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AN ALTERNATIVE THERMONUCLEAR n-CAPTURE PATH
TO THE SUPERHEAVY ISLAND*

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MASTER

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Abstract

An alternative thermonuclear n-capture path to the superheavy island by Heiner W. Meldner (Department of Physics and IPAPS, University of California at San Diego, La Jolla, California, U. S. A. 92037), and John Nuckolls and Lowell Wood (Lawrence Livermore Laboratory, University of California, P. O. Box 808, Livermore, California, U. S. A 94551).

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Thermonuclear neutron-capture approaches to heavy transuranium element synthesis are investigated. Certain features of capture experiments using thermonuclear macro-explosives are re-analyzed in view of recent results of nuclear shell-structure theory and theoretical developments in laser-energized fusion micro-explosions. The micro-explosion conditions for prompt multiple neutron capture are estimated for the simplest moderation techniques. We speculate on some practical neutron-capture paths to the superheavy island, especially a hybrid approach, using both macro- and micro-explosion events.

1. Introduction

Our paper is presented in the r-process section of this Nobel Symposium; however, we should stress the word 'alternative' in the title. We are concerned with an alternative both in the neutron-flux exposure time relative to ordinary astro-physical concepts and therefore also in the path to heavy elements. Obviously this 'path' is very different from that in heavy-ion-fusion approaches.

In this alternative, we circumvent the two common difficulties of heavy-ion fusion; i. e., for one, excessive excitations of compound nuclei possibly destroying any helpful shell structure, and, on the other hand, neutron deficiencies possibly causing heavy-ion approaches to systematically miss the 'island' to the north-west. Instead, we might get stranded in the south-east because of fission depletions and problems in arranging underground shots.

In putting much emphasis on this thermonuclear neutron-capture approach for the past several years [1,2], we have obviously indicated our preferences as to the relative merits of various superheavy-element production schemes. In fact, this excellent Symposium has reminded us quite strongly that all real nuclear physics is concerned with the problem of 'model-dependent models', i. e., choices of models of many-body formalisms dependent on one's election of models of internucleon forces. Superheavy-element physics checks in the most sensitive

way whether any of these models are 'realistic' because we are speculating about their predictions; in fact, we are talking about fairly long-range extrapolations. At this point we are nonetheless prepared to place an infinitesimal bet on the success of our approach relative to the heavy-ion game.

In this paper we shall briefly sketch four topics:

1. basic (laboratory) prompt neutron-capture concepts;
2. predictions of shell and other structure effects for neutron-rich heavy isotopes;
3. new neutron sources, micro-explosives;
4. experimental suggestions.

2. Neutron Capture

The r-process essentials have been described in several previous papers. Let us just very briefly recall a few in order to indicate the main differences found in thermonuclear (micro-)explosives. The basic two steps are--as shown in Fig. 1--neutron capture and subsequent increase of proton numbers by β -decays. All this generally in competition with fission. The basic nuclear theory problems in this area are qualitatively similar to those encountered in astrophysical r-process calculations [3,4]. That is, we are mainly concerned with the stability structure resulting from the interference of closed curves for both equi-fission barrier heights and binding energies [5] in the N-Z plane.

Closed fissility contours result from surface symmetry terms in nuclear mass formulas [6]. Superposition of closed curves for binding and fissility then results in an 'r-process region' which is a strip roughly parallel to and to the right of the beta stability line in the N-Z plane. [See Fig. 1.] In a rough approximation where shell structure is neglected, this strip region seems to be limited to $Z \leq 103$. It is determined as follows: for heavy elements, neutron-induced fission occurs near beta stability since the neutron binding energies are larger than the fission barriers. At some point to the neutron-rich side of beta stability, neutron binding and fission barriers are equal; these points are on the '(n, f) line', the left-hand limit of the r-process region. In some area to the right of this, barriers are higher than the decreasing neutron binding (or Fermi) energy. At still higher neutron excess, the fissility increases so much that spontaneous fission takes over; i. e., we have reached the right-hand limit of the r-process region.

It is important, however, to note the significant time-scale differences between prompt neutron capture in laboratory-scale experiments and astrophysical r-process situations. For example, in laser-energized fusion devices, we consider a so-called 'prompt' capture which is not only faster than intermediate g-decays but faster by factors of at least 10^{11} than the 'rapid' neutron capture sequence in conventional astrophysical r processes. For all but the end points of a capture chain, this short neutron-flux exposure allows neutron capture to compete with fission modes of decay that are 10^{11} times faster than the fastest tolerable ones in astrophysical r processes.

3. Shell Effects

Figure 1 describes an over-simplified picture where shell effects are neglected. In reality, we expect shell effects to complicate the situation as qualitatively shown in Fig. 2; i. e., there are localized areas of isotopes with short spontaneous fission half-lives also between the beta-stability and (n, f)-line. Note, this structure is a common feature of all predictions presented at this Symposium. The structure of neutron fission contours is, of course, similar to these spontaneous fission contours (see W. M. Howard [7, 8]). This is a major new and encouraging result which tells us that previous experiments did not fail in producing elements with $Z > 100$ because fission took over along capture chains-- on the way out to neutron richness--but rather because the yield was cut off during the decay back to beta-stability where the decay paths hit areas of strong spontaneous fission competition. This picture indicates at the same time that there should be a 'bridge' to the superheavy island. This is shown in some more quantitative detail in Fig. 3 which refers to odd Z-odd A elements only. [See also Ref. 9 and Fig. 10.]

Most of the data in this contour plot are taken from Refs. 9 and 4. As a first-order estimate of the odd nucleon-number effects, we have simply added five orders of magnitude to the fission half-lives calculated for even-even nuclei ('pessimistic' case of Ref. 9 and S. G. Nilsson, private communication; for a more recent model prediction, see Fig. 10, below). The neutron-induced fission (n, f) line and the beta-stability

results are those of Ref. 4. We see that long capture chains are possible for $Z = 92 \pm 1$. Initially, a long chain is also necessary since beta decay will otherwise drive the path into the fission areas as depicted in Figs. 2 and 3 [10]. We have to circumvent especially the 'area I' (also called 'Bay of Pigs', cf. Refs. 7,8) indicated in the semi-quantitative plot of Fig. 2. Theoretically, it seems to be well established now that shell structure effects generate these disconnected areas I and II of low fission barriers. Most interestingly, as seen clearly in Fig. 3, this disconnection causes the building of a 'bridge' supported by the $N = 184$ shell closure. After an initial 'long shot' the latter bridge should allow several short capture/decay zig-zags that finally approach the superheavy-element region. Beta lifetimes are quite short in this very neutron-rich area; i. e., we have to rely on nuclear structure effects also in order to have sufficiently high hindrance factors for the weak decays.

In leaving this brief semi-quantitative survey of recent nuclear-structure predictions let us again emphasize that the necessity of an initial capture of 20 to 30 neutrons followed by much shorter capture/decay zig-zags is a common feature to all current model predictions. The question is therefore: can we foresee a neutron-source technology that might allow the required controlled neutron exposure?

4. Neutron Sources

There is a substantial amount of literature available on the use of (underground) thermonuclear macro-explosives for capture [10,11]. It seems obvious that a Hutch-type device is necessary for the first step which consists of a very long capture chain. From there on exposure in micro-explosives might help us; however, it is a more futuristic concept.

The best introduction to laser-energized fusion micro-explosives is probably an article in this month's issue of Scientific American [12]. This is just the most recent of a series of publications [13-15] on fusion in a strongly pre-compressed thermonuclear fuel pellet created by a "spherical ablation implosion rocket, externally energized by a power-programmed laser." The idea is that roughly symmetrical laser heating of the full surface area of the atmosphere surrounding the pellet and the reaction forces associated with the ensuing explosively rapid ablation of the pellet's surface drives a strong compression wave into the unheated material ahead of the thermal conduction wave and the hydrodynamic rarefaction wave. Complex computer simulation codes show that very large pellet compressions may be achieved in this way by subjecting the pellet's atmosphere to laser light of very rapidly increasing intensity in such a fashion as to result in all acoustic signals from the ablating pellet surface arriving at the center of convergence at essentially the same time [14], producing nearly shockless, isentropic compression.

This obviously requires an elaborate laser-pulse shaping so that the pellet compression history is in 'resonance' with the implosion-driving laser pulse intensity. It is also important to keep the electrons of the pellet essentially Fermi-degenerate during the compression; this reduces the implosion pressure requirement from 10^{12} to 10^{11} atmospheres [14], resulting in an order of magnitude saving in laser pulse energy. If the implosion is continued until the pellet's density-radius product is significantly greater than $0.3 \text{ grams cm}^{-2}$, charged particles from the fusion reaction are absorbed within the dense pellet, and the ion-electron coupling time becomes significantly shorter than the inertial confinement time. In such optimal configurations, the fuel 'ignites' in the sense of a high explosive: a thermonuclear detonation front propagates outward from the hydrodynamically ignited pellet center in a fraction of the pellet's hydrodynamical disassembly time, thus requiring thermonuclear ignition conditions to be produced in only a very small ($\leq 1\%$ by mass) core of the compressed fuel. In comparison to the energy-break-even condition for uncompressed DT pellets ($> 10^9$ joules) the required laser input can typically be reduced to about 10^3 joules at 10^4 -fold compression. In terms of explosive size, this requires a laser energy of the order of a kilojoule per microgram of fuel. Nuclear reactions then occur at local densities which--as demonstrated for 'typical examples' below--might be high enough to produce moderated neutron fluxes useful for the short capture/decay zig-zags discussed earlier.

The calculations leading to the simple pocket formulas quoted here are based on scaling of the thermonuclear burn processes into these highly compressed situations under the ρR variant. Teller [13] has noted that this simplification is not entirely justified, as the density dependence of the Coulomb logarithm and inverse bremsstrahlung is neglected. For the typical compression situations we consider, however, binary collision processes are completely dominant. Compton scattering of bremsstrahlung photons by plasma electrons are significant for some aspects of thermonuclear burn for density-radius products in excess of about 20 grams cm^{-2} [13 and x]. However, in the area of interest to us here, all of these effects produce only minor changes in the neutronic aspects of the thermonuclear burn.

Let us look at the simplest analytical estimates. For example, the diffusion moderation result for bare solid DT pellets demonstrates immediately the difficulties in obtaining a significant slow (< 0.5 MeV, or so) neutron flux relative to the fast flux: The time-integrated fast neutron flux, i. e., the fast fluency is of order

$$\bar{\phi}_f \approx 10^{24} \rho R \quad (1)$$

in CGS units, and the moderated or slow one is of order

$$\bar{\phi}_s^\infty \approx 10^{23} (\rho R)^2 \quad (2)$$

for DT, if we make the over-optimistic assumption of having an essentially infinite moderation time available. In reality, moderation is, of course, terminated within the pellet's hydrodynamical disassembly

time of order 10^{-10} sec. [14, 15]. But even if we neglect any reduction of this estimate for the slow fluency, we have a rather distressing result: We need ρR 's around 30 gm/cm^2 to have the slow flux exceed the fast one by a factor of three or so--the latter being a reasonable minimum requirement for neutron capture synthesis without excessive fast fission losses. At this time, achievement of implosions with such high ρR 's seems to be very challenging in terms of confidently foreseeable technology. In the terminology of Ref. 12, this region is reserved for the 'second generation' of laser-fusion devices. On the other hand, we have the clear indication from our theory as well as from macro-explosion neutron-capture experiments that fast fission is a major obstacle in this approach to heavy-element synthesis. That is, moderation techniques are absolutely crucial in order to avoid hopeless depletion of high Z target material by the initial burst of unmoderated high energy neutrons generated in the burning fusion fuel. In view of this, it seems hopeless to attempt any significant neutron capture with a simple pure DT scheme. This dilemma is illustrated in Figs. 4 and 5 where we actually plot a somewhat more realistic diffusion estimate, namely,

$$\phi_s^{aR/v} = \phi_s^\infty \left\{ 1 - \frac{12}{\pi^2} \sum_{v=1}^{\infty} \frac{(-)^v}{v^2} \exp \left[- \frac{v^2 \alpha_c}{\rho R} \right] \right\} \quad (3)$$

This fluency estimate takes into account finite moderation time by a parameter α multiplying R/v , a mean value of the time in which a

neutron crosses the pellet. Realistic moderation times are of order $R/3v$ to R/v . (For DT, we have $c = 13 \text{ gm/cm}^{-2}$.)

Figure 4 shows that sufficiently high total moderated fluencies can be expected for ΦR 's around 10 gm/cm^2 (cf. the fluency levels of the thermonuclear macro-explosion experiments Anacostia and Par). However--as anticipated from the low $\phi_{\text{slow}}/\phi_{\text{fast}}$ result for the over-optimistic case--we see in Fig. 5 that the slow fluencies are considerably lower than the fast ones for moderation times of $R/3v$ or less, even if there is no restriction in the magnitude of achievable ΦR 's.

One solution of this problem is to homogeneously admix substantial quantities of moderators such as hydrogen with a high quality fuel such as DT. Note that $\Phi R/v$ as introduced above is the characteristic time for the burning fuel. Disassembly of the whole compressed pellet will take several times longer than that of the initially ignited core. That is, the curves labelled R/v in Figs. 4 and 5 provide an estimate for the possible improvement by such configurations.

More quantitative results can be obtained from computer-based neutron transport calculations for realistic pellet implosion-explosion processes, using a Monte Carlo scheme. The achievable fluencies for pure DT and for DT with 90% hydrogen are plotted in Fig. 6.

The actual neutron spectrum for a 25 gm/cm^2 pellet with 90% hydrogen is shown in Fig. 7. This brief sample shows us that one can expect schemes of this type to achieve fluxes useful for multiple neutron capture. Computer optimized pellet configurations are not homogeneous; for example, they may have a DT core, DD as the major fuel component and target material surrounded by a homogeneous moderator.

Note, however, that the total slow neutron fluencies cannot compete with those of Par- let alone Hutch-type devices, especially if one considers the 'first generation' [12] laser-fusion pellets of order 5 to 10 gm/cm². Micro-explosions are, on the other hand, easily contained since the pellet masses in question are so small that the momentum transferred to standard-size container walls (cf. Refs. 12 and 14) is comparable to that of a firecracker in spite of their thermonuclear explosive power ranging up to hundreds of pounds of TNT. Laser-fusion devices are thus very attractive neutron sources for fast recovery and re-exposure purposes.

5. Experiments

Experimental suggestions involving the use of laser-energized fusion are naturally speculative at this time. However, reasonably short-range extrapolations of technological developments for schemes as indicated above lead us to believe that controlled sequential neutron capture, preferably in odd-Z elements, is a promising approach to transuranium nucleosynthesis.

As we have mentioned earlier when discussing the implications of Fig. 3, elements near ²³⁸U seem to be optimal as targets for the first step. Let us therefore focus more strongly on this part of the N-Z plane since we have some experimental evidence on neutron-rich elements in this area, as indicated in Fig. 8.

A capture chain starting with ^{242}Np has an advantage of four neutrons over ^{238}Pa ; however, as shown in Fig. 3, it more closely approaches the low (n, f) threshold area near $N = 166$ than is the case for the Pa chain, which has about 1 MeV higher thresholds everywhere. All this suggests that a mixed target is optimal, consisting of ^{238}U and ^{242}Pu in an arrangement which maximizes the ratio of (n, p) reactions to (initial) fast fissions.

Projecting current technology to about the year 1976, we might conceive the following steps in a multiple-capture experiment:

- 1) Exposure of ≥ 10 grams of ^{238}U and ^{242}Pu in a Hutch-type underground device, designed for maximum (n, p) reactions and rapid partial recovery. Recovering a fraction of $10^{-8 \pm 1}$ of the neutron-richest products would be ideal; 10^{-12} , i. e., $\sim 10^6$ atoms, seems to be the lowest tolerable limit.
  - 2) Re-exposure of the recovered neutron-rich elements--which should include $Z \sim 93$ to 97 and $N \geq 174$ --in a laser-fusion device within minutes to hours of (underground) production.
- Note: The longest expected half-lives of beta decays driving the produced isotopes towards the critical spontaneous fission areas (Figs. 2 and 3) are in the range of minutes. A relatively fast, high efficiency recovery of all transuranic isotopes seems feasible since laser fusion yields a contained plasma.

In the preceding paper, Mike Howard⁸⁾ has shown us a plot of (n, f) thresholds which has a 'Cape Farewell' close to this area (his Fig. 5). That is, the r process would be terminated at a point perhaps as low as $Z = 98$, $N = 184$, and certainly near $Z = 106$, $N = 184$. However, in comparing this plot with our Fig. 8, we see that empirical evidence indicates the real thresholds to be about 1 MeV higher. But it looks as if controlled prompt capture can handle even his pessimistic prediction simply because it allows us to 'jump' over some critical areas using the spontaneous-fission 'bridge' seen in Fig. 10. For example, we can zig-zag up to near $Z = 99$, $N = 186$ and then (beta) jump from this eastern point on 'Cape Farewell' to the shore of the island at $Z = 107$, $N = 178$. According to our Fig. 10, spontaneous fission is so slow on this path across the odd-A 'bridge' that depletion would be quite negligible.

- 3) Final exposure of target material containing elements with $Z \sim 107$, $N \geq 176$. Capture of only four to seven neutrons and subsequent beta decay then can lead to superheavy products.

Naturally, we have to expect losses of one or two orders of magnitude per recovery and exposure. Starting step 2) with only 10^6 atoms should then still produce detectable yields under rather pessimistic assumptions. There is no need to recover the favorable isotopes only in intermediate steps; the useless ones will automatically be 'separated' by fission. A more comprehensive study is expected to be available soon [2].

In theory, this approach sounds rather straightforward. In practice, the above steps are likely to require the efforts necessary for 'giant leaps'. As a first stage, it might be desirable to have an international collaboration on analyzing the data from an underground shot designed for rapid recovery of neutron-rich isotopes to test structures as described in our Figs. 8 and 10. Such an experiment would not only check the feasibility of step 1, above, but also test the predictive power of our theory at a distance equal to that from known isotopes to the superheavy island.

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Figure Captions

Fig. 1 Simplified pattern of

(1) beta stability (dashed dotted),

(2) neutron induced fission, nf (dotted), and

(3) spontaneous fission lines, sf (solid),

in the N - Z plane for the heavy isotopes of interest neglecting shell effects. Prompt capture breeding requires a recovery of isotopes between the nf and sf lines and re-exposure within their beta-decay times which typically range from seconds to hours.

Fig. 2 Simplified structure of current estimates for a stability map

of odd Z /odd A isotopes emphasizing two major features;

(1) regions of low fission barriers above $Z \approx 93$, around $N \approx 166$ ('Bay of Figs') and above $Z \approx 99$, around $N \approx 174$;

(2) a shell effect, producing high barriers for $N \approx 178$ to 184 for all Z 's in the area shown. Lines have the same meaning as in Fig. 1.

Fig. 3 Estimated half-life contours for odd Z /odd A nuclei in the

N - Z plane. The thin solid zig-zag paths refer to capture experiments as discussed in the text.

- Fig. 4 Total neutron fluencies as function of ρR for different values of the moderation time. These flux estimates are compared with the flux levels shown as dashed lines of the thermonuclear devices, 'Par' and 'Anacostia' (which produced capture of at least 10 and 8 neutrons, respectively, in a ^{238}U target).
- Fig. 5 Diffusion estimate of the fluency ratios obtainable at various moderation times.
- Fig. 6 Monte Carlo calculation of neutron fluency ratios for DT with and without admixture of 90% hydrogen (by volume).
- Fig. 7 Neutron spectrum for a 25 gm/cm^{-2} DT pellet with 90% H.
- Fig. 8 Semi-empirical slow neutron fission threshold contours for all nuclei in the N-Z plane. Heavy lines indicate where it is presently possible to normalize to fission data from macro-explosions [11] or other experiments [16]. The dashed lines are extrapolations using the contour shapes predicted by several recent calculations [7, 9, 17] with a κ_{eff} around 2. ($Q = 19$ set, at $Z \approx 94$, $A \approx 280$) For $N < 170$, this semi-empirical plot is expected to be accurate to within $\pm 0.5 \text{ MeV}$.

Fig. 9 Estimated odd Z effect in yield curves of macro-explosion experiments (adapted from Ingley, Ref. 11). Note the decrease in slope for the odd Z chains, starting from Pa and Np relative to the yield curves for the even elements U and Pu.

Fig. 10 Preliminary results from Ref. 17 calculated with a modified oscillator model for odd Z /odd A nuclei. The numbers in circles and squares are logarithms of spontaneous fission half lives (in years). Spin and parity of the most probable groundstate configuration are given on the right. Circles refer to nuclei with a 'first minimum' groundstate (weakly deformed or spherical); squares denote nuclei with large initial deformations.

Fig. 1

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