

RADIOASSAY OF LOW-LEVEL, LOW-ENERGY BETA ACTIVITY IN MULTILABELED SAMPLES CONTAINING HIGH-ENERGY BETA IMPURITIES USING LIQUID SCINTILLATION SPECTROMETRY

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ABSTRACT. We have developed an optimized multiple window liquid scintillation counting technique, combined with internal standardization and spectrum unfolding for the determination of low-level, low-energy beta activity in multilabeled samples containing high-energy β impurities. We use a software program or spreadsheet for manipulating the spectral data and reconstructing the distinct spectral contributions for every individual radionuclide and impurity. The most important advantage of this method is that it does not require setting up quench correction curves. Furthermore, the knowledge of exact reference activity is not required, thus eliminating two sources of introduced error in calculating final results. We have used the technique successfully on mixtures of ^{99}Tc and ^{63}Ni contaminated with ^{60}Co and ^{137}Cs over a wide range of quenching and activity ratios.

INTRODUCTION

Standard procedures to analyze a complex mixture of beta-emitting radionuclides by liquid scintillation counting (LSC) usually involve chemical separations to isolate all radioisotopes of interest in the sample, followed by an analysis of β activity in a predefined counting window or on the total β spectrum. Although these procedures are accurate in most cases, they fail to give satisfactory results for samples contaminated with a large surplus activity of impurities (a thousandfold or more), even with separation factors up to 99.9%. Known examples are those taken from typical nuclear waste streams produced by pressurized water reactor (PWR) nuclear plants where activity ratios between key nuclides (such as ^{60}Co and ^{137}Cs) and critical nuclides (such as ^{14}C , ^{63}Ni , ^{99}Tc and ^{129}I) exhibit values between 10^3 to 10^6 (Deconinck *et al.* 1993).

In a previous program (Verrezen and Hurtgen 1992) we noted that radioactive waste samples, chemically prepared for the measurement of ^{63}Ni , ^{99}Tc or ^{129}I by LSC, still contained amounts of ^{60}Co and in some cases ^{137}Cs large enough to cause considerable interference. Especially with ^{99}Tc measurements, ^{60}Co contamination is extremely difficult to trace in LSC because of the very small difference in β energy. Therefore, the commonly used multiple-window techniques for routine measurements of multiple-labeled samples (Matsui and Takiue 1991; Snipes and Lengeman 1971; Takiue *et al.* 1990; Veen 1974) fail to give correct results. Further, using quench correction curves to analyze samples with highly variable radiochemical composition (such as radioactive waste samples) requires the establishment of a vast set of calibration curves. For the same reason, spectrum-unfolding procedures (Grau, Martin-Casallo and Grau 1991) are too labor-intensive to be interesting for occasional, nonroutine measurements.

In this paper, we describe an optimized multiple-window procedure that combines internal standardization and spectrum-unfolding techniques, eliminating the need for quench correction curves. The only requirements are an LS spectrometer and a set of measuring vials containing 1 background vial, 1 or more sample vials and 1 sample vial with internal reference for each radionuclide present in the solution (both contaminant and nuclide of interest). One must know only the activity concentration of the reference (standard) for the radionuclide of interest.

THEORY

When a solution containing two or more β -emitting radionuclides is measured by LS spectrometry, the spectrum obtained is the sum of the individual contributions: IMP (impurity), NOI (nuclide of interest) and BGD (background). Figure 1 illustrates the effect of contamination by ^{137}Cs of a ^{63}Ni sample.

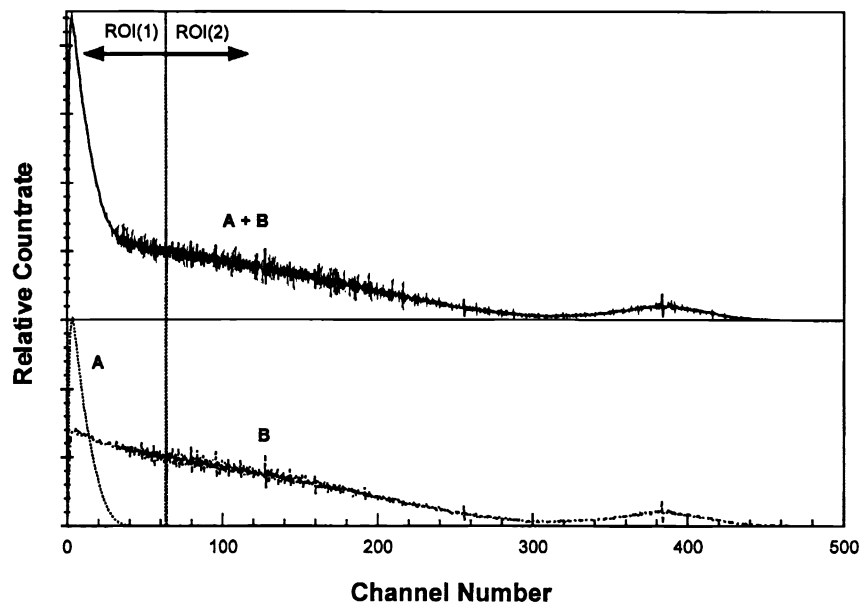


Fig. 1. Effect of contamination of a ^{63}Ni sample with ^{137}Cs on spectral shape. A = ^{63}Ni contribution; B = ^{137}Cs contribution; A + B = resulting spectrum

Therefore, the net contribution of the nuclide of interest is

$$\text{NOI}(\text{net}, i) = \text{SMP}(\text{net}, i) - \text{IMP}(\text{net}, i). \quad (1)$$

Here $\text{SMP}(\text{net}, i)$ equals the net contribution (obtained from the gross contribution $\text{SMP}(\text{gross}, i)$ after correction for the background $\text{BGD}(i)$) of the sample in a specified region of interest (i) as obtained from the spectrum. $\text{NOI}(\text{net}, i)$ and $\text{IMP}(\text{net}, i)$ are the net contributions of the nuclide of interest and the impurity to the β spectrum in the same region of interest. The background contribution $\text{BGD}(i)$ can be obtained by integrating the spectrum of a blank sample (with identical chemical and physical properties as the sample being measured) in the correct region of interest. The net count rate of the impurity in the sample on the contrary has to be simulated from a reference spectrum (REF). For this, the following hypothesis can be used:

$$\frac{\text{IMP}(\text{net}, \text{low})}{\text{IMP}(\text{net}, \text{high})} = \frac{\text{REF}(\text{net}, \text{low})}{\text{REF}(\text{net}, \text{high})} = K(Q). \quad (2)$$

This hypothesis states that the ratio of the net count rate in two fixed regions of interest for a single radionuclide is a constant for a given level of quenching. The indices *low* and *high* refer to two different regions of interest, one in the low energy part of the spectrum, the other in the high energy part. By setting the lower limit of the region of interest in the high energy part of the spectrum well

behind the endpoint of the spectrum for the nuclide of interest (so that $\text{NOI}(\text{net,high}) = 0$), this equation is transformed to the more useful form

$$\text{IMP}(\text{net, low}) = \text{REF}(\text{net, low}) \cdot \frac{\text{SMP}(\text{net, high})}{\text{REF}(\text{net, high})} \quad (3)$$

Using this relation one can rebuild the net spectrum of the impurity in the low-energy part of the sample spectrum by resolving the above equation for every channel in the observed β spectrum. The net countrate of the nuclide of interest (in the low-energy region of the spectrum) is given by

$$\text{NOI}(\text{net, low}) = \text{SMP}(\text{net, low}) - K'(Q) \cdot \text{REF}(\text{net, low}) \quad (4)$$

where

$$K'(Q) = \frac{\text{SMP}(\text{net, high})}{\text{REF}(\text{net, high})} \quad (5)$$

This equation can be expanded for samples containing more than one contaminant

$$\text{NOI}(\text{net, low}) = \text{SMP}(\text{net, low}) - \sum_{k=1}^n K'_k(Q) \cdot \text{REF}_k(\text{net, low}) \quad (6)$$

For samples containing n contaminants ($n + 2$), β spectra are needed to solve this equation: the sample spectrum, the background spectrum and n reference spectra (one for every contaminant present). The only prerequisite is that all spectra must be characterized by the same quenching level. This can be obtained by using quench calibration curves and spectral transformation functions (Grau, Martin-Casallo and Grau 1991) to shift all spectra to a common quenching level. However, this is a rather labor-intensive procedure.

The method that we propose here does not require any quench calibration curves and allows for easily obtained activity values, even for samples with high levels of contamination and/or quenching. All that is needed is an LS spectrometer and a set of counting vials with well-defined composition to assure that the quenching level in all vials is identical.

METHODS

All samples were prepared using OptiPhase HiSafe™ 3 LS cocktail (LKB/Pharmacia). Aqueous solutions of ^{63}Ni , ^{60}Co , ^{99}Tc and ^{137}Cs (Canberra-Packard) were used with activity concentrations of 5.5, 55 and 550 Bq ml^{-1} . Deionized, reversed-osmosis water was used for sample preparation. Carbon tetrachloride was used as a quencher to prepare the quenched samples.

All radioactivity measurements were carried out with a Packard TriCarb® 2250CA low-background LS spectrometer operating at a controlled temperature ($10 \pm 2^\circ\text{C}$). All samples were prepared and measured in 20 ml polyethylene vials. The quench parameter transformed spectral index of the external standard (tSIE) was obtained from an external gamma-ray source of ^{133}Ba .

For every measurement of a contaminated sample, we prepared a set of counting vials consisting of 1 background vial, 3 replicate vials of the actual sample and 1 internal reference vial for every radionuclide present in the sample (including the nuclide of interest). The background vial contained 2 ml of reversed-osmosis water and 18 ml of OptiPhase HiSafe™ 3 LS cocktail. Each sample vial contained 1 ml of sample, 1 ml of reversed-osmosis water and 18 ml of OptiPhase HiSafe™ 3. The

reference vials contained 1 ml of sample, 1 ml of an aqueous reference solution of the appropriate radionuclide present in the sample and 18 ml of OptiPhase HiSafe™ 3. With the possible exception of the background vial, all vials thus contained mixtures with identical chemical and physical properties (only the isotopic composition was different). Therefore, all vials were characterized by the same quenching parameter. No further quench correction was needed. In cases where the difference in chemical composition of the sample and pure water caused a considerable difference in quench-behavior (e.g., waste stream samples), a background solution was prepared with the same chemical composition to simulate as close as possible the quench behavior of the actual sample.

In the experiments in which the influence of quenching was examined, appropriate amounts of CCl₄ were added to the scintillation cocktail before the cocktail was added to the measuring vials.

All vials from one set were transferred into the sample changer of the LS counter and given a 2-h thermal equilibration period. All vials were then counted successively for 60 min each and the β spectra were recorded on disk. After counting, all spectra were transferred to a desktop computer and imported into a spreadsheet for further analysis.

Because all spectra are inherently characterized by the same quenching level, the net values of the spectral data can be obtained by simple mathematical calculations using the equations stated below

$$\text{SMP (net)} = \text{SMP (gross)} - \text{BGD} \quad (7)$$

$$\text{REF(net)} = \text{SMP}^*(\text{gross}) - \text{SMP}(\text{gross}) = \text{SMP}^*(\text{net}) - \text{SMP}(\text{net}). \quad (8)$$

Here, SMP*(gross) is the spectrum obtained for the vial containing 1 ml of sample to which 1 ml of reference solution (for the contaminating radionuclide) is added.

If the lower limit of the region of interest in the high energy part of the spectrum is set well behind the edge of the spectrum of the radionuclide to be measured (NOI(net,high) = 0), then IMP(net,high) can be calculated in the same way:

$$\text{MP}(\text{net,high}) = \text{SMP}(\text{gross,high}) - \text{BGD}(\text{high}) = \text{SMP}(\text{net, high}). \quad (9)$$

The previous equations can subsequently be used to rebuild the spectrum of the contaminant in the lower energy range using the spectral shape of the reference spectrum and the calculated ratio K'(Q). Finally, the net spectrum of the nuclide of interest can be restored.

The spectrum of the reference vial to which the standard of the nuclide of interest was added can be used similarly to obtain the net spectrum of the standard. If the activity concentration is known, this spectrum can be used to calculate the counting efficiency in the proper region of interest and, consequently, to calculate the activity of the nuclide of interest present in the sample from the previously obtained net contribution. In contrast to methods based on quench calibration curves, the exact activity concentrations of the reference solutions for the different contaminants do not have to be known.

The validity of the obtained results can be monitored by fitting the net spectrum of the nuclide of interest to its reference spectrum. Both spectra are normalized and the residual term (calculated as the difference between the normalized net spectrum and reference spectrum on a channel to channel base) is plotted. A good fit is characterized by a symmetrical difference plot (located at zero-level). Figure 2 shows examples of a good fit (upper part) and an unreliable fit (lower part). This plot also shows the statistical uncertainty on the contribution of the sample spectrum (3 σ on the observed

number of counts per channel). Accurate results are obtained if the difference function (residual) lies within the $\pm 3\sigma$ range.

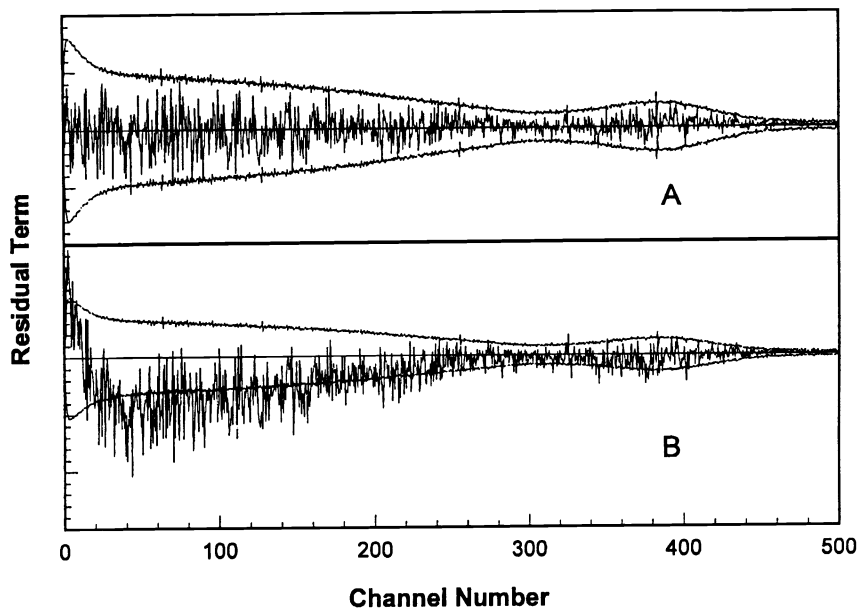


Fig. 2. Residual plots as an indication of the validity of the fit. A: valid fit; B: invalid fit.

RESULTS AND DISCUSSION

Table 1 summarizes the results for a typical case (^{63}Ni samples contaminated with ^{137}Cs). The ratio of the activity assayed by the present method to the theoretical ^{63}Ni activity present in the sample is given in the last three columns. The stated uncertainties for the ^{63}Ni recovery are the overall (propagated) counting errors, calculated from the number of counts observed in the spectra of BGD, Sample and Reference vials for the 95.5% confidence level (2σ), but not taking into account the uncertainty on the standard. For example: the first sample in run C contained 2.8 Bq of ^{63}Ni (radioelement to be assayed) and an equal amount of ^{137}Cs (contaminant). In the case where 5.5 Bq of ^{63}Ni standard solution was used as internal standard and 5.5 Bq of ^{137}Cs solution for the reference spectrum, the activity of ^{63}Ni in the sample was calculated to be 2.74 ± 0.14 Bq ($98 \pm 5\%$ recovery). Taking into account the uncertainty on the ^{63}Ni standard ($\pm 7\%$) the overall uncertainty on the analysis would be 2.74 ± 0.24 Bq ($98 \pm 9\%$ recovery).

Values are given for each experiment where 5.5, 55 or 550 Bq of the ^{63}Ni standard or the ^{137}Cs reference solution were added to the sample. The quench level is constant for every given set of vials. The numerical data of runs C, I and K indicate that good results can be obtained, even when the contaminant is present in a 10- or 100-fold excess, provided that the activity of the reference added is greater than the activity of the contaminant present. If the activity of the reference added is insufficient, no valid fit could be established.

The amount of added standard (^{63}Ni) obviously is also important, as indicated by the results of runs Q and S. Best results are obtained if the added amount of standard activity is at least equal to the activity of the nuclide of interest in the sample solution.

TABLE 1. ^{63}Ni recovery in ^{137}Cs contaminated samples. Effect of sample composition, contamination level and quenching level.

Run	Sample composition		Quench level (tSIE)	^{63}Ni standard added (Bq)	Percent ^{63}Ni recovery		
	^{63}Ni (Bq)	^{137}Cs (Bq)			5.5 Bq ^{137}Cs standard added	55 Bq ^{137}Cs standard added	550 Bq ^{137}Cs standard added
C	2.8	2.8	494 ± 1	5.5	98 ± 5	99 ± 4	99 ± 4
	2.8	2.8	494 ± 1	55	99 ± 4	99 ± 1	99 ± 4
	2.8	2.8	493 ± 2	550	98 ± 5	98 ± 5	98 ± 5
I	2.8	28	491 ± 2	5.5	*	100 ± 5	100 ± 4
	2.8	28	490 ± 1	55	*	102 ± 6	102 ± 4
	2.8	28	490 ± 1	550	*	104 ± 6	101 ± 8
K	2.8	280	488 ± 3	5.5	*	*	99 ± 16
	2.8	280	488 ± 3	55	*	*	107 ± 18
	2.8	280	488 ± 3	550	*	*	106 ± 18
Q	28	2.8	490 ± 2	5.5	100 ± 1	96 ± 1	100 ± 1
	28	2.8	489 ± 2	55	101 ± 1	97 ± 1	101 ± 1
	28	2.8	489 ± 2	550	99 ± 1	96 ± 1	99 ± 1
S	280	2.8	486 ± 3	5.5	*	*	*
	280	2.8	486 ± 3	55	104 ± 0.4	104 ± 0.4	104 ± 0.4
	280	2.8	487 ± 3	550	101 ± 0.4	101 ± 0.4	101 ± 0.4
M	2.8	2.8	254 ± 3	5.5	98 ± 5	99 ± 4	98 ± 4
	2.8	2.8	255 ± 3	55	97 ± 5	98 ± 4	97 ± 4
	2.8	2.8	255 ± 3	550	96 ± 5	98 ± 4	96 ± 4
E	2.8	2.8	148 ± 0.8	5.5	98 ± 11	99 ± 9	97 ± 9
	2.8	2.8	148 ± 0.8	55	99 ± 11	100 ± 9	98 ± 9
	2.8	2.8	148 ± 0.8	550	99 ± 12	100 ± 9	99 ± 9
O	2.8	2.8	68 ± 0.5	5.5	104 ± 24	99 ± 19	100 ± 18
	2.8	2.8	68 ± 0.5	55	106 ± 24	101 ± 19	102 ± 19
	2.8	2.8	68 ± 0.5	550	104 ± 24	99 ± 19	100 ± 18

*No valid fit possible

The results of runs M, E and O illustrate the ruggedness of the present method toward quenching. Even at high quenching levels (tSIE = 68 corresponding to 3% (v/v) CCl_4 in the sample) excellent results were obtained. Table 2 summarizes the data for a similar case (^{63}Ni contaminated with ^{60}Co). As indicated by runs J, L and T, the amount of added reference or standard activity must be at least equal to the activity present in the sample to obtain satisfactory results. Preferably, a tenfold excess should be maintained.

In a subsequent experiment, the present method was used to assay a ^{99}Tc sample contaminated with ^{60}Co . Because of the small difference in energy between the primary β particles ($E_{\text{max}} = 294$ keV for ^{99}Tc (Martin 1986b) and $E_{\text{max}} = 318$ keV for the 99.87% β particle of ^{60}Co (Martin 1986a)), the spectral overlap between ^{99}Tc and ^{60}Co is high (Fig. 3). In the present study the high energy part of the ^{60}Co spectrum (generated by the 0.12% β particle with $E_{\text{max}} = 1488$ keV) was used to fit the lower energy part of the same spectrum. Table 3 gives the results of this assay. Although the uncertainty on the experimentally obtained values is high, the agreement with the theoretical values is good.

TABLE 2. ^{63}Ni recovery in ^{60}Co contaminated samples. Effect of sample composition, contamination level and quenching level.

Run	Sample composition		Quench Level (tSIE)	^{63}Ni standard added (Bq)	Percent ^{63}Ni recovery		
	^{63}Ni (Bq)	^{60}Co (Bq)			5.5 Bq ^{60}Co standard added	55 Bq ^{60}Co standard added	550 Bq ^{60}Co standard added
	D	2.8			2.8	495 \pm 2	5.5
	2.8	2.8	496 \pm 1	55	103 \pm 8	101 \pm 7	100 \pm 7
	2.8	2.8	496 \pm 1	550	99 \pm 7	100 \pm 7	99 \pm 7
J	2.8	28	490 \pm 1	5.5	*	100 \pm 15	99 \pm 11
	2.8	28	490 \pm 1	55	*	103 \pm 15	102 \pm 11
	2.8	28	490 \pm 1	550	*	103 \pm 16	102 \pm 12
L	2.8	280	491 \pm 2	5.5	*	*	96 \pm 35
	2.8	280	490 \pm 2	55	*	*	97 \pm 36
	2.8	280	491 \pm 2	550	*	*	98 \pm 36
R	28	2.8	489 \pm 2	5.5	97 \pm 1	100 \pm 1	97 \pm 1
	28	2.8	489 \pm 2	55	102 \pm 1	103 \pm 1	101 \pm 1
	28	2.8	489 \pm 2	550	100 \pm 1	103 \pm 1	100 \pm 1
T	280	2.8	485 \pm 2	5.5	94 \pm 2	94 \pm 1	94 \pm 1
	280	2.8	486 \pm 3	55	100 \pm 0.4	100 \pm 0.4	100 \pm 0.4
	280	2.8	488 \pm 2	550	100 \pm 0.4	99 \pm 0.4	99 \pm 0.2
N	2.8	2.8	257 \pm 5	5.5	99 \pm 4	99 \pm 3	99 \pm 3
	2.8	2.8	257 \pm 5	55	101 \pm 4	101 \pm 3	101 \pm 3
	2.8	2.8	256 \pm 5	550	101 \pm 4	100 \pm 3	100 \pm 3
F	2.8	2.8	148 \pm 0.5	5.5	100 \pm 10	99 \pm 8	99 \pm 8
	2.8	2.8	148 \pm 0.8	55	99 \pm 10	98 \pm 8	98 \pm 8
	2.8	2.8	148 \pm 0.8	550	101 \pm 10	99 \pm 8	98 \pm 8
P	2.8	2.8	69 \pm 0.5	5.5	97 \pm 9	101 \pm 7	104 \pm 7
	2.8	2.8	69 \pm 0.5	55	99 \pm 9	102 \pm 7	105 \pm 7
	2.8	2.8	69 \pm 0.9	550	96 \pm 9	100 \pm 7	103 \pm 7

*No valid fit possible

TABLE 3. ^{99}Tc recovery in ^{60}Co contaminated samples. Effect of sample composition and contamination level.

Run	Sample composition		Quench level (tSIE)	^{99}Tc standard added (Bq)	Percent ^{99}Tc recovery		
	^{99}Tc (Bq)	^{60}Co (Bq)			5.5 Bq ^{60}Co standard added	55 Bq ^{60}Co standard added	550 Bq ^{60}Co standard added
	G	2.8			2.8	490 \pm 2	5.5
	2.8	2.8	490 \pm 2	55	103 \pm 19	103 \pm 17	105 \pm 15
	2.8	2.8	490 \pm 2	550	102 \pm 19	102 \pm 17	105 \pm 15
H	2.8	28	487 \pm 2	5.5	*	102 \pm 40	98 \pm 30
	2.8	28	487 \pm 2	55	*	107 \pm 42	99 \pm 32
	2.8	28	488 \pm 2	550	*	108 \pm 43	99 \pm 32

*No valid fit possible

This method was tested on a triple-labeled sample (^{63}Ni sample, contaminated with both ^{60}Co and ^{137}Cs). Table 4 lists the results for this experiment. As in previous experiments, satisfactory results were obtained, even with excess contaminant.

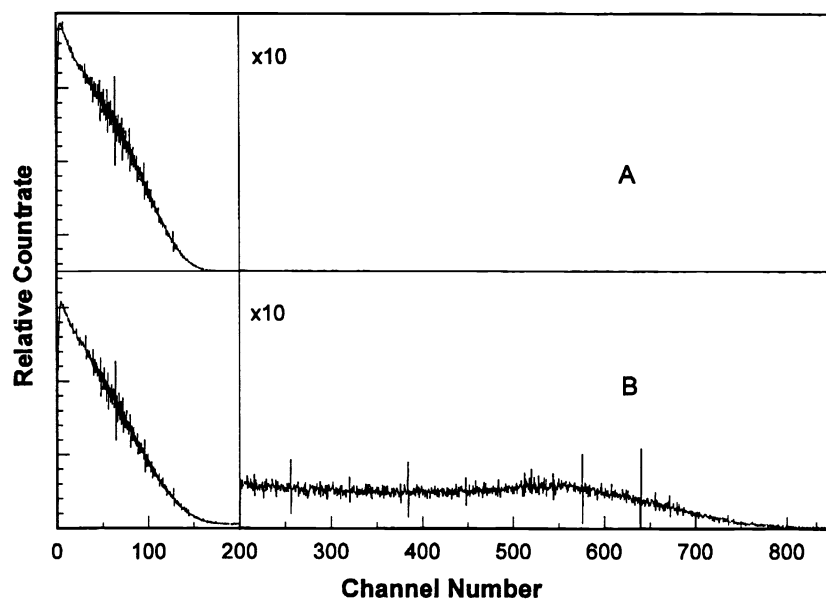


Fig. 3. β spectra of ^{99}Tc (A) and ^{60}Co (B) showing the high degree of spectral overlap in the low energy part of the spectrum. The right side (y-axis expanded by a factor of 10) shows the 0.12% β -particle intensity in the ^{60}Co spectrum, distinguishing it from the ^{99}Tc spectral shape.

TABLE 4. ^{63}Ni recovery in ^{60}Co and ^{137}Cs contaminated samples. Effect of sample composition and contamination level.

Run	^{63}Ni (Bq)	^{60}Co (Bq)	^{137}Cs (Bq)	Quench level (tSIE)	^{63}Ni standard added (Bq)	Percent ^{63}Ni recovery		
						^{60}Co and ^{137}Cs reference added (Bq each) 5.5	^{60}Co and ^{137}Cs reference added (Bq each) 55	^{60}Co and ^{137}Cs reference added (Bq each) 550
A	2.8	1.38	1.38	494 ± 2.3	5.5	105 ± 6	105 ± 5	105 ± 5
	2.8	1.38	1.38	493 ± 2.3	55	102 ± 6	102 ± 5	102 ± 5
	2.8	1.38	1.38	493 ± 2.3	550	102 ± 6	102 ± 5	102 ± 5
B	0.92	4.14	4.14	493 ± 2.2	5.5	105 ± 44	102 ± 31	102 ± 30
	0.92	4.14	4.14	493 ± 2.0	55	104 ± 44	101 ± 30	101 ± 29
	0.92	4.14	4.14	493 ± 2.6	550	105 ± 44	102 ± 31	102 ± 30

CONCLUSION

The method presented in this paper can be successfully applied to the radioassay of low-energy β activity in samples contaminated with (multiple) high-energy β impurities, provided that the qualitative radionuclide composition is known and a background sample with similar quench behavior is available. We obtained good results, even when the amount of impurity present largely exceeded the activity of the nuclide of interest or with highly quenched samples. The method offers considerable advantages over existing double-window or spectrum-unfolding techniques, as no quench correction curves are needed and only the activity concentration of the standard solution (the nuclide of interest) needs to be known. The activity concentration of the reference solutions (one for every contaminant) is not important, provided the activity added to the reference vials is at least equal but preferably higher than the activity of the impurity present in the sample.

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