

METASTABILITY OF  $\alpha$ -STABLE NEUTRON-RICH NUCLEI

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Cluster radioactivities of nuclei far from beta-stability line in the neutron-rich region, where  $\alpha$ -decay is not allowed, have been systematically studied within analytical superasymmetric fission model. Input mass tables calculated by various authors were used to determine the released energy. Half-lives shorter than  $10^{30}$  seconds have been estimated for very neutron-rich parent nuclei with atomic numbers from 57 to 86. The emitted clusters are also proton-deficient nuclei, as for example  $^{22}\text{O}$ ,  $^{46}\text{S}$ ,  $^{50}\text{Ar}$ ,  $^{76}\text{Fe}$ ,  $^{78}\text{Ni}$ , etc.

*1. Introduction*

A cluster ( $A_e, Z_e$ ), may be spontaneously emitted from a parent nucleus ( $A, Z$ ), leaving a daughter ( $A_d, Z_d$ ) if released energy

$$Q = M - (M_e + M_d) \quad (1)$$

is a positive quantity. In this equation  $M$ ,  $M_e$ ,  $M_d$  are the atomic masses of the parent, emitted cluster and daughter nuclei, respectively, expressed in units of energy. The corresponding nucleus is metastable with respect to splitting into two fragments.

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An important observable is the parent nucleus lifetime,  $T$ , relative to this disintegration mode. If  $T$  is low enough, the phenomenon has been detected by using presently available experimental techniques, and the kinetic energy of the emitted cluster  $E_k = Q A_d/A$  is also measured. Up to now the longest observed lifetime is of the order of  $10^{26}$  seconds.

Three fission models and one cluster preformation model have been used in our papers of 1980 to predict new decay modes intermediate between  $\alpha$ -decay and fission (see the review papers 1 and 2 and the references therein). In order to be able to take into consideration the large number of combinations parent - emitted cluster (of the order of  $10^5$ ), we developed since 1980 the analytical superasymmetric fission model (ASAFM) and used it to make the first predictions of nuclear lifetimes. In 1984, before any other model for calculation of the lifetime was developed, we published the estimated half-lives and branching ratios relative to  $\alpha$ -decay for more than 150 decay modes, including all cases experimentally confirmed up to now (see the review papers 3 and 4 and also the Refs. 5 and 6 on  $^{14}\text{C}$ ,  $^{20}\text{O}$ ,  $^{24-26}\text{Ne}$ ,  $^{28,30}\text{Mg}$ , and  $^{32,34}\text{Si}$  radioactivities). A comprehensive table was produced by performing calculations within that model. Subsequently, the numerical predictions of ASAFM have been improved by taking better account of the pairing effect in the correction energy. Cold fission fragments were also considered in another version of the tables<sup>7)</sup>. The above mentioned systematics was further extended in the region of heavier clusters with mass numbers  $A_e > 24$ . Recently, we developed a new semiclassical method to calculate cluster preformation probability as the penetrability of the precession part of the barrier, and on this basis we have shown that fission models are equivalent with preformed cluster models.

The half-life estimations within ASAFM have been updated and the region of parent nuclei expanded far from stability and toward superheavy elements<sup>8)</sup> using the 1988 mass tables<sup>9)</sup> as input data for the  $Q$ -value calculation. The purpose of this paper is to investigate a relatively small area of the nuclear chart, with neutron-rich isotopes of the elements having  $Z = 57 - 97$ , sometime approaching the neutron-drip line, where  $\alpha$ -decay is not allowed because  $Q_\alpha < 0$ . We have previously concluded<sup>10)</sup> that even some »stable« nuclei could be cluster emitters with long half-lives. Now we are presenting shorter half-lives of  $\alpha$ -stable nuclides far from stability.

## 2. Results

The half-life of a parent nucleus against cluster radioactivity is calculated with analytical relationships derived from

$$T = [(\hbar \ln 2)/(2E_v)] \exp(K) \quad (2)$$

$$K = \frac{2}{\hbar} \int_{R_c}^{R_b} \{2\mu [(E(R) - E_{cor}) - Q]\}^{(1/2)} dR \quad (3)$$

where  $\mu = m A_e A_d/A$  is the reduced mass,  $m$  is the nucleon mass, and  $E(R)$  is the interaction energy of the two fragments separated by the distance  $R$  between centers. It is known that the fission barrier heights are too large within Myers-Swiatecki's variant of the liquid drop model.  $E_{cor}$  is a correction energy, similar

to the Strutinsky shell correction that allows getting a more realistic, lower and thinner barrier. It also takes into account the fact that nuclear inertia in the overlapping region is different from the reduced mass<sup>11)</sup>.  $R_\alpha$  and  $R_\beta$  are the turning points of the WKB integral,  $h$  is the Planck constant, and  $E_v$  is the zero-point vibration energy. For practical reasons (to reduce the number of fitting parameters) we took  $E_v = E_{cor}$  though it is evident that, owing to the exponential dependence, any small variation of  $E_{cor}$  induces a large change of  $T$ , and thus plays a more important role than the preexponential factor variation due to  $E_v$ .

The accuracy of the three masses in Eq. (1) plays a special role. Any erroneous increase of the  $Q$ -value is reflected in a lower value of  $K$ , producing a drastic decrease of the half-life, because of the exponential dependence shown in Eq. (2). Consequently we need mass values as close as possible to the true ones. This is the reason why we prefer using the measured masses (mass code  $C = 0$ ) or those determined from systematics ( $C = 1$ ) by Wapstra et al.<sup>12)</sup>. Otherwise we try the mass estimations<sup>9)</sup> one after the other, and then we compare the results. Following mass codes are conventionally adopted:  $C = 3$  for Jänecke and Masson estimations<sup>13)</sup>;  $C = 2$  for Masson and Jänecke<sup>14)</sup>;  $C = 4$  for Spanier and Johansson<sup>15)</sup>;  $C = 5$  for Tachibana et al.<sup>16)</sup>;  $C = 6$  for Satpathy and Nayak<sup>17)</sup>;  $C = 7$  for Comay et al.<sup>18)</sup>, and  $C = 8$  for Möller et al.<sup>19)</sup>.

In fact we made calculations for seven input mass tables. The first one possessing a central region around the line of beta-stability where  $C = 0$  or  $C = 1$ , is bordered on the neutron-rich and proton-rich sides by  $C = 3$  masses. In all other six input mass tables we shall only consider the regions beyond the core with »pure«  $C = 2, 4, 5, 6, 7, 8$ , respectively. From results of a large number of systematic calculations (25685 input masses times about 200 possible emitted clusters), we usually selected those which have  $T < 10^{30}$  s.

A small difference in the mass value of one, two or three partners obtained with different mass formulas, produces corresponding shifts in  $Q$ -values and induces a large variation of the lifetimes  $T$ . In some cases even the most probable emitted cluster may differ from table to table. In spite of these differences, one can draw some reliable conclusions concerning cluster emission rates.

The regions of  $\alpha$ -stable nuclei, according to different mass formulas, can be seen in Fig. 1. At the bottom we got a large area because not only  $C = 3$  nuclei beyond the Wapstra et al. table, but also  $C = 0$  or  $C = 1$  are contributing. The narrower surfaces presented by tables with mass codes  $C = 6$  and  $C = 8$  appear as a consequence of a smaller area of the corresponding input masses. In the proton-deficient region they are not extended, like the other, down to the neutron drip line. In a similar manner, nuclides with  $Z < 51$  are not included in mass table  $C = 4$ .

From the results of calculations performed in such a manner, we have selected the most optimistic ones, predicting  $T \leq 10^{30}$  s. In the same time we are looking for  $\alpha$ -stable nuclei, hence  $Q_\alpha < 0$ . Tables  $C = 4, 5$  and  $8$  gave no output fulfilling the selection criteria. Also, for  $C = 6$  we have obtained only one case, namely  $^{46}\text{S}$  radioactivity of  $^{179}\text{Dy}$  ( $C_d = 0$ ;  $C = C_e = C_{d\alpha} = 6$ ):  $Q_\alpha = -5.52$  MeV,  $E_k = 1.08$  MeV/nucleon,  $T = 10^{29.6}$  s, and the daughter is  $^{133}\text{Sn}$ . In the following we shall present the output coming from  $C = 2, C = 3$  and  $C = 7$ .

The most probable emitted clusters are neutron-rich nuclei. For only few of them ( $^{22}\text{O}$  and  $^{26}\text{Ne}$ ) the atomic masses have been measured ( $C = 0$ ). The others:

$^{46,48}\text{S}$ ;  $^{49}\text{Cl}$ ;  $^{50,52}\text{Ar}$ ;  $^{53}\text{K}$ ;  $^{56}\text{Ca}$ ;  $^{62}\text{Ti}$ ;  $^{74}\text{Cr}$ ;  $^{75}\text{Mn}$ ;  $^{74,76}\text{Fe}$ ;  $^{77}\text{Co}$  and  $^{78-82,84,86}\text{Ni}$  are so far away from the line of  $\beta$ -stability, that we had to use the estimated masses for all partners involved, except  $\alpha$ -particle:  $C_e = C = C_d = C_{d\alpha}$ .

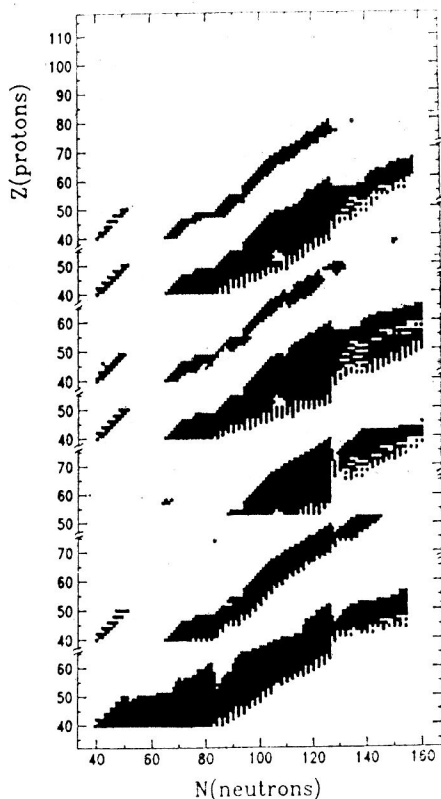


Fig. 1. Regions of  $\alpha$ -stable parent nuclei according to different mass tables, in the following order from top to bottom:  $C = 8, 7, 6, 5, 4, 2, 3$ . The contributions of  $C = 0$  and  $C = 1$  are included in the last plot.

The highest frequency of appearance of doubly magic  $^{78}\text{Ni}$  and of  $^{76}\text{Fe}_{50}$  as the most probable emitted cluster is certainly a consequence of shell and pairing effects acting around  $N_e = 50$  and  $Z_e = 28$  magic neutron and proton numbers of the light fragment, and in a less extent of the magicity  $Z_d = 50$  of the daughter number of protons.

There are two regions of cluster emitters beyond the area we are presenting here, where the double magicity of the daughter plays an important role:  $Z_d = 82$ ,  $N_d = 126$  for cluster radioactivities already experimentally confirmed, and  $Z_d = N_d = 50$  for a new island of proton rich-parents<sup>20)</sup> which could be produced by using radioactive beams.

The lightest clusters (O, Ne, S, Cl, Ar, K and Ca proton-deficient isotopes) are predicted to be emitted from  $Z = 57 - 70$  neutron-rich parents, by using the mass table with  $C = 2$ . Some of them are not present in Fig. 2, where we set the limits  $Z = 65 - 87$ ,  $N = 130 - 156$ , but they could be seen in Fig. 3 and Table 1. In the left-side of Fig. 2, there are only heavier emitted clusters from  $C = 2, 3, 7$ . The lighter ones, due to  $C = 6$  should be ignored from the present discussion, because they do not belong to  $\alpha$ -stable nuclei. We have included  $C = 6, 8$  owing to the unusual feature of very high branching ratio with respect to  $\alpha$  decay  $b = T_\alpha/T > 1$ . In fact this is a part of a smooth transition from  $b \ll 1$  to  $b > 1$  and finally to  $b \rightarrow \infty$ .

The sharp decrease of lifetime with increasing neutron number (Fig. 3) for emitted clusters lighter than  ${}^{76}\text{Fe}$ , is the corresponding increase of  $Q$ -value, a

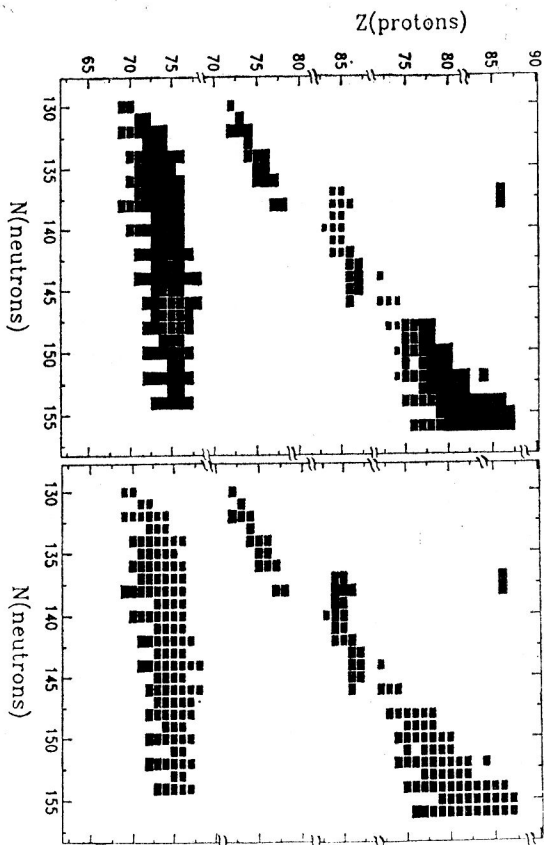


Fig. 2. Atomic numbers (left-side) of the most probable cluster emitted from parent nuclides with  $Z$  protons and  $N$  neutrons, and the corresponding half-lives (right-side) according to different mass tables, in the following order from top to bottom:  $C = 8, 7, 6, 2, 3$ . Only  $C = 7, 2, 3$  refer to  $\alpha$ -stable nuclei. For  $C = 8, 6$  branching ratios relative to  $\alpha$ -decay  $b > 1$  have been selected. Light and heavy points correspond to low  $Z_e$  and short  $\log T$  and high  $Z_e$  and long  $\log T$ , respectively. The highest  $Z_e$  is 28 and the longest  $T$  is  $10^{25}$  years.

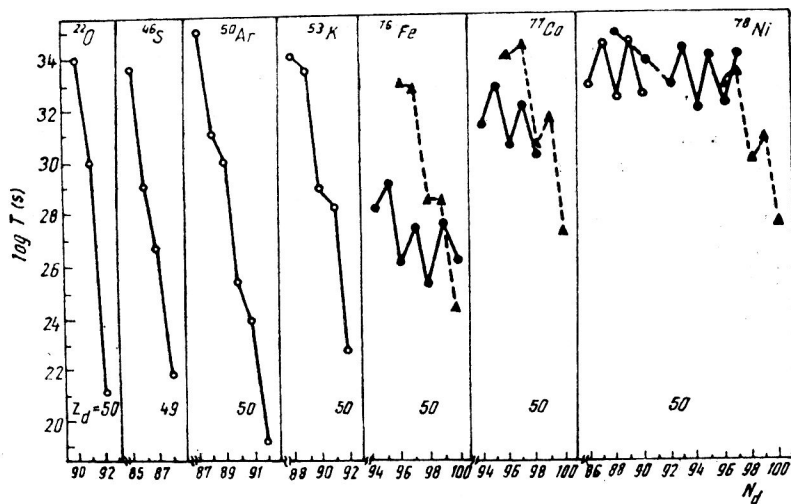


Fig. 3. Half-lives for some cluster radioactivities versus the neutron number of the daughter in the region of neutron-rich  $\alpha$ -stable emitters. Open points are obtained with  $C = 2$ , full points with  $C = 3$  and triangles with  $C = 7$ .

TABLE 1.

Z	A	$Q_\alpha$ MeV	$Z_e$	$A_e$	$Z_d$	$N_d$	$E_{\kappa}/A_e$ (MeV/nucleon)	log T(s)
58	163	-5.74	8	22	50	91	0.93	30.00
	164	-6.06	8	22	50	92	1.03	21.20
60	170	-7.55	10	26	50	94	1.12	25.20
62	176	-9.45	16	48	46	82	0.98	22.20
64	180	-9.08	16	46	48	86	1.08	23.00
66	184	-8.33	16	46	50	88	1.11	26.30
	186	-9.44	16	46	50	90	1.19	17.80
68	189	-8.10	18	50	50	89	1.13	29.90
	190	-9.09	18	50	50	90	1.16	25.40
	191	-9.93	18	50	50	91	1.19	23.90
	192	-11.06	18	50	50	92	1.22	19.20
70	194	-13.29	18	52	50	92	1.24	11.10
	196	-10.50	20	56	50	90	1.11	29.70

Cluster emission from  $\alpha$ -stable neutron-rich nuclides. Estimations based on  $C = C_d = C_{da} = 2$  mass tables. Also  $C_e = C$ , except  $^{22}\text{O}$  and  $^{26}\text{Ne}$ , for which  $C_e = 0$ .

trend which is reversed in comparison with what happens when clusters closer to the line of beta stability are emitted. We have, for example, a decreasing  $Q$  versus  $N_d$  for  $^{16}\text{O}$  emission, and an increasing one for  $^{26}\text{O}$  radioactivity. A smaller variation of half-life was obtained for heavier clusters ( $^{76}\text{Fe}$ ,  $^{77}\text{Co}$  and  $^{78}\text{Ni}$ ). The odd-even effect is also evident, leading to a staircase shape for light emitted clusters and large oscillations for heavier ones.

TABLE 2.

Z	A	Z <sub>e</sub>	A <sub>e</sub>	Z <sub>d</sub>	N <sub>d</sub>	C	Q <sub>α</sub> MeV	E <sub>β</sub> /A <sub>e</sub> MeV/n	log T(s)	C	Q <sub>α</sub> MeV	E <sub>β</sub> /A <sub>e</sub> MeV/n	log T(s)
72	204	28	78	44	82	2	-10.44	0.94	20.5	3	-3.09	0.91	25.2
	222	28	86	44	92	3	-12.74	0.90	5.6				
73	205	28	78	45	82	2	-8.77	0.93	27.10	3	-3.01	0.93	28.70
75	224	26	76	49	99	3	-11.20	1.03	25.50	7	-5.53	1.01	29.60
76	224	26	76	50	98	3	-10.03	1.04	25.50	7	-4.50	1.03	28.40
	225	26	76	50	99	3	-10.54	1.04	27.70	7	-4.93	1.04	28.50
	226	26	76	50	100	3	-10.99	1.04	26.30	7	-5.36	1.05	24.50

Cluster emission from α-stable neutron-rich nuclides. Estimations based on  
 $C = C_e = C_d = C_{da} = C_{da} = 2,3,7$  mass tables.

The differences from table to table are sometimes even of qualitative nature. For example the most probable emitted cluster from  $^{218}\text{Hf}$  should be  $^{84}\text{Ni}$  according to  $C = 3$ , and  $^{48}\text{Ar}$  from  $C = 7$ . Similar examples can be mentioned for other parents, like:  $^{219,221}\text{Ta}$ ,  $^{208,222,224,226}\text{W}$ ,  $^{211,225-227,229}\text{Re}$ , and  $^{228,230}\text{Os}$ . In Table 2 we present examples of qualitative agreement.

For heavier elements ( $Z = 77 - 86$ ), only  $C = 7$  led to results fulfilling the above mentioned criteria (see Fig. 2). Some of them are shown in Table 3. A comprehensive table is published elsewhere<sup>8)</sup>.

TABLE 3.

Z	A	$Q_\alpha$ MeV	$Z_e$	$A_e$	$Z_d$	$N_d$	$E_k/A_e$ (MeV/nucleon)	log T(s)	
78	226	-3.49	28	78	50	98	1.07	30.00	
	228	-4.29	28	78	50	100	1.09	27.70	
	229	-4.46	28	78	50	101	1.09	28.30	
	230	-5.00	28	78	50	102	1.10	25.20	
	231	-5.30	26	76	52	103	1.11	25.20	
	232	-5.66	26	76	52	104	1.13	21.40	
	234	-5.93	26	76	52	106	1.16	16.60	
	235	-6.59	26	76	52	107	1.17	16.80	
	80	232	-4.65	28	78	52	102	1.13	28.80
		233	-4.92	28	78	52	103	1.14	29.30
234		-5.11	28	78	52	104	1.15	26.20	
235		-5.21	28	78	52	105	1.16	26.10	
236		-5.13	26	76	54	106	1.17	22.60	
82	237	-5.79	26	76	54	107	1.18	22.70	
	236	-3.72	28	78	54	104	1.19	29.20	
	237	-3.81	28	78	54	105	1.20	29.20	
	238	-3.72	28	78	54	106	1.21	25.80	
84	239	-4.38	28	78	54	107	1.22	26.60	
	238	-0.80	28	78	56	104	1.24	29.50	
	239	-0.89	28	78	56	105	1.25	29.50	
	240	-0.73	28	78	56	106	1.27	26.00	
	241	-1.36	28	78	56	107	1.26	26.80	

Cluster emission from  $\alpha$ -stable neutron-rich nuclides. Estimations based on  $C = C_e = C_d = C_{da} = 7$  mass tables.

In conclusion, the half-lives of neutron-rich nuclei against cluster radioactivities are very sensitive to small variations of the atomic masses of the three partners involved (parent, daughter and emitted cluster). There is a need for an accurate mass formula producing estimations very close to the measured values. On the basis of a large number of systematic calculations, using different input mass tables to find the released energy, we believe that an island of cluster emission should exist in the neutron-deficient  $\alpha$ -stable region, where the neutron-rich neutron ( $N_e = 50$ ) and proton ( $Z_e = 28$ ) magic numbers of the emitted cluster, as well as (in a less extent) the proton magic number of the daughter ( $Z_d = 50$ ) are playing an important role.



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## METASTABILNOST $\alpha$ -STABILNIH JEZGRI BOGATIH NEUTRONIMA

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Originalni znanstveni rad

Sistematski je istraživana radioaktivnost atomskih jezgri s emisijom nukleonskih grozdova u području bogatom neutronima izvan linije beta stabilnosti, koristeći analitički supersimetrijski fizijski model. U određivanju oslobođene energije upotrebljavane su masene tablice različitih autora. Za jezgre bogate neutronima s atomskim brojevima između 57 i 86 procijenjena su vremena poluraspada na manje od  $10^{30}$  s. Emitirani grozdovi su također jezgre siromašne protonima, kao na primjer  $^{22}\text{O}$ ,  $^{46}\text{S}$ ,  $^{50}\text{Ar}$ ,  $^{76}\text{Fe}$ ,  $^{78}\text{Ni}$  itd.