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Published in:
Physics Letters. Section B: Nuclear, Elementary Particle and High-Energy Physics

1972

Reprinted from

PHYSICS LETTERS

Volume 40B, No. 5, 7 August 1972

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pp. 517–521

NORTH-HOLLAND PUBLISHING COMPANY

AMSTERDAM

ON THE TERMINATION OF THE R-PROCESS AND THE SYNTHESIS OF SUPERHEAVY ELEMENTS

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Received 14 June 1972

Predictions for spontaneous and neutron-induced fission, alpha and beta decay have been made on the basis of potential-energy surfaces for $Z \geqslant 90$ nuclei on the neutron-rich side of the beta stability line. In spite of their considerable quantitive uncertainties, these predictions appear to negate the possibility of superheavy element synthesis via a one- or two-step r-process from super novae. In addition artificial superheavy element synthesis by two consecutive exploding nuclear devices appears improbable from these results. Neutron induced fission seems to be even more decisive than spontaneous fission for the early termination of the r-process.

Nuclei heavier than ^{209}Bi and in particular the actinide elements are considered to be synthesized in stars by the so-called rapid neutron capture process (r-process), as suggested in a 1957 paper by Burbridge, Burbridge, Fowler and Hoyle [1]. In a subsequent paper Fowler and coworkers [2] concluded that probably the neutron capture process along the r-process path towards increasingly heavier elements was broken off by the competing process of fission at approximately $Z \approx 93, A \approx 275$. This conclusion was based on the use of a liquid-drop formula [3] for the estimate of masses and fission half-lives along the r-path. More recently Fowler and Schramm [4] reversed this conclusion and conjectured that the path for the r-process could continue beyond $N = 184$ and that eventually the superheavy island of stability could be populated by beta-decay from elements on the r-path. The reversal of the earlier prediction was based on the fact that effects of nuclear shell structure – empirical shell energies from the actinide region and theoretically predicted ones in the $Z = 114, N = 184$ region (as by ref. [5]) – were in a gross way included in the fission half-life estimates. This was achieved solely by the use of an empirical fission half-life formula containing a shell-structure dependent fissility parameter x.

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The use of such a semiempirical formula for fission half-lives was found to work satisfactorily in the actinide region. On the other hand one may argue that the actinide region is a rather homogeneous one, all elements exhibiting similar ground state deformations and all barriers being of similar two-peak type.

Among the relevant r-process elements, on the other hand, there is reason to presume a greater variety in ground state shapes and barrier characters. We have therefore attempted to estimate fission, alpha and beta half-lives on the basis of a nuclear potential-energy surface calculated from the modified-harmonic-oscillator potential. We have thus employed the potential of ref. [6],

$$V = V_{\text{osc}} + V'$$

where $V_{\text{osc}} = \frac{1}{2}\hbar\omega\rho^2(1 - \frac{2}{3}\epsilon P_2 + 2\epsilon_4 P_4)$
and $V' = -\mu'\hbar\omega_0 [l_t^2 - \langle l_t^2 \rangle] + \kappa' \boldsymbol{s} \cdot \nabla V_{\text{osc}} \times \boldsymbol{p}$.

In a few test cases we have also included terms proportional to $\epsilon_3 P_3$ and $\epsilon_5 P_5$. The A-dependent κ' and μ' values are chosen in accordance with the conclusion of ref. [6]. In some of the cases the $\boldsymbol{s} \cdot \nabla V \times \boldsymbol{p}$ term was replaced with the one more commonly used $\boldsymbol{l}_t \cdot s$ term.

The potential-energy surface is then obtained as a function of ϵ and ϵ_4 by the use of the Strutinsky shell correction method [7]. The energy minima give the ground state masses from which alpha and beta Q-values are readily obtained.

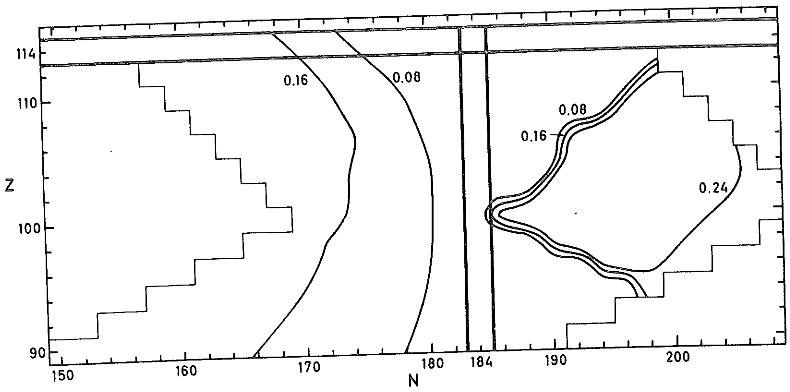

Fig. 1. Contour plot of the absolute values of the equilibrium quadrupole deformations in the (*N*, *Z*) plane.

The ground state quadrupole deformations are given as function of *N* and *Z* in fig. 1.

On the basis of available theoretical ground state masses alpha half-lives are readily determined with satisfactory accuracy. In addition alpha decay for most of these neutron-rich elements is hardly a competitive process. The crucial processes are spontaneous and neutron-induced fission. We shall discuss first spontaneous fission.

In considering barrier penetration we have limited ourselves to the one-dimensional penetration problem and assumed the WKB formula to hold for the projected barrier. Thus ϵ_4 is only employed to find a minimal static potential barrier.

Usually the barrier is two- or three-peaked. A contour plot of the height of the highest barrier is shown in fig. 2. Sometimes the outer minimum at $\epsilon \approx 0.4-0.7$ is lower than the inner one at $\epsilon \approx 0-0.03$. We have then assumed the ground state to be the one shielded by the least penetrable barrier. For cases when the outer minimum is lower than the inner one, but according to the above criterium the inner one still corresponds to the ground state, we associate with the latter a half-life corresponding only to the penetration of the first barrier. The rationale for this is that in that case the nucleus has a large probability to be

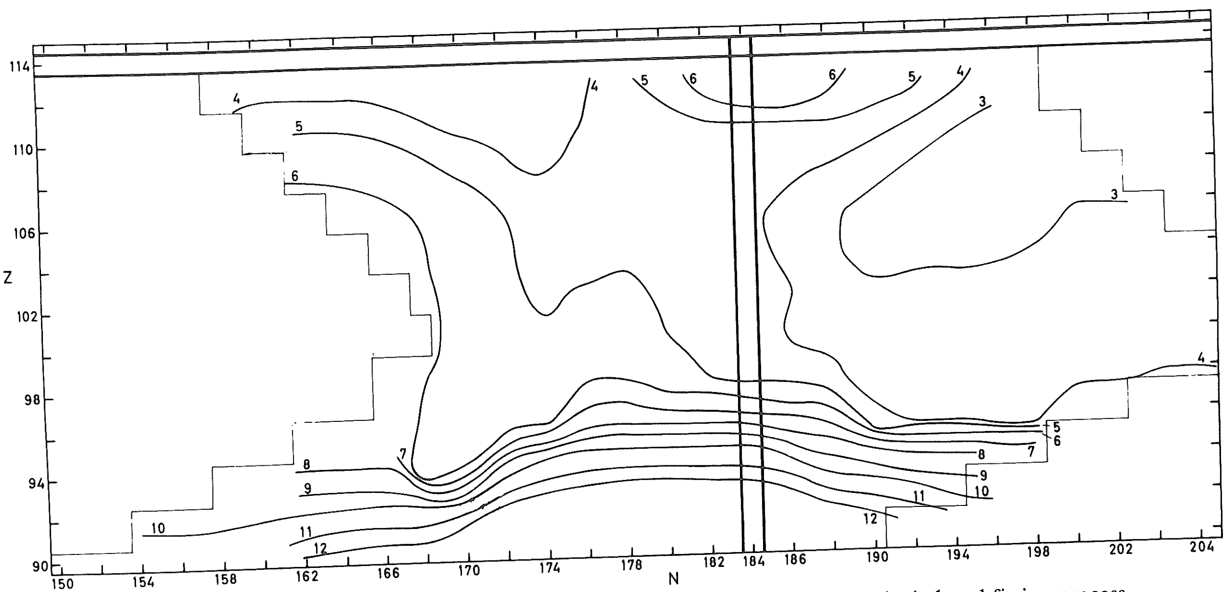

Fig. 2. Contour plot in the (*N*, *Z*) plane of the height of the barrier dominant in the induced fission process.

captured in the outer minimum state, from which it subsequently decays in an independent fashion.

Furthermore, the mass parameter is assumed to be independent of ϵ and to have the semiempirical value given by Moretto and Swiatecki [8],

$$B/\hbar^2 = 0.054\,A^{5/3}\ \mathrm{MeV}\,.$$

The spontaneous fission half-lives obtained on the basis of these model calculations are exhibited by the solid lines of fig. 3. This figure is highly reminiscent of one produced by Ford, Seeger and Nix on the basis of the modified harmonic oscillator model and reported by Cowan in 1969 [9]. The "bay" of short half-lives extending from the upper right corner of the picture corresponds to a region of nuclei where the outer minimum is lower than the inner one.

To predict the β^--decay half-lives we applied the gross theory developed by Yamada, Takahashi and Koyama [10] and recently extended by Takahashi [11] to include the first forbidden transitions. In this theory the strength function, defined as an appropriate average of the squared nuclear matrix element times the final level density, is assumed to be composed of the single-particle strength functions which are determined with the aid of the sum-rule considerations. The method of summing up these single-particle contributions is given in the paper of Koyama et al. [10]. Two forms of the single-particle strength functions were employed: Gaussian and modified Lorentz forms with the representative values of σ_N being 5 MeV and 12 MeV respectively. These values seem to be reasonable, judging from the analysis of the beta decay strength functions and half-lives (see e.g. refs. [11, 12]).

Generally, the results of the gross theory are expected to become more reliable and more stable as the Q-value increases. In particular, the fine fluctuations from the gross structure that are usually pronounced in the low-lying daughter states will be relatively small in the case of the even-even parent because of the large final level density.

It appears from fig. 3 that the r-process, if it proceeds by a path as that suggested by Fowler and

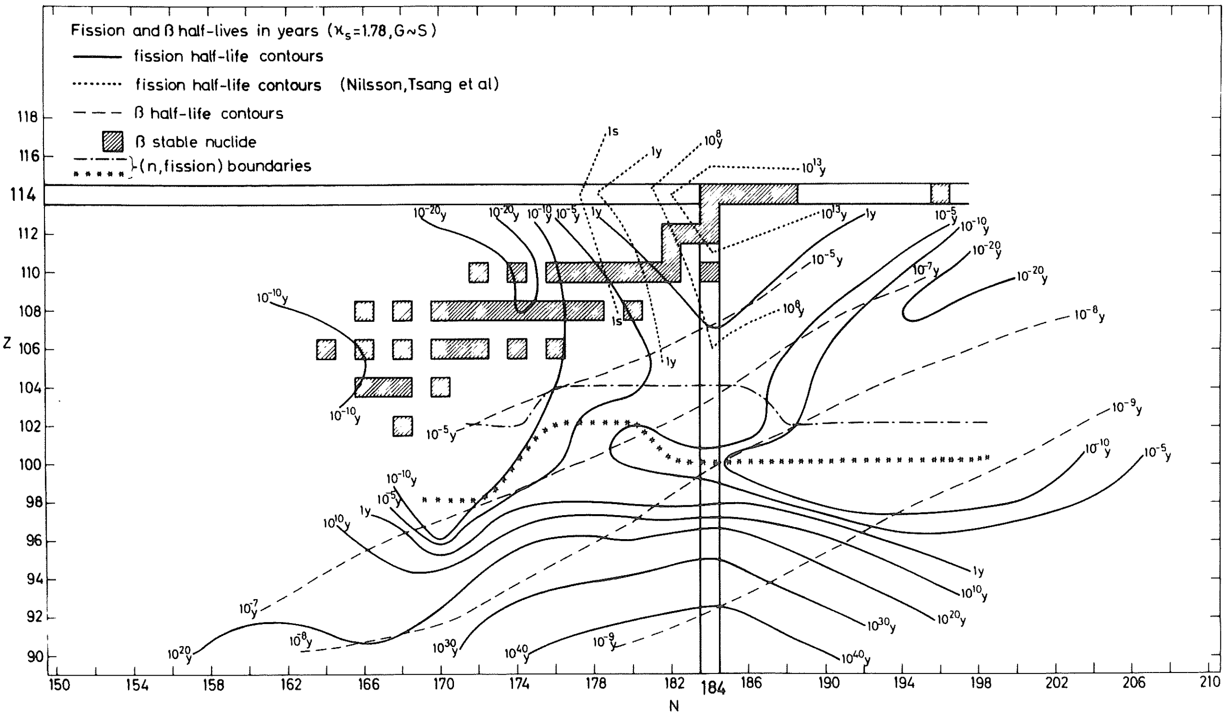

Fig. 3. Fission and beta-decay half-life contours and (n, fission) boundaries for $\kappa_s = 1.78$, $G \sim$ surface area. See the text for explanation.

Schramm [3], in terms of this particular model is terminated at approximately $Z = 98, N = 186$ even for the weak surface symmetry coefficient suggested by Myers and Swiatecki in 1967 [13]. Below this point there could occur some beta-decay chains particularly from neutron-rich elements with $N = 184$ for which the (n, γ) cross section is low. These consecutive beta-decays proceed to, say, $Z = 100, N = 178$. Beyond this the beta-decay products reach the channel of instability between the normal actinides and the superheavy region, unless a possible new burst of neutrons causes a resumption of the r-process from about $Z = 100, N = 178$. This is a situation that is reminiscent of the suggestion put forth by Meldner [14] of employing neutron showers produced by two-consecutive exploding nuclear devices. However, even this two-step process should be terminated near $Z =$ 102, 104 by neutron-induced fission processes as marked by a star-line and a dot-dash line in fig. 3. These lines correspond to a pessimistic and an optimistic estimate of the energy for which the neutron-induced fission balances gamma decay back to the ground state after neutron capture. The two lines correspond to $W - S_n = 0.8$ MeV and 0 MeV respectively, where W is the height of the highest barrier and S_n the neutron separation energy. The kinetic energy of the neutrons is usually assumed to be of the order of 0.2 MeV in the r-process.

The quantitative results of this investigation are highly tentative and may be modified by serveral factors. The influence of one of these factors on the spontaneous fission half-lives, from an alternative choice of the surface symmetry coefficient, is shown in fig. 4. In this alternative we have used $\kappa_s = 2.53$ (with the other parameters of the MS set unchanged) and assumed that the pairing matrix elements are independent of distortion instead of proportional to the surface area. These two sets of assumptions lead to similar barriers in the stable actinide region. For the neutron deficient region the second set leads to reduced barriers and spontaneous fission half-lives (see fig. 4) which blocks the access to the SHE region at an even earlier point.

Since we do not have a consistent total set of liquid-drop parameters in the second case we have not calculated neutron separation energies in the latter case.

The asymmetric $P_3 + P_5$ degrees of freedom are additional unfavourable factors. Calculations with these degrees of freedom included have been performed for some neutron-rich Th, U and Pu nuclei resulting in

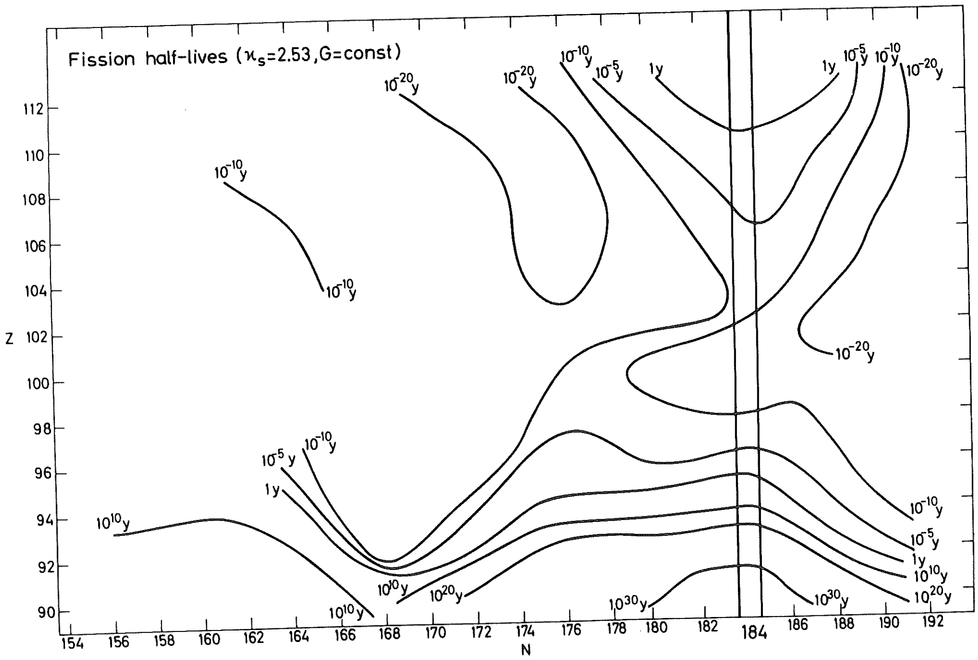

Fig. 4. Fission half-life contours for $\kappa_s = 2.53$, G = constant.

spontaneous fission half-life reduction factors of 10 to 10^5.

These additional blocking factors may be more than balanced by a possible underestimate of the shell effects connected with $N = 184$. Thus in the single-particle potential models employed by Nix and Strutinsky and coworkers with s.p.-levels fitted to the Pb region rather than to the observed single-particle levels of the deformed regions and also another distortion dependence of the spin-orbit term, the $N = 184$ gap comes out considerably larger. Thus, as Fiset and Nix [15] have pointed out, according to the calculations of their group the SHE island has a longer north-south extension than it has on the basis of the modified harmonic-oscillator model. The same conclusion is arrived at by Brack et al. [16] and by Lukasiak et al. [17].

Such shell effects, if more correctly predicted by these groups than by us, probably make it possible to come closer to the SHE island of stability by either the one- or the two-step r-process, but it would appear that all of the calculations [15–17] based on a study of the potential-energy surface suggest that the SHE island is unreachable by any r-process. The decisive factor blocking the path is then the neutron induced-fission process even more than the spontaneous-fission process.

We are grateful for helpful communications from J.R. Nix and early access to his results. Conversations with W.A. Fowler are also acknowledged. One of us (R.B.) is grateful to the Swedish International Development Authority (SIDA) for financial support, and another (R.K.S.) for the award of a Nordita Professorship and the hospitality of the Department of Mathematical Physics of the Lund Institute of Technology and the Niels Bohr Institute.

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