

How to detect the heaviest man-made isotopes*

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The average number of neutrons emitted per spontaneous fission is predicted to increase substantially for very heavy nuclei. This suggests the use of neutron-multiplicity counters for the early detection of heavy isotopes produced by prompt neutron capture. We discuss the possibilities and limitations of such a technique.

NUCLEAR REACTIONS Fission; calculated $\bar{\nu}$ of very neutron rich isotopes produced in thermonuclear explosions; proposed method for early detection of heavy isotopes produced.

I. INTRODUCTION

Heavy elements up to ^{257}Fm have been made in thermonuclear explosions by prompt multiple-neutron capture on uranium targets.¹⁻⁵ Current nuclear-structure theory of heavy neutron-rich isotopes suggests that neutron capture on uranium continues past mass 257 with the consequent production of much heavier isotopes. The apparent limit of $A=257$ observed in all exposures to date is therefore possibly due to the relatively long time required for recovery and mass analysis of the products rather than to intrinsic difficulties associated with producing heavier nuclei. In the "Hutch" experiment, for example, more than 10^{17} atoms of mass 262 could have been made initially.² In an experiment with a neutron flux somewhat higher than in Hutch, perhaps up to 10^{16} atoms of mass 265 and 10^{12} atoms of mass 270 could be produced initially,⁶ as seen in Fig. 1.

Some problems of neutron-capture synthesis can be understood from Fig. 2, which summarizes a common feature of all recent nuclear-fission predictions presented, for example, at the 1974 Nobel Symposium on Superheavy Elements.^{7,8} In particular, above $Z \approx 94$, and $N \approx 166$ there exists a region, hereafter called the 94-166 region, where fission barriers are relatively low and consequently spontaneous-fission half-lives are relatively short. Because of this, any initial yield y_0 of isotopes with $A \geq 258$ after prompt capture will be cut off during the decay back to β stability when the decay path hits this area of strong spontaneous-fission competition. (The same structure calculations indicate that there may be a "bridge"—the

possibility of a jump to superheavy nuclei—at $N \approx 184$.⁷⁻⁹) It is understandable from Fig. 2 why the yield curve of nearly β -stable isotopes ν_0 should terminate at $A=257$. Given this situation, a crucial experiment is a Hutch-type underground shot designed for the *early* detection of neutron-rich isotopes, which would then test the structure illustrated in Fig. 2. In this way, a shell-structure effect related directly to the one that led to the prediction of an island of superheavy nuclei

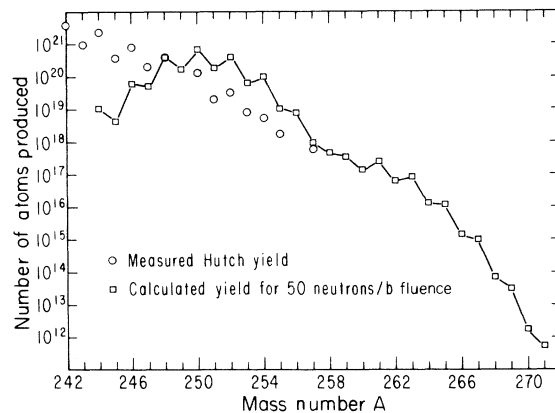


FIG. 1. Number of atoms produced in a thermonuclear explosion as a function of the mass number A . The circles give the experimental yield for the Hutch experiment (Refs. 3 and 4). The squares connected by line segments are calculated by Bell (Ref. 6) for a ^{238}U target of 5×10^{22} atoms and for a fluence (time-integrated neutron flux) of 50 neutrons/b. This size target is 11% larger than that used in Hutch, and this fluence represents some improvement over that in Hutch, by perhaps up to a factor of 2.

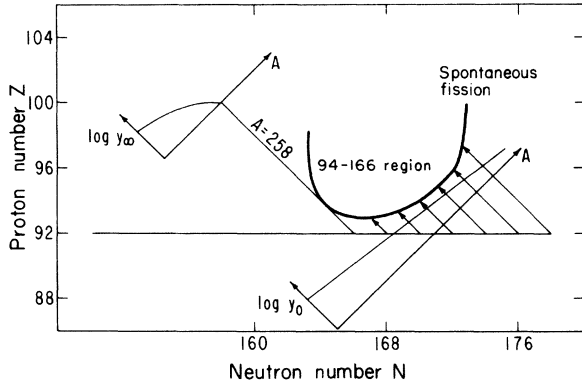


FIG. 2. Qualitative effect of an area of short spontaneous-fission lifetimes (above $Z \approx 94$ and $N \approx 166$, as indicated) on the yield curve of elements near β stability.

could be tested.

Conventional mass analysis, even when combined with the most rapid recovery techniques, seems inadequate for the detection of a yield curve similar to the one denoted by y_0 in Fig. 2. Because most β decays in this area do not take more than a few seconds, the isotopes of interest undergo spontaneous fission before any processing can be completed.

Obviously, we need some very early counting technique which would check characteristic spectral features, for which we propose the use of a *neutron-multiplicity counter*.¹⁰⁻¹³ Such a technique should be possible because the average number of neutrons $\bar{\nu}$ emitted per fission is predicted to increase substantially for very heavy nuclei. The theory of this effect is discussed in Sec. II, and Sec. III describes neutron-multiplicity counters that may be used for this purpose. In Sec. IV we estimate the expected neutron-multiplicity counting rates and neutron background in connection with the problem of resolving the neutrons emitted from the various sources. Our conclusions are presented in Sec. V.

II. AVERAGE NUMBER OF NEUTRONS PER FISSION

The average total number of neutrons $\bar{\nu}$ per fission is determined primarily by the average total excitation energy \bar{E}_{ex} and neutron separation energy \bar{B} of the fission fragments. In particular, as discussed in Ref. 14,

$$\bar{\nu} = \bar{E}_{ex} / (\bar{B} + \bar{\eta}_{c.m.}) - 1,$$

where the average neutron kinetic energy $\bar{\eta}_{c.m.}$ (in the reference frame of the moving fission fragment) is also a function of the fragment excitation energy. The quantity “-1” in this expression takes into account approximately the terminal deexcitation of the fission fragments by γ decay rather

than by neutron emission.

As the neutron number N of the nucleus undergoing fission increases, the average neutron separation energy \bar{B} of the fission fragments in general decreases, which increases the average number of neutrons $\bar{\nu}$.

The average fission-fragment excitation energy \bar{E}_{ex} is equal to the difference between the energy release in fission and the average fission-fragment kinetic energy. An increase in the proton number Z increases the total energy release and in general increases both the fission-fragment kinetic energy and excitation energy separately. Therefore, the average number of neutrons $\bar{\nu}$ usually increases with increasing Z .

However, if the fission fragments are close to a doubly magic nucleus such as ^{132}Sn , their special stability against deformation leads to an anomalously high kinetic energy and correspondingly low excitation energy. In this case $\bar{\nu}$ may actually decrease with the addition of protons. For the fission of nuclei close to the valley of β stability this effect is taken into account semiempirically in the work of Schmitt and Mosel by adjusting the stiffnesses of fission fragments in a static two-spheroid scission model.¹⁵ However, these estimates do not apply to the heavy neutron-rich nuclei in which we are primarily interested. For example, when the mass number is near 264, the nuclei of primary interest to us are from a region given by $N = Z + 69 \pm 4$ and $A = 265 \pm 4$ (where fission fragments are far removed from ^{132}Sn) rather than $Z = 100$ and $N = 164$. For this extremely neutron-rich region of nuclei, single-particle effects at the scission point are expected to be much smaller, and the resulting division of the total energy release into kinetic energy and excitation energy should follow more closely the prediction of the liquid-drop model, which yields a smooth dependence on fissioning nuclei. Because of this, we obtain the average fission-fragment kinetic energy from Viola's semiempirical relationship¹⁶

$$\bar{E}_{kin} = (0.1071Z^2/A^{1/3} + 22.2) \text{ MeV}.$$

The estimation of the energy release (and hence the excitation energy) involves large uncertainties associated with how rapidly the nuclear surface energy decreases with the addition of neutrons. This decrease is often represented in terms of a surface-asymmetry constant κ , which is defined so that the surface energy $E_s^{(0)}$ of a spherical nucleus is given by

$$E_s^{(0)} = a_s A^{2/3} \left[1 - \kappa \left(\frac{N-Z}{A} \right)^2 \right].$$

Although in recent semiempirical nuclear mass

formulas^{17, 18} the effect of the neutron excess is actually somewhat more complicated, the effective value of κ determined by these mass formulas is approximately 2.8 ± 0.5 . There is still considerable uncertainty in this quantity.

Increasing the value of κ leads to a larger energy release for very neutron-rich nuclei and hence to a larger number of neutrons per fission. Conversely, decreasing the value of κ leads to a smaller number of neutrons per fission. There are also uncertainties in the neutron separation energy for very neutron-rich nuclei, which arise from uncertainties in both the volume-asymmetry and surface-asymmetry constants in the nuclear mass formula.

Because of this dependence of $\bar{\nu}$ on the constants of the nuclear mass formula, the measurement of a high neutron multiplicity in fission can in principle be used *both* to detect the production of heavier nuclear species and to determine more accurately the effective surface-asymmetry constant (*if* Z assignments are possible); thus, one shot might answer two crucial questions of superheavy-element theory.

As an example of the expected increase in $\bar{\nu}$ for very heavy nuclei, we show in Fig. 3 results calculated with the recent mass formula of Seeger and Howard,¹⁸ which includes shell-structure corrections. The values of $\bar{\nu}$ increase from about 4 for mass numbers close to 254 to about 8 for $A \approx 266$ and 10 for $A \approx 276$. The calculations are

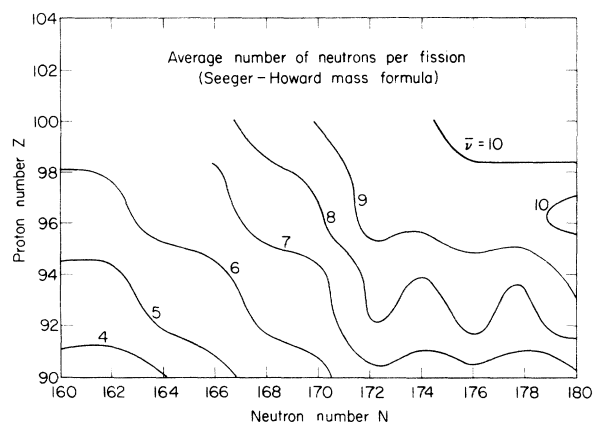


FIG. 3. Contour plot of the average number of neutrons per fission vs the neutron number N and proton number Z . The results are calculated for the spontaneous fission of even-even nuclei into two equal fragments by use of the mass formula of Seeger and Howard (Ref. 18), which includes shell-structure corrections calculated with Strutinsky's method. The values for nuclei near $Z=100$ and $N=164$ are expected to be lower than those calculated here because fission fragments near the doubly magic nucleus ^{132}Sn are especially stable against deformation.

performed for the spontaneous fission of even-even nuclei, with the energy release calculated for division into two equal fragments. The slightly smaller energy release in the case of odd-odd fragments is responsible for part of the fluctuations in the contours, although additional fluctuations arise from single-particle effects on the ground-state masses. These are calculated in the mass formula of Seeger and Howard by use of Strutinsky's method from the single-particle levels of a modified harmonic-oscillator potential.

III. NEUTRON-MULTIPLICITY COUNTERS

Neutron-multiplicity counters have been developed recently in connection with searches for superheavy elements in nature.^{10, 11} As will be discussed in the next section, the usefulness of these counters is restricted by the large size of their gates of about 100 μs (highly moderated neutrons are detected) and their vulnerability to γ background. The latter, in fact, excludes the counter described in Ref. 10 for our purposes. The ^3He counter of Ref. 11, on the other hand, appears to be the least γ sensitive instrument developed thus far. In addition, its originally low efficiency has just been improved by some modifications.

A polyethylene cylinder was inserted that contained 10 additional ^3He detectors, bringing the total up to 30. The resulting sample chamber is 10 cm in diameter and 61 cm long. This alteration increases the single-neutron efficiency ϵ from 0.30 to 0.46, which improves considerably the detection of high multiplicities. The ^3He counter is significantly less sensitive to γ rays than a scintillation counter detecting thermalized neutrons. Halperin *et al.*¹² were able to count tantalum plates showing 5000 R/h near the surface by raising the lower-energy window and surrounding the Ta plates with relatively small amounts of Pb and Al. Because they were looking for a few counts a day of multiplicities ≥ 4 , rare pileup events would have been significant. On the other hand, for the experiment suggested here shielding should not be necessary for samples showing up to about 10^4 R/h.

Another possibly useful instrument for our purposes is a fast-neutron-multiplicity counter that has been suggested by Macklin; components of this system are now being developed.¹³ This scintillation counter would count fast neutrons by detecting proton recoils with a time window of about 30 ns after the first neutron. For this gate, neutron losses are about 0.9% and single neutrons with energies down to 0.1 MeV should be detected. Decreasing the gate to 13 ns cuts off neutrons below 0.5 MeV. Both the sample cavity and the outside of the containers of the scintillator plus

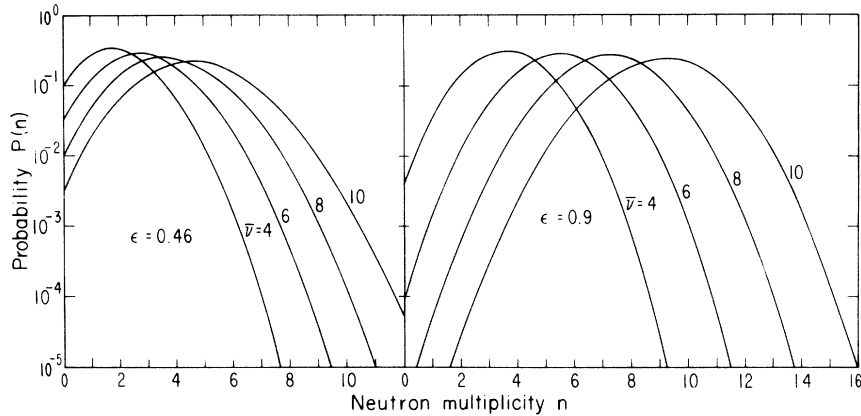


FIG. 4. Probability $P(n)$ for observing events of neutron multiplicity n , for four values of the average number of neutrons $\bar{\nu}$ per fission. The results are calculated by use of Eq. (1) for single-neutron detection efficiencies $\epsilon=0.46$ and 0.9 , which correspond to the two types of neutron-multiplicity counters discussed in Sec. III.

hydrogenous material would have dodecahedron shapes with a phototube against each outside face. It is anticipated that efficiencies of some 0.9 or so can be obtained with a reasonable thickness of scintillator medium. Unfortunately, the fast counter cannot tolerate any significant γ background.

The probability $P(n)$ for observing events of neutron multiplicity n is a function both of the single-neutron detection efficiency ϵ and the intrinsic distribution $p(\nu)$ for emitting ν neutrons per fission. In particular,¹⁹

$$P(n) = \sum_{\nu=n}^{\infty} p(\nu) \frac{\nu!}{n!(\nu-n)!} \epsilon^n (1-\epsilon)^{\nu-n}. \quad (1)$$

For $p(\nu)$ we assume a Gaussian^{19,20}

$$p(\nu) = \frac{1}{\sqrt{2\pi}\sigma_{\bar{\nu}}} \exp\left[-\frac{(\nu-\bar{\nu})^2}{2\sigma_{\bar{\nu}}^2}\right].$$

The width $\sigma_{\bar{\nu}}$ is obtained by linearly extrapolating experimental values of $\sigma_{\bar{\nu}}$, viz.^{19,20}

$$\sigma_{\bar{\nu}} = 1.05 + 0.04\bar{\nu}.$$

Several cases of interest here are shown in Fig. 4.

IV. EXPECTED SPECTRA AND RESOLUTION PROBLEMS

Because the nuclei produced in a thermonuclear explosion by prompt multiple-neutron capture are far from the line of β stability, the lifetimes corresponding to the first several β decays should be very short. In particular, the initial β -decay lifetimes are expected to be short compared to the spontaneous-fission lifetimes.^{8,9} The heavy neutron-rich nuclei that are produced should therefore undergo a succession of rapid β decays until they reach the 94-166 region, where the spontaneous-fission lifetimes suddenly drop well

below the β -decay lifetimes (see again Fig. 2). As can be seen from the allowed β transitions listed in Table I, the β decay times required to reach the 94-166 region are expected to lie within the range of 10 to 1000 s, once account is taken of the hindrances arising from forbidden transitions (see also Refs. 8 and 9). After the final β decay the resulting nuclei should rapidly undergo spontaneous fission, which will be accompanied by the emission of several neutrons per fission. It is the detection of these events of high neutron multiplicity in the presence of strong delayed-neutron and delayed γ backgrounds to which we now turn our attention.

The number of nuclei that are produced in a thermonuclear explosion and that reach the 94-166 region can be estimated according to the methods of Refs. 2, 5, and 6. Although these calculations do not take into account explicitly the fission of nuclei from excited states following β decay, this depletion in the yield is accounted for approximately by adjusting the neutron fluence and neutron-capture cross sections so that experimental yields of nuclei close to β stability are reproduced approximately.

The results calculated by Bell⁶ for a thermonuclear explosion whose neutron flux is somewhat higher than in the Hutch experiment¹⁻⁴ are shown in Fig. 1. For example, for mass numbers 260, 265, and 270, the calculated total number of atoms N_0 that are produced in the explosion and that arrive at the 94-166 region is about 10^{17} , 10^{16} , and 10^{12} , respectively.

By means of a recovery pipe leading from the surface down to the underground explosion, a small fraction f of the total debris is to be recovered rapidly and analyzed in one or more neutron-multiplicity counters. In a system designed

TABLE I. Allowed β -decay lifetimes for some isotopes originating from the P_α -chain calculated from Eq. (12) of Kodama-Takahashi (Ref. 27), using the Seeger-Howard mass formula (Ref. 18).

A	Z	τ_β (s)
261	91	0.2
	92	1.7
	93	0.6
	94	2.5
	95	2.0
	96	8.3
263	91	0.1
	92	1.0
	93	0.4
	94	4.8
	95	1.6
	96	5.5
	97	4.5
	98	14.5
	100	27.6
265	91	0.1
	92	0.7
	93	0.2
	94	2.7
	95	0.9
	96	3.8
	97	3.8
	98	8.5
	99	8.1
	100	27.6
	101	22.9
267	91	0.1
	92	0.5
	93	0.2
	94	1.6
	95	0.5
	96	5.9
	97	2.1
	98	7.8
	99	7.0
	100	13.5
	101	22.9

for maximum recovery, the fraction f could be as large as perhaps 10^{-8} . However, as we will see momentarily, the large delayed-neutron and delayed γ backgrounds require that much smaller fractions ($f \approx 10^{-13}$) be used during at least the first few minutes of counting. The sample size can of course be increased as the backgrounds drop with increasing time in order to search for longer-lived nuclei. For sample sizes up to even perhaps 10^{-10} , a relatively simple recovery pipe attached parasitically to an explosion designed primarily for another purpose should be feasible.

At time t after the explosion, the number of atoms of a given mass number that remains in the sample being counted is

TABLE II. Delayed-neutron and delayed- γ backgrounds for a sample whose size represents a fraction f of a thermonuclear device that releases 10 kt of its explosive energy from the fission of ^{239}Pu . The γ background refers to a distance that is 5 cm from a point source.

Time (min)	Neutron background (neutrons/100 μs)	γ background (R/h)
0	$3.6 \times 10^{17}f$...
1	$1.5 \times 10^{15}f$	$1.3 \times 10^{14}f$
2	$2.9 \times 10^{14}f$	$6.2 \times 10^{13}f$
5	$1.0 \times 10^{13}f$	$2.4 \times 10^{13}f$
10	$2.0 \times 10^{11}f$	$1.1 \times 10^{13}f$
15	4.0×10^9f	$7.4 \times 10^{12}f$
20	8.5×10^7f	$5.4 \times 10^{12}f$

$$N(t) = N_0 f \exp(-t/\tau_\beta), \quad (2)$$

for the case in which the lifetime τ_β for the last β decay prior to the 94-166 region is much longer than those for the preceding β decays. This represents the total number of coincidence events of high neutron multiplicity for this mass number that will be recorded after the neutron and γ backgrounds become sufficiently low to permit counting to begin at time t .

We estimate the delayed-neutron and delayed- γ backgrounds on the basis of a thermonuclear explosion that releases 10^{13} cal (equivalent to 10 kt of TNT) of its explosive energy from the fission of ^{239}Pu , which is equivalent to the fission of approximately 1.46×10^{24} nuclei.^{21, 22} This corresponds to a conservatively large fission contribution sufficient for a Megaton-size thermonuclear device.^{22, 23} For times less than about 20 min, the delayed-neutron background can be approximated by the sum of six terms that decrease exponentially in time; the corresponding half-lives range from about 0.2 s for the shortest-lived component to about 55 s for the longest-lived component.²⁴ For times longer than about 20 min, additional long-lived delayed neutrons arising from (γ, n) reactions with various nuclei and from photofission of ^{239}Pu also become important.²⁵ Because we are interested primarily in the shorter times, we estimate the delayed-neutron background on the basis of the sum of six exponential terms.²⁴ However, even at shorter times the interaction of the thermonuclear neutrons with the material in the immediate vicinity of the explosion leads to reaction products which contribute to both the delayed-neutron and delayed γ backgrounds. Unfortunately, quantitative estimates of this latter effect cannot be made without detailed specifications of the device. For the time interval between 1 and 120 min the delayed- γ background decreases with time t approximately²⁴ as $t^{-1.05}$.

The delayed-neutron and delayed- γ backgrounds estimated in this way^{21-24,26} are given in Table II for a neutron-multiplicity counter with a 100 μ s gate that is located 5 cm from a point source. As discussed in Sec. III, a counter with these characteristics and a single-neutron detection efficiency $\epsilon = 0.46$ has already been developed for other purposes. (Although the gate for this counter could be reduced to at least 50 μ s and possibly to even 10 μ s with negligible effect on the single-neutron detection efficiency, we adopt 100 μ s as a conservative value. If a shorter gate is used, the delayed-neutron background per gating interval is reduced accordingly.)

We now present an example of how such an existing detector, which we stress has *not* been optimized for high neutron and γ backgrounds, might already be used for the detection of mass numbers up to about 265. As we will see, this should be possible provided the total lifetime $\Sigma\tau_\beta$ for all β decays before spontaneous fission in the 94-166 region is about 2 min or longer.

From Table II we see that for a recovery fraction $f = 10^{-13}$, the delayed-neutron background is down to one neutron per gating interval 5 min after the explosion, and drops rapidly with increasing time. The delayed- γ background after 5 min is only 2.4 R/h, which is well below the permitted level for this detector. For mass number $A = 265$, where $\bar{\nu} \approx 8$ and $N \approx 10^{16}$, about 100 events of high neutron multiplicity should be recorded provided that $\Sigma\tau_\beta \gtrsim 2$ min. The total number of counts can be increased significantly by increasing continuously the fraction f to the optimum size permitted by the backgrounds as time progresses.

Because most of the delayed neutrons, especially those in the long-lived component, are below 0.5 MeV in energy,^{22,24} the delayed-neutron background can be reduced by a factor of 2 to 3 by rejecting those events in which the neutron energy is below about 0.5 MeV. This can be achieved, of course, only at the expense of some reduction in single-neutron counting efficiency.

Further improvements can be made in the size gate and detection efficiency of the counter that we have been considering. But the key to substantial increases in detection possibilities for very heavy nuclei lies in the development of a neutron-multiplicity counter with a much smaller gate that at the same time can tolerate the fairly high γ background that is present. For example, for a counter with a 10 ns gate and for a fraction $f = 10^{-12}$, we see from Table II that the neutron background is down to 0.15 neutrons per gating interval after 1

min. However, at 1 min the γ background at 5 cm is 130 R/h. By the use of shielding and/or moving the detectors to a larger distance, it is possible to reduce this background below the level of about 0.1 R/h that can be tolerated with this type detector. We would then be able to record over 10^3 events for $A = 265$ provided that its lifetime $\tau_\beta \gtrsim 30$ s.

V. SUMMARY AND CONCLUSION

On the basis of the predicted increase in the average number of neutrons released per fission for very heavy nuclei, we have proposed the use of neutron-multiplicity counters to detect very heavy nuclei produced by prompt multiple-neutron capture in thermonuclear explosions. For an experiment with a neutron flux somewhat higher than that in Hutch, it should be possible to detect the production of nuclei with mass numbers up to about 265 by means of an existing neutron-multiplicity counter provided that β lifetimes add up to about 2 min or more. With a neutron-multiplicity counter that is designed specifically to reduce the effects of the delayed-neutron and delayed γ backgrounds, the lifetime limit could be reduced to about 30 s. Because of the relatively small fraction of the total debris that would be required (as small as 10^{-13}), an experiment of this type could be performed parasitically in connection with a thermonuclear explosion designed primarily for another purpose.

In addition to demonstrating the production of nuclei with mass numbers up to about 265 by prompt multiple-neutron capture, this type of experiment is valuable in testing two crucial theoretical predictions concerning heavy neutron-rich nuclei. The first involves the rate at which the nuclear surface energy decreases with increasing neutron excess, and the second involves the single-particle effects associated with the short spontaneous-fission half-lives in the 94-166 region. A test of these two predictions is crucial in making firmer theoretical calculations on such important questions as the likelihood of producing super-heavy nuclei by either multiple-neutron capture or in heavy-ion reactions.

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