CHAPTER 62

Low-Level Scintillation Counting with a LKB Quantulus Counter Establishing Optimal Parameter Settings

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The use of spectrophotometric quartz vials with a square or rectangular cross section was reported in 1977.[1] The original application was in a counter with a single loading drawer (Intertechnique LS 20). Subsequently, counters from different manufacturers were used with the same type of vials. Today, all commercial counters have multiple sample loading mechanisms designed to handle standard glass vials. Our square vials are smaller than the standard vial; therefore, we need a holder with the outside dimensions of the standard vial to stabilize the square vials. These vial holders are lifted into the counting chamber by a piston. In some counters the lifting piston does not have a fixed rotational position. Since it is important that the vial face is always perpendicular to the photomultiplier (PM) tube axes, the loading mechanism had to be redesigned to insure that the vial always assumes the same position with no rotation. Some counters have a complicated system of shutters to protect the energized PM tubes from ambient light sources. The presence of shutters makes the design may be quite involved.

This modification of the piston mechanism was particularly simple for the Quantulus. The loading piston is part of a lever system and does not rotate. A V-shaped groove was cut in the flat surface of the piston. A ridge on the bottom of the black nylon holder fits in this groove and determines the vial position precisely.

The holder has the size of a standard counting vial (28 mm in diameter and 60 mm in length). A rectangular window is cut through the holder, along and centered around the PM tube axis. The quartz vial is positioned in this window and always exposes the same surface area to the PM tubes. The inner surfaces of the holder are made reflective by attaching a thin film of aluminized mylar. The benzene filling level in the vial is kept constant, and different sample volumes are accommodated by using vials with different volumes. We have vials available with volumes of 4.5, 3.0, 1.5, 0.6, and 0.3 cc.

The advantage of this design lies in the straight and direct optical link between vial and PM tubes. Optical conditions are precisely the same after each loading cycle. In contrast, most round standard vials are not precise cylinders. It is especially difficult to produce round quartz vials without distortions.

We prepare our sample benzene for scintillation counting by adding 0.91 wt% of Butyl-PBD primary scintillator. The precise amount of scintillator powder is added directly to the sample benzene. For our smallest vial (0.3 cc) the exact scintillator weight is 2.35 mg. This amount can be weighed routinely and introduced into the vial with an error of ±5%. Recently, new scintillation techniques have been introduced which allow for the photon pulse length modifications of beta decays. This is achieved by adding a small percentage of a secondary scintillation fluor to the primary scintillator. We do not believe that the extremely small amounts of required secondary fluor can be weighed with sufficient precision to accommodate our laboratory methods; therefore, we have not investigated secondary fluors. The advantages of adding the scintillation powder directly to the sample are: (1) the full volume of the vial is available for sample benzene and (2) a potential source of contamination is avoided. Our concern lies with impurities in the cocktail solution that may cause quenching or contain trace amounts of ^{14}C.

Recent design modifications of the Quantulus counter are making possible a wide range of discriminator adjustments for pulse height difference (PAC) and variation of pulse length (PSA). The range of the digitally controlled adjustment is between 1 and 256 for both discriminators. With the PAC discriminator, pulses from the left and the right PM tubes are compared. Dissimilar pulses are rejected and the rigor of comparison becomes stricter as the setting number is increased. The PSA discriminator passes longer pulses as the setting number is increased.

We have varied both settings over nearly their full range and also made some tests without the PSA discriminator. Four sets of measurements were made. A clear, fully transparent vial was used for measuring a background (anthracite) and a modern standard (NBS oxalic acid) solution. The series was repeated with a vial which had the outside of the quartz faces frosted with no. 400 grinding powder. These measurements were made without pulse energy restriction, i.e., the energy window was open to the full range of the observed beta energy.

Results are shown in Figures 1 to 3. Figure 1 presents the background benzene measurements. The benzene was synthesized in our laboratory from anthracite coal. Each measurement represents about 12 hours of counting time and has a standard error of about 0.02 cpm. Some scatter in the data was produced by the use of different vials and background solutions. One has to bear in mind that the very low background of the Quantulus counter will cause the above mentioned differences to appear as major fluctuations. We have picked from our tabulation of data two series of PSA values, one for PAC set at 5 (pulses with large amplitude difference are passed) and one for PAC set at

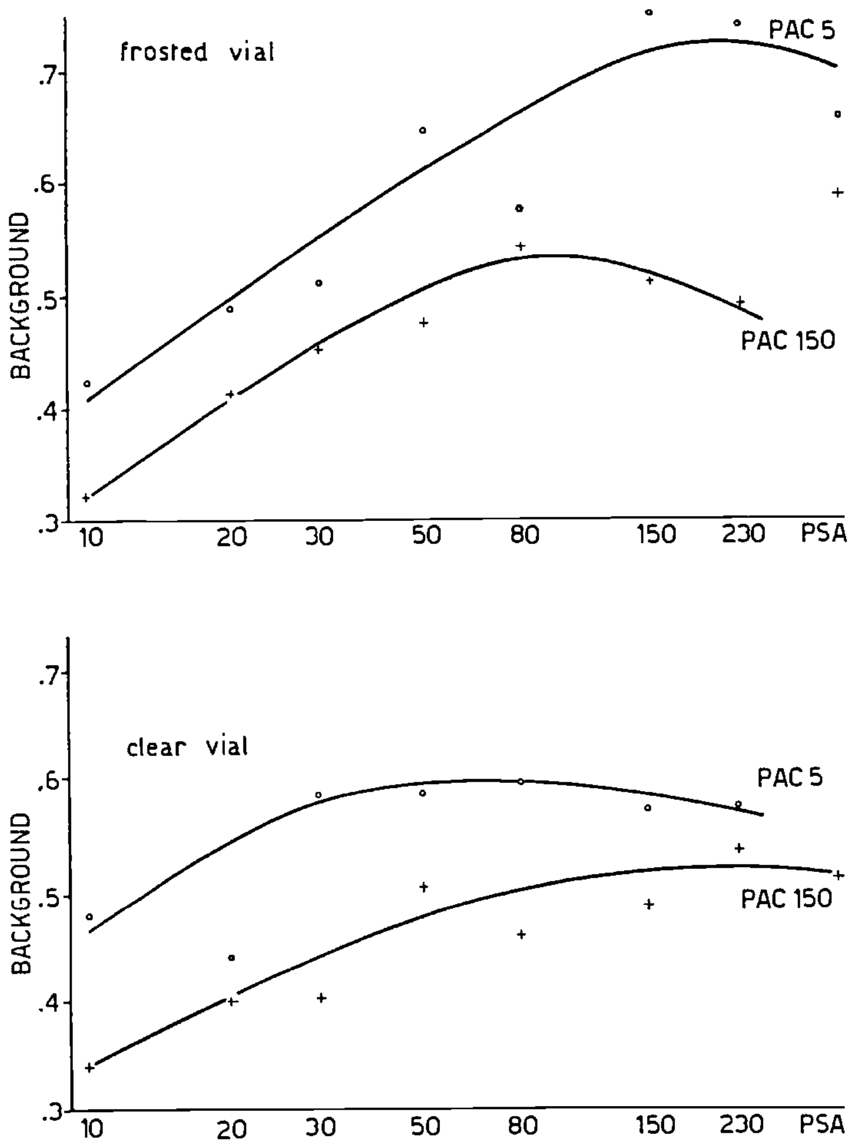

Figure 1. Background in cpm of a square quartz spectrophotometric vial with a light path of 10 mm, filled with 3 cc ^{14}C-free benzene and 0.9% butyl PBD scintillator. Pulse shape discriminator PSA restricts length of PM pulses strongly with setting 10 on scale and weakly with setting 230. Points on far right are measurements made without PSA discriminator. PAC curves show effect of discriminator for pulse amplitude differences. With PAC = 5 curve (open circles) only extreme differences are filtered out, with PAC = 150 (crosses) also smaller differences are filtered out.

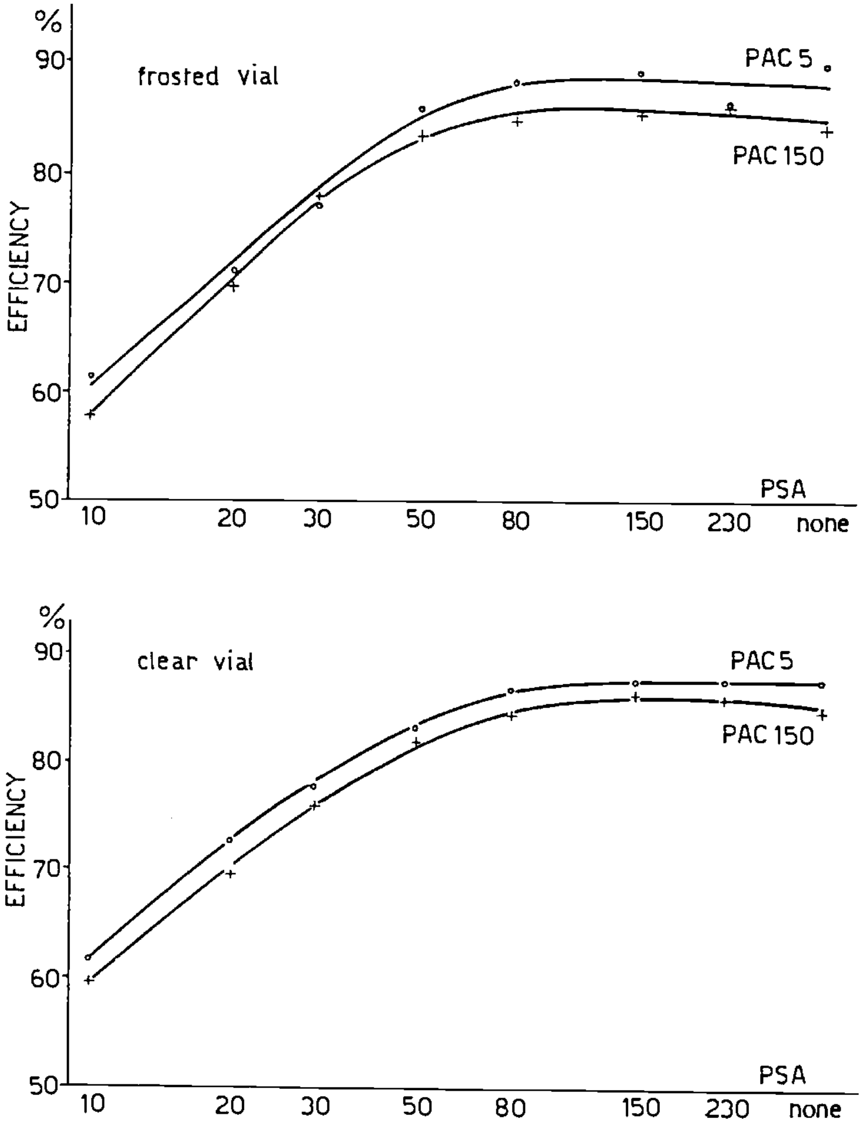

Figure 2. Counting efficiency measured with modern ^{14}C standard. Percent efficiency is based on the ratio of measured net count rate over theoretical rate of decay of the standard. Parameters shown are discussed in caption to Figure 1.

150 (moderately small pulse difference). We have also recorded two measurements with the PSA discriminator switched off. These results showed the largest deviation from the expected trend, causing some points to lie outside of the background graph. Pulse amplitude discrimination has a substantial influence on lowering the background. At PAC = 150 a larger number of pulses is

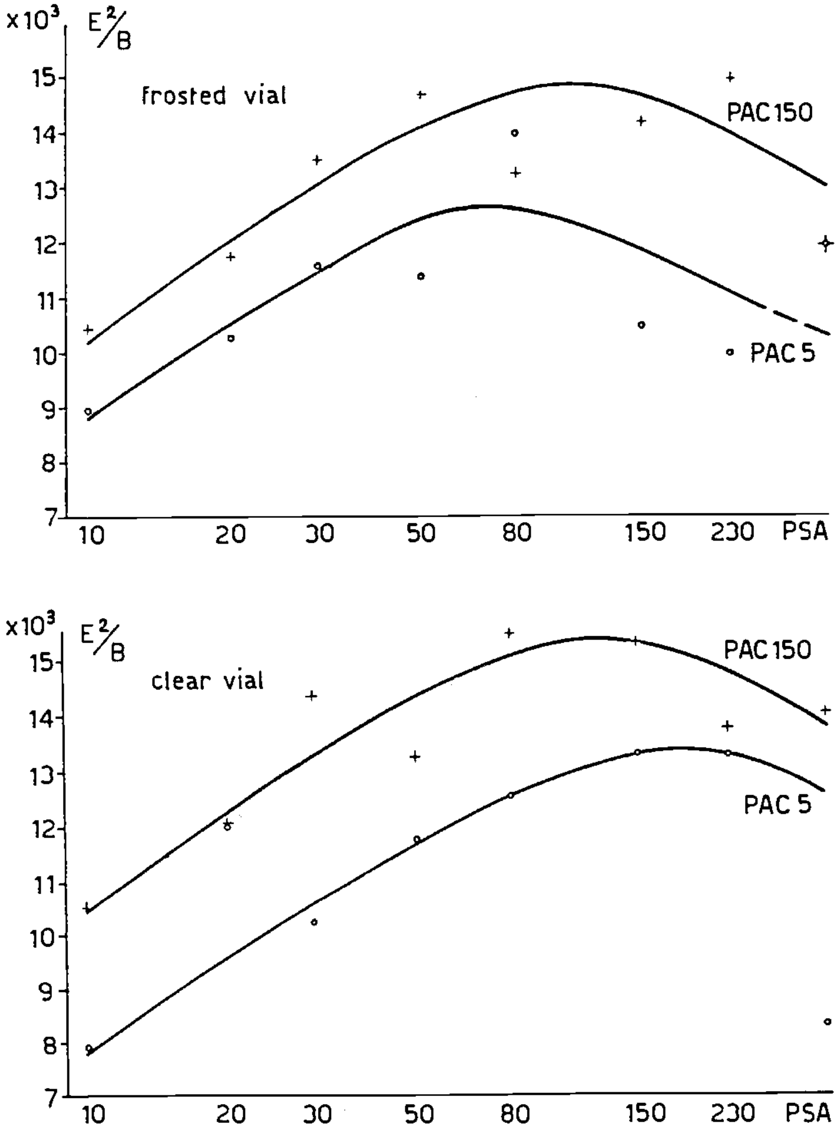

Figure 3. Figure of merit E^2/B. E is efficiency shown in Figure 2 and B is background shown in Figure 1.

rejected, which suggests that assymetrical pulses are abundant. These may originate from the glass envelope of either PM tube rather than from the vial. Frosted vials appear to increase the pulse height assymmetry. This is shown by the higher background level when the discrimination setting is at PAC = 5. Even at PAC = 150 a slightly higher background is observed for frosted vials.

Figure 2 shows the relation of counting efficiency to the PAC and PSA adjustments. Settings are the same as in the background study. The count rate of the modern standard used in this experiment is about fifty times higher than background; therefore, the scatter of the background data is only an insignificant contribution. The small separation of the two curves shows that pulse amplitude discrimination plays only a minor role. This implies that the vials, set in their holders, are radiating the photons equally from both windows of the holder, regardless of the point of origin of the photons within the vial. The large dependence of efficiency on the pulse length discrimination shows a substantial rate of ^{14}C pulse rejection up to a setting of 80.

The difference in efficiency between frosted and clear vials is insignificant. At PAC = 5 the frosted vials are more efficient by about 3%. At PAC = 150 the difference disappears. Clear vials have a lower background; thus, we have given preference to this vial type. Furthermore, clear vials have the added advantage of allowing sample filling and withdrawal process observation and residual matter detection in the vials. For example, wax-like coatings tend to build up over a 6-month period and can be seen in clear vials as a faint greyish layer on the inside up to the benzene filling level. This contaminant is a byproduct from the catalytic conversion of acetylene to benzene.[2]

The E^2/B, or figure of merit, is not controlled by the counter alone. It is strongly influenced by the vial and the reflectors installed around the vial. Figure 3 clearly shows the advantage of using pulse amplitude discrimination at a level of about 150. Additional data, which is not shown here, indicates that higher settings will not substantially decrease background but will negatively affect efficiency, causing a decline in E^2/B.

The ideal PSA setting is more difficult to assess. Efficiency and E^2/B graphs indicate an optimal setting of PSA = 80 or slightly higher; however background is substantially higher at the PSA = 80 setting than at the PSA = 30 setting, i.e., 0.5 compared to 0.43 cpm. This may be an important consideration if small or old age samples with net count rates of 0.1 cpm or less are to be dated.

CONCLUSION

Simple mechanical adaptations make it possible to use precisely manufactured square and rectangular quartz vials in most counters. Archaeological samples and geochemical probes (water and atmosphere) are usually small and seldom yield more than 4 cc of synthesized benzene; therefore, the reduced volume of these vials is not a disadvantage.

Our detailed study of this vial type in a Quantulus counter demonstrates the usefulness of electronic pulse filtering. Choosing clear vial faces, a sample volume of 3 cc benzene, a pulse amplitude (PAC) discrimination setting of 150 and pulse length discrimination setting of 80, and using no pulse energy limitations in the range of ^{14}C pulses, one can obtain a background of 0.48 cpm, an efficiency of 84% and a E^2/B factor of 15,000. For 0.6 cc vials we recently measured a background of 0.14 cpm, an efficiency of 75% and an E^2/B factor of 40,000.

Additional improvements on these numbers are possible by restricting the energy windows for the ^{14}C pulses to the main portion of the peak and cutting off the low energy tail of this peak. A brief, current study involving 3 cc vials indicates a lowering of background by 0.1 cpm, a decrease in efficiency of 4.5%, and a slight increase in E^2/B of about 2000.

The major application of the Quantulus installation will be in dating old archaeologic samples, as well as small carbon samples in the weight range of 200 to 600 mg.

ACKNOWLEDGEMENT

We acknowledge the support received from the NSF archeometry program with the grants BNS-8845200 and BNS-8801697.

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