Superheavy Element Synthesis*

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We propose to overcome the present difficulties of superheavy-element production by utilizing neutron capture at time constants between those associated with "prompt" and "rapid" processes, i.e., by employing suitably controlled thermonuclear explosives.

Despite all efforts, the production or detection of superheavy elements has so far proved impossible.[1] Recent results of conservative step-by-step approaches with heavy-ion accelerators are, however, cause for optimism; e.g., odd-A Rf and odd-odd Ha isotopes have unexpectedly long overall half-lives. One can therefore expect a marked increase in stabilization at odd nucleon numbers for the superheavy elements. This corroborates our prediction[2] that it will be an *odd-odd* superheavy nucleus—such as $^{294}111$, $^{296}113$—which is most stable (provided that its β decay does not dominate strong decays).

Theoretical work has now converged to a point where practically all approaches agree in the prediction of a shell closure at $Z=114$, which was first suggested over six years ago.[3] Experimental *non*confirmation of 114 would establish completely unexpected effects, i.e., strong nonlinearities absent in our present shell-structure theory.

Standard heavy-element production methods involving heavy-ion fusion suffer from two basic difficulties in all possible compound nuclei produced, viz., (1) strong neutron deficiency, and (2) high excitation energy. Attempts to overcome the first of these by accelerating neutron-rich fission fragments have been done at low flux with Cf fragments.[4] Two-step fission-fusion processes might also be realized with nuclear reactions at energies above several GeV. Some of the neutron-rich "chips" produced in proton or heavy-ion induced reactions at very high energies will be in the appropriate range of charge/mass and kinetic energy for fusions with other nuclei in a (thick) target, thereby forming superheavy compound systems.[5]

It is, of course, quite possible that no compound nuclei can be formed with sufficiently *low excitation* by any of the above methods. This difficulty is crucial. It is only a shell closure that can create a superheavy element "island" of relative stability. Gross structure stabilizations are negligible in this area compared to shell effects—the latter being at best of order 10 MeV in nuclear (spectral) gaps or fission barriers. According to recent estimates,[6] any compound nucleus—formed in reactions similar to Ge + Th —will have an excitation energy of the order of 30 to 80 MeV. Empirical evidence unfortunately points to at least 60 MeV[7]—wiping out possible shell-structure stabilization and optimism for producing superheavy elements this way.

The above difficulties could be avoided by prompt-to-rapid[8] capture in a "laboratory super r process" simulating to some extent the conditions of the transuranium or transeinsteinium part of the astrophysical r process[9] (and thus producing similar capture/decay paths). Such experiments certainly pose formidable technological problems; we have no comment on this part of the method except to mention that some experts give it a reasonable chance.[10]

The major nuclear theoretical question is whether capture chains of useful length can be realized at high Z. In all previous experiments, the terminal point proved to be ^{257}Fm. High neutron flux can dramatically improve the yield, as was shown with the Hutch device,[11] but does not lead to charges higher than those produced in low-flux exposures or even reactors. The main hurdle is the increase of the strong decays along capture chains, especially the instability with respect to neutron-induced fission. This trend is suggested by conventional mass formulas with surface symmetry terms.[12] Fission barriers decrease more rapidly than neutron separation (threshold) energies with increasing neutron excess. The region useful for rapid-to-prompt capture/decay production paths is thus a band moved to the neutron-rich side of the β-stability line by normally more than 10 neutron numbers with a width of 10 to 20.[13] The most detailed investigations that have so far been published about this apply to the astrophysical r process.[9,14]

For the type of experiment we propose here —*capture of neutrons from thermonuclear explosions controlled to allow some intermediate β decays*—the maximum neutron excess limit is practically the same as in the r process. (It is not exactly the same due to different average neutron temperatures and fluxes in prompt and rapid capture.) The total bandwidth could unfortunately be much less than 20 neutrons since the minimum neutron excess limit has to be moved up to a point where β decay is not much slower than the longest achievable total exposure time or the longest time lapse between (hot) multiple exposures.

The real capture-chain cutoffs, of course, have shell structure[15] and are much sharper than expected from average trends due to strong odd-even effects in the high-Z region: For example, the overall half-life of ^{258}Fm is ten orders of magnitude shorter than that of its odd-A neighbor, ^{257}Fm. Similarly increased odd-even effects are exhibited in the half-lives of the heaviest elements produced so far, e.g., in Rf and Ha isotopes, as mentioned earlier. Thus, even-even (ee) isotopes constitute a major set of hurdles in the region of interest: Not only is β decay slowest, but (strong) nuclear decay is enhanced by many orders of magnitude. This causes neutron capture-decay chains to be strongly depleted or even terminated when passing through high-Z, ee nuclei. A sizable part of the staggering observed in prompt-capture yield curves might therefore be due to odd-even effects in the fission competition, as was suggested some time ago by Dorn and Hoff.[16] Fortunately, we can construct many capture/decay paths that circumvent ee nuclei, as indicated in Fig. 1: Favored are all cases where even numbers of neutrons are captured at odd Z alternating with even numbers of intermediate β decays.

Present capture data indicate an additional helpful nonmonotonic behavior with increasing neutron excess, namely, the existence of unlinked islands of relative stability in transuranium isotopes. As was pointed out in Eccles's review,[11] the use of targets of Np, Pu, and Am showed that each

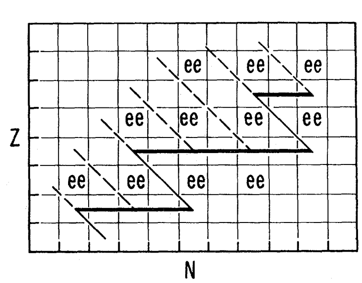

FIG. 1. Capture/decay paths circumventing even-even nuclei.

of these $Z > 92$ capture chains suffered from fission to a significant degree. On the other hand, the production of elements up to ^{257}Fm from U targets after prompt capture—through a baryon number conserving reaction—must pass through isotopes which all have strong decay half-lives that are sufficiently long to allow β-decay competition.

These findings imply that, *everywhere* along capture chains with $92 \leq Z \leq 100$ and $A < 258$, the spontaneous fission lifetimes are long. For sufficiently large neutron excess, additional neutron capture does not lead to high excitation so that (low-energy) neutron-induced fission should not be much faster than spontaneous fission (at $N+1$) at least for even N isotopes. This suggests the existence of several unlinked or "island" Am and Pu isotopes of relative stability disconnected from the Am/Pu chains by ($A \gtrsim 247$) isotopes with strong decay half-lives which are too short for β-decay competition. Similar areas of stability were introduced in calculations of r-process superheavy-element production.[9,17]

On the basis of the above, we suggest the following two stages for prompt-to-rapid-capture superheavy-element synthesis:

First, a Hutch-type experiment arranged to allow quick access to the milligram quantities produced by Es and Fm isotopes. Capture in U targets is well understood[11] and, in fact, singularly effective in yielding *macroscopic* quantities of Es/Fm. In all previous experiments, over 90% target material losses were due to the initial fast (14-MeV) neutron-induced fission. Additional yield improvements might therefore be possible by the use of effective fast-flux shielding or moderation.

The second stage would be the exposure of Es/Fm targets to thermonuclear fluxes in an arrangement that allows fast intermediate β decays; for example, by catching a part of the plasma emerging from an initial shot at the site of a subsequent explosion ("sequential exposure") or by extending the duration of just one low-energy, 20-keV equivalent, neutron flux exposure, or by combining these methods.

As indicated schematically in Fig. 2, a typical double shot, with time lapse $> 10^{-2}$ sec, should allow about four intermediate β decays, thus yielding neutron-rich Lr and Rf isotopes which, upon capture of about fourteen neutrons, can decay to $Z \sim 111$. Sequential exposures introduce large losses. However, useful yields are still possible because of the macroscopic nature of

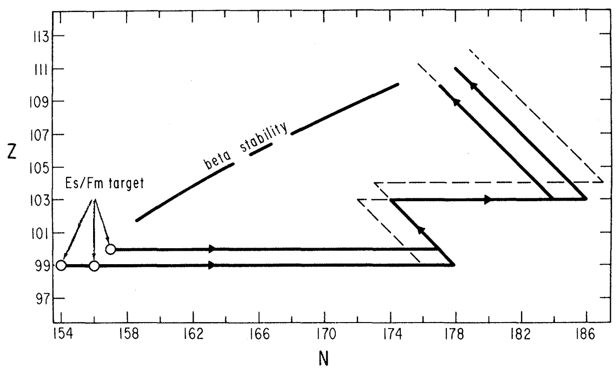

FIG. 2. Paths in a "double-shot" experiment. Two sequential nuclear explosions are used to provide intense neutron fluxes that are separated in time by about 0.01 sec. During this time, some of the neutron-rich nuclei produced in the first irradiation should undergo about four β decays. This leads to moderately unstable heavy nuclei with large proton numbers that serve as targets for the next irradiation. In this two-step, or a more general multistep, process, the magnitude of $(N-Z)^2/A^2$ (governing fission depletion) for the most neutron-rich nuclei that must be produced in order to reach the island of superheavy nuclei is much smaller than in ordinary one-step nuclear explosions.

such experiments (producing up to 10^{17} times as many nuclei as heavy-ion reactions).

The lowest useful exposure-time extension is determined by the fastest realizable β decays for transcalifornium elements up to Rf with $A \sim 273$ to 281. Our lower-limit estimates range from 17 to 0.5 msec according to the simple formula of Ref. 9, using the β energies of a recent mass-formula extrapolation[18] which is consistent with current nuclear matter theory. Extension by a factor of 10^3 relative to normal (20-keV equivalent prompt neutron) flux duration is therefore the very lowest reasonable limit. A possible advantage is that multimillisecond exposures can be successful at neutron fluxes an order of magnitude below those of Hutch.[11] The extended exposure allows smaller bandwidth capture/decay paths, more "zig zags" than Fig. 2, which is desirable in order to minimize strong decay depletion at the neutron-rich corners.

The upper limit for the time lapse beween subsequent exposures depends on the intermediate depletion that can be tolerated. This time constant is therefore given by the fastest strong decays for the non-ee neutron-rich isotopes of elements with $99 \leq Z \leq 105$. The uncertainty in the fission lifetime extrapolations based on Nilsson-Strutinsky calculations[19] and other current phenomenologies is so large that it automatically embraces the range of practical interest, namely, $10^{-1\pm1}$ sec; i.e., one order of magnitude above fastest β decays, and of the order of shock-wave travel times for the usual distances (cf. the Marvel event[11,20]).

Note: The more or less prompt methods proposed here can tolerate competition of much faster strong decays than conventional r processes —hypothesized to reach the superheavy island.[9] Heavy-ion fusion might succeed earlier in "naming" some short-lived neutron-deficient superheavy elements. However, neutron capture from explosives could very well be the only method reaching the *center* of the predicted island while yielding *macroscopic* quantities of superheavy elements.

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[1]Cf. talks of G. T. Seaborg and W. Greiner, in the Third International Transplutonium Element Symposium, Argonne National Laboratory, 20 October 1971 (unpublished); also G. T. Seaborg, in Fourth International Conference on Peaceful Uses of Atomic Energy, Geneva, 1971 (unpublished), and Annu. Rev. Nucl. Sci. 18, 53 (1968); J. R. Nix, LASL Report No. LA-DC-11825, 1970 (unpublished), and in Proceedings of the International Conference on Properties of Nuclei far off the Region of Beta-Stability, Leysin, Switzerland, 1970 (unpublished), Vol. 2, p. 605.

[2]H. W. Meldner and G. Hermann, Z. Naturforsch. 24a, 1429 (1969).

[3]See Greiner, Ref. 1; W. D. Myers and W. J. Swiatecki, UCRL Report No. 11980, 1965 (unpublished), and Nucl. Phys. 81, 1 (1966); H. W. Meldner, UCRL Report No. 16843, 1965 (unpublished), and Ark. Fys. 36, 593 (1967), and Phys. Rev. 178, 1815 (1969). A self-consistent HF calculation [W. H. Bassichis and A. K. Kerman, Phys. Rev. C 2, 1768 (1970)] led to a $Z = 120$ prediction. However, this is possibly due to certain deficiencies of the Tabakin interaction leading to a very unrealistic competition of Coulomb and nuclear forces [W. H. Bassichis, private communication; see also, P. Signell, Phys. Rev. C 2, 1171 (1970)]. Another recent HF calculation (G. Saunier and B. Rouben, to be published) supports *both* 114 and 120. All current "realistic" models, fitting the known energy gaps or mass discontinuities and the related shell-model spin assignments, predict $Z = 114$.

[4]E. Cheifetz, R. C. Gatti, R. C. Jared, S. G. Thompson, and A. Wittkower, Phys. Rev. Lett. 24, 148 (1970).

The intensity problem could be solved by combining a high-flux reactor with a heavy-ion accelerator.

[5]L. Westgaard, private communication, and in the Sixth European Conference on the Interactions of High Energy Particles and Complex Nuclei, Kitzbuhel, Austria, 26 September to 2 October 1971 (unpublished). He estimated a production rate of about 5000 nuclei per month for the reaction ^{238}U(^{56}Ca, xn)$^{294-x}$112 when using CERN's 25-GeV 10^{12}-protons/sec beam. A. Ghiorso and W. J. Swiatecki recently proposed a "low-energy" reaction in order to increase the N/Z ratio in heavy ions. At energies of at least 8-MeV per particle, projectiles such as Ca or Ge can pick up a few neutrons (or be stripped of few protons) in grazing collisions with a Th nucleus prior to compound system formation in the same thick Th target.

[6]W. J. Swiatecki, in Proceedings of the Nordic-Dutch Accelerator Symposium, Ebeltoft, Denmark, 19 May 1971 (unpublished), and private communication.

[7]M. Lefort, private communication, and in the Sixth European Conference on the Interactions of High Energy Particles and Complex Nuclei, Kitzbuhel, Austria, 26 September to 2 October 1971 (unpublished).

[8]By this we mean process characterized by millisecond time constants rather than microseconds (prompt) or seconds (rapid), as discussed in the text.

[9]D. N. Schramm and W. A. Fowler, Nature 231, 103 (1971).

[10]E. Teller and D. W. Dorn, private communication; G. A. Cowan, private communication.

[11]S. F. Eccles, UCRL Report No. 72167, 1969 (unpublished), and private communication; G. A. Cowan, in *Proceedings of the Thirteenth Robert A. Welch Foundation Conference, Houston, Texas, 1969,* edited by W. O. Milligan (Robert A. Welch Foundation, Houston, Texas, 1969); R. A. Heckmann, in *Proceedings of the Symposium on Engineering with Nuclear Explosives, Las Vegas, Nevada, 1970,* CONF-700101 (U. S. AEC Division of Technical Information, Springfield, Va., 1972).

[12]See, e.g., Fig. 4 of J. R. Nix, LASL Report No. LA-DC-12488, 1971 (unpublished).

[13]Heavy-element production paths cannot move too far from β stability since strong decays would then cause serious depletion. On the other hand, they have to have a minimum neutron excess in order to prevent β decays from becoming too slow relative to overall exposure times.

[14]G. I. Bell, Rev. Mod. Phys. 39, 59 (1967).

[15]Uranium capture chains are considerably longer than those of the first few neighboring elements with higher *and* lower Z (Ref. 11). This is mostly because of the $Z=92$ subshell influence.

[16]D. W. Dorn and R. W. Hoff, Phys. Rev. Lett. 14, 440 (1965). See also G. I. Bell, Phys. Rev. 158, 1127 (1967); and J. S. Ingley, Nucl. Phys. A124, 130 (1969). Eccles (Ref. 11) was, in fact, able to reporduce all yield curves by using *measured* (n,p) and (n,α) cross sections together with extrapolated capture rates.

[17]See also E. E. Berlovich, Izv. Akad. Nauk SSSR, Ser. Fiz. 34, 2286 (1970) [Bull. Acad. Sci. USSR, Phys. Ser. 34, 2038 (1970)].

[18]K. A. Brueckner, J. H. Chirico, and H. W. Meldner, Phys. Rev. C 4, 732 (1971).

[19]Table 5 of Nilsson *et al.*, Nucl. Phys. A131, 1 (1969), reveals an error in half-life prediction which is roughly one order of magnitude per extrapolated nucleon number—typical factors are 10^{-2} at $Z=94$ (^{242}Pu) and 10^6 at $Z=102$ (^{254}No)—in the *known* region. Extrapolation by about ten units in Z then can be expected to be uncertain by at least 10 orders of magnitude. See also M. Bolsterli, E. O. Fiset, J. R. Nix, and J. L. Norton, Phys. Rev. Lett. 27, 681 (1971), and Phys. Rev. C (to be published).

[20]H. D. Glenn and B. K. Crowley, J. Appl. Phys. 41, 689 (1970).

Nuclear Surface Energy and Neutron-Star Matter*

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Direct calculations of the nuclear surface energy are made for a Hamiltonian containing the Skyrme nucleon-nucleon interaction. A plane surface separating nuclear matter and a neutron gas or a vacuum is considered in Hartree-Fock and Thomas-Fermi approximations. These surface energies are incorporated in the compressible liquid-drop model to obtain properties of neutron-star matter. The Hartree-Fock results lead to Z values for the nuclei roughly constant at around $Z \sim 36$–38.

Neutron-star matter at densities between 4×10^{11} gm/cm^3 and approximately nuclear densities consists of neutron-rich nuclei immersed in a gas of pure neutrons. The size of the nuclei is determined by competition between the nuclear surface energy and the electrostatic Coulomb energy, and the atomic number Z is directly proportional to the energy per unit area of the nuclear surface. In the calculation of Baym, Bethe, and Pethick[1] (BBP) the surface energy inserted into their compressible liquid-drop model was estimated on the basis of dimensional arguments. The resulting