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MEASUREMENT OF THE 46.5 keV GAMMA-RAY EMISSION PROBABILITY OF ^{210}Pb AND ITS APPLICATION AS A STANDARD FOR THE CALIBRATION OF ^{129}I MONITORS

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A reverse electrode coaxial Ge detector has been used to determine the 46.5 keV gamma-ray emission rates of several sources of ^{210}Pb. The effect of the escape peaks was carefully taken into account when determining the areas of the full-energy peaks for the calibration of the spectrometer. Source activities were measured by means of an improved $2\pi\alpha$ counting technique. The emission probability of the 46.5 keV gamma ray was found to be 0.0426 ± 0.0007. This nuclide can be used as a low-energy gamma-ray standard as demonstrated in the calibration of ^{129}I monitors.

1. Introduction

^{210}Pb(RaD) is often used as a standard source in alpha- and/or beta-particle studies, since the daughter nuclide ^{210}Bi(RaE) undergoes pure beta decay with a maximum energy of 1.16 MeV and the grand-daughter nuclide ^{210}Po(RaF) emits 5.3 MeV alpha-particles. Approximately 80% of the very soft beta emission from ^{210}Pb populate the 46.5 keV excited state of ^{210}Bi; the gamma transition from this nuclear level undergoes significant internal conversion to L-electrons so that the gamma-ray emission probability is only a few percent. No other gamma rays are observed in the decay chain of ^{210}Pb(RaD)–^{210}Bi(RaE)–^{210}Po(RaF)–^{206}Pb(RaG). Hence, the 46.5 keV gamma ray would be a useful standard if the gamma-ray emission probability per decay were known with reasonable accuracy. Furthermore, ^{210}Pb has a long half-life of 22.3 yr, making this radionuclide especially suitable as a gamma-ray standard.

Few measurements of the 46.5 keV gamma-ray emission have been reported using modern instruments, except for the studies by Debertin and Peßara [1]. Therefore, an accurate determination of this emission probability has been carried out with a combination of improved $2\pi\alpha$ counting and gamma-ray spectroscopy. A high-purity germanium detector was used in the spectroscopic studies; the crystal has a thin ion-implanted front contact and produces a nearly flat response below 100 keV. Subsequent studies have also shown ^{210}Pb to be a suitable gamma-ray standard for the calibration of ^{129}I monitors.

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2. Activity measurements of ^{210}Pb

2.1. Improved $2\pi\alpha$ counting system

The decay scheme for ^{210}Pb(RaD)–^{210}Bi(RaE)–^{210}Po(RaF)–^{206}Pb(RaG) is shown in fig. 1 [2]: ^{210}Pb decays to the excited and ground states of ^{210}Bi by two beta emissions with maximum energies of 15 and 61 keV, respectively. However, measurements of such low-energy beta-particles are unsuitable for the determination of source activities, and 2π counting of the alpha-particles emitted from grand-daughter ^{210}Po is preferred. Secular equilibrium is assumed to be established between the ^{210}Pb and ^{210}Po nuclides. The half-lives of ^{210}Bi and ^{210}Po are 5.0 are 138.4 d, respectively, and more than four years (1460 d) are required after source preparation before secular equilibrium will be established to an accuracy of better than 0.1%. Several ^{210}Pb sources were used (12 mm diameter) in the present study that had been electrodeposited 30 years ago in the form of PbO_2 on a palladium-coated silver disc.

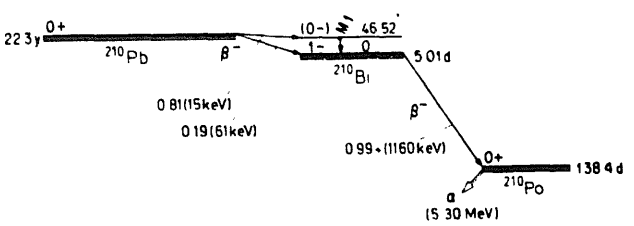

Fig. 1. The decay chain of ^{210}Pb(RaD)–^{210}Bi(RaE)–^{210}Po(RaF)–^{206}Pb(RaG) from ref. [2].

The top half of a rectangular box-shaped 4π counter was operated as a $2\pi\alpha$ counting system, using research grade methane (99.99%) as the flowing gas. This device had an α-plateau region extended from 2000 to 2500 V with a slope of 0.05%/100 V. Count rates at operating potentials of 2300, 2350, 2400 and 2450 V were averaged after extrapolating the data to the zero discrimination level. Under equilibrium conditions, the $2\pi\alpha$ count rate (n_α) can be related to the disintegration rate ($(n_0)_{Pb}$) of ^{210}Pb by the following equation:

$$(n_0)_{Pb} = 2n_\alpha \frac{(T_{1/2})_{Pb}}{(T_{1/2})_{Pb} - (T_{1/2})_{Po}} \left(\frac{1}{1-a}\right)\left(\frac{1}{1+b}\right), \quad (1)$$

where $(T_{1/2})_{Pb}$ and $(T_{1/2})_{Po}$ represent the half-lives of ^{210}Pb(22.3 yr) and ^{210}Po (138.4 d), respectively. The factors a and b represent the corrections for the self-absorption and backscattering of the alpha-particles.

2.2. Correction for self-absorption

Self-absorption (a) of a uniformly distributed source of superficial density (d) can be estimated by the equation:

$$a = d/(2R), \quad (2)$$

where R is the range of the alpha-particles in the source material [3]; R has a value of 16.5 mg/cm^2 for 5.3 MeV alpha-particles in PbO_2 [4]. The effective thickness (superficial density) of the source was determined by alpha-particle spectrometry using a 50 mm^2 Si surface barrier detector mounted in a vacuum chamber. Typical spectra are shown in fig. 2 for a thin ^{241}Am and an electrodeposited ^{210}Pb source. The distance from the source to the detector was 5 cm, and the energy resolution was 14 keV for the 5.486 MeV alpha-particles of ^{241}Am. A correction can be made for the intrinsic resolution to give a spectral line width that is a measure of the source thickness; the effective thickness (d) of the source material can then be obtained from

$$d = \sqrt{\Gamma_{obs}^2 - \Gamma_i^2}/S, \quad (3)$$

where Γ_{obs} and Γ_i are the observed and intrinsic line widths (FWHM), respectively, and S is the stopping power of the source material for the specified alpha-particle. The Bragg additivity rule and the tabulations of Ziegler [4] were used to calculate a stopping power of 294 keV cm^2/mg for 5.30 MeV alpha-particles in PbO_2. Measured thicknesses for the sources ranged from 0.59 to 1.10 mg/cm^2, corresponding to self-absorption values from 1.79% to 3.33%.

The validity of the data was checked by $2\pi\beta$ counting to measure the the energetic beta-emissions from ^{210}Bi. Both the alpha-particles from ^{210}Po and the low-energy beta-particles from ^{210}Pb were absorbed by covering the sources with an aluminum absorber (13.53 mg/cm^2). The ratio of the alpha and beta count rates was calculated with and without self-absorption corrections, and the results are shown in fig. 3: N_α/N_β is consistent to within 0.2% after correction (B), while considerable scatter occurs in the uncorrected data (A). This constancy in the ratio of the count rates supports the validity of the self-absorption correction process.

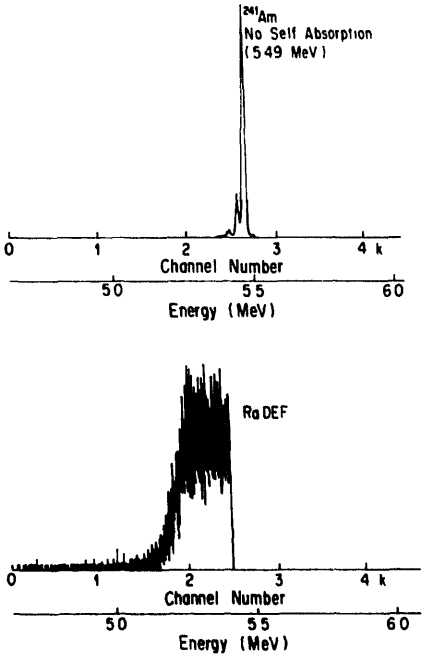

Fig. 2. Top: Alpha-particle spectrum for a thin ^{241}Am source measured with a Si surface barrier detector. Bottom: Alpha-particle spectrum for a ^{210}Pb source in equilibrium with ^{210}Po.

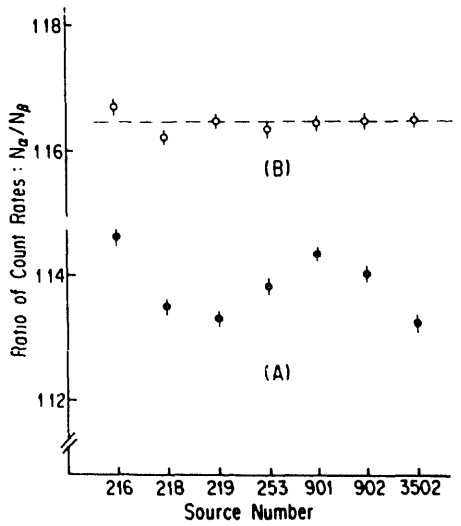

Fig. 3. Ratio of the $2\pi\alpha$-count rate of a bare source to the $2\pi\beta$-count rate of the same source covered with an aluminium foil of 13.53 mg/cm^2. (A) uncorrected for self-absorption; (B) corrected for self-absorption.

Table 1
Estimate of the overall uncertainty in the radioactivity measurements of ^{210}Pb

Sources of error	Uncertainty (1σ) in final result	
	Type A [%]	Type B [%]
Counting statistics	0.1	–
Self-absorption estimation	0.1	0.2
Backscatter estimation	–	0.3
Extrapolation to zero discrimination level	0.1	–
Equilibrium	–	0.1
Overall	0.17	0.38
	0.42	

2.3. Correction for backscatter

Extensive experimental studies have been undertaken by Hutchinson et al. [5] on the backscattering properties of the 5.30 MeV alpha-particle from ^{210}Po for various metals and for two different surface conditions. Results were compared from a Robinson and a 2π counter, and the backscatter was shown to be a linear function of the atomic number of the backing material.

A $2\pi\alpha-\gamma$ coincidence counting technique was used in the current studies to measure the backscatter of alpha-particles emitted from electrodeposited sources of ^{241}Am. The data were consistent with the results of Hutchinson et al. [5], except that they were not significantly dependent on the surface condition. Our experimental procedures and the results are described in the Appendix.

The source mounts were made of palladium-coated silver, and the backscatter correction was estimated to be 2.23% from the data described above. Scattered alpha-particles were emitted in a very small angle close to the surface of the mount, and their absorption can be treated separately as described by Ballaux [6]. However, the absorption of the scattered alpha-particles was neglected in this work since d/R was less than 0.07. Contributions to the uncertainty in the activity measurement are listed in table 1, and the overall value is estimated to be 0.4% (1σ).

3. Measurement of the 46.5 keV gamma-ray emission rate

3.1. HPGe spectrometer

The 46.5 keV gamma-ray emission probability was measured with a high-purity germanium spectrometer (EG&G Ortec LO-AX HPGe coaxial-type). Significant crystal sensitivity (to a depth of 13.3 mm), a thin inactive layer (less than 0.3 μm) and beryllium window (0.5 mm thick) ensure a nearly flat response in the energy range from several to 100 keV. The diameter of the active crystal is 35.7 mm, with an energy resolution of better than 270 eV FWHM at 5.9 keV and 540 eV FWHM at 122 keV. A horizontal cryostat cools the detector, which is shielded in a box of 6 cm thick lead lined with 5 cm of old steel, 5 mm copper and 5 mm Perspex. The distance between the source and cryostat surface is maintained at 20 cm by means of a cylindrical aluminum jig. Pulse-height analysis was carried out using a Seiko–EG&G 7800 multichannel analyser with a memory size of $2 \times 4K$.

3.2. Calibration

The amplifier gain of the HPGe spectrometer was set to give 0.073 keV/channel. Appropriate point sources of 10^4 Bq were standardised by means of a $4\pi\alpha-\gamma$ or $4\pi X-\gamma$ coincidence technique (table 2), and they were used to calibrate the spectrometer. Five standards were prepared of each radionuclide; each source was covered with 0.1 mm (27 mg/cm^2) thick aluminum, and corrections were made for the absorption in this aluminum cover, the air between the source and the detector, and

Table 2
Standard sources to calibrate the HPGe spectrometer

Nuclides	Method of standardisation	Uncertainty [%]	Energy [keV]	Emission probability [6, 7*]
^{111}In	$4\pi X-\gamma$	0.45	22.3 (K_α)	0.679 ±0.025
			26.0 (K_β)	0.145 ±0.006
			171.28 (γ)	0.903 ±0.009*
			245.35 (γ)	0.939 ±0.006*
^{139}Ce	$4\pi X-\gamma$	0.40	33.3 (K_α)	0.647 ±0.021
			165.86 (γ)	0.7999±0.0016*
^{241}Am	$4\pi\alpha-\gamma$	0.20	59.54 (γ)	0.359 ±0.004
^{133}Ba	$4\pi X-\gamma$	0.65	80.99 (γ)	0.346 ±0.008*
^{57}Co	$4\pi X-\gamma$	0.30	122.06 (γ)	0.8533±0.0027*
			136.47 (γ)	0.1085±0.0008*

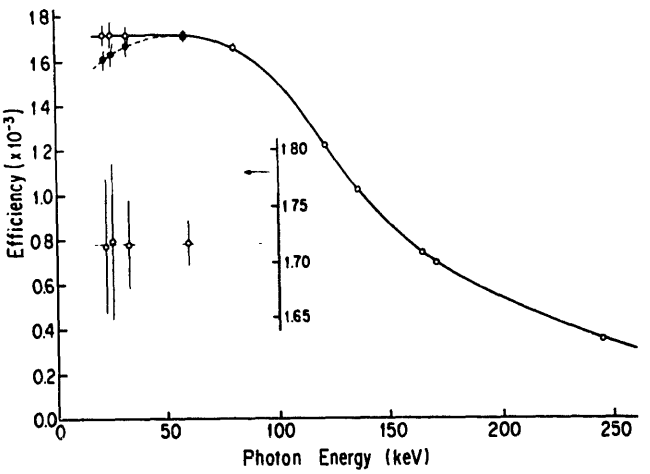

Fig. 4. Efficiency calibration of the HPGe spectrometer corrected for absorptions in the aluminum cover of the sources, in the air, and in the dead layer of the germanium crystal (source–detector distance of 20 cm). The dotted line is for the full-energy peak, while the solid line is for the full-energy peak plus the escape peak.

the dead layer of the germanium crystal. The peak-area analyses involved a subtraction of the background continuum, and consideration of the different shapes of the X- and gamma-ray peaks due to the large differences in the natural line widths [8]. No collimator system was used during these experiments, and corrections were made for the coincidence summing effect in the spectra of ^{111}In and ^{139}Ce. The full-energy peak efficiency curve is shown in fig. 4 as a dotted line that decreases with decreasing photon energies below 30 keV. This effect is mainly caused by the escape of the K-X-rays of germanium from the crystal; the escape peak is observed in the pulse-height spectrum approximately 10 keV below the full-energy peak, and the loss of peak area in the efficiency curve can be corrected by adding the area of this escape peak. The solid line in fig. 4 represents the complete efficiency curve after the escape peak correction. This curve is nearly constant over the energy range of interest, and therefore increases confidence in the reliability of the efficiency interpolation [9]. It should also be noted that the accuracy of the calibration curve in the region about 50 keV is determined primarily by the 59.5 keV gamma-ray emission from the ^{241}Am standard. The efficiency at 46.5 keV was determine to be $(1.716 \pm 0.023) \times 10^{-3}$ (1σ) by a least squares fitting of the data to a linear function.

3.3. Measurement of ^{210}Pb gamma-ray spectrum

An example of the pulse-height spectrum for ^{210}Pb is shown in fig. 5. The gamma-ray emission rate for each ^{210}Pb source was determined with reference to the calibrated efficiency. A correction of 0.045% was made to the data to allow for the difference in size between the ^{210}Pb sources (12 mm diameter) and the point sources used in the calibration.

The gamma-ray spectra for four ^{210}Pb sources were accumulated over a total counting time of 300000 s for each source (~ 600 Bq). An emission probability of 0.0426 ± 0.0007 (1σ) was determined for the 46.5 keV gamma ray, after correction for X-ray escape (1.3%) and attenuation in the aluminum cover (0.60%), in the air (0.09%) and in the dead layer of the crystal (0.06%). The stated uncertainty (1σ) was estimated from the various components listed in table 3, and the resulting data are compared with other measured values and evaluations

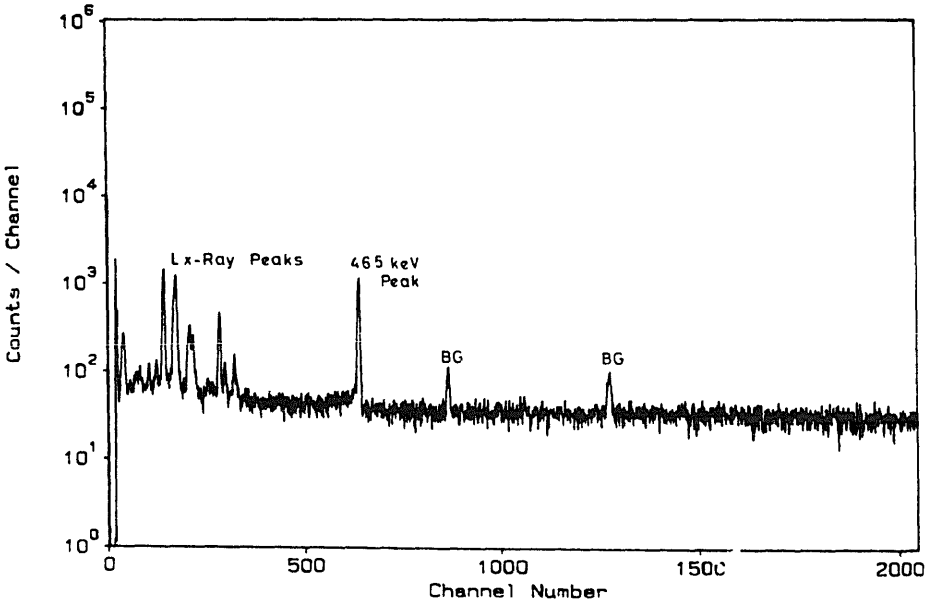

Fig. 5. Pulse-height spectrum for ^{210}Pb measured with the LO-AX HPGe spectrometer (counting time: 150000 s).

Table 3
Estimation of the overall uncertainty in the 45.6 keV gamma-ray emission probability

Source of error	Uncertainty (1σ) in final result	
	Type A [%]	Type B [%]
Decay data of standard source	–	1.4
Interpolation	–	0.1
Statistics of calibration points	0.43	–
Statistics of ^{210}Pb spectroscopy	0.55	–
Escape peak correction	0.1	0.2
Absorption corrections for		
Al cover	–	0.1
air	–	0.05
dead layer	–	0.05
Radioactivity measurement of ^{210}Pb	0.17	0.38
Overall	0.73	1.50
	1.65	

in table 4. Within the stated uncertainties, our value is in agreement with the data reported by Debertin and Peßara [1], which was also adopted in NCRP-58 [3]. The other evaluations [13–15] recomended lower values that are based on the older measurement of Krause [12].

4. Application of ^{210}Pb as a standard for the calibration of ^{129}I monitors

^{129}I is an important radionuclide in environmental surveillance, with a very long half-life of 1.57×10^7 yr. Routine measurements can be performed by monitoring for the 39.6 keV gamma-ray (emission probability 0.075 ± 0.002 [14]) with a low-energy photon spectrometer (LEPS) or a scintillation detector. ^{210}Pb could be used to calibrate spectrometers designed for ^{129}I measurements, because the gamma-ray emissions from these two nuclides have an energy difference of only 6.9 keV, and most of photon detectors have a nearly flat response in this region. Satisfactory results have been obtained by preparing standard sources of ^{210}Pb distributed uniformly in a charcoal filter for the calibration of ^{129}I dust monitors.

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Appendix

Measurements of the backscatter of 5.48 MeV alpha-particles from metals

An electrodeposited source of ^{241}Am with negligible self-absorption can be used in conjunction with $2\pi\alpha$–γ coincidence counting to determine the backscatter of the 5.48 MeV alpha-particles from the source mount. The backscatter (b) can be calculated from the equation:

$$b = (n_c/n_\gamma - 0.5)/0.5,$$

where n_c and n_γ are the count rates in the coincidence- and gamma-channels after corrections for accidental coincidences and dead time effects [6]. This technique was used in the current studies to measure the backscatter of 5.48 MeV alpha-particles from polished and unpolished mounts of aluminum, stainless steel, copper, palladium, silver and platinum. A Buchler Ecomet III lapping machine was used to polish some of the mounts with 6 μm diamond powder and 1 μm alumina powder on a buff. ^{241}Am was dissolved in ammonium sulphate

Table 4
Emission probability of the 46.5 keV gamma ray: Comparison of the present result with other reported measurements and evaluations

Author/reference	Method	Emission probability
Damon and Edwards (1954) [10]	thin NaI scintillation spectrometer and α proportional counter	0.038 ± 0.006
Fink (1957) [11]	NaI scintillation spectrometer and α proportional counter	0.045 ± 0.004
Krause (1958) [12]	NaI scintillation spectrometer and ZnS scintillation counter	0.0405 ± 0.0008
Debertin and Peßara (1981) [1]	Ge spectrometer	0.0418 ± 0.0008
Nucl. Data Sheets (1981) [13]	evaluation	0.0405 ± 0.0008
Lagoutine et al. (1985) [14]	evaluation	0.0406 ± 0.0008
Table of Radioactive Isotopes (1986) [15]	evaluation	0.0405 ± 0.0020
NCRP Report No. 58 (1985) [3]	evaluation	0.0418 ± 0.0023
Present work	see text	0.0426 ± 0.0007

Note added in proof: U. Schötzig [16], this Symposium, emission probability of 0.0424 ± 0.0005.

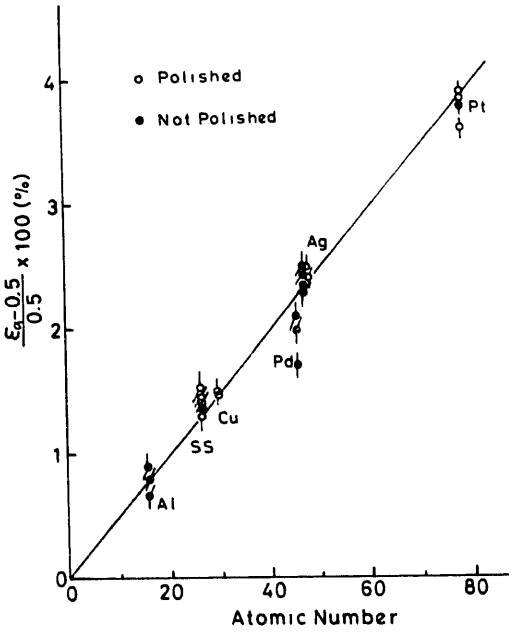

Fig. 6. Backscatter measurements for 5.48 MeV ^{241}Am alpha-particles by the $2\pi\alpha-\gamma$ coincidence counting technique (open circles represent polished source mounts, and the closed circles denote unpolished source mounts).

solution and electrodeposited onto these metal backings (source diameter 20 mm, thickness 0.15 $\mu g/cm^2$). Finally, $2\pi\alpha$ γ coincidence measurements were undertaken by means of a conventional $4\pi\beta-\gamma$ coincidence counting system.

The results are shown in fig. 6, in which the open circles represent the polished mounts and the closed circles denote the unpolished mounts. While there is good agreement between these data and the measurements of Hutchinson et al. [5] for polished mounts, the considerable deviations reported in ref. [5] for unpolished mounts are not apparent in the current studies. The surface effects noted in the earlier experiments were found to be less important as shown by the close proximity of the corresponding open and closed circles in fig. 6.

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