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Approximating the r-Process on Earth with Thermonuclear Explosions: Lessons Learned and Unanswered Questions

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Abstract

During the astrophysical r-process, multiple neutron captures occur so rapidly on target nuclei that their daughter nuclei generally do not have time to undergo radioactive decay before another neutron is captured. The r-process can be approximately simulated on Earth in certain types of thermonuclear explosions through an analogous process of rapid neutron captures known as the "prompt capture" process. Between 1952 and 1969, 23 nuclear tests were fielded by the US which were involved (at least partially) with the "prompt capture" process. Of these tests, 15 were at least partially successful. Some of these tests were conducted under the Plowshare Peaceful Nuclear Explosion Program as scientific research experiments. It is now known that the USSR conducted similar nuclear tests during 1966 to 1979. The elements einsteinium and fermium were first discovered by this process. The most successful tests achieved 19 successive neutron captures on the initial target nuclei. A review of the US program, target nuclei used, heavy element yields, scientific achievements of the program, and how some of the results have been used by the astrophysical community is given. Finally, some unanswered questions concerning very neutron-rich nuclei that could potentially have been answered with additional nuclear experiments is presented.

1.1 Introduction

Nuclear explosions have been used in a number of scientific investigations and application (see e.g. Dorn 1970). One such study was the Heavy Element Program conducted under the US Atomic Energy Commission's Peaceful Nuclear Explosion Program, Project Plowshare, which had as its objectives the production of heavy transuranic elements, and the investigation of the nuclear properties of very neutron-rich isotopes (Ingley 1969; Eccles 1970; Eberle 1972). The Soviet Union also conducted a similar research program (Adamskii et al. 1996; Nordyke 1998). For such experiments, an intense flux of neutrons is required, which is primarily produced by the D(T,n)^{4}He reaction, although the D(D,n)^{3}He reaction, the ^{9}Be(n,2n)2^{4}He reaction (if Be is present) and neutrons produced from fissions in the target nuclei also contribute. These neutrons are born at high energies (up to 14.1 MeV), and it is necessary to thermalize them to $10-20$ keV before significant multiple neutron captures can occur in target nuclei such as ^{232}Th, ^{238}U, and ^{242}Pu. Care must also be taken to minimize the effect of unwanted neutron absorbers like the ^{3}He(n,p)T reaction. The intense flux of neutrons is present only for 10 to 20 nanoseconds, and consequently, the target

Table 1.1. *Comparative Neutron Exposure Environments*

Type of Exposure	Flux ($n\bar{v}$)	Duration Δt	Fluence ($n\bar{v}\Delta t$)	Temperature
HFIR	5×10^{15}/cm^2 s	~ 0.5 yr	10^{23}/cm^2	2.5×10^{-5} keV
s-process	$\sim 10^{16}$/cm^2 s	$< 10^3$ yr	$< 10^{26}$/cm^2	10 to 30 keV
r-process	$> 10^{27}$/cm^2 s	1 to 100 s	$> 10^{27}$/cm^2	~ 100 keV
Prompt capture	$> 10^{32}$/cm^2 s	$< 10^{-7}$ s	$\sim 10^{25}$/cm^2	10 to 20 keV

nuclei undergo multiple neutron captures at constant atomic number, Z since the time span is too short to allow beta decay. However, at the same time, some daughter nuclei may experience fission or the (γ,n) reaction which interrupts the capture chain. A parallel capture chain may also be created on a Z–1 target through the (n,p) reaction. The rapid multiple captures of neutrons by target nuclei in a thermonuclear explosion has been given the name "prompt capture" (Cowan 1967). Once the "prompt capture" phase is over, the neutron-rich nuclei transform into longer-lived nuclei primarily through a series of beta decays back to the line of relative beta stability. For example: ^{257}U($\beta^-\bar{\nu}$) ^{257}Np($\beta^-\bar{\nu}$) ^{257}Pu… ^{257}Fm. The term "decay back" is used to refer to the series of beta decays to the line of beta stability experienced by a neutron-rich nucleus (Wene and Johansson 1974). The "decay back" process may be interrupted by other decay processes such as spontaneous fission, β-delayed fission, and β-delayed neutron emission.

Table 1.1 (modified from Eccles 1970) compares the environments for neutron capture of the astrophysical s- and r-process to their man-made analogues in the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory and the "prompt capture" process. For neutron capture in the HFIR and the s-process, the respective time scale for beta decay is much less than that for successive neutron captures, which results in the production of nuclei close to the line of beta stability. These two capture processes differ most with the neutron temperature, where the HFIR environment is much cooler than that of the s-process. In contrast, both the r-process and the "prompt capture" process undergo neutron captures so rapidly that multiple neutron captures can occur before the onset of beta decay, thus producing neutron-rich nuclei far from the line of beta stability. The r-process differs from the "prompt capture" process in that it takes place on a longer time scale and at hotter temperatures, and the (γ,n) reaction plays a bigger role. The net effect of these differences is that the target nuclei neutron capture cross sections are smaller for the r-process than for the "prompt capture" process and the longer time scale allows some beta decays to occur during the r-process at the same time neutron captures are still occurring. The similarities between the r-process and the "prompt capture" process are, however, sufficiently close that the "prompt capture" process can be thought as a fair approximation of the r-process, and data from a number of nuclear tests have been used to calibrate the nuclear physics used in r-process nucleosynthesis codes (see, e.g., Cowan, Thielemann, and Truran 1991).

1.2 Highlights of the Heavy Element Nuclear Test Program

Table 1.2 gives information about the nine most successful nuclear tests in the US Heavy Element Program. A total of 23 nuclear tests were involved with the program of which 15 tests were successful in producing neutron-rich isotopes. The fact that heavy transuranic isotopes could be produced in a thermonuclear device was discovered serendipitously through the analysis of the Mike test debris. All told, 15 new isotopes, shown in Table

Table 1.2. *Significant Nuclear Tests in the U.S. Heavy Element Program*

Event	Date	Yield	Fluence (moles/cm^2)	Target	Notes
Mike	10/31/52	10.4 Mt	2 to 3	^{238}U	1) 15 new isotopes discovered 2) 2 new elements (Es, Fm) discovered 3) 17 neutron captures achieved
Anacostia	11/27/62	<20 kt	$2.5-4$	^{238}U	Achieved Mike-like heavy element results, but with much less explosive yield
Par	10/09/64	38 kt	11	^{238}U	1) First test to outperform Mike heavy element results 2) ^{250}Cm discovered 3) 19 neutron captures achieved
Barbel	10/16/64	<20 kt	11	^{238}U	Duplicated Par results with different design
Tweed	05/21/65	<20 kt	12	^{237}Np, ^{242}Pu	Results not as good as Par, only 13 neutron captures achieved
Cyclamen	05/05/66	12 kt	18	^{238}U, ^{243}Am	1) Heavy element production exceeded Par and Barbel, but no new isotopes 2) 17% of the target converted to A$\geq$242
Kankakee	06/15/66	20-200 kt	12	^{238}U	Results similar to Par, different design
Vulcan	06/25/66	25 kt	12	^{238}U	Repeat of Tweed with different target, achieved Par-like results
Hutch	07/16/69	20-200 kt	$35-45$	^{238}U, ^{232}Th	1) Heavy element production exceeded Cyclamen, but no new isotopes 2) 19% of the target converted to A$\geq$242

Table 1.3. *New Isotopes Discovered in the Mike Debris*

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

1.3 (Diamond et al. 1960; Cowan 1967) and 2 new elements, einsteinium and fermium, were discovered (Ghiorso et al. 1955). A maximum of 17 successive neutron captures took place on some of the ^{238}U target nuclei, which became ^{255}U, and it underwent 8 beta decays to become ^{255}Fm, 20.1 h half-life. The estimated thermal neutron fluence on the target was 2 to 3 moles/cm^2.

The discoveries from the Mike test led to the development of the US Heavy Element Program conducted in part under the Peaceful Nuclear Explosion Program. Almost 10 years were to elapse before the results of the Mike test were approximately reproduced in the much lower yield Anacostia test (Hoff and Dorn 1964). The success of the Anacostia test showed that the program could be conducted in contained underground nuclear explosions (as opposed to an unconstrained atmospheric test) and at a nuclear yield much easier to deal with. New discoveries were made with the Par and Barbel tests (fielded respectively by the Lawrence Livermore Laboratory and the Los Alamos Scientific Laboratory), where both achieved an estimated thermal neutron fluence on the target of 11 moles/cm^2. The very neutron-rich isotope ^{250}Cm was discovered and 19 successive neutron captures were achieved on some of the ^{238}U target nuclei, which became ^{257}U, which later underwent 8 beta decays to become ^{257}Fm, 100.5 d half-life (Bell 1965; Los Alamos Radiochemistry group 1965; Combined Radiochemistry Group 1966). With the Tweed test, new target isotopes were tried to see if using an odd Z isotope (^{237}Np) or a heavier isotope (^{242}Pu) could produce better results than had been achieved with ^{238}U. Despite a higher estimated thermal neutron fluence of 12 moles/cm^2, the Tweed heavy element production was inferior to that of Par and Barbel. It was later determined that all of the ^{237}Np and most of the ^{242}Pu were fissioned by the high energy neutrons before the neutrons were thermalized. (At that time, the neutron cross sections for these two isotopes were not well known.) In spite of the target destruction, some of the ^{242}Pu survived to undergo 13 successive neutron captures to become ^{255}Pu, which later underwent 6 beta decays to become ^{255}Fm (Bell 1967a; Ingley 1969). The Tweed test was repeated with a ^{238}U target in the Vulcan test and the results were slightly better than for Par and Barbel (Ingley 1969; Eccles 1970).

The Cyclamen test achieved a significant increase in the thermal neutron fluence to 18 moles/cm^2 and its target nuclei were ^{238}U and ^{243}Am (an odd-Z isotope). Expectations were that isotopes with atomic mass, A of 259 and 261 would be produced and detected, and a heroic effort was made to get samples via drillback from the underground test to the laboratory in 36 hours. Unfortunately, no isotopes heavier than ^{257}Fm were detected, but a record 17% of the target isotopes were converted to isotopes with A$\geq$242. Most of the ^{243}Am was fissioned by the high energy neutrons before they were thermalized (Hoffman 1967; Ingley 1969). This was the last heavy element test conducted by the Los Alamos Scientific Laboratory. It is interesting to note that the most successful reported heavy element experiment conducted by the Soviet Union took place with the July 29, 1966 Zond test, where the target nuclei were also ^{238}U and ^{243}Am. The Zond results were somewhat similar to those of Anacostia with ^{252}Cf being the heaviest isotope produced, representing 14 neutron captures (Adamskii et al. 1996). The Kankakee test, with a different design concept, achieved results similar to that of Par and Barbel (Ingley 1969; Eccles 1970). A final attempt by the Lawrence Livermore Laboratory to produce isotopes with A$>$257 was made with the Hutch test. With a predicted thermal neutron fluence of 35 moles/cm^2 and target nuclei of ^{232}Th and ^{238}U, it was expected that isotopes out to A=265 would be produced and detected. Unfortunately, the fact that drillback samples did not reach the laboratory for analysis until after 7 days did not help, and as was the case for Cyclamen, no isotopes heavier than ^{257}Fm were detected, but a record 19% of the target was converted to isotopes with A$\geq$242. The ^{232}Th target was not very susceptible to fissioning from the high energy neutrons, but it showed evidence of at best, only 13 neutron captures to ^{245}Th, which later underwent 6 beta decays to become ^{245}Cm (Eccles 1970; Hoff and Hulet 1970; Eberle 1972). The Hutch subsidence crater at the Nevada Test Site is shown in Figure 1.1. The inability to produce new isotopes heavier than ^{257}Fm eventually led to a loss of interest in the Heavy Element Program and there were no further tests conducted by the US.

1.3 Target Nuclei

The ideal target nucleus for the "prompt capture" process should have its neutron capture cross section exceed its neutron induced fission cross section for all neutron energies.

Fig. 1.1. A picture of the Hutch subsidence crater at the Nevada Test Site. Deep below the surface lies the greatest concentration of ^{250}Cm on Earth.

In addition, the progeny of the target should have the same properties. In reality, no nuclei are known with these exact properties, but some come close enough in that a significant fraction are able to survive the initial presence of high energy neutrons and that the surviving nuclei can then undergo multiple neutron captures once the neutrons are thermalized. The isotopes ^{232}Th, ^{238}U, ^{237}Np, ^{242}Pu, and ^{243}Am have been used as targets in the US Heavy Element Program with different degrees of success.

The most successful results have been accomplished with ^{238}U, which has been used in the majority of the Heavy Element tests. In 6 such tests, 19 neutrons captures were achieved, and for Cyclamen and Hutch respectively, 4% and 8% of the initial ^{238}U was converted into isotopes with A$\geq$244. Experimental results show that the uranium capture cross section does suffer some thermal fission losses at atomic number 239, 241, 249, and 251 (Ingley 1969; Eccles 1970).

Both ^{232}Th and ^{242}Pu were used once, and a fraction of each were able to experience 13 neutron captures. ^{232}Th and its daughters showed the least sensitivity to fission, but the capture chain was apparently severely blocked at atomic numbers 242 and 244, where the neutron capture cross section became very small (Eccles 1970). In contrast, ^{242}Pu was very susceptible to high energy neutron induced fission and only about 0.3% of it was converted to neutron-rich isotopes. In addition, for Pu there was significant thermal fission depletion at atomic numbers 243, 245, 247, 249, 251, and 253 (Bell 1967a; Ingley 1969).

The isotopes ^{237}Np and ^{243}Am are the only odd Z isotopes tested in the Heavy Element Program and they produced disappointing results. At the time they were fielded, their cross sections were not well determined and it was not known that they did not come close to meeting the definition of ideal target nuclei. The initial high energy neutron population fissioned all of the ^{237}Np and almost all of the ^{243}Am targets. A tiny trace of the ^{243}Am survived to experience up to 2 neutron captures to become ^{245}Am, which later underwent 1 beta decay to become ^{245}Cm (Hoffman 1967).

Other target nuclei, such as ^{226}Ra, ^{231}Pa, and ^{252}Cf have been suggested but not used (Hoffman 1967; Seaborg 1968; Eccles 1970). Other isotopes could be considered, but only the ones listed above have the advantage of being potentially available in macroscopic quantities. Both ^{226}Ra and ^{231}Pa offer the possibility of seeing if more than 19 captures can be achieved, since there is plenty of room for captures before the "apparent" A=257 barrier. In addition, ^{231}Pa provides an odd Z isotope to study that is likely to survive the high energy neutron phase. ^{226}Ra would be a challenging sample to work with because of its radiological properties, but it is not unworkable. If it can survive the initial high energy neutron phase, ^{252}Cf might offer the possibility of having multiple captures on it to break the "apparent" A=257 barrier.

1.4 Heavy Element Yields

Figure 1.2 shows the isotopic abundances of the transformed target nuclei as a function of atomic mass for three landmark experiments, Par, Cyclamen, and Hutch (Ingley 1969; Hoffman and Hulet 1970). The heavy element production curves for all tests except for Hutch show similar behavior in that isotopes with A<250, the odd nuclei are less abundant than the even nuclei, but for A>250, this behavior is reversed. The initial behavior of the abundance curves to A=250 can be understood from the fact that for even Z nuclei (like U), because of pairing effects, the neutron capture cross section is larger when there are an odd number of neutrons in an isotope, compared to the case where there are an even number of neutrons. There are two competing theories to explain the behavior of the abundance curves for A>250. The odd Z hypothesis argues that a parallel capture chain on Pa is established through the reaction ^{238}U(n,p)^{238}Pa (Bell 1967b) during the high energy neutron phase. Calculations show that if 10^{-5} to 10^{-2} of the initial target nuclei are converted into odd Z nuclei, the relative abundance behavior for A>250 could be explained (Ingley 1969; Eberle 1972). The other theory proposes the phenomena of β-delayed fission for reversal of the odd-even abundance pattern (Wene and Johansson 1974). β-delayed fission was proposed after the last US Heavy Element test and it is different from spontaneous fission. It occurs when the Q value for beta decay becomes so large that the nucleus can decay into an excited state whose energy is comparable to the fission barrier, which allows the excited state the possibility of fissioning before it decays by another channel (such as another beta decay). Wene and Johansson argue that β-delayed fission would affect even-even nuclei worse for A>250. Both theories predict that the point where the reversal of odd-even behavior in the abundance distribution occurs would shift to a larger atomic mass as the neutron fluence increases, which is consistent with the Hutch curve. At present, both theories do about equally well in explaining past results (Hoff 1986).

Figure 1.2 also shows that as A increases, the relative abundance of an isotope decreases exponentially, but that the rate of decrease is affected by the size of the thermal neutron fluence on the target. Hutch target nuclei experienced about twice the neutron fluence of Cyclamen (35−45 versus 18 moles/cm^2), yet it produced (after adjusting for different target masses), over 120 times more ^{250}Cm and over 1600 times more ^{257}Fm. The abundance sensitivity to neutron exposure is shown even more clearly in Figure 1.3 (Eccles 1970). Hutch produced about 50 mg of ^{250}Cm, most of which is still underground underneath the crater in Figure 1.1.

1.5 Scientific Achievements

The US Heavy Element Program was responsible for the discovery of 2 new elements and 16 new transuranic isotopes (see e.g., Hoff 1978). The "prompt capture" process proved to be an efficient method of production for very neutron-rich isotopes like ^{250}Cm,

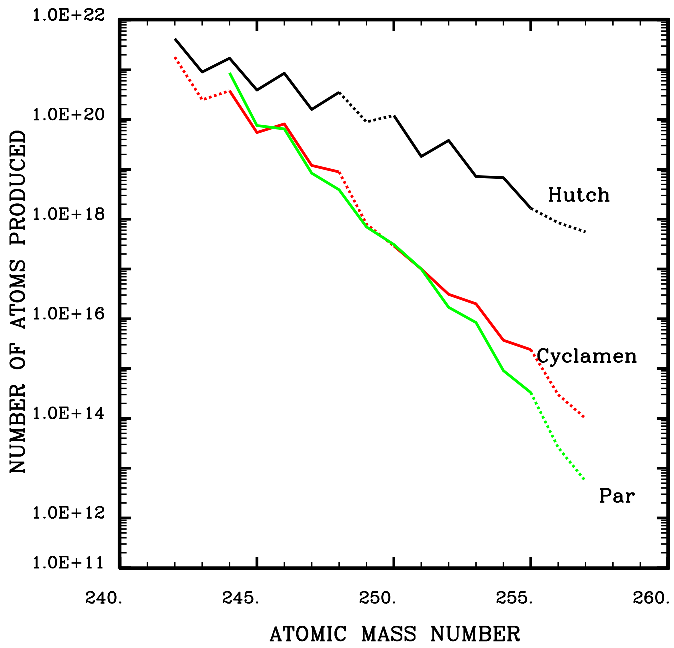

Fig. 1.2. Isotopic abundances of the transformed target nuclei as a function of atomic mass for the Par, Cyclamen, and Hutch tests. A dashed line is used when an isotope was not directly detected between two detected isotopes.

^{254}Cf, and ^{257}Fm, which are difficult to make in quantity by any other methods (Hoff and Hulet 1970). Two of these isotopes (^{250}Cm and ^{251}Cf) have half-lives of sufficient length (9700 y and 900 y, respectively) that it still would be feasible to extract new samples from the Hutch and Cyclamen test sites. Detailed studies were performed on the nuclear structure and properties of many of the isotopes that were extracted; sufficient quantities of ^{250}Cm and ^{257}Fm were obtained to allow them to be used as targets in accelerators. For example, the study of ^{258}Fm produced from Hutch ^{257}Fm showed that it decayed much faster than expected (0.37 ms half-life versus a predicted 2 hr half-life) by spontaneous fission (Hulet et al. 1971). The nuclear properties of the short-lived isotopes in the U and Pu "prompt capture" chains were deduced from the relative abundances of the "decay back" products. Neutron capture cross sections, fission to capture ratios, and information about the size of the fission barrier were deduced (Ingley 1969; Eccles 1970; Eberle 1972; Wene and Johansson 1974; Cowan, Thielemann, and Truran 1991). The Heavy Element Program remains the only man-made probe of nuclear properties far into the neutron-rich region of the transuranic elements, and it is the only available direct experimental test for theoretical predictions of β-delayed fission (Hoff 1987).

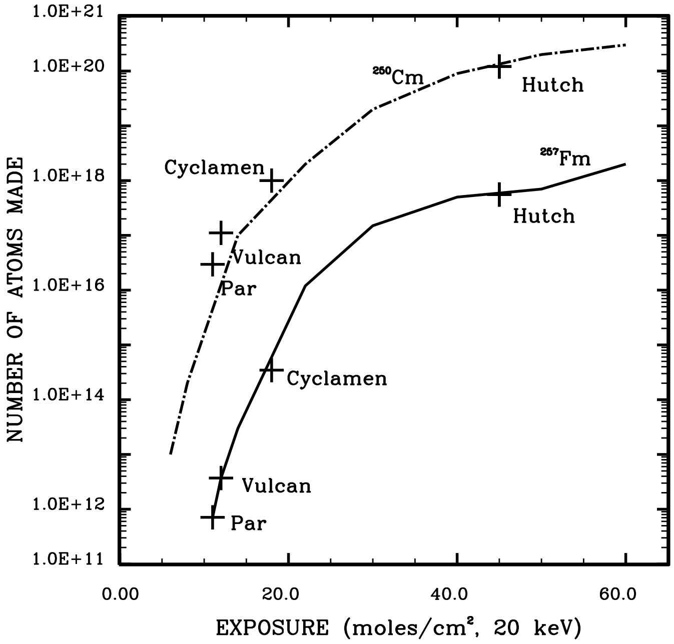

Fig. 1.3. Amounts of ^{250}Cm and ^{257}Fm produced in various nuclear tests as a function of neutron exposure on a ^{238}U target. The amount produced is normalized to the ^{238}U Hutch target (4.5×10^{22} atoms). The curves are predictions for a ^{238}U target.

1.6 Astrophysical Applications

The “prompt capture" process has provided detailed information on neutron capture chains from seed nuclei of ^{238}U and ^{242}Pu, which can be used to calibrate r-process codes (see e.g., Cowan, Thielemann, and Truran 1991). For example, in the 1980’s, comparison between the “prompt capture” process data for U, and r-process code predictions, showed that for A in the 252 to 257 range, the effect of β-delayed fissions were seriously overestimated in the U decay chain. The effect of this overestimate in the calculations also resulted in serious overestimates of the Milky Way Galactic age based on abundance ratios of chronometric pairs ^{232}Th/^{238}U, ^{235}U/^{238}U, and ^{244}Pu/^{238}U (Hoff 1987). The solution to this conflict was to raise the fission barrier of these neutron-rich nuclei which then reduced the effect of β-delayed fission and allowed agreement between the “prompt capture” process data and r-process calculations (Cowan, Thielemann, and Truran 1991).

1.7 Unanswered Questions

Although the Heavy Element Program has had many accomplishments, it did leave behind a number of unanswered questions, which unfortunately can only be answered by future nuclear experiments. One such question, as discussed in section 1.4, is the change in the abundance curve between the relative abundances of the even and odd isotopes for

A$>$250 due to the odd Z parallel capture chain hypothesis, the effect of β-delayed fission, or a combination of both? A test using the odd Z isotope ^{231}Pa as the target nucleus could potentially answer this question as the two theories predict different results. Another question is whether the isotope ^{257}Fm is as far as one can go with the "prompt capture" process? For the Cyclamen and Hutch tests, expectations were that nuclei perhaps out to A=265 would be produced, and if their half-lives were greater than 12 hr, they could be detected, yet nothing new was found. We now know that our inability to detect new isotopes in this mass range is due to the very short spontaneous fission half-lives of the daughter isotopes near the end of the "decay back" process. For example, ^{260}Fm spontaneously fissions with a 4 ms half-life. If it had instead beta-decayed to ^{260}Md which has a 32 d half-life, it would have been detected. The neutron-drip line for U is calculated to be at about A=277 (Möller 2003) and captures past A=257 should be possible. An improved Hutch design with a significantly increased thermal neutron fluence on the target might be able to drive the capture chain past A=265 and allow the "decay back" process to reach a portion of the line of relative beta stability that might have longer half-lives due to such effects as a possible neutron shell closure or getting to the vicinity of the "island" of relatively stable superheavy elements. During the "decay back" process from this extended capture chain, the isotopes would still have to survive possible destruction due to the effects of β-delayed fission or β-delayed neutron emission. Whether a super Hutch-like experiment would actually produce new detectable neutron-rich nuclei remains an interesting question to debate, and one that cannot be resolved in the current era of no nuclear test experiments.

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