

ALPHA DECAY AND CLUSTER RADIOACTIVITY OF HEAVY SUPERHEAVY NUCLEI

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Abstract. Superheavy nuclei of interest for the forthcoming synthesis of the isotopes with $Z = 119, 120$ are investigated. One of the very interesting experiment was performed at the velocity filter SHIP (GSI Darmstadt) trying to produce $^{299}120$ in a fusion reaction $^{258}\text{Cm}(^{54}\text{Cr}, 3n)^{299}120$. We report calculations of α decay half-lives using four models: AKRA (Akrawy), ASAF (Analytical Super-Asymmetric Fission), UNIV (Universal Formula), and semFIS (Semi-empirical formula based on Fission Theory). The released energy, Q , is calculated using the theoretical model of atomic masses WS4, see Phys. Rev. C **84** (2011) 014333 and Phys. Lett. B **734** (2014) 215. For $^{92,94}\text{Sr}$ cluster radioactivity of $^{300,302}120$ we predict a branching ratio relative to α decay of -0.10 and 0.49 , respectively, which is expected to be measured in a forthcoming experiment.

Key words: Alpha decay; Heavy-particle decay; Lifetimes.

1. INTRODUCTION

The interest for α decay (α D) is strongly stimulated by the search for heavier and heavier superheavies – nuclides with $Z > 103$, produced by fusion reactions, which may be identified easily if a chain of α D leading to a known nucleus may be measured. Superheavy nuclei (SHN) [1–3] with atomic number Z up to 118, have been produced by two kinds of fusion reactions:

(1) almost cold fusion (with one evaporated neutron) at GSI, Germany [4, 5] and RIKEN Japan [6] based on the doubly magic target ^{208}Pb or its neighbour ^{209}Bi , and

(2) hot fusion (with three or four evaporated neutrons) at JINR Dubna, Russia and Livermore Nat. Lab., USA [7, 8] with the ^{48}Ca projectile.

One of the very interesting experiment was performed at the velocity filter SHIP (GSI Darmstadt) trying to produce $^{299}120$ in a fusion reaction $^{258}\text{Cm}(^{54}\text{Cr}, 3n)^{299}120$ [9, 10].

Wang *et al.* [11] compared 20 models of atomic masses and 18 relationships for calculation of α decay half-lives. They found that “*SemFIS2 (semi-empirical based on fission theory) formula is the best one to predict the alpha-decay half-lives ... In addition, the UNIV2 (universal formula) formula with fewest parameters ... work well in prediction of the superheavy nuclei (SHN) alpha-decay half-lives*”. Among these, an important role is played by [12, 13].

Wang *et al.* recommend the atomic mass model WS4 [14, 15]. In case of $^{297,299}_{119}$ nuclei we could not get the Q-values by using the model WS4, hence the KTUY05 model [16] was used instead.

From the attempts to synthesize $Z = 119, 120$ isotopes we selected [9, 10] dealing with $Z = 120$, without any positive result until now; we shall study one of the chains starting with $^{299}_{120}$ and ending with the fissioning nucleus $^{283}_{84}\text{Rg}$.

In order to calculate α D half-lives, we shall use semFIS, UNIV, ASAF [17–23] and AKRA [24]. A FORTRAN77 computer program [25] gives us the possibility to improve the parameters of the ASAF model in agreement with a given set of experimental data. The UNIV model was updated in 2011 [26].

A parent nucleus, AZ , disintegrates with emission of a light particle, $^{Ae}Z_e$, and a heavy daughter $^{Ad}Z_d$



The kinetic energy of the α particle is related to Q-value by the relationship $E_k = QA_d/A$ and Q-value is calculated from the atomic masses.

In the region of superheavy nuclei the majority of researchers prefer to use the Viola-Seaborg formula [27]. For nuclei with $Z = 84 - 110$ and $N = 128 - 160$, for which both Q_{α}^{exp} and T_{exp} values are available, new optimum parameter values [12] have been determined. A new semiempirical formula for the alpha decay half-lives [22] was developed. The analytical and numerical superasymmetric fission models (ASAF and NUSAF) [23] were used together with fragmentation theory developed by the Frankfurt School, and with penetrability calculations, to predict cluster (or heavy particle) radioactivity [28, 29]. The extended calculations, *e.g.* [30] have been used to guide the experiments and as a reference for many theoretical developments. For some isotopes of SHs, with $Z > 121$, cluster decay modes may compete with α D and spontaneous fission [31, 32].

A self-consistent model for light nuclei was developed, showing binding energies for 8 nuclei, *e.g.* ^4He , ^8Be , ^{12}C , ^{16}O up to ^{36}Ar [33]. Superheavy nuclei and their α -decay have been studied in a chiral SU(3) Model [34]; calculations were performed in the mean-field approximation with parameters fitted to properties of finite nuclei. $Z = 120$ and $N = 172, 184, 198$ are predicted to be magic numbers.

The heaviest uranium isotopes with neutron numbers in the range (170 – 260) have been investigated within relativistic mean field and Skyrme-type approaches

[35].

Interesting calculations have been also performed by Polish scientists [36, 37].

2. THE MODELS

In the following we shall give informations concerning the AKRA, ASAF, UNIV, and semFIS models [24, 38]. More details can be found in the e-print [39]. We express the half-lives in decimal logarithm of the values in seconds, $T = \log_{10} T_{1/2}(s)$. The half-life of a parent nucleus AZ against the split into a cluster $A_e Z_e$ and a daughter $A_d Z_d$

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

is calculated by using the WKB quasiclassical approximation, according to which the action integral is expressed as [40]

$$K = \frac{2}{\hbar} \int_{R_a}^{R_b} \sqrt{2B(R)[E(R) - Q]} dR \quad (3)$$

with $B = \mu$ – the reduced mass, $K = K_{ov} + K_s$ (overlapping and separated fragments), and $E(R)$ is the total deformation energy. R_a, R_b are the turning points, defined by $E(R_a) - Q = E(R_b) - Q = 0$. The overlapping integral, K_{ov} is the action integral from the initial separation distance between centers (parent configuration) up to the scission point. The separation integral, K_s , is the action integral taken from the part of the barrier when the fragments are separated. K_{ov} is interpreted as the cluster preformation factor.

The AKRA model [24] was derived by adding few parameters to the one developed by Royer [41]; this formula is defined as

$$T_{1/2} = a + bA^{1/6} \sqrt{Z} + \frac{cZ}{\sqrt{Q_\alpha}} \quad (4)$$

with initial parameters $a = -27.657; -28.408; -27.408,$ and $-24.763,$
 $b = -0.966; -0.920; -1.038,$ and $-0.907,$ and $c = 1.522; 1.519; 1.581,$ and 1.410
for e-e (even-even), e-o (even-odd), o-e (odd-even), and o-o (odd-odd), respectively.

The new relationship is obtained by introducing $I = (N - Z)/A$ and the new parameters d and e :

$$T_{1/2} = a + bA^{1/6} \sqrt{Z} + \frac{cZ}{\sqrt{Q_\alpha}} + dI + eI^2, \quad (5)$$

where after a fit with a comprehensive set of experimental data [24] we got $a = -27.989, b = -0.940, c = 1.532, d = -5.747, e = 11.336.$

For ASAF we replace in Eq. 3 $E(R) - Q$ by $[E(R) - E_{corr}] - Q$. E_{corr} is a correction energy [42] taking into account the fact that Myers-Swiatecki's liquid

drop model [43] overestimates fission barrier heights. The turning points of the WKB integral are:

$$R_a = R_i + (R_t - R_i)[(E_v + E^*)/E_b^0]^{1/2} \quad (6)$$

$$R_b = R_t E_c \{1/2 + [1/4 + (Q + E_v + E^*)E_l/E_c^2]^{1/2}\} / (Q + E_v + E^*), \quad (7)$$

where E^* is the excitation energy concentrated in the 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, $R_j = r_0 A_j^{1/3}$ ($j = 0, e, d$; $r_0 = 1.2249$ fm) are the radii of parent, emitted, and daughter nuclei, and $E_b^0 = E_i - Q$ is the barrier height before correction. The interaction energy, in the presence of a non negligible angular momentum, $l\hbar$, is given by:

$$E(R) = E_C + E_{Y+E} + E_l = e^2 Z_e Z_d / R + E_{Y+E} + \hbar^2 l(l+1) / (2\mu R^2) \quad (8)$$

where

$$E_C = \frac{2\pi}{3} (\rho_{eH}^2 F_{CH} + \rho_{eL}^2 F_{CL} + 2\rho_{eH}\rho_{eL} F_{CHL}) \quad (9)$$

with ρ_{eH} and ρ_{eL} the charge densities of the heavy and light fragment, respectively, F_{IJ} are shape dependent quantities, and

$$E_{Y+E} = \frac{c_s}{4\pi r_0^2} \int_0^{2\pi} \int_{R_i}^{R_f} \int_{R_i}^{R_f} E_{YH}(z) E_{YL}(z, z') d\phi dz dz', \quad (10)$$

where c_s is the surface tension coefficient, $r_0 = 1.2$ fm. E_{YH} and E_{YL} depend on the heavy and light fragment shapes.

The two terms of the action integral K are calculated by analytical formulas (approximated for K_{ov} and exact for K_s in case of separated spherical shapes within the LDM):

$$K_{ov} = 0.2196 (E_b^0 A_e A_d / A)^{1/2} (R_t - R_i) \left[\sqrt{1 - b^2} - b^2 \ln \frac{1 + \sqrt{1 - b^2}}{b} \right] \quad (11)$$

$$K_s = 0.4392 [(Q + E_v + E^*) A_e A_d / A]^{1/2} R_b J_{rc}; \quad b^2 = (E_v + E^*) / E_b^0 \quad (12)$$

$$J_{rc} = (c) \arccos \sqrt{(1 - c + r) / (2 - c)} - [(1 - r)(1 - c + r)]^{1/2} + \sqrt{1 - c} \ln \left[\frac{2\sqrt{(1 - c)(1 - r)(1 - c + r)} + 2 - 2c + cr}{r(2 - c)} \right], \quad (13)$$

where $r = R_t / R_b$ and $c = r E_c / (Q + E_v + E^*)$. In the absence of the centrifugal contribution ($l = 0$), one has $c = 1$.

The choice $E_v = E_{corr}$ allows to get a smaller number of parameters. Shell and pairing effects are included in $E_{corr} = a_i(A_e)Q$ ($i = 1, 2, 3, 4$ for even-even, odd-even, even-odd, and odd-odd parent nuclei). For a given cluster radioactivity there

are four values of the coefficients a_i , the largest for even-even parent and the smallest for the odd-odd one. The shell effects for every cluster radioactivity is implicitly contained in the correction energy due to its proportionality with the Q value, which is maximum when the daughter nucleus has a magic number of neutrons and protons.

The UNIV (Universal Formula) was obtained starting with the decay constant $\lambda = \ln 2/T$, expressed as a product of three (model dependent) quantities $\lambda = \nu S P_s$, where ν is the frequency of assaults on the barrier per second, S is the preformation probability of the cluster at the nuclear surface, and P_s is the quantum penetrability of the external potential barrier. By assuming that the frequency ν remains practically constant and the preformation depends only on the mass number of the emitted particle, A_e one has a single straight line:

$$\log T = -\log P_s - 22.169 + 0.598(A_e - 1), \quad (14)$$

where $-\log P_s = c_{AZ} \left[\arccos \sqrt{r} - \sqrt{r(1-r)} \right]$ with $c_{AZ} = 0.22873(\mu_A Z_d Z_e R_b)^{1/2}$, $r = R_t/R_b$, $R_t = 1.2249(A_d^{1/3} + A_e^{1/3})$, $R_b = 1.43998 Z_d Z_e / Q$, and $\mu_A = A_d A_e / A$.

For α -decay of even-even nuclei, $A_e = 4$, one has

$$\log T = -\log P_s + c_{ee}, \quad (15)$$

where $c_{ee} = \log S_\alpha - \log \nu + \log(\ln 2) = -20.375$. We can find new values for c_{ee} and we also can extend the relationship to even-odd, odd-even, and odd-odd nuclei, by fitting a given set of experimentally determined alpha decay data.

The semFIS (Semiempirical relationship based on fission theory of α -decay) was derived in order to improve the behaviour in the vicinity of magic numbers

$$\log T = 0.43429 K_s \chi - 20.446 \quad (16)$$

where $K_s = 2.52956 Z_{da} [A_{da}/(A Q_\alpha)]^{1/2} [\arccos \sqrt{x} - \sqrt{x(1-x)}]$, and the numerical coefficient χ , close to unity, is a second-order polynomial $\chi = B_1 + B_2 y + B_3 z + B_4 y^2 + B_5 y z + B_6 z^2$ in the reduced variables y and z , expressing the distance from the closest magic-plus-one neutron and proton numbers N_i and Z_i : $y \equiv (N - N_i)/(N_{i+1} - N_i)$; $N_i < N \leq N_{i+1}$ $z \equiv (Z - Z_i)/(Z_{i+1} - Z_i)$; $Z_i < Z \leq Z_{i+1}$ with $N_i = \dots, 51, 83, 127, 185, 229, \dots$, $Z_i = \dots, 29, 51, 83, 115, \dots$, and $Z_{da} = Z - 2$, $A_{da} = A - 4$. The coefficients B_i are obtained by fit with experimental data, using a computer program [25] in which the parameters are determined to assure a minimum standard rms deviation

$$\sigma = \left\{ \sum_{i=1}^n [\log(T_i/T_{exp})]^2 / (n-1) \right\}^{1/2} \quad (17)$$

3. RESULTS AND CONCLUSIONS

A global indicator for a given model is the weighted mean value, *e.g.*

$$\sigma_{semFIS534} = \frac{173\sigma_{e-e} + 134\sigma_{e-o} + 123\sigma_{o-e} + 104\sigma_{o-o}}{534} = 0.40803, \quad (18)$$

to compare calculations within semFIS with experimental data for 534 emitters: 173 even-even, 134 even-odd, 123 odd-even, and 104 odd-odd.

We give in Table 1 the cluster emission with Q-values calculated using WS4 model, and half-lives with ASAF model. The most interesting results are those obtained for the heaviest nuclides: $^{300,302}_{120}$ with branching ratios $B_\alpha = -0.10$ and 0.49, respectively, $^{299,301}_{120}$ with $B_\alpha = -1.49$ and -1.17 , $^{297,299}_{119}$ with $B_\alpha = -1.99$ and -3.21 , and $^{300}_{119}$ with $B_\alpha = -3.75$.

Few possible alpha decay chains of even-even and odd-mass SH emitters are given in Figs. 1 and 2, Tables 2, 4, and Table 5, respectively. As mentioned in the caption of Fig. 1, the kinetic energies are calculated with WS4 model and the decay half-lives with ASAF model. Figure 1 illustrates the alpha decay chains with high branching ratios relative to α -decay of even-even nuclei $^{300}_{120}$ and $^{302}_{120}$ (-0.10 and 0.49, respectively). For odd mass (A) and atomic number (Z) $^{297}_{119}$ and $^{299}_{119}$ (Fig. 2) the branching ratios are lower with at least one order of magnitude.

Table 1

Cluster radioactivities of even-even emitters. Q-values obtained using WS4 model, and half-lives with ASAF model.

A	Z	A_e	Z_e	Q_c (MeV)	$\log_{10} T_c(s)$	$B_\alpha = T_\alpha - T_c$
300	120	92	38[Sr]	321.36	-5.73	-0.10
302	120	94	38[Sr]	320.04	-5.26	0.49

Table 2

Cluster radioactivities of even-odd emitters. Q-values obtained using WS4 model, and half-lives with ASAF model.

A	Z	A_e	Z_e	Q_c (MeV)	$\log_{10} T_c(s)$	$B_\alpha = T_\alpha - T_c$
299	120	91	38[Sr]	321.48	-2.70	-1.49
301	120	93	38[Sr]	320.58	-3.86	-1.17

In Table 1 we present the results obtained by calculating with ASAF model the half-lives T_c and branching ratios relative to alpha-decay B_α and the Q_c -values with the WS4 model for ^{38}Sr radioactivity of $^{300}_{120}$ and of $^{302}_{120}$ nuclei. The branching ratios of -0.10 and 0.40 looks very promising for future experiments.

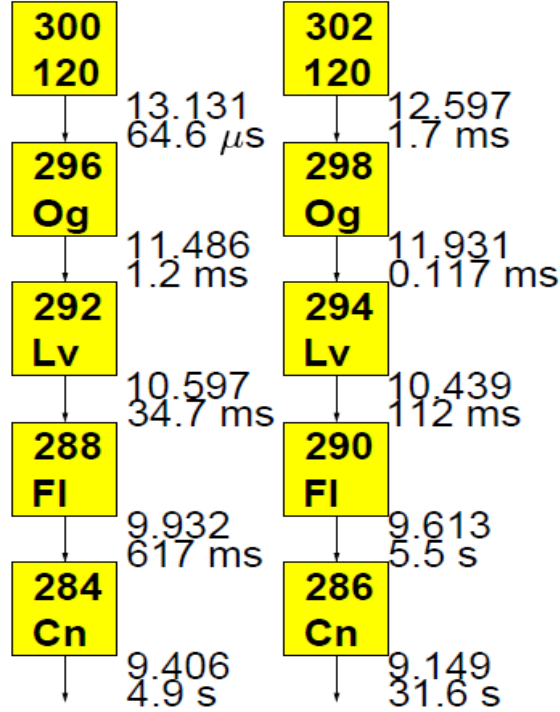


Fig. 1 – Few alpha decay chains of even-even SH emitters. We give the two emitters for which the branching ratios of cluster decay with respect to α decay is close to unity. We give the kinetic energy (MeV) and the half-life of the parent nucleus. Kinetic energies are calculated with WS4 model, and half-lives with ASAF model.

Table 3

Comparison of alpha decay half-lives obtained with four different models.

A	Z	Q_α (MeV)	$\log_{10} T_\alpha$ (s) ASAF	$\log_{10} T_\alpha$ (s) AKRA	$\log_{10} T_\alpha$ (s) UNIV	$\log_{10} T_\alpha$ (s) SemFis
297	119	12.210	-3.695	-0.450	-4.124	-3.717
299	119	12.696	-4.730	-1.357	-5.142	-4.739
300	120	13.308	-5.833	-4.415	-6.113	-5.765
302	120	12.766	-4.769	-4.769	-5.117	-4.682

In case of $^{299}120$ and $^{301}120$ shown in Table 2, the branching ratios are lower with more than one order of magnitude (-1.49 and -1.17 , respectively).

A comparison of half-life calculation for four superheavy nuclei ($^{297}119$, $^{299}119$, $^{300}120$, and $^{302}120$) using the ASAF, AKRA, UNIV and SemFIS models is presented in Table 3. One can see that compared with the SemFIS results one can get for $^{297,299}119$ large discrepancies with AKRA model ($\log T_\alpha = -0.450$ compared to -3.717 and -1.357 compared to -4.739 , respectively). On the other hand ASAF

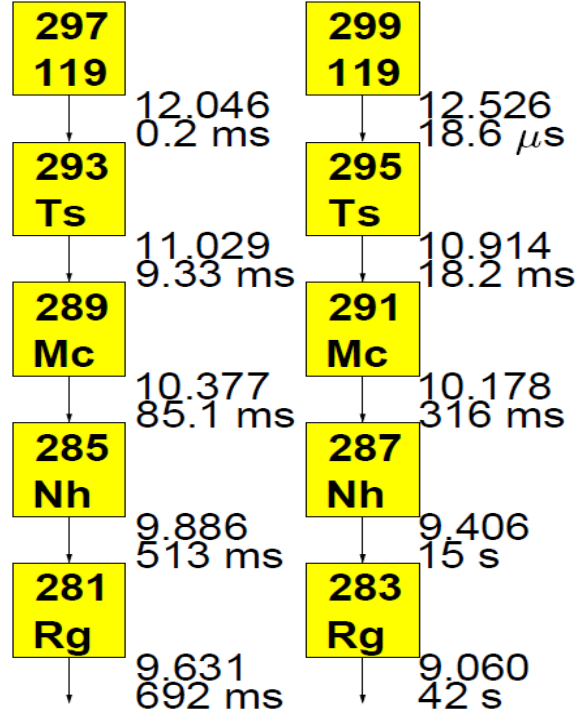


Fig. 2 – Alpha decay chains of odd-mass SH emitters. The branching ratios of cluster decay with respect to α decay is less than unity. Calculations are performed with the same models as for Fig. 1.

Table 4

Alpha decay chains of $^{299,300,302}_{120}$ nuclei. Kinetic energy and the half-life of every decay mode are given. WS4 and ASAF models are used.

A	Z	A_e	Z_e (symbol)	E_k (MeV)	T_α
300	120	296	118 (Og)	13.131	64.60 μ s
296	118	292	116 (Lv)	11.486	1.20 ms
292	116	288	114 (Fl)	10.597	34.70 ms
288	114	284	112 (Cn)	9.932	617 ms
284	112	280	110 (Ds)	9.406	4.9 s
302	120	298	118 (Og)	12.597	1.70 ms
298	118	294	116 (Lv)	11.931	0.117 ms
294	116	290	114 (Fl)	9.613	5.5 s
290	114	286	112 (Cn)	9.149	31.6 s

Table 5

Alpha decay chains of $^{297,299}119$ nuclei. Kinetic energy and the half-life of every decay mode are given. WS4 and ASAF models are used.

A	Z	A_e	Z_e (symbol)	E_k (MeV)	T_α
297	119	293	117 (Ts)	12.046	0.20 ms
293	117	289	115 (Mc)	11.029	9.33 ms
289	115	285	113 (Nh)	10.377	85.10 ms
285	113	281	111 (Rg)	9.886	513 ms
284	112	280	109 (Mt)	9.631	692 s
299	119	295	117 (Ts)	12.526	18.60 μ s
295	117	291	115 (Mc)	10.914	18.20 ms
291	115	287	113 (Nh)	9.406	15 s
287	113	283	111 (Rg)	9.060	42 s

and UNIV give much closer results (-3.695 and -4.124 compared to -3.717 and -4.730 and -5.142 compared to -4.739). For the even-even nuclei $^{300,302}120$, the discrepancies are much smaller. As expected, the best results are given by SemFIS and ASAF models. From these results we may give the following values of the error bars: 1.5 and 0.4 orders of magnitude for the half-lives of $^{300}120$ and $^{302}120$, respectively, and 3.7 and 3.8 orders of magnitude for $^{297}119$ and $^{299}119$, respectively.

In Table 4 we present the results obtained with ASAF for alpha decay half-lives T_α and with WS4 for the kinetic energy E_k for 9 superheavy nuclei. The corresponding daughter nuclei are denoted with A_e and Z_e . The shortest half-life is 64.60μ s for $^{300}120$ and the daughter ^{118}Og and the largest one is 31.6 s for ^{290}Fl with ^{112}Cn daughter. A similar Table is Table 5 for other 9 superheavy nuclei. Here the minimum half-life is 18.60μ s for $^{299}119$ with daughter ^{117}Ts and maximum is 692 s for ^{284}Cn with the daughter ^{109}Mt .

A comparison between the data obtained at GSI Darmstadt by Sigurd Hofmann and co-workers [10] and our calculations using the model WS4 for E_k and ASAF for half-lives for α decay is made in the Table 6. E_k^S and T_α^S are taken from the Ref. [10]. We can see that the kinetic energies are comparable, but the half-lives are sometimes very different, *e.g.* 0.0645 s (our calculation) compared to 5.4 s given in Ref. [10] for the alpha emitter $^{299}120$, 29.7 s compared to 0.78 s, for $^{297}114$, 9.42 h *versus* 0.0184 s for $^{267}104$, etc. Maybe some of these discrepancies are due to the fact that there are isomeric states that have not been taken into account up to now. Among the best agreement one has 0.0373 s compared to 0.0184 s for $^{291}116$, 14.70 *versus* 19.9 for $^{287}113$, 0.85 and 0.42 for $^{279}110$, 0.0912 and 0.0184 for $^{286}114$, 0.154 and 0.261 for $^{293}116$, etc.

We introduced a weighted mean value of the rms standard deviation, allowing

Table 6

Comparison between the data given in Ref. [10] and our calculations using the model WS4 for E_k and ASAF for half-lives for α decay. E_k^S and T_α^S are taken from the Ref. [10].

A	Z	E_k^S (MeV)	E_k (MeV)	T_α^S (s)	T_α (s)
299	120	13.14	12.88	5.4	0.0645
295	118	11.81	11.54	0.261	0.0059
291	116	10.70	10.74	0.0184	0.0373
		10.74		0.026	
		11.81		0.261	
297	114	10.025	10.02	0.78	29.7
		10.70		0.0184	
287	113	10.14	9.41	19.9	14.70
283	112	9.521	9.71	6.5	34.3
		10.70		0.0184	
279	110	9.706	9.94	0.42	0.85
294	118	13.14	11.65	5.4	0.000485
290	116	11.81	10.84	0.261	0.00961
286	114	10.70	10.23	0.0184	0.0912
282	112	10.70	9.97	0.0184	0.112
275	108	10.70	9.30	0.0184	9.88
271	106	10.70	8.76	0.0184	73.70
267	104	10.70	7.77	0.0184	9.42 h
292	116	11.81	10.63	0.261	0.035
288	114	10.70	9.93	0.0184	0.616
284	112	10.70	9.41	0.0184	4.86
293	116	11.81	10.53	0.261	0.154
289	114	10.70	9.83	0.0184	192.
285	112	10.70	10.37	0.0184	1.71
281	110	10.70	9.19	0.0184	287.
277	108	10.70	10.68	0.0184	0.056

to compare the global properties of a given model. We made few predictions concerning possible α D decay chains of future SHs. A comparison between the data reported in the Ref. [10] and our calculations shows either a good agreement (*e.g.* $^{291,293}_{116}$, $^{286}_{114}$, $^{287}_{113}$, and $^{293}_{116}$) or a large disagreement (*e.g.* $^{299}_{120}$, $^{297}_{114}$, and $^{267}_{104}$). In the future it would be useful to take into account a detailed structure of different states, some of them being isomeric states.

The error bars for the half-lives of even-even nuclei are lower (0.4 - 1.7 orders of magnitude) than for odd atomic number $^{297,299}_{119}$.

In conclusion, we recommend to the experimentalists to synthesize the $^{300,302}120$ nuclei in order to observe for the first time that cluster radioactivity (spontaneous emission of $^{92,94}\text{Sr}$ clusters) could be comparable to α decay (tentatively -0.10 and 0.49 branching ratios).

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