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 $\alpha$-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 $\alpha$-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 considered 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 analyzers of the $^{232}$Th nucleus is in a range of $1.5 - 2.5 \text{ g/(l or kg)}$ in the samples.

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

**INTRODUCTION**

The radiation pollution, which is caused by exotic nuclei including $\alpha$-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 $\mu$g/l. The radioactivity concentration of uranium in this area is about 1.7 - 160.1 Bq/l$^3$. In a report of the United Nations Scientific 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$^{-3}$ 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 defictions 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
bone marrow, 9%; calcified 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. 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 activity and critical concentration of the nuclides in the $^{232}$Th $\alpha$-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 $\alpha$-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). 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 efficiency. 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 $\alpha$-decay half-lives are re-examined via few methods such as the semi-empirical formulae of the Viola-Seaborg relationship, the Poenaru method, and the Royer approach 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:

$$T = \log_{10}(T_{1/2}) = (aZ + b)\frac{1}{\sqrt{Qa}}$$

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 $\alpha$-decay Q-value ($Q$) of the nuclei. Akarawy model was developed by G. Royer with additional parameters so that the quantity is as a function of $Z$, $Q$ and the mass number, $A$, as

$$T = \log_{10}(T_{1/2}) = a + 0.6\sqrt{Z} + \frac{cZ}{\sqrt{Qa}}$$

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 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

$$T = \log_{10}(T_{1/2}) = a + b\frac{1}{\sqrt{Qa}} + dI + fI^2,$$

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

$$a = -27.989; b = -0.940; c = 1.532; d = -5.747; f = 11.336. $$

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 analytist 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, $\lambda$, as

$$R = \lambda N = \frac{\ln 2}{T_{1/2}}.$$
threshold of 0.01 Bq/(l or kg) of the alpha spectrometers, the maximum decay constants, $\lambda_{\text{max}}$, are applied to the formula as

$$
N_{\text{cr}} = \frac{0.01}{\lambda_{\text{max}}}.
$$

This 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 $\alpha$-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, $\delta \lambda_i (i = 1(\text{Viola}), 2(\text{Royer}), 3(\text{Poenaru}) \text{ and } \text{NuDat})$, 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, $\lambda_{\text{Av}}$, as

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

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 defined as

$$
\begin{align*}
\delta \lambda_{\text{Min}} &= \text{Min}(\delta \lambda_1, \delta \lambda_2, \delta \lambda_3) \\
\delta \lambda_{\text{Max}} &= \text{Max}(\delta \lambda_1, \delta \lambda_2, \delta \lambda_3) \\
\delta \lambda_{\text{NuDat}} &= \delta \lambda_{\text{NuDat}}.
\end{align*}
$$

Since there is only one data point, the minimum and maximum deviations of the NuDat data should be the same. Hence, we notice that $\delta \lambda_{\text{NuDat}} = \delta \lambda_{\text{NuDat}}$. Since the radioactivity is directly proportional to the decay constants, its uncertainty can be estimated by the deviations of the constants, $\delta$. By using the quantities of $\delta \lambda_{\text{Min}}, \delta \lambda_{\text{Max}}$, and $\delta \lambda_{\text{NuDat}}$ estimated in Table 2, the radioactivity uncertainty of the daughters in the $\alpha$-decay chain could be predicted to be varied in the range of $\delta = 1\% - 120\%$ depending on the type of the decay mode.

![Figure 1: The $\alpha$-decay chain of the $^{232}$Th isotope.](image)
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  | 5.33    | 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 $\delta_{\text{Min}}$, $\delta_{\text{Max}}$, and $\delta_{\text{NuDat}}$ (in %) are respectively the minimum, maximum, and NuDat decay-constant deviations normalized by the average values.

| Element | Z | A | $\lambda_{\text{Viola}}$ (s$^{-1}$) | $\lambda_{\text{Royer}}$ (s$^{-1}$) | $\lambda_{\text{Poenaru}}$ (s$^{-1}$) | $\lambda_{\text{Av}}$ (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.

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 gives 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 first two models whilst the nuclear asymmetry is concerned in the Poenaru one. Ths 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. Ths phenomenon should be paid attention in the investigation of the radioactivity for the element analysis of the environmental samples when using the alpha analyzers. 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 modifications 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, $m_{crit}$, of the isotopes in the $^{232}$Th chain for the analysis using the alpha spectrometers with 100% of the detection efficiency

| Element | Z | A  | $m_{crit}$ (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. This 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 efficiency 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 μg/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 first 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 analysis of the mother isotope, $^{232}$Th should be considered whilst it is not necessary to be concerned for the daughters.

**ABBREVIATIONS**

$\alpha$: 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 conflict 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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