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Original Research

Critical concentration of the exotic nuclei in the ^{232}Th chain for the alpha analysts

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ABSTRACT

The ^{232}Th (Thorium) isotope and its α-decay daughters are ubiquitous in soil, rock, and water environments that are harmful to living organisms if accidentally up-taking. Therefore, it is necessary to detect these isotopes and measure their concentrations in the environments in order to protect living organisms from the contaminated areas. In the measurements using alpha analyzers, the estimation of the critical concentration of the isotopes in the samples is highly demanded to confirm the feasibility of the analysis. In this work, we evaluated the critical concentrations of the radioactive nuclei in the ^{232}Th chain based on their half-lives and radioactivities for the alpha analysis. In particular, we examined the α-decay half-lives of the mentioned isotopes by using the semi-empirical formulae proposed by Viola-Seaborg, Royer, and Poenaru to estimate their radioactivities and decay constants. The predicted half-lives were then normalized by their average values and compared with the NuDat data of the National Nuclear Data Center (Brookhaven National Laboratory). The relationships between the estimated half-lives and the NuDat data are deduced as linear functions. The critical concentration of the isotopes is determined based on the decay constants following the decay rule. The results show that most of decay half-lives are in consistent with those obtained from the NuDat data except for the multi-decay-mode isotopes. However, there is a large variation in the average values, 15% - 95% (in decimal logarithmic scale), of the half-lives evaluated by different formulae, that consequently leads to large decay constants deviations of 1% — 120% from the average values. From these results, we confirmed that the large radioactivity uncertainty because of the half-life deviations should be concerned for the preparation of the environmental samples when using alpha spectrometers. By assuming an efficiency of 100%, we found that the critical concentration for the alpha analysts of the ^{232}Th nucleus is in a range of $1.5 - 2.5$ μ g/(l or kg) in the samples.

Key words: alpha-decay, half-life, radiation safety, radioactivity, Thorium

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INTRODUCTION

The radiation pollution, which is caused by exotic nuclei including α-decay of heavy isotopes, has emerged as one of the most environmental interests in recent decades. A group of scientists found water sources from deep wells and hot springs in Northern Greece polluted by Uranium with concentration of 0.15 - 7.66 μg/l. The radioactivity concentration of uranium in this area is about 1.7 - 160.1 mBq/l[1]. In a report of the United Nations Scientifi Committee on the effects of atomic radiation, the average uranium weight of 0.3 - 11.7 mg/kg was found in soil[2]. In another area of Morocco, the average radioactivities of Uranium and Thorium in water along Oum Er-Rbia River was 12 - 37 Bq.m^{-3} and 2 - 10 Bq.m^{-3} [3] and their concentrations in some Egyptian rocks were 4.0 - 35.0 ppm and 11.0 - 124 ppm[4], respectively.

Thorium assimilates well into different types of environments such as soil, water, and air. It is able to integrate into different types of soil or rocks with high concentration: 1 - 4 mg per/kg of and esite, less than 3.5 mg/kg of gabbro, and less than 1.0 mg/kg of basalt[5]. It is also found highly immigrated in granitic rocks with a typical rate of 10 - 40 mg/kg. It is reported that the Thorium assimilates into soil more than water with 10 - 13 mg/kg in soils and higher than 0.1 g/liter in water[5]. The Thorium was also detected in the air in north Norway with a concentration of 8.15 mBq/g[6]. As the thorium decays, a series of exotic daughters (*e.g.*, radium, radon, polonium, *etc.*) is produced. These exotic nuclei assimilated in the soil, water, or air can easily access into the human body via contaminated food such as meat, vegetables, cereals, etc or even air. Consuming these contaminated materials may cause diseases or biological defections in living organisms, especially the human body. According to the study of A. Kaul *et al.*[7], the ^{232}Th isotope can be absorbed by the human body with a typical tissue distribution as follows: liver, 59%; spleen, 29%; red

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bone marrow, 9%; calcifi d bone, 2%; lungs, 0.7%; kidneys, 0.1%. For example, about 15% of German exposed to a radioactive dose of 25 rad/year of ^{232}Th exhibited liver cancer symptoms[8]. Therefore, investigation of radioactivities of the exotic nuclei in the suspected areas should be conducted to preserve the environment and to protect the living organisms from harmful radiation. In the present work, we illustrated a method to estimate the uncertainty of the radioactivity and critical concentration of the nuclides in the ^{232}Th α-decay chain, as shown in the following **Figure 1**.

There are few techniques for analyzing nuclear isotopes based on gamma or alpha radiation. For the α-decay nuclei such as ^{232}Th the alpha spectrometers are commonly applied to investigate the concentration of the isotopes. It is necessary to estimate the concentration of the interest isotopes in the environmental samples together with the limit of the alpha analysis. It should be noted that the typical radioactivity detection limit of the alpha spectrometers is about 0.01 Bq/liter (or kilogram) of the samples of water (or soil)[9]. In such a case, the uncertainty of the radioactivity is one of the important parameters impacting on the precision of the analysis because the concentration is proportional to the decay rates and the detection effici cy. Since the radioactivity strongly depends on the half-life of the nuclei, studies of the half-life for the concentration are highly demanded. In this work, we estimate the half-life to deduce the critical concentrations of the isotopes in the samples corresponding to the limit of the analysis. The α-decay half-lives are re-examined via few methods such as the semi-empirical formulae of the Viola-Seaborg relationship[10], the Poenaru method[11–13], and the Royer approach[14,15] which have been improved to increase the accuracy of the half-life prediction. The uncertainties of the half-lives, decay constants and the radioactivity of the exotic nuclei in the ^{232}Th decay chain are also investigated.

THEORETICAL FRAMEWORK

The calculation methods for alpha half-life ($T_{1/2}$) have been improved by a few semi-empirical approaches. It can be evaluated by using Viola-Seaborg formula[10]:

$$T = \log_{10}\left(T_{1/2}\right) = (aZ+b)\frac{1}{\sqrt{Q_\alpha}} + cZ + d + f \quad (1)$$

with initial parameters $a = 1.6618; b = -8.5166; c = -0.2022; d = -33.9069$ and $f = 0$(even − even); $f = 0.772$(odd-even); $f = 1.066$(even-odd); $f = 1.114$(odd-odd).

In this method, the half-life depends only on the number of the proton (Z) and the α-decay Q-value (Q) of the nuclei. Akarawy model[12] was developed by G. Royer[14,15] with additional parameters so that the quantity is as a function of Z, Q and the mass number, A, as

$$T = \log_{10}\left(T_{1/2}\right) = a + bA^{1/6}\sqrt{Z} + \frac{cZ}{\sqrt{Q_\alpha}} \quad (2)$$

where $a = -27.657, -28.408, -27.408$ and -24.763; $b = -0.966, -0.920, -1.038$ and -0.907; $c = 1.522, 1.519, 1.581$ and 1.410 for *even-even*, *even-odd*, *odd-even* and *odd-odd* isotopes, respectively.

The models mentioned above strongly depend on the even-odd property of the number of proton Z and neutron N. Since the heavy nuclei often have a strong deformation which is dependent on the asymmetry of the nuclei, D. N. Poenaru[11–13] proposed a new model based on the Royer formula by adding two factors in terms of the asymmetric factor, $I = (N-Z)/A$. The new formula reads

$$T = \log_{10}\left(T_{1/2}\right) = a + bA^{1/6}\sqrt{Z} + \frac{cZ}{\sqrt{Q_\alpha}} + dI + fI^2, \quad (3)$$

in which the fitting parameters based on experimental data could be deduced as

$$a = -27.989, b = -0.940, c = 1.532, d = -5.747 \text{ and } f = 11.336$$

Notice here that Poenaru model in **Equation (3)** is dependent on the asymmetry of the nuclei and the even-odd property has been removed.

In the quantitative analysis, the radioactivities (R) of the exotic isotopes are measured by using alpha analyst to determine the element amount in a sample. The amount of the concerned isotopes can be deduced based on the classical relationship between the quantity of the nucleus, N, and the decay constant, λ, as

$$R = \lambda N = \frac{\ln 2}{T_{1/2}} N. \quad (4)$$

According to **Equation (4)**the radioactivity uncertainty is directly proportional to the uncertainty of the decay constant or the half-life of the nucleus. Since the number of a nucleus is a constant in a sample, the radioactivity uncertainty is decided by the decay-constant deviation. In order to examine the critical amount of isotopes, $N_{cr.}$, required for the detection

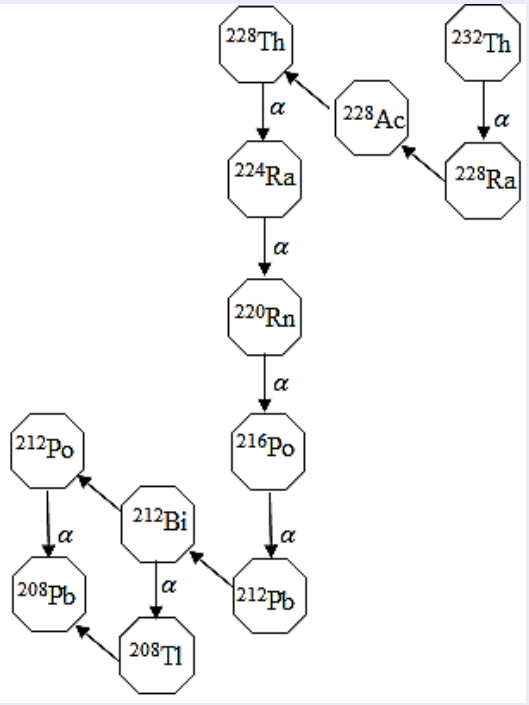

Figure 1: **The α-decay chain of the ^{232}Th isotope.**

threshold of 0.01 Bq/(l or kg) of the alpha spectrometers, the maximum decay constants, λ_{max}, are applied to the formula as

$$N_{cr.} = \frac{0.01}{\lambda_{\max}}. \quad (5)$$

Th s critical value is useful for estimating the weight of the environmental samples in the preparation of the alpha analysis.

RESULTS

As mentioned, the nuclei in the α-decay chain derived by ^{232}Th are considered since they may exist in environments of water or soil. The half-lives of these isotopes are calculated with various models described in **Equations (1), (2) and (3)**to determine the minimum and maximum deviations from the average values. The lower and upper uncertainties of the radioactivity can be estimated based on the half-life deviation. **Table 1** shows the estimated half-lives (in second) of the isotopes in the ^{232}Th chain. The decay constants and their deviations are presented in **Table 2**. The deviations, $\delta\lambda_i (i = 1(Viola), 2(Royer), 3(Poenaru)$ and $NuDat)$,of the estimated values by using the methods in Equations (1), (2) and (3) and the data in Ref.[16] (NuDat Database) are normalized by the average calculated values, λ_{Av}, as

$$\delta\lambda_i = \frac{|\lambda_i - \lambda_{Av}|}{\lambda_{A_y}} \times 100(\%). \quad (6)$$

In order to examine the variation range of the decay constants, we consider the minimum and maximum deviations of the calculated values determined by the Viola-Seaborg, Royer, and Poenaru models which are defi ed as

$$\begin{cases} \delta_{Min} = \text{Min}\,(\delta\lambda_1, \delta\lambda_2, \delta\lambda_3) \\ \delta_{\text{Max=Max}(\delta\lambda_1, \delta\lambda_2, \delta\lambda_3)} \end{cases}. \quad (7)$$

Since there is only one data point, the minimum and maximum deviations of the NuDat data should be the same. Hence, we notice that $\delta_{\text{NuDat}} = \delta\lambda_{\text{NuDat}}$.

Since the radioactivity is directly proportional to the decay constants, its uncertainty can be estimated by the deviations of the constants, δ. By using the quantities of δ_{Min}, δ_{Max}, and δ_{NuDat} estimated in Table 2, the radioactivity uncertainty of the daughters in the α-decay chain could be predicted to be varied in the range of $\delta = 1\% - 120\%$ depending on the type of the decay mode.

In order to improve the validation in the use of the formulae, the calculated half-lives are compared with

Table 1: The estimated half-life (in logarithmic scale) of various α-decay nuclei in the ^{232}Th chain

Element	Z	A	Q (MeV)	T-Viola	T-Royer	T-Poenaru	T-Average	T-[NuDat]
Th	90	232	4.082	17.70	17.43	17.43	18.02	17.64
Th	90	228	5.520	7.92	7.95	7.99	8.43	7.78
Ra	88	224	5.789	5.53	5.60	5.68	6.09	5.50
Rn	86	220	6.405	1.80	1.94	2.05	2.42	1.75
Po	84	216	6.906	-1.02	-0.84	-0.70	-0.36	-0.84
Po	84	212	8.954	-7.10	-6.73	-6.55	-6.26	-6.52
Bi*	83	212	6.207	2.36	1.42	2.03	2.56	3.56

(*) The multi-decay mode nuclei.

Table 2: The estimated decay constant of various α-decay nuclei in the ^{232}Th chain. The values of δ_{Min} δ_{Max} and δ_{NuDat} (in %) are respectively the minimum, maximum, and NuDat decay-constant deviations normalized by the average values δ_{Min}

Element	Z	A	λ_{Viola} (s^{-1})	λ_{Royer} (s^{-1})	$\lambda_{Poenaru}$ (s^{-1})	λ_{Av} (s^{-1})	(%)	(%)	(%)
Th	90	232	1.383E-18	2.586E-18	2.549E-18	2.173E-18	17.3	36.3	27.7
Th	90	228	8.370E-09	7.032E-09	7.831E-09	7.744E-09	1.1	9.2	48.4
Ra	88	224	2.040E-06	1.454E-06	1.733E-06	1.742E-06	0.5	17.1	26.8
Rn	86	220	1.096E-02	6.136E-03	7.978E-03	8.356E-03	4.5	31.1	49.2
Po	84	216	7.324E+00	3.437E+00	4.810E+00	5.190E+00	7.3	41.1	7.9
Po	84	212	8.660E+06	2.467E+06	3.682E+06	4.936E+06	25.4	75.4	53.0
Bi*	83	212	3.031E-03	6.435E-03	2.616E-02	1.187E-02	45.8	120.3	98.4

(*) The multi-decay mode nuclei.

the NuDat data. **Figure 2** shows the correlation between Viola-Seaborg, Royer, Poenaru, and average estimated half-lives and the data in Ref. [16]. The relationships of the values are described as linear functions with the fitting coeffici ts listed in **Table 3**.

For the analysis using the alpha spectrometers, the required amounts of the isotopes in the samples are investigated by using Equation (5) and considering the molar mass of the isotopes. The necessary amounts of each isotope in the α-decay chain of the ^{232}Th nucleus in a liter (or a kilogram) of water (or soil) are listed in **Table 4**.

DISCUSSION

The results indicate that the half-lives estimated by the Viola-Seaborg and Royer models are almost similar to each other but larger than the ones determined by the Poenaru approach and close to the data obtained in Ref. [16]. The estimation by using various models give a wide dispersion range of 15% — 95% of the average values (in the decimal logarithmic scale). Notice that the odd-even property of the alpha emitters is taken into account in the fi st two models whilst the nuclear asymmetry is concerned in the Poenaru one. Th s may cause a difference in the decay half-lives calculated by the three models. The half-lives estimated by the models are more different from the data obtained from the Ref. [16] for the case of the ^{212}Bi isotope. Th s phenomenon should be paid attention in the investigation of the radioactivity for the element analysis of the environmental samples when using the alpha analysts. We realized that the formulae in **Equations (1), (2) and (3)**give less half-life uncertainty (less than 30%) for the ^{232}Th ^{228}Th ^{224}Ra, and ^{220}Rn isotopes. In addition, these semi-empirical models are more reliable if used for the exotic isotopes which have only the α-decay mode such as ^{228}Th and ^{224}Ra. In other words, these equations still need more modifi ations to apply well for the multi-decay mode (*e.g.* combination of the alpha-, beta-decay, and spontaneous fission) nuclei.

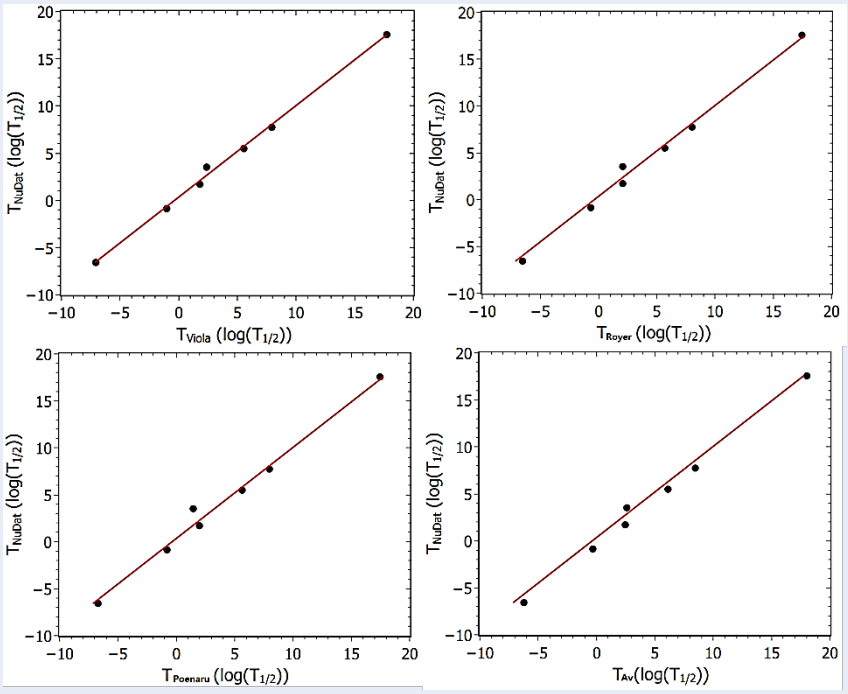

Figure 2: **The functions describing the relationship of the NuDat data and the half-lives estimated by the models are determined by the chi-square fitting based on the data in Table 1.**

Table 3: The fitting parameters of the linear functions in forms of $y = ax + b$ describing the relationship between the NuDat data and the estimated values of the half-lives

Parameters	Viola-Seaborg	Royer	Poenaru	Average
a	0.96967 ± 0.02495	0.99732 ± 0.03786	0.98399 ± 0.04807	0.98646 ± 0.03404
b	0.35235 ± 0.20372	0.18145 ± 0.30413	0.35439 ± 0.38554	-0.23541 ± 0.28321

Table 4: The critical amounts, m_{cr}., of the isotopes in the ^{232}Th chain for the analysis using the alpha spectrometers with 100% of the detection efficiency

Element	Z	A	m_{cr} (μg/(l or kg))	m_{NuDat} (μg/(l or kg))
Th	90	232	1.5E+00	2.5E+00
Th	90	228	4.5E-10	3.3E-10
Ra	88	224	1.8E-12	1.7E-12
Rn	86	220	3.3E-16	2.9E-16
Po	84	216	4.9E-19	7.5E-19
Po	84	212	4.1E-25	1.5E-24
Bi*	83	212	1.4E-16	1.8E-14

The comparison of the estimated results based on the semi-empirical formulae with the NuDat Data shows that the Royer model is more reliable than the others for the calculation of the α-decay half-lives of the nuclei in the ^{232}Th chain. Th s result is almost consistent with those from the NuDat database.

As can be seen in **Table 4**, the required concentration of the daughters in the environmental samples is much less than the one of the mother nucleus. By assuming 100% of the detection effici cy for the alpha analysis systems, the critical concentration of the ^{232}Th isotopes necessary for the alpha analysts is in the range of 1.5 – 2.5 μmg/(l or kg) in the environmental samples. The results also indicate that the alpha analyzers can be well applied to quantitative analysis of the daughters in the ^{232}Th chain without regarding their detection limit of 0.1 Bq/(l or kg).

CONCLUSION

The critical concentration of the isotopes in the ^{232}Th decay chain needed for the alpha analysts was estimated based on the half-lives. The half-lives were re-examined by the semi-empirical models for the estimation of the radioactivity uncertainty. By comparing with the database NuDat, the results calculated by the two fi st formulae, in which the odd-even property is concerned, are more reliable than the ones obtained by the model in terms of the nuclear asymmetry. We also found that the formulae are well applied to the mono–decay mode. The radioactivity uncertainty estimated based on the half-lives calculated by the mentioned models has a deviation range of 1% - 120%. The critical concentration required for alpha analysts of the mother isotope, ^{232}Th should be considered whilst it is not necessary to be concerned for the daughters.

ABBREVIATIONS

α: alpha particle, 4He

Bq: Becquerel (one of the units of the radioactivity), Bq = decay/s

s: second

T1/2: half-life

NuDat: National Nuclear Data Center (Brookhaven National Laboratory)

COMPETING INTERESTS

The authors declare that there is no confli t of interest regrading the publication of this paper.

AUTHORS' CONTRIBUTIONS

All the authors contribute equally to the paper including the research idea, data analysis, and writing manuscript.

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REFERENCES

1. Samaropoulos I, Efstathiou M, Pashalidis I, Ioannidou A. Determination of uranium concentration in ground water samples of Northern Greece. EPJ Web of Conferences. 2012;24:03005.
2. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation: UNSCEAR 1993 Report to the General Assembly, with Scientific Annexes; 1993.
3. Amrane M, Oufni L. Determination for levels of uranium and thorium in water along Oum Er-Rabia river using alpha track detectors. Journal of Radiation Research and Applied Sciences. 2017;10:246–251.
4. Hussein A. Determination of uranium and thorium concentration in some Egyptian rock samples. Journal of Radioanalytical and Nuclear Chemistry. 1994;188:255–265.
5. ;. Available from: http://weppi.gtk.fi/publ/foregsatlas/text/.
6. Kolb W. Thorium, Uranium and Plutonium in Surface Air at Vardo,. Journal of Environmental Radioactivity. 1995;31(1):1–6.
7. Kaul A. Thorotrast Kinetics and Radiation Dose Results from Studies in Thorotrast Patients and from Animal Experiments. Radiation and Environmental Biophysics. 1978;15:241–259.
8. National Research Council . Health Risks of Radon and Other Internally Deposited Alpha-Emitters: BEIR IV. Washington, DC. The National Academies Press; 1988. Available from: https://doi.org/10.17226/1026.
9. Pommé S. Typical uncertainties in alpha-particle spectrometry. Metrologia. 2015;52:S146–S155.
10. Viola VE, Seaborg GT. Nuclear systematics of the heavy elementsII Lifetimes for alpha, beta and spontaneous fission decay. Journal of Inorganic and Nuclear Chemistry. 1966;28(3):741–761.
11. Poenaru DN. Estimation of the alpha decay half-lives. Journal of Physique. 1983;44:791–796.
12. Akrawy DT, Poenaru DN. Alpha decay calculations with a new formula. Journal of Physics G: Nuclear and Particle Physics. 2017;44:10.
13. Poenaru DN, Gherghescu RA. -decay and cluster radioactivity of nuclei of interest to the synthesis of Z = 119, 120 isotopes. Physical Review C. 2018;97:044621.
14. Royer G. Alpha emission and spontaneous fission through quasi-molecular shapes. Journal of Physics G: Nuclear and Particle Physics. 2000;26:1149.
15. Royer G. Analytic expressions for alpha-decay half-lives and potential barriers. Nuclear Physics A. 2010;848:279.
16. National Nuclear Data Center. Brookhaven National Laboratory;. Available from: https://www.nndc.bnl.gov/NuDat2/.