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CRITICALITY RESEARCH IN SUPPORT OF CHEMICAL REPROCESSING IN THE THORIUM FUEL CYCLE TECHNOLOGY PROGRAM-BASIC PROCESS DESCRIPTION

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[redacted]

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ABSTRACT

The basic processes for reprocessing thorium based light water reactor type fuels are defined for use in identifying criticality data needs. The Reference Thorium Fuel Cycle is used as the primary fuel cycle. Material forms and compositions are described for each major processing step. These forms consist of nitrates and oxides of Th - U - Pu combinations. Fuel fabrication and fuel pool storage facilities are also defined to the extent they interact with fuel reprocessing.

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CRITICALITY RESEARCH IN SUPPORT OF CHEMICAL REPROCESSING IN THE THORIUM FUEL CYCLE TECHNOLOGY PROGRAM - BASIC PROCESS DESCRIPTION

INTRODUCTION

In the design and operation of fuel reprocessing plants, a sound understanding of the criticality parameters involved is required to achieve a safe and economical facility. The purpose of this document is to establish a basic process description for use in identifying criticality data needs in the thorium based Light Water Reactor (LWR) type fuels.

Several fuel cycle options are currently being considered for these thorium based fuels.(1) Practically all combinations of ^{235}U, ^{238}U, ^{233}U, Pu, and Th are covered in these various options. As the initial basis for establishing the criticality data needs in these fuel cycles, the Reference Thorium Fuel Cycle(1) has been chosen and is the primary fuel cycle considered in this document.

PROCESS DESCRIPTION

The flow diagram for the proposed Reference Thorim Fuel Cycle[1] is shown in Figure 1. The fuel cycle consists of six different types of facilities. These are:

- A - Uranium and Thorium mines
- B - Uranium Enrichment Plant
- C - Fuel Fabrication Facilities
- D - Light Water Reactors
- E - Spent Fuel Storage Facilities
- F - Chemical Reprocessing Plants

The top row of blocks in Figure 1 represent the present LWR fuel cycle. The facilities indicated in the additional blocks are needed to implement a complete thorium based fuel cycle. Although this document is primarily concerned with chemical reprocessing plants, fuel fabrication and fuel storage facilities are also covered because of their close interactions with fuel reprocessing.

The fuel fabrication plants include both remote operations, for recycled thorium/uranium (C3) and plutonium/thorium (C4) fuel, and contact operations for non-recycled thorium/uranium fuel (C2). Currently, an oxide fuel form is the primary candidate for the thorium based fuels because of the greater experience with oxide fuel irradiation. The oxide form could be either as a sintered powder or as compacted microspheres.

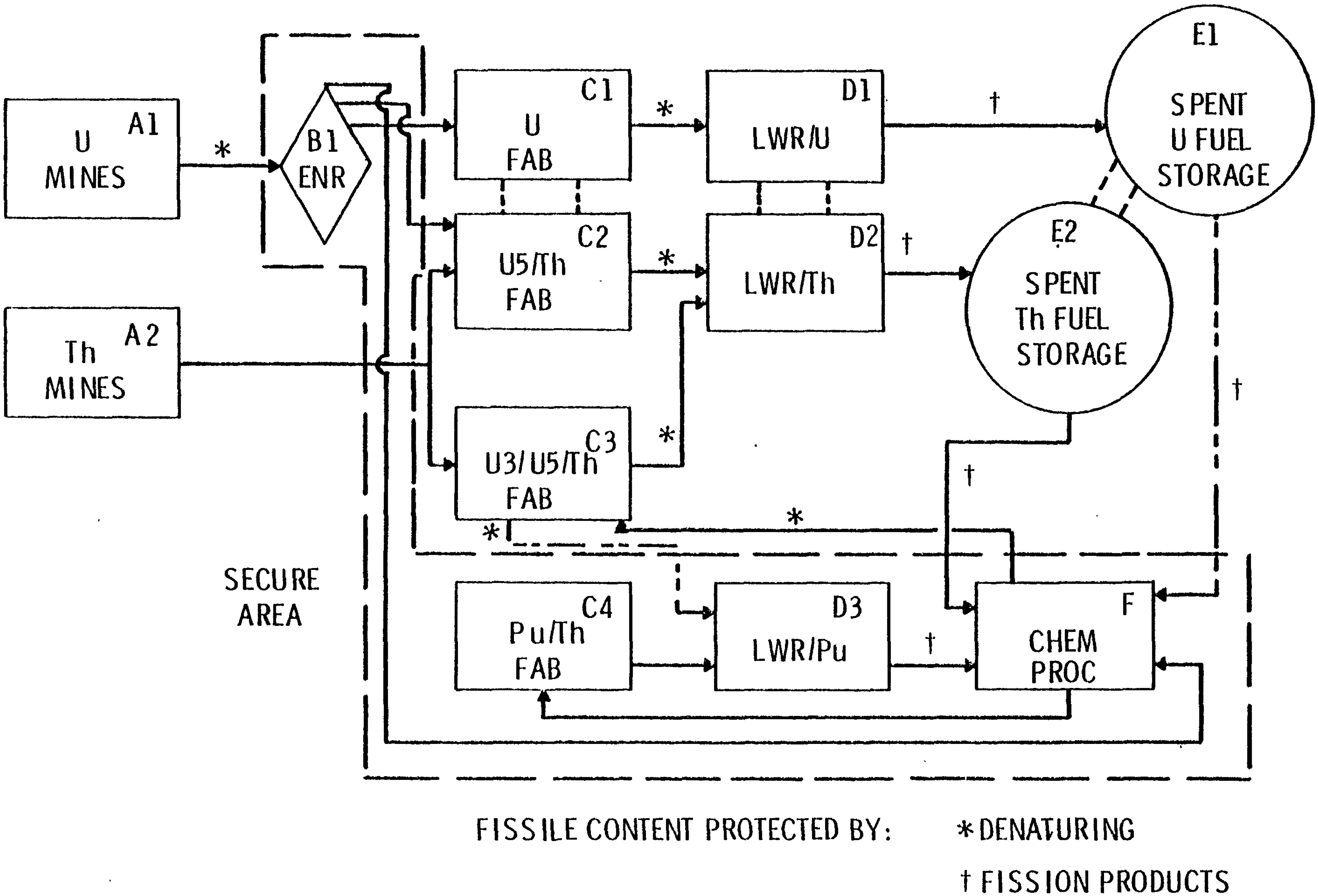

FIGURE 1. Proposed Reference Thorium Fuel Cycle(1)

A typical flow sheet for fabricating LWR sintered oxide fuel pellets is shown in Figure 2.[2] A flow sheet for the Oak Ridge Sol-gel process for microsphere fuel fabrication is shown in Figure 3.[3] The sol-gel oxide production process could be located either in the fabrication plant or reprocessing facility. The flow sheet for this process is shown in Figure 4.[3] The fissile material form at various process steps for fuel fabrication and sol-gel preparation are presented in Appendix A.

The criticality safety limits placed on spent fuel storage pools at reactor sites or in facilities dedicated to storage are similar to those which could be used in the storage pool of a chemical reprocessing plant. The reprocessing plants in the referenced fuel cycle (F in Figure 1) must have the capability for reprocessing three types of fuel; uranium, uranium/thorium mixtures, and plutonium/thorium mixtures. Although one plant could be built to handle all fuels, equipment designed for thorex solvent extraction will not be optimum for Purex operation, and vice versa. All fuels will be considered to contain uranium, plutonium, and thorium. A flow diagram for uranium/plutonium/thorium reprocessing is shown in Figure 5.[2] The solvent extraction employs the acid thorex process. A conceptual flowsheet for this process is given in Figures 6 and 7. The fissile material form at various process steps for reprocessing are presented in Appendix B.

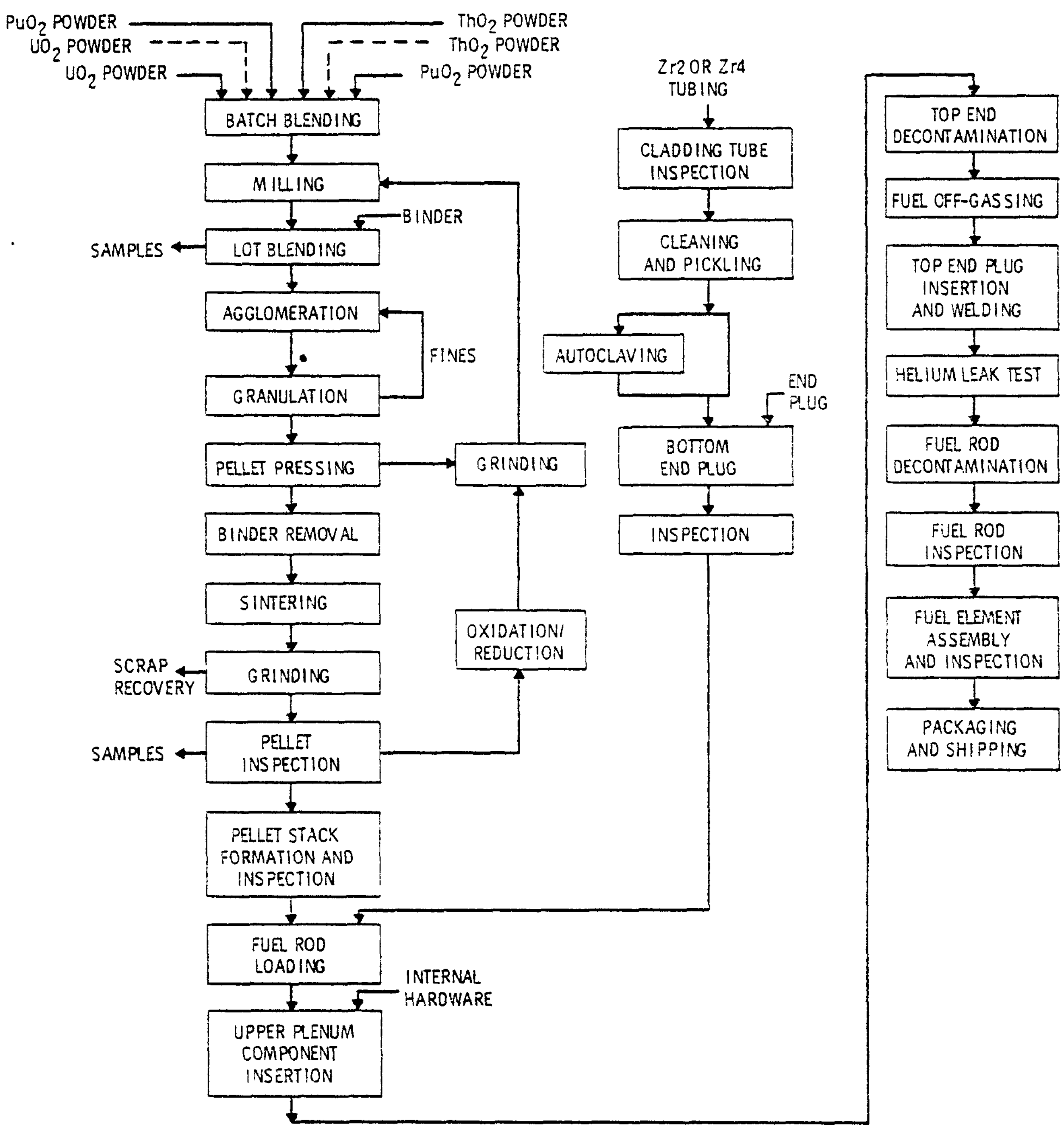

FIGURE 2. Oxide Pellet Fuel Element Fabrication(2)

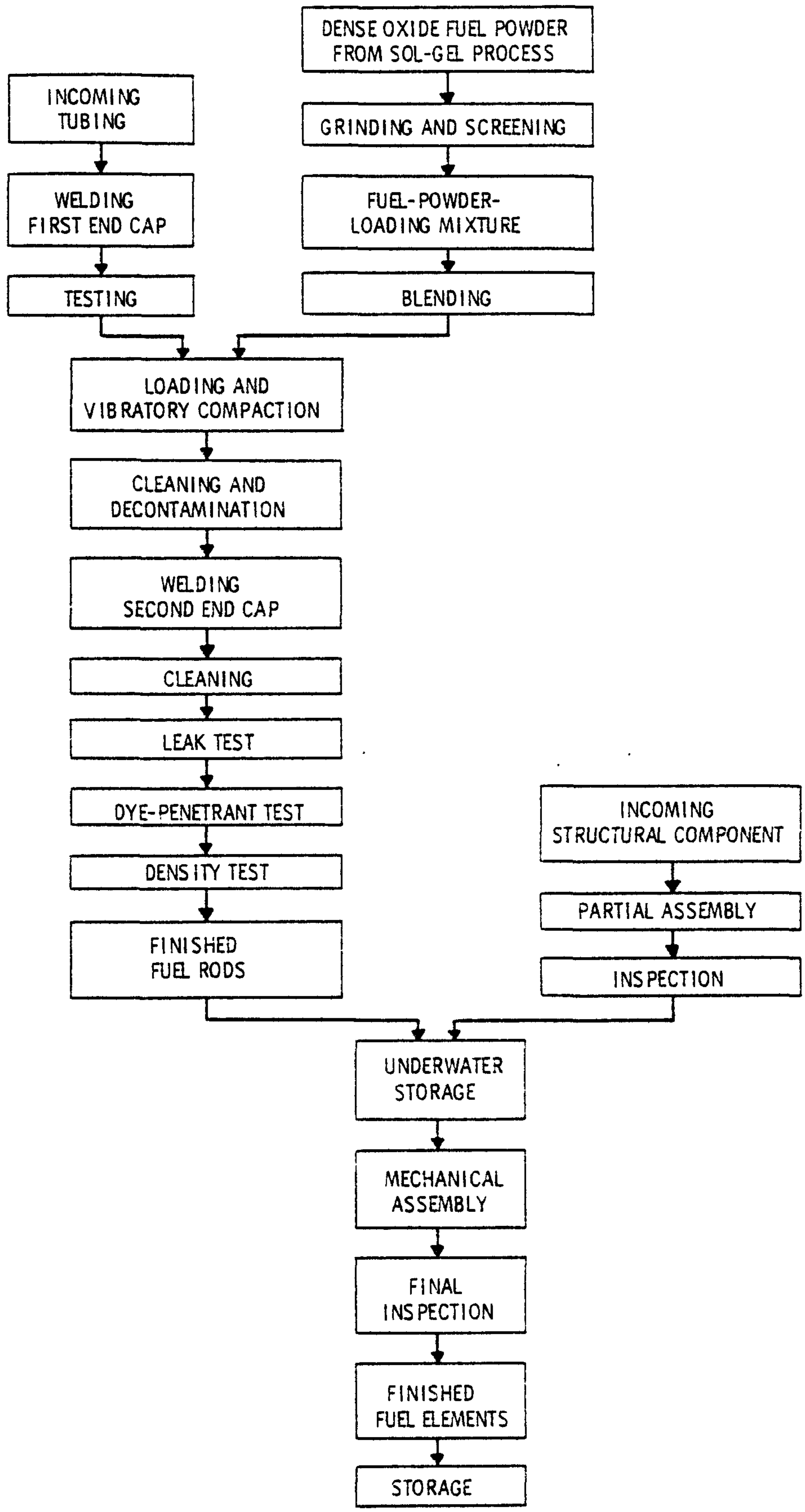

FIGURE 3. Sol-gel Fuel Element Fabrication[3]

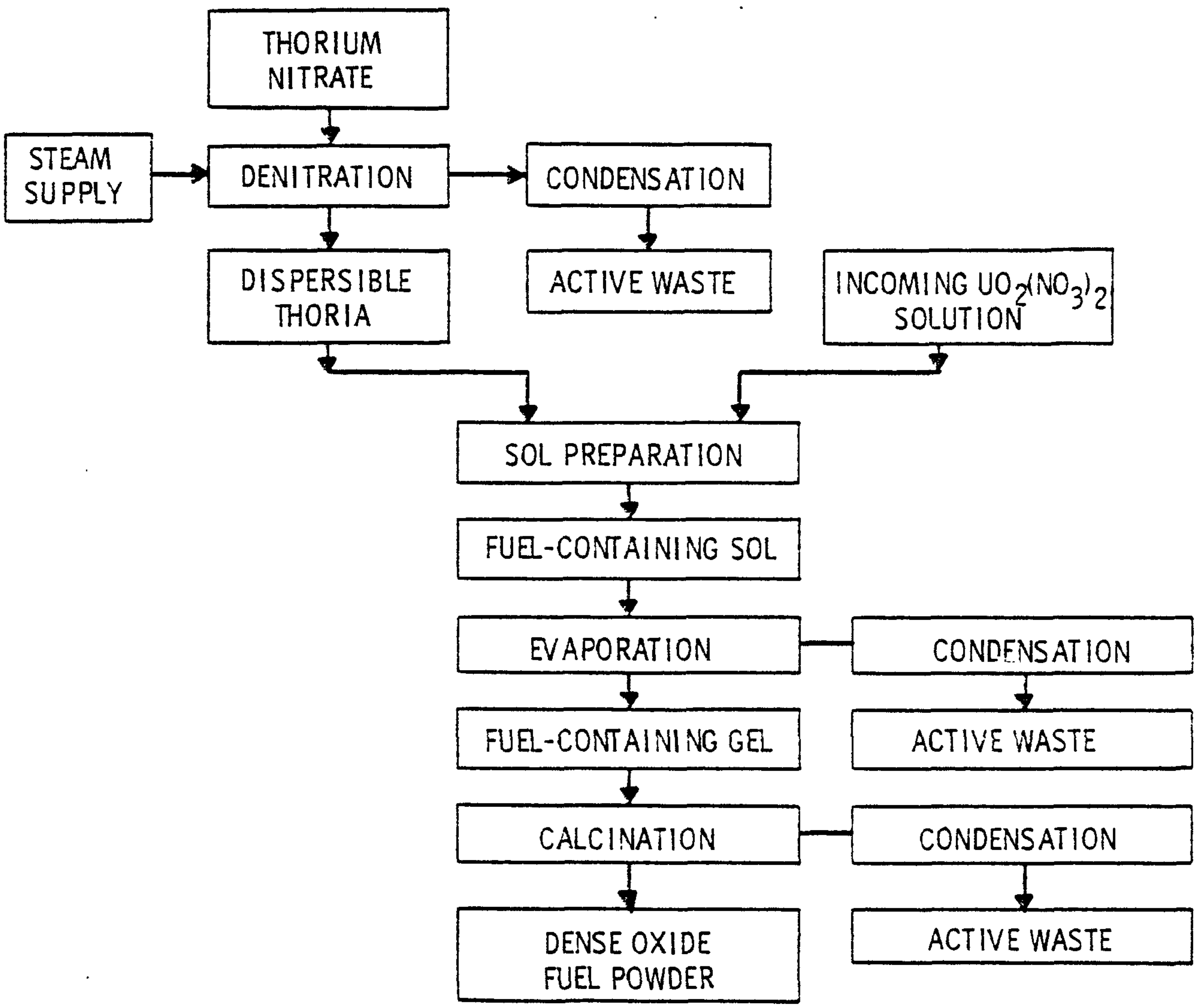

FIGURE 4. Sol-Gel Microsphere Preparation[3]

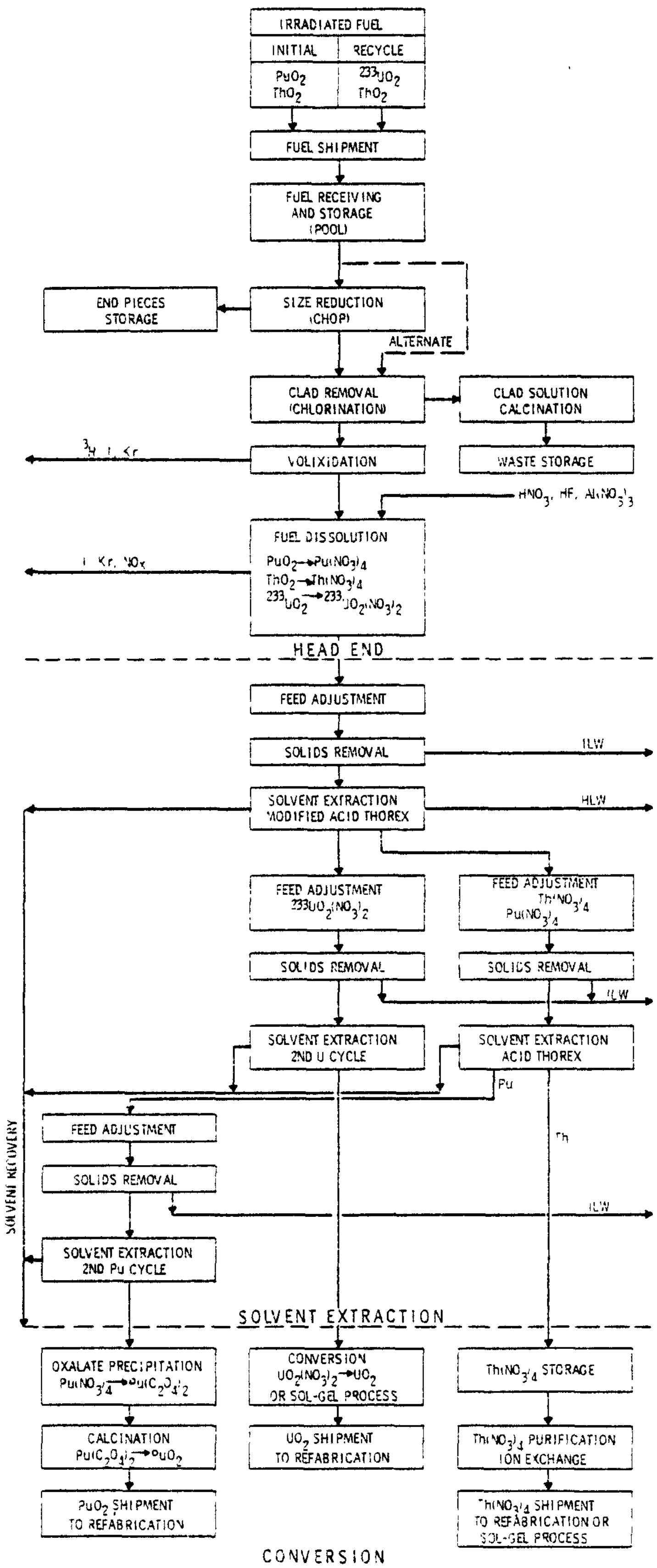

FIGURE 5. Uranium/Plutonium/Thorium Fuel Reprocessing(2)

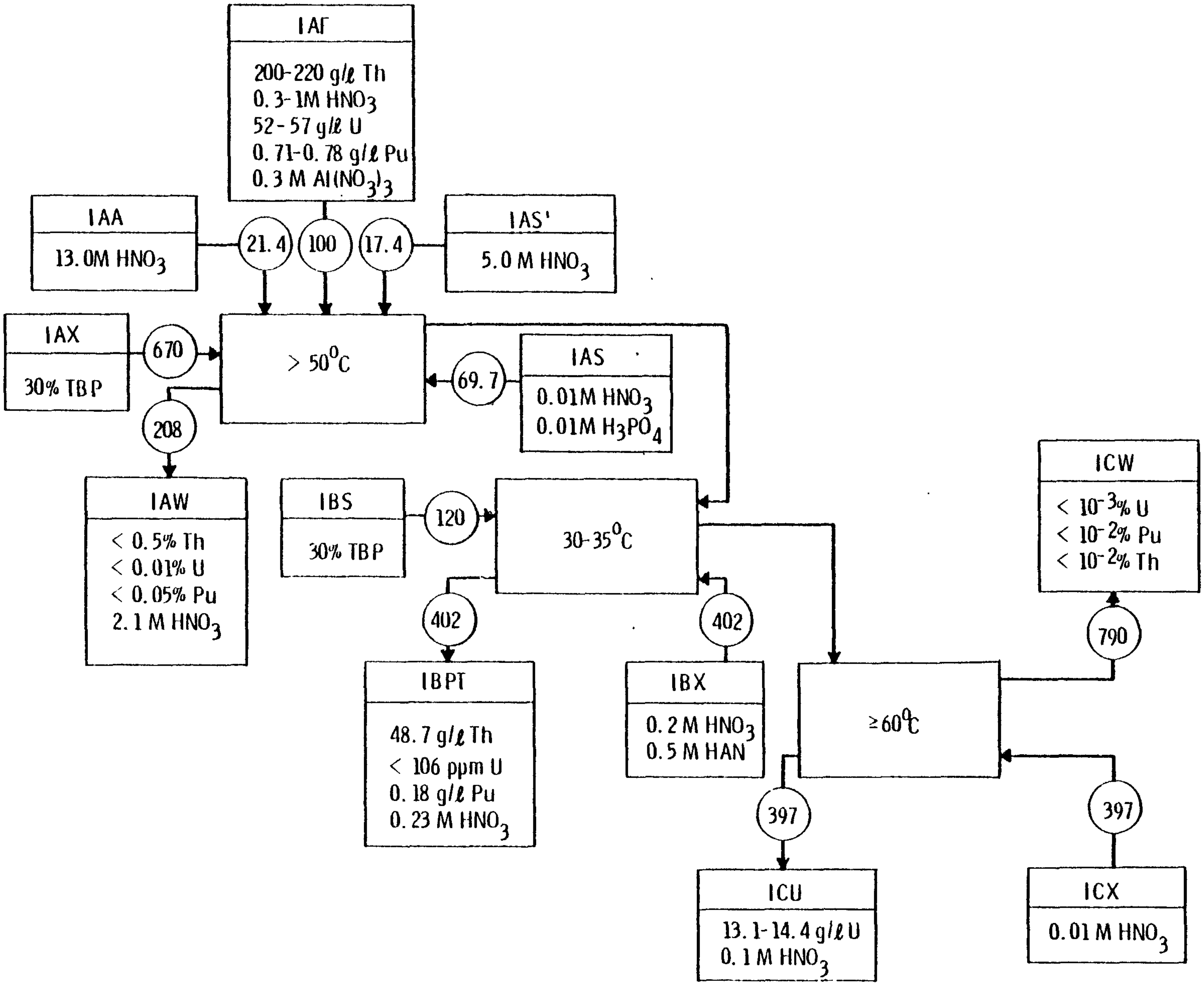

FIGURE 6. Acid Thorex - First Cycle

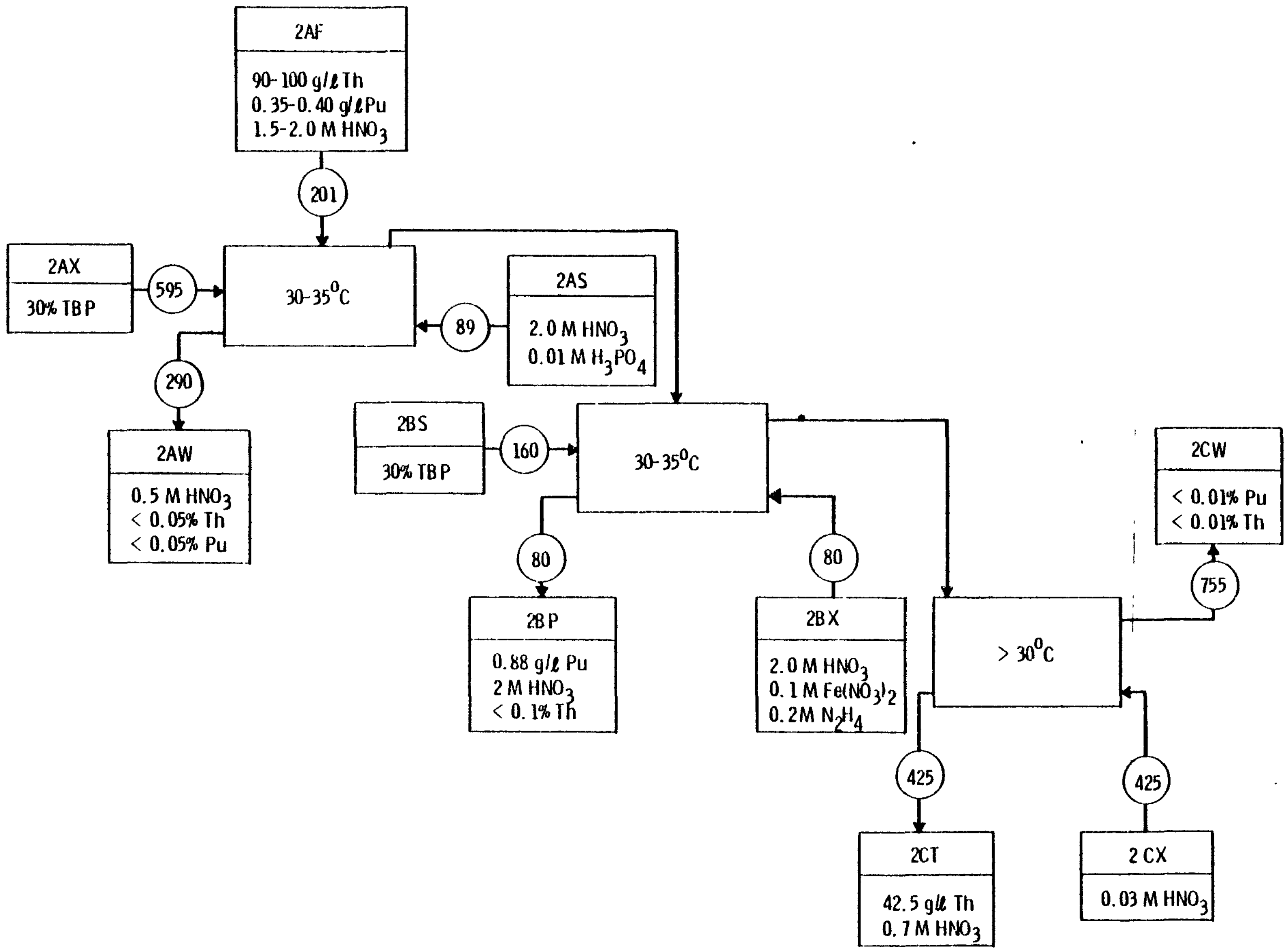

FIGURE 7. Acid Thorex - Second Th/Pu Cycle

Estimated isotopic compositions for uranium/thorium and plutonium/ thorium fuel cycles are given in Appendix C. These abundances are given only to indicate ranges of concentration ratios which might be expected in the fabrication and reprocessing facilities.

CONCLUSIONS AND SUMMARY

The TFCT program involves a wide range of fuel forms. These include powders, solutions, pellets, rods and fuel rod bundles. Chemical forms include nitrates and oxides. A summary of the expected forms is given below:

- Powders - UO_2 - ThO_2 and PuO_2 - ThO_2 powders with low and medium moderation: Possible feed material in fuel fabrication and end product in reprocessing.

- Pellets - UO_2 - ThO_2 and PuO_2 - ThO_2 both dry and water moderated: Present fuel fabrication and possibly in head end of reprocessing.

- Rods & Fuel Rod Bundles - UO_2 - ThO_2 and PuO_2 - ThO_2 with and without fixed and soluble poisons: Present in shipping containers, fuel fabrication, and spent fuel storage pools.

- Fuel Rods in Fissile Solution - UO_2 - ThO_2 and PuO_2 - ThO_2 mixtures in acid fissile solutions: Present in dissolvers. Rods may be clad or unclad and soluble poisons may be present.

- Solutions - $UO_2(NO_3)_2$ - $Th(NO_3)_4$ and $Pu(NO_3)_4$ - $Th(NO_3)_4$ solutions: Present in solvent extraction. Possibility of soluble poisons being present. Both over— and under— moderated systems possible. Organic and acid present.
- Fluidized Bed - $UO_2(NO_3)_2$ - $Th(NO_3)_4$ with trace plutonium present in fluidized bed. The bed may consist of oxide or stainless steel spheres coated with sprayed $Th(NO_3)_4$. Normally only depleted uranium is present, but should the solvent extraction system fail, greater than trace quantities of Pu and U could be present.

All the above uranium/thorium mixtures contain ^{238}U with either ^{235}U or ^{233}U and ^{235}U. Criticality data are needed on all these fuel forms to achieve the safest and most economical thorium based LWR fuel cycle.

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APPENDIX

APPENDIX A - FUEL FABRICATION

I. Oxide Pellet Fuel Fabrication

Process Step	Fuel Cycle Type	Fissile Material Form
1. Receipt and Storage of UO_2, ThO_2, and/or PuO_2	U5/Th	dry UO_2 powder
	U3/U5/Th	dry UO_2 powder dry ThO_2 powder with trace quantities of plutonium[a] and uranium
	Pu/Th	dry PuO_2 powder dry ThO_2 powder[a]
2. Blending, Milling, Agglomeration of oxide powders	U5/Th or U3/U5/Th	Mixtures of UO_2 and ThO_2 powders[a] combined with binder
	Pu/Th	Mixtures of PuO_2 and ThO_2 powders combined with binder
3. Pellet pressing, sintering, grinding, inspection, storage	U5/Th or U3/U5/Th	UO_2-ThO_2 pellets - normally dry with possibility of moderation trace
	Pu/Th	PuO_2-ThO_2 pellets - normally dry with possibility of moderation

(a) Should solvent extraction fail, significant concentrations of plutonium and/or uranium could be present in recycle thorium.

APPENDIX A - FUEL FABRICATION

Process Step	Fuel Cycle Type	Fissile Material Form
4. Fuel rod loading, off-gassing, welding, inspection, packaging, storage, shipping.	U5/Th or U3/U5/Th	Normally dry rods containing pellets of UO_2-ThO_2. Possibility of moderation.
	Pu/Th	Normally dry rods containing pellets of PuO_2-ThO_2. Possibility of moderation.
5. Scrap Recovery; dissolution, precipitation, centrifuging, calcination.	U5/Th, U3/U5/Th, and/or Pu/Th	Steps similar to reprocessing plant: Scrap dissolved and thorium, uranium, and plutonium separated. The uranium and plutonium will be precipitated and calcined to obtain oxide powders for reintroduction at top of flow.

II. Sol-gel Fuel Element Fabrication

The fissile material forms encountered in sol-gel fuel element fabrication are similar to those for oxide pellet fuel element fabrication except no pellets are produced. Instead, the powder is directly vibratory compacted into the fuel elements. Hence, all fissile material forms except pellets are present.

APPENDIX A - FUEL FABRICATION

III. Sol-Gel Microsphere Preparation(4)

Process Step	Fuel Cycle Type	Fissile Material Form
1. Receipt and storage of UO_2 $(NO_3)_2$ and Th $(NO_3)_4$ from purification.	U5/Th	UO_2 $(NO_3)_2$ solution only
	U3/U5/Th	UO_2 $(NO_3)_2$ solutions Th $(NO_3)_4$ containing trace quantities of fissile uranium and plutonium.[a]
2. Denitration	U5/Th	None
	U3/U5/Th	Recycle Th $(NO_3)_4$ with trace quantities of fissile uranium and plutonium.[a] Product is ThO_2. Fluidized bed denitration consists of Th $(NO_3)_4$ sprayed onto oxide or stainless steel spheres. Air jets remove solidified oxide.
3. Sol preparation	U5/Th or U3/U5/Th	Mixtures of UO_2 $(NO_3)_2$ and ThO_2. Trace plutonium may be present.[a] Product UO_3-ThO_2 sol.

[a] Should solvent extraction fail, significant concentrations of plutonium and/or uranium could be present in recycle thorium.

APPENDIX A - FUEL FABRICATION

Process Step	Fuel Cycle Type	Fissile Material Form
4. Gelation and Calcination	U5/Th or U3/U5/Th	UO_3-ThO_2 is evaporated and calcined. Product is dense, dry UO_2-ThO_2.[a]

[a] Should solvent extraction fail, significant concentrations of plutonium and/or uranium could be present in recycle thorium.

APPENDIX B - FUEL REPROCESSING

I. Uranium/Thorium Fuel Reprocessing (Both U5/Th and U3/U5/Th)

Process Step	Fissile Material Form
1. Receipt and storage of UO_2-ThO_2 irradiated fuel elements.	$^{235}UO_2/ThO_2$ and $^{233}UO_2/ThO_2$ rods in bundles. Probably stored in water basins.
2. Chopping of fuel rods and clad removal by zirflex and sulfex.[5]	Dry UO_2/ThO_2 rods and mixtures of rods in acid solution with oxide fines. Zircaloy clad removal in boiling 6M NH_4F-1MNH_4NO_3. Stainless steel clad removal refluxing 4M H_2SO_4.
3. Voloxidation to remove volatile fission products and tritium.	Oxidation heating to cause UO_2 change to U_3O_8.
4. Dissolving[6]	Change of UO_2 to $UO_2(NO_3)_2$ and ThO_2 to $Th(NO_3)_4$ in solution of 12M HNO_3, 0.05M HF and $Al(NO_3)_2$. Rods and fissile solutions will be present simultaneously. Also, soluble poisons might be present. Low concentrations of plutonium are also present.

Process Step	Fissile Material Form
5. First Cycle solvent extraction.	200-220 g/ℓ Th with 52-57 g/ℓ U and 0.71-0.78 g/ℓ Pu feed. Also present HNO_3 and 30% TBP. Fixed poisons may be present.
6 Second cycle solvent extraction (Th/Pu).	90-100 g/ℓ Th with 0.35 to 0.4 g/ℓ Pu in 1/5-2.0 M HNO_3, Trace U. Fixed poisons may be present.
7. Second cycle solvent extraction (U).	Over moderated UO_2-$(NO_3)_2$ solution with trace Th and Pu. Fixed poisons may be present.
8. UO_2 $(NO_3)_2$ conversion or transfer to sol-gel facility.	UO_2 $(NO_3)_2$ solution converted to either UF_6 or UO_2.
9. Th $(NO_3)_4$ purification and shipment to refabrication or sol-gel facility.	Th $(NO_3)_4$ with trace Pu content.[a]
10. Pu conversion	Pu $(NO_3)_4$ converted to Pu $(C_2O_4)_2$ and then to Pu O_2.

II. Plutonium/Thorium Fuel Reprocessing

The reprocessing of plutonium/thorium fuel is similar to uranium/thorium fuel. The difference is in the abundances for uranium and plutonium. Similar process steps are employed. Appendix C gives estimated isotopic concentrations.

[a] Should second Th-Pu cycle solvent extraction fail, significant Pu could result.

APPENDIX C

Appendix C

FUEL ISOTOPIC DISTRIBUTIONS

Fuel Compositions (wt%)

U5/Th Fuel[1]	Initial Load	Irradiated (29,000 MWd/Mt)
Th	0.7785	0.7916
^{235}U	0.0443	0.0179
^{238}U	0.1772	0.1769
^{233}U	-	0.0108
Fissile Pu	-	0.0028

U3/U5/Th Fuel[2]	Recycle Load
Th	0.708 - 0.7785
^{235}U	0.0 - 0.0443
^{238}U	0.1772- 0. 57
^{233}U	0.0 - 0.035

Pu/Th Fuel[7].	Recycle Load	Irradiated
Th	0.877	0.900
^{233}U	-	0.011
^{239}Pu	0.040	0.017
^{240}Pu	0.040	0.034
^{241}Pu	0.024	0.019
^{242}Pu	0.019	0.019

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