Basics of α/β–discrimination for liquid scintillation counting.

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Introduction

The α/β-discrimination is of major importance for measurement of environmental samples and for the control of nuclear power stations. Samples in these areas often contain α- and β-emitters in the same sample. For the successful use of the α/β-discrimination you need some experience with liquid scintillation counters and the corresponding applications. This application note describes the α/β-discrimination including the pulse analysis (pulse decay analysis = PDA) but other methods of the classical scintillation technique are also important such as time-resolved liquid scintillation counting[1,2,3].

Pulse decay analysis

We can detect α-particles with almost 100% counting efficiency in a LSC instrument but the ratio of the number of photons formed during the scintillation process to the energy of the α-particle is only 1/10 of the ratio that we observe during a β-decay. Because we calibrate the energy scale of the LSC with β-sources we will see an α-particle at 1/10 of its energy. A 5 MeV α-particle will appear at approximately 500 KeV. Therefore there will be substantial overlap of many α-emitters with β-particles in the multichannel analyzer, so discrimination only on the basis of the decay energy is not possible.

In order to understand how the separation of α-decays from β-decays is accomplished, it is necessary to examine the process at a molecular level. α- and β-events may be distinguished from one another in a liquid scintillation counter by examining the electronic pulses that are produced at the PMT anode of the detector.[4, 5]
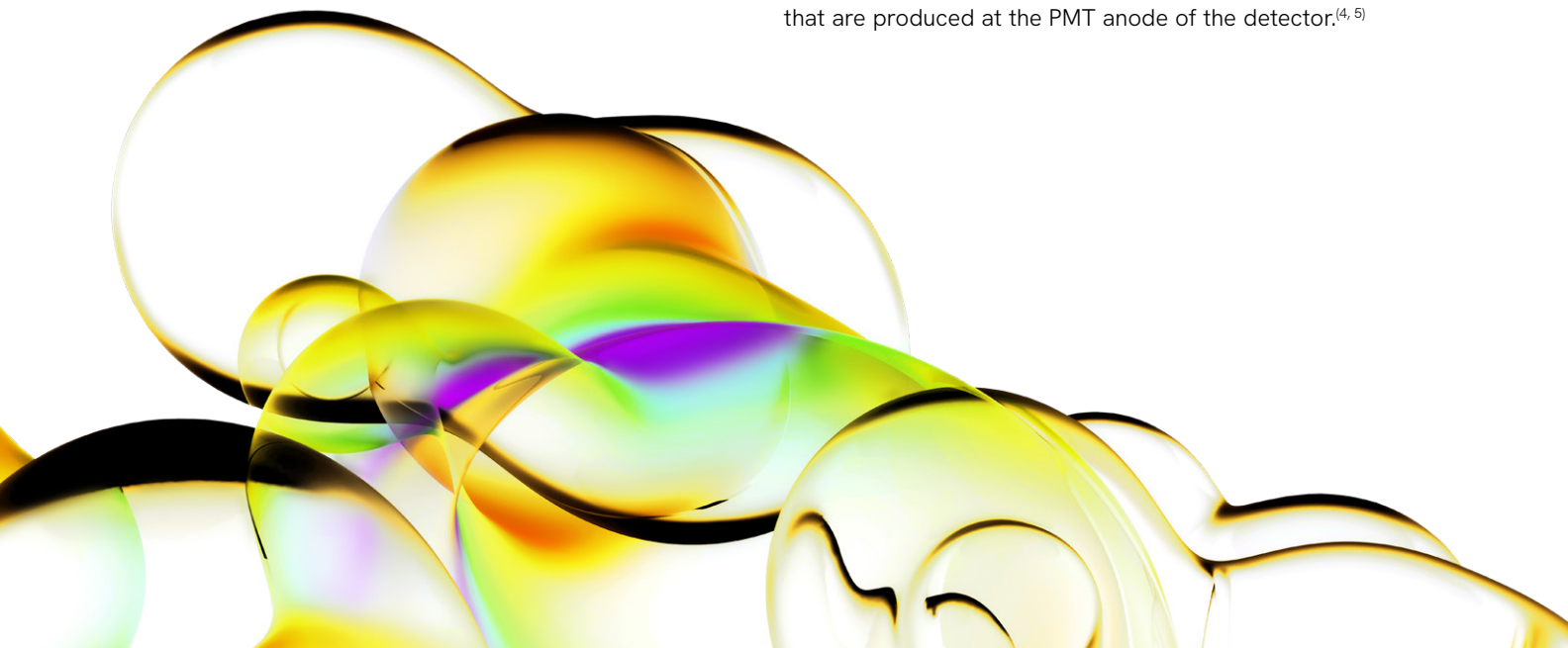
These pulses are made up of two major components: The prompt component and the delayed component.[6] These components occur in different proportions in α- and β-pulses with the result that α-pulses are longer than β-pulses (see figure 1). We already mentioned the low photon conversion rate of a-particles. This is due to the fast component that contains a large amount of ionization quench and does not result in photons.

The delayed component, compared to the fast component, is much larger in α-decays than in β-decays. The longer lifetime of signals from α-particles in the electronic circuits is the basis of the α/β-discrimination. α-pulses can be 30-40 ns longer than β-pulses.
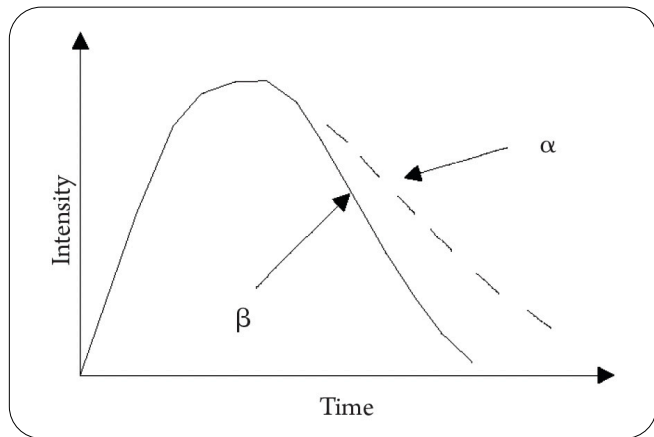

Figure 1: Different pulse length of α- and β-pulse

PDA and TR-LSC

Pulse decay analysis is based on the zerocross pulse timing technique. In general, a zero cross sensing circuit has consistently proven to provide excellent α/β-separation.[7] The zerocross method is illustrated in figure 2 and 3. To further optimize the separation of α-events from β-/γ-decays, the anode pulses are integrated, slowed and further shaped by passing through a low pass filter. This stretches the pulses as represented graphically in figure 2.

A derivative of this wave function produces a zero crossover pulse as indicated in figure 3. From this information, an optimum pulse decay discriminator (PDD) may be set between the zerocross time points of the two types of pulses.

Because α- and β-pulses have a different pulse length, setting optimized PDD values allows separating between α- and β-particles.
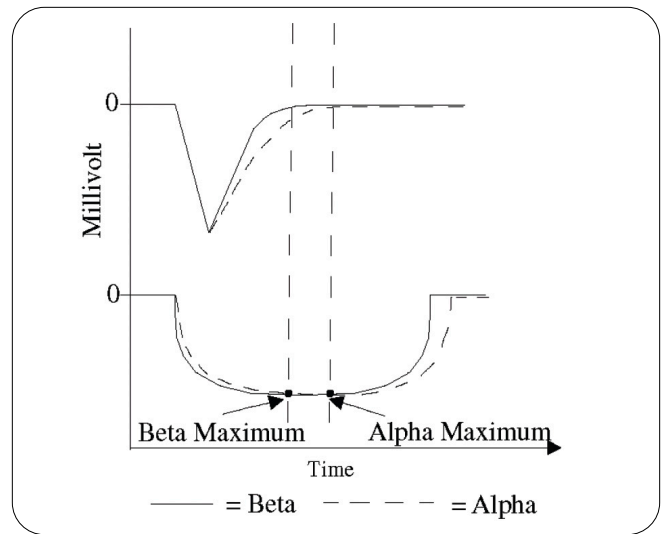

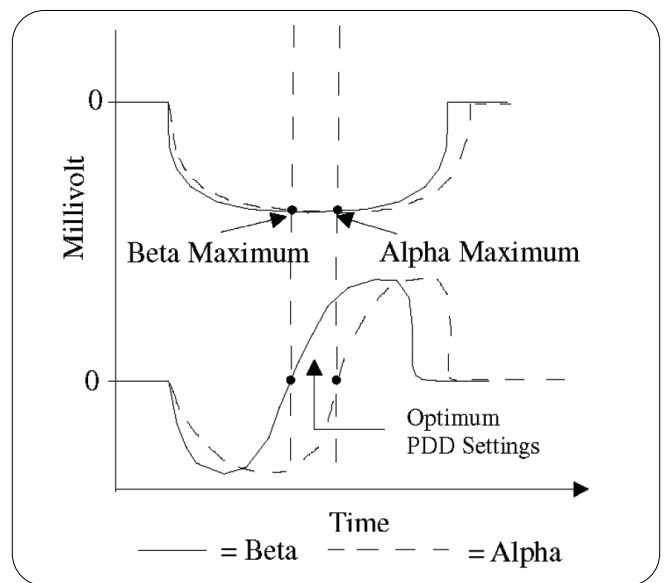Figure 2: Low pass filtered pulse processed with pulse decay analysis

Figure 3: Zero crossover Puls for the Determination of PDD Settings

Pulses which decay in a shorter time than indicated by the PDD values will be classified as β-events and consequently will be saved in the β-multi channel analyzer (MCA); pulses which are longer will be saved in the α-MCA.

You can minimize the misclassification of α-particles as β-particles and vice versa.

Additional background discrimination or guard detectors are not necessary for α-detection. High energy cosmic rays, interacting with the scintillator, will be discriminated by the PDA in the same way as β-radiation.

TR-PDA (time resolved PDA) is the combined use of TR-LSC background discrimination and pulse decay analysis. By applying TR-LSC to PDA, α-events misclassified as β-events are discriminated by TR-LSC since the long "tails" (delayed component) of α-pulses resemble background events. Thus, α-decays misclassified as β-decays will be eliminated from the β-MCA. In addition, the misclassification of β-decays will also be reduced since the TR-PDA causes a shift to a higher optimum PDD value. At higher PDD values, fewer β-events can satisfy the time requirement for a α-pulse which reduces the β-spill into the α-MCA. The net result of TR-PDA is a reduction of two- to five-fold in the misclassification of β- and α-events.

Alpha/Beta calibration

In order to optimize α/β-separation performance, it is essential to determine the correct PDD setting as already described. On Revvity TriCarb® models with α/β-option, the optimum setting is the setting where there is equal and minimum spill of α-pulses into the β-MCA and β-pulses into the α-MCA.

Figure 4 illustrates the percent spillover or percent misclassification of [241]Am and [36]Cl samples in ULTIMA™ Gold AB, a cocktail specifically designed for α/β-separation. The determination of an optimum PDD requires two standards, one pure α-emitter and one pure β-emitter of interest. For the most accurate results, the standards must be as near identical as possible to the unknown samples in their chemistry, volume, vial type etc. For gross α and gross β measurements where the particular radionuclides may not be known, a α- and β-standard of similar energy to the α- and β-nuclides in the samples is desirable.

To find the optimum PDD settings, each standard is counted individually at a range of PDD settings and the percent misclassification of α into the β-MCA and vice versa are plotted against PDD on the same graph.

When only the β-emitter is of interest, a PDD value below the instrument determined optimum may be used, which minimizes the misclassification of α-activity into the β-MCA at the expense of reducing the β counting efficiency. Similarly, when only the α-emitter is of interest, a PDD value greater than the optimum can be used. This minimizes the misclassification of β-events into the α-MCA at the expense of reduced a counting efficiency. As mentioned previously, the instrument determined optimum PDD is at the intersection of the two curves which corresponds to the minimum misclassification of α- and β-events.
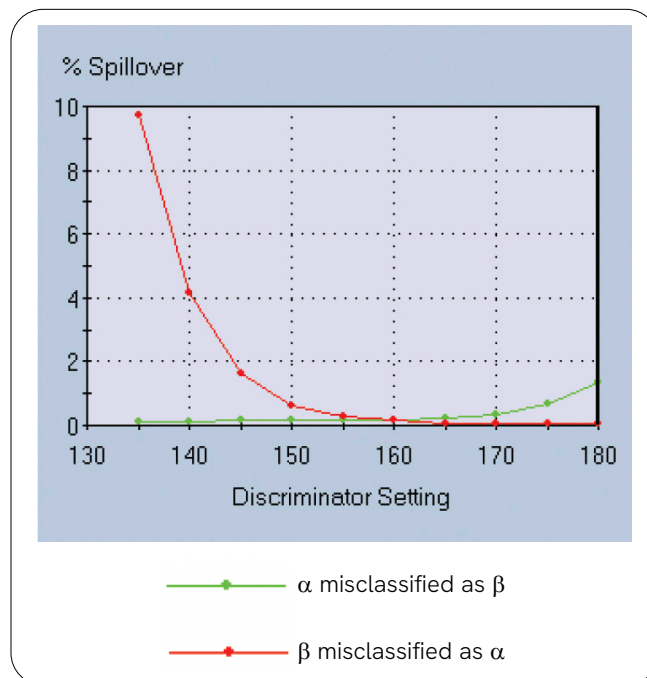

Figure 4: Misclassification plot for the determination of optimum PDD settings

Factors influencing the α/β-discrimination

Quench

We have seen how to determine the optimized PDD settings. However, a number of factors have an influence on the PDD settings and a correction of the determined PDD settings might be necessary in some cases. Strong quench in your sample may have an influence on the PDD settings. The PDD settings as well as the amount of misclassification can change with increasing quench. In general the amount of misclassification is increasing with increasing quench.

The optimum PDD settings for the example shown in figure 5 have been determined as 188 and the spill of α-pulses into the β-MCA and vice versa were 0.4% in each case. The same but heavier quenched sample can be seen in figure 6. The PDD settings as well as the misclassification have changed significantly. The optimum PDD value is now 123 and the β-spill into the α-MCA as well as the α-spill into the β-MCA is 1.15%.

When the degree of quenching for a particular set of samples varies, there are two possible approaches. The first approach is to produce a single pair of α/β-standards which are similar in their quench (tSIE) value to the least quenched sample.

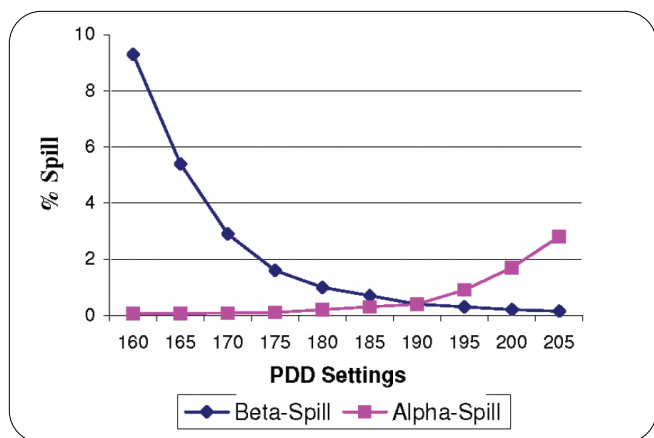Figure 5: Misclassification of ^{36}Cl and ^{241}Am without quench
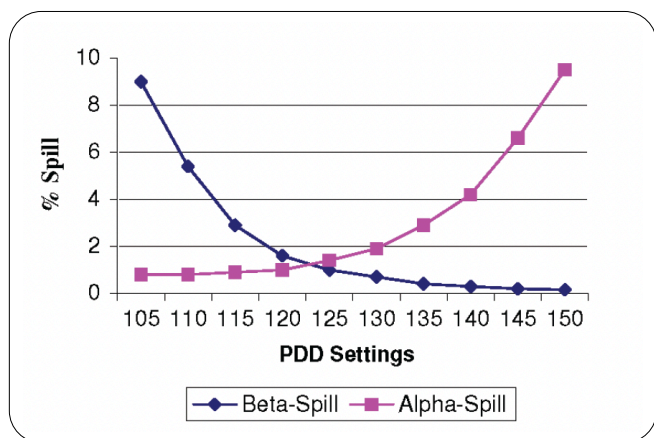

Figure 6: Misclassification of ^{36}Cl and ^{241}Am with strong quench

The next step is to progressively quench them and reoptimize the PDD at each quench level. This produces a series of PDD values and percentage misclassifications for a range of quenching (tSIE values).

The degree of sample quenching in each unknown sample may then be measured by making a short count on each sample to determine the value of the tSIE quench indicating parameter. Then, samples have to be counted at their individual optimum PDD conditions, which could conceivably require a separate protocol for each sample.

The alternative approach is to prepare a pair of standards which are equivalent in quenching to the least quenched samples, determine the optimum PDD and then progressively quench the standards. However, in this approach, the misclassification is always determined at the original PDD setting of the least quenched sample. This allows the construction of a plot of percentage misclassification versus tSIE for a single PDD setting.

All samples may then be counted within a single protocol and a correction for misclassification as a function of quench is applied.

This approach would be in addition to a quench curve which relates quenching to detection efficiency. Figure 7 demonstrates the construction of such a correction plot.

Cocktails

Above we discussed that some mathematical operations are necessary to separate α– from β–pulses. Standard cocktails for β-counting applications which employ fast solvents, such as xylene, pseudocumene, toluene and alkylbenzenes, are less efficient for separating α– from β–activity. Figure 8 illustrates the misclassification of ^{238}Pu and ^{90}Sr/^{90}Y which was achieved using the old InstaGel® (xylene cocktail, todays Insta-Gel Plus is based on pseudocumene). This would be a typical misclassification value for a cocktail employing a fast solvent. To overcome the poor separation, 20% naphthalene has been added to standard cocktails for α–/β–separation applications.[8]
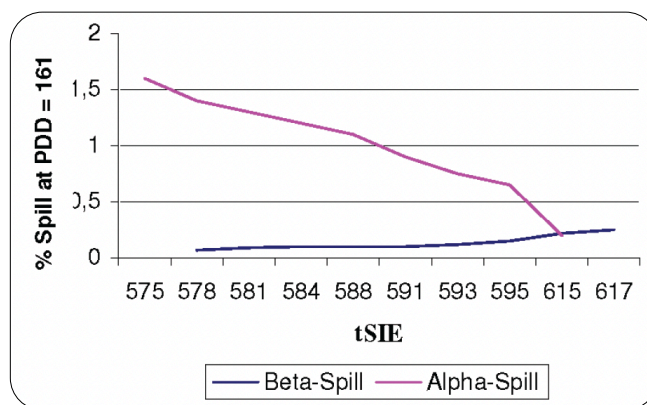

Figure 7: Influence of quench on the misclassification

Naphthalene improves α–/β–separation and reduces misclassification of Insta-Gel cocktails by acting as an intermediate in the energy transfer process between the solvent and fluor.[9,10] This route increases the production efficiency of excited fluor molecules. Figure 9 illustrates the improvement in separation which is obtained when 20% naphthalene is added to Insta-Gel.

Other cocktails, such as the ULTIMA Gold series of cocktails, are based on diisopropylnaphthaline (DIPN). These cocktails have some advantages over fast classical cocktails with 20% naphthalene. ULTIMA Gold is non-toxic, non-flammable and biodegradable, and is therefore preferable in cocktail manufacture to naphthalene.[11]

ULTIMA Gold AB, the first cocktail specifically developed for α-/β–discrimination applications, has also exhibited superior α-/β–separation compared to fast cocktails with 20% naphthalene, and has an excellent water and acid holding capacity.

Table 1 indicates that the addition of ULTIMA Gold F (UG F) to samples in ULTIMA Gold AB reduces the misclassification.
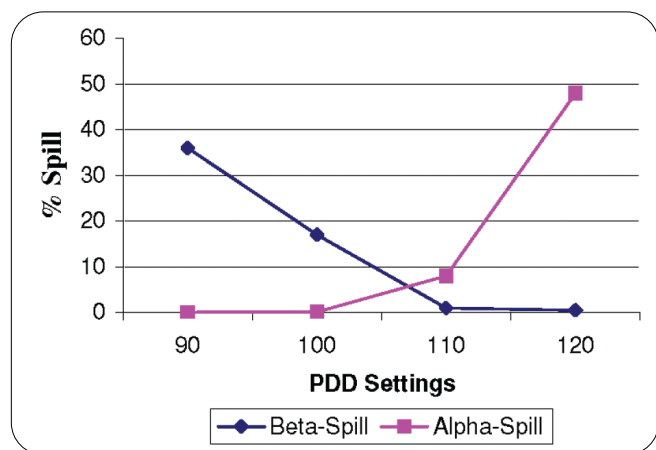
However, this reduction of misclassification will be achieved at the expense of a lower sample uptake capacity because we replace the emulsifying cocktail ULTIMA Gold AB with a pure organic cocktail ULTIMA Gold F. Therefore the uptake capacity for aqueous and strong ionic solutions will be reduced drastically with an increasing amount of ULTIMA Gold F.

Figure 8: Misclassification of ^{238}Pu and ^{90}Sr/^{90}Y in Insta-Gel
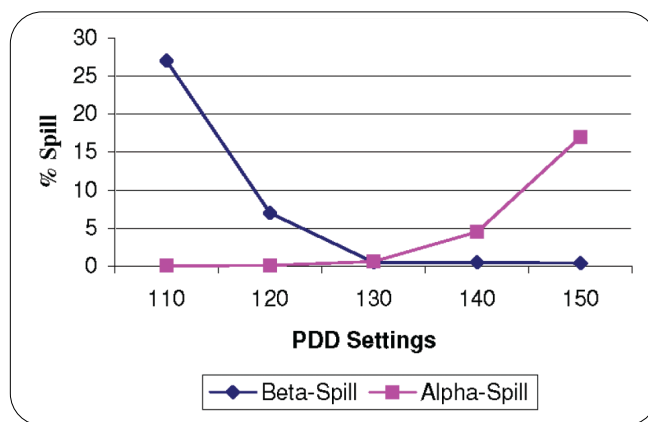
Abb. 9: Misclassification of ^{238}Pu and ^{90}Sr/90Y in Insta-Gel + 20% Naphthalin

Table 1: Uptake capacity of UG AB/UG F and corresponding misclassifications

Sample type	ULTIMA gold AB	85 Vol % UG AB, 15 Vol % UG F	75 Vol % UG AB, 25 Vol % UG F	50 Vol % UG AB, 50 Vol % UG F
Water Sample uptake Misclassification	0,2-10,0 mL 0,74-1,87%	0,2-2,25 mL 0,77-4,21%	0,2-2,25 mL 0,58-0,68%	0,2-0,5 mL 0,42-0,47%
1 M HCL Sample uptake Misclassification	0,2-5,5 mL 0,87-1,73%	0,2-2,0 mL 0,64-0,79%	0,2-1,25 mL 0,64-0,75%	0,2-0,4 mL 0,43-0,52%
2 M HCL Sample uptake Misclassification	0,2-5,5 mL 0,87-1,73%	0,2-1,25 mL 0,56-0,76%	0,2-1,0 mL 0,49-0,60%	0,2-0,3 mL 0,38-0,49%
1 M HNO$_3$ Sample uptake Misclassification	0,2-3,25 mL 0,75-3,85%	0,2-1,75 mL 0,66-1,51%	0,2-1,25 mL 0,55-0,98%	0,2-0,4 mL 0,48-0,53%
2 M HNO$_3$ Sample uptake Misclassification	0,2-2,25 mL 0,77-4,21%	0,2-1,5 mL 0,60-1,91%	0,2-1,0 mL 0,70-1,17%	0,2-0,3 mL 0,54-0,62%

Sample volume

Several experiments have been performed to determine the effect of sample volume on α–/β–separation. The Plots were acquired with ^{36}Cl and ^{241}Am in Insta-Gel + 20% naphthalene. The percentage misclassification and optimum PDD values are presented in table 2. These demonstrate virtually constant PDD over the entire volume range and only a small increase in the misclassification at the 1 and 2 mL volumes.

However, this can be accounted for by the increasing importance of the background count rate in the misclassification calculation since background subtraction is not performed in the calculation of the misclassification spill plot.

Vial type

Table 3 illustrates the change in percentage misclassification of events over a period of 24 days using α- and β-standards prepared in 7 mL and 20 mL glass vials, 20 mL plastic vials and 20 mL low diffusion vials (Revvity low diffusion vials are specially prepared polyethylene vials with a thin Teflon®-like inner coating). There appears to be no significant difference in performance between standard 7 mL and 20 mL glass vials, and no change in misclassification over time. On the other hand, the misclassification from standard plastic vials systematically increases throughout the time-course of the experiment. The low diffusion vials are similar in response to plastic vials except that there is a lag period of about two days before there is an increase in misclassification.

Table 2: Dependency of PDD and %Spill on the volume

Sample volume (mL)	Alpha spill (%)	Beta spill (%)	Optimum PDD settings
16	0,69	0,70	101
10	0,65	0,62	101
8	0,64	0,57	101
4	0,68	0,67	101
2	0,85	0,74	100
1	1,14	1,02	98

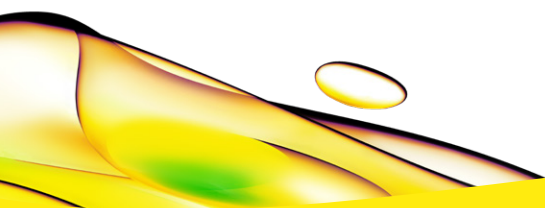Table 3: α/β – misclassification as a function of time and vial type

Days after sample formulation	Percentage misclassification of α–particles in the β–MCA/β–particles in the α–MCA			
	20 mL glass vial	7 mL glass vial	20 m plastic vial	20 mL low diffusion vial
0	0,47/0,51	0,56/0,58	0,56/0,65	0,46/0,53
1	0,42/0,51	0,49/0,62	0,59/0,90	0,43/0,61
2	0.49/0,51	0,48/0,62	0,96/1,01	0,53/0,63
6	0,41/0,55	0,47/0,55	1,39/1,51	0,68/0,81
9	0,38/0,51	0,43/0,54	1,61/1,78	0,96/0,95
16	0,41/0,49	0,46/0,56	1,94/2,24	1,22/1,28
24	0,49/0,51	0,49/0,54	2,18/2,53	1,40/1,67

The effect of the plastic vials is due to diffusion of some component into the vial wall, since decanting the vial contents into a glass vial restores the misclassification to that which is typically observed in a glass vial. The diffusion of cocktail into the plastic wall results in a stretching of the beta pulses which is known for solid scintillators, thus reducing the difference in the pulse length between α– and β–pulses.

Further improvement can be obtained by etching the inner wall of glass vials. Etching the vials improves the light output and thus improves the ability to separate α– and β–events. This procedure also improves the spectral resolution.

Literature

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