Applied Radiation and Isotopes 200 (2023) 110953

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Measurement of the intensities of the long-range alpha particles from ^{212}Po

A. Martín Sánchez *, R. Mora Rodríguez

Departamento de Física, Universidad de Extremadura, E-06006, Badajoz, Spain

ARTICLE INFO

Keywords:
Long-range alpha particles
^{212}Po
Excited energy-levels
Nuclear decay scheme
Alpha spectrometry

ABSTRACT

^{212}Bi partially decays by β^- populating excited levels of ^{212}Po. Some of these excited states of ^{212}Po decay with very low probability by direct alpha-particle emissions instead of a gamma-alpha cascade. This effect was known since the earliest times after the discovery of radioactivity. Emission energies of these long-range alpha particles were measured in the past, but the activity ratios were not accurately determined. Relative intensities for these decays have now been experimentally determined. Results agree with data previously reported. It is the first time that an uncertainty estimate is provided for such experiment.

1. Introduction

Among the radiations from ^{212}Bi sources, Rutherford and Wood (1916) noted a few particles whose range is about 11.6 cm in air, markedly in excess of the 8.6 cm range of the well-known 8.78517 (11)-MeV alpha-particle emissions (Nichols, 2011) from ^{212}Po to ^{208}Pb. It is now well known that these long-range alpha particles are emitted from excited levels of ^{212}Po (Evans, 1985). In this nucleus the partial lifetime for alpha decay is comparable with the partial lifetime for gamma decay (about 300 ns for both cases). Consequently, a small fraction of the excited ^{212}Po nuclei will undergo alpha decay directly from an excited level to the ground level of ^{208}Pb (Fig. 1).

Although experimental measurements have been performed since long-range alpha particles were discovered, not many revisions for results have been made. The goal of this paper is the experimental check of the relative number of these particles compared with the most usual (connecting ground levels from both ^{212}Po and ^{208}Pb) α_0 transitions. Experimental results include the corresponding uncertainty budget.

2. Historical review

Rutherford and Wood (1916), studying samples of ThC (^{212}Bi), measured ranges of 5 cm for alpha particles (emitted from ^{212}Bi) and 8.6 cm (from ^{212}Po) in air. They also found two homogeneous sets of alpha particles, with ranges 10.2 and 11.3 cm in air. The number of these long-range alpha particles was about 1/10000 of the total particles emitted by ThC, of which 2/3 of this proportion corresponds to the particles with 11.3 cm, and 1/3 to those with 10.2 cm.

After this discovery, measurements of these ^{212}Po long-range alpha-particle groups were independently performed by Rutherford (1921), and Wood (1921), using mica and aluminium absorbers. Bates and Rogers (1923, 1924) measured by scintillation these long-range alpha-particle groups also reporting particles with longer range, but objections were raised against their methods, and the particles with range greater than 11.5 cm were later discarded. New range measurements were made by Meitner and Freitag (1926), and Nimmo and Feather (1929) using Wilson chambers, reordering the particles with greater range in only two groups (see Table 1), but they showed discrepancies about the relative intensities of these two groups. Their results were later checked by Rutherford et al. (1931), although in this work only two groups of long-range alpha particles from ThC' (^{212}Po) were observed at that time (see Table 1). Emission energies for the long-range alpha particles were estimated in these works. Rosenblum and Valadares (1932) studied alpha spectra giving some results for the long-range alpha-particle groups (indicating the possible existence of a third group), which were later checked by Lewis and Bowden (1934) with magnetic spectrometers. Energies and relative intensities for the long-range alpha-particles from ^{212}Po were summarized by Rytz (1953) in a classical work. Briggs (1954) tabulated three long-range alpha--particles from ThC' following the studies performed previously by Rytz (1951).

Based on several works after the tables by Rytz (1953), Emery and Kane (1960) presented the level scheme including not only the energy values and transition intensities, but also spin-parity assignments, making a global evaluation considering the electromagnetic character of the gamma transitions involved. Some calculations considering the

* Corresponding author. Departamento de Física, Universidad de Extremadura, E-06006, Badajoz, Spain.
E-mail address: ams@unex.es (A. Martín Sánchez).

https://doi.org/10.1016/j.apradiso.2023.110953
Received 9 May 2023; Received in revised form 21 June 2023; Accepted 17 July 2023
Available online 19 July 2023

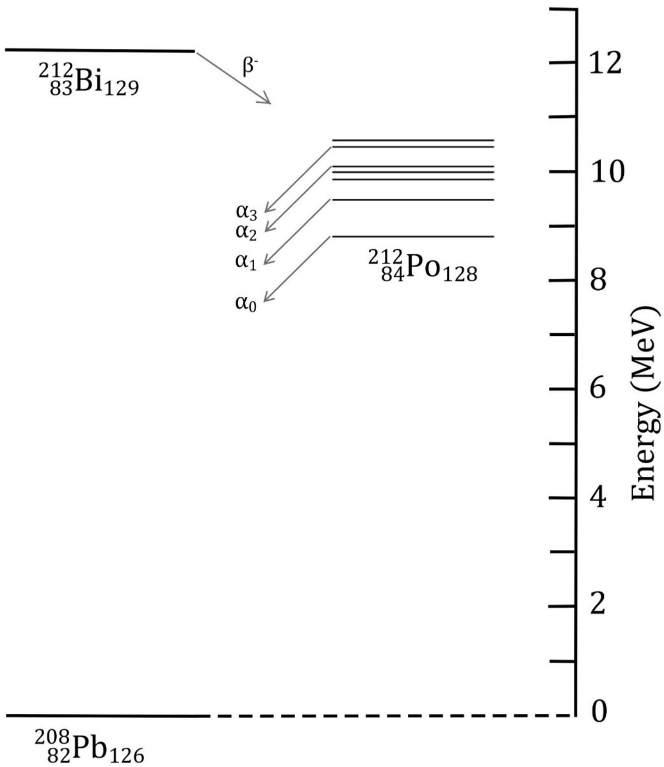

Fig. 1. Simplified decay scheme of the final part of the thorium series.

alpha barrier penetrability were performed by Griffioen and Rasmussen (1961) estimating some slight theoretical changes in the energy values.

Experimental measurements using coincidence techniques were made by Flack and Johnson (1962), but they only studied the associated gamma emissions. Bertolini et al. (1962) obtained experimental results for the relative intensities of these long-range alpha particles measuring with an ionization chamber and solid-state detectors. A revision of these data was performed by Leang (1965), who also measured energies and intensities by magnetic spectrometry. These values have remained practically unchanged in the tables and compilations published thereafter. Surprisingly, in all the experimental works, and even in the compilations, quoted relative intensity values were given without uncertainties, presumably because the quantities involved for the abundance of the long-range alpha particles and those decaying from the ground level of ^{212}Po (α_0) differ by 4 orders of magnitude.

Pancholi and Martin (1972) adopted values for the relative intensities of the long-range alpha particles as the unweighted average of several former works (see Table 1). In fact, later compilations of data have continued giving practically the same value for relative intensities (without uncertainties), although some adjustments for the energy-emission values have been reported (Pancholi and Martin, 1972; Browne et al., 1978; Martin, 2007; Browne and Firestone, 1986). Long-range alpha-particle emissions from ^{212}Po were not included in the nuclear data tables edited by Rytz (1979, 1991). Recent evaluations (Artna-Cohen, 1992; Browne, 2005; Martin, 2007) have found values that are practically the same as those previously tabulated.

However, in the Nuclear Data Tables from LNHB, a global weighted uncertainty (see Table 1) for the total group of long-range alpha particles has been estimated by Nichols (2011) taking into consideration the data given formerly by Rytz (1951), Bertolini et al. (1962) and Leang (1965). Now, in this work, experimental measurements for the relative intensities of the long-range alpha particles from ^{212}Po are presented.

3. Materials and methods

The technique of alpha-particle spectrometry was used to make the measurements. The source provided by CIEMAT was a collimated disc with an active area of 20 mm^2, containing 258 ± 10 Bq of ^{232}U in equilibrium with its daughters. A 50 mm^2 silicon-PIPS detector from Canberra was placed in front of the source, with a source-to-detector distance of 65 mm inside a vacuum chamber with low geometry (Caro Marroyo et al., 2014). A magnet was positioned between the source and the detector to discriminate alpha-beta coincident events, as described in Section 4.

Fig. 2 shows an example of the alpha-particle spectra obtained. The three energy regions for the long-range alpha particles (from ^{212}Po) are indicated. However, the analysis of the measured spectra presented several difficulties.

4. Experimental problems

Compared with α_0 emissions, long-range alpha particles are relatively rare. In addition, several experimental problems are present. A first problem was that possible alpha-gamma coincidences could arise, and then, this point was studied using the device described elsewhere (Caro Marroyo et al., 2014). Alpha-gamma coincidences were observed in the energy range up to 8.8 MeV. However, no such coincidences were detected above this energy range. This suggests that the excited states of ^{212}Po decay either by alpha-gamma cascade emissions or by direct alpha emissions only, as depicted in the energy diagram in Fig. 1. Therefore, the possibility of alpha-gamma coincidences being a problem was shown to be non-existent.

Secondly, when long-time measurements must be taken, some shifting caused by environmental (temperature variations) and electronical (heating, baseline instability, etc.) effects can cause spectral quality to deteriorate. A possible solution for this problem consists in fractioning measurements to avoid these spurious effects performing partial analysis. This was the method followed in this work, although measuring time produces low counting statistics.

The experimental study encountered a new problem caused by the short life of the ^{212}Po levels. Coincidence summing occurred between the emitted alpha particles and the beta particles originating from the ^{212}Bi ground level, creating interferences in the analysis of the energy region above 8.8 MeV where long-range particles are expected (Bland and Martín Sánchez, 1990; Martín Sánchez et al., 1990). While these interferences could not be fully eliminated, the use of a magnet between the source and the detector helped to reduce their impact.

The enlarged-spectrum part in Fig. 2 shows that emissions α_2 and α_3 are registered in a region far away the zone for beta-alpha coincidences, and they have no problems for analysis. However, α_1 must be detected in the zone with alpha-beta coincidences, presenting an important interference to study what events are really corresponding to the searched decays.

5. Results

Two series of measurements were taken. In the first set, 30 (22 one-day, 2 two-days, 4 three-days, and 2 four-days) spectra were measured. The second set included 57 (55 one-day, and 2 two-days) spectra. All spectra were energy calibrated considering the channels corresponding to the maxima of the emissions of the ^{232}U and daughters. A simple integration method adding the counts obtained between the channels corresponding to the emission α_0 (including the low-energy tail) was applied. The total counts obtained for each individual spectrum for α_0 emissions were added to obtain the global counting. In the first case, more than 8×10^5 events were registered for α_0, whereas about 10^6 events were registered in the second set.

Counts registered in each energy region corresponding to α_2 and α_3 were added for all the spectra. To surmount the difficulty presented by

Table 1
Relative intensities (I) for the long-range alpha particles produced in the decay $^{212}Po \rightarrow {}^{208}Pb$ [d] α from the natural Thorium series. The columns R/E mean if data are coming from Range (R, cm) or from Energy (E, MeV) measurements. In the column Type the meaning is: M, data coming from measurements; C, data from calculations; E, data found in compilations or evaluations appearing in nuclear data tables. The results reached in the present study are at the end, including the uncertainties.

α_0		α_1		α_2		α_3		R/E	Type	Reference
R/E	I	R/E	I	R/E	I	R/E	I			
8.6	10^6	10.2	~33	–	–	11.3	~66[a]	R	M	Rutherford and Wood (1916)
8.6	10^6	–	–	–	–	11.5	220	R	M	Bates and Rogers (1923, 1924)
8.6	10^6	9.5	70	–	–	11.5	200	R	M	Meitner and Freitag (1926)
8.6	–	9.9	–	–	–	11.7	—[b]	R	M	Nimmo and Feather (1929)
8.788	10^6	9.512	33.6	–	–	10.624	189	E	M	Rutherford et al. (1931)
8.770	–	9.4(3)	–	(9.6)	–	10.5(2)	–	E	M	Rosenblum and Valadares (1932)
8.778(8)	10^6	9.491(2)	34	–	–	10.541(8)	190	E	M	Lewis and Bowden (1934)
8.805[d]	10^6	9.522	35	10.454	20	10.574	170	E	M	Rytz (1951)
8.7801(4)	10^6	9.4923(4)	35	10.422	20	10.5432 (4)	180	E	M	(1954)
8.805[e]	10^6	9.522	35	10.454	20	10.574	180	E	M	Emery and Kane (1960)
8.810	10^6	9.522	35	10.452	20	10.573	180	E	C	Griffioen and Rasmussen (1961)
8.777	10^6	9.500	45	10.360	17	10.470	167	E	M	Bertolini et al. (1962)
8.785	10^6	9.499(4)	34	10.432(4)	10	10.550(4)	160	E	M	Leang (1965)
8.7848(3)	10^6	9.497(1)	37	10.426(5)	16	10.549(2)	172	E	E	Pancholi and Martin (1972)
8.78437(7)	10^6	9.503(4)	34	10.436(4)	10	10.554(2)	160	E	E	Rytz (1979)
8.78437(7)	10^6	9.495(3)	35	10.422(3)	20	10.543(3)	170	E	E	Browne et al. (1978)
8.78437(7)	10^6	9.4969(7)	37	10.427(4)	16	10.5487 (7)	172	E	E	Martín Sánchez et al. (1990)
8.78437(7)	10^6	9.4969(7)	40	10.427(4)	16	10.5487 (7)	170	E	E	Browne and Firestone (1986)
8.78486(12)	10^6	9.503(4)	34	10.436(4)	10	10.554(2)	160	E	E	Artna-Cohen (1992)
8.78517(11)	10^6	9.49878 (11)	38	10.43294 (11)	16	10.5521 (2)	166[c]	E	E	Nichols (2011)
8.78517 (11)[f]10^6	9.49878 (11)	36(14)	10.43294 (11)	21(4)	10.5521 (2)	156(11)	E	M	This work	

[a] Plus other particles with greater range.
[b] Greater-range particles discarded.
[c] Nichols (2011) gives as mean value 219(15) for the total quantity of long-range alpha particles from ^{212}Po.
[d] Global energies involved in the decay were given; the energies quoted here have been calculated taking into consideration recoil.
[e] Energy values assumed from Rytz (1951).
[f] Energy values assumed from LNHB (Nichols, 2011).

the interferences in the region of alpha-beta coincidences with the counts corresponding to the α_1 emission, data coming from each single spectra were fitted to exponential curves. When the counts corresponding to the channel for the α_1 emissions were above the fitted value, these counts were considered, discarding those cases in which this condition was not accomplished. Fig. 3 shows an example of this procedure. Normalizing to 10^6 events for α_0, the relative intensities for the long-range alpha-particles detected for each emission are given in Table 1.

An uncertainty budget was estimated considering the effects affecting the final values obtained in the measurements. Not many factors or variables can be considered because the searched results are relative quantities measured in the same conditions. Some effects were added to the (counting Poisson) statistics uncertainties. Studying the shifting observed in the measurements of individual spectra by comparison between them the case in which a greatest shift was observed could be estimated to introduce an uncertainty of about 3% in the regions for the α_2 and α_3 emissions. Fitting experimental results to an exponential curve in the region of α_1 is the part adding the greatest component for the uncertainty. All the individual spectra were fitted, and the greatest difference found between the values of the experimental results and the corresponding fitted value (calculated as a percentage) for the energy corresponding to the α_1 emission was considered as the worst value obtained for fitting effects and included in the uncertainty estimation (see Table 2). Although the efficiency for alpha-particle spectrometry with semiconductor detectors can be considered constant versus energy, and measurements of this effect were no made in our case, an uncertainty of about 1% was prudently assigned. Finally, the combined standard uncertainty was assigned to each relative intensity.

6. Summary and conclusions

Long-range alpha particles emitted by ^{212}Po, first discovered by Rutherford over a century ago, have been measured by several authors in the past. The most recent experimental measurements were conducted by Leang (1965), and no other results have been published since then (to the best of our knowledge).

In this study, alpha-particle spectrometry was used to measure long-range alpha particles, and our results for the relative intensities agree (with k = 2 confidence level) with previous published values. This is the first time that experimental values for the associated uncertainties of the relative intensities have been reported. We estimate the uncertainty budget based on several experimental effects for the relative intensities of the long-range alpha particles. ((Our results are expected to be included in the new tables on nuclear data.))

CRediT authorship contribution statement

A. Martín Sánchez: Writing – original draft, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **R. Mora Rodríguez:** Writing – review & editing, Validation, Supervision, Software, Investigation, Formal analysis.

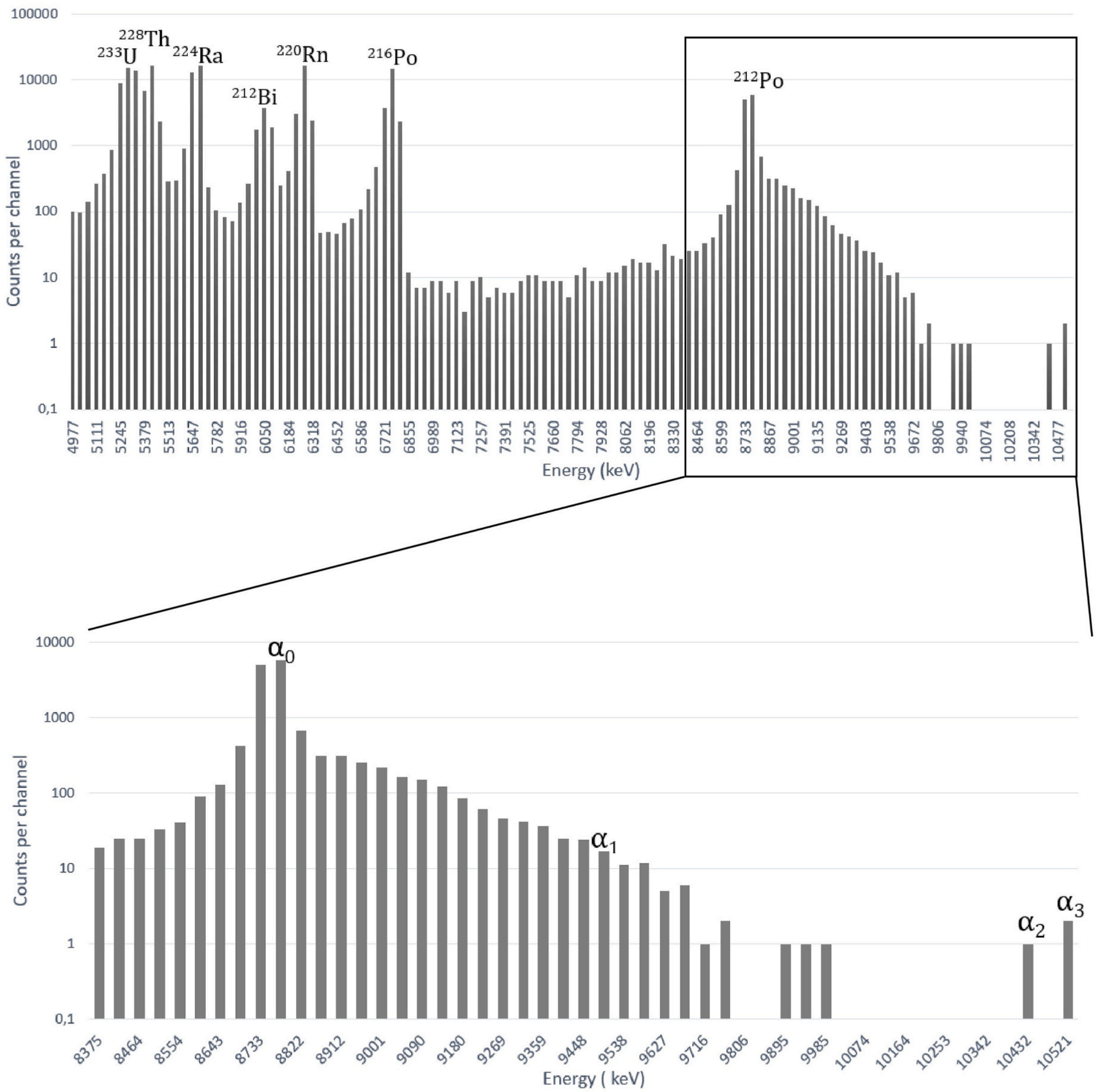

Fig. 2. Experimental alpha-particle spectrum of the ^{232}U source (with daughters) and enlarged part of the most energetic emissions showing the regions where the long-range alpha-particles were measured.

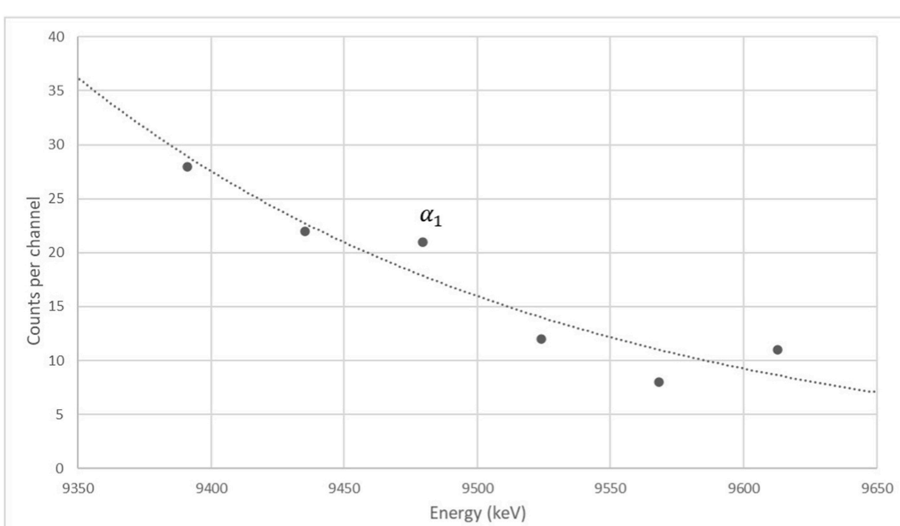

Fig. 3. An example of one of the measured spectra showing the region where the α_1 emissions are registered. The best fit of the experimental data to an exponential function is also shown in the plot.

Table 2
Uncertainty budget to estimate final values for the relative intensities of the long-range alpha particles from ^{212}Po.

Uncertainty component	Type	Relative standard uncertainty (%) (k = 1)		
		α_1	α_2	α_3
Counting (Poisson)	A	15	15	6
Shifting	A	–	3	3
Fitting	A	34	–	–
Efficiency	B	1	1	1
Combined standard uncertainty		37	16	7

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

The authors wish to thank to Junta de Extremadura (with a partial finance from the European Regional Development Fund, EDRF) for supporting this work through Grant GR21002.

References

Artna-Cohen, A., 1992. Nuclear data sheets for A=212. Nucl. Data Sheets 66, 171.
Bates, L.F., Rogers, J.S., 1923. Long range alpha-particles. Nature 112, 938.
Bates, L.F., Rogers, J.S., 1924. Particles of long range emitted by the active deposits of radium, thorium and actinium. Proc. Roy. Soc. A 105, 97–116.
Bertolini, G., Capellani, F., Restelli, G., Rota, A., 1962. Excited states of Tl-208 and Po-212. Nucl. Phys. 30, 599–612.
Bland, C.J., Martín Sánchez, A., 1990. A direct measurement of ^{228}Th activity by alpha-beta coincidence counting. Nucl. Instrum. Methods Phys. Res. 286, 375–378.
Briggs, G.H., 1954. The energies of natural alpha particles. Rev. Mod. Phys. 26, 1–6.
Browne, E., Dairiki, J.M., Doebler, R.B., Shihab-Eldin, A.A., Jardine, L.J., Tuli, J.K., Buyrn, A.B., 1978. In: Lederer, C.M., Shirley, V.S. (Eds.), Table of Isotopes, seventh ed. Wiley, New York.
Browne, E., Firestone, R.B., 1986. In: Shirley, V.S. (Ed.), Table of Radioactive Isotopes. Wiley, New York.
Browne, E., 2005. Nuclear data sheets for A = 212. Nucl. Data Sheets 104, 427–496.
Caro Marroyo, B., Martín Sánchez, A., Jurado Vargas, M., 2014. Improvements to alpha-particle spectrometry techniques. Appl. Radiat. Isot. 87, 328–330.
Emery, G.T., Kane, W.R., 1960. Gamma-ray intensities in the Thorium active deposit. Phys. Rev. 118, 755–762.
Evans, R.D., 1985. The Atomic Nucleus (Reprint Edition 1982of 14^{th} Printing 1972). Krieger, Florida.
Flack, F.C., Johnson, J.E., 1962. The gamma radiation from ^{212}Po (ThC'). Proc. Phys. Soc. 79, 10–13.
Griffioen, R.D., Rasmussen, J.O., 1961. Analysis of long-range alpha-emission data. Phys. Rev. 121, 1774–1778.
Leang, C.-F., 1965. Spectres α de long parcours des poloniums 214 (RaC') et 212 (ThC'). Compt. Rend. Acad. Sci. Paris 260, 3037–3040.
Lewis, W.B., Bowden, B.V., 1934. An analysis of the fine structure of the α-particle groups from Thorium C and of the long-range groups from Thorium C'. Proc. Roy. Soc. A 145, 235–249.
Martin, M.J., 2007. Nuclear data sheets for A = 208. Nucl. Data Sheets 108, 1583–1806.
Martín Sánchez, A., Vera Tomé, F., Bland, C.J., 1990. Recent measurements of ^{228Th} activity by alpha-beta coincidence counting. Nucl. Instrum. Methods Phys. Res. 295, 273–275.
Meitner, L., Freitag, K., 1926. Über die α-Strahlen des ThC+C' und ihr Verhalten beim Durchgang durch verschiedene Gase. Z. Phys. 37, 481–517.
Nichols, A.L., 2011. ^{212}Bi-Comments on Evaluation of Decay Data. LNE-LNHB/CEA Table de Radionuclèides.
Nimmo, R.R., Feather, N., 1929. An investigation of the ranges of the long range α-particles from Thorium C and Radium C, using an expansion chamber. Proc. Roy. Soc. A 122, 668–687.
Pancholi, S.C., Martin, M.J., 1972. Nuclear Data Sheets for A =212. Nucl. Data Sheets B8, pp. 165–191.
Rosenblum, S., Valadares, M., 1932. Sur la structure fine des rayons α du ThC. C. R. Acad. Sci. Paris 194, 967.
Rutherford, E., Wood, A.B., 1916. Long range alpha particles from Thorium. Phil. Mag. Ser. 6 31, 379–386.
Rutherford, E., 1921. The mass of the long-range particles from Thorium C. Phil. Mag. 41, 570–574.
Rutherford, E., Wynn-Williams, C.E., Lewis, W.B., 1931. Analysis of the α-particles emitted from thorium C and actinium C. Proc. Roy. Soc. A 133, 351–366.
Rytz, A., 1951. Nouvelles experiences sur le spectre magnetique alpha du Thorium C et des longs parcours du Thorium C'. Compt. Rend. 233, 790.
Rytz, A., 1953. J. Recherches Centre Natl. Recherche Sci., Labs. Bellevue (Paris) 25, 254.
Rytz, A., 1979. New catalogue of recommended alpha energy and intensity values. Atomic Data Nucl. Data Tables 23, 507–533.
Rytz, A., 1991. Recommended energy and intensity values of alpha particles from radioactive decay. Atomic Data Nucl. Data Tables 47, 205–239.
Wood, A.B., 1921. Long-range particles from thorium active deposit. Phil. Mag. 41, 575–584.