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Synthesis of Neutron-Rich Nuclides

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Synthesis of Neutron-Rich Nuclides*

by

George A. Cowan

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SYNTHESIS OF NEUTRON-RICH NUCLIDES

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ABSTRACT

The technique of multiple neutron capture in nuclear explosives is compared with alternative methods for the production of heavy elements. Some recent data from the "Cyclamen" experiment are cited to summarize the present state-of-the-art and prospects for further progress are reviewed.

Introduction

Thermonuclear explosions in which no significant amount of neutron-absorbing material is present generate very large neutron exposures (time-integrated flux); they are capable of manufacturing heavier multiple neutron capture products than have yet been synthesized. The effectiveness of this source was first demonstrated by the production of fifteen previously undiscovered nuclides (Table I), including two new elements, in the debris from the "Mike" device in 1952.[1,2] Subsequent progress in the detailed understanding of the processes involved has encouraged the A.E.C. and scientists at the Livermore and Los Alamos laboratories to conduct, over the past ten years, a number of small-yield thermonuclear experiments designed to improve on the "Mike" source. The design efforts have been under the general direction of Carson Mark at Los Alamos and David Dorn at Livermore.

TABLE I. NEW PRODUCTS IDENTIFIED IN THE "MIKE" THERMONUCLEAR DEBRIS

Pu:	244, 245,* 246
Am:	246
Cm:	246, 247, 248
Bk:	249
Cf:	249, 252, 253, 254
Es:	253, 255
Fm:	255

*^{245}Pu was identified in 1954.

The first experiment of this kind, using a neutron source specifically designed to produce neutron-rich nuclides, was conducted at Eniwetok in 1958. With the beginning of underground tests in 1961, more experiments were planned, and, to date, 15 underground experiments have been conducted by the two laboratories. Analysis of the debris and interpretation of the data have been carried out collaboratively by nuclear chemists and physicists at Argonne National Laboratory, Lawrence Radiation Laboratory at Berkeley and Livermore, and Los Alamos Scientific Laboratory. Although the individual experiments have met with varying degrees of success, the neutron exposure has been steadily increased (Fig. 1). The last experiment in this series ("Cyclamen," May, 1966, 11-kt yield) produced a neutron exposure twice the size of that observed in 1952, at 10^{-3} the energy yield.[3] A small target of ^{238}U was immersed in this source; at this exposure level, new heavy element species, in particular, ^{258}Fm, would have been observed had the

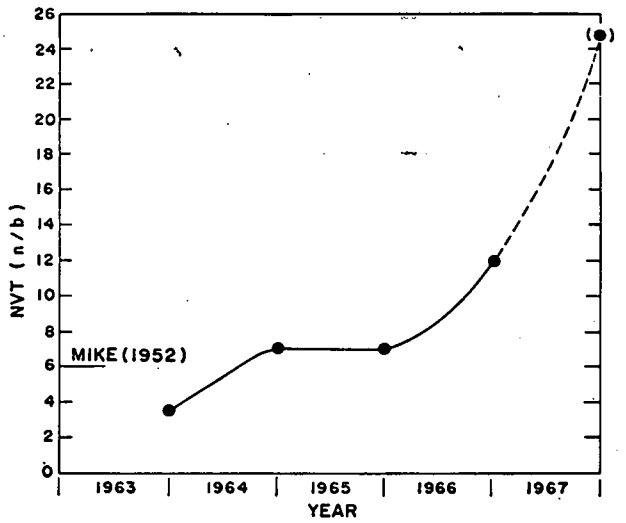


Fig. 1. The improvement in thermonuclear neutron sources with time.

decay properties accorded with some previous predictions. That neither ^{259}Fm nor ^{261}No was identified is probably related either to the fact that lifetimes for spontaneous fission and alpha decay in these species are either longer or much shorter than predicted earlier from extrapolation of known half-lives or to the unexpectedly short spontaneous fission lifetimes of species in the long beta-decay chains leading toward the line of beta stability.

In this review we briefly compare the technique of multiple neutron capture in nuclear explosives with other experimental methods for the production of neutron-rich nuclides, summarize the present state-of-the-art with relevant data from "Cyclamen," and offer some comments on future prospects for heavy element synthesis by neutron capture.

Techniques for Production of Heavy Elements

1. Charged-particle bombardment

Historically, the most successful process for making new elements has been bombardment of a suitable target with α particles.[4] In recent years, as the masses of the known elements have moved past the region accessible by this technique, it has become necessary to use heavier ions (e.g., $^{12,13}C$, ^{22}Ne) as projectiles. With the help of new machines, it should be possible to attempt the synthesis of superheavy products by bombardment of any element in the periodic table with any other element.[5]

Charged-particle bombardments of an element such as carbon or neon generally produce products on the neutron-poor side of beta stability so that the product of highest Z is made first, followed by the growth of daughter products of lower Z as the initial product decays by alpha or positron emission, electron capture, or spontaneous fission. Where a neutron-rich target becomes available in sufficient quantity, it may be possible to use this technique to produce the same heavy nuclides as would be produced by neutron capture (e.g., $^{250}Cm + {}^{13}C \rightarrow {}^{261}No + 2n$). However, in general, the mass region at a given Z available for exploration by charged-particle bombardment is distinctly different and lower than the mass region at the same Z which is open, in principle, to investigation by multiple neutron capture.

Charged-particle bombardment will probably continue to be the most useful and successful method for making new elements. It should continue to build elements beyond the presently known species unless the half-lives of the products become too short to permit identification.

2. Multiple neutron capture in reactors

Neutron bombardment of heavy elements in reactors produces nuclides close to the line of beta-stability. The successive capture products move toward the neutron-rich side as buildup proceeds during prolonged bombardment in high-flux reactors.

Usually several mass numbers of the same element are formed at even-Z species (Pu, Cm, Cf) and very few at odd-Z species. When sufficiently neutron-rich to decay by beta emission, these capture products decay to daughters of higher Z. If all the heavier products decay very quickly by alpha-particle emission or by spontaneous fission, they cannot be transformed significantly to a higher mass number by neutron capture, and the process of element-building is effectively terminated. Apparently this limit has been reached at mass 258. According to measurements by Hulet et al.,[6] ^{258}Fm decays so quickly, probably by spontaneous fission, that it blocks further neutron buildup. It seems unlikely, therefore, that elements beyond fermium will be synthesized in detectable quantities by this technique. The program will, however, make large amounts of

heavy target materials available for heavy ion bombardment. It is possible that some of the heavy products will be useful as target materials for further multiple neutron capture in thermonuclear sources. This question will be discussed later.

3. Prompt multiple neutron capture

Since neutron capture in explosions occurs on a time scale which is orders of magnitude shorter than the time scale for the most energetic beta decay, we choose to call it "prompt" capture. Such capture differs from thermal capture in reactors in the following ways:

a. Although most captures occur under "thermal" conditions in the source, "thermal" in an explosion is in the keV energy region, and equilibrium conditions are never realized. The capture cross sections behave more systematically than those in thermal reactors since they are averaged over resonance structure and can be calculated from statistical theory.(7)

b. All captures occur in a target which has no time to change significantly in Z by beta decay. The target may be initially changed in Z by reactions with high-energy neutrons or recoiling charged particles, but, during the subsequent history of neutron capture, higher mass numbers are synthesized at a constant proton number. As a result, all long-lived species of a given element occur in ratios determined almost entirely by the exposure and capture cross sections of the parents. These ratios are not affected significantly by competition with beta decay or, presumably, spontaneous fission. Thus, in contrast to thermal reactors, the ratio of ^{244}Pu to lower mass plutonium isotopes is not decreased by the short half-life for beta decay in ^{243}Pu. Similarly, the ratio of ^{250}Cm to ^{248}Cm and of ^{254}Cf to ^{252}Cf is much higher in prompt capture targets than in reactor targets.

As with neutron capture in reactors, element-building by prompt capture has not yet proceeded past ^{257}Fm, not because ^{258}Fm is very short-lived (here the β-decay half-life is unimportant) but for another reason to be discussed later in this paper, as will be the question of fissionability of the neutron-rich species of the target during neutron capture.

The process of prompt multiple neutron capture may be described as occurring when target atoms are immersed in a sea of neutrons or a neutron "gas." The atoms and neutrons remain in contact until nearly all of the atoms have captured neutrons. The amount of the various capture products can then be calculated from a series of differential equations of the form

$$dN_i = N_{i-1} nv\sigma_{c(i-1)} dt - N_i nv\sigma_{d(i)} dt.$$

This problem has a simple solution if all the cross sections are equal. The solution is the Poisson distribution: $N_i/N_o^o = x^i e^{-x}/i!$ All the cross sections are not equal, however, and the actual abundances are calculated by computer methods. Nevertheless, it is useful conceptually to regard the process as resulting in a Poisson distribution in mass numbers in which x is equal to nvtσ and is the average number of neutrons captured per nucleus.

An interesting property of the Poisson distribution is that if the flux is doubled, resulting in a doubling of x from, say, 3 to 6, the fraction of target which captures 20 neutrons increases from 10^{-11} to about 10^{-8}; that is, the yield increases about 10^5 for this mass number when the source is improved by a factor of two. In the more rigorous calculation the increase is about 10^4, the less favorable factor being due to a decrease in capture cross sections with increasing neutron number.(8)

In experiments of this type, we are interested in neutron exposures such that x is much greater than 1, say 10, which permits the production of detectable quantities of products which have captured 25 neutrons or more.

One may think of the quantity x as equal to $\sigma v \int n(t)dt$, where σv is, to a first approximation, a constant (if fission channels are closed) and is equal to about 10^{-16} cm^3/sec for many heavy elements. In the very high flux of HFIR, the neutron density, n, is about 10^{10}/cm^3. Thus, if we wish x to be equal to 10, t in the reactor must be about 10^7 sec, contrasting to an exposure time in a thermonuclear device of $<10^{-6}$ sec.

In a thermonuclear explosion one creates a neutron gas at much higher densities and then attempts to confine the gas for a "long" time in order to maximize the product $\int n(t)dt$. The principle is

simple but the techniques are not, and, since the techniques become involved with classified design information, the source is not described further. It is possible, however, to consider the basic problems without a detailed description of the neutron source.

The "Cyclamen" Experiment

When a nuclear explosion occurs underground, about 1 kt of soil is melted per kt (H. E. equivalent) of energy released. The yield of "Cyclamen" was 11 kt. A corresponding number of kilotons of soil melted, flowed to the bottom of a momentarily standing cavity to form a puddle, and was covered by the collapsing cavity. After reentry by drilling, which takes approximately 20 hr at this depth (~ 1000 ft), samples containing about 10^{-10} debris per gram of soil were brought up in special sampling tools. When a few kilograms of this material were recovered, samples were flown to Los Alamos, Argonne, Berkeley, and Livermore where they were dissolved and prepared for various kinds of chemistry.

The most important sample was the one on which actinide chemistry was performed since it contained all the new heavy-element nuclides through Z = 103. At Los Alamos, the soil was dissolved in HF and HNO_3, and the actinides were removed in an insoluble fluoride fraction; the fluorides were dissolved and extracted with tri-butyl phosphate, back-extracted, and reextracted with HDEHP (another phosphate ester), separated from lanthanides on an alcoholic HCl ion exchange column, and separated into the individual actinides on a cation column with hot α-HIB (isobutyric acid) eluant. This procedure took about 12 hr. Other samples were processed for elements 104 and 105, and still others were made available for the mass separator.

The principal actinide samples prepared at Los Alamos are shown in Table II. The chief point evident from inspection of this table is that although reasonable numbers of mass 259 atoms and some mass 261 atoms should have been contained in most of these samples, none were observed. The possibility of seeing masses 258 and 260 is discounted since, presumably, the spontaneous fission half-lives of these species are too short.

The entire "Cyclamen" mass distribution is shown in Fig. 2. The mass 259 and 261 predictions of the previous table are derived from Bell's calculations, as is the value of 12 n/b for the neutron exposure.[8]

TABLE II. CYCLAMEN SAMPLES

No.	Fraction	Separation Time	Atoms 257	Predicted 259	Predicted 261
1	4×10^{-12}	36 h	4×10^{2}	12	---
2	5×10^{-9}	58 h	5×10^{5}	1.5×10^{4}	50
3	2×10^{-8}	few d	2×10^{6}	6×10^{4}	2×10^{3}
4	5×10^{-8}	few d	5×10^{6}	1.5×10^{5}	5×10^{3}

One of the most interesting features of these curves is the alternating variation in yield, depending on whether the mass is odd or even. Up to about mass 250 the even-mass numbers are made in relatively larger yield as might be expected since the odd-mass species of uranium have capture cross sections about twice as large as the even-mass species. Above mass 250 this saw-tooth variation reverses, and the relatively larger yields occur at odd-mass numbers. Diamond and Fields of Argonne National Laboratory first suggested that this indicated a change in Z of the target nucleus. Bell[7] calculated the production of the odd-Z elements protactinium and neptunium in uranium targets and showed that it is entirely reasonable to assume that these odd-Z elements are responsible for the production of most of the heavier products. They are important because their average cross sections are several times larger than those of uranium, and even

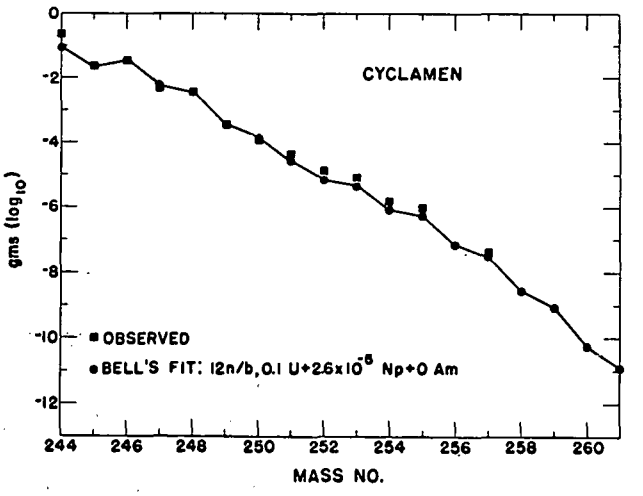


Fig. 2. The "CYCLAMEN" yield distribution.

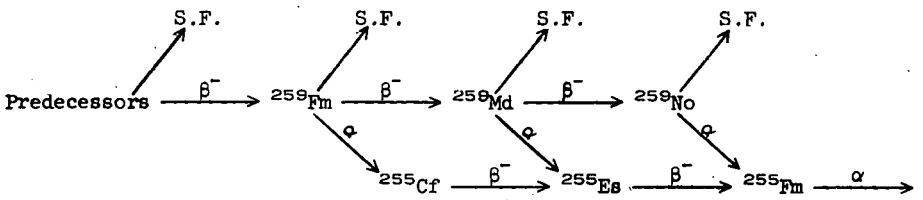
a 10^{-3} fraction of these atoms is sufficient to establish them as principal contributors to nuclides of mass >250.

For this reason we attempted in "Cyclamen" to make use of an odd-Z target, ^{243}Am, mixed with the ^{238}U target. There is no evidence, however, that it contributed significantly to the production of heavy elements. Calculations by Bell[8] indicate that the failure of americium to contribute is probably due to its excessive neutron-induced fissionability. The consequent depletion of the target material at several steps in the multiple capture chain quickly reduces the contribution of americium to a negligible level.

Conclusions

A discussion on the significance of our failure to find mass 259 follows.

The possible decay modes for the mass 259 chain are:



If spontaneous fission did not badly deplete the chain during β^- decay before ^{259}Fm, our limits on the spontaneous fission half-life for ^{259}Fm would be ≤5 hr or ≥7.5 yr. If, in fact, its spontaneous fission half-life is very long, we should have seen ^{259}Fm decay by α emission with a predicted half-life of 20 yr but we observe a limit on the α half-life of ≥100 yr. A long-lived β emitter should result either in the appearance of spontaneous fission activity from ^{259}Md or evidence of α emission together with the growth of an α group from ^{255}Fm which must be made by any α-emitting predecessor at ^{259}Fm, ^{259}Md, or ^{259}No. In nearly a year of counting the ^{259}Fm in the LASL sample, we have failed to see any significant growth of α particles from ^{255}Fm.

If fast beta decay has occurred in ^{259}Fm to ^{259}Md, the above limits on ^{259}Fm α and spontaneous fission decay do not apply, but we should have seen evidence for ^{259}Md. Our failure to see any such evidence corresponds to limits on the spontaneous fission half-life of ≤5 hr or ≥15 yr. The limit on the α-decay half-life is ≥200 yr.

We discount the possibility of β decay in ^{259}Md to ^{259}No.

The corresponding limits in mass chain 261, if it decayed to ^{261}Md, are ≤7 hr, or ≥0.5 yr for spontaneous fission and ≥2 yr for α decay. If the β-decay half-life of ^{261}Md is ≤9 hr, the chain could have gone to ^{261}No before our measurements began. The corresponding limits on ^{261}No would then be ≤9 hr or ≥1 yr for spontaneous fission and ≤14 hr and ≥1 yr for α decay.

Our observations do not exclude the following possibilities at mass 259:

1. Fast spontaneous fission in the mass 259 chain in competition with several successive β decays before ^{259}Fm.
2. Fast spontaneous fission at ^{259}Fm.
3. Long-lived ^{259}Fm.
4. Fast beta decay to ^{259}Md followed by fast spontaneous fission.
5. Long-lived ^{259}Md.

On the basis of decay systematics we tend to discount Nos. 2, 4 and 5. Eventually we hope to check on the existence of a nearly stable mass 259 by analysis of the fermium and mendelevium fractions in a highly sensitive mass spectrometer.

Perhaps the most likely hypothesis is No. 1 -- that fast spontaneous fission occurred before ^{259}Fm in one or more of the relatively neutron-rich members of the 259 chain. Consequently, so little mass ^{259}Fm was made that we failed to observe it. We hope in a next experiment to increase the amount of mass 259 by several thousand so that even if only a small fraction survives spontaneous fission competition to reach ^{260}Fm, it will be measurable.

As in the past, it is contemplated that all of

the nuclear data derived from the "Cyclamen" experiment, including results not referred to here, will be published by investigators at the four participating laboratories.

The chief problem in assessing prospects for the future is the lack of a good theoretical basis for predicting spontaneous fission half-lives. Although the several efforts presently underway to construct a more solid basis for calculating shell effects may prove helpful, the absence of a reliable model emphasizes the need for additional experimental data to establish the stability trend of the new products.

If the next experiment achieves the expected neutron exposure of 25 n/b, the distribution of products predicted by Bell[8] will be as shown in Fig. 3. In such an experiment, masses of 265 and, perhaps, even heavier masses will become detectable if they survive spontaneous fission for a few hours. If they do not, at least, sufficient ^{257}Fm will have been made to enable us to perform some new and interesting measurements that will provide information on decay systematics for species of mass up to 260.

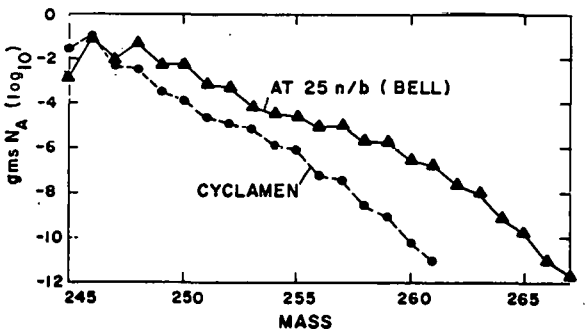


Fig. 3. A predicted yield curve at an exposure level of 25 n/b compared to "CYCLAMEN."

Separation of the actinides in a ton of debris would provide more than 10^{12} atoms of ^{257}Fm and permit measurements on products from some or all of the reactions indicated in Table III.

As a minimum result of the next experiment, new information on spontaneous fission half-lives should be obtained which will indicate whether heavier nuclides live long enough to be successfully identified in debris from prompt neutron capture experiments.

TABLE III. SOME POSSIBLE LABORATORY-PRODUCED NUCLIDES WITH ^{257}Fm TARGET

^{257}Fm:
$(n\gamma)^{258}Fm$
$(dn)^{258}Md$
$(d2n)^{257}Md$
$(tp)^{259}Fm$
$(tn)^{259}Md$
$(\alpha n)^{260}No$
$(\alpha 2n)^{259}No$
$(np)^{259}Es$

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