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DOE/ID-10422

Weapons-Grade Plutonium Dispositioning
Volume 1

Executive Summary

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Published June 1993

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Prepared for the
National Academy of Sciences
and for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-76ID01570

ABSTRACT

The Secretary of Energy requested the National Academy of Sciences (NAS) Committee on International Security and Arms Control to evaluate dispositioning options for weapons-grade plutonium. The Idaho National Engineering Laboratory (INEL) assisted NAS in this evaluation by investigating the technical aspects of the dispositioning options and their capability for achieving plutonium annihilation levels greater than 90%. Additionally, the INEL investigated the feasibility of using plutonium fuels (without uranium) for disposal in existing light water reactors and provided a preconceptual analysis for a reactor specifically designed for destruction of weapons-grade plutonium. This four-volume report was prepared for NAS to document the findings of these studies.

Volume 2 evaluates 12 plutonium dispositioning options. The INEL believes that if plutonium annihilation levels greater than 90% are desired, only those options that reprocess irradiated fuel can reasonably achieve this goal. The four options achieving the highest rating, in alphabetical order, are the Advanced Light Water Reactor with plutonium-based ternary fuel, the Advanced Liquid Metal Reactor with plutonium-based fuel, the Advanced Liquid Metal with uranium-plutonium-based fuel, and the Modular High Temperature Gas-Cooled Reactor with plutonium-based fuel.

Volume 3 considers a concept for a low-temperature, low-pressure, low-power-density, low-coolant-flow-rate light water reactor that quickly destroys plutonium without using uranium or thorium. This reactor concept does not produce electricity and has no other mission than the destruction of plutonium.

Volume 4 addresses neutronic performance, fabrication technology, and fuel performance and compatibility issues for zirconium-plutonium oxide fuels and aluminum-plutonium metallic fuels. Only the fabricability issues of carbide fuels were addressed. In addition, the effects of adding gadolinium, erbium, and europium were evaluated for obtaining negative temperature coefficients. For both the oxide fuels and metallic fuels, erbium was the best additive to develop negative temperature coefficients.

CONTENTS

TABLES

ACRONYMS

ABC	Accelerator-Based Conversion System
ALMR	Advanced Liquid Metal Reactor
ALMR-MB	Advanced Liquid Metal Reactor Maximum Burner Cycle
ALMR-R	Advanced Liquid Metal Reactor Reference Cycle
ALWR	Advanced Light Water Reactor
ALWR-MOX	Advanced Light Water Reactor with Mixed Oxide Fuel
ALWR-T	Advanced Light Water Reactor with Ternary Fuel
CISAC	Committee on International Security and Arms Control
DOE	U.S. Department of Energy
INEL	Idaho National Engineering Laboratory
MHTGR	Modular High Temperature Gas-Cooled Reactor
MOX	mixed oxide
MSR	Molten Salt Reactor
MT	metric ton
MW(t)	megawatt thermal power
NAS	National Academy of Sciences
PBR	Particle Bed Reactor
SS	stainless steel

INTRODUCTION

In 1992, the Secretary of Energy requested that the National Academy of Sciences (NAS) Committee on International Security and Arms Control (CISAC) evaluate various methods available for plutonium dispositioning. The Idaho National Engineering Laboratory (INEL) volunteered to provide technical support for the NAS evaluation and the CISAC Reactor Panel requested the following tasks be completed by the INEL:

1. Provide an independent comparison of the various reactor vendor options for the destruction of plutonium

2. Evaluate the feasibility of a nonuranium-bearing plutonium fuel for use in existing commercial light water reactor plants

3. Provide a conceptual analysis for a reactor specifically designed to fission plutonium without producing electricity or tritium.

The INEL prepared Volumes 1–4 in response to these requests.

The INEL is not advocating any specific reactor concept or technology discussed herein. The results are based on conceptual studies and analyses conducted by the INEL and on information provided to the U.S. Department of Energy (DOE) by organizations advocating specific concepts.

The effort included research by scientists, engineers, and staff of EG&G Idaho, Inc.; Westinghouse Idaho Nuclear Company; and the DOE Idaho Operations Office. EG&G Idaho had the lead responsibility for overall management and publication of the report. A summary of each task, as detailed in Volumes 2 through 4, follows.

VOLUME 2 SUMMARY

In response to a specific request from NAS, INEL staff gathered and evaluated information from the sponsors of reactor and accelerator-based options on the capability of their systems to annihilate plutonium (destruction of 90% to 99.9% of all plutonium isotopes). Sponsors for the various concepts were:

- Brookhaven National Laboratory—Particle Bed Reactor (PBR)
- General Atomics—Modular High Temperature Gas-Cooled Reactor (MHTGR)
- General Electric/Argonne—Advanced Liquid Metal Reactor (ALMR)
- Los Alamos National Laboratory—Accelerator-Based Conversion System (ABC)
- Oak Ridge National Laboratory—Molten Salt Reactor (MSR)
- Westinghouse Savannah River Company—Advanced Light Water Reactor (ALWR).

The following four areas were identified as the basis for the evaluations:

- Fuel status
- Reactor and accelerator system status
- Waste-processing status
- Waste-disposal status.

To supplement the original proposals, a set of questions was developed in each of these areas and transmitted to the sponsors. The answers to these questions, as well as the original sponsor proposals, were reviewed and evaluations of option capabilities were made. A summary of results for each of the four areas follows.

Fuel Status

Two fuel forms have been proposed for both the ALMR and ALWR options. A uranium-plutonium-based metal fuel was proposed for the ALMR reference fuel cycle (referred to as ALMR-R) and a plutonium-based metal fuel for a maximum burner fuel cycle (ALMR-MB). Both a mixed oxide (MOX) fuel and a ternary (T) fuel have been proposed for the ALWR to provide more rapid plutonium annihilation.

Fuel development of the ALWR-MOX and ALMR-R is essentially complete. However, experience with plutonium-based fuel fabrication is limited and requires demonstration and certification. As a result, the fabrication of plutonium-bearing fuels will be on the critical path if annihilation of a high percentage of all plutonium isotopes (90% or greater) is desired as a requirement.

Fuel that will allow annihilation of high percentages of plutonium in reasonable time frames is at the same stage of development for the ALMR-MB, ALWR-T, and MHTGR options. Significant development work will be required for the PBR fuel. Insufficient information on fuel development was provided by sponsors of the ABC and MSR, but the INEL's review indicates that substantial fuel development will be required.

The INEL believes that sponsor estimates for fuel fabrication facility costs and schedules are optimistic because experience with uranium oxide fuel cannot be directly extrapolated to plutonium-based fuel fabrication.

Operating costs and fuel development schedules are expected to be greater for plutonium-based fuel because plutonium must be handled remotely. However, operating costs are a small part of the overall costs of the plant.

Reactor and Accelerator-Based System Status

For proliferation reasons the capability to annihilate all plutonium isotopes was used as the measure of a concepts' effectiveness rather than the capability to annihilate just ^{239}Pu and ^{241}Pu isotopes. Therefore, the INEL chose total plutonium annihilation as a measure of a reactor's effectiveness in destroying plutonium.

The original ALWR option description proposed using MOX fuel for the dispositioning of plutonium. Use of MOX fuel to annihilate 50 MT of plutonium requires long periods of time or large numbers of reactors, and numerous fuel-reprocessing cycles. The ALWR-T and MHTGR options are both capable of annihilating high percentages (90%) of ^{239}Pu without reprocessing. However, for the ALWR-T, high annihilation percentages can only be reached through in reactor fuel assembly resident times significantly longer than current light water reactor fuel assembly experience. Further investigation is required to determine what effects the long exposure times will have on fuel assembly materials. A modified fuel management scheme must be used in the MHTGR to achieve annihilation of large fractions of all plutonium isotopes. This fuel management scheme moves the graphite blocks outside the primary shield for secondary irradiation. The PBR plutonium annihilation rates, derived by the concept sponsor, seem to be inconsistent with the known exposure history.

Calculations by option sponsors indicate that concepts with reprocessing can achieve near total plutonium annihilation in a shorter duration than nonreprocessing options (see Table 1).

There are technology development issues that must be resolved for all reactor and accelerator systems. For all options, criticality and reactivity control during reactor operation must be examined when annihilation of large percentages of plutonium is desired. Specific technology development issues for the ABC, PBR, and MSR options were not identified because these concepts are in a preliminary stage of conceptual development. It is clear that significant issues relating to materials, design, and fabrication would have to be resolved before these concepts could be constructed.

Table 1. Years required to annihilate various fractions of 50 MT plutonium for reactor and accelerator systems assuming reprocessing of the irradiated fuel.

Concept	MW(t)	90%	99.9%
ABC	4260	32.8	42
ALMR-R	4239	144.1	159.9
ALMR-MB	4239	37.9	42.1
ALWR-T	3636	NR	NR
MHTGR	4050	59.8	U
MSR	3030	53.2	59
PBR	3600	38.0	49.7

NR—no reprocessing proposed.
U—this level of annihilation is unachievable.

The INEL believes sponsor estimates for system development, construction costs, and schedules are optimistic. Sponsor estimates for startup and operational costs are expected to be similar to those of current operating facilities on a per reactor basis.

Waste-Processing Status

Waste processing is an integral part of the ABC, ALMR, and MSR. Technical development of the ALMR waste processing is under way at Argonne National Laboratory.

Waste processing for the ABC and MSR are not well developed for a plutonium-based fuel requiring process and component development. The PBR waste processing is not necessary because the particles would be packaged and sent to a waste-disposal facility. Waste-disposal issues for the other reactor concepts are mature for uranium-based fuels; however, many technical issues remain for plutonium-based fuels.

A detailed technical assessment of the waste-processing area was not performed as part of this report.

Waste-Disposal Status

Several waste-disposal options are possible using reactors or an accelerator. Each possible waste option has the following issues that must be considered in a comparative evaluation process:

- **Repository Availability.** The likelihood of waste going to the first geological repository is very low and plans for a second repository have not been initiated. Monitored storage of plutonium or its denatured form will be required for several decades.
- **Repository Control.** There are two key variables in the control of material in a repository—(1) control and containment of radioactive material (2) and control of criticality. Both require evaluation for all proposed options.
- **Waste Forms and Characterization Programs.** Any new waste forms will require characterization and waste-handling equipment performance testing prior to acceptance at a future geologic repository.

Although it does not appear that any concept has a notable advantage in the waste characterization area, the ABC, ALMR, and MSR concepts all have decreased repository requirements, such as radioactive lifetime of their final fuel form and minimal criticality control issues. Further study is required to characterize the implications of long-term storage of these waste streams.

Because insufficient time and information was available to the INEL to perform detailed comparisons of each concept, the INEL recommends further study of what it believes are the top four concepts—ALMR-MB (Advanced Liquid Metal Reactor Maximum Burner), ALMR-R (Advanced Liquid Metal Reactor Reference Cycle), ALWR-T (Advanced Light Water Reactor with Ternary Fuel), and MHTGR (Modular High Temperature Gas-Cooled Reactor).

VOLUME 3 SUMMARY

NAS requested that the INEL examine concepts focusing only on a mission of destroying plutonium and not producing electricity or other isotopes. Not considering other missions freed the INEL from several constraints used by proponents of other plutonium dispositioning concepts. Because of the design goal—a reactor having the sole function of plutonium destruction, INEL reactor designers had unusual flexibility in specifying reactor geometry and materials.

High reactor coolant temperatures are required to produce electricity efficiently. Although an estimated $12 billion per unit revenue from the sale of electricity is lost, some capital costs are reduced by eliminating turbines, generators, some support facilities, possibly some backup safety systems, and a thick-walled pressure vessel. Reactor design becomes simpler and more flexible, also reducing costs. The mission of destroying plutonium is not impeded by electricity load demand concerns and outages for maintenance of electrical generation and distribution systems.

Light water reactor technology was adopted to minimize technology development risk, ensure the greatest chance of success, and reduce costs. Low power densities, temperatures, pressures, and coolant flows were selected to enhance reactor safety.

All reactors without fertile materials (uranium or thorium) operating at the same power level and capacity factor annihilate plutonium at the same rate. In addition, this rate is faster than the rate of any reactor containing fertile materials.

Fuel forms chosen for use in any type of plutonium burner should:

- Possess the highest burnup capability
- Provide the highest degree of operational safety
- Offer the most cost-effective and efficient fabrication methods
- Minimize hazardous waste generation
- Offer the most cost-effective and efficient end-of-life disposal option.

Only two plutonium-only fuel forms have been tested for any length of time in a nuclear reactor; they are plutonium dispersed in an aluminum matrix and a plutonium oxide kernel embedded in carbon and sealed in a silicon carbide shell.

A major disadvantage of removing ^{238}U is the reduction or elimination of a prompt negative Doppler reactivity coefficient and a negative moderator temperature coefficient. A pure plutonium fuel type is not desirable in light water reactors because of the low mass loading per fuel rod (yielding short fuel cycles) and strong positive temperature coefficients. Any workable fuel composition must have a negative prompt temperature coefficient (i.e., reactor power decreases as temperature increases) for safety and control purposes.

A low-power-density, plutonium-burning reactor cooled by low-temperature, low-pressure light water flowing at low velocity was recommended. Primary coolant system flow is provided by pumps, but even greater safety advantages would be attained if a natural circulation cooling system could be employed. A reactor employing these concepts has multiple thermal-hydraulic and safety advantages including:

- The margin to critical heat flux is very large
- The time required to raise fuel temperatures to damage and melting point are significantly longer than current light water reactors
- Coolant flowing vertically upward in the core ensures initiation and continuance of natural circulation core cooling should the primary coolant pumps fail

- The open, nonchannelized, core flow arrangement avoids safety issues associated with flow instability in parallel channels

- The low energy stored in the coolant and structure minimizes the requirements placed on the containment

- The low pressure coolant system significantly reduces the safety risk due to loss-of-coolant accidents and is compatible with passive safety injection systems.

Table 2 presents conceptual parameters for the reactor; the ranges indicate the flexibility that exists in the concept to optimize fuel fabrication costs within acceptable safety limits.

Costs associated with construction, operation, decontamination and decommissioning, and waste processing and disposal must be considered for all facilities. These nuclear facility costs demand that methods, processes, and requirements be critically reviewed and improved if these facilities are to be affordable.

If and when several identified cost-reduction ideas are implemented, the overall cost of designing, constructing, and operating a reactor can be reduced significantly.

Several ideas are identified in the body of the report as having a potential for high reductions in net costs of a plutonium-burning concept.

Additional design studies are recommended in the areas of reactor physics and neutronics; materials; thermal transport system; accident scenarios; characterization of spent fuels; economics; and multiple use of the reactor for production of medical isotopes, tritium, and ^{238}Pu or for burning actinides.

Table 2. General parameters for a plutonium-burning reactor concept.

Parameters	Baseline	Possible range
Reactor power [MW(t)]	1000	—
Fuel material	PuAl	PuO_2/ TRISO
Cladding material	Al	SS or ZR
Coolant type	Light water	—
Moderator type	Light water	—
Fuel cycle length (yr)	3	1–5
Batch resident time (yr)	12	5–12

VOLUME 4 SUMMARY

Use of plutonium fuels in existing commercial light water reactor plant designs is attractive because it enables maximum exploitation of existing technology and infrastructure. The potential exists for weapons-grade plutonium dispositioning with the minimum development costs, shortest schedule, and minimal development risk. Plutonium fuels without uranium or thorium are most desirable because production of additional weapons materials can be avoided and the plutonium destruction rate is maximized. Unfortunately, the absence of fertile materials generally results in unacceptable (positive) temperature coefficients of reactivity. Acceptable temperature coefficients can probably be obtained with other resonance absorbers. Rare-earth elements gadolinium, erbium, and europium are attractive candidates.

Three categories of plutonium fuels are considered for use in commercial light water reactor designs: plutonium oxide fuels, aluminum-plutonium metallic fuels, and plutonium carbide fuels. Preliminary evaluations of the neutronic performance of the first two fuel types have been completed. Important neutronic performance characteristics examined include plutonium mass loading, resulting cycle length, prompt fuel temperature reactivity coefficients, isothermal temperature coefficients, and plutonium isotopic compositions. In general, reactivity coefficients are examined only at beginning of life.

Fabrication and performance issues have been examined for the three fuel categories.

Oxide Fuels

Plutonium oxide fuels compositions considered in the neutronic analyses include plutonium-zirconium oxides with thorium and/or the burnable poison additives of gadolinium, erbium, and europium. A standard pressurized water reactor uranium oxide (UO_2) fuel was analyzed as a reference case for comparison purposes.

The plutonium mass loadings obtainable in the fuel forms containing only PuO_2 and ZrO_2 are unacceptably low and the prompt temperature coefficients are positive. Acceptable plutonium mass loadings and negative reactivity coefficients can be obtained with plutonium fuels containing thorium, but the liability of ^{233}U production must be accepted. High plutonium mass loadings and negative prompt temperature coefficients are obtained with any of the three rare-earth additives. For the light water reactor lattice configuration and compositions examined, accepted isothermal coefficients are obtained with gadolinium. With europium as an additive, the isothermal temperature coefficient is negative over the operational temperature range. At low europium mass loadings, the isothermal temperature coefficient is positive at low temperatures.

Fabrication processes for traditional mixed uranium-plutonium oxide fuels are well established and believed adaptable to fabrication of zirconium-plutonium oxide fuels containing rare-earth additives. The use of ZrO_2 as a diluent for PuO_2 creates a potential problem in obtaining a homogeneous fuel form. Some development work will be required regardless of the process selected. Three powder preparation techniques are in current use for mixed oxide fuels. They are the coconversion process using thermal microwave denitration, the integrated dry route using mechanical blending, and the coprecipitating process. The latter two are the currently favored processes, but are unsuitable if fuel reprocessing is ultimately required to achieve the desired burnup. The coconversion process appears appropriate whether or not fuel reprocessing is required. Conventional fabrication techniques can be adapted to manufacture pellets from the mixed oxide powders. The preferred technique will depend on the powder fabrication method that is used and on the desired fuel pellet properties and performance characteristics.

Metallic Fuels

Aluminum-plutonium metallic fuels are examined for use in existing light water reactor designs operated at low temperature and low pressure. The emphasis is on use of the plutonium-aluminum fuel in a typical light water reactor lattice configuration. The fuel consists of plutonium-aluminum [$PuAl_4(Al)$] and the cladding is aluminum metal. Additives of gadolinium, erbium, or europium are explored to achieve negative temperature coefficients. Plutonium mass loadings are slightly higher than for the pure PuO_2-ZrO_2 fuel form, but still unacceptably low. The prompt temperature coefficients are small but negative for plutonium. The isothermal temperature coefficient is strongly positive. High plutonium mass loadings are obtained with any of the three rare-earth additives considered. Prompt fuel temperature coefficients are negative for both erbium and europium; the gadolinium case was not examined. The isothermal temperature coefficients are strongly negative for erbium. The beginning-of-cycle isothermal temperature coefficient with europium as an additive is slightly positive for the examined cases . Negative temperature coefficients are expected when ^{153}Eu can be included in the evaluation.

Several plutonium-aluminum fuel forms and fabrication techniques were developed or explored during plutonium recycle studies about 30 years ago. Fabrication techniques vary depending on the plutonium-aluminum composition. The eutectic composition is approximately 2 at% (15.64 wt%) plutonium. Compositions with plutonium content lower than the eutectic composition are reasonably ductile and can be formed by rolling or extrusion. Compositions with higher plutonium content tend to be brittle and difficult to form. Low plutonium content alloys suitable for use in a plutonium-burning light water reactor have been fabricated in several forms. Successful techniques included hot extrusion of plutonium-aluminum alloy fuel cores for insertion into zircaloy tubes, coextrusion of plutonium-aluminum fuel cores with aluminum cladding, cylindrical-shaped aluminum cladded plates, and plates fabricated using the picture-frame technique. Fabrication of high plutonium content alloys is more difficult. If high plutonium content alloys were necessary, the composition ductility can be improved by the addition of nickel, zirconium, or titanium. High plutonium-content alloys have been successfully fabricated into plate fuels. Fabrication processes will need to be more automated than previously employed to meet dose exposure requirements and to minimize wastes.

Carbide Fuels

Uranium-plutonium carbide fuels were considered because of their higher thermal conductivity and lower operating temperatures compared to oxide fuels. Because of the limited plutonium carbide fabrication experience base, fabrication must be assessed based on existing experience with the uranium carbide fuels. Two fuel forms are believed to be suitable—one where carbide pellets are contained in a metal jacket and the other consisting of carbide particles dispersed in a graphite matrix. Although this appears to be a reasonable extension of uranium carbide experience, substantial development would be required.

In summary, this study focused on three fuel forms (oxides, aluminums, and carbides) to contain plutonium for dispositioning in a nuclear reactor. The plutonium oxide fuel form is for use in a commercial light water reactor: the aluminum-plutonium fuel form is for a noncommercial, low-temperature reactor; and the plutonium carbide fuel form is for a graphite reactor. In all fuel forms, the highly reactive weapons-grade plutonium must be balanced with a thermal resonance absorber (i.e., a fertile heavy metal material such as depleted uranium or thorium or a burnable poison such as gadolinium, erbium, europium). If the requirements for the fuel necessitates the use of a burnable poison in place of depleted uranium or thorium, a fuel development program should be initiated very early in the program.

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8/24/94

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