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MIXED OXIDE CONVERSION FACILITY ALTERNATIVE CONCEPTUAL DESIGNS

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ABSTRACT

Allied-General Nuclear Services has developed alternative design concepts for three mixed oxide conversion facilities. The design, relative proliferation resistance, and cost of these concepts are compared.

SUMMARY

Allied-General Nuclear Services recently performed studies to evaluate alternative proliferation-resistant flowsheets of the uranium-based LWR fuel cycle. The alternatives evaluated consist of coprocessing schemes with either a gamma or a heat spike added. A literature search and evaluation were performed to select a process technology for mixed oxide coconversion. The COPRECAL process was chosen as the most suitable conversion process technology.

Three alternative mixed oxide conversion facility design concepts were prepared based on the COPRECAL technology. These alternative concepts are compared to a pure plutonium conversion facility. Facility designs, relative proliferation resistance, and cost estimates are discussed.

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INTRODUCTION

Allied-General Nuclear Services (AGNS) recently performed studies for the Department of Energy (DOE) to evaluate alternative flowsheets that could feasibly increase the proliferation resistance of the uranium-based LWR fuel cycle. These studies were performed in support of DOE programs which provided information to the International Nuclear Fuel Cycles Evaluation (INFCE).

The alternative flowsheets evaluated were combinations of uranium/plutonium partitioning options and spiking variations. The options which were considered consisted of complete partitioning (100% plutonium which is the classic Purex process), partial partitioning (75% uranium-25% plutonium), and no partitioning (99% uranium-1% plutonium). The spiking variations included adding (or leaving) highly radioactive fission products with the fissile material or increasing the heat content of the plutonium.

Of the numerous possible combinations of these alternatives, three were chosen for conceptual design development. All three alternatives involve partial partitioning (or coprocessing) of the plutonium and uranium (75% uranium-25% plutonium). In the first alternative, referred to as the low-decontamination factor (low-DF) alternative, the mixed nitrate product stream from the separations process is highly contaminated with fission products which remain with the plutonium-uranium mixture through conversion and fuel fabrication. In the second alternative, referred to as the high-DF alternative, the mixed nitrate product is decontaminated from fission products. The mixed nitrate is converted to the mixed oxide which is blended with a solid Co-60 spike, equivalent to the fission product level in the low-DF alternative. The Co-60 spike is added immediately following conversion but prior to storage, shipment, or fuel fabrication. In the third alternative, referred to as the heat spike alternative, an increased level of Pu-238 is induced into the spent fuel by mixing recovered Np-237 into the fuel prior to irradiation in an LWR. These alternatives are compared to the classic Purex process which is referred to as the base case. The base case and alternative flowsheets are summarized in Figure 1.

CONVERSION PROCESS

The interim product stream from the separations process for all three alternatives considered is an aqueous mixture of plutonium and uranium nitrate. This is compared to the base case where the interim product stream is pure aqueous plutonium nitrate solution. The processes normally used to convert plutonium from the nitrate to the oxide (oxalate or peroxide precipitation) were not considered feasible for conversion of plutonium-uranium mixtures. Therefore, a study was performed to identify and compare processes to coconvert the mixed nitrate solution to the mixed oxide.

A literature search was conducted to identify candidate process technologies. A series of criteria was developed to provide an objective comparison of these processes. The following processes were identified as being capable of coconverting mixed nitrate solutions:

(1) Microsphere Production by Sol-Gel (ORNL)
(2) Coprecipitation with Ammonium Hydroxide (NUMEC)
(3) Coprecipitation with Oxalic Acid (LASL)
(4) Direct Fluid Bed Codenitration (Argonne)
(5) Coprecipitation Direct Calcination (GE).

The following basic criteria were selected to provide the evaluation technique for comparison of the processes:

(1) The process must enhance diversion resistance.
(2) The final product form must be suitable for reactor fuel.
(3) The process must be technically feasible.

Enhancement of diversion resistance was considered to be the capability of the process to coconvert the mixed nitrate to the mixed oxide with minimal possibility for plutonium, uranium, or spike separation. The mixed oxide product, to be acceptable as reactor fuel, must be sinterable to a minimum of 90% of theoretical density and must be homogeneous. The technical feasibility criterion was subdivided into categories as follows:

- Stage of development
- Relative complexity
- Continuous versus batch operation
- Ease of control
- Recycle stream compatibility
- Relative throughput
- Materials of construction required
- Impurity decontamination
- Product solubility
- Inherent process safety.

In general, a process must meet all three of the basic criteria to qualify as the technical basis for the design. Failure to meet either the diversion resistance or the fuel suitability criteria disqualified the process from consideration. Failure to meet the technical feasibility criteria was considered less significant, because further development or process alterations can result in solutions to technical problems.

At the time of this study (January 1978), each process was evaluated as described in the open literature or by direct contact with the developers of the process. The results of this evaluation are presented in Table 1.

The Sol-Gel, ammonium hydroxide coprecipitation, and oxalic acid coprecipitation processes fail the proliferation resistance criterion. These processes either require complete separation of the plutonium from the uranium or provide an easy means for separation within the process. The direct codenitration process meets the proliferation resistance criterion but, at the current stage of development, has failed to produce a product that is acceptable as reactor fuel. This leaves the coprecipitation-direct calcination (or COPRECAL) process as the most suitable one. The COPRECAL process meets all three of the basic criteria and was, therefore, selected as the technical basis for the design concept.

The COPRECAL process offers no easy means of plutonium-uranium separation. Separation can be readily accomplished in other processes such as those involving precipitation and filtration by simply adjusting chemical flows. Plutonium-uranium separation could only be accomplished in a facility utilizing the COPRECAL technology after extensive equipment and piping modifications. The time and level of effort needed to perform such modifications to in-cell process equipment and accomplish separation results in an extremely low probability for successful diversion of fissile material without timely warning.

The COPRECAL process was originally developed by the General Electric Company. Process development is continuing at the Savannah River Laboratory. The process utilized in these alternative conceptual designs represents a scale-up from the General Electric development work. The following is a description of the scaled-up COPRECAL process which is shown schematically in Figure 2.

The mixed nitrate product solution from the separations process is characterized as follows:

- 100 grams per liter plutonium
- 300 grams per liter uranium
- 2 to 6 molar nitric acid
- 200 kilograms of heavy metal (uranium plus plutonium) per day.

Because of separation process variations, the first step of the COPRECAL process involves adjustment of the heavy metal and nitric acid concentrations as required. The mixed nitrate solution is combined with excess ammonium hydroxide at ambient temperature and high pH ($\leq$11). The plutonium and uranium precipitate as plutonium hydroxide and ammonium diuranate, respectively. The precipitation reaction is very rapid and essentially complete.

The resultant slurry is injected into a fluid bed calciner where it is calcined to UO_3-PuO_2. The UO_3-PuO_2 flows to a second fluid bed where it is contacted with a dilute hydrogen gas stream and is reduced to UO_2-PuO_2. The UO_2-PuO_2 flows to a third fluid bed where it is contacted with carbon dioxide to stabilize the mixed oxide to approximately $UO_{2.07}$-PuO_2. The stabilized mixed oxide flows to a fluid bed cooler where it is cooled to less than 38°C. The mixed oxide product is

blended, sampled, and then transferred to bulk storage or prepared for shipment. Off-gas from the fluid beds is collected and scrubbed, passes through a condenser, and is filtered. Scrubber solution and condensate are recycled.

The COPRECAL process is a suitable technology for conversion of the mixed nitrate to the mixed oxide in all three alternative designs. The initial process development was carried out using mixed plutonium-uranium compounds corresponding to the high-DF alternative. The quantity of fission products in the low-DF alternative is small and does not affect the chemistry of the COPRECAL process. The amount of additional heat, hydrogen, and alpha radiation generated by the process materials in the heat spike alternative would have no effect on the COPRECAL process.

FACILITY DESIGN COMPARISON

The three alternative design concepts, high-DF, low-DF, and heat spike, were developed using the COPRECAL technology to compare the relative proliferation resistance and cost. The designs were sized to support a 1500 MTU/year separations facility. Because the basic process technology is unaffected by the alternative, the process equipment is very similar in all cases. In all three designs, the conversion process is accomplished in four separate independent parallel conversion lines with a combined instantaneous processing rate of 360 kilograms of heavy metal per day. Feed solution receipt and adjustment, mixed oxide storage, and waste treatment are similar for all three alternatives.

The high radiation levels emitted from LWR recycle plutonium makes glovebox conversion processing impractical. Therefore, in all three alternatives, the process operations are conducted in remotely operated shielded cells. The differences among the alternative designs are in the amounts and types of shielding and in the maintenance modes. In the low-DF alternative, maintenance is generally carried out remotely. A limited amount of maintenance can be carried out by contact or in shielded gloveboxes following removal of process materials and extensive decontamination. Remote repair and decontamination facilities are expensive to build and operate. The high-DF and heat spike alternatives offer somewhat wider latitude for operation and maintenance than the low-DF alternative. Operations involving very dilute process materials or where flushing of process materials can be done readily may be carried out in shielded gloveboxes. Contact or glovebox maintenance is considerably less expensive than remote maintenance.

In all three alternative designs, the facility consists of a central core of cells. An example of the central cell core arrangement is shown in Figure 3. The cells contain all processing equipment including the feed adjustment tankage, conversion equipment, mixed oxide powder handling equipment, bulk MOX storage tankage, off-gas treatment

equipment, and waste recycle and disposal equipment. The cells are surrounded by viewing and operating galleries at appropriate levels. The galleries, in turn, are surrounded by various nonoperating support areas. The building is a single, monolithic reinforced concrete structure for all three designs.

PROLIFERATION RESISTANCE

Coprocessing with 25% plutonium/75% uranium is common to the three reprocessing alternatives considered. This is compared to the classic reprocessing cycle which completely separates the plutonium and uranium. The net effect of coprocessing is dilution of the fissile plutonium with nonfissile uranium. At this 25/75% dilution, diversion of an estimated 200 kilograms is required to obtain a weapon-usable quantity of fissile material. This amount of fissile material is considerably above the safeguards significant quantity of special nuclear material, which is 8 kilograms of plutonium or 32 kilograms of mixed oxide. A nuclear material accountancy system for a mixed oxide conversion facility such as the ones described here would be designed for timely detection of the diversion of less than 32 kilograms of mixed oxide. This more than satisfies the proposed accountancy requirements of 10 CFR 70.

Spiking with a gamma emitting radionuclide at the proposed deterrent level of 1000 R/hour greatly increases the difficulty of both protracted covert and abrupt overt diversion. Diversion of gamma spike protected material requires heavy shielding and remote operation for the diversion and for subsequent purification activities. The probability for detection of diverted nuclear material is significantly increased by the high level of radiation emitted by the gamma spike.

In the low-DF alternative, the fissile material is always protected by the gamma spike. In the high-DF alternative, the plutonium and uranium are initially decontaminated from the fission products then a gamma spike, such as Co-60, is added following conversion, prior to mixed oxide storage, fabrication, or transport. The difference in proliferation resistance between the continuously protected low-DF alternative and the high-DF alternative is minimal. In both cases, process operations are carried out in inaccessible, remotely operated cells. Access to the fissile material is greatly decreased in both alternatives by physical barriers such as reinforced concrete cell walls and steel process vessels. The marginal increase in proliferation resistance in the low-DF alternative is accompanied by a significant increase in cost and operational difficulties. The increased biological shielding required, the increased number of operations carried out in shielded cells, and the problems associated with remote decontamination and maintenance all serve to significantly raise capital and operating costs for the low-DF alternative. Additionally, the presence of the highly radioactive gamma spike during the conversion process, as in the

low-DF alternative, increases the difficulty in obtaining analytical data hence reduces the accuracy of input data for a material accountancy system.

The heat spike alternative consists of increasing the Pu-238 content of reactor-generated plutonium above usual levels. This is achieved by recycle of Np-237 and U-236. The heat spike alternative would increase the proliferation resistance of the fuel cycle by decreasing the attractiveness of the fissile material as weapons material. While it does not render the plutonium useless for weapons purposes, it significantly increases the difficulty of weapons fabrication and assembly, requires a continuous heat removal system, and increases the quantity of plutonium required to fabricate a weapon. The heat spike alternative has the advantage over the other alternatives considered in that the spike (Pu-238) cannot be removed by chemical means.

Colocation of the subsequent fuel fabrication facility with the conversion and reprocessing facilities would eliminate the need for transport of fissile material except in the form of reactor fuel rods. Any of the three alternative conversion facility designs described here would be vulnerable to only the most sophisticated threats.

COST COMPARISON

Capital cost estimates were developed for each of the three alternative conversion process facilities. These cost estimates reflect the relative processing difficulty associated with the means of protecting the fissile material. These cost estimates were made in 1978 and 1979 and are escalated to mid-1980 dollars. They include the total cost of engineering and construction. Sufficient design effort has been expended to support these cost estimates to an accuracy ±25%.

All three processing alternatives, low-DF, high-DF, and heat spike, involve much the same basic process equipment, central cell structure, support equipment, and general facility layout. All three alternatives provide for receipt of the mixed nitrate solution, conversion of the mixed nitrate to the mixed oxide, storage capacity for one year's production of mixed oxide, and capability to load the mixed oxide into shipping containers for transport off site. The major differences among the alternatives are in the areas of shielding, remote operations, and related processes. Comparative capital costs are summarized in Table 2.

The low-DF alternative is the most costly facility to construct, estimated at $272 million. The high level of gamma radiation requires significantly more shielding. The low-DF cells require 36 inches of concrete wall thickness compared to approximately 24 inches in the high-DF and heat spike alternatives and 18 inches for the reference plutonium conversion facility. Additionally, the high level of radiation emitted by the gamma spiked process material requires that more of the support operations be conducted in remote operation cells.

Increased remote maintenance and remote decontamination equipment are likewise required.

The high-DF alternative conversion facility is estimated to cost $236 million to build. The reduced cost, compared to the low-DF facility, reflects the lower amount of shielding required and the greater utilization of glovebox and contact-operated equipment. The high-DF alternative includes equipment to receive, prepare, and blend a solid gamma spike material with the mixed oxide. This spike preparation equipment is estimated to add $15.5 million to the cost of the high-DF facility.

The heat spike alternative conversion facility is estimated to cost $221 million to build. Processing heat spiked (increased Pu-238) recycled LWR mixed oxide presents only minor differences compared to nonheat-spiked LWR mixed oxide. The added heat generated by the nuclear material is small compared to process heat generation. Heat removal is significant only in storage areas where large quantities of process materials are located. The heat spike has no effect on the conversion process. Shielding wall thicknesses increase only 11% for the heat spike alternative compared to the high-DF alternative. The lower total cost of the heat spike alternative compared to the high-DF alternative reflects the elimination of the gamma spike preparation equipment and the heavy shielding required for the gamma spiked mixed oxide in the bulk storage and shipping container preparation cells.

The heat spike conversion facility contains a separate process to convert neptunium nitrate to the oxide. This process is included in support of the neptunium recycle portion of the heat spike alternative fuel cycle. The neptunium conversion process is estimated to add $16.5 million to the total cost for the heat spike alternative conversion facility.

To put these costs into perspective, they are compared with the classic LWR fuel cycle. The estimated cost to construct a pure plutonium conversion facility of comparable capacity designed for LWR recycle plutonium is $166 million. These cost estimates are summarized in Table 2.

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TABLE 1

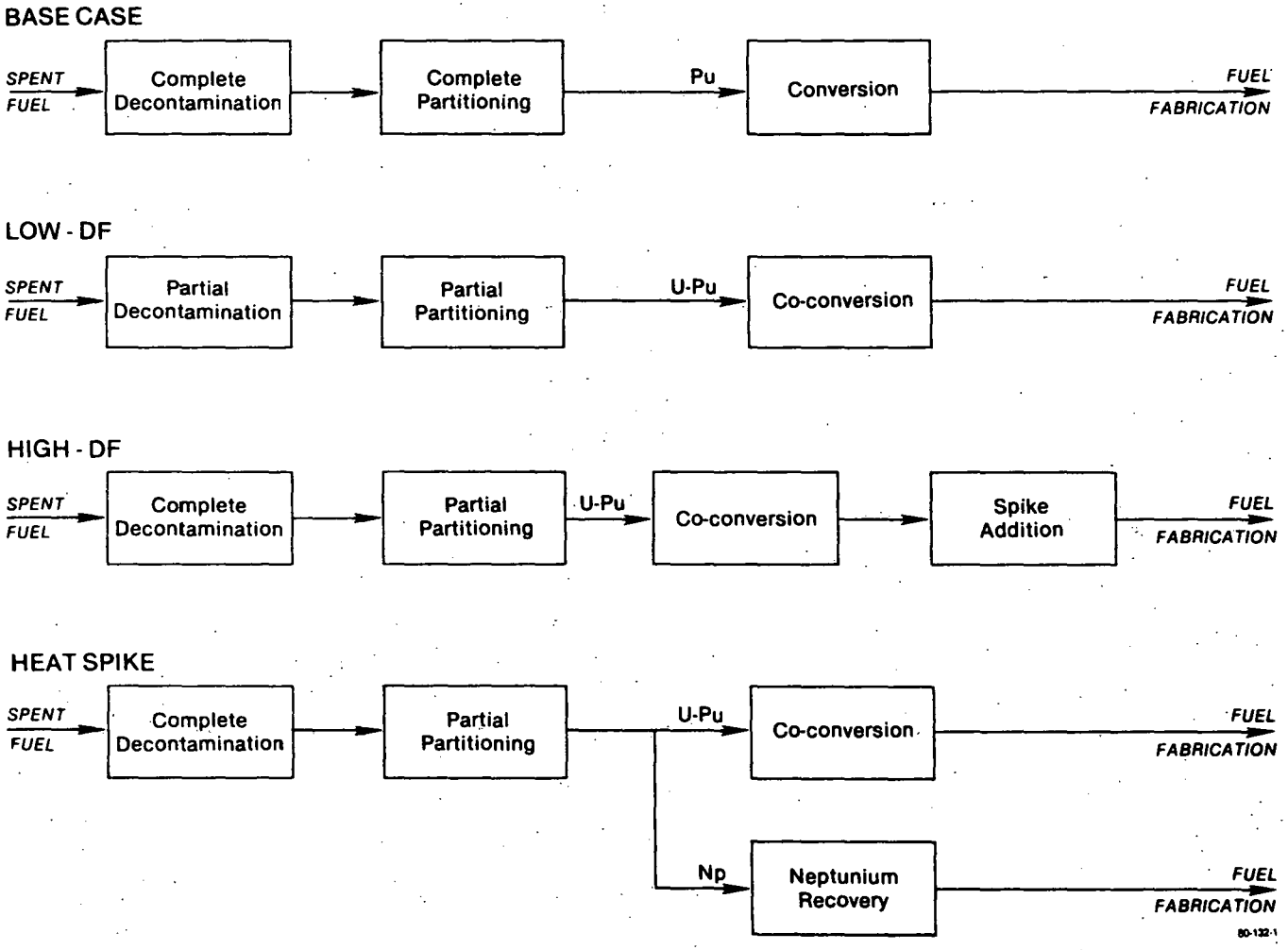
PROCESS EVALUATION

Process / Criteria	Sol-Gel	NH_4OH Co-ppt	Oxalic Co-ppt	Direct Denitration	COPRECAL
Proliferation Resistance	Unfavorable	Unfavorable	Unfavorable	Favorable	Favorable
Product Suitability	Unfavorable	Favorable	Unknown	Unfavorable	Favorable
Technical Feasibility	Unfavorable	Favorable	Favorable	Favorable	Favorable

TABLE 2

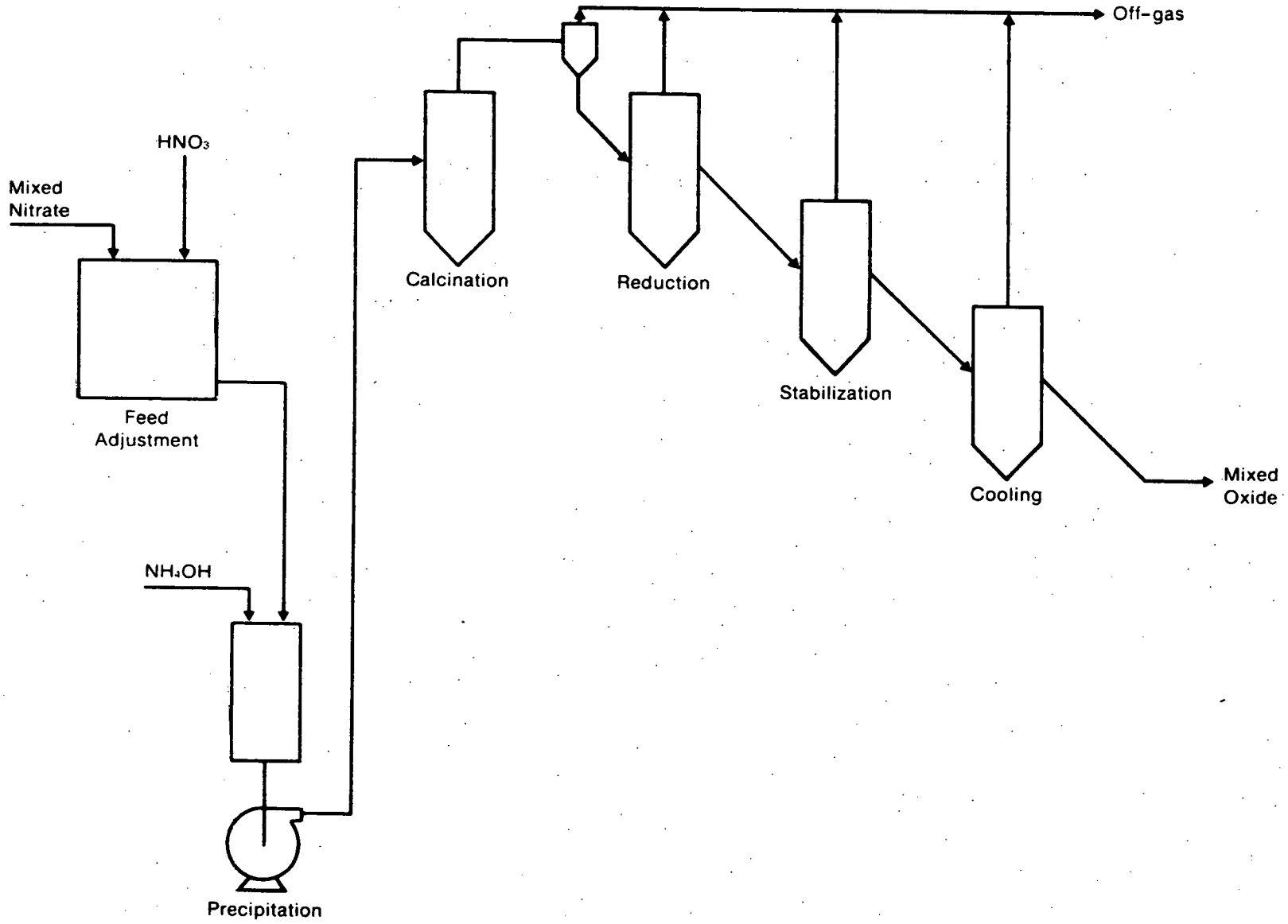
CAPITAL COST COMPARISON

Alternative	Capital Cost (mid-1980 dollars x 10^6)
Low-DF	272
High-DF	236
Heat Spike	221
Plutonium Conversion	166



COPROCESSING ALTERNATIVES

FIGURE 1



COPRECAL PROCESS

FIGURE 2

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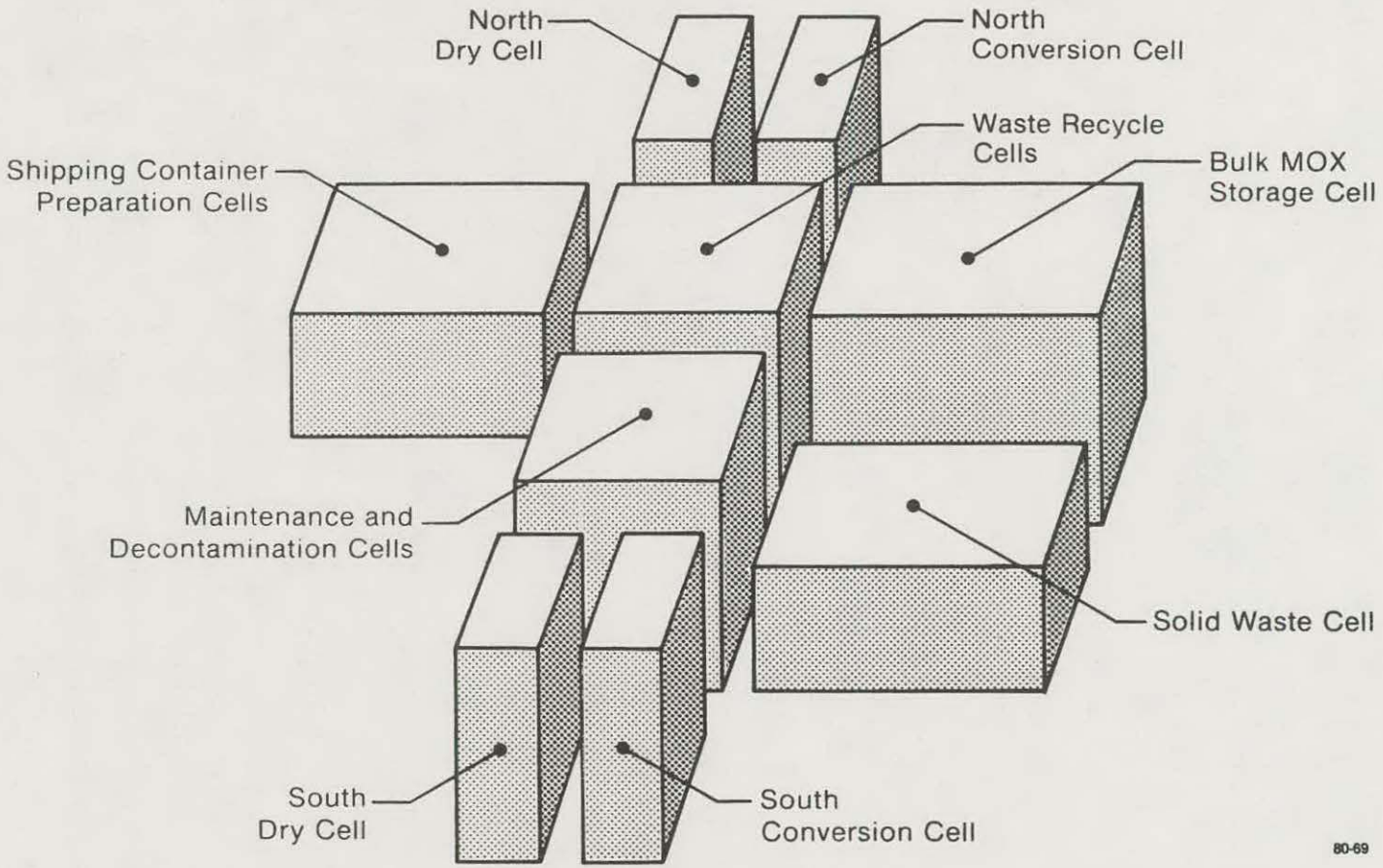

CENTRAL CELL ARRANGEMENT

FIGURE 3

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