PRODUCTION OF BERKELIUM AND CALIFORNINIUM

by

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ABSTRACT

By the end of 1974 about 2 grams of $^{252}$Cf and 0.2 grams of $^{249}$Bk had been produced in the high flux reactor programs at Savannah River and Oak Ridge and similar quantities of these nuclides were under irradiation. Production methods by accelerators and thermonuclear explosions were also well developed, but have proved a more expensive route to those nuclides which are available from the reactors. Multi-kilogram quantities of berkelium and californium are an eventual possibility from burnup of the byproduct actinides from the power reactor fuel cycles.
INTRODUCTION

Certainly one of the most unexpected technological success stories of the twenty-five years which have followed the original discovery of berkelium and californium must be the transformation of these elements from invisible flyspecks at the forefront of a new technology to regular if not quite mundane articles of commerce. In this transformation production has risen from the few thousand atom levels described in the preceding talks to the milligram and then the gram levels, with kilogram levels for the power reactors a possibility which may even change the emphasis from allocating a rare resource to disposal and destruction of an unwanted surplus. This development has involved most of the world's nuclear sites, and my talk will be mainly historical in describing their various contributions. However, as I emphasize at the end of the talk, a continuing development can still be expected.

Slides 1 and 2 list all the known isotopes of berkelium and californium which have half lives of more than an hour, that is those isotopes which have been or might be the goals of major production programs. These slides also list the chief production routes to each isotope. All the routes basically involve increasing the atomic weight of a naturally occurring heavy nuclide such as $^{238}\text{U}$ by five to fifteen or more mass units through bombardment with nuclear projectiles. The atomic number is adjusted automatically, and some times inconveniently, by beta decay. Charged particle bombardments are the only route to the lighter, neutron-deficient isotopes such as $^{247}\text{Bk}$ and $^{246}\text{Cf}$, but the less expensive neutron bombardments are normally used for the heavier isotopes - including $^{249}\text{Bk}$ and $^{252}\text{Cf}$, the main production goals to date. Milking a radioactive parent such as $^{249}\text{Bk}$ or $^{254}\text{Es}$ is a particularly useful intermediate step for obtaining isotopes such as $^{249}\text{Cf}$ or $^{250}\text{Bk}$ in pure form, but must depend on prior irradiations to form the intermediates.
CHARGED PARTICLE PRODUCTION

I'm going to say relatively little about charged particle production of the heavy elements, mainly because of woeful ignorance on my part, but also because this technique has so far proven more useful for research and discovery than production. The best candidate isotope of berkelium and californium for charged particle production is $^{247}\text{Bk}$, at a 1380 year half life (1) the longest lived berkelium isotope, but one not formed by neutron reactions, which instead yield the 314 day $^{247}\text{Bk}$(2). In turn, the best approach to forming $^{247}\text{Bk}$ seems to be to bombard neutron product $^{244}\text{Cm}$ with alpha particles. Several interactions give the desired isotope, including $\alpha,p$ and $\alpha,n$ (with following $\beta$ decay) reactions on the $^{244}\text{Cm}$ and the appropriate $\alpha,pxn$ or $\alpha,xn$ neutrons on the higher curium isotopes (3). However, efficiencies are still very low. The main difficulty is the 24.5 Mev coulomb barrier to the alpha particles at the charged nucleus. Overcoming this barrier requires accelerating the alpha particles to 30-50 Mev, and, as shown by Slide 3, reaction cross sections are then very low, in millibarns, with numerous side reactions. Even though the desired reactions to $^{247}\text{Bk}$ are endothermic, with negative Q's around - 13 Mev, the accelerated alpha particles, once they are in the nucleus, contribute so much energy that fission and a variety of spallation neutron boiloffs can take place in addition to the desired reactions, which are mainly knock ons of a single neutron or proton from $^{244}\text{Cm}$. As a result costs are high and yields are low. Thus, even though the charged particle reactions are well developed and excellent facilities exist as the various cyclotrons, Van de Graaffs and Linacs, accelerator production of $^{247}\text{Bk}$ is calculated to cost several hundred million dollars or more per gram.
Neutron buildup of the heavy elements has the advantages that no coulomb barriers have to be overcome, that copious supplies of neutrons are available at comparatively low costs from nuclear reactors, nuclear explosions, radio-nuclide sources, and accelerators, and that much of the production effort can eventually be made a byproduct of power reactor operation. Hence costs are much lower than for charged particle reactions. However, it is only fair to point out that neutron production of berkelium and californium requires multiple neutron additions with losses at each step, so overall yields are not necessarily better than in a single-step charged particle reaction.

The main neutron routes for producing berkelium and californium are illustrated in Slide 4. The different neutron sources differ by orders of magnitudes in their neutron flux levels and reaction rates. Man-made neutron synthesis of berkelium and californium was started by the appropriately modest high flux reactor route in August 1952\(^{(4)}\). However, before the year was out the "Mike" thermonuclear explosion \(^{(5)}\) provided synthesis fluxes surpassing even those postulated for supernovae and the massive stars and quasars. Analysis at Argonne, Los Alamos, and Berkeley of the "Mike" debris promptly led to the discovery of \(^{249}\)Bk, \(^{249}\)Cf, \(^{252}\)Cf, \(^{253}\)Cf, and \(^{254}\)Cf.

After the "Mike" shot the thermonuclear explosion route continued to be exploited for transplutonium element production in a series of underground tests performed by the Livermore and Los Alamos Laboratories\(^{(6,7)}\). Slide 5 illustrates some of the microchemistry being performed with bulldozers and drilling rigs to obtain samples from these tests\(^{(8)}\).

In the thermonuclear explosions essentially the entire reaction must take place in less than \(10^{-6}\) seconds, by which time the device blows itself apart. Both theoretical considerations and the product yields show that there is no time for beta decay of the target nucleus in this short time period, so the initial reaction is straight out the single element line of increasing atomic
weight, with decay occurring only after the explosion. The beta decay products are listed along the top axis of Slide 6, which gives the production versus mass number for two of the shots, Hutch with a neutron exposure of 40 moles/cm$^2$ and Cyclamen with 15 moles/cm$^2$. The experimental points are very well calculated for an average neutron moderation to about 20 kev and for calculated reaction cross sections averaging about one barn. There is a noticeable nucleon spin pairing effect favoring production of nuclides with even neutron numbers in the original production chain. Higher neutron exposures favor higher neutron addition numbers and higher molecular weights, with, to a first approximation, the distribution of neutron additions following a Poisson distribution around the most likely neutron addition number. In studying possible californium production by this route, the developers calculated that a production regime could be developed to produce about 10g of $^{252}$Cf per shot from about a 1% conversion of a $^{238}$U target. However, even though the thermonuclear techniques are well developed, the recovery techniques for separating the desired products from the underground nuclear debris need much more work, and there is presently no such effort in progress.

**NEUTRON PRODUCTION IN REACTORS**

Thus while the "Mike" thermonuclear shot successfully upstaged the initial reactor efforts to produce berkelium and californium by neutron addition, the reactors have in fact rather cornered the business since that time. The by-now familiar nuclide path for the reactor irradiations is shown in Slide 7. With the fortunate (in terms of fission losses) partial exceptions of $^{241}$Pu and $^{249}$Bk, the irradiations follow the beta stability path in the often multyear reactor exposures. The additions occur easily except for partial holdups due to low cross sections at $^{246}$Cm and $^{248}$Cm. However, competition from fission reactions, particularly at $^{239}$Pu, $^{241}$Pu, $^{245}$Cm, $^{247}$Cm, $^{249}$Cf, and $^{251}$Cf consume most of the reaction intermediates. As shown by the slide, something like 1800 neutrons
and slightly under a thousand atoms of $^{238}$U target are required to produce one atom of $^{252}$Cf in a typical reactor irradiation of this heaviest naturally occurring nuclide. Slide 7 incidently is one which I have used frequently, the only trouble being that each time I use it I have to redo it because the cross sections have changed again. This time is no exception, although the changes were generally small. As the result of cooperative work at Argonne, Los Alamos, Oak Ridge and Savannah River, the first full multigroup cross section set for this nuclide path had just become available; it will be presented by Savannah River at the Conference on Nuclear Cross Sections and Technology in Washington next March.

The possibility of reactor production of high atomic number actinides was realized even before the onset of reactor operations, so a major interlaboratory program was undertaken to accomplish it as soon as the achievement of irradiation fluxes in excess of $10^{14}$ n/cm$^2$-sec in the Materials Testing Reactor (MTR) at Arco, Idaho offered reasonable conversion rates$^4$. The rather unusual irradiation specimens, aptly called napkin rings, are illustrated in Slide 8$^9$. About 100 of them were fabricated at the Argonne National Laboratory from plutonium-aluminum alloys prepared at Los Alamos. The plutonium alloys were fully clad with aluminum and the rivets inserted in the corners to act as spacers for cooling water flow. Samples of these irradiation specimens were widely distributed to Los Alamos, Berkeley, Knolls, Chalk River, and Harwell as well as being processed at Argonne. In the seven year program from 1952-1959 the MTR produced about 30 µg of $^{252}$Cf as well as smaller amounts of $^{249}$Bk and the other californium isotopes. This material proved of sufficient interest that larger scale production programs were then undertaken at Savannah River and Oak Ridge.

Slide 9 shows all the major transplutonium production programs undertaken to date including the invididual thermonuclear shots and the earlier MTR programs. The large scale production got underway in June 1959 with plutonium irradiations
in the production lattices of the Savannah River reactors. Two separate campaigns were run, Transplutonium-I and Transplutonium-II, which between them produced 930g $^{242}\text{Pu}$, 300g $^{243}\text{Am}$, and 330g $^{244}\text{Cm}$ (10). At that time, it was thought that the actual production of berkelium, californium and the other higher actinides from these intermediates would be carried out almost exclusively at Oak Ridge in the new High Flux Isotope Reactor, or HFIR, then under design, and in fact the Transplutonium campaign nuclides did eventually provide the startup targets for the HFIR. However, before that startup took place, the Transplutonium program received a major boost from an independent SRP program to produce about 3 kg of $^{244}\text{Cm}$ for use in developing isotopic heat sources (10-13). This program was performed in two steps. The first step, Curium I, carried out in 1964, burned most of the fission heat from a plutonium irradiation lattice, and the second step, Curium II, carried out in 1965-1967, used a high flux lattice to transmute part of the reconstituted plutonium targets to $^{244}\text{Cm}$ and other higher actinides. Between the two Curium campaigns a High Flux Demonstration Lattice (14) run as a pilot for Curium II, irradiated about 525g of $^{242}\text{Pu}$ in fluxes up to $5.4 \times 10^{15}$ n/cm$^2$-sec. About 1 mg of berkelium and 11.8 mg of californium were obtained from these campaigns.

Meanwhile, the HFIR started up in September 1966 and has continued to operate ever since that time as the mainstay of the AEC's research production of actinides. I will return to the HFIR operation in more detail in a few minutes.

The Savannah River transplutonium programs were resumed in 1969 with a large scale program to make $^{252}\text{Cf}$ available to industry. This program used the residues of the Curium II operations in a new high-flux lattice, Californium I, to produce about 2 grams of $^{252}\text{Cf}$ in 16 months of operation (13). This material, plus that produced in HFIR by reirradiating Cf-I targets, presently constitutes the world's primary supply of californium. At the conclusion of Cf-I the targets were transferred to low flux positions in the regular Savannah River lattices where just
enough new californium is made to balance decay; the targets have been removed for reprocessing as the demand required. Approximately 0.75 g of $^{252}$Cf has been recovered to date from this material.

The Californium-I campaign was paralleled by a new plutonium-burning campaign intended to produce replacement $^{242}$Pu, $^{243}$Am, and $^{244}$Cm at considerably lower costs than in the Curium-I and II campaigns. The savings result from performing the irradiations at comparatively low fluxes over long time periods rather than in a crash high flux campaign. The products serve as a general purpose actinide stockpile as well as potential targets for berkelium and californium production.

Slide 10 is a drawing of a Savannah River reactor$^{(12)}$. Even though these reactors, designed back in 1952, are rapidly approaching great granddaddy status they have proved almost ideal for the transplutonium programs just because of their relative lack of modern specialization. Basically each reactor is simply a tank of heavy water about 18.5 feet in diameter with a 16 foot water depth. It is provided with heavy shielding, with an unusually flexible control system of some 427 separate control and safety rods, and with a powerful D$_2$O coolant flow. Lattice positions are large and well spaced from each other and can accommodate a variety of assembly types. This seemingly unsophisticated and unspecialized design puts very few constraints against the design of very sophisticated and very specialized lattices to perform specific transplutonium production tasks. An example is the high flux demonstration lattice illustrated in Slide 11$^{(14)}$ which still holds the reactor flux record of $6.1 \times 10^{15}$ n/cm$^2$-sec. In this lattice only the very center of the reactor tank is loaded with fuel and targets, and all the cooling flow is concentrated into this area so as to allow heat removal rates as high as $2.66 \times 10^6$ Btu/hr-ft$^2$. Under these conditions fuel life is very short, only a few days, but the reactor is provided with a rapid loading and unloading system which can changeover the lattice in one day or less.
The HFIR reactor, illustrated in Slide 12\(^{(16)}\) makes an interesting contrast to the Savannah River reactors. Also a very successful design, it is intended not as a large, general purpose reactor, but as a very small, single purpose irradiator specialized to the very particular task of transplutonium isotope production and research. Fluxes of up to \(5 \times 10^{15} \text{n/cm}^2\text{-sec}\) are achieved with a power of about 100 megawatts and a fuel loading of about 9.4 kg of highly enriched uranium, which lasts for about 23 days of operation. The target area is about 5 inches in diameter and the capacity is about 300 grams of target isotopes.

**CHEMICAL SEPARATIONS**

HFIR’s sister facility, the Transuranium Processing Plant or TRU, illustrated in Slide 13\(^{(17)}\), has perhaps been of even more importance than HFIR to the berkelium and californium programs, since it has been used not only to recover the HFIR production, but also the production to date from the Savannah River Cf-I lattice. Quantities of these materials recovered through 1974 are listed in Slide 14\(^{(18)}\). They amount to about 218 mg \(^{249}\)Bk and 2017 mg \(^{252}\)Cf as well as the other actinides through \(^{257}\)Fm. About half a gram of californium is now being produced each year, together with a tenth that quantity of berkelium.

Separations technology has been improving steadily throughout the transplutonium campaigns, and a large number of different processes have been used at various times at Oak Ridge and Savannah River as well as for the more specialized recovery programs at Argonne, Berkeley, Livermore, Los Alamos, and other sites. Slide 15 illustrates one of the latest flow sheets, that for the completed, but as yet unused Multipurpose Processing Facility (MPPF) at Savannah River. Like TRU this is a manipulator facility, and is tucked into unused space behind the 5.5 foot thick concrete walls of one of the SRP process canyons. Capacity is large, up to 260 mg of \(^{252}\)Cf at each manipulator facility at any one time. The separations are built around cation ion exchange chromatography.
with very finely divided resins to provide high separations factors. Pressurized columns are required for reasonable flow rates\(^{(20)}\).

If the original targets contain much plutonium, the first step is to remove this plutonium plus the nonlanthanide fission products by conventional solvent extraction. Then, after feed adjustment to the desired concentration and acidity (and the addition of NaNO\(_2\) to help complex ruthenium), the feed solution is loaded onto DOWEX 50W-X8\* resin saturated with a Zn\(^{2+}\) barrier ion, and elution is begun with 0.05M DTPA buffered to pH 6 with NH\(_4\)OH. In the presence of the DTPA chelating agent the resin has less affinity for zinc than for the actinides and more affinity for NH\(_4^+\) so the displacement development gives self-sharpening bands. Es and Fm, Cf, and Bk come off in that order in the Zn band, and there is good separation between this band and the following Cm/Am, lanthanide, and NH\(_4^+\) bands. With some recycle it is possible to obtain separate Es/Fm/Cf and Bk fractions at this time.

Final purification of the separate Cf and Bk fractions proceeds by essentially duplicate elution chromatography processes. First the DTPA is removed by conventional ion exchange, sorbing the transplutonium elements on the column, washing through the DTPA, and eluting the sorbed ions with 5 M HNO\(_3\). Next the actinides are separated from the zinc barrier ion introduced in the displacement chromatography step and from similar cationic impurities by carrier precipitation with lanthanum hydroxide, which is followed by redissolution in 3 molar nitric acid. Then the solution is loaded onto the NH\(_4^+\) form of DOWEX 50W-X8\* and eluted with 0.4 molar \(\alpha\)-hydroxyisobutyrate, varying the pH stepwise from 3.4 to 5.0 to vary the effective concentration of this complexing agent. The complexed actinides are bound to the column more tightly than the NH\(_4^+\) ion and are only slowly displaced by it. Four actinide fractions are obtained, Fm/Es, Cf, Bk and Cm/Am, in that order. Finishing of the individual actinide fractions

\* Trademark of the Dow Chemical Company
is accomplished by extracting them onto a resin column and then calcining the resin. In the case of californium, californium oxysulfate, \( \text{Cf}_2 \text{O}_2 \text{SO}_4 \), is obtained by calcining at 800°C and californium oxide, \( \text{Cf}_2 \text{O}_3 \), by calcining at 1250°C\(^{21}\).

One of the particular features of the MPPF is the MOSES computer control system for the ion exchange operations, where MOSES is an acronym for Monitoring On-Line System for Elemental Separations\(^{22}\). It is illustrated in Slide 15. Basically the MOSES system works by using gamma and neutron monitoring of shielded, completely contained loops from the process streams to determine the concentrations of the actinides of interest. These inputs are then fed to a PDP-9* computer for the actual process control. Three detector systems are used, \( \text{BF}_3 \) counters to measure neutrons from the spontaneous fissions of \( ^{252}\text{Cf}, ^{250}\text{Cf}, \) and \( ^{244}\text{Cm} \), a sodium iodide scintillation detector to monitor the 1.27 Mev gamma ray from \( ^{154}\text{Eu} \) and \( ^{160}\text{Tb} \), and a lithium-drifted germanium detector to give specific responses to the gamma photopeaks for \( ^{244}\text{Cm}, ^{243}\text{Am}, ^{154}\text{Eu}, ^{160}\text{Tb} \) and \( ^{144}\text{Ce} \). Two or more detectors detect all isotopes of interest except for \( ^{243}\text{Am} \) to which only the Ge (Li) detector responds. The usefulness of the system has been well demonstrated in the SRP curium programs, which included the separation of about 10 mg of californium.

FUTURE PRODUCTION

Changing the perspective now from the existing production programs for berkelium and californium to possible future production programs, the primary change is in the production starting point. The original programs had quite literally to start at square one with \( ^{238}\text{U} \). As we saw earlier, this involved multiple processing steps and the investment of hundreds or thousands of neutrons and target atoms for each atom of berkelium or californium produced. However,

*Trademark of the Digital Equipment Corporation
as shown by Slide 17, in making approximately 2 grams of $^{252}\text{Cf}$ available to date we have in fact also done most of the work towards producing an additional 50 grams or so of berkelium and californium by producing a high grade stockpile of curium intermediates. Expressing the irradiations in terms of gram atomic weights of neutron required, we see the existing stockpile has already absorbed 238 moles of neutrons and only about 7 more are required to turn it into about 50 grams of californium. Further, these high-grade targets are backed up by about 10 kg of americium and curium (unseparated) from a still continuing SRP irradiation program.

Over the still longer range the berkelium and californium will come from byproduct production in power reactor fuel cycles. Some years ago I surveyed this possibility in a paper grandiosely entitled Tons of Curium and Pounds of Californium. It turns out I was much too modest in my estimates.

As shown by Slide 18 the AEC predicts the power reactors to generate about $10^{10}$ megawatt days of heat between now and the year 2000\(^{(23)}\), and, even without breeder reactors, at least 0.5 grams of plutonium will be produced for each megawatt day. The total amount of plutonium produced between now and the year 2000 is thus predicted to be 5000 metric tons or more. It is assumed that eventually all of this plutonium will serve as reactor fuel, much of it by burnup in the same fuel element in which it is formed, the rest by chemical recovery and recycle into other reactors. However, as we saw earlier in Slide 7, about 7% of this plutonium will in the process transmute to americium and curium. Again part of this transplutonium residue will be burned as it is produced and part recovered, and again it is assumed that, in order to avoid possible escape of this very long lived activity into the biosphere, it will be necessary to recycle the recovered material for neutron burnup. Thus power reactor operation between now and the year 2000 carries an eventual commitment to burn up some 360 metric tons of americium and curium. From Slide 7, we find this process will produce 5 tons - not pounds - of berkelium and californium. While it is unlikely that more than a tiny fraction of this production can or should be recovered - after all the
basic purpose of the program is to burn out the actinides - its existence means that berkelium and californium deliveries can be set entirely by demand and not capacity restrictions. These demands, particularly for $^{252}\text{Cf}$, will be discussed in the next talk.
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23. USAEC Office of Planning and Analysis, WASH-1139(74) (1974)
<table>
<thead>
<tr>
<th>ISOTOPE</th>
<th>HALF LIFE</th>
<th>DECAY</th>
<th>CHARGED PARTICLE (ACCELERATOR)</th>
<th>NEUTRON</th>
<th>DECAY PARENT</th>
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<tr>
<td>$^{243}$Bk</td>
<td>4.6 h</td>
<td>EC</td>
<td>$^{241}$Am ($\alpha,2n$), $^{243}$Am ($\alpha,4n$), $^{242}$Cm (d,n)</td>
<td>-</td>
<td>-</td>
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<tr>
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<td>4.4 h</td>
<td>EC</td>
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<td>-</td>
<td>-</td>
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<td>4.98 d</td>
<td>EC</td>
<td>$^{243}$Am ($\alpha,2n$), $^{242}$Cm ($\alpha,p$), $^{244}$Cm (d,n)</td>
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<td>-</td>
</tr>
<tr>
<td>$^{246}$Bk</td>
<td>1.8 d</td>
<td>EC</td>
<td>$^{243}$Am ($\alpha,\beta$), $^{244}$Cm ($\alpha,pn$)</td>
<td>-</td>
<td>-</td>
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<tr>
<td>$^{247}$Bk</td>
<td>1380 y</td>
<td>$\beta$</td>
<td>$^{244}$Cm ($\alpha,p$), $^{245-6}$Cm ($\alpha,pxn$)</td>
<td>-</td>
<td>$^{247}$Cf</td>
</tr>
<tr>
<td>$^{248}$Bk</td>
<td>16 h</td>
<td>$\beta$,EC</td>
<td>-</td>
<td>$^{247}$Bk (n,γ)</td>
<td>-</td>
</tr>
<tr>
<td>$^{249}$Bk</td>
<td>314 d</td>
<td>$\beta$</td>
<td>-</td>
<td>Fuel Path</td>
<td>-</td>
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<tr>
<td>$^{250}$Bk</td>
<td>3.22 h</td>
<td>$\beta$</td>
<td>-</td>
<td>$^{249}$Bk (n,γ)</td>
<td>$^{254}$Es</td>
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### Calenornium Production Routes (Isotopes with Half Life >1 hr)

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<tr>
<td><strong>Isotope</strong></td>
<td><strong>Half Life</strong></td>
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<tr>
<td>246 Cf</td>
<td>35.7 h</td>
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<tr>
<td>247 Cf</td>
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<td>248 Cf</td>
<td>350 d</td>
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<tr>
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<td>352 y</td>
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<td>251 Cf</td>
<td>900 y</td>
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<td>254 Cf</td>
<td>60.5 d</td>
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<td>255 Cf</td>
<td>1.5 h</td>
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ACCELERATOR PRODUCTION OF $^{247}$Bk

REATIONS

$^{244}$Cm ($\alpha$,p) $^{247}$Bk

$^{244}$Cm ($\alpha$,n) $^{247}$Cf $\rightarrow$ EC $^{247}$Bk

$^{243+x}$Cm($\alpha$,xn) $^{247}$Cf $\rightarrow$ EC $^{247}$Bk

CROSS SECTIONS FOR $\sim$30 Mev $\alpha$ PARTICLES ON $^{244}$Cm

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<th>Cross Section</th>
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<tr>
<td>$\alpha$,2n</td>
<td>17 mb</td>
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<tr>
<td>$\alpha$,3n</td>
<td>5 mb</td>
</tr>
<tr>
<td>$\alpha$,4n</td>
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<tr>
<td>$\alpha$, p3n</td>
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### Neutron Flux Sources for Transuranium Synthesis

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<tr>
<th>Neutron Addition Process</th>
<th>Neutron Flux $n/cm^2$-sec</th>
<th>Reaction Time SEC</th>
<th>Exposure n/barn</th>
<th>Neutron Energy Ave kev</th>
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<tr>
<td>High Flux Reactor</td>
<td>$5 \times 10^{15}$</td>
<td>$10^6 - 10^8$</td>
<td>0.15</td>
<td>$\sim 2.5 \times 10^{-5}$</td>
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<tr>
<td>Steller R Process</td>
<td>$&gt; 10^{27}$</td>
<td>1-100</td>
<td>1000</td>
<td>$\sim 100$</td>
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<tr>
<td>Nuclear Explosion</td>
<td>$&gt; 10^{31}$</td>
<td>$10^{-6}$</td>
<td>50</td>
<td>$\sim 20$</td>
</tr>
</tbody>
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MASS YIELD CURVES IN THE HUTCH AND CYCLAMEN NUCLEAR EXPLOSIONS
Solid Lines are Results of Calculational Fits

\[ N = N_0 \frac{(\sigma \phi)^n e^{-\sigma \phi}}{n!} \]

- \( n \) = number neutrons added
- \( \sigma \) = cross section
- \( \phi \) = neutron exposure

HUTCH
40 moles of neutrons/cm²

CYCLAMEN
15 moles of neutrons/cm²
REACTOR PRODUCTION OF CALIFORNIUM

ATOMS REQUIRED/ATOM $^{252}\text{Cf}$ PRODUCED

1.0 AFTER CAPTURE AND DECAY

FISSION

$^{249}\text{Cf} \rightarrow ^{250}\text{Cf}$

$^{251}\text{Cf}$

$^{252}\text{Cf}$

FISSION

$^{249}\text{Bk} \rightarrow ^{250}\text{Bk}$

FISSION

$^{249}\text{Am} \rightarrow ^{250}\text{Am}$

FISSION

$^{239}\text{Pu} \rightarrow ^{240}\text{Pu}$

$^{241}\text{Pu} \rightarrow ^{242}\text{Pu}$

$^{243}\text{Pu}$

$^{238}\text{U} \rightarrow ^{239}\text{U}$

NEUTRONS REQUIRED/ATOM $^{252}\text{Cf}$ PRODUCED

$^{1799}$ $^{812}$ $^{566}$ $^{319}$ $^{248}$ $^{176}$ $^{105}$ $^{36.0}$ $^{25.7}$ $^{16.4}$ $^{12.5}$ $^{8.7}$ $^{5.0}$ $^{1.3}$ $^{0}$ $^{0}$

SLIDE 7
NAPKIN RING IRRADIATION ASSEMBLY
PRINCIPAL TRANSPLUTONIUM PRODUCTION PROGRAMS

Calendar Year

| Production Site          | 52 | 53 | 54 | 55 | 56 | 57 | 58 | 59 | 60 | 61 | 62 | 63 | 64 | 65 | 66 | 67 | 68 | 69 | 70 | 71 | 72 | 73 | 74 |
|-------------------------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|    |
| Materials Test Reactor |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
| Savanna River           |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
| Oak Ridge               |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |
| Thermonuclear           |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |    |

- Plutonium Irradiations
- Transplutonium I
- Transplutonium II
- Cm I
- Cm II
- High Flux
- Cf I
- Pu Burn
- High Flux Isotopes Reactor
- Mike
- Barbel
- Tweed
- Cyclamen
- Kankakee Vulcan
- Hutch
ORNL HIGH FLUX ISOTOPE REACTOR

TARGET RODS
(Targets 0.247"D. x 20"H)

BERYLLIUM REFLECTOR

BEAM TUBE

INNER CONTROL ROD

OLTER CONTROL ROD
(18.6" O.D.)

INNER FUEL PLATES
(5" I.D., 10" O.D., 20" H.)

OUTER FUEL PLATES
(11.5" I.D., 17" O.D., 20" H.)

FUEL CONSTRUCTION
<table>
<thead>
<tr>
<th>YEAR</th>
<th>$^{249}$Bk, mg</th>
<th>$^{252}$Cf, mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>1966</td>
<td>0.03</td>
<td>0.1</td>
</tr>
<tr>
<td>1967</td>
<td>1.0</td>
<td>5.3</td>
</tr>
<tr>
<td>1968</td>
<td>1.4</td>
<td>7.0</td>
</tr>
<tr>
<td>1969</td>
<td>3.2</td>
<td>22</td>
</tr>
<tr>
<td>1970</td>
<td>14.3</td>
<td>117</td>
</tr>
<tr>
<td>1971</td>
<td>50.5</td>
<td>403</td>
</tr>
<tr>
<td>1972</td>
<td>47.4</td>
<td>476</td>
</tr>
<tr>
<td>1973</td>
<td>62.0</td>
<td>586</td>
</tr>
<tr>
<td>1974</td>
<td>38.0</td>
<td>401</td>
</tr>
</tbody>
</table>

Total: 217.8 mg, 2017 mg
TRANSPLUTONIUM SEPARATIONS FLOW SHEET

TRANSFER SOLUTION FROM SRP SEPARATIONS PLANT OR MPPF DISSOLVER

PRESSURIZED ION EXCHANGE
DISPLACEMENT DEVELOPMENT

LANTHANIDES

Am-Cm

Cf,Es,Fm

Bk

DTPA

SELF RADIOLYSIS

HNO₃

H₂C₂O₄

PRECIPITATION

HNO₃

CALCINING

HNO₃

TARGET FABRICATION

DTPA - 0.05M Diethylenetriaminepentaacetic Acid
AHIB - Alphahydroxyisobutyrate
SRP PRODUCTION OF ACTINIDE INTERMEDIATES

<table>
<thead>
<tr>
<th>SRP ACTINIDE INTERMEDIATES</th>
<th>GRAMS</th>
<th>MOLES NEUTRONS INVESTED</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Cf-I</strong> ($\bar{A} = 244.84$)</td>
<td>2338</td>
<td>238</td>
</tr>
<tr>
<td><strong>Pu BURN</strong> ($\bar{A} = 243.3$)</td>
<td>9900</td>
<td>865</td>
</tr>
</tbody>
</table>

**252**$_{\text{Cf}}$ PRODUCTION

| SEPARATED TO DATE                   | 2.0   | 14.3                   |
| TO BE SEPARATED                    | 1.5   | 10.7                   |
| FROM Cf-I INTERMEDIATES            | 50.0  | 6.5*                   |
| FROM Pu BURN INTERMEDIATES         | 140.0 | 99*                    |

* ADDITIONAL IRRADIATION REQUIRED TO PRODUCE $^{252}$$_{\text{Cf}}$. 
\(^{252}\text{Cf}\) Production from Power Reactor Actinide Burnup

Expected Nuclear Power Generation in the USA through Year 2000

\[0.8 - 1.4 \times 10^{10}\text{ Megawatt Days}\]

(from AEC forecast of nuclear power growth, Wash-1139(74))

Expected Plutonium Generation in the USA through Year 2000

\[\sim 0.5 \times 10^{10} \text{g} \quad ^{239}\text{Pu}\]

(based on 0.5g/megawatt day in LWRs. Counts both recovered Pu and that burned in reactor before recovery)

\(^{252}\text{Cf}\) Production if require burnup of all actinides rather than waste disposal

5000 kg \(^{252}\text{Cf}\)

(from slide 7 - 1 atom of \(^{252}\text{Cf}\) will be produced from each 986 atoms \(^{239}\text{Pu}\) burned)