Heavy-particle radioactivity

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Abstract. The competition of heavy particle radioactivity (HPR) and α decay is investigated in the region of superheavy (SH) nuclei with atomic numbers Z = 104–124. Calculations of half-lives within analytical supersymmetrical fission (ASAF) model are performed by using different theoretical mass tables to determine Qc, the energy released. For α decay the calculations are made using semi-empirical Fission (semFIS) model and AME11 mass table. A trend toward shorter half-lives and larger branching ratios relative to alpha decay for heavier SHs is observed.

1. Introduction
Superheavy (SH) elements with atomic numbers Z = 104–118 (see the review papers [1, 2] and the presentations at this Conference by Ch. E. Düllmann [3] and N. Schunck [4]) have been synthesized with cold fusion reactions [5, 6] or with hot fusion induced by ⁴⁸Ca projectiles [7, 8]. The names and symbols for Z = 104–112, 114, 116 are: Rf, Db, Sg, Bh, Hs, Mt, Ds, Rs, Cu, Fl, and Lv, respectively. The elements 113, 115, 117, and 118 are still waiting to be named. Many of them are identified through α decay chains. Previously we also discussed the competition of α decay and heavy particle radioactivity (HPR) [9], which may be important [10, 11] in the region of the heaviest SHs. In this process, from one parent nucleus ZA, one obtains an emitted particle ZAe, and a daughter ZAd:

\[ A Z \rightarrow A_e Z_e + A_d Z_d \]  (1)

Alternative theories are available [12, 13, 14, 15, 16]. A universal decay law based on R-matrix theory was recently introduced for α emission and HPR [17].

Starting with 1984 [18] many HPR have been experimentally confirmed [19, 20] in heavy parent nuclei with Z = 87 to 96: ¹⁴C, ²⁰O, ²³F, ²²²⁴–²⁶Ne, ²⁸³⁰Mg, and ³²³⁴Si. The measured half-lives are in good agreement with predicted values within the ASAF model (see the review [21] and references therein). The largest branching ratio relative to α decay, bα = Tα/Tc, of 10⁻⁸⁻⁹ was observed for ¹⁴C radioactivity of ²²³Ra. Usually in this region of the nuclear chart HPR is a rare process in a huge background of α particles.

The measurable quantities are: the kinetic energy of the emitted cluster Ek = QcA_d/A or the released energy Qc and the half-life Tc or the branching ratio bα = Tα/Tc. With the existing techniques it is possible to measure half-lives shorter than the limit Tc < 10⁻³² s and branching ratios large enough bα > 10⁻¹⁷. In the present work we give new details concerning the use of different mass tables to estimate the competition of α decay and HPR.
2. Calculations
A strong shell effect of the doubly magic daughter $^{208\text{Pb}}_{82}$ was observed. In order to study its importance in the region of SHs with $Z > 110$ we changed the concept of HPR, previously [22] associated with a maximum $Z_{e}^{max|\text{old}} = 28$. Now we allow

$$Z_{e}^{max} = Z - 82$$  \hfill (2)

The accuracy of half-life calculation is essentially dependent on the precision with which we know the $Q$-value

$$Q = [M - (M_e + M_d)]c^2$$  \hfill (3)

obtained as a difference between the parent, $M$, and the two decay product masses, $M_e$ and $M_d$, in units of energy; $c$ is the light velocity. $Q$-value should be positive in order to allow a spontaneous decay.

The decay constant

$$\lambda = \ln 2/T_c = \nu SP_s$$  \hfill (4)

is expressed by a product of three model dependent quantities $\nu$, $S$ and $P_s$ where $\nu$ is the frequency of assaults on the barrier per second, $S$ is the preformation probability and $P_s$ is penetrability of external barrier. According to our method [23] the preformation in a fission theory is given by the penetrability of the internal part (overlapping stage of the two fragments) of the barrier.

We developed our ASAF model starting with the Myers-Swiatecki liquid drop model [25] adjusted with a phenomenological correction. The half-life is given by

$$T = [(h \ln 2)/(2E_v)]exp(K_{ov} + K_s)$$  \hfill (5)

It is calculated by using the WKB quasiclassical approximation

$$K = \frac{2}{\hbar} \int_{R_a}^{R_b} \sqrt{2B(R)E(R)}dR$$  \hfill (6)

with $B = \mu$ (the reduced mass), $K = K_{ov} + K_s$, and $E(R)$ replaced by $[E(R) - E_{corr}] - Q$ where $E_{corr}$ is a correction energy similar to the Strutinsky shell correction. The turning points of

![Figure 1. Chart of nuclides for which calculated masses are available according to the MySw94 mass tables. 8824 masses for $Z = 1 - 136, N \leq 236$. The Green approximation of the beta stability is marked by full squares.](image)

the WKB integral are: $R_a = R_t + (R_t - R_i)[(E_v + E^*)/E_b^{0.5}]$ and $R_b = R_tE_c \{1/2 + [1/4 + (Q + E_v + E^*)E_b/E_c^{2.5}]\}/(Q + E_v + E^*)$ where $E^*$ is the excitation energy concentrated in the
Figure 2. Chart of nuclides for which calculated masses are available according to the APDT95 mass tables. 7508 masses for $Z = 10 - 130, N \leq 206$. The Green approximation of the beta stability is marked by full squares.

Figure 3. Chart of nuclides for which calculated masses are available according to the DuZu96 mass tables. 9321 masses for $Z = 2 - 122, N \leq 207$. The Green approximation of the beta stability is marked by full squares.

separation degree of freedom, $R_i = R_0 - R_e$ is the initial separation distance, $R_t = R_e + R_d$ is the touching point separation distance, $R_j = r_0 A^{1/3}_j$ ($j = 0, c, d$; $r_0 = 1.2249$ fm) are the radii of parent, emitted, and daughter nuclei, respectively, and $E^0_i = E_i - Q$ is the barrier height before correction. The interaction energy at the top of the barrier, in the presence of a non-negligible angular momentum, $l\hbar$, is given by $E_i = E_c + E_d = e^2 Z_e Z_d / R_t + h^2 l(l+1) / (2 \mu R_t^2)$. The two terms of the action integral $K$, corresponding to the overlapping $K_{ov}$ and separated $K_s$ fragments, are calculated analytically [11, 22]. The potential barrier shape similar to that which we considered within the ASAF model was calculated by using the macroscopic-microscopic method [24].

3. Mass tables

Half-life calculations are very sensitive to the $Q$-values. The closest to reality are the updated table of evaluated experimental masses AME11 [26], but many masses are still not available for new SHs in this table. We have also used calculated masses:

- Macroscopic-Microscopic: FRDM95 (Möller, Nix, Myers, Swiatecki) [27],
- Thomas-Fermi: MySw94 [28],
- Phenomenological/Hybrid: KTUY05 (Koura, Tachibana, Uno, Yamada) [29],
• Hartree-Fock: **APDT95** (Aboussir, Pearson, Dutta, Tondeur) [30],
• Semiempirical Shell Model: **LiMaZe01** (Liran, Marinov, Zeldes) [31, 32],
• Shell Model: **DuZu96** (Du, Zuker) [33].

We show in figures 1, 2, and 3 the nuclides for which calculated masses are available according to the tables MySw94, APDT95, and DuZu96, respectively. For the region of interest \(Z = 104 - 124\) the beta stability line goes through \(N_g = 106\) for \(Z = 104\) and \(N_g = 206\) for \(Z = 124\). It is clearly seen that the most complete mass table of these three is MySw94. The other two will allow us to make calculations only for some of the neutron deficient heaviest SHs.

When using calculated masses for parent and daughter nuclei we take into account the nuclides stable against one proton, two protons, one neutron and two neutrons spontaneous emissions which leads to a smaller number of parent nuclei than those shown in figures 1-3.

4. **A trend toward shorter half-lives for heavy superheavies**

Besides the emitted clusters with \(Z_e \leq 28\) (Be, Ar, Ca, Ti, V, Cr, Mn, Fe, and Ni), many other types of new HPR with \(Z_e > 28\) appear when we use the MySw94 calculated mass table to determine the \(Q\)-values: Cu, Zn, Ga, Ge, As, Se, Br, Kr, Rb, Sr, Y, Zr, Nb, and Mo (see figure 4). Many of the SH nuclides are \(^8\)Be emitters, but they have a very low branching ratio \(b_\alpha\). Most frequently occurs the doubly magic \(^{78}\)Ni radioactivity.

![EMITTED CLUSTERS](image)

**Figure 4.** (color online) Chart of superheavy cluster emitters with atomic numbers \(Z = 104 - 124\). The \(Q\) values are calculated using the MySw94 mass table. Black squares mark the Green approximation of the line of beta stability.

An even-odd staggering of HPR half-lives was observed [11] leading to shorter \(T_c\) for even \(N\) nuclides compared to the neighboring odd \(N\) ones. In order to avoid such a complication we consider in figure 5 for HPR only the odd \(N\) isotopes. Two important trends are observed: (1) both \(T_c\) and \(T_\alpha\) are shorter for SHs with larger atomic number, and (2) for some of the isotopes of \(Z = 122, 123\) and \(Z = 124\) elements cluster decay half-life may be shorter than that of \(\alpha\) decay: \(T_c < T_\alpha\) (or the branching ratio \(b_\alpha = T_\alpha/T_c\) becomes larger than unity when the atomic number of the parent nucleus increases over \(121\)). Very large values of \(\log_{10} T_\alpha\) (over 13) occurring at \(N = 195\) for \(Z = 121 - 124\) and at \(N = 193, 195\) for \(Z = 124\), due to very low value of \(Q_\alpha\) have been removed from the plot.

We observe in table 1 that large differences in \(Q\)-values for HPR calculated with various mass tables occur very frequently and make an important contribution to the broad range of branching ratios \(b_\alpha\) for any particular nucleus. In this table we kept for \(\alpha\) decay the \(Q_\alpha\)-value determined using AME11 experimental masses and the half-life \(T_\alpha\) calculated within semFIS model. The semFIS calculations are in good agreement with experimental values for \(\log_{10} T_\alpha\) of \(292, 293, 294\) which are \(-1.74\) s, \(-1.84\) s, and \(-3.05\) s, respectively. By taking as the optimum AME11, the closest values of \(Q_\alpha\) (and consequently of \(T_\alpha\), and \(b_\alpha\)) are obtained when we use the KTUY05 mass table. The most optimistic results are based on the LiMaZe01 table and the most pessimistic on the FRDM95 masses. The sensitivity of half-life values to a
Figure 5. Decimal logarithm of the half-lives of superheavy nuclei with atomic numbers \(121 - 124\) against \(\alpha\) decay (open circles) and HPR for odd-neutron isotopes (open squares) versus the neutron number of the parent nucleus. \(Q\) values are calculated using the MySw94 mass tables. Vertical lines correspond to \(N = 186, 196\).

Figure 6. Comparison of alpha-decay half-lives calculated with ASAF (full squares) and semFIS models vs. the neutron number of the parent nucleus. Both even and odd \(N\) values are considered. Vertical lines correspond to \(N = 186, 196\). \(Q\)-values are calculated using the MySw94 mass tables.

A small variation of the \(Q_c\) value is also evident. Let us take for example the case of \(^{296}\)Lv\(_{116}\): the decrease of \(Q_c\) with 1.17 MeV from 284.90 (AME11) to 283.73 (APDT95) leading to an increase of 1.32 orders of magnitude in \(T_c\), whereas a decrease with 4.19 MeV (FRDM95) gives 4.72 orders of magnitude increase in \(T_c\).

More elaborate models should be used (see, e.g., [34, 4]) in order to estimate the competition of spontaneous fission.

An estimation of the accuracy gives the standard rms deviation of \(\log_{10} T\) values:

\[
\sigma = \left( \frac{1}{n-1} \sum_{i=1}^{n} \left[ \log_{10}(T_i/T_{exp}) \right]^2 \right)^{1/2}
\]  \hspace{1cm} (7)
Table 1. $Q$ values in MeV, half-lives and branching ratios for the most probable HPR of SH nuclei with $Z = 116, 117, 118$, $N = 176$ obtained by using seven mass tables: AME11, DuZu96, LiMaZe01, APDT95, KTUY05, MySw94, and FRDM95.

| Parent  | Mass table | $Q/\alpha$ | $\log_{10} T_\alpha (s)$ | Emitted cluster | $Q/c$ | $\log_{10} T_c (s)$ | $\log_{10} b_\alpha$ |
|---------|------------|------------|--------------------------|-----------------|------|-------------------|---------------------|
| $^{292}_{116}$ | AME11 | 10.809 | -1.55 | $^{84}$Se | 284.90 | 0.53 | -2.08 |
| | DuZu96 | 281.09 | 4.82 | -6.37 |
| | LiMaZe01 | 288.20 | -3.21 | 1.66 |
| | APDT95 | 283.73 | 1.85 | -3.40 |
| | KTUY05 | 284.39 | 1.10 | -2.66 |
| | MySw94 | 282.02 | 3.77 | -5.32 |
| | FRDM95 | 280.71 | 5.25 | -6.80 |
| $^{293}_{117}$ | AME11 | 11.183 | -2.03 | $^{85}$Br | 293.73 | 1.16 | -3.19 |
| | DuZu96 | 290.40 | 4.90 | -6.93 |
| | LiMaZe01 | 296.73 | -2.24 | 0.21 |
| | APDT95 | 292.57 | 2.47 | -4.50 |
| | KTUY05 | 293.52 | 1.39 | -3.42 |
| | MySw94 | 291.58 | 3.57 | -5.60 |
| | FRDM95 | 290.04 | 5.31 | -7.34 |
| $^{294}_{118}$ | AME11 | 11.811 | -3.31 | $^{86}$Kr | 303.69 | -2.01 | -1.30 |
| | DuZu96 | 300.22 | 1.86 | -5.17 |
| | LiMaZe01 | 306.27 | -4.91 | 1.60 |
| | APDT95 | 302.23 | -0.39 | -2.93 |
| | KTUY05 | 303.11 | -1.37 | -1.94 |
| | MySw94 | 302.06 | -0.20 | -3.12 |
| | FRDM95 | 300.29 | 1.78 | -5.09 |

Compared to calculations within ASAF model, lower values of $\sigma$ for $\alpha$ decay half-lives may be obtained [35] within our UNIV (universal curve) [36] and semFIS (semiempirical) models. For 44 even-even nuclei we obtained $\sigma = 0.164$ within semFIS, $\sigma = 0.267$ within UNIV and $\sigma = 0.402$ within ASAF model. For 25 odd-odd nuclei $\sigma = 0.451, 0.456$ and 0.795, respectively.

A comparison of $\log_{10} T_{\text{ASAF}}$ with $\log_{10} T_{\text{semFIS}}$ is shown in figure 6. Very large values of $\log_{10} T_\alpha$ (over 13) occurring at $N = 195, 196$ for $Z = 121 - 124$ and at $N = 193 - 196$ for $Z = 124$, due to very low value of $Q_\alpha$, have been removed from the plot. We assume that semFIS calculations are closer to reality, particularly in the vicinity of the neutron and proton magic numbers. From figure 6 one can see that odd-even staggering are more pronounced for semFIS than ASAF model for odd-$N$ values in the neighborhood of $N = 184$ when $N \leq 186$. The agreement between ASAF and semFIS values is rather good.

In conclusion we found that calculated half-lives $T_c$ against HPR and the branching ratios relative to $\alpha$ decay are showing a trend toward shorter $T_c$ and larger $b_\alpha$ for the heaviest SHs. In the vicinity of neutron magic number $N = 184$ the half-lives for $\alpha$ decay calculated within ASAF model are shorter than those determined with semFIS which takes into account the influence of closed shells. There is a need of extending atomic mass models in the region of heaviest neutron-rich superheavy nuclides. The accuracy of calculated masses in the region of heaviest SHs should be improved in order to make reliable predictions of half-lives.
Acknowledgments
This work was partially supported within the IDEI Programme under Contracts No. 43/05.10.2011 and No. 42/05.10.2011 with UEFISCDI, Bucharest.

References
[1] Sobiczewski A 2011 Radiochimica Acta 99 395
[2] Greiner W and Poenaru D N 2010 Cluster Structure of Atomic Nuclei ed Brenner M (Research Signpost, Trivandrum, India) chap 5, pp 119
[3] Düßlmann C E 2012 Search for superheavy elements at GSI, Invited talk at this Conference
[4] Schunck N 2012 Density functional theory approaches to fission, Talk at this Conference
[5] Hofmann S 2011 Radiochim. Acta 99 405
[6] Morita K et al 2007 J. Phys. Soc. Jpn. 76 045001
[7] Oganesian Y T 2011 Radiochimica Acta 99 429
[8] Düßlmann C E et al 2010 Phys. Rev. Lett. 104 062503
[9] Düßlmann C E et al 2010 Phys. Rev. Lett. 104 252701
[10] Poenaru D N, Gherghescu R A and Greiner W 2012 Phys. Rev. C 85 034615
[11] Blendowske R, Flissbach T and Walliser H 1987 Nucl. Phys. A 464 75
[12] Lovas R G, Liotta R J, Insolia A, Varga K and Delion D S 1998 Phys. Rep. 294 265
[13] Adamian G G, Antonenko N V and Zubov A S 2005 Phys. Rev. C 71 034603
[14] Kuklin S, Adamian G and Antonenko N 2005 Phys. Rev. C 71 014301
[15] Denisov V Y and Khudenko A A 2004 Atomic Data Nucl. Data Tables 85 185
[16] Qi C, Xu F R, Liotta R J and Wyss R 2009 Phys. Rev. Lett. 103 072501
[17] Myers W D and Swiatecki W J 1966 Nucl. Phys. A 81 1
[18] Aboussir Y, Pearson J M, Dutta A K and Tondeur F 1995 Atomic Data Nucl. Data Tables 61 127
[19] Liran S, Marinov A and Zeldes N 2000 Phys. Rev. C 62 047301 (Preprint nucl-th/0006045, nucl-th/0102055v1)