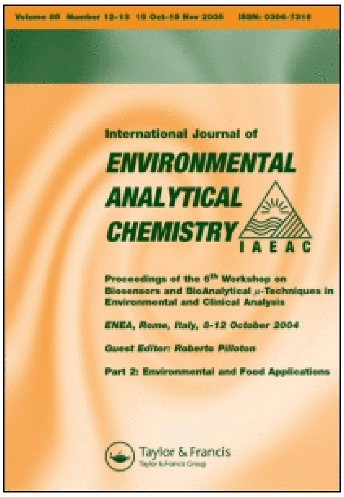

International Journal of Environmental Analytical Chemistry
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713640455

An Application of Marinelli Geometry in Gamma-Ray Spectrometry

D. Kut[a]; A. F. Akgün[a]
[a] Cekmece Nuclear Research and Training Centre, Havaalani, Istanbul, Turkey

To cite this Article Kut, D. and Akgün, A. F.(1985) 'An Application of Marinelli Geometry in Gamma-Ray Spectrometry', International Journal of Environmental Analytical Chemistry, 22: 3, 161 — 168
To link to this Article: DOI: 10.1080/03067318508076419
URL: http://dx.doi.org/10.1080/03067318508076419

Intern. J. Environ. Anal. Chem., 1985, Vol. 22, pp. 161–168
0306-7319/85/2204-0161 $18.50/0

An Application of Marinelli Geometry in Gamma-Ray Spectrometry

D. KUT and A. F. AKGÜN

Çekmece Nuclear Research and Training Centre, P.O. Box 1, Havaalani, Istanbul, Turkey

(*Received December 19, 1984; in final form February 21, 1985*)

The absolute efficiency calibration of the gamma counting system was obtained for two different sample-detector geometries for the analyses of environmental bulk samples.

The system was first calibrated by using standard point sources and the equation of efficiency curve was found and plotted. The second calibration was made by replacing the point sources with one-liter Marinelli beakers capable of using the detector active volume. The calibration standards were solution standards of Am-241, Cs-137 and Co-60. This Marinelli beaker geometry was successfully used in the leakage test of fission products which were accumulated in the spent fuel of the TR-1 Reactor.

KEY WORDS: Marinelli geometry, low-level counting, efficiency calibration, spent fuel transfer.

INTRODUCTION

For the effective measurement of low-activity environmental bulk samples such as sea and rain water, sample preparation is an important fact. Concentration of samples is a time-consuming process and may cause losses of radioisotopes of interest.[1–3] Besides, in counting low-activity samples, the best sample-detector geometry for the maximum efficiency has to be used.[3,4] In this case, Marinelli geometry which enables using the entire active volume of the crystal, will provide the highest absolute detector efficiency.[5,6]

Two calibration procedures were carried out using point source-detector and Marinelli geometries, respectively. The absolute efficiency equations were found. The efficiencies and lower limit of detection for various isotopes were calculated and a comparative evaluation was performed.

At Çekmece Nuclear Research and Training Centre, the 1-MW TR-1 Reactor was shut down in 1977 in order to increase its power to 5 MW. MTR type spent fuels were loaded in a IU-04 type transfer cask provided by Transnucleaire Company and sent to the U.S. for reprocessing. The leakage of the fission products from fuels into the cask water was measured using this Marinelli geometry.

EXPERIMENTAL AND RESULTS

Counting system

The gamma spectrometer used in the experiments consisted of a Canberra M7229 HP Ge detector coupled to a Canberra M85 4096 channel analyzer and related electronic units. The crystal volume of the detector was about 60 cm^3, the resolution was 1.87 keV at 1332.5 keV and the relative efficiency was 10%. Detector shielding was 5 cm thick lead lined with 0.3 cm copper.

Efficiency calibration

First, counting geometry was set up at a detector-source distance of 7.5 cm (Figure 1*a*) and efficiency calibration was performed using Radiochemical Centre (Amersham, U.K.) standard sources such as Co-57 (122, 136 keV), Cs-137 (661.6 keV), Co-60 (1173.2, 1332.5 keV) and Y-88 (1836 keV). One-liter Marinelli beakers made of thin polyethylene were used to perform the second calibration (Figure 1*b*). Calibrated standard solutions were prepared by diluting National Bureau of Standards (NBS) and Radiochemical Centre, standard solutions to have about a few nCi of each radionuclide per liter.

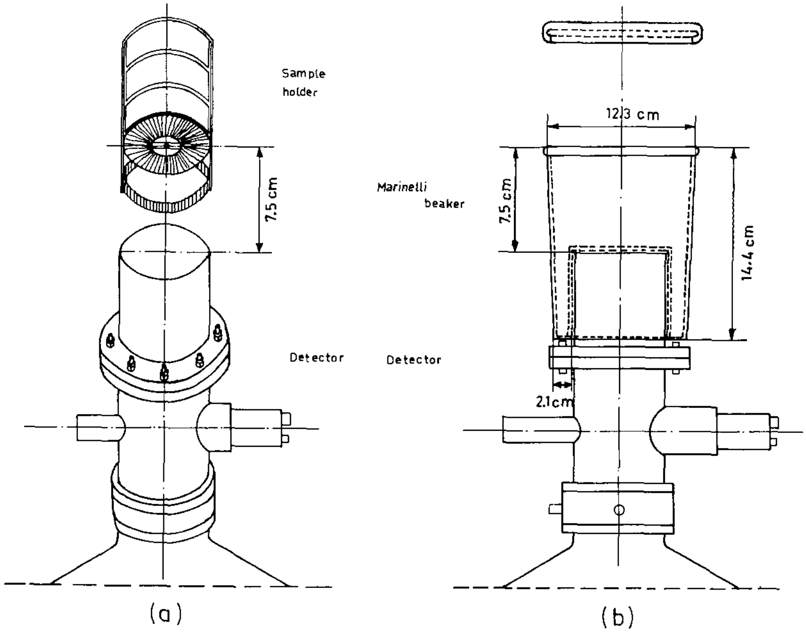

FIGURE 1 Source-detector geometry for: (a) point source; (b) Marinelli beaker.

All experimental efficiency values of the standards used were obtained by using the following formula:

$$\varepsilon(E)=\frac{N}{T\times\gamma(E)}$$

where $\varepsilon(E)$: the detector efficiency at energy E,
N: the net peak area,
$\gamma(E)$: the current gamma emission per sec at energy E.

Efficiency curves of the counting system were plotted for point sources and Marinelli geometry for these experimental data (Figure 2). Equations of efficiency curves were derived as $y=0.54\ x^{-0.90}$ and $y=0.09\ x^{-0.48}$ for point source and Marinelli geometry, respectively. By using these equations, absolute efficiencies for some of the environmentally important radionuclides were calculated (Table I). It has to be noted that in these derivations the experimental data for

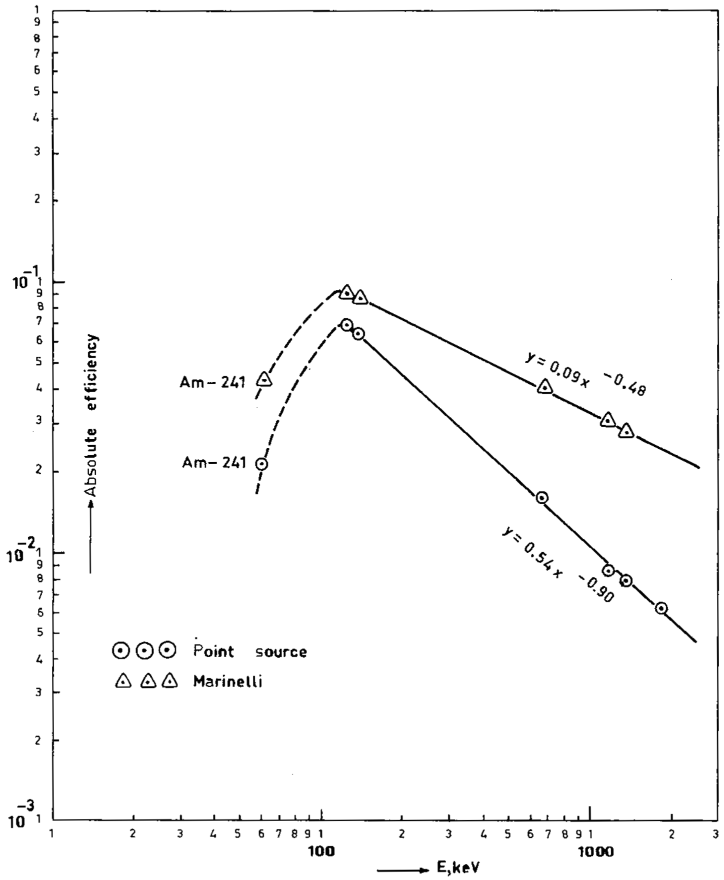

FIGURE 2 Detector efficiency curves.

Am-241 have not been used, but have been given in Table I and Figure 2, because these equations are valid only for the higher energy region of the efficiency curves.

Lower limit of detection (LLD)

The lower limits of detection of some isotopes for point sources and Marinelli geometry were determined and compared (Table II). The

TABLE I

Calculated absolute efficiencies of the counting system for different energies (Am-241 values are experimental).

Isotope	Gamma energy (keV)	Efficiency ($\times 10^{-2}$) Point source[a]	 Marinelli geometry[b]
Am-241	59.5	0.2112	0.4467
U-235	186	0.479	0.7537
Th-232 (Tl-208)	583	0.1707	0.4348
U-238 (Bi-214)	609	0.1641	0.4258
Cs-137	661.6	0.1523	0.4091
U-238 (Bi-214)	1120	0.09460	0.3175
Co-60	1173.2	0.09071	0.3105
Co-60	1332.5	0.08085	0.2920
K-40	1460	0.07443	0.2795
U-238 (Bi-214)	1764	0.06273	0.2551

[a] Using $y = 0.54x^{-0.90}$.

[b] Using $y = 0.09x^{-0.48}$.

TABLE II

LLD values for some isotopes.

Isotope	Gamma energy (keV)	LLDs (Bq) Point source $T = 1000$ m	 Marinelli geometry $T = 1000$ m	 $T = 140$ m
U-235	186	3.7	1.1	2.7
Tl-208	583	3.3	0.8	1.3
Bi-214	609	4.4	1.5	3.7
Cs-137	661.6	2.7	0.7	2.0
Co-60	1173.2–1332.5	3.2	0.8	1.0
K-40	1460	51.8	10.4	29.2

general formula for calculating the LLD values is[2,7,8]

$$\mathrm{LLD}=\frac{4.66S_B}{2.22\varepsilon(E)fT}$$

where LLD: the lower limit of detection (pCi) at 95% confidence level,

S_B: standard deviation of the background $(\text{background})^{1/2}$,
$\varepsilon(E)$: the detector efficiency at energy E,
f: the branching factor of E,
T: the counting time (minutes).

If the background is given as pCi, it may be expressed as $\mathrm{LLD}\cong 4.66S_B$. The lower limits of detection given in Table II were calculated according to this expression. Background measurements taken with empty sample containers for point source and Marinelli geometry did not change. However, as can be seen from Table II, different LLD values were found, because LLD depends directly on detector efficiency.

Application

Before starting the leakage test of spent fuels, the efficiency calibration of the system was made for Marinelli geometry and the counting method was checked with samples having Cs-137 and Co-60 in the order of 100 Bq.

Meanwhile, 33 spent fuel and 3 fresh fuel samples from the TR-1 reactor were loaded in a IU-04 type transfer cask and the cask was emptied and refilled with water. One hour after filling, a one liter water sample was taken (Sample 1) with a Marinelli beaker from the lower tap of the cask and counted. The counting geometry was as shown in Figure 1*b*. Four hours after the first sampling, another water sample was taken from the same place and counted (Sample 2). A third water sample was taken from the storage pool of spent fuels and analyzed in the same way (Sample 3).

Due to the long storage of spent fuels (about 7 years), the fission and activation products having relatively short half-lives, such as Ce-144 (284 days), Cs-134 (2.07 years), Zr-95 (65 days) and Sb-124 (60.2 days), had disappeared and Cs-137 (30.2 years) and Co-60 (5.27 years) activities were determined. Results are given in Table III.

TABLE III

Analyses (±S.D.) of water samples taken from cask and storage pool of spent fuels.

Sample no.	Cs-137 (661.6 keV) (Bq/l)	Co-60 (1173 and 1332 keV) (Bq/l)
1	3.70±0.74	1.85±0.37
2	4.44±1.11	1.48±0.37
3	36.26±5.18	6.66±0.74

DISCUSSION

Although for the transport regulation the measurement of the Cs-137 activities were enough, we have also measured the Co-60 activities as a further verification. According to the permissible limits, the Cs-137 activity difference between the two samples should be zero or in the order of the lower limit of detection and both results should be less than 0.037 Bq/ml. As can be seen from Table III, this is true for the Cs-137 activities of the first and second sample. As can also be seen from this table, statistically there is no difference between the two Co-60 activities. The activity of the sample taken from the spent fuel storage pool was higher. This may be explained by diffusion of activity from fuel cladding during the cooling time.

In conclusion, it was shown that spent fuel claddings had no crack, break or corrosion and, therefore, they could be transferred safely.

References

1. E. Gilat, Y. Laichter and N. H. Shafrir. In: *Proc. Radiol. Impacts of Releases from Nuclear Facilities into the Aquatic Environment*, Otaniemi, Finland, 1975, pp. 63–76, IAEA, Vienna, 1975, STI/PUB/406.
2. Health and Safety Laboratory, *HASL Procedures Manual HASL-300*, Sections D-07 (1977) and D-08 (1976), U.S. Atomic Energy Commission.
3. National Centre for Radiological Health, *Radioassay Procedures for Environmental Samples*, PHS Publication No. 999-RH-27, Washington, DC, 20402 (1967).
4. C. E. Crouthamel, F. Adams and R. Dams, *Applied Gamma-Ray Spectrometry* (Pergamon Press, Oxford, 1970), 2nd ed.

5. J. A. Gonzalez. In: *Proc. Intern. Symp. of Methods of Low-Level Counting and Spectrometry*, Berlin, 1981, pp. 59–74, IAEA, Vienna, 1981, STI/PUB/592.
6. Nuclear IEEE Standards, *IEEE Std.* 680 (1978).
7. J. A. Cooper, *Nucl. Instr. and Meth.* **82,** 273 (1970).
8. H. Hötzl and R. Winkler. In: *Proc. Intern. Symp. on Methods of Low-Level Counting and Spectrometry*, Berlin, 1981, pp. 77–91, IAEA, Vienna, 1981, STI/PUB/592.