

A BETA SPECTROMETER FOR MONITORING ENVIRONMENTAL MATRICES

M. Palazzolo, P. Prati, G. Ricco and M. Taiuti*

Abstract—In environmental matrices, pure beta emitters are often present with other beta-gamma emitters. This work describes a beta spectrometer that allows measurement of pure beta-emitter activities of about 10^{-2} Bq. The detection system consists of a plastic scintillator surrounded in a 4π geometry by large anticoincidence NaI(Tl) scintillators. Solid, liquid, and gaseous samples can be analyzed, and no chemical separation is necessary. The system is particularly useful for ^{90}Sr measurement.

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Key words: monitoring, environmental; ^{90}Sr ; radiation, beta; beta spectrometry

INTRODUCTION

THE CHERNOBYL accident demonstrated the need for rapid and accurate analysis of environmental matrices. In particular, pure beta emitters, such as ^{90}Sr , must be strictly monitored; their direct detection is complicated due to the number of beta-gamma emitters present. ^{90}Sr is usually measured by radiochemical procedures that are time-consuming and require that the $^{90}\text{Sr}/^{90}\text{Y}$ equilibrium is achieved (Martin 1987). Methods based on anticoincidence measurements have been suggested (Brini et al. 1990).

We have developed a spectrometer utilizing a plastic scintillator surrounded in all directions by large NaI(Tl) anticoincidence detectors. This system allows beta-energy measurement with gamma anticoincidence. The plastic scintillator also can be operated in coincidence with NaI(Tl) detectors. In this configuration, beta spectra in coincidence with gamma peaks on the NaI(Tl) detectors can be measured. Complete beta and beta-gamma spectrometry can be performed. Within a few hours, ^{90}Sr activities on the order of 10^{-2} Bq can be discriminated in a sample of ~ 0.5 g.

SPECTROMETER DESCRIPTION

A detector diagram is provided in Fig. 1. A cylindrical plastic scintillator, NE102A (80-mm diameter

and 25-mm thick), is optically cemented to a NaI(Tl) scintillator (80-mm diameter and 60-mm thick). The two detectors are coupled to a fast photomultiplier[†] and are surrounded by a large cylindrical NaI(Tl) scintillator (254 mm \times 254 mm with an 80-mm-diameter hole) coupled to eight photomultipliers.[‡]

Another NaI(Tl) cylindrical detector (76.2-mm diameter and 76.2-mm thick) with its own photomultiplier[§] is positioned in front of the plastic scintillator and completes the 4π NaI(Tl) anticoincidence. The sample is inserted near the plastic scintillator as a thin layer deposited on a Lucite support (solid samples) or as absorbed into a carbon fiber plug (liquid samples). Air concentrations can be measured by counting air particulate filters. The system is shielded in all directions with 10 cm of lead and an internal layer of 2 cm of electrolytic copper.

Signals from the cemented plastic and NaI(Tl) scintillators are analyzed by a pulse-shape discriminator (PSD) circuit, which measures the input pulse width, in order to discriminate between events occurring in the plastic and NaI(Tl) scintillators. Signal time constants from plastic and NaI(Tl) scintillators are ~ 20 ns and 250 ns, respectively. For anticoincidence, pulses from the plastic scintillator are distinguished when they are: a) recognized by the PSD as generated only by the plastic scintillator; and b) in anticoincidence with all other NaI(Tl) detectors.

Because the plastic scintillator thickness is greater than the range of beta particles emitted by the most important radioactive isotopes (for example, the maximum range in Ne102A for ^{90}Y beta particles is about 20 mm), pure beta emitters only produce signals in the plastic detector. The beta signals from beta-gamma decays are detected by the plastic, but the attendant gamma rays are detected by the NaI(Tl) scintillators. With anticoincidence, coincident events are automatically rejected. The system can be used to seek signals on the plastic scintillator that are in coincidence with gamma-ray peaks from NaI(Tl) detectors. In this way, beta-gamma spectrometry can be performed.

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[†] Thorn EMI Electron Tubes Ltd., Model 9822B, Bury Street Ruislip, Middlesex HA47TA, England.

[‡] Thorn EMI Electron Tubes Ltd., Model 9758, Bury Street Ruislip, Middlesex HA47TA, England.

[§] Philips Scientific Inc., Model 46AVP, 305 Island Road, Mahwah, NJ 07430.

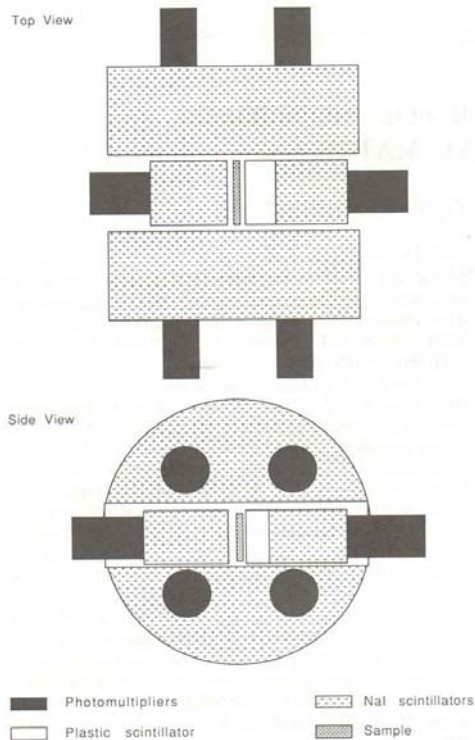


Fig. 1. Spectrometer composition and geometry.

ELECTRONICS

The electronics schematic is shown in Fig. 2. The dynode signal from the cemented plastic and NaI(Tl) scintillators, and the summed anode pulses from the other NaI(Tl) detectors, are amplified by two different linear amplifiers¹ and stored on two gated 8k-memory multichannel analyzers (MCA) connected through an RS232 serial port to a computer.² The PSD and a coincidence circuit produce the trigger for the signal acquisition. The pre-amplified dynode signals are further amplified and shaped by a delay line amplifier.³

The clipped pulses have rise and decay times pro-

¹ Silena SpA, Model 7611/L and Model 7616, Via Firenze 3, Cernusco S/N (MI), 20063 Italy.

² Digital Equipment Corporation, Model VAX11/780, 129 Parker Street, Maynard, MA 01754.

³ EG&G Ortec, Model 460, 100 Midland Road, Oak Ridge, TN 37831.

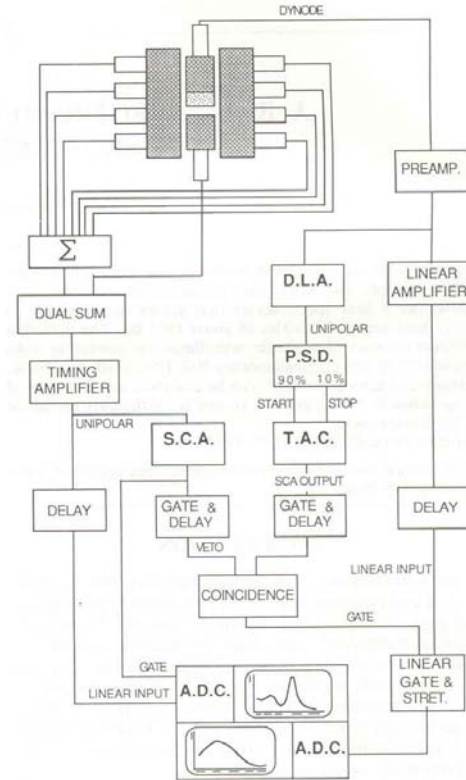


Fig. 2. Spectrometer electronics scheme.

portional to the dynode pulse width and amplitude proportional to the particle energy. These pulses are then discriminated by a pulse-shape and timing single-channel analyzer.⁴ This, coupled with a time-to-amplitude converter (TAC),⁵ permits discrimination through the decay time of the amplified input pulse.

The TAC single-channel analyzer (SCA) output—with pulses of a given minimum and maximum—through a selected window provides the signal that can be discriminated by a fast coincidence⁶ module, either in anticoincidence or alternatively in coincidence with

⁴ EG&G Ortec, Model 552, 100 Midland Road, Oak Ridge, TN 37831.

⁵ Enertec Scumberger S.A., Model 7151, Branche Instrumentation Nucléaire, 1, Pard de Tanneries, 67380, Lingolsheim, France.

⁶ EG&G Ortec, Model 414A, 100 Midland Road, Oak Ridge, TN 37831.

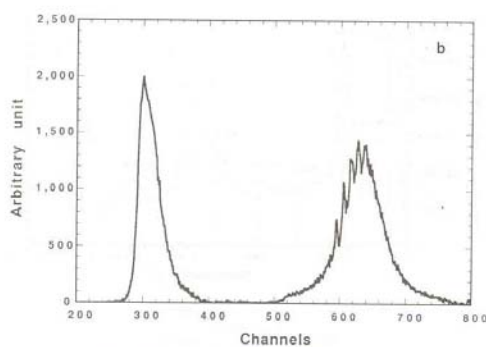
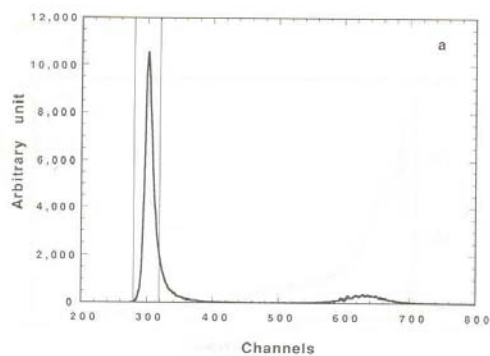


Fig. 3. a) PSD spectrum of a $^{90}\text{Sr} + ^{90}\text{Y}$ beta source. The two vertical lines define the pulse-width window. The lower peak is from background events in NaI(Tl) (cosmic rays) and by bremsstrahlung photons produced by high-energy ^{90}Y electrons. b) PSD spectrum of a ^{137}Cs beta-gamma source.

Table 1. Measured and calculated efficiencies for the cemented plastic and NaI(Tl) scintillators for two gamma sources 12 cm from the detectors.

	Percent detection efficiency			
	Measured		Calculated	
	Plastic	NaI (Tl)	Plastic	NaI (Tl)
^{137}Cs	0.39	1.41	0.38	1.50
^{54}Mn	0.33	1.49	0.36	1.41

a gate pulse from the NaI(Tl) detectors. The gate signal from the NaI(Tl) detectors is produced by a single-channel analyzer⁸⁸ whose window can be set to select

⁸⁸ Silena, Model 7616, Via Firenze 3, Cernusco S/N (MI), 20063 Italy.

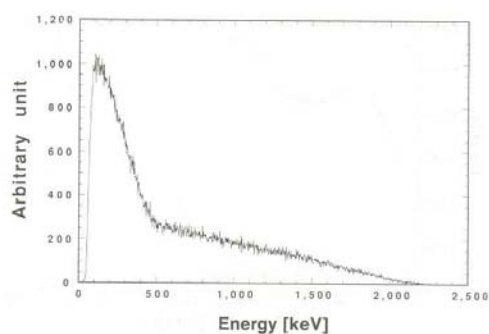


Fig. 4. Energy spectrum of a $^{90}\text{Sr} + ^{90}\text{Y}$ source. The double beta decay of ^{90}Sr and ^{90}Y is clearly detected. Counts are expressed in arbitrary units.

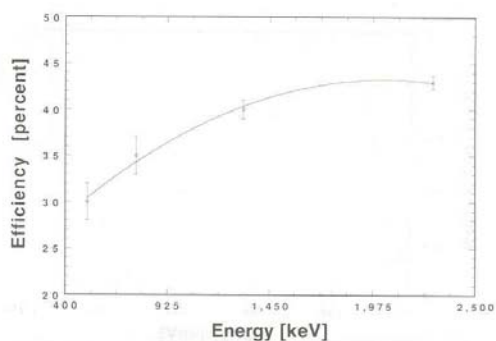


Fig. 5. Efficiency η_{β} vs. the maximum energy of beta emitters. In the range from 500 up to 2280 keV, the efficiency is well-fitted by the curve $\eta_{\beta}(E) = 20.72 + 2.16 \times 10^{-2} E - 5.12 \times 10^{-6} E^2$.

Table 2. Anticoincidence rejection efficiency vs. gamma energy for some common isotopes. The relatively high uncertainty is due to the calibration standards.

Isotope	Gamma energy (keV)	Rejection efficiency (%)
^{203}Hg	279	93 ± 5
^{137}Cs	662	96 ± 5
^{54}Mn	834	90 ± 5
^{60}Co	1,173–1,332	97 ± 4
^{88}Y	898–1,836	96 ± 5

either all energies or a chosen gamma peak. Logic and linear signals are synchronized by a quad gate/delay generator.¹¹

¹¹ Philips Scientific, Model 794, 305 Island Road, Manwah, NJ 07430.

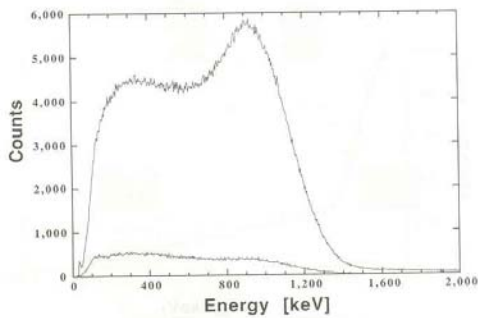


Fig. 6. ^{60}Co energy spectra on the plastic scintillator with the NaI(Tl) anticoincidence on (lower plot) and off (upper plot). Measure time is 600 s in both cases.

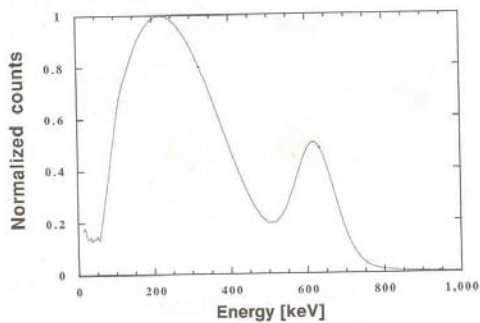


Fig. 7. ^{137}Cs energy spectrum on the plastic scintillator. The beta energy distribution and the internal conversion electron peak at about 620 keV are apparent. The source is a thin layer deposited on a metallic disc, and counts are normalized to the maximum value.

SPECTROMETER PERFORMANCE

The pulse-shape discrimination of the signals from the cemented plastic and NaI(Tl) scintillators is the key to proper operation. In Fig. 3a and b, the PSD spectra of beta ($^{90}\text{Sr} + ^{90}\text{Y}$) and beta-gamma (^{137}Cs) sources are shown. Beta particles are detected by the plastic scintillator; gamma rays interact with both the scintillators. Events occurring in both scintillators are clearly resolved. Interactions in plastic and in NaI(Tl) produce two peaks. Radiations detected in both the detectors produce pulses in the second peak or in the region between the two peaks.

The discriminator threshold (width of the TAC window) is set using a $^{90}\text{Sr} + ^{90}\text{Y}$ source. Fig. 3a shows the window limits. The PSD input threshold is set to

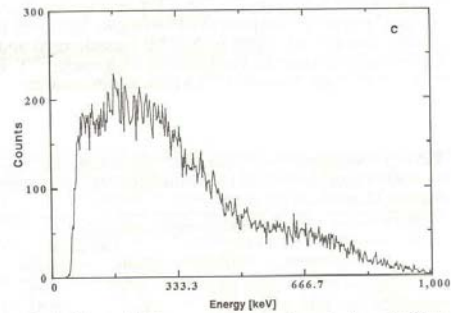
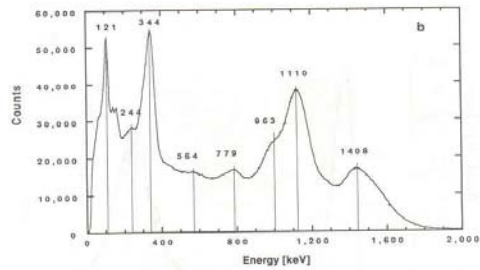
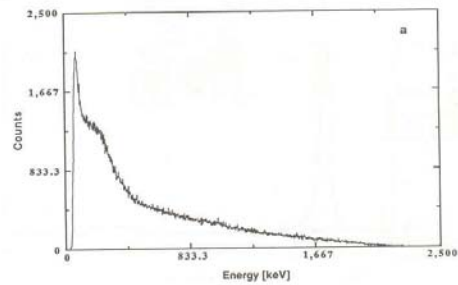


Fig. 8. a) $^{90}\text{Sr} + ^{152}\text{Eu}$ spectrum on the plastic scintillator. Count time is 22,000 s. b) $^{90}\text{Sr} + ^{152}\text{Eu}$ spectrum on NaI(Tl) detectors. Count time is 21,600 s. c) $^{90}\text{Sr} + ^{152}\text{Eu}$ spectrum with the plastic scintillator in coincidence with the 779-keV peak on NaI(Tl) detectors. Count time is 6,200 s. The main feature is the ^{152}Eu beta decay with a maximum energy of 700 keV.

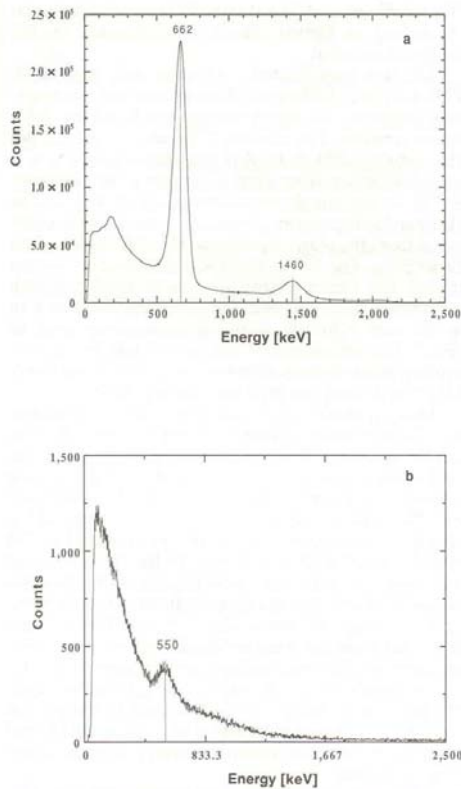


Fig. 9. a) Spinach spectrum on NaI(Tl) detectors with ^{137}Cs and ^{40}K peaks. Count time is 60,000 s and sample weight is 55 g. b) Spinach spectrum (0.3 g) on the plastic scintillator. Count time is 236,000 s.

40 mV, which corresponds to a minimum detectable energy of about 60 keV with the plastic scintillator. The ability to resolve events from both detectors without efficiency losses has been demonstrated with a Monte Carlo code (Corvisiero et al. 1981). Calculated and measured peak efficiencies (events occurring only in plastic and only in NaI) for two sealed and calibrated gamma sources are in agreement (see Table 1).

The measured spectrum of a low-activity (~ 150 Bq) $^{90}\text{Sr} + ^{90}\text{Y}$ source is shown in Fig. 4. The source was a thin layer deposited on a plastic disc (50-mm diameter, 12 mg cm^{-2} total thickness), and the detection efficiency (η_β) was 43%. In Fig. 5, the detection efficiency η_β , measured using some calibrated sources, is

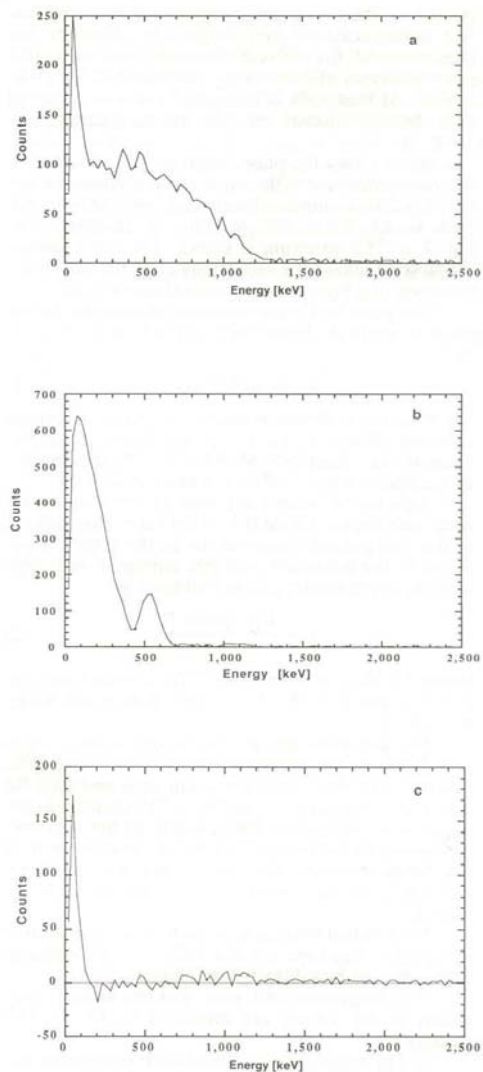


Fig. 10. Spinach spectrum on the plastic scintillator after three different corrections: a) ^{137}Cs standard spectrum subtraction. The remaining counts follow the typical ^{40}K shape. b) ^{40}K standard spectrum subtraction. The remaining counts follow the typical ^{137}Cs shape. c) ^{137}Cs and ^{40}K standard spectra subtraction. The remaining spectrum closely resembles background. The low-energy residual peak is due to the unfolding process statistics and to the beta-particle self absorption within the sample.

plotted vs. the maximum energy of the beta emitters. The anticoincidence gamma-rejection efficiency has been evaluated for a 30-keV threshold using calibrated gamma sources with an energy range from 0.3 MeV to 2 MeV. At least 85% of the gamma rays are detected and, hence, rejected by the anticoincidence (see Table 2).

Fig. 6 shows the pulse-height spectra obtained in the plastic detector with and without anticoincidence for ^{60}Co . The Compton distribution, with anticoincidence, is only 9% of that without anticoincidence. In Fig. 7, a ^{137}Cs spectrum is shown. The good gamma suppression allows the measurement of the beta spectrum with the internal conversion electron peak.

Beta particles from coincident beta-gamma decays are detected by the plastic with an efficiency ($\eta_{\beta\gamma}$) given by

$$\eta_{\beta\gamma} = \eta_{\beta}(1 - \eta_{\gamma r}), \quad (1)$$

where η_{β} = beta detection efficiency and $\eta_{\gamma r}$ = gamma rejection efficiency. $\eta_{\beta\gamma}$ is typically lower than 5%. Gamma rays, from 0.06 MeV to 2 MeV, are detected by the plastic with an efficiency lower than 0.3%.

Background counts are due to photomultiplier noise with the rate (B) as 0.33 ± 0.01 cps. Sixty percent of the background counts occur in the region below 300 keV. The minimum pure beta activity, σ , detectable with the spectrometer can be evaluated as

$$\sigma = \frac{3 \times \text{sqrt}(B/T)}{\eta_{\beta}}, \quad (2)$$

where T is the count time. For ^{90}Sr , $\eta_{\beta} = 0.43$ and σ is 0.07 Bq and 0.02 Bq for 1- and 10-h count times, respectively.

The sensitivity also can be calculated for energies over 300 keV. The detection efficiency for ^{90}Sr is 29%, and σ is 0.06 Bq with a 1-h count time and 0.02 Bq with a 10-h count time. The $^{90}\text{Sr} + ^{90}\text{Y}$ spectrum in the region over 300 keV is less sensitive to the beta self-absorption in the sample and to a certain number of interfering beta-gamma emitters. (Electrons from radioactive decays usually have maximum energies below 1 MeV.)

The residual beta-gamma emitters' contribution in the plastic spectrum (in the following, the residual spectrum) can be subtracted as follows:

1) Beta-gamma and pure gamma activity contained in the sample are measured by the NaI(Tl) detectors.

2) The residual spectrum of each recognized isotope is either subtracted, if the residual spectra of the involved isotopes are known, or directly measured in coincidence with a selected gamma peak on the NaI(Tl) detectors, properly set with the SCA window (see Fig. 2). With this procedure, a complete beta, gamma, and beta-gamma spectrometry can be performed and the pure beta activity in the sample can be determined.

Two applications are provided as examples: 1) a liquid sample containing a mixture of $^{90}\text{Sr} + ^{90}\text{Y}$ and

^{152}Eu solutions; and 2) a thin solid layer of dried spinach, picked in Genoa (Italy), contaminated by the Chernobyl accident.

The very complicated ^{152}Eu decay scheme (ORNL 1973) is a good challenge to demonstrate the spectrometer's capability to reject events produced by beta-gamma emitters. The solution (^{90}Sr activity: 12 ± 1 Bq; ^{152}Eu activity: 453 ± 14 Bq) was deposited on a 0.4-mm-thick carbon fiber plug. The steps to discriminate the ^{90}Sr in the sample are summarized in Fig. 8. The ^{152}Eu standard spectrum (separately measured) was subtracted from the plastic spectrum (Fig. 8a) to obtain the ^{90}Sr activity. The ^{152}Eu contribution was also counted through the plastic scintillator in coincidence with gamma peaks on the NaI(Tl) detectors, as shown in Fig. 8b and c for the 779-keV gamma-ray peak of ^{152}Eu .⁵⁵ The subtraction of this partial interfering contribution allowed measurement within 30% uncertainty of the ^{90}Sr activity on the entire energy range.

Dried spinach (0.3 g) was deposited on a Plexiglas disc (7-cm diameter) and counted for ~66 h. The NaI(Tl) spectrum (Fig. 9a), obtained with the same dried spinach but in a larger quantity (55 g), showed two total absorption peaks at 662 keV (^{137}Cs) and 1,460 keV (^{40}K) with activities of 116 ± 6 Bq g^{-1} and 133 ± 1 Bq g^{-1} , respectively. The plastic spectrum (Fig. 9b) showed a peak at 550 keV due to the ^{137}Cs internal conversion electrons with some absorption in the sample. Internal conversion electrons from ^{137}Cs are emitted with energy of about 620 keV (see Fig. 7). The plastic spectrum was then unfolded, searching for other beta emitters, after subtracting standard spectra of ^{137}Cs and ^{40}K sources (Fig. 10a and b). The subtracted standard spectra were corrected for the average energy lost by electrons within the sample. This is evaluated based on the position of the internal conversion peak in the plastic spectrum.

After unfolding the residual plastic spectrum (Fig. 10c), with background subtraction, only low-energy pulses were noted; we concluded that no other beta emitter was present in the sample within the spectrometer sensitivity. A check on the portion of the unfolded spectrum above 1,500 keV showed a slope different from that of ^{90}Y and indistinguishable from background measurements. $^{90}\text{Sr} + ^{90}\text{Y}$ sensitivity, with this long count time, was about 0.008 Bq. It was concluded that ^{90}Sr activity in the sample, if present, was less than 0.026 Bq g^{-1} . This result, in spite of the relatively large sample aliquot, cannot be immediately correlated with ^{90}Sr in fresh spinach after the Chernobyl accident. The drying process could have resulted in the loss of ^{90}Sr .

CONCLUSIONS

A sensitive beta spectrometer was produced using a plastic scintillator operated in anticoincidence with

⁵⁵ The 779-keV gamma-ray peak is associated with about 50% ^{152}Eu beta activity.

4 π NaI(Tl) detectors. ^{90}Sr activities as low as 10^{-2} Bq can be measured in a few hours. Sample preparation is minimal, and the monitoring of pure beta emitters in environmental matrices can be performed in a short time.

The spectrometer performance can be improved in two ways:

1) A better beta-gamma events rejection could be achieved by substituting NaI(Tl) with BGO detectors with a greater gamma detection efficiency.

2) Pure gamma interference and background could be attenuated interposing, between the sample and the plastic, a very thin silicon detector operated in coincidence with the plastic detector.

More work is necessary to improve sample preparation and to automate data reduction.

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