

## CHAPTER 47

# Calculational Method for the Resolution of $^{90}\text{Sr}$ and $^{89}\text{Sr}$ Counts from Cerenkov and Liquid Scintillation Counting\*

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### INTRODUCTION

Environmental samples taken around reactor facilities potentially contain both 50.4 day half-life strontium-89 ( $^{89}\text{Sr}$ ) and 29 year half-life strontium-90 ( $^{90}\text{Sr}$ ). The health risk associated with exposure to  $^{90}\text{Sr}$  is considered to be a factor of 20 greater than that associated with  $^{89}\text{Sr}$  due to its longer half-life and its energetic daughter yttrium-90 ( $^{90}\text{Y}$ ). It is therefore often important to distinguish between these two isotopes when monitoring for health and environment protection. Most common methods for determining both  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  are based on time allowed ingrowth of  $^{90}\text{Y}$  after the total strontium is chemically purified and counted. This usually requires from 7 to 14 days to allow sufficient  $^{90}\text{Y}$  ingrowth for adequate statistics.

A method has been developed that determines both  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  without requiring an ingrowth period for  $^{90}\text{Y}$ ; therefore, the results can be obtained much sooner. The method is similar to that developed more than a decade ago by Randolph<sup>1</sup> which uses sequential Cerenkov and liquid scintillation counting after chemical purification. A new calculational method has been employed, however, which corrects for the fraction of the Cerenkov counts due to  $^{90}\text{Sr}$  and the fraction of both counts due to the early ingrowth of  $^{90}\text{Y}$ . The method does not rely on the spectral resolution of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ , and  $^{90}\text{Y}$  through the use of channel ratios, as has previously been required with the combined liquid scintillation of a Cerenkov technique. Therefore, higher detection efficiencies are obtained which result in better sensitivity.

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**Table 1. Nuclide Characteristics**

Nuclide	<sup>89</sup> Sr	<sup>90</sup> Sr	<sup>90</sup> Y
Half-life	50 d	29 Y	64 h
Max. beta energy (keV)	1491	546	2284
Avg. beta energy (keV)	583	196	934
Rel. health risk	1	20	—
M.P.C. (pCi/L) drinking water	80	8	—

## THEORY

Cerenkov radiation is produced when a charged particle passes through a transparent medium at a velocity greater than the speed of light in that medium. A threshold condition must be met which is defined by

$$vn/c = 1, \quad (1)$$

where  $v$  is the velocity of the particle,  $c$  is the speed of light, and  $n$  is the refractive index of the transparent medium. Furthermore, the Cerenkov photon yield increases proportionally as the energy of the particle increases above the threshold.

The nuclear decay characteristics for <sup>89</sup>Sr, <sup>90</sup>Sr, and <sup>90</sup>Y are shown in Table 1. The combined Cerenkov and liquid scintillation counting technique takes advantage of the large difference in average beta energies of <sup>89</sup>Sr and <sup>90</sup>Sr to discriminate between the two after the <sup>90</sup>Y is separated chemically. The threshold energy for the beta particle stimulation of Cerenkov radiation in water is 263 keV; therefore, in a modern liquid scintillation counter, Cerenkov counting provides over 40% efficiency for the 583 keV average beta energy <sup>89</sup>Sr, while providing less than 1% counting efficiency for the 196 keV average beta energy <sup>90</sup>Sr. The total strontium activity is determined by liquid scintillation counting, which provides over 90% counting efficiency for both isotopes. The combined information obtained from these two counts yields the activity of each isotope.

## OPERATIONAL METHOD

### Chemical Separation

A standard procedure, similar to that used in EPA Method 905.0,<sup>2</sup> is used to chemically purify the sample. Calcium and magnesium are removed by precipitation of the strontium as the nitrate. Barium and radium are removed by a chromate precipitation. Yttrium and other impurities are removed with hydroxide scavenging. A final strontium precipitation is made as the carbonate

**Table 2. Experimental Efficiencies and Backgrounds**

Nuclide	Cerenkov	Liquid Scint
	Eff. (%)	Eff. (%)
<sup>89</sup> Sr	40	95
<sup>90</sup> Sr	1	91
<sup>90</sup> Y	61	95
	(cpm)	(cpm)
<b>Background</b>	<b>15</b>	<b>62</b>

in a tared liquid scintillation vial. The precipitate is washed, dried, and weighed to calculate the recovery of the standardized strontium carrier. The average chemical recovery is approximately 80%.

### Counting

The precipitate is dissolved in 5 mL of 0.3 M HCl, and the aqueous solution is counted on a liquid scintillation counter without scintillator for 20 min for Cerenkov radiation, primarily from <sup>89</sup>Sr. Seventeen mL of "Atomlight" liquid scintillator is then added and mixed with the sample. The solution is again counted for 20 min on the liquid scintillation counter for the measurement of both <sup>89</sup>Sr and <sup>90</sup>Sr.

Randolph used optimized discriminator channel settings to provide energy windows for each count. He then used the count ratios in each widow to calculate factors for correcting the fraction of the Cerenkov counts due to <sup>90</sup>Sr and the fraction of both counts due to the early ingrowth of <sup>90</sup>Y. However, due to the severe energy spectrum overlap of each of the isotopes, the optimized windows only contain a fraction of the total counts from the isotope of interest. Therefore, the sensitivity is reduced.

In the present experiment, the novel calculational method is used to correct for the fraction of the Cerenkov counts due to <sup>90</sup>Sr and the fraction of both counts due to the early ingrowth of <sup>90</sup>Y. The discriminators were set for each count such that the energy window contained the entire peak of the isotope or isotopes of interest, thus improving the sensitivity of the method. Beckman LS-9800 liquid scintillation counters were used in this study. The experimental efficiencies for each isotope and the backgrounds obtained for each counting method are shown in Table 2.

### CALCULATIONAL METHOD

The Cerenkov counts are primarily from <sup>89</sup>Sr, with a small contribution from <sup>90</sup>Sr and the early ingrowth of <sup>90</sup>Y. The net Cerenkov count rate (cpm),  $C_1$ , is given by

$$C_1 = {}^{89}\text{Sr cpm}_c + {}^{90}\text{Sr cpm}_c + {}^{90}\text{Y cpm}_c, \quad (2)$$

where  $\text{cpm}_c$  represents the Cerenkov counts per minute obtained from each nuclide. The liquid scintillation counts are due primarily to both  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$ , with a small contribution from the early ingrowth of  $^{90}\text{Y}$ . The net liquid scintillation count rate (cpm),  $C_2$ , is given by

$$C_2 = {}^{89}\text{Sr cpm}_1 + {}^{90}\text{Sr cpm}_1 + {}^{90}\text{Y cpm}_1, \quad (3)$$

where  $\text{cpm}_1$  represents the liquid scintillation counts per minute obtained from each nuclide. The count rate due to early ingrowth of  $^{90}\text{Y}$  can be defined in terms of the  $^{90}\text{Sr}$  count rate by knowing the elapsed time from chemical separation of  $^{90}\text{Y}$  to counting. Equations 2 and 3 then become

$$C_1 = {}^{89}\text{Sr cpm}_c + ({}^{90}\text{Sr cpm}_c) (A_1) \quad (4)$$

and

$$C_2 = {}^{89}\text{Sr cpm}_1 + ({}^{90}\text{Sr cpm}_1) (A_2), \quad (5)$$

where

$$A_1 = 1 + (D_1)(1 - e^{-\lambda t_c}) \quad (6)$$

and

$$A_2 = 1 + (D_2)(1 - e^{-\lambda t_l}), \quad (7)$$

where  $D_1$  is the  $^{90}\text{Y}$  Cerenkov efficiency to  $^{90}\text{Sr}$  Cerenkov efficiency ratio,  $t_c$  is the time from chemical separation to Cerenkov counting,  $D_2$  is the  $^{90}\text{Y}$  liquid scintillation efficiency to  $^{90}\text{Sr}$  liquid scintillation efficiency ratio,  $t_l$  is the time from chemical separation to liquid scintillation counting, and  $\lambda$  is the  $^{90}\text{Y}$  decay constant.

The Cerenkov and liquid scintillation count rates for each nuclide are related by their counting efficiency ratio of each for the two counting methods such that

$${}^{89}\text{Sr cpm}_c = ({}^{89}\text{Sr cpm}_1) (B_1) \quad (8)$$

and

$${}^{90}\text{Sr cpm}_c = ({}^{90}\text{Sr cpm}_1) (B_2), \quad (9)$$

where  $B_1$  is the  $^{89}\text{Sr}$  Cerenkov efficiency to  $^{89}\text{Sr}$  liquid scintillation efficiency ratio and  $B_2$  is the  $^{90}\text{Sr}$  Cerenkov efficiency to  $^{90}\text{Sr}$  liquid scintillation efficiency ratio. By replacing these terms in Equation 4,

$$C_1 = ({}^{89}\text{Sr cpm}_1) (B_1) + ({}^{90}\text{Sr cpm}_1) (B_2) (A_1). \quad (10)$$

If the ratios of counting efficiencies are determined by counting pure standards, Equations 5 and 10 contain two unknowns and can be solved simultaneously to yield

**Table 3. Results from Measuring Mixtures of Isotopes (Lower Level)**

Sample/ Nuclide	Known (pCi/L)	Measured (pCi/L)		2 Sigma CL	Ratio
1 <sup>89</sup> Sr	120	130	+/-	11	1.08
1 <sup>90</sup> Sr	0	0.7	+/-	5	—
2 <sup>89</sup> Sr	120	125	+/-	15	1.04
2 <sup>90</sup> Sr	220	234	+/-	11	1.06
3 <sup>89</sup> Sr	240	219	+/-	16	0.91
3 <sup>90</sup> Sr	220	259	+/-	13	1.18
4 <sup>89</sup> Sr	120	130	+/-	19	1.08
4 <sup>90</sup> Sr	440	491	+/-	16	1.11
5 <sup>89</sup> Sr	0	8	+/-	12	—
5 <sup>90</sup> Sr	220	260	+/-	10	1.18

$${}^{90}\text{Sr cpm}_1 = [(B_1)(C_2) - C_1] / [(B_1)(A_2) - (B_2)(A_1)] \quad (11)$$

and

$${}^{89}\text{Sr cpm}_1 = C_2 - (A_2)({}^{90}\text{Sr cpm}_1). \quad (12)$$

The activity concentration of each nuclide can then be determined by Equations 11 and 12 and by

$${}^{90}\text{Sr (pCi/L)} = ({}^{90}\text{Sr cpm}_1) / [(E_1)(V)(R)(2.22)] \quad (13)$$

and

$${}^{89}\text{Sr (pCi/L)} = ({}^{89}\text{Sr cpm}_1) / [(E_2)(V)(R)(2.22)] \quad (14)$$

where  $E_1$  = <sup>90</sup>Sr liquid scintillation efficiency,  
 $E_2$  = <sup>89</sup>Sr liquid scintillation efficiency,  
 $V$  = Sample Volume (L), and  
 $R$  = Chemical Recovery.

## RESULTS

Known mixtures of <sup>89</sup>Sr and <sup>90</sup>Sr, ranging to 100% of each, have been measured by this technique at both low and moderate activity levels. The results are shown in Tables 3 and 4. While it appears from the lower level data that a slight positive bias may exist for <sup>90</sup>Sr, it is likely that the apparent bias is due to counting uncertainty since the bias disappears at the higher activity level. The error due to uncertainty in correction for the early ingrowth of <sup>90</sup>Y is minimized by counting the samples as soon as possible after separation.

Since this method requires two counts, the uncertainty must be considered when estimating the total measurement due to counting statistics. An estimate of the standard deviation,  $s$ , due to counting uncertainty can be made by

Table 4. Results from Measuring Mixtures of Isotopes (Higher Level)

Sample/ Nuclide	Known (pCi/L)	Measured (pCi/L)		2 Sigma CL	Ratio
6 <sup>89</sup> Sr	2044	2042	+/-	47	1.00
6 <sup>90</sup> Sr	0	2	+/-	41	—
7 <sup>89</sup> Sr	2044	2041	+/-	53	1.00
7 <sup>90</sup> Sr	2199	2045	+/-	44	0.93
8 <sup>89</sup> Sr	4088	3938	+/-	71	0.98
8 <sup>90</sup> Sr	2199	2145	+/-	60	0.98
9 <sup>89</sup> Sr	2044	2074	+/-	66	1.01
9 <sup>90</sup> Sr	4398	4371	+/-	52	0.99
10 <sup>89</sup> Sr	0	38	+/-	36	—
10 <sup>90</sup> Sr	2199	2177	+/-	26	0.99

$${}^{90}\text{Sr cpm, } s = \frac{\{[(B_1)^2(G_2/T_2 + F_2/T_2) + G_1/T_1 + F_1/T_1]\}}{[(B_1)(A_2) - (B_2)(A_1)]^2}^{1/2} \quad (15)$$

and

$${}^{89}\text{Sr cpm, } s = [(G_2/T_2 + F_2/T_2) + (A_2)^2({}^{90}\text{Sr cpm, } s)^2]^{1/2}, \quad (16)$$

where  $G_1$  = gross Cerenkov count rate (cpm),  
 $G_2$  = gross liquid scintillation count rate (cpm),  
 $F_1$  = background Cerenkov count rate (cpm),  
 $F_2$  = background liquid scintillation count rate (cpm),  
 $T_1$  = Cerenkov counting time (min), and  
 $T_2$  = liquid scintillation counting time (min)

when the background and sample counting times are equal. Similarly, the lower limit of detection (LLD), considering only counting statistics, can be estimated by

$$\text{LLD (cpm)} = 2.71/T_2 + 4.65(\text{sb}) \quad (17)$$

where sb is the standard deviation of the blank as defined by

$${}^{90}\text{Sr sb} = \frac{\{[(B_1)^2(F_2/T_2) + F_1/T_1]\}}{[(B_1)(A_2) - (B_2)(A_1)]^2}^{1/2} \quad (18)$$

and

$${}^{89}\text{Sr sb} = [F_2/T_2 + (A_2)^2({}^{90}\text{Sr sb})^2]^{1/2} \quad (19)$$

The liquid scintillation counters have a higher background than the low-background gas-proportional counters typically used for counting environmental strontium samples. This results in reduced sensitivity. However, the higher efficiencies obtained with liquid scintillation counters allow lower limits of detection which are only a factor of 2 to 3 higher than those obtained using

**Table 5. Calculated Lower Limits of Detection**

<b>Cerenkov and Liquid Scintillation Count</b>	
$^{90}\text{Sr}$	= 12.8 cpm = 7.9 pCi/L
$^{89}\text{Sr}$	= 15.9 cpm = 9.4 pCi/L
<b>Traditional 14-Day Two-Count Method</b>	
$^{90}\text{Sr}$	= 1.4 cpm = 2.4 pCi/L
$^{89}\text{Sr}$	= 2.0 cpm = 3.6 pCi/L

Note: All LLDs assume 20 min counts on 1 L samples and 80% chemical recovery.

the same count time on gas-proportional counters. The lower limits of detection obtained in this study are shown in Table 5 with the LLDs obtained with the traditional 14-day two-count method (assuming 1-L samples counted for 20 min and 80% chemical recovery for both). If low-background liquid scintillation counters are used, the lower limits of detection may be even lower.

## CONCLUSION

The described method provides a useful way to analyze both  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  without a 14 day wait for  $^{90}\text{Y}$  ingrowth. The use of the derived equations provides a correction for early  $^{90}\text{Y}$  ingrowth and the  $^{90}\text{Sr}$  Cerenkov counts, without the use of energy window ratios. Better sensitivity has been provided than has previously been possible with this technique.

The sensitivity of this method is adequate for monitoring plant effluents and many environmental level samples. Lower limits of detection, below the maximum contaminant concentration allowed for drinking water, can be achieved when a 1-L sample and a 20 min count time are used. If monitoring for lower levels of contamination are necessary, a larger sample or a longer count time may be used.

## REFERENCES

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