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A calculation method to estimate partial half-lives for exotic radioactivities¹

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Abstract - Careful analysis of our previous semiempirical model for the cluster radioactivity of translead nuclei suggests that a simple function of some characteristics of the emitted cluster and the daughter nucleus can account for the trends observed in the half-lives of these exotic decay processes. The half-life $T_{1/2}$ is found to be expressed in terms of the atomic numbers of the product nuclei and the Q-value of the two-body disintegrating system as $\tau = \log T_{1/2}(s) = (aZ_{\rm C} + b)(Z_{\rm D}/Q)^{1/2} + (cZ_{\rm C} + d)$, by using a unique set of four parameters a, b, c and d, their values being determined from the fitting of this expression to the available data. About 85% of measurements are reproduced within one order of magnitude, and only in 5% of cases the calculated half-lives differ from the experimental ones by more than two orders of magnitude. It is also shown that, for some selected cases of cluster emission not yet measured, the method presented here anticipates results which are comparable to the ones obtained from systematic studies, making it a useful tool for fast estimation of half-life values of exotic radioactivities.

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Exotic decay, or cluster radioactivity, the decay process by which heavy nuclear fragments such as C, O, F, Ne, Mg, and Si isotopes are emitted from some translead parent nuclei, is a rare nuclear phenomenon well established since the middle eighties of the past century when the emission of ¹⁴C ions from ^{222–224,226}Ra isotopes [1–7] and ²⁴Ne from ²³⁰Th, ²³¹Pa, and ^{232–234}U isotopes [8–13] were detected in different laboratories. Such a rare spontaneous disintegration process was reported for the first time by de Carvalho *et al.* [14, 15] ten years before the first successful half-life measurements. These latter were accomplished after the first comprehensive, preliminary half-life predictions given by Săndulescu, Poenaru, and Greiner [16] of a number of nuclear clusters ranging from ¹⁴C up to ⁴⁶Ar that were expected to be emitted from heavy nuclei up to ²⁵²Cf.

Today, twenty-seven distinct cases of heavy-ion emission from twenty different translead parent nuclei are known from a collection of fifty-nine measured half-life values of these decays [17–19]. In theoretical and/or semiempirical descriptions of such an exotic decay process researchers have agreed and recognized that cluster radioactivity is a natural manifestation of the nuclear shell structure present in either the daughter nucleus produced or cluster emitted, or both, but mainly governed by the structure of 126 neutron shell closure of the daughter nucleus, *i.e.*, the doubly magic ²⁰⁸Pb or its neighbours.

Such studies have led to semiempirical formulae which in general reproduce successfully the decay data, and are also used to give half-life estimates (within 1–2 orders of magnitude) for decay cases not yet experimentally observed. The ASAFM —analytical superasymmetric fission model, for instance, is a widely used calculation method which has been quite recently extended to exotic decays that are expected to take place in the region of superheavy nuclei [20].

In the present note we report on an alternative calculation routine which resulted from our previous studies to systematize the half-life data of known cluster decays [17, 21]. These calculation models rely entirely on a quantum mechanical, tunneling mechanism through a potential barrier where, besides the basic Coulomb contribution to the total height of the barrier, both the centrifugal (when angular momentum, ℓ , differs from zero) and overlapping effects have been also considered to half-life evaluations. In doing so, we realized that for all these decay cases the centrifugal contribution to the potential barrier could be safely neglected in view of the simultaneous small values of angular momentum ($\ell < 5$ in all cases) and large values of half linear momentum squared, $\mu_0 Q$, where μ_0 and Q are the reduced mass and total available energy, respectively, of the two-body disintegration system. In this way, if one expresses lengths in fm, masses in u, energies in MeV, and time in second, the partial half-life, $T_{1/2}$, for a particular mode of decay by cluster emission can be evaluated from a compact expression as

$$\tau = \log T_{1/2}(\mathbf{s}) = \tau_0 + \tau_1 + \tau_2 \,, \tag{1}$$

in which

$$\tau_0 = -22 + \log a + \frac{1}{2} \log \left(\mu_0 / Q \right) \tag{2}$$

is the term related to the frequency of assaults on the barrier,

$$\tau_1 = 0.19 \, w \, g \sqrt{\mu_0 Q} \cdot F(u) \tag{3}$$

is the contribution from the overlapping barrier region, and

$$\tau_2 = 0.27358 Z_{\mathsf{C}} Z_{\mathsf{D}} \sqrt{\frac{\mu_0}{Q}} \cdot G(u) \tag{4}$$

is the one corresponding to the external, separation barrier region. The functions

$$F(u) = \sqrt{\frac{1}{u} - 1} \quad \text{and} \quad G(u) = \arccos\sqrt{u} - \sqrt{u(1 - u)} \tag{5}$$

contain the dependence upon the ratio $u = Q/V_c$, where $V_c = Z_C Z_D e^2/c$ is the potential energy in the configuration at contact of the preformed fragments ($e^2 = 1.4399652$ MeV·fm is the square of the electronic elementary charge, and Z_c and Z_D are the atomic numbers of the emitted cluster and daughter nucleus, respectively). The lengths $a = R_P - R_C$, $c = R_D + R_C$, and $w = c - a = 2R_C - (R_P - R_D)$ represent the inner turning point, the distance between the centres of the fragments at contact, and the width of the overlapping region, respectively (here, R_P , R_C and R_D denote the nuclear radii of the parent nucleus, cluster emitted, and daughter nucleus, respectively). Finally, g is the only adjustable parameter of the model, the value of which being thus determined from a set of half-life measurements. The g-value depends upon the source of data adopted for both nuclear mass and radius as well as the set of measured half-life values for the cluster decays under consideration. The g-value is related through equation (3) to the cluster preformation probability (for details see [21]).

Now, both the functions F(u) and G(u) decrease monotonically towards zero as u increases to 1. In addition, the quantities $\log a$, c, and w do not vary significantly in cases for which the same cluster is being considered to be emitted. Therefore, from inspection on equations (1)–(5) it can be easily shown that τ will decrease with the increasing of u together with the decreasing of the quantity $\mu_0 Z_{\rm C} Z_{\rm D}$. Hence, for the same emitted cluster from a set of parent nuclei, τ should be expected to vary strongly upon the quantity $Z_{\rm D}(\mu_0/Q)^{1/2}$. On

the other hand, the reduced mass $\mu_0 = A_{\rm C}(1 - A_{\rm C}/A_{\rm P})$ for the most probable cluster emission cases, *i.e.* those cases for which the daughter nucleus is the double magic ²⁰⁸Pb or its neighbours, is shown to depend only upon the cluster mass number, $A_{\rm C}$, and scales as $\sim A_{\rm C}^{0.9}$, so that a choice for the variable ξ to describe the half-life trends seems to be $\xi = Z_{\rm D} Q^{-1/2}$. Indeed, if one plots τ against $Z_{\rm D} Q^{-1/2}$ a number of straight lines emerges, each one for the same cluster, when different clusters from C up to Si are considered to be emitted from translead and transuranium parent nuclei.

Afterwards, we have found that the values of $\tau = \log T_{1/2}^{\exp}$ could much better be described by using the quantity $\xi = (Z_D/Q)^{1/2}$. This is illustrated in figure 1 where the data have been fitted by the straight lines



$$\tau = (aZ_{\mathsf{C}} + b)(Z_{\mathsf{D}}/Q)^{1/2} + (cZ_{\mathsf{C}} + d).$$
(6)

Figure 1: The logarithm of the cluster decay half-life $\tau = \log T_{1/2}(s)$ plotted against the quantity $\xi = (Z_D/Q)^{1/2}$ for all 59 half-life values measured to date (circles, grouped by clusters of the same element). The straight lines result from the present calculation method [equations (6) and (7)]. The open circles correspond to the three measurements that are not reproduced by the present method (see text). The inset graph shows, for the fifty-six measurements, the distribution of the difference $\Delta \tau$ between calculated and experimental half-life values.

The values of parameters a, b, c and d, determined by using the current version of the LSM program recently developed by Mamczur [24], are

$$a = 12.8717, \quad b = -5.1222, \quad c = -4.6496, \quad d = -73.3326.$$
 (7)

Differences between experimental and calculated half-life values following formula (6) and parameter values (7) result smaller than two orders of magnitude in practically 95% of cases, and within one order of magnitude in 85% of cases. The distribution of the quantity $\Delta \tau = \log(T_{1/2}^{c}/T_{1/2}^{e})$ is shown in the inset of fig. 1. Only for two cluster emission cases, namely, ²³¹Pa \rightarrow ²³F and ²³⁰U \rightarrow ²²Ne it resulted $|\Delta \tau| > 2$. However, this faint agreement should not be thought as so surprising. The first half-life result was obtained based on only one event recorded [25], and also it is the only exotic decay case of an odd-Z cluster known to date. As for ²²Ne emission from ²³⁰U only two events have been attributed to ²²Ne ions in the experiment reported in [26], but without comparison with accelerator calibrations. In addition, as discussed by Bonetti *et al.* [27], ²²Ne exhibits some degree of deformation, and such a particular characteristic may lead to a poor agreement for this case. The $\Delta \tau$ distribution for the fifty-six measurements (shaded histogram in fig. 1) is seen to be normally centered at $\Delta \tau = 0$ with standard deviation $\sigma = 0.713$.

The four-parameter formula (6) has been applied to estimate half-lives of yet unmeasured cluster emission cases. Results are shown in Table 1 (last column), and they can be compared to previously calculated values from systematic studies as indicated in columns 4–6. The differences between each other calculated τ -values for cases listed in Table 1 are found centered at $\overline{\Delta \tau} \approx 0.21$ with standard deviation $\sigma = 1.31$. Altogether, this means that evaluation of half-life values for cluster radioactivities can be obtained within an average uncertainty of two orders of magnitude.

In summary, in this work we have reported on an alternative method for calculating the exotic decay half-life of translead parent nuclei. The method uses a simple function of the following three characteristics of the nuclei involved in the decay process: the atomic numbers of both the emitted cluster and the daughter nucleus, and the *Q*-value of the twobody disintegrating system. With a unique set of four parameters, it has been possible to reproduce reasonably well the majority of the available experimental half-life data. Besides that, for some selected cases of cluster emission not yet measured, the calculation routine here presented, despite its simplicity, anticipates results which are comparable to the ones obtained from systematic studies. This makes it a useful tool for fast estimation of half-life values of cluster radioactivities.

		Q-value ^a	Calculated half-life, $\tau = \log T_{1/2}(s)$			
No.	Decay case	(MeV)	Ref. [17]	Ref. [21]	Ref. [22]	This work
1	$^{222}\mathrm{Fr} \rightarrow {}^{14}\mathrm{C} + {}^{208}\mathrm{Tl}$	30.187	16.76		18.2	16.89
2	$^{223}\mathrm{Fr} \rightarrow {}^{14}\mathrm{C} + {}^{209}\mathrm{Tl}$	29.110	18.79		19.0	19.05
3	225 Ra \rightarrow 14 C + 211 Pb	29.576	19.06		20.0	18.84
4	$^{224}\mathrm{Ac} \rightarrow ^{15}\mathrm{N} + ^{209}\mathrm{Pb}$	37.877	17.30		18.7	19.16
5	$^{227}\mathrm{Ac} \rightarrow {}^{14}\mathrm{C} + {}^{213}\mathrm{Bi}$	28.174	23.17		23.1	22.53
6	$^{224}\text{Th} \rightarrow {}^{14}\text{C} + {}^{210}\text{Pb}$	33.043	13.75		13.1	12.36
7	$^{226}\mathrm{Th} \rightarrow {}^{18}\mathrm{O} + {}^{208}\mathrm{Pb}$	45.876	18.51		18.0	20.29
8	$^{227}\mathrm{Th} \rightarrow {}^{18}\mathrm{O} + {}^{209}\mathrm{Pb}$	44.351	21.29		22.6	22.52
9	$^{229}\mathrm{Th} \rightarrow ^{20}\mathrm{O} + ^{209}\mathrm{Pb}$	43.552	24.31		26.1	23.74
10	$^{232}\mathrm{Th} \rightarrow {}^{26}\mathrm{Ne} + {}^{206}\mathrm{Hg}$	56.146	29.24		29.4 ^b	27.70
11	$^{225}\text{Pa} \rightarrow ^{15}\text{N} + ^{210}\text{Po}$	40.326	14.88		14.8	16.77
12	$^{233}\mathrm{U} \rightarrow {}^{28}\mathrm{Mg} + {}^{205}\mathrm{Hg}$	74.446	25.54		27.4	25.68
13	$^{235}\mathrm{U} \rightarrow {}^{28}\mathrm{Mg} + {}^{207}\mathrm{Hg}$	72.380	28.15		27.3 ^b	27.87
14	$^{235}\mathrm{U} \rightarrow ^{29}\mathrm{Mg} + ^{206}\mathrm{Hg}$	72.706	28.45		27.4^{b}	27.52
15	$^{238}\mathrm{U} \rightarrow {}^{34}\mathrm{Si} + {}^{204}\mathrm{Pt}$	86.062	30.22	30.02	28.0^{b}	28.25
16	$^{235}\mathrm{Np} \rightarrow ^{28}\mathrm{Mg} + ^{207}\mathrm{Tl}$	77.322	23.07		24.0	23.72
17	$^{236}\mathrm{Np} \rightarrow ^{28}\mathrm{Mg} + ^{208}\mathrm{Tl}$	75.373	25.50		28.1	25.68
18	$^{236}\mathrm{Np} \rightarrow ^{32}\mathrm{Si} + ^{204}\mathrm{Au}$	88.467		27.58	26.9 ^b	27.02
19	$^{237}\mathrm{Np} \rightarrow ^{30}\mathrm{Mg} + ^{207}\mathrm{Tl}$	75.043	27.15		28.3	26.02
20	$^{237}\mathrm{Np} \rightarrow ^{32}\mathrm{Si} + ^{205}\mathrm{Au}$	87.960		28.16	30.0	27.50
21	$^{239}\mathrm{Pu} \rightarrow {}^{34}\mathrm{Si} + {}^{205}\mathrm{Hg}$	91.095	27.16	26.84	29.0	25.65
22	$^{240}\mathrm{Pu} \rightarrow {}^{34}\mathrm{Si} + {}^{206}\mathrm{Hg}$	91.291	26.83	26.52	27.4	25.47
23	$^{241}\mathrm{Am} \rightarrow {}^{32}\mathrm{Si} + {}^{209}\mathrm{Tl}$	90.920		27.50	28.9	26.83
24	$^{241}\mathrm{Am} \rightarrow {}^{34}\mathrm{Si} + {}^{207}\mathrm{Tl}$	94.192	25.01	24.53	25.8	23.93
25	$^{243}\text{Am} \rightarrow {}^{34}\text{Si} + {}^{209}\text{Tl}$	91.037		28.34	29.9	26.72
26	$^{240}\text{Cm} \rightarrow {}^{32}\text{Si} + {}^{208}\text{Pb}$	97.825	21.48	20.96	21.2	21.87
27	$^{240}\mathrm{Cm} \rightarrow {}^{34}\mathrm{Si} + {}^{206}\mathrm{Pb}$	95.738		24.40	25.1	23.61
28	$^{242}\text{Cm} \rightarrow ^{32}\text{Si} + ^{210}\text{Pb}$	93.885		25.42	26.0	25.20

Table 1 - Intercomparison between different partial half-life predictions and the presentones for cases of nuclear cluster decay.

^a Screening effects included (for details see Ref. [17]).

 $^{\mathsf{b}}$ Quoted in Ref. [23].

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