an independent estimate of the absolute efficiency ε. For counting at 100% efficiency the pairs rate is proportional to the total count rate but, if ε is less than 100%, then the pairs rate is proportional to ε^2, on the assumption that each alpha particle is independently detected with efficiency ε. In table 1, the last entry for batch 5 shows an efficiency calculated in this way. It compares favourably with the immediately preceding entry for the total count measured at the same time. It may be noted that this measurement was extended for enough time to record 2,500 pairs.

So far as finding the thorium concentration by pairs counting is concerned, an efficiency of less than unity is not very serious if the data are to be used for dose rate calculations, for the reasons given earlier. However, if the thorium concentrations are to be compared with those found by other means e.g. neutron activation analysis or alpha spectrometry, the efficiency factor cannot be neglected. The late John Hutton (personal communication) had carried out an extensive comparison of thorium analyses made by various methods and arrived at an empirical correction to be applied to pairs counts. It has turned out to be almost the same as the ε^2 efficiency factor.

Acknowledgements

The authors are grateful to the Australian Research Council and the Australian Institute of Nuclear Science and Engineering for assistance provided during the preparation of this paper. We would also like to thank Professors DJ Huntley and MJ Aitken for their many helpful comments. JMW thanks the Department of Physics and Mathematical Physics at University of Adelaide for a Summer Scholarship.

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PR D. Huntley

This work presents a convincing statement that all is not well. I am not convinced that the problem entirely lies in the ZnS screens rather than elsewhere. The different "efficiencies" obtained for different ZnS batches are strong evidence for the former; there is, however potential for other problems. The uncertainties in the alpha particle ranges appear to be of the order of 5%, as judged by differing values published over the years. In this respect calculations using the later ranges of Ziegler (1985) are needed. My experience with uranium standards is that the suppliers will not provide quantitative information about the degree of secular equilibrium, and this is most important. In addition there is the question of radon escape. McCorkell (1986) showed that it varied from 1.7 to 15.5% in the Canadian standards, was 7.5% in one DOE (USA) standard, and up to 34% in others. It was negligible in glassed standards. Does anyone know where the radon goes in a sealed TSAC cell? The situation with thorium standards is much better because of the short half lives, apart from the potential for radon escape. A glassed thorium standard should relieve this problem, and thus be the best. If the ZnS is imperfect why did Huntley et al. (1986) get such good agreement (~3%) between measured and calculated count rates on glassed thorium standards? And why did Aitken (1985, pp 306-7) get such good agreement with powdered standards? I don't know.

Turning now to the slow pair count rates, we have, over the years, found measured pair rates to be consistently lower than those calculated from Th concentrations determined by NAA. For this reason I use the figure 2.05r instead of 2.31r that is in Table 3 of Huntley et al. (1986).

This is a good opportunity to point out that the corresponding constants for fast pairs in that table are too large because no allowance was made for the fact that the first alpha decay is to an excited state which engenders a delay of the emission of the second alpha particle.

Some notes on the experimental side :

a) For the same purpose as the use of "Hanstand" we have used a glassed standard for many years. This is placed directly onto the ZnS and does not significantly contaminate the ZnS when left on it for a few hours. It is stored in a desiccator.

b) If the amplifier gain is set relatively large, some pulses will exceed 10 V (this will be the case for Fig. 1). Whether or not these pulses are counted will depend on the details of the electronics, and presents the potential for missed counts.

If the amplifier gain is set relatively small, the discriminator zero error (i. e. zero discriminator setting does not correspond to zero volts) becomes amplified and it is necessary to determine where zero volts is on the discriminator.

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