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HALIDE SLAGGING OF URANIUM-PLUTONIUM ALLOYS

by

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ABSTRACT

Nearly complete extraction of cerium (a stand-in for all highly electropositive fission products) from molten uranium-10 w/o plutonium-10 w/o fission alloys by molten halide salts has been realized within one hour at 1150°C. MgCl₂ in a CaCl₂-BaCl₂ flux was used to oxidize the cerium and thereby effect its transfer to the molten salt phase. The molten phases have been satisfactorily contained in a beryllia crucible. Flux and metal phases have been separated by cooling the system to about 860°C to solidify the metal phase, and then by pouring off the still-molten salt layer. The metal phase was subsequently remelted and poured into a different mold. An alternative procedure is to pour both phases together and subsequently break apart the solidified metal and salt phases.

Yields of purified metal ingots have been about 98%. When 110% of the amount of MgCl₂ required to oxidize all of the cerium to CeCl₃ was employed, 98% of the cerium was removed along with less than 0.5% of the plutonium. This represents a cerium-plutonium separation factor of about 7000. Very little plutonium (less than 0.1%) has been vaporized.

I. INTRODUCTION

The Chemical Engineering Division of Argonne National Laboratory has been working for a number of years on the development of pyrometallurgical processes for fast reactor fuels. To demonstrate the engineering feasibility of such processes and obtain cost information on them, they will be used to process fuel materials discharged from Argonne's Second Experimental Breeder Reactor (EBR-II). For this purpose, a processing plant known as the EBR-II Fuel Cycle Facility has been built next to the reactor. (1)

A melt-refining process* will be used to recover the first core loading of EBR-II fuel,⁽¹⁾ which is an enriched uranium alloy (46 w/o U²³⁵) containing about 5 w/o fissionium.**

After initial testing of the EBR-II reactor with the enriched uranium alloy, it is proposed that the reactor be loaded with a fuel containing plutonium as the fissionable material. This may be a metal fuel consisting of uranium containing 10 to 20 w/o plutonium and about 10 w/o fissionium. The melt-refining process to be used for recovery of the first core loading is also applicable to this fuel,⁽²⁾ but a halide-slugging procedure (in which rare earth and other highly electropositive fission products are selectively oxidized and extracted from molten fuel alloy into an overlying molten salt phase) offers some advantages. These advantages are (1) higher product yields (>98% versus 90 to 95%), (2) a lower operating temperature (1150°C versus 1400°C), and (3) a shorter processing time (1 hour or less versus 1 to 3 hours). The objective of the work reported herein was to investigate the separation of a typical rare-earth element (cerium) from plutonium by a halide-slugging procedure.

An indication that rare earth, alkaline earth, and alkali metals may be removed selectively from uranium and plutonium is shown by the standard free energies of formation of their most stable chlorides and fluorides given in Table I.⁽³⁾ Uranium and plutonium halides may be seen to be appreciably less stable than representative rare earth and alkaline earth halides (and also less stable than alkali metal chlorides, but not fluorides). The free energies of formation are only a qualitative indication that separations are possible since it is necessary to know activity coefficients of the species of interest in the molten salt and liquid metal phases in order to compute accurately the partition coefficients. The most pertinent deviations from ideality occur for cerium in uranium, and plutonium in uranium.^(4,5) Although the deviations for both of these elements are positive, the activity coefficient for cerium in uranium is larger than that for plutonium. Because of these deviations from ideality, the separation of rare earths from plutonium calculated solely from the standard free energies of formation may be considerably in error.

*In this process, clad, chopped fuel pins are melted in a lime-stabilized zirconia crucible, held at 1400°C for 1 to 3 hours, and poured into a mold to form an ingot which is subsequently used to prepare new fuel pins. While the fuel is molten, fission products are removed by (1) escape of noble gases, (2) volatilization of some elements, and (3) selective oxidation of the highly electropositive elements and deposition of them along the wetted crucible surfaces. Noble-metal fission products, such as molybdenum, ruthenium, and zirconium, are not removed and remain in the fuel as alloying elements.

**Fissionium is the name given to the mixture of noble-metal fission products that remain in the fuel as alloying elements. The estimated equilibrium concentrations of these were used in the preparation of the first core loading,⁽¹⁾ namely; molybdenum-2.5 w/o, ruthenium-2.0 w/o, rhodium-0.26 w/o, palladium-0.19 w/o, zirconium-0.1 w/o, and niobium-0.01 w/o.

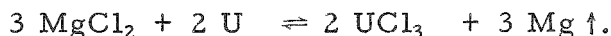
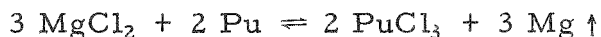
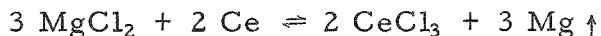
Table I

STANDARD FREE ENERGIES OF FORMATION AT 1200°C(3)

Chlorides	$+\Delta F^\circ$ (kcal/gm-atom of chlorine)	Fluorides	$+\Delta F^\circ$ (kcal/gm-atom of fluorine)
CdCl ₂	-21	UF ₃	-86
ZnCl ₂	-32	UF ₄	-86
UCl ₄	-43	KF	-92
UCl ₃	-47	NaF	-95
MgCl ₂	-50	PuF ₃	-97
PuCl ₃	-54	MgF ₂	-100
CeCl ₃	-58	CeF ₃	-111
CaCl ₂	-72	BaF ₂	-115
BaCl ₂	-76	CaF ₂	-116
NaCl	-78		

As shown by Table I, separations of highly electropositive fission products from uranium and plutonium are possible in both the chloride and fluoride salt systems. However, fluoride salt systems are considerably more corrosive than chloride salt systems on potential container materials, e.g., alumina and beryllia. Therefore, work was limited to the use of chloride systems. The chloride salts making up these systems should be stable in the presence of uranium and plutonium metals and should be relatively nonvolatile at temperatures up to 1300°C. These two criteria limit the major slag constituents to a few alkaline-earth chlorides, e.g., BaCl₂ and CaCl₂.

A sufficient amount of oxidizing agent must be added to the carrier slag to oxidize rare earth and other highly electropositive fission product elements to their respective chlorides. Magnesium chloride is a practical oxidant because it is less stable than rare-earth element chlorides and because it has a reasonably low volatility at operating temperatures of 1100 to 1300°C. The MgCl₂ reacts with a rare earth (cerium), plutonium, and uranium by the following reactions:



Because magnesium is removed by vaporization, these reactions should be driven completely to the right, the $MgCl_2$ serving, in effect, only as a source of chlorine. The final distribution of cerium, plutonium, and uranium is then decided by equilibria involved in the following reactions



This study has consisted of (1) preliminary measurements of the relative extractions of rare earths and plutonium into a halide slag as a function of the amount of oxidant employed, (2) the loss of plutonium by vaporization, and (3) some observations on separation of slag and metal phases and on the performance of beryllia crucibles.

II. PREVIOUS EXPERIMENTAL WORK

A. Oxide Slags

The use of oxide slags has several difficulties. Chief among these are the scarcity of combinations of stable oxides with sufficiently low melting points for practical use, and the absence of container materials that are resistant to attack by molten oxides and molten uranium. For these reasons, after a few preliminary experiments^(6,7) this approach was not pursued at Argonne National Laboratory.

Subsequently Petit and co-workers in France found several relatively low melting (1100 to 1400°C) combinations of BeO , BaO , and SrO , and reported that high-fired BeO and Al_2O_3 crucibles offered some possibility for the containment of a molten oxide-uranium system, despite some dissolution of the crucible in the molten oxide. By use of a small amount of UO_2 in the flux as an oxidant, decontamination factors of greater than 1000 were realized for barium and lanthanum tracers.

In further work along this line at Argonne National Laboratory with uranium-fissium alloys,⁽⁹⁾ the high removals of an electropositive element (in this case, cerium) were confirmed, but the attack of BeO and Al_2O_3 crucibles was considered to be too severe to warrant further work along this line.

B. Halide Slagging

Several early investigations were conducted on the use of liquid halide slags for extracting fission products from molten uranium alloys (10-16)

In each of these studies, trace quantities of plutonium in neutron-irradiated uranium and large excesses of oxidant (MgCl_2 , UCl_3 , UCl_4 , UF_3 , and UF_4) were used. The separations observed were consistent with thermodynamic arguments; i.e., the alkali and alkaline-earth fission products were extracted more efficiently than plutonium, but attempts were not made to separate plutonium from rare earths. Noble-metal fission products remained in the uranium phase. Extraction of more than 90% of the plutonium was shown to be feasible.

The above oxidants were employed in fluxes of alkali and alkaline-earth chlorides and fluorides. Fluoride fluxes were generally employed in the early work, probably because thermodynamic estimates indicate a more efficient extraction of plutonium by UF_3 than by UCl_3 . Recently, chloride salts have received the most attention because they are less corrosive than the fluoride systems.

Previous work on the selective extraction of highly electropositive elements from uranium and plutonium alloys is described in the following paragraphs.

1. Extraction of Rare Earths from Uranium Fuels with Chloride Fluxes

The use of chloride fluxes (often containing some fluoride) was investigated at Argonne National Laboratory for the selective extraction of rare earths (represented by cerium) from uranium-5% fission fuels.⁽⁹⁾ Beryllia, which is resistant to attack by uranium, chloride fluxes, and combinations of these, was used as a containment material.*

Five runs were made in which two different halide fluxes were used: calcium chloride containing 13 m/o magnesium fluoride, and calcium chloride containing 2 m/o uranium tetrachloride. Cerium in concentrations of between 0.5 and 0.8 w/o was present in the 700-g charges of uranium-5% fission alloy. The liquation temperatures were varied between 1100 and 1300°C. These runs are summarized in Table II. With the uranium tetrachloride oxidant, cerium removals were about 94%; with the magnesium fluoride oxidant, a cerium removal of 99% or better was obtained.

Table II

MELT REFINING OF URANIUM FUEL UNDER CHLORIDE FLUXES⁽⁹⁾

700 grams uranium-5 w/o fission containing 0.5 to 0.8 w/o cerium melt refined under 200 grams of flux for 1 hour in beryllia crucible, and poured at 1300°C

Flux	Liquation Temp (°C)	Yield (%)	Ce Removal from Ingot (%)	U Loss in Flux (% of U in charge)
$\text{CaCl}_2 + 2 \text{ m/o } \text{UCl}_4$	1100	89.6 (pouring temp- 1100°C)	94.3	0
$\text{CaCl}_2 + 13 \text{ m/o } \text{MgF}_2$	1100	97.4	99	0.03
$\text{CaCl}_2 + 13 \text{ m/o } \text{MgF}_2$	1200	97.6	99.3	0.003
$\text{CaCl}_2 + 2 \text{ m/o } \text{UCl}_4$	1200	90.7	93.9	3.85
$\text{CaCl}_2 + 13 \text{ m/o } \text{MgF}_2$	1300	95.7	~100	1.35

*After a brief investigation, fluoride fluxes were eliminated from consideration because no satisfactory containment material could be found.

Because a poor product yield (90%) was obtained in the first run on pouring at 1100°C, subsequent pours were made at 1300°C. Yields on pouring at 1300°C varied between 95.7 and 97.6%. Before the metal was poured, the flux was removed by freezing the metal, pouring off the flux, and vaporizing the residual flux under reduced pressure. The loss of uranium in the flux was 0.03% or less at a liquation temperature of 1100°C. A single isopressed beryllia crucible was used for three runs and did not suffer any apparent damage.

2. Extraction of Rare Earths from Plutonium-rich Fuels

Leary and co-workers^(17,18) have successfully extracted rare earths from plutonium-rich fuels that were under investigation for use in the Los Alamos LAMPRE reactors (Los Alamos Molten Reactor Experiments). Their charge material was an alloy consisting of 92.5 w/o plutonium, 2.5 w/o iron, and 5 w/o of a mixture of fission products representing the major fission products after 10% burnup of the plutonium. In one experiment, approximately 100 grams of the alloy was contacted with 13 grams of a KCl-LiCl slag or flux, to which was added a quantity of MgCl₂ 10% in excess of that required to oxidize the rare-earth elements. More than 99% of the cerium and more than 97% of the lanthanum were transferred to the salt phase, along with less than 1% of the plutonium. In another experiment, the same transfer of cerium and lanthanum was realized with a PuCl₃ oxidant in a NaCl slag. The use of PuCl₃ as an oxidant would enable replacement of the fission products oxidized with plutonium.

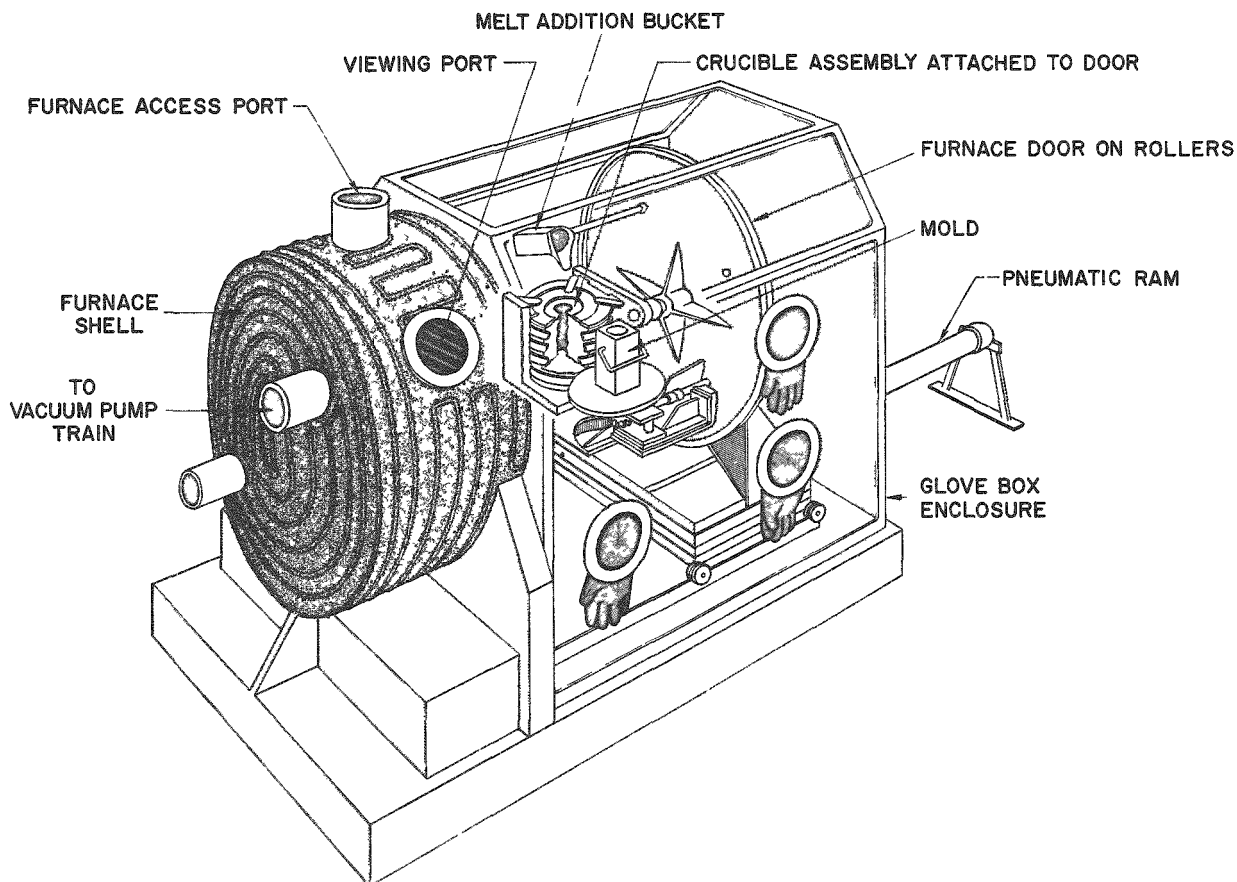
III. EQUIPMENT, MATERIALS, AND PROCEDURE FOR PRESENT INVESTIGATIONS

The equipment used was an induction furnace with an integral glovebox (Figure 1). In this furnace the crucible, induction coil, and mold are mounted on a movable furnace door which rolls on tracks within the glovebox to permit convenient handling of alpha-active materials. The furnace is connected to a vacuum system which can reduce the pressure in the furnace to 2×10^{-4} mm Hg. The induction coils are made of solid copper.⁽¹⁹⁾

The immediate application of a halide slagging process is to the second core loading of the Experimental Breeder Reactor-II. The exact composition of this material has not yet been established. An alloy consisting of 80 w/o uranium-10 w/o plutonium-10 w/o fission and melting between 910 and 920°C⁽²⁰⁾ was used (see actual composition in Table III, p. 9). It is believed that the alloy finally selected will not vary significantly in composition from this experimental alloy.

Figure 1

SCHEMATIC DIAGRAM OF INDUCTION FURNACE AND GLOVE BOX
FOR MELT REFINING OF URANIUM-PLUTONIUM-FISSIUM ALLOY



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Table III

MELT REFINING OF PLUTONIUM CONTAINING FUELS BY USE OF HALIDE FLUXES

All runs liquated at 1150°C for 1 hr under argon flux poured off at 860°C
and ingot poured at 1300°C

Metal Charge Composition (w/o)	80.97 uranium 10.02 plutonium 9.01 fissium plus cerium in amounts listed in column 3 of this table
Fissium Composition (w/o)	2.87 molybdenum 3.20 ruthenium 0.54 rhodium 1.13 palladium 0.93 zirconium
Flux Composition	81 w/o BaCl ₂ 19 w/o CaCl ₂
Ratio of Charge to Flux	7:2
Oxidant	MgCl ₂
Crucible	Isopressed high density high-purity BeO

Run No	Metal Charge		Excess Oxidant (%)	Ingot Yield (%)	Ce Removal from Ingot (%)	Pu Transferred to Flux (% of Pu in charge) ^a	Pu Vaporized (% of Pu in charge)	Ce/Pu Separation Factor ^a
	Wt (g)	Ce (w/o)						
HVC-5	737	0.335	No oxidant added	97.1	62.3	0.05	0.09	-
HVC-7	743	0.335	10	98.3	98.2	0.45	~0.002	7400
HVC-10	716	0.468	10	97.5	97.6	0.29	~0.006	7200
HVC-11	691	0.751	100	96.9	99.1	1.88	~0.085	4100

^a Separation Factor $\frac{\text{Ce in Flux/Ce in Metal}}{\text{Pu in Flux/Pu in Metal}}$ Ce and Pu values are weight percent at the end of a run

In the preliminary experimental work on uranium fuels, no attempt was made to optimize the flux composition. Consequently, although the $\text{CaCl}_2\text{-MgF}_2$ salt worked satisfactorily from a chemical standpoint, a less volatile flux which is more readily separable from the uranium matrix would be advantageous. After experimentation with several different salts, the two-component salt system, $\text{BaCl}_2\text{-CaCl}_2$, was found to possess the required characteristics. As shown by the phase diagram,⁽²¹⁾ a 70 m/o $\text{BaCl}_2\text{-CaCl}_2$ mixture has a melting point of $\sim 800^\circ\text{C}$. This figure was checked experimentally. Since the melting point of the 80-10-10 uranium-plutonium-fissium alloy is estimated to be $910\text{-}920^\circ\text{C}$ ⁽²⁰⁾, the use of the 70 m/o $\text{BaCl}_2\text{-CaCl}_2$ flux would enable the separation of the flux and metal phases by pouring off the molten flux from the solidified metal at a temperature of about 860°C . No concerted attempt was made to optimize the composition of the flux, but the composition chosen has the desirable characteristics of having a melting point lower than that of the metal phase, of being readily separable from the metal when both are frozen together, and of having low volatility.

In the experimental procedure used, the charge, flux, and oxidant were loaded into an isopressed, high-density, high-purity beryllia melt crucible. An Alundum mold crucible for the flux, and a specially designed magnesia-washed copper mold crucible* for the ingot, were placed in their proper positions on the rotary mold table within the furnace (see Figure 1). The furnace door was then closed pneumatically, and the furnace was evacuated to a pressure of about 0.1 micron. The furnace was flooded with argon to a pressure of 6-10 in. of water and heated by induction to 1150°C over about a 3-hour period. The melt was maintained at this temperature for one hour. Temperatures were measured by a Pt/Pt-10% rhodium thermocouple encased in a beryllia protection tube. The furnace was then cooled to 860°C , at which temperature the still molten flux was separated from the solid metal by being poured into the Alundum mold crucible.** The metal, which had remained behind, was then heated to 1300°C , at which temperature it was poured into the copper mold. Samples of both the ingot and the flux were taken for cerium and plutonium analyses. An estimate of the extent of plutonium vaporization was obtained by means of a 1-in.-wide tantalum condenser suspended over the top of the beryllia crucible. To date, one beryllia crucible has been used eight times, including both experimental and alloy-preparation runs.

*The special features of the copper mold were as follows: (1) To eliminate the dangers of external sampling of plutonium-containing fuels, the bottom of the copper mold had four small indentations, approximately $1/8$ in. in diameter by $1/4$ in. long. When the uranium was poured, these indentations filled with metal, producing protrusions which were later sheared off in the glove box for samples. This technique has been shown to give representative samples.⁽²²⁾ (2) A second problem in the design of a mold is to ensure easy and certain removal of a comparatively small ingot from a massive mold. This was accomplished by tapering the mold, splitting it horizontally along the bottom face of the ingot, and adding two machine screws which, when tightened, push against two steel inserts and thereby lift the bottom part of the mold away from the ingot. The ingot is then left in the upper part of the mold from which it is easily removed by slight tapping.

**Alternatively, the flux and metal may be poured simultaneously, and the solidified phases broken apart by gentle tapping.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Four runs with plutonium-containing fuels have been conducted. Experimental conditions and results of these runs are given in Table III. The runs included a blank run in which no MgCl_2 oxidant was added to the flux, two runs with 10% excess MgCl_2 (i.e., 10% in excess of the quantity to react stoichiometrically with the cerium present), and one run with 100% excess oxidant. The percentage of excess oxidant was varied to determine the sensitivity of plutonium transfer to the amount of oxidant added.

The results shown in Table III are encouraging. The loss of plutonium by vaporization was negligible in all runs. A cerium extraction of 62% in the blank run probably resulted from the presence of oxidizing impurities in the flux, and, in conjunction with a low plutonium transfer (0.05%) to the salt, indicates the ease with which cerium may be removed selectively.

In the runs made with 10% excess oxidant, plutonium transfer to the salt was also small (<0.5%). Of particular significance is the cerium-to-plutonium separation factor, which averages 7300 for these two runs. Both the ingot yields and the cerium removal were about 98%.

When the oxidant was increased to a 100% excess, the plutonium transferred to the flux increased (to ~2%), and the cerium removal increased (to ~99%). The relatively high plutonium transfer in this run indicates that the amount of oxidant added will have to be controlled within rough limits which will depend on the amount of fission products formed. Since burn-up, and hence the amount of fission products formed, can be predicted with an accuracy of 20% or better by either routine analytical methods or calculation, the feasibility of the process for actual plant operation appears to have been established.

V. CONCLUSIONS AND RECOMMENDATIONS

The experimental work that has been conducted has shown the following:

- 1) Highly electropositive elements may be selectively extracted from uranium-plutonium alloys and into a molten halide salt phase. Excessive extraction of plutonium may be avoided by controlling the amount of oxidizing agent employed.
- 2) Plutonium losses by vaporization are small.
- 3) The salt and metal phases may be cleanly separated.
- 4) The metal fuel alloy may be separated in high yields (>98%).
- 5) High-density beryllia crucibles show considerable promise as containment vessels.

Because of the lower temperature of operation (as compared to the present EBR-II melt-refining process), further work on this procedure is warranted.

Included in the further work should be the development of information on the following items:

- 1) The chemistry and equilibrium of the reactions involved.
- 2) The activity coefficients of all the species present in the salt and metal phases. Such information would enable the calculations of a theoretical separation factor, which, in turn, could be used as a guide to establish optimum operating conditions.
- 3) Flux composition. Although the $\text{BaCl}_2\text{-CaCl}_2$ mixture works satisfactorily, a more extensive program than has been carried out might result in a better flux.
- 4) Containment materials. The most important problem remaining is proof that the same type of isopressed beryllia crucible used in these experiments will be satisfactory in full-scale operations. High-density, isopressed beryllia crucibles to handle the full charge from the second-core EBR-II loading have been ordered and will be tested. These crucibles are required to handle a fuel alloy charge of 10 to 15 kg. Probably the largest charge of material which could be handled would be about 50 kg (somewhat less than half the critical mass of the fuel alloy). Such batch sizes would be practical for processing the fuel discharged from large, advanced, power reactors. Besides pressed crucibles, beryllia-lined crucibles fabricated by plasma-spraying beryllia on an inexpensive substrate material, such as silicon carbide, may also be practical.

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