LIQUID SCINTILLATION STANDARDIZATION OF ^{133}Ba

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ABSTRACT. We present three parametric coincidence and anticoincidence methods using liquid scintillation counting (LSC) for measuring the electron capture (EC) radionuclide ^{133}Ba. We observed different slopes of the extrapolation curves for three energy windows of the gamma spectrum. We used an LS spectrometer in coincidence and anticoincidence to accumulate X-ray and Auger electron spectra. The results of ^{133}Ba activity measurements for the three parameters are presented and uncertainties are discussed.

INTRODUCTION

The electron capture and gamma emitter ^{133}Ba is important in γ spectrometry. However, ^{133}Ba is difficult to measure, as the decay comprises X-rays and Auger electrons of various energies, and exhibits a γ-ray spectrum with a high degree of γ-ray conversion. This yields conversion electrons of low energies. Figure 1 illustrates the decay scheme of ^{133}Ba (Lederer 1967). Here, we present the 4 π(x,e)-γ coincidence and anticoincidence method as a continuation of earlier work (Chylinski 1971; Chylinski & Radoszewski 1975). Application of LS spectrometry assured the highest possible counting efficiency for γ-rays and electrons. The three parameters, as three parts of the γ-ray spectrum, minimize extrapolation uncertainties. In addition, application of the anticoincidence method confirmed our results.

METHODS

The counting equipment comprises a four-fold detector and a circuit developed earlier and applied to ^{22}Na measurement (Chylinski & Radoszewski 1992). The four-fold detector consists of 2 opposed photomultipliers for X-ray and electron counting and 2 NaI(Tl) detectors for γ-ray counting. The electronic circuit contains a two-stage coincidence system. The first stage coincidence unit forms the (x,e) channel. The second stage unit is formed by the output of the (x,e) channel and two γ-scintillation counters working in the summed mode for the γ channel. The radioactive solution was composed of 25 mg Ba as $BaCl_2$ in 1 ml of 1 M HCl. "Atomlight" liquid scintillator (New England Nuclear) was used for sample preparation. The counting sample consisted of 0.5 ml of 0.04 M EDTA solution mixed with an aliqout of ^{133}Ba solution and with 10 ml of liquid scintillator. We use the following notation for the equations of 4π(LS)-γ coincidence counting:

N_o	decay rate in the sample
N_1	counting rate in the LS channel
N_2	counting rate in the γ channel
N_C	coincidence count rate
N_{AC}	anticoincidence counting rate
e_1	counting efficiency of Auger electrons and X-rays
e_2	counting efficiency of γ-rays in γ-channel
$e_{1\gamma}$	counting efficiency of γ-rays, γ-ray conversion electrons and Compton electrons in the LS channel
$e_{\gamma\gamma}$	counting efficiency for "second-type coincidences" caused by the γ-ray (when Compton effect or γ-ray conversion effect in γ-ray cascade occur; this means Compton electron or conversion electron in LS) channel and the scatter or the second γ-ray in the γ channel.

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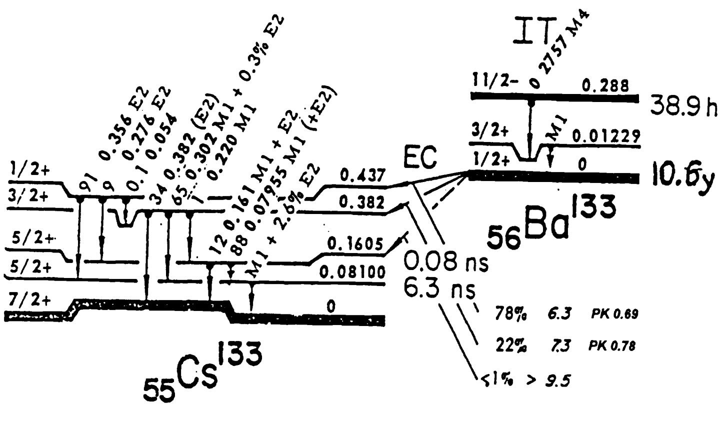

Fig. 1. Decay scheme of ^{133}Ba (Lederer 1967)

With this notation, the following relations can be presented

$$N_1 = N_o\left[\varepsilon_1 + (1-\varepsilon_1)\,\varepsilon_{1\gamma}\right] \tag{1}$$

$$N_2 = N_o \varepsilon_2 \tag{2}$$

$$N_c = \left[\varepsilon_1 \varepsilon_2 + (1-\varepsilon_1)\varepsilon_{\gamma\gamma}\right] \ . \tag{3}$$

Transformation of equations (1), (2) and (3) yields

$$\frac{N_1 N_2}{N_c} = N_o \left(\frac{1 + \dfrac{1-\varepsilon_1}{\varepsilon_1}\,\varepsilon_{1\gamma}}{1 + \dfrac{1-\varepsilon_1}{\varepsilon_1}\,\dfrac{\varepsilon_{\gamma\gamma}}{\varepsilon_2}} \right) \tag{4}$$

which can be transformed by dividing the expression in brackets as a polynomial division

$$\frac{N_1 N_2}{N_c} = N_o \left[1 + \frac{1-\varepsilon_1}{\varepsilon_1}\left(\varepsilon_{1\gamma} - \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2}\right) + \left(\frac{1-\varepsilon_1}{\varepsilon_1}\right)^2 \left[\left(\frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2}\right)^2 - \varepsilon_{1\gamma}\frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2}\right] + \ldots \right] \ . \tag{5}$$

This is sufficient for analysis, as further expressions also have the expression $(1 - \varepsilon_1)/\varepsilon_1$.

After extrapolation of $\varepsilon_1 \rightarrow 1$, which means that $(1 - \varepsilon_1)/\varepsilon_1 \rightarrow 0$, the basic relation is

$$N_o = \frac{N_1 N_2}{N_c} \tag{6}$$

and for the anticoincidence method

$$N_o = \frac{N_1 N_2}{N_2 - N_{AC}} \quad . \tag{7}$$

We may denote

$$\varepsilon_{1\gamma} - \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2} = \Phi$$

which designates the slope of the extrapolation curve of N_o to $\varepsilon_1 = 1$, or $(1 - \varepsilon_1)/\varepsilon_1 = 0$. In most cases, the slope, Φ, is positive, but in some cases, where $\varepsilon_{\gamma\gamma}/\varepsilon_2 > \varepsilon_{1\gamma}$, the slope can be negative. We obtained different slopes for ^{133}Ba measurement when different windows of the γ-ray spectrum were counted in the γ channel.

RESULTS

First, we chose three windows of the γ-ray spectrum (Fig. 2) as parameters: 275–500 keV, 100–500 keV and 100–275 keV. We chose the lower 100 keV threshold to avoid the inclusion of the X-ray sum peak. To choose the proper range of extrapolation, which we performed by lowering the high voltage in the LS channel, the anode differential characteristic was taken (Fig. 3). Figure 4 shows the results of extrapolation, for the ranges mentioned above, for a given source. The different slope of the extrapolation curves (from positive parameters 1 and 2, to negative parameter 3) can be explained only by a different value of

$$\Phi = \varepsilon_\gamma - \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2} \quad .$$

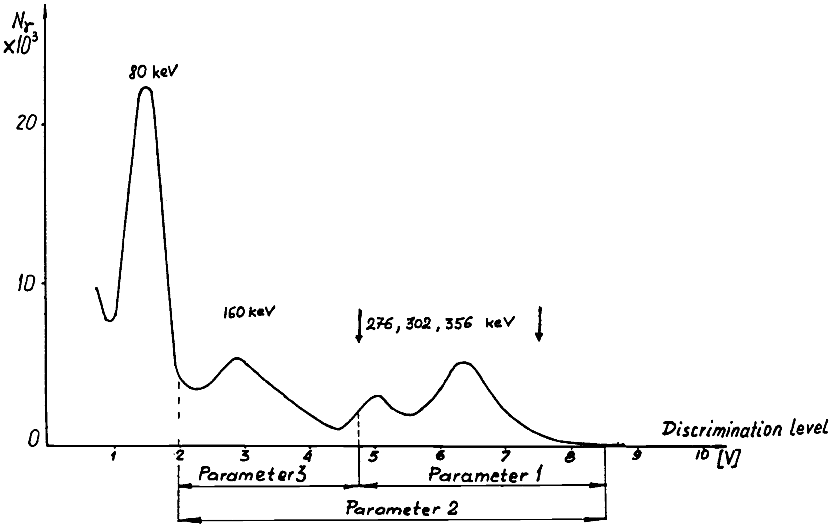

Fig. 2. γ spectrum of ^{133}Ba for parameter 1 (275–500 keV), parameter 2 (100–500 keV) and parameter 3 (100–275 keV)

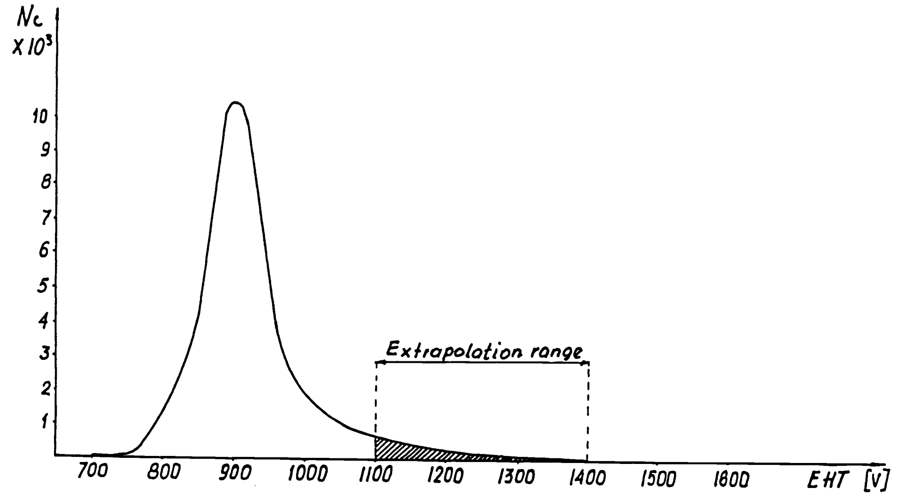

Fig. 3. The differential anode characteristic in LSC for ^{133}Ba

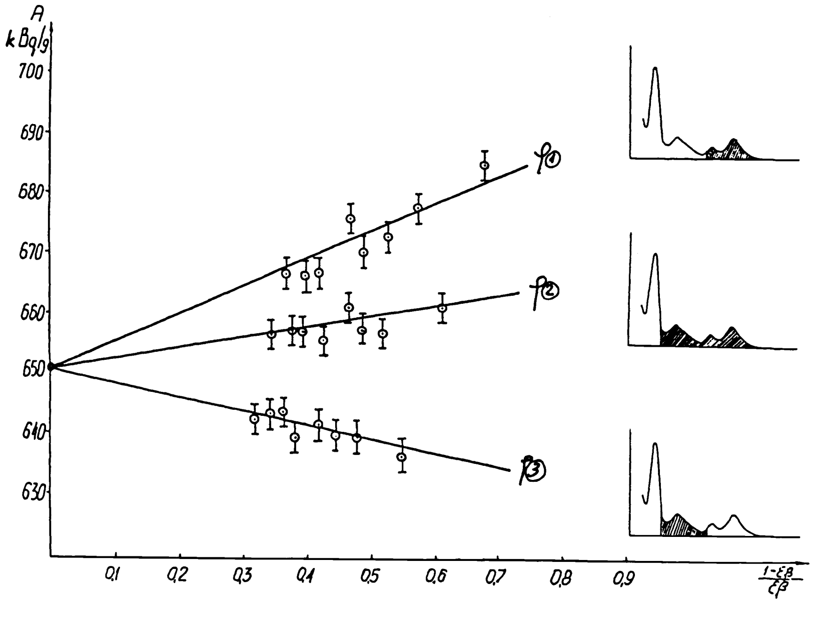

Fig. 4. Extrapolation curves for a single source of ^{133}Ba for the three parameters

That is, for a positive slope, the value is

$$\varepsilon_{1\gamma} > \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2}$$

and for a negative slope

$$\varepsilon_{1\gamma} < \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_2} \quad .$$

Figure 4 also shows that the counting points start earlier in parameter 3 than in the others. That is because the counting efficiency in the LS channel (ε_1) in our calculation is defined as N_c/N_γ. This depends partly on the second-type coincidences ($\varepsilon_{\gamma\gamma}$). Therefore, the highest value of N_c/N_γ is obtained for a negative slope of the extrapolation curve. Table 1 presents the counting results. We tabulate activities obtained for six sources counted in coincidence and anticoincidence for each parameter; counting uncertainties are also given.

TABLE 1. ^{133}Ba Activities for Coincidence and Anticoincidence Methods and Three Parameters

					Activity (kBq g^{-1})	
No.	Mass (mg)	ε_β	ε_γ	Φ	Coincidence	Anticoincidence
Parameter 1						
1	9.212	0.731	0.0525	0.0952	637.249	636.277
2	9.212	0.733	0.0531	0.0817	656.175	655.321
3	18.441	0.725	0.0528	0.0818	644.806	641.658
4	8.844	0.732	0.0528	0.0632	652.638	652.158
5	18.398	0.724	0.0531	0.0856	647.214	643.250
6	8.650	0.732	0.0530	0.0813	647.281	645.528
					647.560	645.698
A'= 646.629 kBq g^{-1}	$S_x = 0.29\%$	$t_\alpha S_x = 1.16\%$				
Parameter 2						
1	9.212	0.742	0.0960	0.0336	641.263	639.889
2	9.212	0.741	0.0962	0.0365	657.392	653.970
3	18.441	0.736	0.0963	0.0336	646.259	643.998
4	8.844	0.741	0.0959	0.0342	644.994	643.372
5	18.398	0.739	0.0961	0.0301	650.171	647.206
6	8.650	0.742	0.0959	0.0306	648.867	647.845
					648.158	646.047
A'= 647.102 kBq g^{-1}	$S_x = 0.22\%$	$t_\alpha S_x = 0.89\%$				
Parameter 3						
1	9.212	0.762	0.0432	-0.0110	635.479	636.380
2	9.212	0.763	0.0431	-0.0240	655.416	657.724
3	18.441	0.756	0.0428	-0.0418	649.514	650.370
4	8.844	0.763	0.0431	-0.0366	649.982	649.653
5	18.398	0.758	0.0431	-0.0342	652.056	654.760
6	8.650	0.759	0.0428	-0.0561	655.637	656.466
					649.681	650.892
A'= 650.287 kBq g^{-1}	$S_x = 0.32\%$	$t_\alpha S_x = 1.30\%$				

TABLE 2. Comparison of Results for Three Parameters

Parameter	A_o [kBq g^{-1}]	Standard error (%)	Maximum Δ (dispersion) (%)
1	646.6	0.29	0.57
2	647.1	0.22	0.57
3	650.3	0.32	0.57

Table 2 compares the results for each parameter. The coincidence result for the three parameters is

$$A_{OC} = 648.5 \text{ kBq g}^{-1}, \text{ with } S_x = 0.23\% .$$

The anticoincidence result for the three parameters is

$$A_{OAC} = 647.5 \text{ kBq g}^{-1}, \text{ with } S_x = 0.24\% .$$

The mean values for coincidence and anticoincidence for the three parameters are

$$\begin{aligned} A_O &= 648.0 \text{ kBq g}^{-1} \\ S_x &= 0.16\% \\ t_\alpha S_x &= 0.66\% \\ \sigma_s &= 0.11\% . \end{aligned}$$

The overall uncertainty is

$$\sigma_t = [(t_\alpha S_x)^2 + \sigma_s]^{½} = 0.67\%$$

where: S_x = standard error
$t_\alpha S_x$ = standard error for confidence level 0.99
σ_s = systematic error
σ_t = overall uncertainty of the radioactive concentration.

CONCLUSION

The three parametric coincidence and anticoincidence methods for radioactive concentration measurement for ^{133}Ba ensure a credible method of measurement, which is especially useful for standard solution determination. Six results can be obtained simultaneously for a given source. This approach decreases counting uncertainty. It should be emphasized that the method has general application to any β-γ, EC-γ, or even α-β emitters with any type of γ-ray spectrum (especially when more than one γ line is present). It is also noteworthy that the influence of γ-rays in a liquid scintillation detector is usually higher than that in a 4π proportional counter ($\varepsilon_{\beta\gamma}$, $\varepsilon_{\gamma\gamma}$). Thus, this influence and the slope of the discrimination curves must be considered in LS spectrometry.

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II. BIOSCIENCE APPLICATIONS