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NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH
Section A

ELSEVIER

Alpha-particle and electron capture decay of ^{209}Po

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Abstract

Gamma-ray and Kα X-ray emissions have been measured from a very pure ^{209}Po source containing less than 0.13% ^{208}Po activity and no detectable ^{210}Po ($\leq 2 \times 10^{-4}$%). The alpha-particle emission rate for this source has previously been determined. Data are presented that confirm alpha decay to the ^{205}Pb excited level at 262.8 keV, with an alpha-particle emission probability (±standard uncertainty) of 0.00559±0.00008. The ratio of K-shell electron capture to total electron capture for the second forbidden unique electron capture decay to the 896.6 keV level in ^{209}Bi was determined to be 0.594±0.018. The electron capture decay fraction was found to be 0.00454±0.00007, while the probabilities per decay for the 896.6, 262.8, and 260.5 keV gamma rays and the Bi Kα and Pb Kα X-rays were measured as 0.00445±0.00007, 0.00085±0.00002, 0.00254±0.00003, 0.00202±0.00005, and 0.00136±0.00005, respectively.

1. Introduction

Considerable efforts have been made to analyze and determine the photon-emission impurities associated with the calibration and production of ^{209}Po solution standards. These standards have been described by Collé et al. [1], and are available [2] as a NIST Standard Reference Material (SRM) 4326. ^{209}Po decay is predominantly by α-particle emission with a weak electron capture branch (second forbidden unique transition) to the first excited state of ^{209}Bi. This pursuit was initially motivated by four issues: (1) the low contaminant level of other Po radionuclides in the present ^{209}Po stock material which allowed precise determinations of the photon emission rate, particularly for the Pb and Bi Kα X-rays; (2) the possibility of refining the ^{209}Po decay scheme by measurements of a source material that had a well-known total α-particle emission rate; (3) discrepancies in the major γ-ray emissions as indicated in the evaluation of Martin [3]; and (4) previous reports of very weak α-particle decay branches whose existence is questionable.

In regard to the first issue of radionuclidic purity, the source material available for this study was determined [1] to contain only 0.00124±0.00010 (value±standard uncertianty) ^{208}Po relative to ^{209}Po activity ratio and no detectable ^{210}Po, with a limit of $\leq 2 \times 10^{-6}$ relative to ^{209}Po. The previous investigations of ^{209}Po decay were based on source material containing ^{208}Po/^{209}Po activity ratios at levels from 1 to 20 [4,5]. Mandal et al. [5] made measurements on source material that contained even larger activity ratios of ^{210}Po/^{209}Po.

Hagee et al. [4] and Mandal et al. [5] deduced the relative intensity ratio of the α-particle emission to the 262.8 keV level and the ground state doublet ($P_{\alpha 2}/P_{\alpha 1+\alpha 0}$) and the P_{EC}/P_{α} decay ratio. The P_{EC}/P_{α} ratio was reported to be 0.0026 by Hagee et al. [4], while Mandal et al. [5] obtained a value of 0.0048; Hagee et al. [4] reported a value of 0.0048 for the α-particle branch ratio $P_{\alpha 2}/P_{\alpha 1+\alpha 0}$ and Mandal et al. [5] obtained a value of 0.0092 as deduced from the γ-ray measurements. There is nearly a factor of 2 difference in the respective ratios as reported. Furthermore, Hagee et al. [4] reported two additional α-particle decay branches to levels in ^{205}Pb at 576 keV (1.5×10^{-6}) and 761 keV (5.56×10^{-6}). Mandal et al. [5] measured the Bi K X-rays and was able to deduce the $P_{L+M+N\cdots}/P_K$ capture ratio, which was in good agreement with the theoretical predictions of Behrens and Bühring [6].

The results reported below build on previously published work from this laboratory [1,7]. These studies take advantage of ^{209}Po source material that is very pure in terms of radionuclidic impurities and is gravimetrically related to an SRM solution with a well defined total α-particle emission rate. The results of the photon emission measurements are presented, including the Kα X-ray doublet of Pb and Bi. Fractional decay branches and

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probabilities per decay are deduced, and a decay scheme is presented.

2. Detectors and measurement conditions

The X-ray and gamma-ray emission probabilities were measured on two different detectors: N (intrinsic n-type) and P (intrinsic p-type) are both coaxial and manufactured from high-purity germanium. All of the significant characteristics are listed in Table 1. The N detector was used for the measurement of the $K\alpha$ X-ray emission rates; this detector is shielded by low-radiogenic lead and has a graded X-ray shield of tin and copper (background runs indicated virtually no Pb $K\alpha$ X-rays and a small but determinable Bi $K\alpha$ X-ray emission).

The source-to-detector distances for the N and P detectors were 10 cm and 25 cm respectively. These distances were chosen so as to reduce correlated summing corrections to values less than a factor of 1.01, and make use of source-to-detector positions that have well-characterized efficiency curves. The source was mounted along the axis of the particular detector crystal in a reproducible manner. When background runs were obtained, the same spacers and source holders were left in front of the detector without the ^{209}Po source present. Three solid point sources, ranging from 401 to 1035 Bq, were prepared by depositing gravimetrically-determined amounts of the stock solution used to prepare SRM solution 4326 on polyester tape of 8.1 mg cm^{-2} thickness, evaporating the deposits to dryness, and covering with the same thickness of polyester tape. The standard deviation of the source measurements was better than 0.05%, and the dry material formed disks no larger than 6 mm diameter. The emission rate measurements were made with the largest activity source, although all three sources (about 2 kBq) were used in the search for depopulating γ-ray emissions from 761 and 576 keV excited levels in ^{205}Pb as would be expected from the results of Hagee et al. [4].

The efficiency calibration for the two germanium detectors is based on NIST SRMs for ^{57}Co, ^{133}Ba, ^{137}Cs and ^{154}Eu. The latter SRM is a gamma-ray emission rate standard, while the first three are activity standards; the gamma-ray emission probabilities from the 1991 IAEA evaluation [8] were adopted.

Table 1
Characteristics of high-purity germanium detectors

	N	P
Type	n-type	p-type
Dimensions	43.6 mm diameter 36.2 mm length	49.9 mm diameter 41.2 mm length
Relative efficiency at 1.33 MeV	9.0%	16.0%
Absorbing layers	0.5 mm Be, ~0.3 μm Ge	1.27 mm Al, 0.7 mm Ge
Pulse shaping time constant	5 μs	4 μs

3. Results

Data were accumulated for a total of about 24 000 min on the N detector and 16 000 min on the P detector. Additionally, for the X-ray analysis, 12 000 min of background spectra were obtained on the N detector. Throughout these measurements, the full width at half maximum of the N detector remained constant at 0.89 keV for the 77.1080 keV Bi $K\alpha_1$ X-ray line and 1.40 keV for the 896.6 keV γ-ray. In terms of peak counts, the most intense lines recorded were the Pb $K\alpha_1$–$K\alpha_2$ and Bi $K\alpha_1$–$K\alpha_2$ X-ray line quartet, which were observed as three peaks in ascending energy order: an easily resolved Pb $K\alpha_2$ at 72.804 keV; an unresolved doublet, the Bi $K\alpha_2$–Pb $K\alpha_1$ at 74.89 keV; and another easily resolved Bi $K\alpha_1$ at 77.108 keV (see Fig. 1). Each of these three peaks was fitted to a single Gaussian shaped photon peak that yielded a corresponding peak count rate. Given the well-characterized and calculable emission ratios for $K\alpha_2/K\alpha_1$ X-rays (as from Scofield [9]) and the three peak X-ray emission rates, one can solve three equations in two unknowns to obtain the individual Pb $K\alpha$ and Bi α X-ray emission rates. The total K X-ray emission rates were deduced from these $K\alpha$ X-ray emission rates using the $K\beta/K\alpha$ ratios from Scofield [9].

The three γ-ray emission rates were measured on the N and P detector systems, and were combined to give grand weighted mean values. Table 2 lists the results of these measurements expressed as the γ-emission rate for the largest activity point source.

Another p-type intrinsic high-purity Ge detector [10] was made available for a short period of time to search for

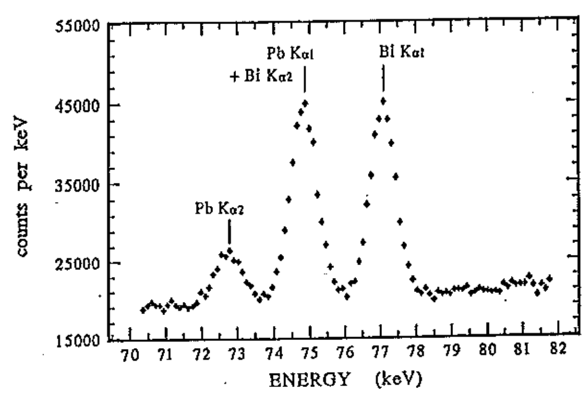

Fig. 1. $K\alpha$ X-Ray peaks for Pb and Bi from the decay of ^{209}Po; the Pb $K\alpha_2$ and Bi $K\alpha_1$ are clearly resolved with an n-type high-purity Ge detector, and the unresolved Pb $K\alpha_1$ and the Bi $K\alpha_2$ peaks were fitted as a single line.

Table 2
Summary of measured X-ray and γ-ray emission rates and the corresponding emission concentrations for the stock solution of ^{209}Po activity

Quantity	Emission rate $[s^{-1}]$	Symbol	Emission rate concentration $[\gamma s^{-1} g^{-1}]$
Pb Kα X-rays	1.40786±0.05018		
Pb K X-rays[a]	1.80501±0.06433	$R_{KX\,Pb}$	0.1496±0.0053
Bi Kα X-rays	2.08936±0.05138		
Bi K X-rays[a]	2.70107±0.06642	$R_{KX\,Bi}$	0.2239±0.0055
260.5 keV[b]	2.6278 ±0.0268	$R_{\gamma 1}$	0.2178±0.0022
262.8 keV[b]	0.8784 ±0.0207	$R_{\gamma 2}$	0.0728±0.0017
896.6 keV[c]	4.6021 ±0.0732	$R_{\gamma 3}$	0.3815±0.0061
α-particles, all energies[d]	–	R_α	85.41 ±0.145

[a] As only the Kα X-ray emission were measured, the total K X-ray emission was deduced from the ratio of Kβ/Kα of Scofield [9].
[b] γ-ray emitted from the depopulation of an excited level in ^{205}Pb.
[c] γ-ray emitted from the depopulation of an excited level in ^{209}Bi.
[d] Emission rate concentration value from Ref. [1].

possible γ-rays that would depopulate the levels in the very weak α-particle branches reported by Hagee et al. [4]. All three point sources were placed on the endcap of the detector and a spectrum accumulated for 11 400 minutes. No other γ-rays, other than those listed in Table 2, were observed from these sources. If one assumes that the depopulation ratios for any such transitions are as found in the β-particle decay of ^{205}Bi to levels in ^{205}Pb [11], a doublet at 759 and 761 keV would be expected with a relative intensity ratio of 3 to 2. They would have an expected peak area 2.3 times the standard uncertainty in the underlying spectral background for the more intense 759 keV photopeak. No such peaks were observed (see Fig. 2). Hence, there was no evidence for the existence of this α-particle branch, with a limit of $<1.8\times10^{-6}$, which is consistent with the α-particle decay hindrance factors [3].

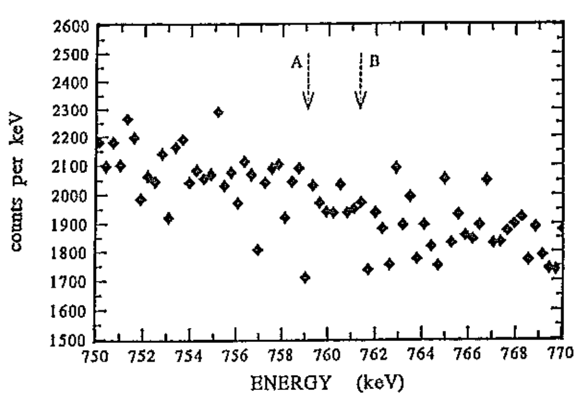

Fig. 2. Detail of the energy region around 760 keV of the γ-ray emissions from all three of the solid ^{209}Po sources. Arrow A is located in the position where the 759.10 keV γ ray would be found, and arrow B is the location of the 761.35 keV γ ray. These two γ rays are the most intense transitions that would depopulate the 761.35 keV level if it were fed by α-particle decay.

As far as the α-particle branch to the 576 keV level in ^{205}Pb, again there is no evidence of any depopulating γ-rays; the analysis of the spectral data yields a limit of $<1.5\times10^{-6}$ for this α-particle branch.

Table 2 summarises the X-ray and γ-ray measurements. Both the photon emission rate per source as well as the emission rate concentrations for the ^{209}Po solution are given. The uncertainties are the estimated standard deviations of the grand weighted mean values; and the estimated standard deviations on the mean values for each detector include the estimated standard deviations of their respective efficiency values which were combined in quadrature. Table 3 lists the typical uncertainty components that were included in the measurement of the 260.5 keV γ-ray. The α-particle emission rate concentration from Ref. [1] is included for comparison in Table 2. On the basis of the

Table 3
The list of relative standard uncertainty components (in percent) for the evaluation of the 260.5 keV gamma-ray emission rate

Component[a]	Detector	
	N	P
Variation in counting[b] (number of measurements)	1.02(6)	2.65(3)
Efficiency function[c]	0.29	2.11
Source position[d]	0.12	0.05
Peak analysis[d]	0.04	0.06
Combined, in quadrature	1.07	3.41

[a] Relative standard uncertainty components arising from half-life, counting losses and source self-attenuation are estimated to be less than 0.01%, and therefore ignored.
[b] Relative standard uncertainty determined from repeated measurements.
[c] Relative standard uncertainty determined from the fit of the efficiency function.
[d] Estimated at the one sigma value.

emission rate concentration quantities defined in Table 2, the following probabilities per decay (P) for the ^{209}Po α-particle and electron capture decay branches may be directly derived:

$$P_{\alpha 2} = (R_{\gamma 1}(1+\alpha_{T1}) + R_{\gamma 2}(1+\alpha_{T2}))/A\,, \quad (1)$$

$$P_{\alpha 1+\alpha 0} = (R_{\alpha} - R_{\gamma 1}(1+\alpha_{T1}) - R_{\gamma 2}(1+\alpha_{T2}))/A\,, \quad (2)$$

and

$$P_{\alpha 2}/P_{\alpha 1+\alpha 0} = (R_{\gamma 1}(1+A_{T1}) + R_{\gamma 2}(1+\alpha_{T2}))/ \times (R_{\alpha} - R_{\gamma 1}(1+\alpha_{T1}) - R_{\gamma 2}(1+\alpha_{T2}))\,, \quad (3)$$

in which A is the ^{209}Po activity concentration;

$$P_{\alpha} = R_{\alpha}/A, \quad \text{and} \quad P_{EC} = R_{\gamma 3}(1+\alpha_{T3})/A\,,$$

hence

$$P_{\alpha} + P_{EC} = 1\,,$$

or

$$A = R_{\alpha} + R_{\gamma 3}(1+\alpha_{T3})\,,$$

where α_{T1}, α_{T2} and α_{T3} are the total internal conversion coefficients for the 260.5, 262.8 and 896.6 keV γ-rays, respectively.

The expression for the ratio of K-shell electron capture to total electron capture (P_K) may be written as

$$P_K = (R_{KX}\mathrm{Bi} - \alpha_{K3}\omega_{KBi}R_{\gamma 3})/((1+\alpha_{T3})\omega_{KBi}R_{\gamma 3})\,, \quad (4)$$

where ω_{KBi} is the K-shell fluorescence yield for Bi, and α_{T3} and α_{K3} are the total and K-shell internal conversion coefficients for the 896.6 keV γ-ray. The Pb K X-ray emission rates are given by:

$$R_{KX\,Pb} = R_{\gamma 1}\alpha_{K1}\omega_K + R_{\gamma 2}\alpha_{K2}\omega_K\,, \quad (5)$$

where α_{K1}, α_{K2} and ω_K are K shell internal conversion coefficients for the 260.5 and 262.8 keV transitions and the K-shell fluorescence yield for Pb, respectively. All emission rate concentrations in Eq. (5) were measured, and furthermore the 260.5 and 262.8 keV transitions have similar multipolarity so that the K-shell internal conversion coefficients will be similar in value, or $\alpha_{K1}/\alpha_{K2} = F$. The experimental studies by Hamilton et al. [11] of the β-particle decay of ^{205}Bi give a value of 1.11±0.22 for this ratio, and the calculated M1 K-shell internal conversion coefficient [12] yields a ratio of 1.025. If $F = \alpha_{K1}/\alpha_{K2}$ is estimated by the latter value, it is possible to solve for each of the K-shell internal conversion coefficients. Thus, one can write the following expressions:

$$\alpha_{K1} = R_{KX\,Pb}/(\omega_{KPb}(R_{\gamma 1} + R_{\gamma 2}/F))\,, \quad \text{and}$$

$$\alpha_{K2} = R_{KX\,Pb}/(\omega_{KPb}(FR_{\gamma 1} + R_{\gamma 2}))\,, \quad (6)$$

where the quantities are defined as for Eq. (5).

Table 4 lists the various emission probabilities per decay and other significant ratios obtained from the current measurements. These data were obtained using the theoretical internal conversion coefficients from Ref. [12], and the evaluated fluorescence yields from Hubbell et al. [13]. The corresponding results of Mandal et al. [5] and Hagee et al. [4] are also presented for comparison. Fig. 3 represents the decay scheme based on the measurements from this laboratory.

Table 4
Comparison of the probabilities per decay and decay mode branches with other publications

Quantity[a]	Present work	Mandal et al. [5]	Hagee et al. [4]
P_{α}	0.9955 ±0.0001	0.9952	0.9974
$P_{\alpha 2}$	0.00551±0.00006	0.0092±0.0005	0.0056
$P_{\alpha 1+\alpha 0}$	0.9900 ±0.0001	0.986	0.9926
$P_{\alpha 2}/P_{\alpha 1+\alpha 0}$	0.00556±0.00006	0.0093	0.0049
P_{EC}	0.00454±0.00007	0.0048	0.00263
P_{EC}/P_{α}	0.00456±0.00007	0.0048	0.00264
P_K	0.594 ±0.018	0.70±0.05	–
$P_{\gamma 1}$	0.00254±0.00003	(100)[b]	(0.00391)[c]
$P_{\gamma 2}$	0.00085±0.00002	(33.3)[b]	(0.00391)[c]
$P_{\gamma 3}$	0.00445±0.00007	(108.1)[b]	0.00263
$\alpha_K(260.5)$	0.538 ±0.020	0.49±0.05[d]	0.495±0.001[d]
$\alpha_K(262.8)$	0.524 ±0.020		

[a] Refer to Table 2 for the definition of these symbols.
[b] Ref. [5] quoted relative intensities, normalized to the 260.5 keV γ-ray as 100.
[c] Ref. [4] was unable to resolve the 260.5 and 262.8 keV γ-ray doublet.
[d] Reported as a combined 260.5 and 262.8 keV transition K-shell conversion coefficient.

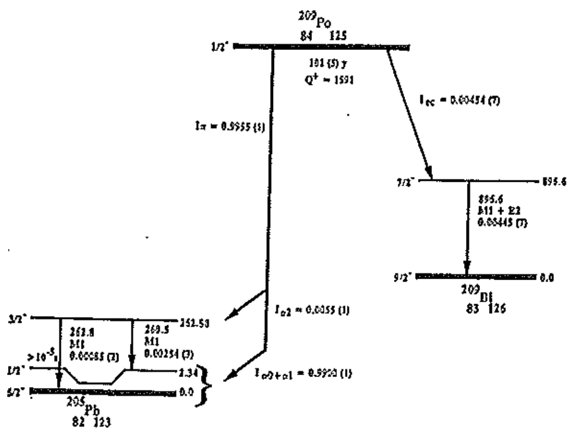

Fig. 3. Proposed decay scheme, in which all of the probabilities per decay arise from the measurement of the laboratory. The other nuclear data are from Martin [3].

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