COMPETITION BETWEEN ALPHA-DECAY AND SPONTANEOUS FISSION IN Rf, Db, AND Sg ISOTOPES

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The most important decay modes for heavy and super-heavy nuclei are their α-decay and spontaneous fission. This work investigates the evolution and the competition of these modes in isotopic sequences. We define, extrapolate and use approximation schemes and methods for obtaining half-lives incorporating the essential physics of decay process. We compile measurements and theoretical half-lives and tabulate recommended values along with total half-lives. We evaluate and compare the alpha decay and fission half-lives using microscopic-macroscopic and phenomenological methods. The alpha and fission half-lives are obtained in terms of a minimal set of parameters determined from the fit of experimental data and results of the shell model rate theory. A summary of the experimental and calculated α-decay and spontaneous fission half-lives of the isotopes of elements Rf, Db, and Sg is presented. Some half-life extrapolations for nuclides not yet known are also obtained. The α-decay and fission are powerful tools for investigating the detailed aspects of nuclear structure and reaction dynamics. The decay properties are strongly connected with the single-particle structure of nuclei.

Key words: Super-heavy nuclei; α-decay, clustering and scattering amplitudes; resonance tunneling; decay-rates systematics.

1. INTRODUCTION

A general question for a given element of how few or how many neutrons can be contained in nucleus to form a bound system, has been the focus of much research on highly unstable nuclei. For super-heavy nuclei (SHN) the situation is quite different for proton rich and neutron rich nuclei. Due to the strong Coulomb repulsion of protons, the proton drip-line is much closer to the valley of stable nuclei than the neutron drip-line. Also, this repulsion increases very rapidly the reaction decay energies when moving out toward the drip-line and can lead to the occurrence of new decay phenomena.

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Lifetimes of SHN are primarily governed by α-decay and spontaneous fission (SF) and in many cases by their tight competition. There is a great interest and effort both from the experimental side where the methods of separation and measurement are constantly improved [1-17], but also from the theoretical approaches [18-24], involving the development of nuclear models and computation codes for the reaction mechanisms, nuclear structure, reaction energies, and decay properties.

Decay studies of SHN close to proton drip-line are particularly interesting from the following points of view: i) – the lower levels of these nuclei are inevitably near the proton emission thresholds and consequently, the proton and α-decay channels are effectively the only open ones; ii) – long α-chains usually terminate by spontaneous fission; iii) – the estimation of half-lives for chains of isotopes and isotones make possible the access to the basic nuclear-ground state properties of new SHN.

During the last decades great progress was made in the study of near-barrier fusion reactions leading to SHN, their main decay properties and structure. A limitation of the “cold” and “hot” fusion reactions with stable beams for producing SHN consists in the fact that they lead to neutron-deficient isotopes having rather short half-lives. The most stable SHN are expected to be located along the stability line in the region of more neutron-rich nuclei, which is unreachable directly by current fusion reactions.

Detailed knowledge of the decay modes and half-lives in a very wide range of neutron and proton numbers is necessary in planning experiments for the production of neutron-rich SHN. Moreover, the study of decay properties may help us to answer some fundamental but open questions: how far may we still move in synthesis of super-heavy elements by the fusion reactions; where the island of stability is centered; what are the properties of the most stable SHN?

2. HALF-LIVES OF SHN

This work is aimed to the analysis of the decay properties of SHN with respect to α-decay SF. For studying the essential features of α decay we use the shell-model rate theory (SMRT) [25]. The SMRT unifies the advantages of the microscopic description of the α-particle preformation process [26] with the ones of the theory of resonance reactions in describing the reaction dynamics.

All the half-lives calculations performed in this paper are based on the α-decay known data summarized in [7] and presented in Fig. 1 and Table 1. The α-decay is characterized by released energy and the corresponding half-life.

The half-life for α-decay can be estimated quite accurately using the formula [26]:

$$\log T_{\alpha}^{SM}(s) = 10.591(Z_d^{0.6} Q_{\alpha}^{-0.5}) - 56.618 \quad (1)$$

rms = 0.078 for (e-e) nuclei,

$$\log T_{\alpha}^{SM}(s) = 10.148(Z_d^{0.6} Q_{\alpha}^{-0.5}) - 53.386 \quad (2)$$
rms = 0.161 for (e-o, o-e) nuclei,

$$\log T_{\alpha}^{SM}(s) = 10.225(Z_d^{0.6} Q_{\alpha}^{-0.5}) - 53.797 \quad (3)$$
rms = 0.047 for (e-e) nuclei,

where Q_α is the effective decay energy including the kinetic energy of fragments and the screening energy, and rms is the root mean square error.

Some extrapolations for unknown α-decay energies are performed using prescriptions [18]. Here, we use the relationship between the Q values of the neighboring SHN having the same mass ($A_1 = A_2$):

$$Q_2 = Q_1 - (\beta_2 - \beta_1)\left[\frac{2}{3} a_c A^{2/3} (\beta + 2) + 8 a_{sym} \beta\right] \quad (4)$$

with $\beta = (N - Z)/A$ denoting the isospin asymmetry, $Z = A(1-\beta)/2$, and $\beta = (\beta_1 + \beta_2)/2, a_c = 0.71$. The mass dependence of the symmetry energy coefficient is given as $a_{sym} = c_{sym}\left(1 + kA^{-1/3}\right)^{-1}$, where c_{sym} is the volume symmetry energy coefficient of the nuclei and k is the ratio of the surface symmetry coefficient to the volume symmetry coefficient. Here $c_{sym} = 31.1$ and k=2.31 are used without including the uncertainty. Thus, Eq. (4) gives the unknown Q_2 value starting from a measured or known value of Q_1 (see Table 1).

The SF of nuclei is a very complicated process. Knowing the multidimensional potential energy surface only is not sufficient for the accurate determination of the corresponding decay time. The most realistic calculations of the SF half-life are based on the search for the least action path in the multidimensional deformation space [23, 24]. Only few examples of such calculations are known that were performed in a rather restricted area of the nuclear map due to long computing times.

In Ref. [27] was proposed the systematics based on idea of the fission barrier. The coefficients of the systematics are determined by the fitting procedure of the experimental data and the realistic theoretical predictions for the region 100 ≤ Z ≤ 120 and 140 ≤ N ≤ 190. The obtained formula reads as

$$\begin{aligned} \log_{10} T_{SF}^{1}(s) = {} & 1146.44 - C_1\left(Z^2/A\right) + 1.63792\left(Z^2/A\right)^2 \\ & - C_2\left(Z^2/A\right)^3 + B_f\left(7.23613 - C_3\left(Z^2/A\right)\right) + h_{e-o} \end{aligned} \quad (5)$$

The parameters $C_1 = 75.3153, C_2 = 0.0119827, C_3 = 0.0947022$ are determined by the fit of known data and theoretical results. In Eq.(4) h_{e-o} are the even-odd (e-o) corrections: h_{e-o} = 0.0 for (e-e) nuclei; h_{e-o} = 0.80822 for (e-o, o-e) nuclei; h_{e-o} = 1.53897 for (o-o) nuclei.

Here, B_f is the fission barrier, which is calculated as a sum of the liquid-drop barrier B_f (LDM) and the ground-state shell correction δU (g.s.), *i.e.* $B_f = B_f$ (LDM) + δU (g.s.). In this paper the best fit of experimental data is obtained using the following parameters:

$$C_1 = 75.4201, C_2 = 0.0119827, C_3 = 0.0947022.$$

The second formula used for the fission half-lives was taken from Ref. [28]:

$$T_{SF}^{2}(s) = \exp\left\{2\pi\left[c_0 + c_1 A + c_2 Z^2 + c_3 Z^4 + c_4 (N-Z)^2 - \right.\right.$$
$$\left.\left. - \left(0.13323\left(Z^2 / A^{0.33}\right) - 11.64\right)\right]\right\} + 10^{h_{e-o}} \tag{6}$$

where, $Q_{sf} = 0.13323\left(Z^2 / A^{0.33}\right) - 11.64$ is the kinetic energy of the fragments and the values of the parameters are c_0=–195.09227, c_1=3.10156, c_2=–0.04386, c_3=1.40301x10^{-6} , and c_4=–0.03199.

The third one is the formula of Ref. [29] given as

$$\log_{10} T_{SF}^{3}(yr) = a\left(Z^2/A\right) + b\left(Z^2/A\right)^2 + c(N-Z)/(N+Z) +$$
$$d\left[(N-Z)/(N+Z)\right]^2 + e + h_{e-o} \tag{7}$$

where the constants are a = –43.25203, b = 0.49192, c = 3674.3927, d = –9360.6, and e = 580.75058. Notice that to both original formulas [28, 29] we add even-odd corrections h_{e-o} .

3. INPUT DATA

The isotopes of elements with Z=107-112 were successfully produced at GSI (Germany), with Z=110-113 at RIKEN (Japan) and with Z=113-118 were produced at JINR-FLNR Dubna (Russia).

The SHN formed in evaporation-fission reactions tend to reach their stability through a series of structural and dynamical modifications. The quantum shell effects, pairing and deformation contribute to forming favorable energetic stable structures.

Figure 1 shows the known data on SHN with Z=104-106. Here, it can be seen that the SHN decay trough α-decay, SF, β-decay, and internal conversion (IC). The dominant decay mode in SHN is α-decay. The α-half-life varies from one nucleus to another showing the differences in nuclear structure and offering information about the gradual or spontaneous changes with Z and N. Regarding Fig. 1 we can note:

– a constant decrease of the α-half-lives with the increase of proton number.
– a considerable increase of α-half-lives with the increase of neutrons number in the isotopic series.
– large differences between half-lives of proton-rich and neutron rich nuclei.
– α and SF-half-lives of odd-odd nuclei, are always greater than of even-even ones.
– a strong competition α-SF is observed in even-even Rf and Sg isotopes and also in odd-even Db isotopes. Moreover, the one neutron addition in the isotopic chain, leads to the alternance of α-SF channels, while the two neutron addition preserves the dominant decay channel.

4. RESULTS AND DISCUSSION

We have systematically calculated the alpha-decay and SF half-lives of Rf, Db, and Sg isotopes by using the Eqs. (1-3, 5-7)]. The used input data are shown in Fig 1. and the detailed results and listed in Table 1. The experimental (Fig.1) and some predicted (Table 1) α-decay energies are used in calculations of the α-decay half-lives. Thus, the partial α-decay half-lives of 20 new nuclides, which are not included in Fig. 1 are predicted by using the calculated α-decay energies from Eq.(4). The agreement between experimental and theoretical α-decay half-lives is quite good for most nuclides (see also [25, 26]).

In general, the α half-lives presented in Table 1 are in a good agreement with most of the existing α-decay data (Fig. 1), and also with calculated results [30-36]. The agreement with the results of different models is also good, if the same additional corrections for screening and even-odd effects are considered. Notice that for a number of nuclei the experimental half-lives are given by empirical estimates and calculated energies which may include some uncertainties. However, appropriate half-life results of different models show that the essential factor determining α half-live is the emission energy.

Table 1 includes the SF half-lives calculated with three different methods [27], [28], [29]. We can see similar values of T_{SF}^{2} and T_{SF}^{3}, which are different from T_{SF}^{1} values. We should note the values of T_{SF}^{1} are very close to experimental data from Fig. 1. Also, we note that these values are in a good accordance with estimation [37-40].

On the basis of the above estimations, we can evaluate the competition between α-decay and SF decay modes. The theoretical and experimental total half-lives are listed in the last two columns. The agreement between experiment and theory appears to be quite good for most nuclei.

Table 1

Alpha-decay and SF half-lives for isotopes of Rf, Db, and Sg elements.
The values of total T_t half-life (s) come from T_α^{SM} and T_{SF}^1 .

Elem.	Z	N	A	E_α (MeV)	Log T_α^{SM} (s)	Log T_{SF}^1 (s)	Log T_{SF}^2 (s)	Log T_{SF}^3 (s)	Log T_t (s)	Log T_t^{exp} (s)
Rf	104	149	253	8.740	1.737	-4.162	-3.092	-3.539	-4.162	-1.886
	104	150	254	8.730	0.946	-4.516	-3.522	-3.115	-4.516	-4.639
	104	151	255	8.910	1.215	0.712	-0.090	-1.251	0.593	0.214
	104	152	256	8.533*	1.607	0.259	-1.388	-1.176	0.240	-2.190
	104	153	257	9.020	0.886	0.583	0.028	0.348	0.408	0.633
	104	154	258	9.050	-0.069	-1.137	0.031	0.091	-1.173	-1.920
	104	155	259	8.870	1.344	-0.559	0.577	1.291	-0.564	0.447
	104	156	260	8.519*	1.662	-1.498	0.735	0.717	-1.498	-1.677
	104	157	261	8.510	2.487	0.273	0.869	1.606	0.271	0.740
	104	158	262	8.062*	3.286	-0.126	0.724	0.729	-0.127	0.361
	104	159	263	7.749*	5.156	1.063	0.560	1.322	1.063	2.819
	104	161	265	7.990*	4.274	3.597	0.018	0.465	3.514	2.176
	104	163	267	7.880*	4.972	4.825	-0.090	-0.938	4.591	3.918
Db	105	150	255	9.560	-0.364	-2.434	-0.092	-4.499	-2.438	0.230
	105	151	256	9.120	0.897	-0.244	0.187	-2.538	-0.274	0.278
	105	152	257	9.160	0.780	-0.166	-0.092	-2.212	-0.213	0.184
	105	153	258	9.200	0.663	0.169	0.189	-0.594	0.048	0.653
	105	154	259	9.470	-0.107	-1.088	-0.077	-0.602	-1.131	-0.292
	105	155	260	9.120	0.904	0.078	0.221	0.688	0.018	0.181
	105	156	261	8.930	1.476	-0.535	0.065	0.360	-0.539	0.653
	105	157	262	8.530	2.758	1.064	0.342	1.338	1.037	1.544
	105	158	263	8.360	3.310	0.492	0.209	0.703	0.484	1.462
	105	160	265	8.234	3.743	2.599	0.067	0.456	2.569	-
	105	161	266	8.100*	4.239	5.097	0.222	0.849	4.182	3.681
	105	162	267	8.000*	4.571	5.319	-0.076	-0.356	4.500	4.219
	105	163	268	7.970*	4.711	6.411	0.189	-0.236	4.702	5.044
	105	164	269	7.951	4.752	4.639	-0.092	-1.708	4.391	-
	105	165	270	7.940*	4.824	4.153	0.187	-1.848	4.069	4.920
	105	166	271	7.915	4.887	2.028	-0.092	-3.575	2.028	-
Sg	106	149	255	10.131*	-1.581	-6.125	-0.092	-8.238	-6.125	-
	106	150	256	9.695*	-1.352	-5.347	-8.120	-7.466	-5.347	-
	106	151	257	9.739*	-0.550	-3.061	-0.092	-5.257	-3.062	-
	106	152	258	9.782	-1.594	-2.905	-5.242	-4.839	-2.926	-2.481

Table 1 (continued)

	106	153	259	9.590	-0.139	-2.187	-0.092	-2.976	-2.191	-0.236
	106	154	260	9.750	-1.501	-2.677	-3.080	-2.895	-2.705	-2.420
	106	155	261	9.560	-0.053	-1.485	-0.090	-1.362	-1.501	-0.638
	106	156	262	9.266*	-0.083	-1.937	-1.633	-1.604	-1.943	-2.096
	106	156	262	9.785*	-1.596	-1.937	-1.633	-1.604	-2.100	-2.096
	106	157	263	9.250*	0.832	-0.034	-0.058	-0.386	-0.090	0.001
	106	158	264	9.029	0.654	-1.179	-0.901	-0.937	-1.186	-1.431
	106	159	265	8.840	2.073	0.836	-0.015	-0.020	0.811	0.903
	106	160	266	8.692*	1.751	1.668	-0.884	-0.865	1.406	-0.443
	106	161	267	8.200*	4.188	4.035	-0.055	-0.237	3.804	-1.721
	106	162	268	8.276*	3.196	4.213	-1.580	-1.364	3.157	-
	106	163	269	8.570	2.941	5.363	-0.089	-1.011	2.940	2.079
	106	164	270	8.320*	3.042	3.615	-2.991	-2.407	2.939	-
	106	165	271	8.540	3.043	3.278	-0.092	-2.317	2.844	2.158
	106	166	272	7.584	5.858	0.985	-5.115	-3.971	0.985	-
	106	168	274	8.178	3.562	-1.682	-7.953	-6.032	-1.682	-

*Estimated E_α values according to prescription of the Ref. [18].

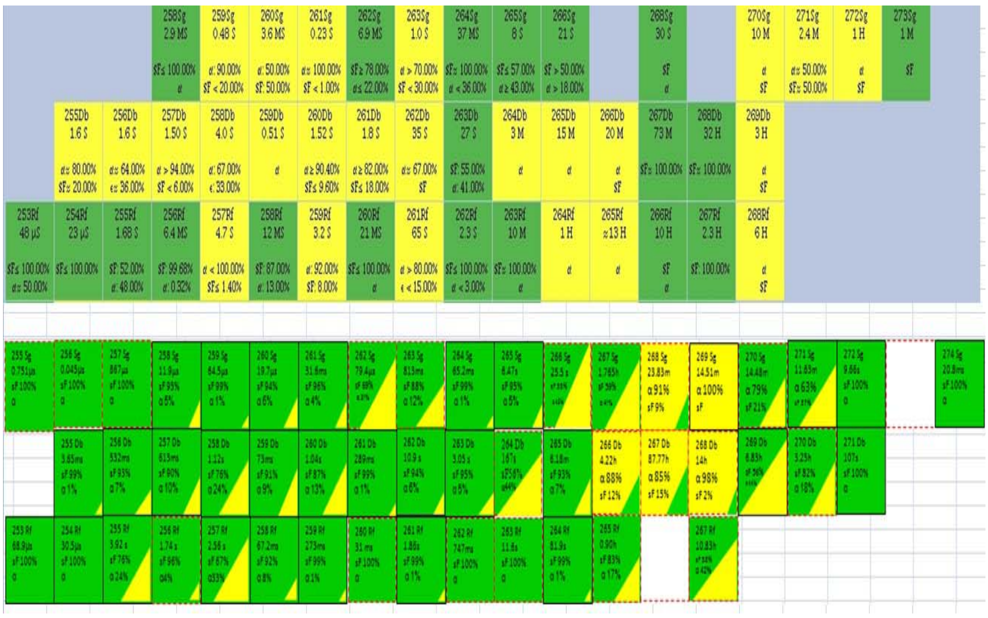

Fig. 1 – Upper part of the figure showing the presently known isotopes of Rf, Db and Sg elements [7]. For each known isotope the element name, mass number, decay modes, branching ratios and half-lives are given. Bottom part includes the results of our calculations for decay modes, branching ratios and half-lives. Extrapolations of some Q_α -values have been made by using the prescription of [18] (the bold dashed lines mark the calculated isotopes).

5. CONCLUSIONS

In summary, we defined and evaluated an approximation scheme that can be used to determine total half-lives of unstable SHN. This scheme takes into account the evolution of the structure in open shell nuclei and the sensible interplay between the microscopic structure and the reaction mechanism. In general, our estimates for half-lives reasonably agree with the experimental data.

The contribution of different effects and corrections (even-odd, shell closures, resonance scattering and screening) on half-lives has been evidenced in different isotopic series and plausible explanations for some discrepancies between calculated and experimental half-lives are given.

For the SHN nuclei it is of importance to predict, even roughly, the radioactive properties of unknown species. We show that such predictions can be made with a fair degree of confidence and this may help in the preparation and identification of new nuclear species in the super-heavy region.

Due to the new experimental and theoretical advances, the α-decay and SF continue to be most important instruments in the investigation of the nuclear structure and reaction mechanisms. Their study offers access to basic characteristics of nuclei such as nuclear mass, energy levels, life times, momentum spins, reaction energies, and emission rates.

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