Simultaneous determination of ^{152}Eu and ^{241}Am in liquid solution by liquid scintillation counting

Xiao-gui Feng · Qian-ge He · Jian-chen Wang · Jing Chen

Abstract ^{152}Eu and ^{241}Am are the most frequently used radiotracers in the separation studies on trivalent minor actinides and lanthanides. In almost all those studies, the determination of ^{152}Eu and ^{241}Am has been based on measuring their γ radiation by using a NaI(Tl) scintillation detector and/or a germanium detector. In this study, based on measuring the β particles and mono-energy electrons from ^{152}Eu and the α particles from ^{241}Am, we provide a new option to simultaneously determine the radioactivities of ^{152}Eu and ^{241}Am by liquid scintillation counting (LSC) with the aid of α/β discrimination. If the count rate ratio of ^{241}Am to ^{152}Eu is within the range of 100:1–1:100, the radioactivities of ^{152}Eu and ^{241}Am in mixed samples can be simultaneously determined by LSC with the errors less than 5 %. In addition, the interferences of ^{241}Am on Eu are divided into two parts: inside and outside the ^{241}Am region of interest. Only if the count rate ratio of ^{241}Am to Eu is more than 10:1, should the latter interference be in consideration.

Keywords ^{152}Eu · ^{241}Am · Liquid scintillation counting · α/β discrimination · Quench

Introduction

The separation of long-lived trivalent minor actinides (An(III), mainly Am(III) and Cm(III)) from lanthanides (Ln(III)) is a key step in nuclear fuel cycle, either for transmutation of minor actinides, or for the volume reduction of α radioactive waste. Many researchers have reported that the ligands containing soft donor atoms such as nitrogen and sulfur are most promising to be used for separation of An(III) from Ln(III) [1–12]. In all those studies, ^{152}Eu and ^{241}Am were chosen as the representative radiotracers of Ln(III) and An(III), and their γ activities were measured by a NaI(Tl) scintillation detector when only one radiotracer was used [1–9] or a germanium detector (Ge(Li) or HPGe) when radiotracers mixture was used [7–12]. It is well known that a germanium detector is very good for the detection of mixed γ-emitters, because of its high energy resolution. However, it is still necessary to provide other options, especially in one of the following cases: (1) when a laboratory has no germanium detectors, but has liquid scintillation counting (LSC) analyzer with the function of pulse shape analysis (PSA); (2) when a germanium detector is not equipped with automatic sample changer, which now is common for many laboratories; and (3) when the γ activity of a sample is so low that the counting will be too time-consuming. In this study, we try to determine ^{152}Eu and ^{241}Am simultaneously using LSC instead of a germanium detector, based on the following considerations: (1) ^{152}Eu and ^{241}Am can be discriminated with the technique of PSA which now is commonly equipped for LSC; (2) the counting efficiencies of LSC are reported as 82.6 % for ^{152}Eu [13] and nearly 100 % for ^{241}Am [14–17] in low quenched conditions, which are much higher than those of germanium detectors; and (3) nowadays almost every LSC has already equipped with automatic sample changer, which can make overnight and weekend hours as productive counting hours and lead to significant increase in the production of radioactive measurement work.

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Experimental

Equipment

An ultra-low background liquid scintillation analyzer (Quantulus 1220) from Perkin Elmer has been used for the measurements. It is provided with a function of PSA which discriminates α from β radiations and directs them separately into α- or β-multi-channel analyzers (MCA). The discrimination results vary with the PSA parameter, which can be set between 1 and 256. Quenching can be monitored with the aid of the external standard's Compton electron spectrum end point (SQP(E)). The SQP(E) is defined mathematically as the channel number of the external standard spectrum, above which 1 % of total intensity of the spectrum is found. The external standard spectrum is derived from the subtraction of the two spectra obtained from a two-step process: two successive measurements (counting time 1:1) of the sample with or without the external standard in the counting chamber.

Materials

Cocktail Ultima Gold AB (Perkin Elmer) and 20 mL polyethylene vials (Perkin Elmer) were employed for LSC measurement. ^{241}Am in 0.5 mol L^{-1} HNO_3 (radiopurity >99.9 %) was provided by China Institute of Atomic Energy. ^{152}Eu in 0.5 mol L^{-1} HCl was purchased from Eckert & Ziegler, with specific activity of 3.7×10^7 Bq mL^{-1} and radioactive impurities of ^{154}Eu (0.979 %) and ^{153}Gd (8.21 %) on November 1, 2011. For the convenience of description, in the next text we use ^{152}Eu to refer pure ^{152}Eu only, and Eu to stand for the mixture of ^{152}Eu, ^{154}Eu, and ^{153}Gd. Because both ^{154}Eu and ^{153}Gd are not α emitters, their radiations, i.e., β particles from ^{154}Eu and mono-energy electrons from ^{154}Eu and ^{153}Gd will be directed into β-MCA together with those of ^{152}Eu, as long as a proper PSA is set. If the total count rates of ^{152}Eu, ^{154}Eu, and ^{153}Gd are assigned to Eu, the difference between the misclassification ratios of α/β discrimination for the pair of ^{241}Am and Eu and those for the pair of ^{241}Am and ^{152}Eu, the count rate of which is equal to that of Eu, will be neglected. This means the results from Eu and ^{241}Am can be also applied to simultaneously determine ^{152}Eu and ^{241}Am.

The quenching agent employed for this study was the simulated solution (referred as SimS) which reflects the quenching effect of acid and salt and represents the main composition of Chinese high level liquid waste. SimS was a mixture of 1.0 mol L^{-1} HNO_3 and metallic ions, the concentrations of which were 18.3, 6.0, 5.7, 2.9 and 1.5 g L^{-1} for Na, Fe, Al, Ni, and Nd.

Method

All the samples were measured with α/β discrimination. The optimum PSA parameter and misclassification ratio at the optimum were determined by the generation of cross-over plot. In order to obtain the optimum PSA parameter at different quench levels, eight runs of measurement were carried out. Four vials (six vials for the first run, see Quenching effect and α/β discrimination), each containing 10 μL ^{241}Am or Eu solution mixed with 10 mL cocktail, were quenched with SimS, by adding 100 μL aliquots at first, and then 200 μL each time from the third run to the eighth run. After each addition, the vials were counted for 10 min at a series of preset PSA parameters.

With the known optimum PSA parameters, six unmixed samples (each sampling volume of which was 10 μL, Table 1) and five mixed samples (Table 2) were counted for 60 min. Afterwards, we repeated the addition of SimS and measurement as described above.

All reported uncertainties correspond to $\pm 1\sigma$. The relative standard deviation of counting rate for any single counting is calculated from $(rt)^{-1/2} \times 100$ %, where r is the count rate, and t is the counting time which is considered as a constant. For all multiple independent measurements, the standard deviation of mean value is calculated with the STDEV.S function of Excel 2010 (Microsoft), as shown in Table 3; Figs. 1 and 4. In addition, error propagation is in consideration for the derived quantities such as the misclassification ratio.

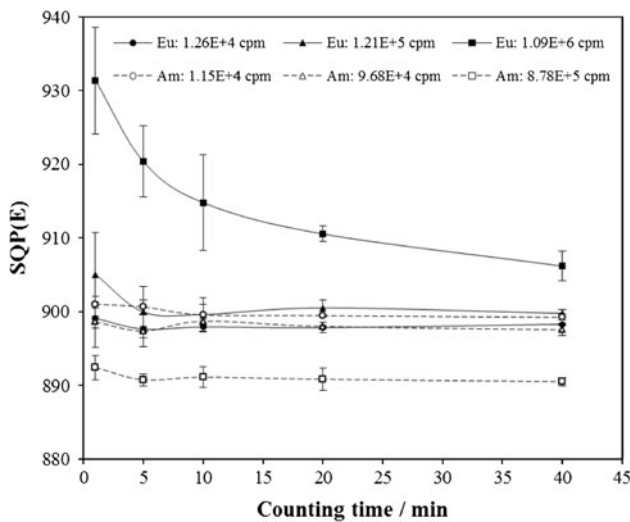Table 1 Six unmixed solutions with different specific activities

Solution ID	Specific activity/Bq mL^{-1}
Eu-H	1.51×10^5
Eu-M	1.46×10^4
Eu-L	1.43×10^3
Am-H	1.19×10^5
Am-M	1.20×10^4
Am-L	1.21×10^3

Table 2 Five mixed samples with different activities

Sample ID	Preparation details	Radioactivity/Bq	
		Eu	^{241}Am
MIX1	10 μL Eu-L + 10 μL Am-H	14.3	1.19×10^3
MIX2	10 μL Eu-L + 10 μL Am-M	14.3	120
MIX3	10 μL Eu-L + 10 μL Am-L	14.3	12.1
MIX4	10 μL Eu-M + 10 μL Am-L	146	12.1
MIX5	10 μL Eu-H + 10 μL Am-L	1.51×10^3	12.1

Table 3 Effects of anticoincidence shield on background and count rates of Eu

Guard detector	Sample ID	Count rate (Channels 1–1,024)/cpm			SQP(E)
		β-MCA	α-MCA	β-MCA + α-MCA	
Active	Blank-1	5.33	0.73	6.06	898.3
	Blank-2	5.86	0.75	6.61	900.6
	Blank-3	4.73	0.53	5.26	897.8
	Blank-average	5.31 ± 0.56	0.67 ± 0.12	5.98 ± 0.68	898.9 ± 1.5
	Eu-1	1.287×10^5	1.034×10^4	1.390×10^5	898.2
	Eu-2	1.184×10^4	8.363×10^2	1.268×10^4	896.7
	Eu-3	1.133×10^5	7.917×10^3	1.212×10^5	901.4
Inactive	Blank-1	23.58	1.80	25.39	897.5
	Blank-2	23.11	2.20	25.31	900.5
	Blank-3	22.88	1.79	24.67	901.4
	Blank-average	23.19 ± 0.36	1.93 ± 0.23	25.12 ± 0.39	899.8 ± 2.0
	Eu-1	1.679×10^5	1.377×10^4	1.817×10^5	902.3
	Eu-2	1.540×10^4	1.105×10^3	1.651×10^4	896.4
	Eu-3	1.481×10^5	1.045×10^4	1.585×10^5	900.4

Fig. 1 The effect of counting time on SQP(E)s for Eu or ^{241}Am samples with different count rates

Fig. 2 Misclassification versus PSA for Eu and ^{241}Am in 10 mL cocktail and 0 μL SimS

Results and discussion

Anticoincidence shield and background

Quantulus 1220 is equipped with anticoincidence shield (or called guard detector). As shown in Table 3, when the anticoincidence shield is inactive, the average background is 23.19 cpm in β-MCA and 1.93 cpm in α-MCA; while when the anticoincidence shield is active, the average backgrounds reduce to 5.31 cpm in β-MCA and 0.67 cpm in α-MCA. The anticoincidence shield not only reduces the backgrounds, but also has different influences on the count rates of different radionuclides [18]. The influences of

anticoincidence shield on the count rates can be neglected for α-emitters, β-emitters and β/γ emitters without β/γ cascade radiations. However, remarkable decreases of count rates are observed when anticoincidence shield is applied for β/γ emitters with β/γ cascade radiations. As for ^{152}Eu, there are some decay branches with β/γ cascade radiations [13, 19], therefore three Eu samples were measured to check the influences of anticoincidence shield. When the anticoincidence shield is set from inactive to active, the count rates in β-MCA + α-MCA of the three Eu samples decrease with almost the same ratio, which is 0.766 ± 0.002 (counts with guard active divided by counts

with guard inactive). Even though the counting efficiencies of Eu decrease 23.4 %, in order to improve the figure of merit, we still chose to set the anticoincidence shield as active in most cases.

## The error of SQP(E) from statistical fluctuations

As the most important factor responsible for reduction in counting efficiency of a given sample/cocktail mixture, quenching must be considered for LSC. For Quantulus 1220, quenching is quantified with the parameter SQP(E); when quenching increases in the sample, the SQP(E) decreases. In the case of low level counting, for which Quantulus 1220 is designed specifically, the influence of statistical fluctuations in the two-step process (refer to Equipment) on the SQP(E) can be ignored; but for high level counting, the influence should be in consideration [20].

We tested a series of Eu or ^{241}Am samples which were prepared with different count rates but to almost the same extent of quenching. Since the statistical fluctuations will decrease with the increase of counting time, the counting time in the two-step process was set ranging from $1 + 1$ min (default) to $40 + 40$ min (Fig. 1). Error bars in Fig. 1 were derived from three repeated measurements for all six samples. Figure 1 shows that the influence of the statistical fluctuations varies with the count rate of the sample. When the count rate is less than 1.5×10^5 cpm, the SQP(E) is approaching to 898.7 ± 1.0 with the increase of the counting time, and the difference between the SQP(E) obtained within $10 + 10$ min and that within $40 + 40$ min can be ignored; but when the count rate is close to 1×10^6 cpm, the SQP(E) is significantly higher (for Eu) or lower (for ^{241}Am) than 898.7 ± 1.0, which implies the SQP(E) may has large uncertainty if the count rate of the sample is too high and the default counting time $(1 + 1$ min) is applied. As a result, in this study most samples were prepared deliberately with count rates less than 1.5×10^5 cpm, and the counting time in the two-step process was set as $10 + 10$ min.

The sampling volume of each sample in Fig. 1 is 10 μL. The SQP(E)s of the four samples with count rates less than 1.5×10^5 cpm are 898.7 ± 1.0, which are almost the same as those of blank samples listed in Table 3. This verifies that 10 μL Eu or ^{241}Am has little influence on the SQP(E).

Quenching effect and α/β discrimination

The six samples used in Fig. 1 were also measured to obtain the optimum PSA parameter when the quenching agent SimS was absent. The results are shown in Fig. 2. The misclassification ratio is defined as: for α emitters, count rates in β-MCA divided by count rates in α-MCA + β-MCA; for β emitters, count rates in α-MCA (here the counting window is
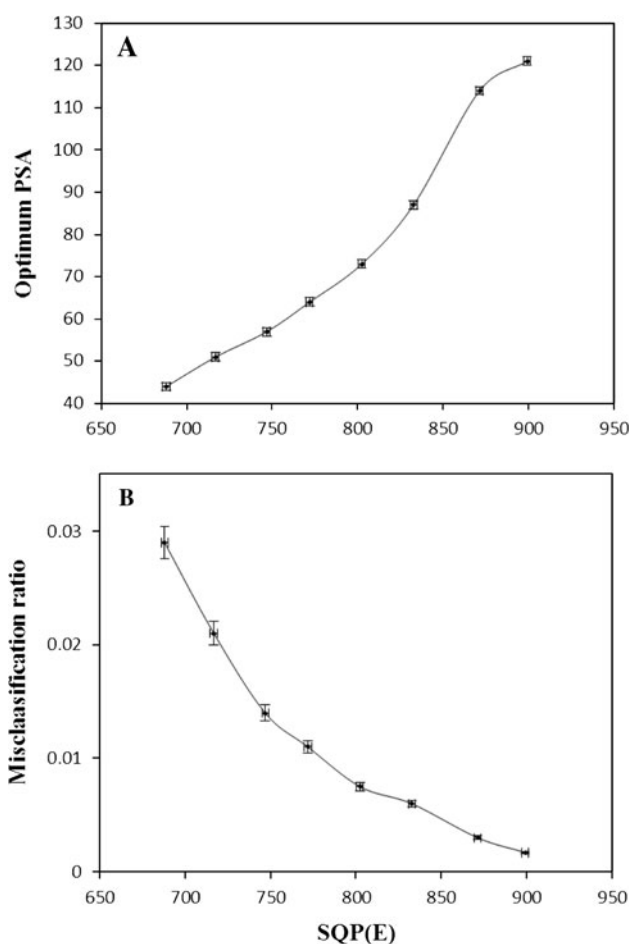

Fig. 3 Changes in optimum PSA (**A**) and misclassification ratio (**B**) over a range of SQP(E) for the pair of Eu and ^{241}Am

only the region of interest (ROI) for the α peak) divided by count rates in α-MCA + β-MCA. The large uncertainties, two of which are too large to be completely displayed in Fig. 3, are come from the low misclassified count rates of the sample "Eu: 1.26 E + 4 cpm" at PSA = 160–180, which are very close to the background in the corresponding ROI.

The optimum PSA is chosen at the crossover point where the misclassification ratio of α event equals that of β event. When the count rate is less than 1.5×10^5 cpm, the crossover point is at almost the same place; but when the count rate is close to 1×10^6 cpm, the crossover point is at an obviously higher place, meaning a higher misclassification ratio. In order to avoid too high misclassification ratios and inaccurate SQP(E)s as described in The error of SQP(E) from statistical fluctuations, it is advisable to prepare LSC samples less than 1.5×10^5 cpm.

In order to study the effect of quenching on α/β discrimination, the four vials, count rates of which were less than 1.5×10^5 cpm, were added with quenching agent SimS and measured by LSC step by step. For each quench
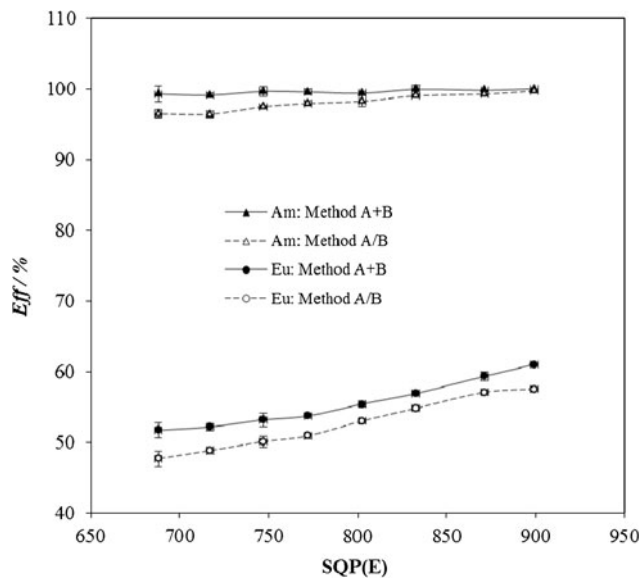
Fig. 4 Effects of SQP(E) on the counting efficiencies of Eu or ^{241}Am

level, the results are similar to those in Fig. 2. The cross-over point is at almost the same place for the two Eu samples and the two ^{241}Am samples, meaning the count rates of Eu and ^{241}Am, if they are less than 1.5×10^5 cpm, have little effect on the optimum PSA and the misclassification ratio. Therefore, the average of optimum PSAs and misclassification ratios at each quench level are shown in Fig. 3. As expected, with the volume of SimS (V_{SimS}) increasing from 0 to 1,300 μL (accordingly the SQP(E) is getting from 899 to 688), the optimum PSA decreases from 121 to 44, and the misclassification ratio increases from 0.0017 to 0.029.

Counting efficiency

Both quench level and α/β discrimination have important effect on the counting efficiency. With the known optimum PSA parameters as shown in Fig. 3, six unmixed samples (each sampling volume of which was 10 μL, Table 1) were counted to obtain counting efficiencies of Eu and ^{241}Am at different quench levels. Figure 4 shows the average counting efficiencies of three Eu samples and three ^{241}Am samples with two data processing methods. "Method A + B" means the counts both in β-MCA and α-MCA are summed to make the total counts for pure α or β emitters; while "Method A/B" means only counts in β-MCA are accepted to make the counts for Eu or those in α-MCA for ^{241}Am. When the SQP(E) is getting from 899 to 688, the counting efficiencies of ^{241}Am (with Method A + B) keep at nearly a constant of 100 %, which is in good agreement with reported results [14–17]. But for the other three cases as shown in Fig. 4, the counting efficiencies decrease with increase of quench level.

The counting efficiencies shown in Fig. 4 were calculated from the specific activities of the six unmixed samples listed in Table 1. The specific activities of Am-H, Am-M, and Am-L are very easy to be determined by measuring 10 μL each sample with LSC [14–17]. As for Eu-H, Eu-M, and Eu-L, we prepared a diluted Eu sample (referred as Eu-ref), which had the same quench level as the six unmixed samples listed in Table 1, to obtain the apparent counting efficiency of Eu as follows.

The preparation of Eu-ref includes two steps: (1) diluting 100 μL original Eu solution with 10 mL 0.5 mol L^{-1} HNO$_3$ to make 10.1 mL stock solution; (2) taking 10 μL stock solution into a vial and adding 10 mL cocktail to make the LSC sample, Eu-ref. Eu-ref was measured using LSC with the anticoincidence shield inactive, and the count rate was $181,656 \pm 135$ cpm on May 5, 2012. The activity of Eu-ref can be figured out with the consideration of dilution factor and half-life correction, based on the above preparation process and the following data: (1) on November 1, 2011, the specific activity of ^{152}Eu was 3.7×10^7 Bq mL^{-1}; and the radioactive impurities were 0.979 % ^{154}Eu and 8.21 % ^{153}Gd; (2) the half-lives of ^{152}Eu, ^{154}Eu, and ^{153}Gd are 13.522 years [19], 8.601 years [21], and 240.4 days [22]. The total activity of Eu-ref on May 5, 2012 was calculated as 3,801 Bq, including 3,569 Bq ^{152}Eu; and the radioactive impurities were 0.997 % ^{154}Eu and 5.10 % ^{153}Gd.

The apparent counting efficiency of Eu by LSC is $181,656 \div 60 \div 3,801 \times 100 \% = 79.7 \%$, the uncertainty of which is about 3.5 %, mainly from the original Eu solution specific activity (3 %) and volume measurement (1 %, three times). With the consideration of the uncertainty and the differences between this study and [13], including quench level, LSC setting, and radioactive impurities, it is acceptable that the apparent counting efficiency of 79.7 % is a little less than 82.6 % [13].

The apparent counting efficiency of 79.7 % is obtained with the anticoincidence shield inactive and the quench level of Eu-ref close to those of blank samples. If the anticoincidence shield is set from inactive to active, the apparent counting efficiency will reduce to $79.7 \times 0.766 \% = 61.1 \%$. When SimS is absent, the three Eu samples have the same quench level as Eu-ref, therefore their average counting efficiency is the same as 61.1 % (Fig. 4). From this counting efficiency and the count rates of the three Eu samples when SimS is absent, the specific activities of Eu-H, Eu-M and Eu-L can be figured out, which is listed in Table 1.

Measurement of mixed samples

All the above experimental results are obtained from unmixed samples. In order to check the feasibility of this study, we also prepared five mixed samples with different

ratios of ^{152}Eu to ^{241}Am (Table 2). The five samples were measured at different quench levels by gradually adding SimS as described in Method. For the convenience of comparison, all the results are expressed as relative error *Err* (Tables 4, 5, 6, 7), which is calculated from Eq. (1):

$$Err = \frac{\frac{r}{60 \cdot Eff} - A}{A} \times 100\,\% \tag{1}$$

where *r* is the count rate in the ROI of ^{152}Eu in β-MCA or of ^{241}Am in α-MCA (cpm); *A* is the radioactivity of Eu or ^{241}Am listed in Table 2 (Bq); and *Eff* is the counting efficiency obtained by linear interpolation at the corresponding SQP(E) on the dashed line as shown in Fig. 4.

Listed in Table 4 are relative errors for the five mixed samples with no overlap correction, just by simple integration between the limits of ROIs. Both the errors for ^{241}Am in MIX1, MIX2, and MIX3 and those for Eu in MIX3, MIX4, and MIX5 are less than 5 %, indicating that no overlap correction is needed if the count rates of ^{241}Am and Eu are in the same order of magnitude, or the count rate of interested nuclide is major in the mixture. On the contrary, if the count rate of interested nuclide is minor in the mixture, i.e., the Eu in MIX1 and MIX2 or the ^{241}Am in MIX4, and MIX5, the errors are getting larger and larger with the increase of quench level or the count rates of uninterested nuclides. On the other hand, the interference of ^{241}Am on Eu is greater than that of Eu on ^{241}Am. Except for Eu of MIX2 in the absence of SimS, all the errors of Eu in MIX1 and MIX2 are greater than 5 %. While the errors of ^{241}Am in MIX4 and MIX5 are less than 5 % in much wider range: SQP(E) > 802 (V_{SimS} < 500 µL) for MIX4 and SQP(E) > 871 (V_{SimS} < 100 µL) for MIX5. This means: if the count rate of ^{241}Am is one order of magnitude higher than that of Eu, overlap correction is usually needed for Eu; while if the highest count rate of Eu is not two orders of magnitude higher than that of ^{241}Am, overlap

correction is not needed when the SQP(E) is greater than 871 (V_{SimS} < 100µL).

For the minor Eu and ^{241}Am, the errors of which are greater than 5 %, we made overlap correction between the lower and upper limits of the ^{241}Am ROI with a linear interpolation at first (Table 5). Table 5 shows after linear correction the errors are significantly less than those listed in Table 4. When the highest count rate of major nuclide is not one order of magnitude higher than that of minor nuclide, all the errors are less than 5 %. However, for Eu in MIX1, all the errors remain at (16.9 ± 0.8 %) no matter how many SimS are included. This is mainly because the Compton electrons continuum resulting from the 59.54 keV γ radiations of ^{241}Am (between channels 50 and 400, and close to the horizontal axis as shown in Fig. 5) are overlapped with the spectrum of Eu, which can be subtracted by Eq. (2):

$$A_{Am/Eu} = \frac{A_{Am} \cdot R_{mis} \cdot (1 - k_{ROI})}{Eff_{Eu}} \tag{2}$$

where $A_{Am/Eu}$ is the part which should be subtracted from the Eu radioactivity (Bq), A_{Am} is the ^{241}Am radioactivity of the sample (Bq), R_{mis} is the misclassification ratio as shown in Fig. 3, k_{ROI} is the ratio of counts in the ^{241}Am ROI to counts in all channels of β-MCA, and Eff_{Eu} is the counting efficiency of Eu as shown by the dashed line in Fig. 4. Because the quench level has little effect on $A_{Am/Eu}$, the least quenched 10 µL Am-H sample (V_{SimS} = 0 µL) is used to calculate $A_{Am/Eu}$ in this study. For Eu in MIX1, $A_{Am} = 1{,}190$ Bq, $R_{mis} = 0.0017$, $k_{ROI} = 0.196$, $Eff_{Eu} = 61.1$ %, therefore $A_{Am/Eu} = 2.66$ Bq. With the subtraction of 2.66 Bq from the Eu radioactivity, the errors of Eu in MIX1 are reduced to less than 5 % as listed in Table 6.

As for ^{241}Am in MIX5, the low error range (<5 %) is extended from SQP(E) > 871 (V_{SimS} < 100 µL) to SQP(E) > 802 (V_{SimS} < 500 µL). When the SQP(E) is

Table 4 Relative errors for five mixed samples with no overlap correction

Sample ID	Nuclide	*Err* (%) vs. V_{SimS} (µL)							
		0	100	300	500	700	900	1,100	1,300
MIX1	^{241}Am	−1.6	−2.1	−1.1	0.0	0.3	0.1	0.4	0.5
	Eu	18.2	51.0	103.9	135.0	213.2	298.5	400.5	398.9
MIX2	^{241}Am	−3.1	−3.3	−2.7	−1.3	−1.1	−1.1	−0.6	−0.8
	Eu	2.4	5.8	8.3	12.4	22.1	29.5	39.0	40.6
MIX3	^{241}Am	−1.9	−3.1	−1.3	−0.4	0.8	0.9	2.2	1.4
	Eu	1.7	1.3	1.7	2.3	3.9	3.5	4.3	4.6
MIX4	^{241}Am	−1.8	−1.1	0.9	4.4	8.7	9.9	14.9	19.3
	Eu	1.0	0.6	−1.0	−0.8	0.8	0.1	0.9	−0.3
MIX5	^{241}Am	1.9	4.3	23.2	51.8	88.3	108.6	144.7	194.4
	Eu	2.6	2.2	0.1	0.3	2.2	1.3	1.7	0.8

Table 5 Relative errors for minor nuclide in four mixed samples with linear correction

Sample ID	Nuclide	Err (%) vs. V_{SimS} (µL)							
		0	100	300	500	700	900	1,100	1,300
MIX1	Eu	17.0	18.2	18.1	16.1	16.4	16.4	16.3	16.9
MIX2	Eu		2.9	−0.7	0.6	1.1	0.0	−1.7	−0.1
MIX4	^{241}Am					−1.5	0.1	1.9	0.2
MIX5	^{241}Am			−3.2	0.2	7.7	14.3	24.4	29.1

Table 6 Relative errors for Eu in MIX1 with linear correction and subtraction of $A_{Am/Eu}$

Sample ID	Nuclide	Err (%) vs. V_{SimS} (µL)							
		0	100	300	500	700	900	1,100	1,300
MIX1	Eu	−1.7	−0.5	−0.5	−2.5	−2.3	−2.2	−2.3	−1.7

Table 7 Relative errors for ^{241}Am in MIX5 with parabolic correction

| Sample ID	Nuclide	Err (%) vs. V_{SimS} (µL)			
		700	900	1,100	1,300
MIX5	^{241}Am	0.8	5.5	4.7	3.5

lower than 802 ($V_{SimS} > 500$ µL), the errors are still greater than 5 %, which needs more complex correction.

Figure 6 shows the effect of the misclassified Eu on the count rate of ^{241}Am at different quench levels. When the quench level is low, for example, $V_{SimS} = 100$ µL, a linear interpolation or even no correction can give sufficient accuracy. While when the quench level is high, for example, $V_{SimS} = 1,300$ µL, the continuum of Eu under the ^{241}Am peak is like a parabola, which means parabolic correction is necessary. Take line 3 in Fig. 6 as an example to show how to make the parabolic correction. Together with the lower and upper limits of the ^{241}Am ROI (channels 460 and 650), one more channel is chosen from 30 to 50 channels at the right of the upper limit (channel 700) to obtain a fitted quadratic polynomial, which is used to calculate the continuum of Eu between the channels 460–650. By subtracting the count rate of the Eu continuum from
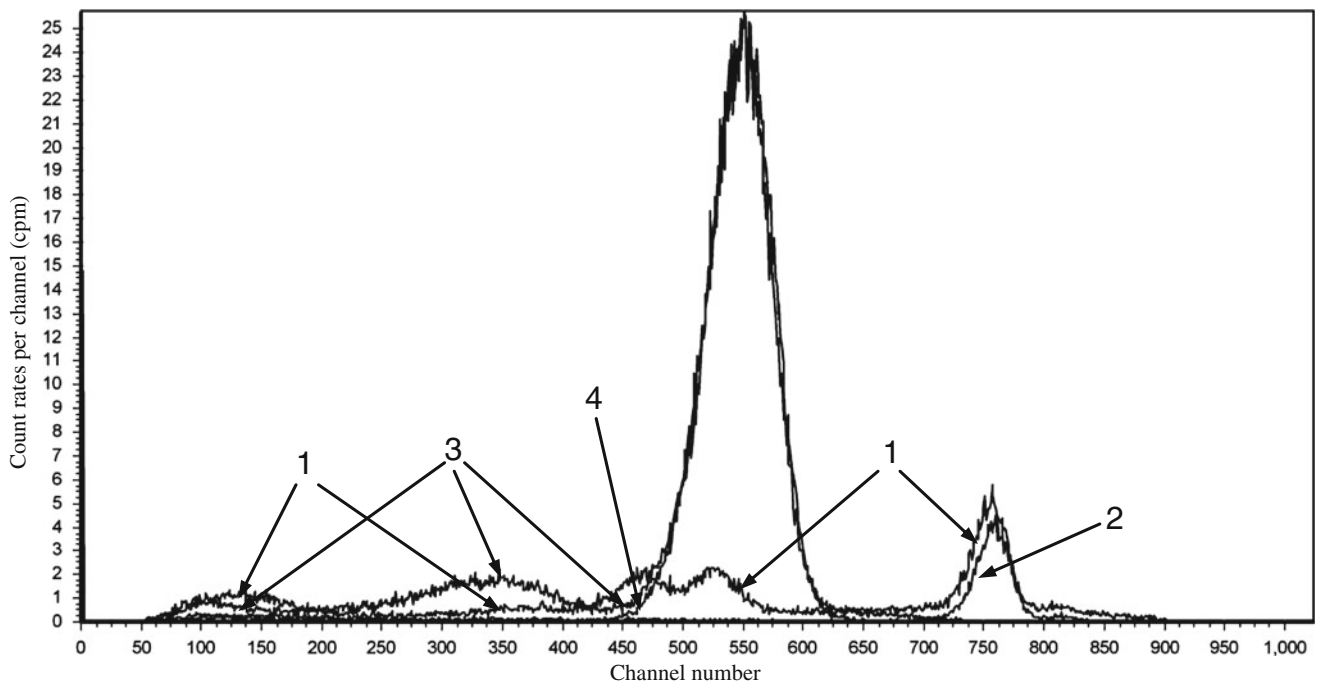
Fig. 5 LSC spectra of MIX1 and 10 µL Am-H in β-MCA at different quench levels *1* MIX1/100 µL SimS, *2* 10 µL Am-H/100 µL SimS, *3* MIX1/1,300 µL SimS, *4* 10 µL Am-H/1,300 µL SimS
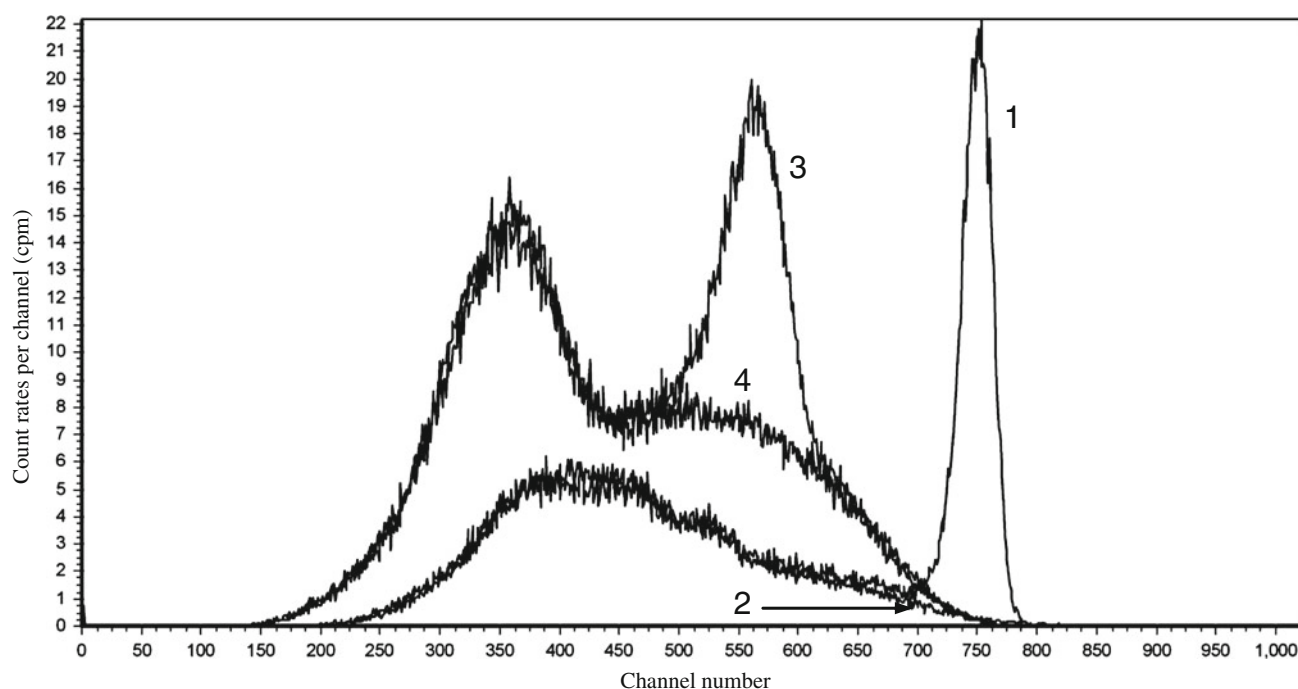
Fig. 6 LSC spectra of MIX5 and 10 μL Eu-H in α-MCA at different quench levels *1* MIX5/100 μL SimS, *2* 10 μL Eu-H/100 μL SimS, *3* MIX5/1,300 μL SimS, *4* 10 μL Eu-H/1,300 μL SimS

overall count rate in the ROI, the net count rate of ^{241}Am is obtained, which is used in Eq. (1) to calculate the relative error. The errors in Table 7 are obtained after parabolic correction, which are lower than those in Table 5, with the biggest one of 5.5 %.

Generally speaking, if the count rate ratio of ^{241}Am to Eu is ranging from 100:1 to 1:100, both ^{241}Am and Eu can be simultaneously determined by LSC with the errors less than 5 % with or without overlap correction. The interferences of ^{241}Am on Eu are divided into two parts: inside and outside the ^{241}Am ROI. If the count rate ratio of ^{241}Am to Eu is less than 10:1, the latter interference can be neglected; otherwise, both interferences should be eliminated.

Conclusions

Based on the counting efficiencies of Eu or ^{241}Am at different quench levels, which obtained from unmixed Eu or ^{241}Am samples, the radioactivities of Eu and ^{241}Am in mixed samples can be simultaneously determined by LSC with the errors less than 5 %, as long as the count rate ratio of ^{241}Am to Eu is within the range of 100:1 to 1:100. If the quench level increases, obvious overlap will be found in the LSC spectrum. Depending on the shape of the spectrum, linear or parabolic correction can be used to decrease the errors. In addition, the interferences of ^{241}Am on Eu are divided into two parts: inside and

outside the ^{241}Am ROI. Only if the count rate ratio of ^{241}Am to Eu is more than 10:1, should the latter interference be in consideration.

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