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Simultaneous Determination of Strontium-90 and Strontium-89 by Cerenkov-Radiation and Liquid-Scintillation Counting: a Re-Evaluation for Low-Level Counting

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^{89}Sr, ^{90}Sr *assay, simultaneous / Liquid-scintillation counting of* ^{89}Sr, ^{90}Sr, ^{90}Y / *Cerenkov-radiation counting of* ^{89}Sr, ^{90}Y

Abstract

Three methods for the simultaneous determination of ^{89}Sr and ^{90}Sr are compared under the aspect of low ^{90}Sr activities and high ^{89}Sr/^{90}Sr ratios as expected in nuclear emergency situations: (i) Liquid-scintillation counting with two pulse height windows, (ii) a combination of Cerenkov-radiation and liquid-scintillation counting with open windows, and (iii) the combination of Cerenkov-light and scintillation counting with two windows set during liquid-scintillation counting, the higher one adjusted to ^{90}Y only. The third method is accurate to ±20% for ^{89}Sr/^{90}Sr ratios up to 15 and at ^{90}Sr disintegration rates down to 0.15 Bq applying a total counting time of about six hours.

1. Introduction

The release of fresh fission products from the damaged Chernobyl reactor station during late April and early May of 1986 has drawn attention to the shortcomings the determination of one of the most hazardous components in fallout, ^{90}Sr with 28.5 y half-life. Since this nuclide decays exclusively by β^--ray emission its assay requires an elaborate chemical enrichment and purification procedure. In fission, ^{90}Sr is always accompanied by 50.5-d ^{89}Sr, again a pure β-ray emitter, which dominates over ^{90}Sr in fresh mixtures of fission products even after prolonged exposure in a reactor. Hence, high ^{89}Sr/^{90}Sr activity ratios are expected in case of a nuclear emergency, but the activity level in the strontium fraction may nevertheless be low at larger distances from the point of release. This is illustrated by fallout data in Germany after the Chernobyl accident: Measured ^{89}Sr/^{90}Sr ratios were 11 [1], 15 and 22 [2], and the ^{90}Sr activities in daily collected samples were, typically, only 8 mBq m^{-3} in air and 0.6 Bq l^{-1} in rain water in the Mainz area [2] where the fallout was at about the average level in Germany.

The standard procedure for distinguishing between the two strontium isotopes is rather simple but time-consuming: ^{90}Sr decays into radioactive 2.67-d ^{90}Y whereas ^{89}Sr has a stable daughter product. By counting the β-radiation of a freshly purified strontium sample the sum activity of both strontium isotopes is obtained. After a waiting period of several days, ingrown ^{90}Y is chemically separated and counted to determine the contribution of ^{90}Sr; the ^{89}Sr activity is obtained as the difference. Since ^{90}Y is also a pure β-ray emitter, its decay curve should be followed over several days to check its radiochemical purity. Hence, a reliable determination of the two strontium isotopes takes about two weeks. An additional problem is the presence of 12.8-d ^{140}Ba in fresh fission products which accompanies strontium through most steps of a chemical separation procedure. Incomplete elimination of this alkaline earth isotope would lead to an overestimation of the ^{90}Sr activity due to the appearance of the ingrowing daughter product, 1.68-d ^{140}La in the milked ^{90}Y sample. Although ^{140}Ba and ^{140}La can be identified by characteristic γ-ray transitions, this may not be possible at the low activity levels considered here.

A waiting time of about two weeks in the ^{89}Sr/^{90}Sr determination is not acceptable in an emergency situation, of course. Hence, a much faster method for the simultaneous determination of ^{89}Sr and ^{90}Sr in fresh fission products and at low concentration levels is urgently needed. One approach is to replace radioactivity measurements by methods based on other principles such as resonance ionization mass spectroscopy with lasers [3]. For radioactivity measurements one may utilize differences in the β^--ray energies to distinguish between the three nuclides: ^{90}Sr emits the softest spectrum with a maximum energy of E_{max} = 546 keV, ^{89}Sr has E_{max} = 1492 keV and ^{90}Y E_{max} = 2282 keV. It may be difficult, however, to obtain reliable results for the softest component if its contribution is small and the counting rate is low. Most attractive is the sequential counting of Cerenkov light and liquid scintillations introduced by Randolph [4] and by Buchtela and Tschurlovits [5]. In this approach, a detailed measurement and analysis of energy spectra is not required. Since the intensity of Cerenkov radiation increases strongly with β^--ray energy [6, 7], ^{90}Sr contributes little compared to the other two nuclides. In a liquid-scintillation counter, however, all three nuclides can be measured with comparable, high ef-

ficiency. In both detector systems the pulse height distributions for the three nuclides are significantly different. This can be utilized by setting pulse height channels for exclusive or preferential counting of individual components.

Randolph [4] uses strontium samples freshly purified from ^{90}Y and counts the Cerenkov radiation with a window setting that covers ^{89}Sr and ^{90}Sr but discriminates the high-energy part of the ^{90}Y spectrum. Then, a scintillator solution is added and the sample is recounted in two pulse-height windows, a lower one covering ^{90}Sr, ^{89}Sr plus ^{90}Y, and a higher one for ^{89}Sr plus ^{90}Y. From this, the ^{90}Sr activity is deduced which is used to correct the Cerenkov radiation rate for contributions of ^{90}Sr/^{90}Y to obtain the ^{89}Sr activity. The method was shown [4] to give reliable results over a wide range of ^{89}Sr/^{90}Sr ratios, between 20:1 and 1:20, at a level of about 120 Bq for the minor component. Ingrowing ^{90}Y did not interfere during the first 16 hours after purification. By extrapolation of counting efficiencies and background rates the author claims that low activity levels as mentioned in the Introduction could be handled but no experimental proof is given.

In the method of Buchtela and Tschurlovits [5] a purification from ^{90}Y is not required. Liquid-scintillation counting with a single, wide window comprises all three nuclides whereas two Cerenkov-light countings, one without and one with a wavelength shifter, are used to differentiate between ^{89}Sr and ^{90}Y. No experimental data on the reliability of the method were presented. As shown by Carmon [6] ^{89}Sr can be estimated in the presence of a large excess of ^{90}Sr by following with a Cerenkov-radiation detector the ^{90}Y activity ingrowing in a freshly purified strontium sample but this is not the situation we are mainly concerned with.

Following Randolph's approach [4] with some modifications we have studied the applicability of the combined Cerenkov-light and liquid-scintillation counting to low activity levels and high ^{89}Sr/^{90}Sr ratios using a commercial liquid-scintillation spectrometer. For comparison we have also made measurements with liquid-scintillation counting only using two different counting channels for the two strontium isotopes.

2. Experimental

A liquid-scintillation spectrometer LS 7800 from Beckman Instruments was used for Cerenkov-radiation and liquid-scintillation counting.

2.1. Two-channel liquid-scintillation counting

The scintillation cocktail "Ready Safe" from Beckman Instruments was used because it was found to be least sensitive against quenching by hydrochloric acid solutions among several commercially available scintillation cocktails. Calibrated solutions (to ±2.5%) of ^{89}Sr and of ^{90}Sr purified from ^{90}Y, 0.1 ml in 0.1 N HCl, were pipetted into high-density, low-pressure polyethylene vials and filled up to 18 ml volume with "Ready Safe". Counting was started one hour after sample preparation to avoid problems with luminescence and electrostatic charging. Optimum windows were set by the spectrometer to channels 0 to 700 for ^{90}Sr and 700 to 1000 for ^{89}Sr at the resulting quench level, as is depicted in Figure 1. Corrections for quenching were made with the automatic quench compensation of the spectrometer using the Compton-edge method [8]. Here the "H-number" indicates the shift in the position of the Compton edge produced by ^{137}Cs γ-quanta in the presence of a quenching substance as compared to an unquenched sample; large H-numbers indicate strong quenching effects. Quenching curves for ^{89}Sr and ^{90}Sr with chloroform as quencher were taken in a range of H = 100 to 500. Up to H = 300, they show little variation of the counting efficiencies for both isotopes in the two selected windows.

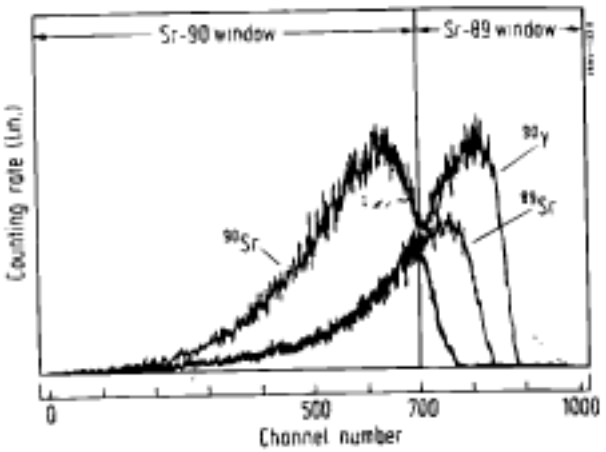

Fig. 1. Pulse height distributions for ^{90}Sr, ^{89}Sr and ^{90}Y in the liquid-scintillation spectrometer and window settings for ^{90}Sr and ^{89}Sr. Quenching monitor readings are H = 81, 97 and 168 for ^{90}Sr, ^{89}Sr and ^{90}Y, respectively. The β^--ray energy on the abscissa is scaled logarithmically.

2.2. Sequential Cerenkov-radiation and liquid-scintillation counting

For Cerenkov-radiation counting the external quench monitor of the spectrometer was switched off. Measured pulse height spectra of the three nuclides are shown in Figure 2. A channel window 0 to 500 was chosen for all of them. The counting efficiency is volume dependent [7] with a flat maximum between 5 and 9 ml for ^{89}Sr and ^{90}Y. A volume of 6 ml was applied. Picric acid which is used in our chemical separation procedure for strontium based on solvent extractions with crown ethers [3] was found to introduce strong quenching and has, therefore, carefully to be elimi-

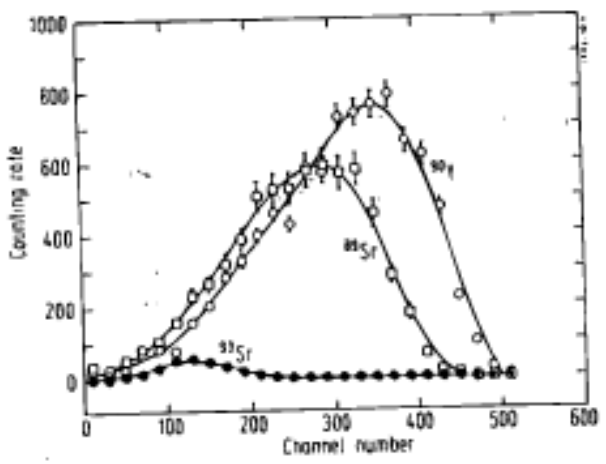

Fig. 2. Pulse height spectra of the Cerenkov radiation produced by the β^--radiations of ^{90}Sr, ^{89}Sr and ^{90}Y in aqueous solution at a constant activity level of 250 Bq for all three nuclides.

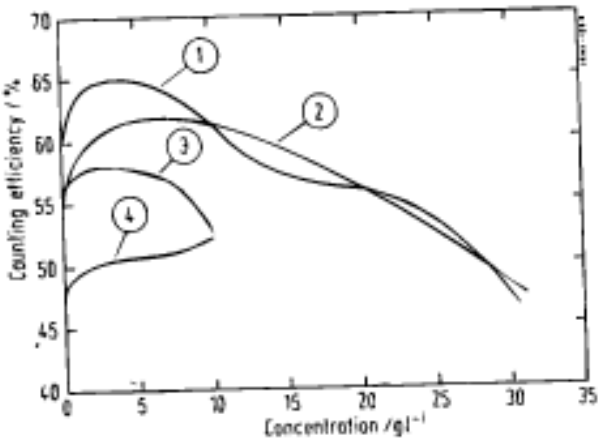

Fig. 3. Efficiency for Cerenkov counting of ^{89}Sr in the presence of different wavelength shifters: (1) 1-naphthylamine-4-sulfonic acid, Na salt, (2) 2-naphthylamine-6,8-disulfonic acid, K salt, 3-naphthylamine-2,7-disulfonic acid, Na salt, (4) 2-naphthylamine-1,5-disulfonic acid. Data points are omitted for sake of clarity.

nated. Since the counting efficiency can be improved by adding a wavelength shifter such as [9] the K-salt of 2-naphthylamine-6,8-disulphonic acid this compound and seven other naphthylamine-sulphonic acids were compared with ^{89}Sr activities. As Figure 3 shows, the Na-salt of 1-naphthylamine-4-sulfonic acid gives the best results. With the optimum concentration, 4.0 g l^{-1}, the counting efficiency is increased by a factor of 1.55 for ^{89}Sr and 3.2 for ^{90}Sr compared to the solution without shifter. Under such conditions, the efficiencies are: ^{90}Sr 6.3%, ^{89}Sr 64%, ^{90}Y 81%, ^{85}Sr 0.3%, and ^{88}Y 0.2%. The γ-ray emitters ^{85}Sr and ^{88}Y are convenient tracers to measure chemical yields in separation procedures. However, as the data indicate spiking of samples with these nuclides may lead to problems in the assay of low activities. With fresh solutions of the wavelength shifter the counting rates are somewhat increased due to chemoluminescence but they are stable between 10 and 50 hours after preparation. Hence, the Cerenkov radiation counting is started about one hour after purification of strontium from ^{90}Y with a sample volume of 6 ml containing 4.0 g l^{-1} of the wavelength shifter taken from an aged solution. After completion of that measurement, 10 ml of the gel scintillator "Ready Solve EP" from Beckman Instruments are added and the liquid-scintillation counting is carried out without window setting. Quenching corrections are made with calibration curves as described in Section 2.1.

The disintegration rates of ^{89}Sr and ^{90}Sr are obtained as follows:

$$D_{89} = \frac{1}{f_\epsilon}\{R_C\epsilon^L_{90} - R_L\epsilon^C_{90}\} \quad (1)$$

$$D_{90} = \frac{1}{f_\epsilon}\{R_L\epsilon^C_{89} - R_C\epsilon^L_{89}\} \quad (2)$$

with

$$f_\epsilon = \epsilon^L_{90}\epsilon^C_{89} - \epsilon^L_{89}\epsilon^C_{90}$$

D_{89}, D_{90} : disintegration rates of ^{89}Sr and ^{90}Sr, dpm

R_C : measured rate in Cerenkov-radiation counting, cpm, corrected for background (18.5 cpm in our setup)

R_L : measured rate in liquid-scintillation counting, cpm, corrected for background (42.8 cpm in our setup)

ϵ^C_{89}, ϵ^C_{90} : Cerenkov-counting efficiencies for ^{89}Sr and ^{90}Sr

ϵ^L_{89}, ϵ^L_{90} : liquid-scintillation counting efficiencies for ^{89}Sr and ^{90}Sr.

The latter two efficiencies ϵ^L_{89}, ϵ^L_{90} are read from a polynomial fit of experimental quenching curves, efficiency ϵ versus quenching monitor reading H, interpolated by the software of the liquid-scintillation spectrometer.

2.3. Low-level Cerenkov-radiation and liquid-scintillation counting

Since during the long counting times required at low activity levels the ingrowing ^{90}Y contributes significantly, the method is modified by applying in the liquid-scintillation counting two windows as shown in Figure 4. Window A, channel 0 to 750, comprises >99% of the ^{89}Sr spectrum and most of ^{90}Sr and ^{90}Y. Window B set at channel 750 to 1000 selects the high energy part of the ^{90}Y spectrum. With these settings the quenching is found to be quite reproducible, H = 204 to 219, after the final steps of our strontium separation procedure [3]: 10 mg of purified strontium carbonate are dissolved in 1 N HCl, evaporated to dryness and heated to 350°C for 15 min to remove traces of picric acid; the residue is taken up by shifter and scintillator solutions. Hence, an

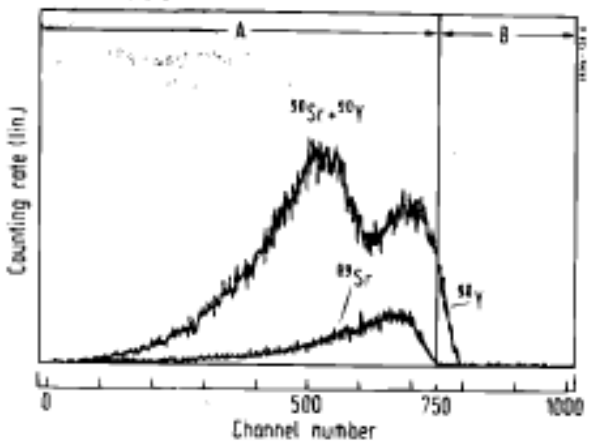

Fig. 4. Pulse height distributions in the liquid-scintillation spectrometer for ^{89}Sr and $^{90}Sr/^{90}Y$ with window settings for ^{89}Sr, ^{90}Sr plus ^{90}Y (A) and for ^{90}Y only (B). Quenching monitor readings are H = 214 for ^{89}Sr and H = 209 for $^{90}Sr/^{90}Y$.

optimization of the two windows in case of variations of quenching is not necessary.

The disintegration rates are calculated as follows:

$$D_{89} = \frac{1}{f'_\epsilon}\left[\epsilon_{90}^{LA}\epsilon_{Y}^{LB}R_C + \epsilon_{90}^{C}\epsilon_{90}^{LA}R_{LB} - \epsilon_{90}^{C}\epsilon_{Y}^{LB}R_{LA} - \epsilon_{Y}^{C}\epsilon_{90}^{LA}R_{LB}\right] \quad (4)$$

$$D_{90} = \frac{1}{f'_\epsilon}\left[\epsilon_{89}^{C}\epsilon_{Y}^{LB}R_{LA} + \epsilon_{Y}^{LA}\epsilon_{89}^{LB}R_C + \epsilon_{Y}^{C}\epsilon_{89}^{LA}R_{LB} - \epsilon_{Y}^{C}\epsilon_{89}^{LB}R_{LA} - \epsilon_{89}^{LA}\epsilon_{Y}^{LB}R_C - \epsilon_{89}^{C}\epsilon_{Y}^{LA}R_{LB}\right] \quad (5)$$

with

$$f'_\epsilon = \epsilon_{89}^{C}\epsilon_{90}^{LA}\epsilon_{Y}^{LB} + \epsilon_{90}^{C}\epsilon_{Y}^{LA}\epsilon_{89}^{LB} - \epsilon_{Y}^{C}\epsilon_{89}^{LA}\epsilon_{90}^{LB} - \epsilon_{90}^{C}\epsilon_{89}^{LA}\epsilon_{Y}^{LB}$$

R_C : measured rate in Cerenkov-radiation counting, cpm, corrected for background (18.5 cpm in our setup)

R_{LA}, R_{LB} : measured rates in liquid-scintillation counting, cpm, in windows A, B corrected for background (A: 42.8, B: 6.9 cpm in our setup)

$\epsilon_{89}^{C}, \epsilon_{90}^{C}, \epsilon_{Y}^{C}$: Cerenkov-counting efficiencies for ^{89}Sr, ^{90}Sr and ^{90}Y

$\epsilon_{89}^{LA}, \epsilon_{90}^{LA}, \epsilon_{Y}^{LA}$: liquid-scintillation counting efficiencies for ^{89}Sr, ^{90}Sr and ^{90}Y in window A

$\epsilon_{89}^{LB}, \epsilon_{Y}^{LB}$: liquid-scintillation counting efficiencies for ^{89}Sr and ^{90}Y in window B.

The efficiencies ϵ^{LA}, ϵ^{LB} are obtained from the polynomial fits to the experimental quenching curves. For $H \geq 212$ we have $\epsilon_{89}^{LB} = 0$ and a simplification of Equation (5).

3. Results and discussion

Conventional liquid-scintillation counting with two counting windows according to Section 2.1 is only of limited potential in the assay of low ^{90}Sr activities in presence of ^{89}Sr. This is evident from Table 1 where samples of 36 Bq ^{90}Sr, purified from ^{90}Y, were counted after mixing with increasing ^{89}Sr activities at three different quenching levels. The ^{90}Sr determination is quite uncertain over the whole range studied with a trend of being much too high. At a ratio $^{89}Sr/^{90}Sr \approx 10$:

Table 1. Determination of ^{89}Sr and ^{90}Sr by liquid-scintillation counting in two pulse height windows with automatic quenching correction

Disintegration rate (Bq)		$^{89}Sr/^{90}Sr$ ratio	Quenching monitor reading H	Deviation (%)	
^{90}Sr	^{89}Sr			^{90}Sr	^{89}Sr
36	4.0	0.11	170	+ 3.3	− 5.6
			262	− 47	− 0.4
			406	− 68	− 4.6
36	35	1.0	170	+ 5.1	+31
			260	+ 26	+17
			406	+ 18	+14
36	176	5	171	+ 24	+ 1.6
			266	+ 51	− 3.9
			403	+ 49	− 6.6
36	281	8	172	+100	−13
			262	+110	−14
			417	+ 73	−19
36	457	13	174	+160	− 4.8
			269	+140	−11
			411	+100	−11
36	1406	39	176	+320	− 7.7
			275	+330	− 8.4

Table 2. Determination of ^{89}Sr and ^{90}Sr by a combination of Cerenkov-radiation and liquid-scintillation counting in samples freshly purified from ^{90}Y

Disintegration rate (Bq)		$^{89}Sr/^{90}Sr$ ratio	Quenching monitor reading H	Deviation (%)	
^{90}Sr	^{89}Sr			^{90}Sr	^{89}Sr
6.9	10	1.4	243	− 2.2	+7.3
			362	− 6.7	+1.8
			486	+ 7.1	+0.9
6.9	20	3	246	+ 8.4	−1.8
			349	+ 7.9	+1.7
			487	− 5.7	−0.6
6.9	46	7	271	+ 6.2	−1.0
			373	− 2.9	−3.6
			493	− 5.5	−2.8
6.9	101	15	244	+ 2.3	−0.9
			363	+25	−0.4
			457	+ 3.2	+1.9
6.9	203	29	244	+91	−3.2
			355	+43	−3.1
			488	−16	−2.2

Table 3. Determination of ^{89}Sr and ^{90}Sr by a combination of Cerenkov-radiation and liquid-scintillation counting in the presence of ^{90}Y equilibrium activities

Disintegration rate (Bq)		$^{89}Sr/^{90}Sr$ ratio	Quenching monitor reading H	Deviation (%)	
^{90}Sr	^{89}Sr			^{90}Sr	^{89}Sr
16	6.6	0.4	211	− 2.2	−10
	13	0.9	209	− 5.0	+ 7.7
	21	1.3	209	− 1.1	+10
	26	1.7	211	+ 2.4	+ 2.9
	33	2.1	207	+ 10	+ 7.6
	39	2.6	210	+ 1.1	+ 5.6
	46	3.0	210	− 5.4	+ 2.2
	53	3.4	209	− 5.7	+ 1.5
	66	4.3	210	− 5.2	− 1.1
	98	6.4	211	+ 3.7	− 0.2
	131	8.5	212	− 1.8	− 0.4
	197	13	217	+ 13	− 3.2
	263	17	224	+ 67	− 4.4
	328	21	220	+ 140	− 6.9
	656	43	224	+1200	−28
1.6	6.6	4.3	196	+ 0.5	+19
	13	8.5	197	+ 2.9	− 6.3
	20	12	201	+ 16	+10
	26	17	204	− 22	+ 4.1
	33	21	204	− 38	+ 1.2
0.15	0.6	4.0	215	+ 13	+14
	1.3	8.1	210	− 2.3	+ 8.6
	1.9	12	215	− 12	+ 3.4
	2.5	16	212	− 23	− 1.6

Table 4. Determination of ^{89}Sr and ^{90}Sr by a combination of Cerenkov-radiation and liquid-scintillation counting in the presence of ingrowing ^{90}Y activities*

Time after purification		Quenching monitor reading H	Deviation (%)	
(h)	($h/T_{1/2}$)		^{90}Sr	^{89}Sr
4.7	0.07	209	+ 8.0	−5.3
23.1	0.36	212	+ 4.8	−2.7
51.8	0.81	214	+10.6	−1.8
119	1.85	216	+ 7.1	+4.9
264	4.1	222	+ 6.6	−5.8
423	6.6	228	+ 9.9	+8.0

* Initial activities: 28 Bq ^{90}Sr and 46 Bq ^{89}Sr, freshly purified from ^{90}Y.

which is likely to occur in fresh fallout the ^{90}Sr activity is overestimated by a factor of two to three. The ^{89}Sr data are more satisfying with a trend of being too low. Obviously the two pulse height distributions with their β^--energy ratio of 2.7 overlap too strongly for the automatic window adjustment by the spectrometer.

Table 2 shows results of similar measurements with the combined Cerenkov-radiation and liquid-scintillation counting after purification from ^{90}Y, Section 2.2, also at three different quenching levels. This method gives much better results although the ^{90}Sr activity is lower by a factor of five than in the preceding table. Apart from the last example with the most extreme $^{89}Sr/^{90}Sr$ ratio, the errors for ^{90}Sr remain at an acceptable level and show no obvious trend in sign. The ^{89}Sr data are very satisfying. This improvement is due to the simplified quenching correction. In the Cerenkov-radiation counting no quenching correction is required at all and in the liquid-scintillation counting with an open window no automatic window adjustment is applied.

In further measurements compiled in Table 3 the ^{90}Sr activities and $^{89}Sr/^{90}Sr$ ratios are varied over some range. Here ^{90}Sr was counted at $H \approx 210$ in radioactive equilibrium with ^{90}Y as described in Section 2.3 with the liquid-scintillation counting immediately following the Cerenkov-radiation counting. The counting time was limited to a maximum of 3 h per measurement or interrupted when in the counting statistics a level of $2\sigma = \pm 2\%$ was reached. The results remain within $\pm 20\%$ of the given activity up to a $^{89}Sr/^{90}Sr$ ratio of about 15 even in the last group of data with only 0.15 Bq of ^{90}Sr activity. When the ingrowth of ^{90}Y in a $^{89}Sr/^{90}Sr$ sample is followed over several half-lives no trend with time is seen in the errors as Table 4 indicates. The quenching number increases somewhat with time because of a slight darkening of the shifter. As a final test, three samples with 15.6 Bq ^{90}Sr and 10 mg Sr carrier were run through the whole chemical separation procedure [3] including burning of traces of picric acid and a gravimetric determination of the chemical yield. The activities found deviate by −5.7%, +13% and −14% from the known value.

4. Conclusion

For the simultaneous determination of ^{89}Sr and ^{90}Sr at low activity levels liquid-scintillation counting with two pulse height windows cannot be recommended. Reliable results are obtained with a combination of Cerenkov-radiation and liquid-scintillation counting that can be carried out with the same spectrometric equipment. Here, either an open window can be applied in liquid-scintillation counting or two pulse height windows with the higher window adjusted to accept ^{90}Y only. The latter method is accurate to $\pm 20\%$ for $^{89}Sr/^{90}Sr$ ratios up to about 15 as may occur in fresh fallout mixtures. Disintegration rates for ^{90}Sr as low as 0.15 Bq, corresponding to 3×10^8 atoms, can be measured with such an error during six hours of total counting time.

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References

1. Hötzl, H., Rosner, G., Winkler, R.: Radiochim. Acta **41**, 181 (1987).
2. Denschlag, H. O., Diel, A., Gläsel, K.-H., Heimann, R., Kaffrell, N., Knitz, U., Menke, H., Trautmann, N., Weber, M., Herrmann, G.: Radiochim. Acta **41**, 163 (1987).
3. Wendt, K., Haub, G., Köhler, S., Kluge, H.-J., Monz, L., Otten, E.-W., Passler, G., Senne, P., Stenner, J., Zimmer, K., Herrmann, G., Trautmann, N., Walter, K., in: *Optoelectronics for Environmental Science* (S. Martellucci, A. N. Chester, eds.). Plenum, New York 1990, p. 163.
4. Randolph, R. B.: Int. J. Appl. Rad. Isot. **26**, 9 (1975).
5. Buchtela, K., Tschurlovits, M.: Int. J. Appl. Rad. Isot. **26**, 333 (1975).
6. Carmon, B.: Int. J. Appl. Rad. Isot. **30**, 97 (1979).
7. Ross, H. H.: Anal. Chem. **41**, 1260 (1969).
8. Horrocks, D. L.: J. Radioanal. Chem. **43**, 489 (1978).
9. Elrick, R. H., Parker, R. P.: Int. J. Appl. Rad. Isot. **19**, 263 (1968).