

Molten Salt Reactors (MSRs): Coupling Spent Fuel Processing and Actinide Burning

Dr. Charles W. Forsberg*
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, Tennessee 37831-6179
Tel: (865) 574-6783
Fax: (865) 574-9512
E-mail: forsbergcw@ornl.gov

Professor Ehud Greenspan
University of California, Berkeley
4107 Etcheverry
Berkeley, California 94720-1730
Tel: (510) 643-9983
E-mail: gehud@nuc.berkeley.edu

Manuscript Date: July 25, 2003
File: MSR.HiltonHead.Combined.2003

Advances in Nuclear Fuel Management III
American Nuclear Society
Hilton Head, South Carolina
October 5–7, 2003

The submitted manuscript has been authored by a contractor of the U.S. Government under contract DE-AC05-00OR22725. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

*Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

MOLTEN SALT REACTORS (MSRs): COUPLING SPENT FUEL PROCESSING AND ACTINIDE BURNING

Charles Forsberg
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, Tennessee 37831
forsbergcw@ornl.gov

Ehud Greenspan
University of California, Berkeley
4107 Etcheverry
Berkeley, California 94720-1730
gehud@nuc.berkeley.edu

Keywords: Molten Salt Reactor, Transmutation, Actinide Burning

ABSTRACT

Molten salt reactors (MSRs) are liquid-fueled reactors that can be used for burning actinides, production of electricity, production of hydrogen, and production of fissile fuels (breeding). Fissile, fertile, and fission products are dissolved in a high-temperature molten fluoride salt with a very high boiling temperature (~ 1400°C). The molten salt serves as both the reactor fuel and the coolant. Heat is generated in the reactor core and transported by the fuel salt to heat exchangers before returning to the reactor core. The MSR is one of the six advanced reactor concepts identified by the Generation IV International Forum as a candidate for cooperative development and is the only fluid-fuel concept.

Liquid-fueled reactors have several unique capabilities in terms of burning actinide elements and thus offer the potential to reduce the long-term radiotoxicity of the waste from production of electricity. This is the basis for much of the renewed interest in MSRs. MSRs (compared with solid-fuel reactors) avoid the complexities of fabricating solid-fuel elements of minor actinides (neptunium, americium, and curium) and many other difficulties. MSRs require fuel processing technologies based on molten fluoride salts. Such processing technologies have the potential for significant improvements in economics compared with traditional aqueous technologies and pyroprocesses for solid spent nuclear fuel.

Actinide burning is a new mission for the fuel cycle; thus, research is just beginning to define the options and alternative paths forward. Some of the characteristics, technical options, and technical challenges associated with molten salt systems are described herein.

I. INTRODUCTION

Historically, there have been two types of fuel cycles: (1) open, with direct disposal of spent nuclear fuel (SNF) and (2) closed, with the processing of SNF to recover and use the fissile and fertile fuel that remains. These two types of fuel cycles and the associated facilities are very different because of their fundamentally contrasting objectives. Today, a third type of fuel cycle is being considered: the destruction of long-lived radionuclides that require geological disposal. Such a process should (1) reduce the risks [both real and perceived] from release of radionuclides in the repository to the environment, (2) expand repository capacity by destruction of the longer-lived heat-generating radionuclides—the actinides, and (3) reduce the potential use of fissile materials for weapons. This third type of fuel cycle is being considered in the context of fuel cycles with and without breeder reactors.

The requirements for such a cycle as well as the unique characteristics of the actinides imply that the optimum reactors and fuel cycle facilities may be very different than those in the other two systems. One candidate for this mission is the molten salt reactor (MSR). The characteristics of an MSR for application to this type of fuel cycle are examined herein.

2. ACTINIDE BURNING

Traditional closed fuel cycles recycle uranium and plutonium. The minor actinides (neptunium, americium, and curium) are treated as wastes. With actinide burning, all of the actinides are to be destroyed. The strategies to achieve this can vary from (1) a traditional closed fuel cycle with plutonium recycle and specialized facilities for destruction of minor actinides which may use some plutonium to fuel this mission to (2) a system to destroy all actinides from light-water reactor (LWR) SNF.

Because of the different nuclear characteristics of the minor actinides that are to be destroyed, a fuel cycle that burns all actinides is significantly different from a traditional fuel cycle. It is these differences that lead to consideration of different reactors and fuel cycles for this mission.

- *Isotopic properties.* Although the recycle of plutonium from the enriched-uranium SNF produced by an LWR is an industrial technology, it is far more difficult and expensive to recycle plutonium that has been recycled multiple times, as well as to recycle americium and curium. The properties of these isotopes (Table 1) indicate why fabrication and irradiation are more difficult. The high heat-generation rates restrict fuel fabrication batch size and create major problems in using any process with heat-sensitive processing operations. The neutron emission requires significant shielding. The extremely high alpha activity makes contamination control difficult, drastically increases the fuel fabrication cost, and results in generation of significant quantities of secondary wastes.

- *Isotopic mixtures.* The minor actinides are generated by multiple neutron capture with different numbers of neutrons required to generate various isotopes. The buildup of each isotope is a strong function of burnup. Small variations in SNF burnup imply large differences in the ratios of various neptunium, plutonium, americium, and curium isotopes. This results in each batch of actinides having very different nuclear properties. This creates major challenges in fuel fabrication and reactor-core management for solid-fuel reactors. In solid-fuel reactors, the energy production of each fuel pin in each location must be tightly controlled over the entire period of irradiation to prevent excess power levels and fuel burnup. This process is relatively simple with plutonium but is very difficult with higher actinides with their much more complex neutronic behavior. The large isotopic variability between batches requires very large scale mixing of batches to minimize the complexities of fuel management and fabrication.

Table 1 Characteristics of actinides

Isotope	Critical Mass ^a (g)	Heat Generation (W/g)	Neutron Rate (no./min-mg)	Alpha Rate (no./Min-mg)
Np-237	20,000	2.07×10^{-5}	0	8.01×10^5
Pu-238	3,000	0.570	155	1.94×10^{10}
Pu-239	450	1.013×10^{-3}	1.35×10^{-3}	6.94×10^7
Pu-240	15,000	7.097×10^{-3}	53.7	2.57×10^8
Pu-241	200	4.06×10^{-3}	0	2.94×10^6
Pu-242	40,000	1.13×10^{-4}	95.3	4.32×10^6
Am-241	16,000	0.115	3.55×10^{-2}	3.88×10^9
Am-242m	13	5.08×10^{-2}	0	5.53×10^7
Am-243	25,000	6.42×10^{-3}	0	2.26×10^8
Cm-244	3,000	2.832	6.87×10^5	9.16×10^{10}
Cm-245	30	5.89×10^{-3}	0	2.00×10^8
Cm-246	Fissile	1.01×10^{-2}	5.58×10^5	3.52×10^8

^aSubcritical mass limits used in criticality safety evaluations for systems without nuclear-grade moderators such as D₂O.

Because of these concerns, renewed interest has emerged in MSRs in which the fuel is dissolved in the coolant. Such reactors offer several advantages in terms of actinide burning:

- *No isotopic blending.* Different lots of SNF have very different plutonium, americium, and curium isotopics. The MSR has a homogeneous liquid fuel. Any fissile material can be fed to the reactor and is homogenized with all the other fuel in the reactor. The very different nuclear characteristics of different batches of higher actinides are addressed by the rate of addition to the homogeneous molten salt. No batch mixing of different batches of actinides is required.
- *No fuel fabrication.* The higher actinides have small critical masses and high rates of decay heat, representing a serious technical and economic challenge for fuel fabrication. However, this is a nonissue for an MSR, because no fuel fabrication is required.

The unique challenges of actinide burning require consideration of the entire fuel cycle. Many types of systems have been proposed; examples include those from Russia and the United States.

- *Russia.* The Kurchatov Institute¹ in Moscow (Fig. 1) has proposed a nuclear system that contains both thermal neutron reactors (LWRs) and fast reactors. The system also contains MSRs for the specific purpose of burning higher actinides while producing electrical power.
- *United States.* The Advanced Fuel Cycle Initiative of the U.S. Department of Energy is considering a system in which LWR fuel is processed, the plutonium and neptunium are recycled for use in LWRs. Several options for destruction of the higher actinides are being considered. In this two-tier system, LWR fuel processing is used to change the waste characteristics to increase the capacity of the planned Yucca Mountain repository and thus to avoid the need to site multiple repositories.

In each of these systems, the actinide burning is performed in separate facilities. This avoids the complexities of minor actinide processing and burning in most of the nuclear power system. All of these systems require a reconsideration of reactors and SNF processing facilities.

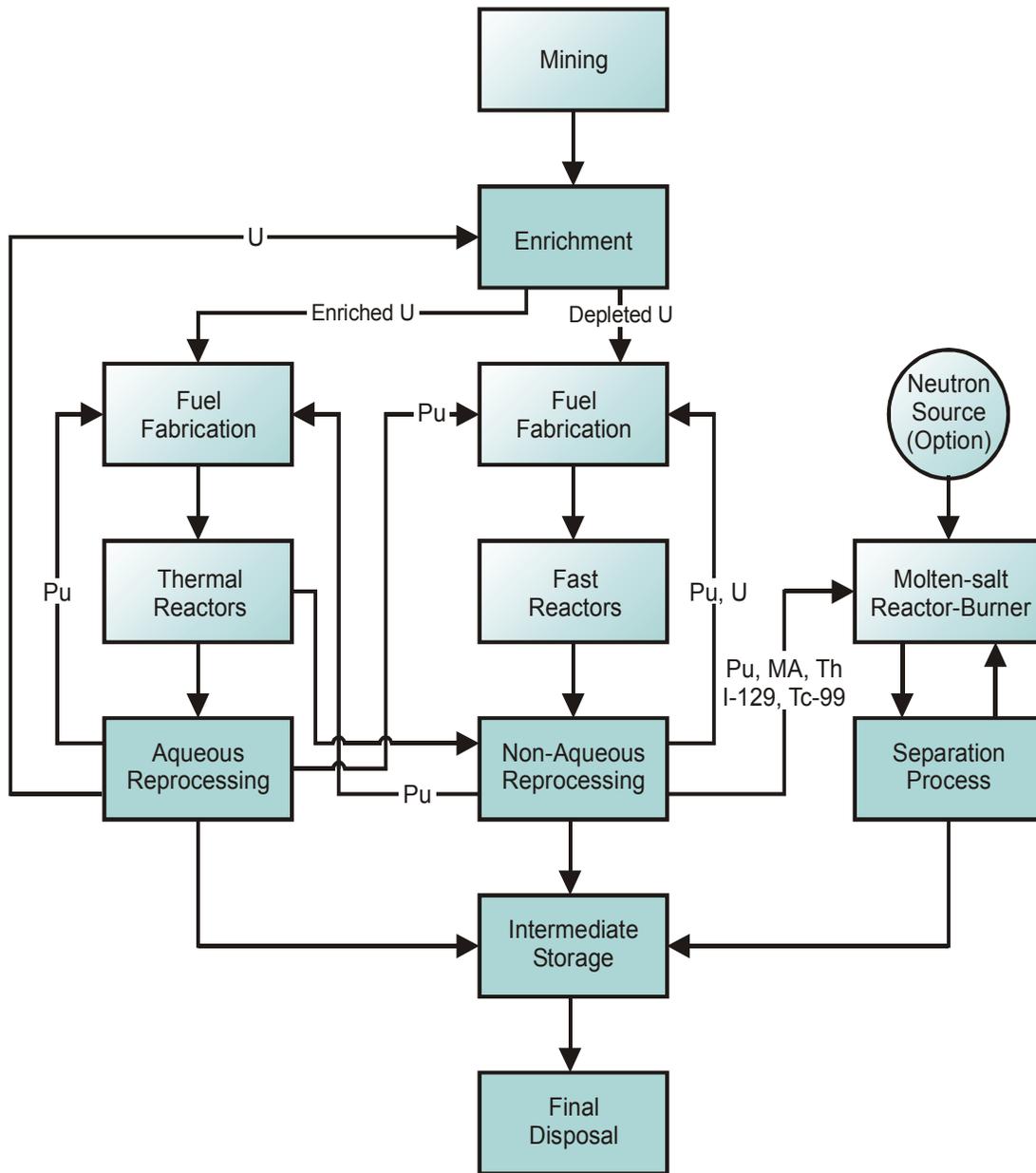


Fig. 1 Kurchatov Long-Term Vision of the Nuclear Fuel Cycle.

3. MOLTEN SALT REACTORS

MSRs were first developed in the late 1940s and the 1950s in the United States for military jet aircraft propulsion.² In 1954, the 2.5-MW(t) Aircraft Reactor Experiment (ARE) demonstrated high-temperature operation and established benchmarks in performance for a circulating fluoride molten salt (NaF-ZrF₄) system with the uranium dissolved in the salt.

Although a nuclear aircraft was never deployed, the ARE was followed in the 1960s by a program to develop a molten salt breeder reactor (MSBR). The Molten Salt Reactor Experiment (MSRE), an 8-MW(t) reactor, demonstrated many of the features required for a power-generating reactor: (1) a ⁷LiF-BeF₂ salt suitable for breeding applications; (2) graphite moderator compatibility with the fluoride salt; (3) stable performance; (4) removal of xenon and krypton from the fuel and subsequent trapping in the off-gas systems; and (5) the use of different fuels, including ²³⁵U, ²³³U, and plutonium. The MSRE successfully operated for 13,000 equivalent full-power hours between 1965 and 1968. A detailed 1000-MW(e) engineering conceptual design of an MSBR was developed. These programs³ of the 1950s and 1960s demonstrated the key technical features of these concepts. However, the MSR program was later canceled when the United States decided to concentrate its reactor development program on a single concept.

A schematic of an MSR is shown in Fig. 2. The fluoride molten salt with dissolved fissile, fertile, and fission isotopes flows through a reactor core moderated by unclad graphite to a primary heat exchanger, where the heat is transferred to a secondary molten salt coolant. The fuel salt then flows back to the reactor core. The heat is generated directly in the molten fuel. In traditional MSR designs, the liquid fuel salt enters the reactor vessel at 565EC and exits at 705EC and - 1 atmosphere (coolant boiling point: - 1400EC). The reactor and primary system are constructed of modified Hastelloy-N or a similar alloy to provide corrosion resistance to the molten salt. Volatile fission products (e.g., krypton and xenon) are continuously removed from the fuel salt. A secondary coolant loop transfers the heat to the power cycle.

The choice of fluoride molten salt depends upon the objectives of reactor operation. For operation as a converter reactor for electricity or hydrogen production or for burning of actinides, a salt (similar to the ARE salt) containing NaF and ZrF₄ could be used. Such salts are inexpensive and are relatively nontoxic. For actinide burning, the molten salt selected to maximize the solubility of actinides. For a breeder reactor, on-line processing is required. For other missions, depending upon specific design details, the processing can be done off-line or off-site. If power production is the goal, a MSR can operate up to 6 years between reactor refuelings.

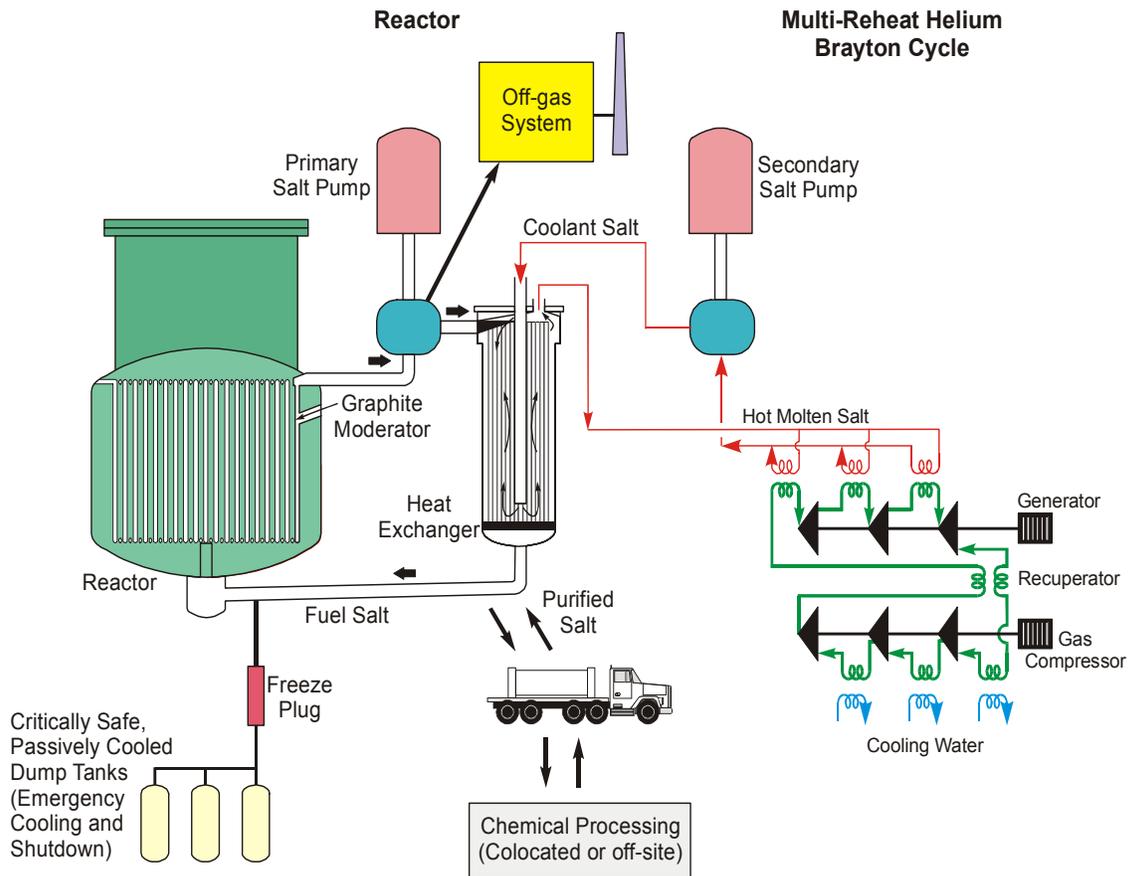


Fig. 2 Molten Salt Reactor

For actinide burning, the MSR offers an important safety characteristics (in addition to avoiding the issues with solid fuels): low actinide inventories. MSRs have a low inventory of fissile materials compared with other reactors because (1) thermal and epithermal neutron reactors require less fissile inventory than fast reactors; (2) the fissile inventory of this fuel cycle outside the reactor is low compared with that for solid-fuel reactors; (3) little excess reactivity is required to compensate for burnup, because fuel is added on-line; (4) direct heat deposition in the fuel/coolant allows very high power densities compared with those for solid-fuel reactors; and (5) certain high-absorption fission products, such as xenon, are continuously removed. This low actinide inventory is important in the context of minimizing the potential accident source term.

Two reactor-plant technical developments⁴ that have occurred since the 1970s have potentially important impacts on the viability of the MSR, particularly for actinide burning.

- *Compact heat exchangers.* In an MSR, the fuel circulates between the reactor core and the heat exchangers; thus, a significant fraction of the inventory of fuel salt is in the heat exchangers rather than in the reactor. The last decade has seen the development of compact heat exchangers for the offshore oil industry. The inventory of fluids in these heat exchangers is a small fraction of that in conventional tube-and-shell heat exchangers. The technology has the potential to dramatically reduce the out-of-core inventory of fuel salt, thus directly reducing the actinide inventory in the reactor and the quantities of fuel salt that must be processed. The technology does require good control of noble metal plate-out on the heat exchangers.
- *Brayton helium cycle.* In the 1970 designs, the MSRs were coupled to a steam cycle because that was the only available power-cycle technology. Since that time, closed-cycle, multi-reheat helium Brayton power cycles have been developed for electricity production. Brayton power cycles, when coupled to MSRs, are expected to increase power plant efficiency and lower costs. They also greatly reduce one of the technical challenges: migration of tritium from the primary system into the secondary system. Sources of tritium in an MSR include fission tritium and activation of the salt. It is relatively easy to trap tritium from the helium in the colder sections of the Brayton cycle. In contrast, it is difficult to (1) prevent tritium from migrating from the primary molten salt to the steam through the high-temperature heat exchangers or (2) manage the tritium if it combines with the steam. This technical benefit of a Brayton cycle gives the reactor designer a greater degree of freedom in the selection of the salt to optimize actinide burning. Minimizing tritium production is not as strong a requirement.

4. REACTOR PHYSICS OF ACTINIDE BURNING

The approach to actinide burning depends upon the system goals. If the primary mission of the MSR is actinide burning, then the principal objective is to minimize the number of MSRs compared with other reactors in the nuclear system. Several studies have been undertaken to evaluate the options⁵⁻¹⁴. Several are reviewed herein.

4.1 Kurchatov Study: Minor Actinide Burner

The Kurchatov Institute in Russia is engaged in the design of molten salt transmuting reactors for the purpose of closing the nuclear fuel cycle⁵⁻⁷ as well as in the measurement of the physical and chemical properties of different molten salts.⁷

The Kurchatov group conducted a feasibility study of the Molten Salt Advanced Reactor Transmuter (MOSART). Cores fueled with different compositions of transuranic (TRU) trifluorides from LWR SNF were considered. Different conceptual core configurations and molten salt systems, as well as different removal processes for the soluble fission products, were considered. The MOSART concept uses a single-fluid system and is fueled by TRU trifluorides without addition of either UF_4 or ThF_4 . The elimination of thorium from the feed offers a number of advantages: (1) maximizing capability for transmutation of actinides by avoiding neutron losses to thorium; (2) easier molten salt processing; and (3) reduced losses of TRU to the waste, due to reduction in the required processing rates and simplification of the fuel salt processing flowsheets.

The overall layout of MOSART is similar to that of the MSBR. It is designed to have a power level of 2400 MW(t) and a molten salt temperature of 550 to 620EC at the entrance to the core. The core design variables considered are the diameter and pitch of the molten-salt channels and the neutron flux amplitude. The flux amplitude is determined by the specific power and the actinide concentration in the molten salt. The graphite lifetime in the core is constrained by radiation damage considerations to a peak fluence of 3×10^{22} n/cm² of neutrons above 50 keV. Several types of molten salt were considered, including different combinations of Li, Be, Na, and Zr fluorides. The preferred combination consists of 15 mol % LiF, 58% NaF, and 27% BeF₂. The solubility of actinides in this molten salt is estimated⁷ to be 2 mol %.

Three TRU feed compositions have been considered: (1) using all TRU elements from uranium dioxide (UO₂) SNF of a commercial pressurized-water reactor (PWR) (60 GWd/t U, 4.9% ²³⁵U/U, 1 year cooling); (2) using all TRU elements from mixed oxide (MOX) SNF (a) fabricated with 7% plutonium from irradiation of UO₂ SNF in a PWR and natural uranium and (b) processed after 10 years of cooling; (3) using a process similar to option 2 but with the plutonium being recycled in PWR's an infinite number of times. This third option, the minor actinide burner, has a very high fraction of minor actinides and a relatively small concentration of plutonium and, especially, fissile plutonium.

An on-line fission products clean-up system has been designed for MOSART. Its efficiency is isotope dependent. Volatile fission products such as Xe and Kr are purged with He gas within - 50 s of their production. The relatively noble fission products (Zn, Ga, Ge, As, Se, Nb, Mo, Ru, Rh, Pd, Ag, Tc, Cd, In, Sn, Sb, and Te) are removed within 2.4 h by plating out on surfaces. The more chemically reactive fission products (Zr, Ni, Fe, Cr, Np, Pu, Am, Cm, Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er, Sm, and Eu) are removed within 1 to 5 years, while Sr, Ba, Rb, and Cs are removed after >30 years.

The preferred design for the minor actinide burner arrived at by the Kurchatov group is a core without a graphite moderator. Such an MSR can be designed to be critical when fed with either one of the three actinide feed compositions considered. Table 2 summarizes selected characteristics of this reactor.

4.2 University of California: Plutonium and Minor Actinide Burner

The University of California at Berkeley (UCB) has been examining the feasibility of transmuting the actinides from LWR SNF in MSRs.⁸ The approach is a once-through fuel cycle proposed by Bowman.⁹ The molten salt is a mixture of NaF + ZrF₄ + fluorinated actinides. The MSR would operate at temperatures between 600 and 700EC. The fuel is made by reacting LWR SNF with fluorine. The UF₆ and the volatile fission products are removed and the remainder is mixed with NaF to form the molten salt fuel (NaF + ZrF₄, with a few percent TRU). This approach uses the clad and fission product zirconium in the LWR SNF as a component of the molten salt. The salt is very similar to that used in the first MSR: the ARE. Fresh molten salt fuel is continuously fed and mixed into the reactor's salt plenum, and an equal volumetric flow rate of the mixed molten salt is continuously removed along with the actinides and fission products to keep the overall salt inventory constant.

The goal of the UCB study is to find the maximum fraction of the actinides fed into the MSR that can be transmuted in one pass through the reactor. The search for the maximum fractional transmutation is done in a parametric study. Four variables are considered in this study: (1) the pitch of the molten salt channels, (2) the diameter of the molten salt channels, (3) the volumetric feed and removal rate of the fuel salt, and (4) the concentration of actinides in the feed salt. The core designs considered are subjected to three constraints: (1) the actinide concentration in the molten salt should be below the solubility limit (for NaF-ZrF₄, this limit was estimated to be 1.56 mol% of actinides); (2) k_{eff} , the neutron multiplication factor, must be above 1.0 for a critical reactor; and (3) the fast neutron fluence in the graphite, measuring radiation damage to the graphite, should not exceed 3×10^{22} n/cm² of neutrons of energy greater than 10 keV. The actinides fed into the MSR are all the heavy metal isotopes in the fuel discharged from PWRs at 33 GWd/MTIHM, excluding 99.9% of the uranium.

Table 2 Selected characteristics of the preferred MOSART design*

Thermal power, MW(t)	2400	MS processing rate, kg/day	347
Electrical power, MW(e)	1100	FP removal time, days	300 EFPD
Capacity factor	0.821	Fraction of actinides transmuted in 60 years**, %	91–92
Core height/diameter, m/m	4/3.5	TRU reactor input, kg/year	740
MS power density, W/cm ³	50	TRU input, mass %	
MS volume in core, m ³	30.4	Np	0.94
MS volume in reactor, m ³	48.4	²³⁸ Pu	7.07
Neutron flux, n/cm ² -s	1 x 10 ¹⁵	²³⁹ Pu	9.41
Fuel salt composition, mole %		²⁴⁰ Pu	38.91
AnF ₃	0.8	²⁴¹ Pu	10.41
NaF	58	²⁴² Pu	20.86
LiF	15	²⁴¹ Am	1.56
BeF ₂	27	²⁴³ Am	5.19
MS mass in core, kg	65,360	Cm	5.65
MS mass in reactor, kg	104,060	TRU going to waste, kg/year	4.8
FP mass in reactor, kg	650	TRU waste composition, %	
TRU mass in reactor, kg	4,820	Np	2.1
TRU mass in reactor after 75 years, Kg		Pu	71.3
		Am	7.7
		Cm	18.4
		Bk	0.1
		Cf	0.4
		Gaseous FP waste, kg/year	360
		Noble metal FP waste, kg/year	235

*MS = molten salt, TRU = transuranic, FP = fission products, EFPD = effective full power days. **Includes actinide inventory remaining in the reactor.

Figures 3 through 6 and Table 3 summarize selected results obtained in the parametric study for a molten salt feed rate of $0.167 \text{ cm}^3/\text{day}$ per 1 MW(t) of reactor power, an actinide feed concentration of 12.87 mol %, and a power density of 390 W per cm^3 of molten salt. The results are calculated for a unit cell that is finite in the axial direction and infinite in the radial direction. The core height is 400 cm. Surprisingly, but fortunately, the highest fractional transmutation (Fig. 5) was obtained for a ratio of graphite (carbon) to molten salt ratio (C/MS) for which k_{eff} is near the maximum attainable ratio (Fig. 3) and the equilibrium concentration (Fig. 4) is near its minimum. This optimal C/MS ratio is between 1 and 3. The corresponding neutron spectrum is highly epithermal (Fig. 6). However, at the power density considered, the neutron flux is on the order of 10^{15} n/cm^2 and the corresponding graphite lifetime is short, on the order of 1 year. It is possible to increase the graphite lifetime by softening the neutron spectrum (increasing C/MS) and, particularly, by reducing the power density. However, both approaches will result in a reduction in k_{eff} .

The k_{eff} values shown in Fig. 3 do not account for radial neutron leakage and for the effect of fission product accumulation in the molten salt. They do not account, as well, for the effect of efficient reflectors. Designing a finite core having 955 molten salt channels that are 7-cm diam. and 420 cm long and that have a C/MS ratio of 3 to be optimally reflected, a k_{eff} of 1.04 was obtained. This 448-cm-diam. core has 4% excess reactivity to compensate for the negative reactivity effect of the fission products that will accumulate in the molten salt. It is yet to be established whether on-line fission product extraction, for example, as proposed by the Kurchatov group (see previous section), can maintain the negative reactivity worth of the fission products residing in the core below 4%.

Rather than increasing the core graphite lifetime by reducing the power density, it is possible to design the nuclear power plant to have multiple cores. At the end of the graphite life in the operating core, the molten salt, with the actinides and fission products, will be transferred to the second core. The graphite structure in the first core will be replaced, while the second core will remain operational. This will permit a relatively high capacity factor to be maintained. The quantity of graphite that will have to be disposed of per given amount of actinides transmuted is independent of the power density and, hence, of the frequency of graphite replacements. The higher the power density and specific power, the shorter the graphite lifetime.

The quality of the plutonium exiting the MSR (Table 3) is significantly poorer than that from an LWR and even more so than that from a liquid-metal reactor. Particularly high is the concentration of ^{238}Pu . The larger the C/MS, the smaller becomes the fraction of ^{239}Pu and ^{241}Pu in the plutonium. Plutonium fed to the MSR is isotopically diluted with the entire inventory in the reactor and immediately degraded. There is no inventory of higher-grade plutonium in the reactor.

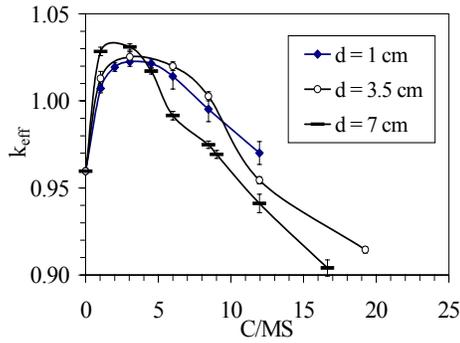


Fig. 3 Dependence of k_{eff} on graphite-to-fuel ratio (C/MS) for different fuel channel diameters.

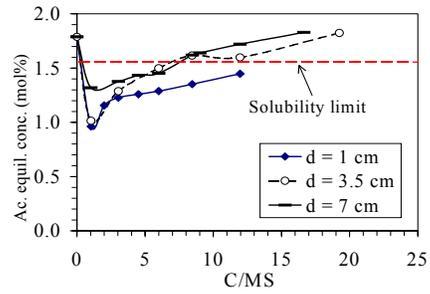


Fig. 4 Dependence of actinide (Ac) equilibrium concentration (mol %) on graphite-to-fuel ratio (C/MS) for different fuel channel diameters.

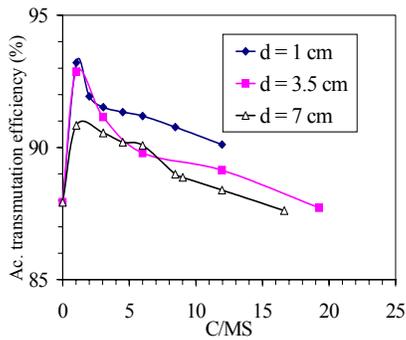


Fig. 5 Actinide transmutation efficiency for different fuel channel diameters and graphite-to-fuel ratios (C/MS).

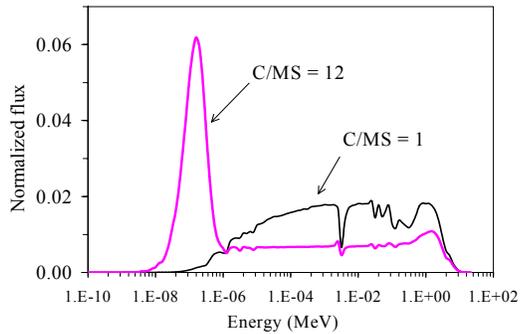


Fig. 6 Normalized total flux in a 1-cm-diam. fuel channels for C/MS = 1 (epithermal spectrum) and C/MS = 12 (thermal spectrum).

Table 3 Isotopic composition (atom %) of plutonium from different reactors

Isotope	Weapons grade	Liquid-metal reactor	LWR	Molten Salt Reactors	
				C/MS = 1	C/MS = 6
²³⁸ Pu	0	0.3	1.6	19	14
²³⁹ Pu	94	70.5	57.6	31	15
²⁴⁰ Pu	6	22.3	26.6	9	15
²⁴¹ Pu	0	2.7	8.8	28	16
²⁴² Pu	0	4.2	5.4	13	40
Total fissile	94	73.2	66.4	59	31

The equilibrium compositions of the actinides in an MSR that has 7-cm-diam. molten salt channels and a C/MS ratio of 3 are presented in Table 4 along with the transmuted fraction of the actinides that have been fed into the reactor. The feed composition used for calculating the data of Table 4 is slightly different from that used for generating the data given in the preceding figures and tables. Nearly 90% of the actinides fed into the MSR are transmuted in a single pass through the reactor: 95% of the ²³⁷Np, 98% of the ²³⁹Pu, 86% of the ²⁴¹Pu, and 98% of the ²⁴¹Am. The concentration of other actinides, notably the heavier isotopes, builds up. By reducing the molten salt feed rate, it is possible to further increase the fractional transmutation without significantly reducing k_{eff} , provided the fission products will be extracted so as to maintain a feed-rate-independent concentration.

The specific power of the reference UCB MSR design (the composition of which is given in Table 4) is very high, nearly 900 kW(e)/kg of heavy metal loaded in the reactor (including an out-of-core inventory that is equal to the in-core inventory). For comparison, the specific power of a typical liquid-metal-cooled reactor is on the order of 100 kW(e)/kg Pu and that of a PWR loaded with MOX fuel is on the order of 300 kW(e)/kg Pu. This indicates that the total inventory of heavy metal loaded into the MSR per unit electricity produced is the lowest possible. The power density of the reference UCB MSR core design (graphite and molten salt) is on the order of that of a PWR; it is higher than the power density of a typical graphite-moderated gas-cooled reactor by a factor of about 20.

Table 4 Equilibrium actinides composition (EC) and fractional transmutation (FT) in a once-through MSR (including short-lived isotopes)*.

Isotope	EC (atoms/cm ³)	FT	Isotope	EC (atoms/cm ³)	FT	Isotope	EC (atoms/cm ³)	FT
²³⁴ U	2.0×10^{17}		²³⁸ Pu	2.8×10^{19}	-0.62	²⁴⁴ Am	5.9×10^{16}	
²³⁵ U	1.4×10^{17}	1.61	²³⁹ Pu	2.4×10^{19}	-0.98	²⁴² Cm	4.1×10^{18}	
²³⁶ U	9.5×10^{16}	0.77	²⁴⁰ Pu	3.0×10^{19}	-0.95	²⁴³ Cm	2.4×10^{17}	0.82
²³⁷ U	1.2×10^{15}		²⁴¹ Pu	2.5×10^{19}	-0.86	²⁴⁴ Cm	6.0×10^{19}	3.62
²³⁸ U	1.4×10^{18}	-0.84	²⁴² Pu	4.7×10^{19}	-0.64	²⁴⁵ Cm	9.0×10^{18}	8.08
²³⁹ U	3.6×10^{13}		²⁴³ Pu	2.8×10^{16}		²⁴⁶ Cm	1.5×10^{19}	95.2
²³⁶ Np	1.7×10^{13}		²⁴⁴ Pu	5.3×10^{16}		²⁴⁷ Cm	1.6×10^{18}	
²³⁷ Np	9.2×10^{18}	-0.94	²⁴¹ Am	2.8×10^{18}	-0.97	²⁴⁸ Cm	2.0×10^{18}	
²³⁸ Np	1.0×10^{17}		²⁴² Am	2.4×10^{16}		²⁴⁹ Bk	6.8×10^{16}	
²³⁹ Np	5.5×10^{15}		^{242m} Am	5.5×10^{16}	-0.89	²⁴⁹ Cf	8.8×10^{15}	
²³⁷ Pu	4.4×10^{13}		²⁴³ Am	1.8×10^{19}	-0.54	²⁵⁰ Cf	7.0×10^{14}	
						Total		-0.89

*Channel diameter = 7 cm; C/MS = 3; molten salt feed-rate = 0.167 cm³/MWD(t)-, power density = 390 W/cm³ molten salt.

As an illustration of the actinide transmutation capability of MSRs consider the United States nuclear energy system. The total quantity of TRU elements that is expected to accumulate in the fuel discharged from LWRs until the year 2030 is estimated to be - 800 tons. Suppose all these actinides are to be transmuted in the reference UCB MSR. A single reference MSR generates - 6 GW(t), slightly larger than the largest PWRs being designed in Europe. Assuming a capacity factor of 0.9, such a reactor can transmute - 2 tons of TRU per year. In 40 years, such a reactor can transmute - 80 tons of TRU. Thus, in 40 years, approximately ten such MSRs can transmute the entire inventory of TRU to be accumulated in the United States until the year 2030 (excluding a small fraction that will end up in the waste stream). If part of the plutonium is be recycled in LWRs, a smaller number of MSRs will be required.

4.3 Observations

Studies of MSRs for actinide burning are at an early stage of development. Only a limited set of design options has been examined. Concepts such as multizone reactors and other more complex core designs have not been investigated and significant uncertainties exist.

If all actinides (plutonium and minor actinides) are to be burnt, the ratio of LWRs to MSRs in terms of thermal energy output is about 10. If the minor actinides are to be burnt with the plutonium recycled in LWRs, the ratio of LWRs to MSRs in terms of thermal energy output may be as high as 45. These ratios are sufficiently high that the option exists to collocate the MSRs with all fuel processing and thus avoid many of the transport issues associated with fissile materials and the higher actinides.

5. COUPLED SOLID SNF AND MSR SPENT SALT PROCESSING

The economic viability of actinide burning depends upon the costs of (1) reactors and (2) SNF processing—both for the MSR fuel salt and for the solid SNF from other reactors. For several reasons, the processing technology for MSR spent salt was developed independently from that used for LWRs and solid-fuel breeder reactors. The original developmental goal for the MSBR was a thermal-spectrum breeder reactor. The fissile inventory of a MSBR is sufficiently small that MSBRs could be started using enriched uranium. The MSBR could be independent of other reactor options. The fissile inventory of a fast-spectrum breeder reactor is 5 to 10 times that of a thermal-spectrum MSBR for comparable power levels. The large fissile inventory requirements to start fast-breeder reactors required a source of fissile material. The only fissile inventory sufficiently large was the plutonium in LWR SNF. Consequently, the processing of LWR and fast reactor systems was coupled.

For actinide-burning missions, there is the new requirement to couple MSRs with solid SNF processing. There are many options for processing MSR spent salt and solid SNF. These are bounded by two sets of options.

The solid SNF is processed using existing technologies to separate the actinides that are to be destroyed. These actinides are then converted into the fluoride form and sent to the MSR system for actinide burning.

The solid SNF and molten-salt fuel are processed in a single facility using many of the same processes. Conceptually, the system contains the following processes.

- *MSR fuel processing.* The molten salt from the MSRs is processed to remove excess fission products using the processes that were partially developed in the 1970s. The excess fission products are solidified into a final waste form.

- *Solid SNF (LWR) front end processing.* The solid SNF is converted into a molten salt to be further processed using the same processes as for MSR salts. The first step is a fluorination process such as fluoride volatility processing. This typically involves fluorination of the fuel—first with hydrogen fluoride and then with fluorine. The uranium is converted to volatile UF_6 , the required chemical form if the uranium is to be re-enriched to recover residual ^{235}U . Most other fission products and actinides are converted to nonvolatile fluorides. Fluoride volatility processes are currently used to convert various uranium feeds from mills into UF_6 . Pilot plants have tested the process for SNF. There are two variants.
 - Zirconium molten salts. For MSRs using a zirconium-based salt, the zirconium clad is fluorinated and becomes part of the molten salt.
 - Non-zirconium molten salts. If the MSR does not use a zirconium-based molten salt, there are two options. The fluorination process can be used only to convert the UO_2 matrix to a fluoride that is dissolved into a molten salt. Alternatively, the zirconium can be fluorinated and the traditional processes to remove excess zirconium from the molten salt can be used to adjust the zirconium content of the final salt.

The second (i.e., integrated) approach is new. *It has become an option because goals have changed.* Actinides are to be transferred from fuel cycles with solid SNF to an MSR. The characteristics of the system create the potential for significantly improved SNF processing but many uncertainties remain. The potential for an improved system compared to other methods to destroy actinides is based on several characteristics of the fluoride system:

- *Fluoride volatility.* Fluoride volatility processes¹⁵ have been developed and tested on a pilot scale for recovery of uranium from solid SNF with high-enriched uranium, with the small quantities of residual plutonium remaining with the fission products as part of the high-level waste stream. Fluoride volatility as a front-end processing option accomplishes two tasks: (1) converts the uranium directly into the required chemical form for recycle and (2) dramatically reduces the quantity of fuel material requiring further processing. Because fluoride volatility processing has potentially major economic advantages over traditional processes, there have been several efforts to develop technology for the processing of low-enriched SNF and recovery of the plutonium. The major technical problems have been the development of efficient methods to separate the plutonium fluorides from the fission product fluorides. However, such separation is not required if the goal is to produce a molten fluoride salt containing plutonium with reduced levels of fission products for a MSR. Equally important, it is a significantly less difficult challenge if the goal is to recover most of the plutonium for use in a solid-fuel reactors, with the minor actinides and some fraction of the plutonium left in the salt to be sent to an MSR.

- *Continuous processing.* The fluoride salts chosen for the MSR must meet nuclear fuel requirements and be good heat transfer fluids. The liquid characteristics of the salt allow development of large-scale continuous chemical processes for removal of fission products from the salt. This has potentially major economic implications. Two approaches have been developed for processing solid fuels: aqueous¹⁶ and pyrometallurgical.¹⁷ In aqueous processes, the solid fuel is first dissolved in a nitric acid solution, and various organic extractants are then used to selectively remove fissile materials from the aqueous solution. Aqueous processing is a high-throughput continuous process with large economics of scale. A variety of pyrometallurgical processes have been developed for fast reactor metal fuel. These processes use liquid metals with high-temperature molten chloride salts in batch operations for separation of fissile materials from fission products. While the pyroprocesses have many advantages (fewer criticality limits without water, group separation of actinides, etc.), the commercial SNF processing technology used worldwide is the aqueous PUREX process because of the current economic advantages of large-scale continuous processing and the production advantage of an oxide product that is compatible with both LWR and fast reactor systems. If the required product is a fluoride (as needed in an MSR), a high-throughput continuous pyroprocessing system becomes a potential candidate because of match between the processing plants and the MSRs. The chemistry of the fluoride salt systems is similar to the chloride system; thus, many of the separations advantages of chloride pyroprocessing systems apply to fluoride systems.
- *Integrated reactor, storage, and processing operations.* Liquid fuel reactors have the unusual characteristic that fuel salts with significantly different chemical and nuclear characteristics can be added to the reactor. If a particular fuel salt from processing LWR fuel has some unique characteristic, one can send that fuel salt to the MSR whose core physics (C/MS ratio) and molten salt inventory are most compatible. For salts that have remained in the reactor for long periods of time and have increased concentrations of actinides such as Californium, there is the option to store the salt for ten to twenty years to allow these short-lived radionuclides to decay away and place back in the reactor. These operational options do not exist in most other reactor systems. The potential gains are not well understood.

Many unresolved issues are associated with coupling the fuel cycles of LWRs and MSRs. If actinide burning in MSRs is undertaken, initial processing operations may use technologies somewhere between the traditional approaches and the approach that is outlined herein. However, important strategic implications are associated with these more advanced systems. Ultimately, decisions on whether to implement actinide burning depend upon societal, economic, and technical factors. To make such decisions, it is important to understand the long-term potential of such systems.

6. CONCLUSIONS

Historically, there have been two fuel cycles: once-through and closed. Today, a third fuel cycle is being considered: the destruction of all actinides. Because the goals are different than a traditional closed fuel cycle, a fundamental rethinking of reactor and processing options may be required. While the near-term option is to modify existing technology for this new mission, in the longer term other options exist. This paper has outlined some of the options for using molten-fluoride-salt technology—including (1) specially designed MSRs for actinide burning and (2) coupled molten-fluoride-salt fuel processing systems for both solid SNF from fast and thermal reactors and the fuel salt from the MSRs. Some of the technologies and operating strategies have been developed; however, many missing links remain in terms of reactor technology, SNF processing technology, and understanding the full system implications.

7. REFERENCES

1. “Power Provision of Mankind’s Sustainable Development, Cardinal Solution of the Nuclear Weapons Non-Proliferation Problem, and the Problem of the Environmental Recovery of the Earth Planet,” IAE-6213/3, Kurchatov Institute, Moscow, Russia (2001).
2. A. P. FRASS and A. W. SAVOLAINEN, “Design Report on the Aircraft Reactor Test,” ORNL-2095, Oak Ridge National Laboratory, Oak Ridge, Tennessee May (1956).
3. *Nuclear Applications & Technology* [entire issue], **8**, February (1970).
4. C. W. FORSBERG, “Molten Salt Reactors (MSRs),” *Proc. Americas Nuclear Energy Symposium, ANES 2002, Miami, Florida, October 16–18, 2002*, American Nuclear Society, La Grange Park, Illinois (2002).
5. O. FEINBERG, V. IGNATIEV, R. ZAKIROV, et al., “Physical and Chemical Feasibility of Fuelling Molten Salt Reactors with TRU's Trifluorides,” *Proc. Global '01*, Paris, France, September (2001).
6. V. IGNATIEV, O. FEYNBERG, A. MYASNIKOV, and R. ZAKIROV, “Reactor Physics & Fuel Cycle Analysis of a Molten Salt Advanced Reactor Transmuter,” *Proc. International Conference on Advanced Power Plants, ICAPP '03*, Cordoba, Spain, May 4–7 (2003).
7. V. IGNATIEV et al., “Physical and Chemical Properties of MSR Fuel/Coolant Salts,” *Proc. International Conference on Advanced Power Plants, ICAPP '03*, Cordoba, Spain, May 4–7 (2003).

8. E. RODRIGUEX-VIEITEZ, M. D. LOWENTHAL, E. GREENSPAN, and J. AHN, "Optimization of a Molten-Salt Transmuting Reactor," *Proc. Int. Conf. on the New Frontiers of Nuclear Technology: Reactor Physics, Safety and High-Performance Computing, PHYSOR-2002*, Seoul, Korea, October 7–10 (2002).
9. C. D. BOWMAN, "Once-Through Thermal-Spectrum Accelerator-Driven LWR Waste Destruction Without Reprocessing," *Nucl. Technol.*, **132**, 66–93, October (2000).
10. K. FURUKAWS et al., "Small MSR with a Rational Th Fuel Cycle," *Nucl. Eng. & Des.*, **136**, 157 (1992).
11. K. FURUKAWA, K. MITACHI, Y. KATO, S. E. CHIGRINOV, A. LECOCQ, and L. B. ERBAY, "Rational Pu-Disposition for ²³³U-Production by THORIMS-NES," IAEA-TECDOC-840, pp.169–181, International Atomic Energy Agency, (1995).
12. Y. HIROSE and Y. TAKASHIMA, "The Concept of Fuel Cycle Integrated Molten Salt Reactor for Transmuting Pu+MA from Spent LWR Fuels," *Proc. Global '01*, Paris, France, September (2001).
13. VERGNES and D. LECARPENTIER, "The AMSTER Concept (Actinides Molten Salt TransmutER)," *Nucl. Eng. Des.*, **216** (1) p. 43 (2002).
14. M. HRON and J. VANICEK, "Project SPHINX Spent Hot fuel Incineration by Neutron Flux: Experimental Verification of Design Inputs for Transmuter with Liquid Fuel Based on Molten Fluorides," *Proc. 11th International Conference on Emerging Nuclear Energy Systems*, September 29–October 4, 2002, Albuquerque, New Mexico (2002).
15. J. T. LONG, *Engineering for Nuclear Fuel Reprocessing*, Gordon and Breach Science Publishers, Inc. (1967).
16. M. BENEDICT and T. H. PIGFORD, *Nuclear Chemical Engineering*, McGraw-Hill Book Co., Inc. (1957).
17. *Actinide and Fission Product Partitioning and Transmutation, NEA–OECD Status and Assessment Report*, OECD, Paris, France (1999).