

A new evaluation of the nuclear decay data of ^{223}Ra

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Abstract. Since 2013, the radionuclide ^{223}Ra is used in nuclear medicine to prepare radiopharmaceuticals for targeted radiotherapy. ^{223}Ra is a member of the natural radioactive series of actinium and decays by alpha-particle emission, populating the excited levels of ^{219}Rn . According to the 2011 nuclear decay data evaluation within the Decay Data Evaluation Project (DDEP), by V.P. Chechev, the half-life of ^{223}Ra is (11.43 ± 0.03) days. The decay scheme is not considered as fully complete, because of the disagreement between the measured and calculated probabilities of some alpha-transitions and incomplete information on several gamma-ray transitions. New high quality measurements of ^{223}Ra nuclear decay data were performed and numerous results were published since 2012 and, consequently, an updated nuclear decay data evaluation was undertaken, according to the DDEP procedures. The main results obtained, focusing on the recommended data improvements, are presented and discussed in this paper.

1 Introduction

The radionuclide ^{223}Ra is an alpha-particle emitter with applications in nuclear medicine. Since 2013, it is used to prepare commercial radiopharmaceuticals ($^{223}\text{RaCl}_2$), used for targeted radiotherapy in bone metastases and bone palliation in prostate cancer of patients from European countries and the USA, [1]. ^{223}Ra is a member of the natural radioactive series of actinium (^{235}U) and decays by alpha-particle emission (100 %), populating the excited levels of ^{219}Rn . According to the evaluation of V.P. Chechev [2], the ^{223}Ra half-life is (11.43 ± 0.03) days (the uncertainty corresponds to a coverage factor $k=1$).

There are three ^{223}Ra production modes, as described in reference [3]: direct production by nuclear reactions, such as proton-induced reactions in natural thorium thin targets; by production and chemical separation of ^{227}Th (18.68 days half-life) – the ^{223}Ra parent, which can be used as a ^{223}Ra generator; by production of ^{227}Ac (21.772 years half-life) – the ^{227}Th parent, to be used as a ^{227}Th generator, [4].

The decay scheme of ^{223}Ra is complex: there are 30 levels of ^{219}Rn populated by the ^{223}Ra alpha decay (from the ground state up to 873 keV), 26 energy groups of alpha-particles emitted in the energy range (5014 - 5872) keV and more than 80 gamma-rays emitted in a wide energy range (4 - 737) keV. Some of the gamma-rays emitted following the ^{223}Ra decay are difficult to detect because of the low energy, very weak emission intensity and/or overlapping with other gamma-rays emitted by the ^{223}Ra decay progeny in equilibrium.

This work presents the main results obtained in the evaluation of ^{223}Ra nuclear decay data, using recently

published experimental data, and based on the DDEP procedures. The new recommended nuclear decay data improvements are underlined.

2 Nuclear decay data evaluation

2.1 The previous evaluations and the incomplete knowledge of the decay scheme

Two previous ^{223}Ra nuclear data evaluations were studied by the authors: within ENSDF [5] and Decay Data Evaluation Project (DDEP), [2], respectively. Also, the important comments about the decay scheme, from [6], were taken into account.

The main problem encountered in the ^{223}Ra nuclear data evaluations, as pointed out by Chechev, is the fact that the decay scheme is not considered as fully completed, because of the disagreement between measured and calculated probabilities of some alpha-transitions and the information about several gamma-ray transitions is incomplete. The calculated probabilities of the alpha-transitions were deduced from the gamma-ray emission measurements, internal conversion coefficients (ICC) computation and decay scheme levels imbalances. Using Chechev's evaluation to compute the total energy of all the emissions from the ^{223}Ra decay, the corresponding decay energy will be (6027 ± 133) keV, for a coverage factor $k=1$. This value has a deviation of about 0.8 % from the evaluated decay energy, $Q_\alpha = (5978.99 \pm 0.21)$ keV, according to the atomic mass evaluation [7].

Most of the experimental data used by Chechev (2011) were published long time ago (from 1954 to

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1998). No absolute measurements of the gamma-ray emission intensities were available until 2011. This lack of recent reliable data, with lower uncertainties, was pointed out in [8].

New high quality experimental data became available since 2015. New measurements were published for the ^{223}Ra half-life [9-11] and gamma-ray emission intensities (probabilities) – using absolute activity standardizations [6], [9], [12-13]. These new experimental results created the opportunity to perform an update of the previous DDEP evaluation (Chechev, 2011). The main results of this new evaluation, together with new experimental requirements for a better knowledge of the decay scheme, are presented and discussed below. Other measurements are underway and, after publishing, the evaluation will be updated and the decay scheme imbalance will be tested.

2.2 New recommended data: ^{223}Ra half-life

The experimental data compiled in order to obtain the new, improved, recommended value for the ^{223}Ra half-life are presented below, in Table 1. Only the last four results (new) were added to those from the previous DDEP evaluation [2]. The new recommended value is: (11.437 ± 0.003) days, $k=1$. According to the DDEP procedures, the value was computed as a weighted mean, by the computer code LWEIGHT4, based on the Limitation of Relative Statistical Weight method.

One must note that the new half-life value is very close to the value given in Chechev’s evaluation (2011), mentioned above. The major improvement refers to the uncertainty, which is now 10 times lower: 0.003 days, instead of 0.03 days. All the uncertainties mentioned in this paper correspond to a coverage factor $k=1$.

Table 1. Experimental data for the ^{223}Ra half-life ($T_{1/2}$).

Reference	$T_{1/2}$ (days)	Uncertainty of $T_{1/2}$ (days)
Hagee et al. [14]	11.685	0.056
Robert [15]	11.22	0.05
Kirby et al. [16]	11.4347	0.0044
Kirby et al. [16]	11.427	0.017
Jordan and Blanke [17]	11.372	0.045
Miller et al. [18]	11.444	0.046
Kossert et al. [9]	11.4362	0.0050
Bergeron and Fitzgerald [10]	11.447	0.006
Bergeron and Fitzgerald [10]	11.445	0.013
Collins et al. [11]	11.4358	0.0028

2.3 New recommended data: gamma-ray intensities following the decay of ^{223}Ra

Four new absolute activity measurements of ^{223}Ra were undertaken in 2015 and 2019, as mentioned above, and this allowed the determination of both the absolute and relative emission intensities of the gamma-rays following this decay, [6], [9], [12-13]. The results reported for the most important gamma-ray emission, of (269.463 ± 0.010) keV, are shown in Table 2.

Table 2. Absolute gamma-ray intensities measured for 269.463 keV photon emission following the decay of ^{223}Ra .

Authors	Value (per 100 decays)
Kossert et al. [9]	13.16 ± 0.15
Pibida et al. [12]	13.24 ± 0.12
Collins et al. [6]	13.37 ± 0.07
Marouli et al. [13]	14.1 ± 0.5

The last measurement result mentioned in Table 2 is slightly higher than the other three. The authors (Marouli et al., 2019) mention possible problems in the activity standardization, but recommend the use of the relative intensities of the gamma-rays in future decay data evaluations. So, this measurement result was not taken into account in the statistical analysis performed. The new recommended value of the absolute emission intensity for the gamma-rays of 269.463 keV is the weighted mean of the first three values from Table 2: (13.31 ± 0.07) per 100 decays.

Table 3 (on two columns) presents the relative emission intensities of the gamma-rays following the decay of ^{223}Ra (the intensity of the 269.463 keV gamma-rays is considered as equal to 100). The experimental data published in references [6], [9], [12-13] and all the references mentioned in the previous DDEP evaluation [2] were taken into account. In Table 3, the significant figures of the uncertainty were written in parenthesis: 106.78 (3) means 106.78 ± 0.03 .

Table 3. Relative emission intensities of the gamma-rays following the ^{223}Ra decay.

Energy (keV)	Relative intensity	Energy (keV)	Relative intensity
103.2 (2)	0.081 (20)	368.56 (12)	0.0998 (35)
103.9 (2)	0.113 (17)	371.68 (2)	3.62 (37)
106.78 (3)	0.162 (8)	372.86 (1)	0.63 (22)
110.86 (1)	0.388 (8)	376.26 (2)	0.0428 (32)
122.32 (1)	9.46 (35)	383.35 (2) *	0.019 (7)

144.27 (2)	24.7 (14)	387.7 (2)	0.105 (40)
154.21 (1)	42.3 (27)	390.1 (2)	0.039 (5)
158.64 (1)	5.42 (18)	430.6 (3) *	0.1792 (43)
175.65 (15)	0.1181 (10)	432.45 (3)	0.231 (11)
177.3 (1)	0.321 (8)	445.03 (1)	9.17 (7)
179.54 (6)	1.200 (10)	487.5 (2)	0.0574 (47)
221.32 (24)	0.225 (8)	500.0 (4)	0.0098 (37)
249.49 (3)	0.280 (7)	523.2 (4)	0.0125 (40)
251.6 (3)	0.474 (11)	527.61 (1)	0.493 (7)
255.2 (2)	0.369 (10)	532.9 (4)	0.013 (5)
269.46 (1) *	100.0 (5)	537.6 (1)	0.0218 (36)
288.18 (3)	1.107 (21)	541.99 (2)	0.0118 (36)
323.87 (1)	27.42 (20)	545.8 (5)	0.014 (6)
328.38 (3) *	1.479 (35)	574.1 (7)	0.010 (5)
334.01 (6)	0.593 (28)	598.72 (2)	0.648 (10)
338.28 (1)	19.60 (14)	609.31 (4) *	0.31 (8)
342.78 (2) *	1.434 (39)	619.1 (2)	0.0388 (46)
355.5 (2)	0.065 (9)	623.68 (4)	0.057 (6)
361.89 (2)	0.246 (47)	711.3 (2)	0.0254 (22)
362.9 (2)	0.140 (7)	728.4 (8)	0.010 (8)

The gamma-rays energies marked with * in the Table 3, can be overlapped with other gamma-rays of close energy, emitted by the progeny of ^{223}Ra . All the recent measurements results ([6], [9], [12-13]) were obtained after the deconvolution of these overlapped peaks identified in the recorded gamma-ray spectra. The deconvolution of the peaks introduce a supplementary component of uncertainty, depending on the experimental data fitting in the gamma-ray spectra. However, in this case, also, the combined uncertainties are significantly lower than in the previous evaluation [2]. The photons of 619.1 keV were observed only by Kossert et al. (2015) and the placement of this gamma-ray in the decay scheme is still uncertain.

3 Conclusions

A new ^{223}Ra decay data evaluation was undertaken. Based on new high quality measurement of the ^{223}Ra half-life and gamma-ray emission intensities, the new recommended data sets have significantly lower uncertainties than the data from the previous evaluations. Despite the reported improvements, new measurements are necessary (especially for the alpha-particle emission probabilities), for a complete and reliable characterization of the ^{223}Ra decay scheme.

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