

CHAPTER VII  
FUEL CYCLE TECHNOLOGIES

VII.A. OVERVIEW OF FUEL CYCLES

VII.A.1. Introduction

The principal role of the fusion breeder reactor is to provide an external source of fissile fuel to support a fission power reactor economy composed of LWRs or other fission reactors. In this role, the fusion breeder is operationally similar to a fissile enrichment plant which requires no fissile feed stream and is an electricity producer rather than a consumer. In contrast with fission breeder reactors (i.e., liquid metal fast breeder reactor (LMRBR) and light water breeder reactor (LWBR)), the neutron rich fusion breeder is a subcritical assembly, produces an order of magnitude more net excess fuel, and is not subject to the neutron balance constraints of conventional fission reactors. As a result, a wide variety of fuel cycles and fuel forms are possible. In particular, the fusion breeder may produce either  $^{233}\text{U}$  or  $^{239}\text{Pu}$  using oxide, metal, carbide, molten salt or other fuel forms.

After the bred fissile material is separated from the fertile fuel form it may be used to provide initial inventories or fissile makeup to fuel conventional light water reactor (LWR) clients operating on one of several possible fuel cycles. Alternatively, the bred fuel could be used to fuel advanced fission converter reactors such as the high temperature gas cooled reactor (HTGR) or the Canadian heavy water reactor (CANDU). If desired, the fusion breeder might also breed fuel to provide initial fissile inventories for liquid metal fast breeder reactors (LMFBR).

Fuel cycle related issues will, in general, depend upon the particular breeding blanket design, the choice of a fertile fuel form, and the choices of a client reactor type and its associated fuel cycle. These issues can have potentially large impact on the cost of bred fuel, the cost of electricity generation, and overall feasibility of the fusion breeder.

In this chapter we first review several candidate fuel cycle options. This review is followed by more detailed discussions relating to the molten salt and thorium oxide suspension blankets considered during this study. Conceptual plant designs and costing analysis relating to molten salt and THOREX fuel reprocessing facilities for TMHR discharge fuel will be presented. These reprocessing technologies are considered to be key elements in the symbiotic electricity generation system. Detailed isotopic generation and depletion results for actinide transmutation species produced in the TMHR blankets will also be presented. The later results have impact upon the possible need to remotely fabricate uranium fuels produced in the TMHR.

#### VII.A.2. Fuel Cycle Options and Issues

A general schematic of the fusion breeder/fission burner reactor fuel cycle is shown in Figure VII.A-1. Although this schematic does not address any specific fuel form, several features are deserving of note. Most importantly, the overall fuel cycle is separated into two distinct fuel cycles which are coupled by the flow of bred fissile material from the fusion breeder to the fission converters. With the exception of issues which bear upon the type of converter fuel fabrication plant required (e.g., the amount of  $^{232}\text{U}$  in bred  $^{233}\text{U}$ ), the breeder and burner fuel cycle issues are entirely separable.

Both the fusion breeder and fission client fuel cycles are closed by reprocessing and recovery of fissile materials. Although direct enrichment, or "refresh" fuel cycles which do not employ reprocessing have been examined in the past,<sup>1,2</sup> these lead to inefficient fissile production in the fusion breeder as well as the disposal and loss of large quantities of valuable fissile resources. Our results indicate that, like the LMFBR, the fusion breeder requires a closed fuel cycle to achieve adequate economic performance. The fusion breeder and fission breeder fuel cycles are also similar in that only small quantities of fertile materials are required for makeup, mining requirements will be minimal, and isotopic enrichment capabilities are not required.

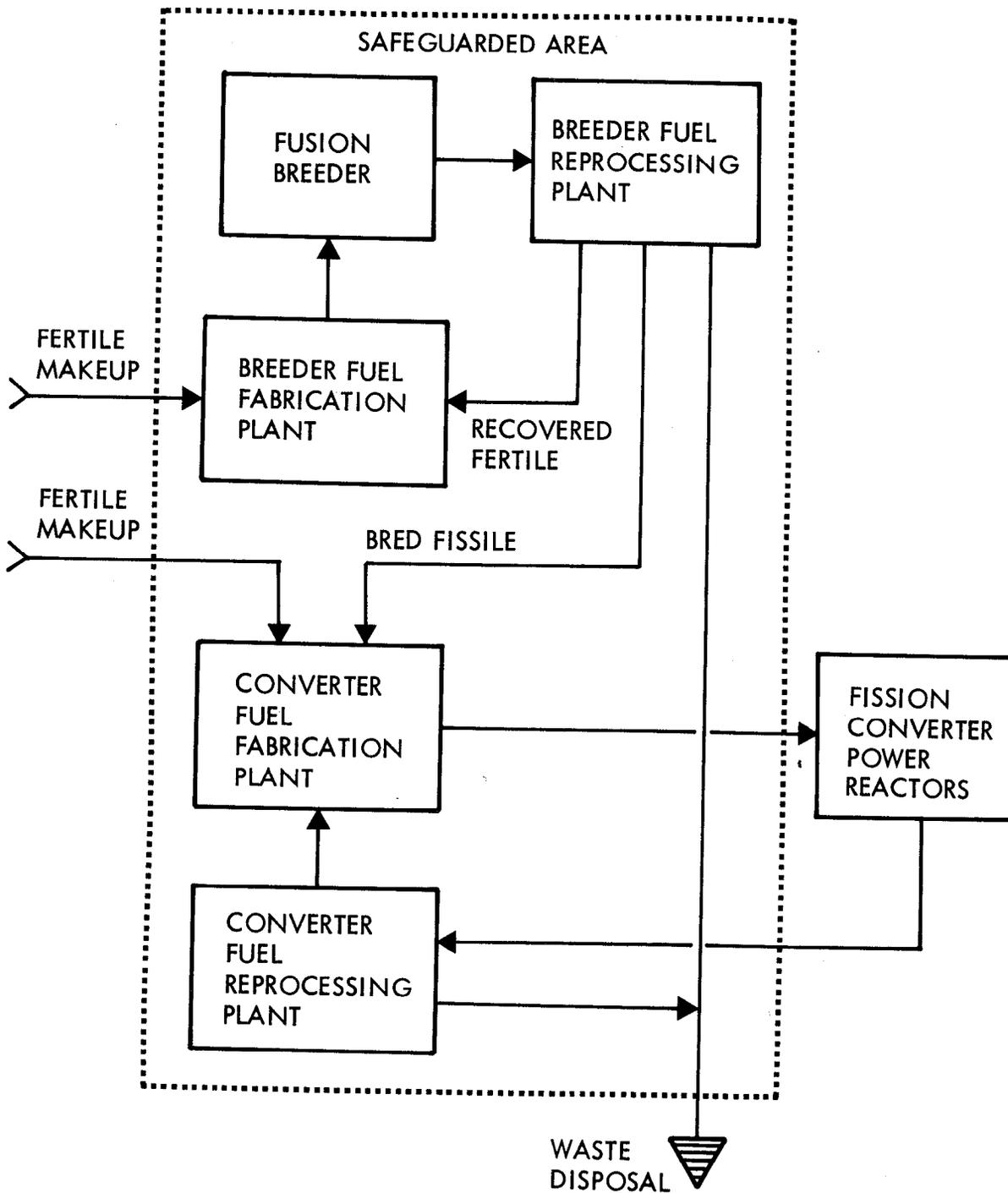


FIGURE VII.A-1. General schematic of the fusion breeder/fission burner reactor fuel cycle.

VII.A.2.a. Overview of Fusion Breeder Fuel Cycles and Issues. Concerning  $^{233}\text{U}$  production from thorium in the fusion breeder, we have considered two fuel cycles. The first of these, chosen for the thorium oxide suspension blanket, requires a THOREX type aqueous chemistry reprocessing plant to extract  $^{233}\text{U}$  from the oxide fuel. A principal concern regarding this technology is cost. In particular, the thorium oxide suppressed fission breeder fuel cycle is unique in the respect that fissile fuel is discharged at very low fission burnup ( $\sim 100$  MWD/MTHM) and at very low concentration ( $\sim 0.6\%$   $^{233}\text{U} + ^{233}\text{Pa}$  in thorium). This leads to an advantage and a disadvantage when the suppressed fission thorium oxide suspension blanket is compared with fast fission blankets. Low burnup, an advantage, leads to lower radioactivity in the discharged fuel with favorable impact on fuel reprocessing and fabrication processes. Conversely, low discharge concentration, can be expected to lead to higher unit costs (i.e.,  $\$/\text{gm}$ ) to recover the bred fuel. This situation is illustrated in Figure VII.A-2 which shows the allowable reprocessing cost to obtain a fixed contribution to the overall cost of bred fuel as a function of the fissile discharge concentration. For instance, in the case of  $\sim 0.5\%$  discharge enrichment, a 20  $\$/\text{gm}$  impact on the cost of bred fissile fuel would require a reprocessing cost of  $\sim 100$   $\$/\text{KgHM}$ . This compares with cost estimates<sup>3</sup> for the reprocessing of high burnup ( $\sim 33,000$  MWD/MTHM) thorium oxide fuels for LWRs which range from 300-500  $\$/\text{KgHM}$ .

Obviously, a major parameter to be optimized in the suppressed fission blanket is the discharge fissile enrichment. As the fissile enrichment builds up, so does the power density and radioactive inventory. The maximum discharge enrichment is constrained by the maximum allowable power density, the design limits of the safety and cooling systems, and the maximum tolerable high level radioactive waste inventory (especially if no emergency cooling system is present). The constraints on fission power density are likely to be quite low in order to capitalize on the improved safety features of suppressed fission blankets. While it is desirable to remove the bred fissile fuel at as low an enrichment as possible, it is more economical to reprocess at higher enrichments.

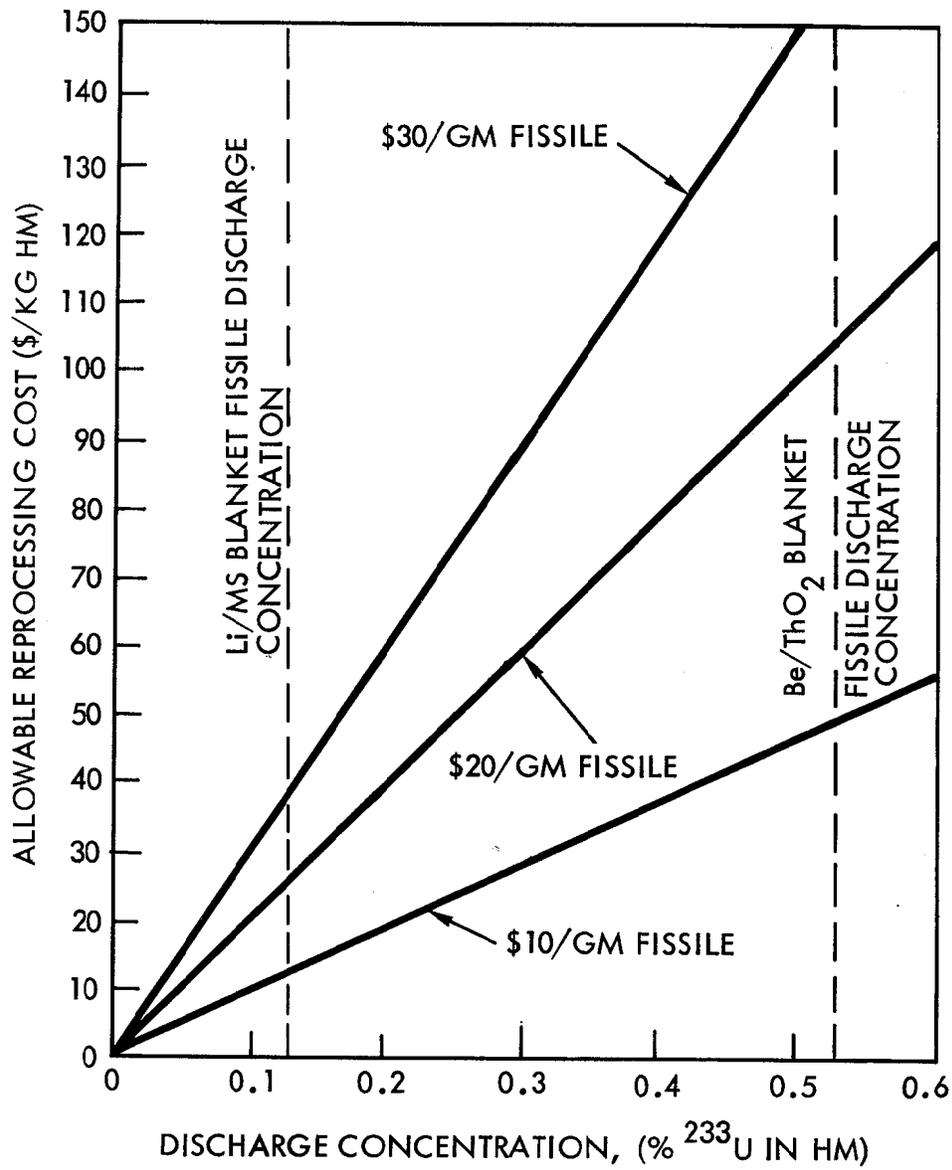


FIGURE VII.A-2. Allowable reprocessing cost to obtain a fixed contribution to the overall cost of bred fuel.

The molten salt reprocessing plant required for the TMHR molten salt blanket is expected to be quite inexpensive and is expected to result in a nearly insignificant increment in the cost of bred fissile from that system. However, the development program required to assure the feasibility of this technology is a concern. The molten salt technology, as adapted to the fusion breeder, is a simplification when compared to the technology requirements for a molten salt breeder reactor (MSBR). In particular, several of the more difficult separation processes can be entirely eliminated and a simple "fluorination only" reprocessing scheme is preferred.

Nevertheless, the use of a molten salt fuel form impacts blanket materials considerations (e.g., corrosion and radiation damage) to the extent that novel approaches are required. These considerations are discussed in detail in Chapter IV.

VII.A.2.b. Overview of LWR Fuel Cycles and Issues. Light water power reactors are expected to dominate nuclear power production when the fusion breeder reactor becomes commercially available and are considered as the principal type of fission client reactor in the symbiotic electricity generation system. The cores of these reactors would be modified to accumulate new types of oxide fuels, but studies suggest that the modified cores might not differ substantially in design or operational characteristics when compared with the current generation of LWRs.<sup>4,5</sup> The fuel cycle facilities required for LWR clients are not generically different than those that would be required to support most other fission reactor clients.

If the fuel bred in the fusion breeder is  $^{233}\text{U}$ , three fuel cycle options are available:

- The thorium fuel cycle (typically 3.3%  $^{233}\text{U}$ , 96.7%  $^{232}\text{Th}$ )
- The denatured thorium fuel cycle (typically 3.2%  $^{233}\text{U}$ , 18%  $^{238}\text{U}$ , 78.8%  $^{232}\text{Th}$ )
- The denatured uranium fuel cycle (typically 2.8%  $^{233}\text{U}$ , 97.2%  $^{238}\text{U}$ )

In comparison, the thorium fuel cycle is most efficient with respect to fissile feed requirements, but the denatured fuel cycles provide some isotopic dilution and have improved intrinsic proliferation resistance.

Regarding fuel processing technologies, both the thorium and denatured thorium LWR fuel cycles will require THOREX fuel reprocessing plant technology as well as a remote and shielded fuel fabrication technology. These technologies have not yet been developed to commercial scale and, although technically straightforward, both processes are expected to result in comparatively high costs per unit of heavy metal throughput. The denatured uranium fuel cycle requires 28% more fissile feed than the denatured thorium fuel cycle, but is compatible with developed PUREX reprocessing plant technology.

If the fuel bred in the fusion breeder is fissile plutonium, denatured fuel cycles are not possible and a mixture of plutonium and  $^{238}\text{U}$  (typically 2.9%  $\text{Pu}_f$ , 97.1%  $^{238}\text{U}$ ) would be used. This fuel cycle is 13% less efficient than the denatured uranium fuel cycle, but circumvents both THOREX and the highly shielded fuel fabrication technology. It is important to note that plutonium burners could be used even if  $^{233}\text{U}$  is bred in the fusion breeder. That is, both the denatured thorium fuel cycle and the denatured uranium fuel cycle produce appreciable quantities of plutonium in the  $^{233}\text{U}$  fueled LWRs.<sup>4</sup> This is a result of neutron absorption in  $^{238}\text{U}$  with subsequent conversion to fissile plutonium. The "secondary" fissile plutonium would, most likely, be recovered and recycled in "secondary" plutonium burning LWRs which might be located within the safeguarded fuel cycle centers.

More detailed information concerning fuel cycle performance and cost estimates as well as the composition of typical symbiotic electricity generation systems is presented in Chapters IX and X. Fuel cycle information for alternate fission reactor clients (i.e., gas cooled pebble bed fission reactors (VHTR and LMFBRs)) is also presented in later chapters.

### VII.A.3. Fuel Cycle Summaries for Reference TMHR Blanket Concepts

In this section important global parameters relating to fuel cycle data for the reference TMHR blanket concepts are presented. Overall fuel cycle parameters for both blankets are presented in Table VII.A-1.

Table VII.A-1. Fuel Cycle Summary for Thorium Oxide and Molten Salt Reference Blanket Designs. (Basis: 3000 MW fusion)

|   | MOLTEN SALT                       |                                |                              |
|---|-----------------------------------|--------------------------------|------------------------------|
|   | Thorium Oxide                     | Fluorination Only <sup>a</sup> | Full Processing <sup>b</sup> |
| Thorium Inventory, MT   | 157                               | 1680 <sup>c</sup>              | 1680 <sup>c</sup>            |
| Fissile Inventories, Kg   |                                   |                                |                              |
| <sup>233</sup> U + <sup>233</sup> Pa In-core                              | 408                               | 2270 <sup>c</sup>              | 2185 <sup>c</sup>            |
| <sup>233</sup> U + <sup>233</sup> Pa Ex-Core <sup>d</sup>                 | 3315                              | 2225                           | 2225                         |
| Fuel Management Mode  | Batch                             | Continuous                     | Continuous                   |
| Fuel Residence Time <sup>e</sup> , yr                                     | 0.123                             | 30                             | 30                           |
| Net Fissile Production <sup>f</sup> , Kg/yr <sup>233</sup> U              | 6630                              | 4450                           | 4450                         |
| Reprocessing Plant Thorium Throughput <sup>f</sup> , MT/yr                | 1326                              | 5400                           | 3340                         |
| Blanket Discharge Concentrations in Thorium <sup>e</sup> , atom %         |                                   |                                |                              |
| <sup>233</sup> U  | 0.20                              | 0.107                          | 0.107                        |
| <sup>233</sup> Pa   | 0.32                              | 0.028                          | 0.023                        |
| Fission Products  | 0.025                             | 0.238                          | 0.214                        |
| <sup>228</sup> Th   | $4.65 \cdot 10^{-8}$              | $8.23 \cdot 10^{-6}$           | $4.31 \cdot 10^{-6}$         |
| <sup>232</sup> U  | $9.78 \cdot 10^{-5}$              | $3.28 \cdot 10^{-4}$           | $1.64 \cdot 10^{-4}$         |
| Reprocessing Plant Discharge Product Concentrations <sup>e</sup> , atom % |                                   |                                |                              |
| <sup>232</sup> U in <sup>233</sup> U                                      | 0.0157 <sup>d</sup>               | 0.236                          | 0.099                        |
| <sup>228</sup> Th in thorium  | $4.67 \cdot 10^{-7}$ <sup>d</sup> | $8.23 \cdot 10^{-6}$           | $4.31 \cdot 10^{-6}$         |

<sup>a</sup>Fluorination only process removes uranium only. Continuous process rate = 0.42 m<sup>3</sup>/hr. Reprocessing plant capacity = 0.60 m<sup>3</sup>/hr.

<sup>b</sup>Full processing removes uranium, protactinium and a small fraction of rare earth fission products. Continuous process rate = 0.24 m<sup>3</sup>/hr. Reprocessing plant capacity = 0.35 m<sup>3</sup>/hr.

<sup>c</sup>In-core fraction = 91%. Fraction in primary loop = 9%.

<sup>d</sup>0.5 yr delay between discharge and reprocessing assumed.

<sup>e</sup>Isotopic accumulation for molten salt blanket based upon 30 year residence time. Thorium is replaced at the rate it is depleted and notes (a) and (b), above, apply.

<sup>f</sup>70% average plant capacity factor included.

VII.A.3.a. Beryllium/Thorium Oxide Blanket Fuel Cycle Summary. The gas-cooled beryllium/thorium oxide suspension blanket operates in a batch fuel reprocessing mode in which the thorium oxide-liquid metal suspension is discharged at relatively frequent intervals of about 45 days (70% average plant capacity factor) when the combined  $^{233}\text{U}$  plus  $^{233}\text{Pa}$  concentration in thorium reaches about 0.52%. Prior to discharge, the suspension may be slowly recirculated within the blanket to insure uniform enrichment and minimize power peaking concerns. Upon discharge, the thorium oxide is separated from the lithium lead heat transfer fluid for reprocessing and the liquid metal is returned to the blanket. This separation may be accomplished using both mechanical and vacuum distillation techniques.

As shown in Table VII.A-1, the 6630 Kg/yr of fissile fuel produced (for a 3000 MW fusion power level) is initially discharged as a mixture of  $^{233}\text{Pa}$  (61%) and  $^{233}\text{U}$  (39%). If the blanket operates at 100% capacity during the fuel residence time, the  $^{233}\text{Pa}$  fraction is even higher (66%), the fuel residence time is reduced from 45 days to 36 days, and the overall fissile discharge enrichment increases to 0.58%. High  $^{233}\text{Pa}$  concentration is considered to be an advantage because less fissioning of bred fissile material is expected.

The amount of fissile material in-core (i.e., the blanket) and ex-core (i.e., the reprocessing loop) is important from the economics perspective. That is, this quantity determines the period of time required before the fusion breeder is able to derive revenues from the sale of its principal product—fuel. Prior to this time, the breeder sells only electricity and accumulates a negative cash flow. The magnitude of this penalty may be put into perspective by assuming that the fusion breeder does not wait the required period before fissile production, but borrows enough fuel to initially operate in steady state on an equilibrium cycle. In this hypothetical case, there is no delay, but the fusion breeder operator would be expected to carry the borrowed resource as a non-depreciating asset and pay an annual carrying charge over the life of the plant.

Assuming typical values for TMHR bred  $^{233}\text{U}$  of 75 \$/gm and a carrying charge of 8.6 %/yr (see Chapter IX), the 3805 Kg inventory associated with the beryllium/thorium oxide blanket would result in a  $25 \cdot 10^6$  \$/yr

operating cost. Typically, over a 30 year plant lifetime, this charge would be about 5% of the overall cost of the TMHR plant capital.

The larger fraction of the total fissile inventory associated with this blanket is the ex-core inventory. This quantity is based upon a 6 month delay prior to reprocessing to allow the fuel to cool and to allow for 99% of the  $^{233}\text{Pa}$  to decay ( $T^{1/2} = 27\text{d}$ ) to  $^{233}\text{U}$  prior to reprocessing. The smaller in-core fissile inventory results from the fertile dilute nature of the blanket (only 157 MT thorium in-core).

Concerning the thorium oxide reprocessing facility, the thorium oxide throughput for a single TMHR will be 1060 MT/yr. This size is comparable to that of typical commercial fuel reprocessing plant designs and is equivalent to the heavy metal throughput rate of about 40 1 GWe LWRs. The above result emphasizes the importance of reprocessing technologies for suppressed fission hybrid blanket concepts.

Additional calculations relating to isotopic generation and depletion as well as the isotopic content of thorium and fissile material discharged from the beryllium/thorium oxide blanket TMHR and its dedicated reprocessing plant will be presented in section VII.C.

#### VII.A.3.b. Lithium/Molten Salt Blanket Fuel Cycle Description.

VII.A.3.b.(1). Batch and Continuous Reprocessing Modes. The lithium/molten salt blanket can operate in either a continuous reprocessing mode or a batch fuel reprocessing mode. In either case, molten salt is removed from the blanket for recovery of bred uranium and (for some processing options), protactinium and, possibly, rare earth fission products.\* All molten salt fuel reprocessing will be performed on-site in a dedicated facility (see Section VII.B).

If batch fuel management is adopted, the molten salt will be charged to the blanket for a fixed period of time required to breed in the desired fissile concentration in the salt. The advantages of this mode of operation are a lower cycle averaged fission rate for the same fissile discharge

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\*Rare earth fission product removal not proposed for TMHR.

concentration (0.11%  $^{233}\text{U}$ ,  $\approx$  0.03%  $^{233}\text{Pa}$  in thorium) and a possible relaxation in reliability requirements for the molten salt process plant. The disadvantage of this mode of operation is the economic penalty of carrying an entire extra load of molten salt (direct cost  $\approx$  \$200 million for the salt inventory in a 3000 MW fusion TMHR).

Isotopic generation for the batch fuel management mode is shown in Figure VII.A-3. As shown, the time to reach a  $^{233}\text{U}$  concentration in thorium of 0.11% (the design level) is 0.54 years. The same period of time is required for  $^{233}\text{Pa}$  in the blanket to reach its equilibrium level of 0.028% in thorium. Since the total molten salt inventory in the blanket and heat transport loop is about 1150  $\text{m}^3$ , the above results indicate that an average salt processing rate of 2130  $\text{m}^3/\text{yr}$  (or 0.243  $\text{m}^3/\text{hr}$ ) is required to limit the amount of salt in process to one re-load. The above processing rate is reasonable and, as will be discussed later, is similar to the processing rate required for continuous reprocessing modes.

If continuous reprocessing modes are adopted a small slip stream of molten salt will be continuously charged and discharged from the molten salt primary coolant loop. In practice, this slip stream would not have to be continuous, but could operate on a time scale which is short in comparison with the batch irradiation time discussed above to limit the total inventory of salt. In this mode, the  $^{233}\text{U}$  and  $^{233}\text{Pa}$  concentrations will reach equilibrium values determined by the process flow rate, the type of reprocessing employed (i.e., the elements that are removed), and other parameters associated with the blanket and the reprocessing plant.

To model this mode of operation we first consider the protactinium mass balance

$$I \frac{dC_p}{dt} = R_p - F_p \eta_p C_p - \lambda_p I C_p \quad (1)$$

where

- $I$  = Total salt inventory in-core and ex-core (g)
- $C_p$  = Protactinium concentration in salt (g Pa/g Salt)
- $R_p$  = Rate of protactinium production via thorium capture and  $^{233}\text{Th}$  decay (g/s)

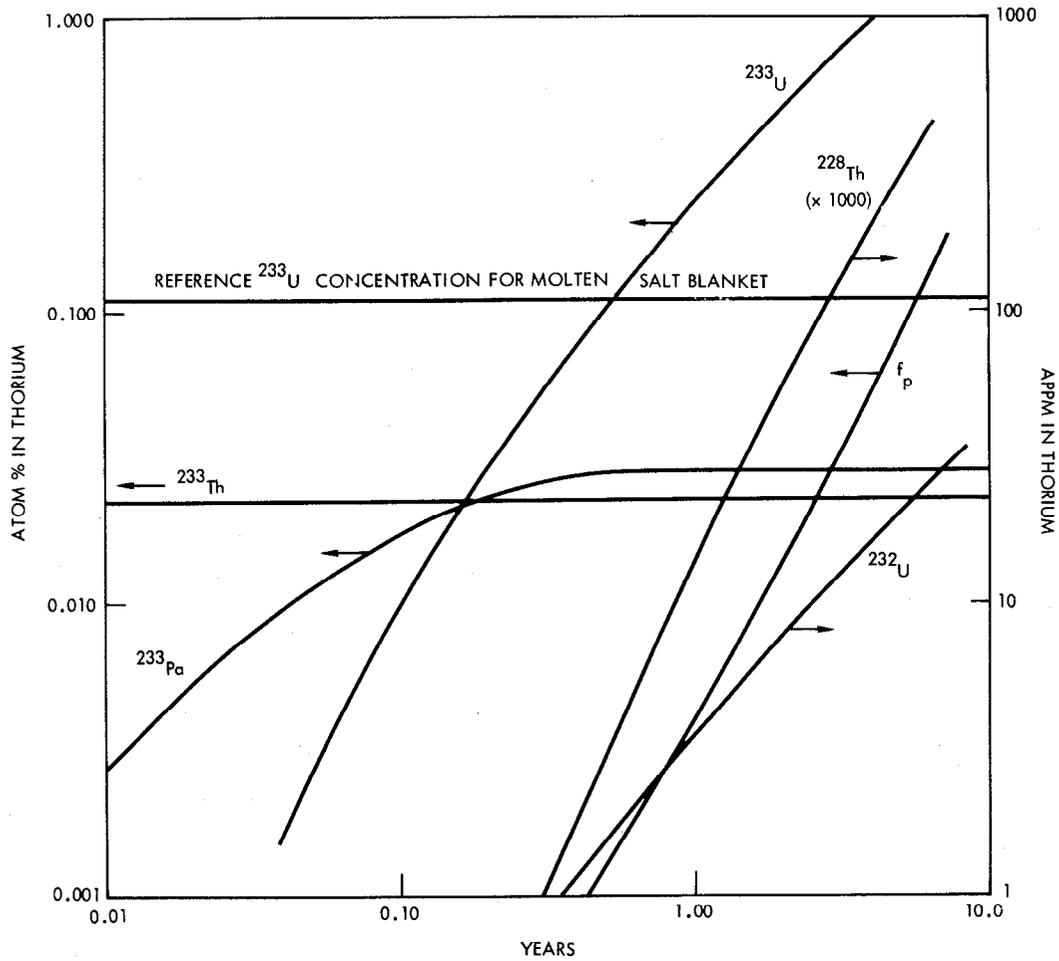


FIGURE VII.A-3. Isotopic generation in molten salt blanket with no fuel reprocessing (batch mode).

$F$  = Salt process rate (g/s)

$\eta_p$  = Protactinium removal efficiency (process dependent)

$\lambda_p$  = Protactinium decay constant ( $s^{-1}$ )

In equation (1), the terms on the right hand side represent nuclear production, process removal, and decay removal, respectively. In equilibrium we have

$$\frac{dC_p}{dt} = 0 \quad (2)$$

and the solution to equation (1) is

$$C_p = \frac{R_p}{F \eta_p + \lambda_p I} \quad (3)$$

To better understand the situation, it is of interest to consider the case where the molten salt process is turned off and the system reaches a natural equilibrium. In this case

$$C_p = \frac{R_p}{\lambda_p I} \quad (4)$$

For the reference lithium/molten salt blanket TMHR (3000 MW fusion) the parameters on the right hand side of equation (4) are

- $R_p = 0.141$  gm/s (at 70% capacity)
- $I = 3.86 \cdot 10^9$  g molten salt ( $1052 \text{ m}^3$  in the blanket,  $100 \text{ m}^3$

in the primary loop)

- $\lambda_p = 2.97 \cdot 10^{-7} s^{-1}$  ( $T^{1/2} = 27d$ )

and the resulting  $^{233}\text{Pa}$  concentration is  $1.23 \cdot 10^{-4}$  gm  $^{233}\text{Pa}$  per gm salt.

A more meaningful quantity, the weight concentration of protactinium in thorium,  $C_p^{\text{Th}}$ , is given by the following

$$C_p^{\text{Th}} = C_p / 0.435 = 2.83 \cdot 10^{-4} \quad (5)$$

where 0.435 is the weight fraction of thorium in the salt. The above quantity represents the equilibrium protactinium concentration in thorium if there were no molten salt process.

We can use equations (3) and (5) to solve for the process flow rate to achieve a specified concentration of  $^{233}\text{Pa}$  in thorium such that

$$F = \frac{1}{\eta_p} \left[ \frac{2.3 R_p}{C_p^{\text{Th}}} - \lambda_p I \right] \quad (6)$$

where  $\eta_p$  is estimated to be 99.5%. For instance, according to equation (6), a slightly reduced  $C_p^{\text{Th}}$  level of 0.023% will require a salt processing rate of 260 gm/s (0.280 m<sup>3</sup>/hr). Since a 70% capacity is assumed, the actual reprocessing capacity should be (0.7)<sup>-1</sup> times larger, or 0.400 m<sup>3</sup>/hr.

Next, consider the uranium mass balance

$$I \frac{dC_u}{dt} = \lambda_p C_p I - F \eta_u C_u \quad (7)$$

where  $F$ ,  $I$ ,  $C_p$ , and  $\lambda_p$  are as previously defined and

$C_u$  = Uranium concentration in salt (g U/g salt)

$\eta_u$  = Uranium removal efficiency (process dependent)

In equation (7), the terms on the right hand side represent protactinium decay to uranium and process removal, respectively. In this analysis, in-core fission as well as the proposed use of small amounts of uranium in the salt to maintain chemical stability are not considered.

Proceeding with equation (7), the equilibrium concentration with the molten salt process is

$$C_u = \frac{\lambda_p C_p I}{F \eta_u} \quad (8)$$

If we use values from above (i.e., a  $^{233}\text{Pa}$  level of 0.023% in thorium, 260 gm/s) and assume a uranium extraction efficiency of 100%, then

$$C_p^{\text{Th}} = C_u / .435 = 0.001 \quad (9)$$

Using the model described above, it is possible to determine the  $^{233}\text{U}$  and  $^{233}\text{Pa}$  concentrations in the molten salt processing loop as a function of the salt processing rate. The results are shown in Figure VII.A-4 for two types of molten salt processing: (1) "full reprocessing" to remove both uranium and protactinium, and (2) a "fluorination only" process to remove

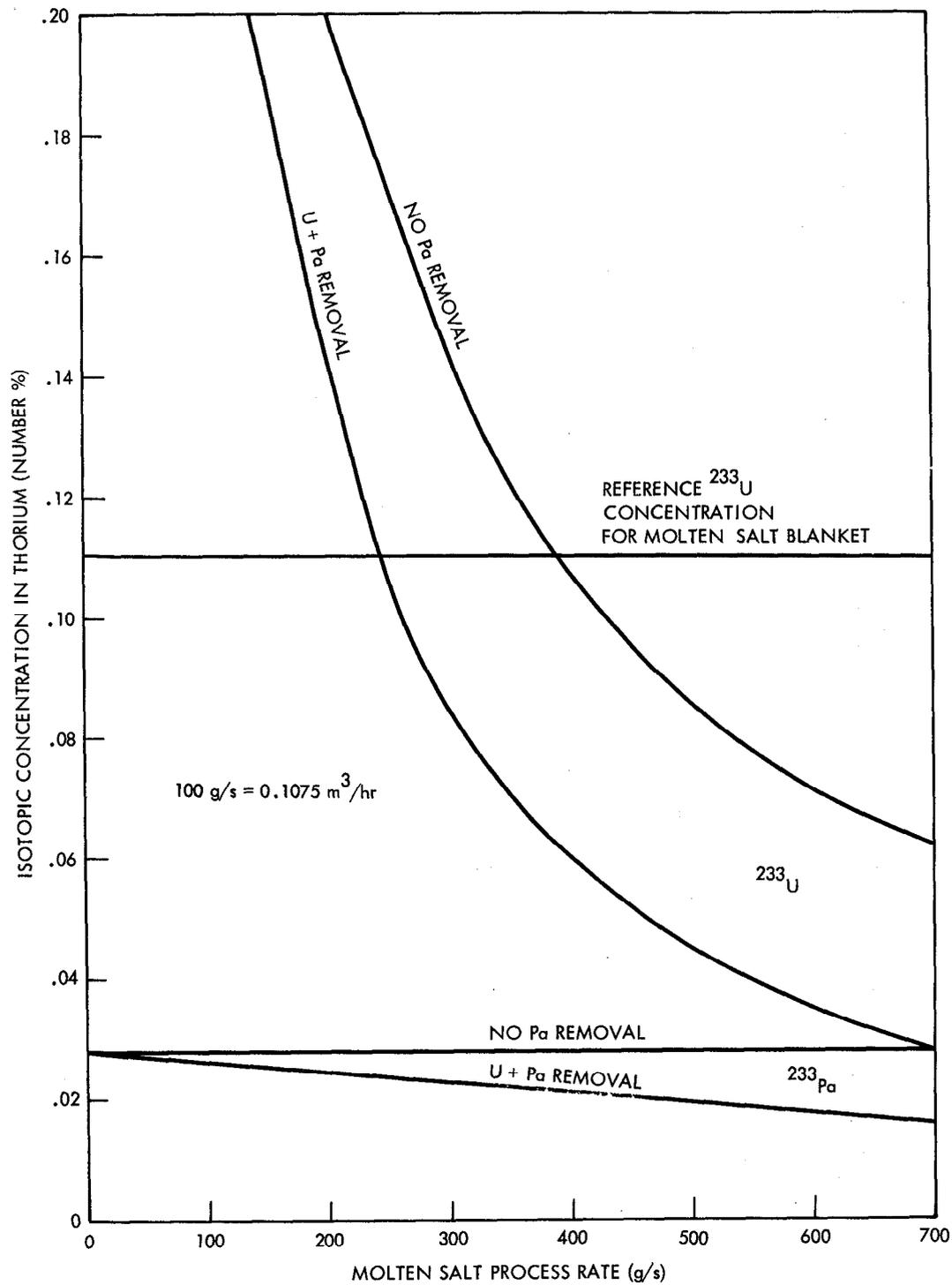


Figure VII.A-4. Equilibrium  $^{233}\text{U}$  and  $^{233}\text{Pa}$  concentrations versus salt process rate.

uranium only with no protactinium removal. The full reprocessing mode is expected to remove 100% of the uranium and 99.5% of the protactinium in the salt per pass through the reprocessing plant, while the less expensive and less technologically demanding fluorination only mode is expected to remove only 75% of the uranium per pass. Both modes are more fully described in Section VII.B.2.

As shown in the figure, for the same processing rate, the U + Pa removal mode results in the lowest  $^{233}\text{U}$  and  $^{233}\text{Pa}$  concentrations in the loop. In this case, a 240 g/s average process rate is sufficient to achieve the reference  $^{233}\text{U}$  concentration of 0.11% in thorium. If the molten salt processing plant has an average capacity factor of 70%, then the design capacity would be about  $240/.7 = 340$  g/s ( $0.365 \text{ m}^3/\text{hr}$ ). For the mode without protactinium removal, a 393 g/s average process rate is required to achieve the reference  $^{233}\text{U}$  concentration and the plant design capacity would be 560 g/s ( $0.600 \text{ m}^3/\text{hr}$ ).

Three considerations are important with respect to a choice between the two possible modes of operation. First, the full processing mode achieves a 20% lower  $^{233}\text{Pa}$  level which should result in marginal increases in fissile breeding (i.e., neutron absorption in  $^{233}\text{Pa}$  is a double loss mechanism) and a small decrease in afterheat due to energy released in protactinium decay. Nevertheless, the  $^{233}\text{Pa}$  concentration is difficult to reduce substantially for reasonable process rates. Second, the fluorination mode is less expensive than the full processing mode despite the much higher process rate. Even so, the more expensive process is estimated to cost less than 5% of the overall direct cost of the reference TMHR. Most importantly, the fluorination only process employs only a batch fluorinator recovery step and eliminates much of the complexity and technological development required for the full process. As a result, the TMHR molten salt processing plant can be less complex than the process required for a Molten Salt Breeder Reactor (MSBR). The choice of the fluorination only process for the TMHR is primarily based upon the later observation. The above considerations are more fully described in Section VII.B.2.

VII.A.3.b.(2). Lithium/Molten Salt Blanket Fuel Cycle Summary. A summary for important fuel cycle parameters for the reference lithium/molten salt blanket TMHR operating in either the fluorination only or full processing mode is given in Table VII.A-1 (Section VII.A.3.a).

As shown, the 4450 Kg/yr of fissile fuel produced is initially discharged as a mixture of  $^{233}\text{Pa}$  (about 19%) and  $^{233}\text{U}$  (about 81%). The fissile inventory associated with either mode of reprocessing is about 4450 Kg and is split about evenly between the ex-core inventory and the in-core inventory. The thorium inventory in the blanket, primary loop, and process plant is 1680 MT. The thorium throughput for a single TMHR will be 5400 MT/yr for the fluorination only process or 3340 MT/yr for the full processing option. Additional calculations relating to isotopic generation and depletion as well as the isotopic content of thorium and fissile material discharged from the lithium/molten salt blanket TMHR and its dedicated reprocessing plant will be presented in Section VII.C.3.

VII.B. THOREX AND MOLTEN SALT FUEL REPROCESSING TECHNOLOGIES FOR TMHR APPLICATION

VII.B.1. Application of Thorex Reprocessing Technology for TMHR Fuel Reprocessing

VII.B.1.a. Overview. As part of the TRW Laser Fusion Breeder Design Study<sup>6</sup> sponsored by DOE during 1980, Bechtel completed a preliminary conceptual re-design and costing study for a THOREX reprocessing plant to address the recovery of fissile material from unclad, low burnup thorium metal fuel discharged from a suppressed fission blanket. The conceptual plant design and process flows were based upon recent commercial reprocessing plant design work<sup>7</sup> (e.g., Barnwell) and plant capital and operating cost estimates were derived as a function of the heavy metal throughput.

Although the thorium metal reprocessing plant technology is not identical to the more difficult technology required to reprocess fuels discharged by the thorium oxide suspension blanket, the technical issues for the metal and the oxide are, for the most part, the same and the description of previous work on thorium metal reprocessing for the fusion breeder in section VII.B.1.b serves as a useful frame of reference. In section VII.B.1.c we use the thorium metal reprocessing plant design and costs as well as other recent reprocessing cost scaling data to obtain a preliminary estimate of the cost to reprocess thorium oxide fuel discharged from a TMHR. The reprocessing plant described in section VII.B.1.b is directly applicable to the thorium metal fueled blanket designs discussed in Appendix A.

Before discussing the plant design it is useful to introduce several design features of the redesigned THOREX plant which could permit substantial reductions in the cost to reprocess thorium metal fusion breeder fuels as opposed to high burnup LWR fuels. These are listed below:

- Lower Burnup ( 100-200 MWd/MT vs 33,000 MWd/MT) - greatly reduces the quantities of fission products and associated effluents.
- Smaller Scope Facility - the reprocessing plant described in Section VII.B.1.a is essentially  $\frac{1}{2}$  the size and has  $\frac{1}{2}$  the equipment of a typical LWR reprocessing plant of similar throughput.

- No High-Enriched Conversion is Required - U is transferred as a solution, hence no calciners and reducers are required.
- Utility Financing Used - considerably lower than usual high risk industry financing.
- Reduced Off-Gas Treatment - due to lower burnup, the off-gas system for this plant is much less extensive than that of typical LWR reprocessing plants.
- No Chop/Shear Equipment - the Th spheres have no cladding and are already in dissolvable size units, hence no shear or clad removal equipment is required.
- No Large Spent Fuel Storage - the spent fuel is stored at the reactor site until ready to be reprocessed. Since the reactors and the reprocessing plant are co-located, only a small surge capacity of spent fuel must be stored at the reprocessing plant.

The above design features will be described in the following discussion.

VII.B.1.b. Thorium Metal Reprocessing Plant. The purpose of the reprocessing plant in the suppressed fission-fusion breeder concept is to recover the fissile uranium ( $^{233}\text{U}$ ) from the irradiated thorium (Th) metal, to transfer the purified uranium product to an on-site light water reactor (LWR) fuel fabricator, to provide for 10-15 years of thorium storage, and to retain, treat, and package the high level waste streams for permanent disposition.\* The basic functions of the reprocessing plant and their interrelations are shown in Figure VII.B.1.

The conceptual design study of a reprocessing plant to accomplish the above objectives was performed by Bechtel as part of the DOE sponsored Laser Fusion Breeder Design Study. The design basis plant was sized to accommodate the fuel discharges from two suppressed fission breeder reactors as described in the study final report.<sup>6</sup> However, the economic

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\*Low-level liquid and solid waste will be transferred to an on-site waste-handling facility which processes all site-generated (reactors, reprocessing, and Th metal fabrication) low-level waste.

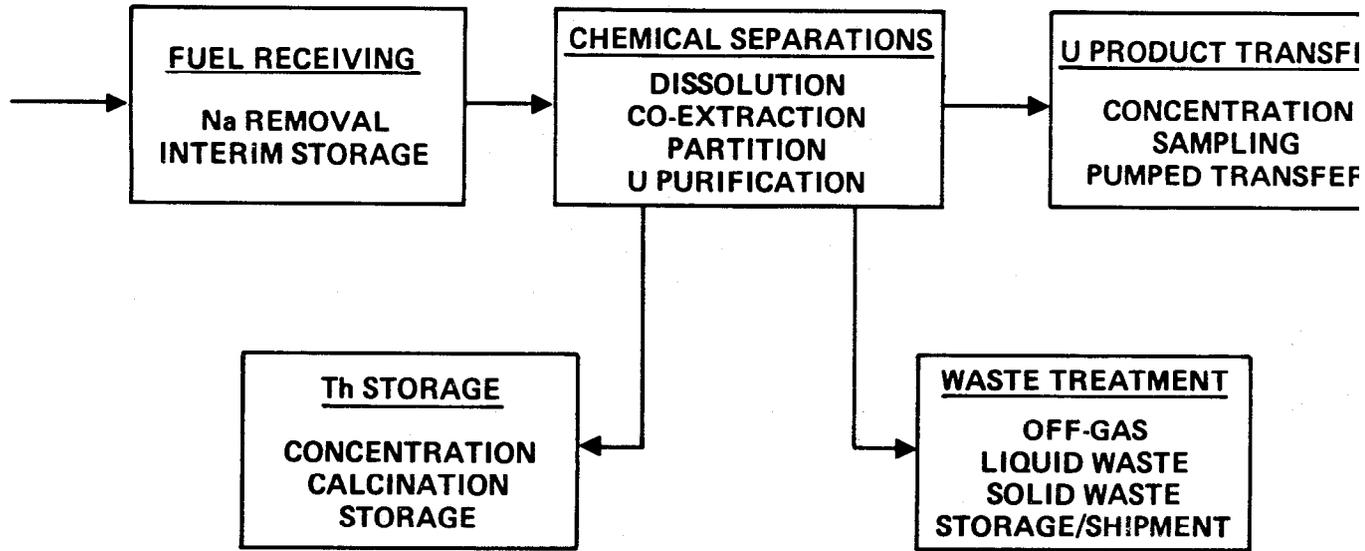


Figure VII.B-1. Thorium blanket reprocessing functions.

impact of smaller and larger reprocessing plants was also determined. The reactors, reprocessing plant, Th metal fabrication plant, and LWR fuel fabrication plant are all assumed to be co-located on the same site.

VII.B.1.b.(1). Reprocessing Plant Design Basis.

Battery Limits. The irradiated thorium fuel storage facilities at the site are not part of the reprocessing plant. Rather, facilities to provide three months of cooling between reactor discharge and chemical processing are assumed to be part of the reactor design. Transfer of the irradiated fuel to the reprocessing plant will be done in a batch operation. Some interim storage of cleaned (Na removed) thorium spheres is provided in the processing plant.

The high-level and intermediate-level liquid waste produced in the reprocessing plant will be concentrated, stored for five years, and solidified prior to shipment to a federal repository. Radioactive solid waste (e.g., spent filters, resin beds, failed equipment) and low-level liquid waste will be transferred to a waste management facility which services the entire complex.

The recovered uranium (essentially 100%  $^{233}\text{U}$ ) will be transferred to the on-site LWR fabrication plant as a concentrated uranyl nitrate solution via a double-walled pipeline. The recovered thorium will be calcined to oxide and stored on-site for 10-15 years.

Reference Plant Design Basis. Several plant designs and flowsheets were surveyed and a composite reference design was selected. To the extent compatible, the fission-fusion hybrid reprocessing plant design, the flowsheet, the equipment, and the operating mode will parallel that described in reference 8 (the flowsheet is a synthesis of those in sections C10 and C12) which is based on the Barnwell Nuclear Fuel Plant (BNFP) design.

Table VII.B-1 lists the design basis and constraints for the fission-fusion hybrid reprocessing plant. These design basis are related to the throughput, the irradiated fuel characteristics, the product specifications, the effluent limits, the allowable personnel exposures, and the structural design criteria.

TABLE VII.B-1. Plant design basis.

|                                 |   |
|---------------------------------|---|
| Operating Period                | 300 days/year   |
| Throughput:                     |   |
| Overall                         | 1220 MTHM/year  |
| Head-End                        | 5 MTHM/day  |
| Solvent Extraction              |   |
| Co-Extraction                   | 5 MTHM/day  |
| U Purification                  | 30 KgU/day  |
| Uranyl Nitrate Pumping          | 4.17 g/hr @ 300 g/g   |
| Th Conversion                   | 5 MTHM/day  |
| Feed Characteristics:           |   |
| Physical and Chemical           | Th metal spheres 2.54 cm in diameter, containing 0.58% <sup>233</sup> U (average)   |
| Burnup (avg) <sup>a</sup>       | 200 MWd/MT  |
| Minimum Cooling Time            | 120 days  |
| Fissile Content (Max. expected) | 0.72% <sup>233</sup> U  |
| Product Specifications:         |   |
| Recovery Factors                | 99.5% of U<br>98% of Th   |
| U Product                       | Th < 0.01%<br>< 0.08% Others  |
| Th Product                      | U < 0.001%<br>< 0.008% Others   |
| Effluent Limits                 |   |
| Liquid                          | No discharge of radioactive liquids   |
| Gas                             | Extensive separation of I and Ru from off gases<br>Kr and <sup>14</sup> C (if present) to be vented along with any volatilized <sup>3</sup> H |
| Solid                           | HLLW to be converted to borosilicate glass within 5 years after separation. <sup>b</sup>  |

<sup>a</sup>Based upon fission product generation.

<sup>b</sup>HLLW = High level liquid waste.

Operation and Maintenance. The hybrid fuel reprocessing plant is envisioned to employ a combination of maintenance philosophies similar to those used in recently constructed or designed LWR reprocessing plants.<sup>7,9</sup> This consists of placing all equipment associated with high levels of gamma radiation in fully remote cells. Equipment exposed to intermediate levels of gamma radiation is located in semi-remote cells where both remote and some direct maintenance work can be performed. Direct maintenance is utilized where no or low radiation levels are present or where radioactive materials are well confined and shielded (e.g., storage areas). Due to the radioactivity associated with the uranium and thorium, remote operation is used throughout the plant. All instrument read-out and control locations will be separated and shielded from the process equipment.

Proliferation Considerations. Proliferation deterrence for this reprocessing plant is provided by hardened physical protection (guards, double-fenced site, remote sensing equipment, etc.) and dynamic material accountability systems. The natural radiation associated with  $^{232}\text{U}$  in the  $^{233}\text{U}$  product also enhances the proliferation deterrence of the products of the reprocessing plant. The  $^{233}\text{U}$  could also be diluted with  $^{238}\text{U}$  during reprocessing, but this would significantly increase the required material throughput and hence the cost of the reprocessing plant and therefore is not specified for this plant design.

#### VII.B.1.b.(2). Plant Description.

Process Flowsheet Description. The process flowsheet is shown in Figure VII.B-2 and a description of the various streams is provided in Table VII.B-2. In addition to the systems detailed in Figure VII.B-2, the plant also contains the following systems: Off-gas treatment solvent recovery, acid and water recovery and make-up, waste treatment, ventilation, process cooling and heating, and thorium and waste storage facilities. Complete material balances were not calculated, however, the flowsheet was derived from previously determined flowsheets<sup>8,10</sup> so that the range of flowrates and conditions can be estimated with enough accuracy to specify equipment and facility size, layout, and cost.

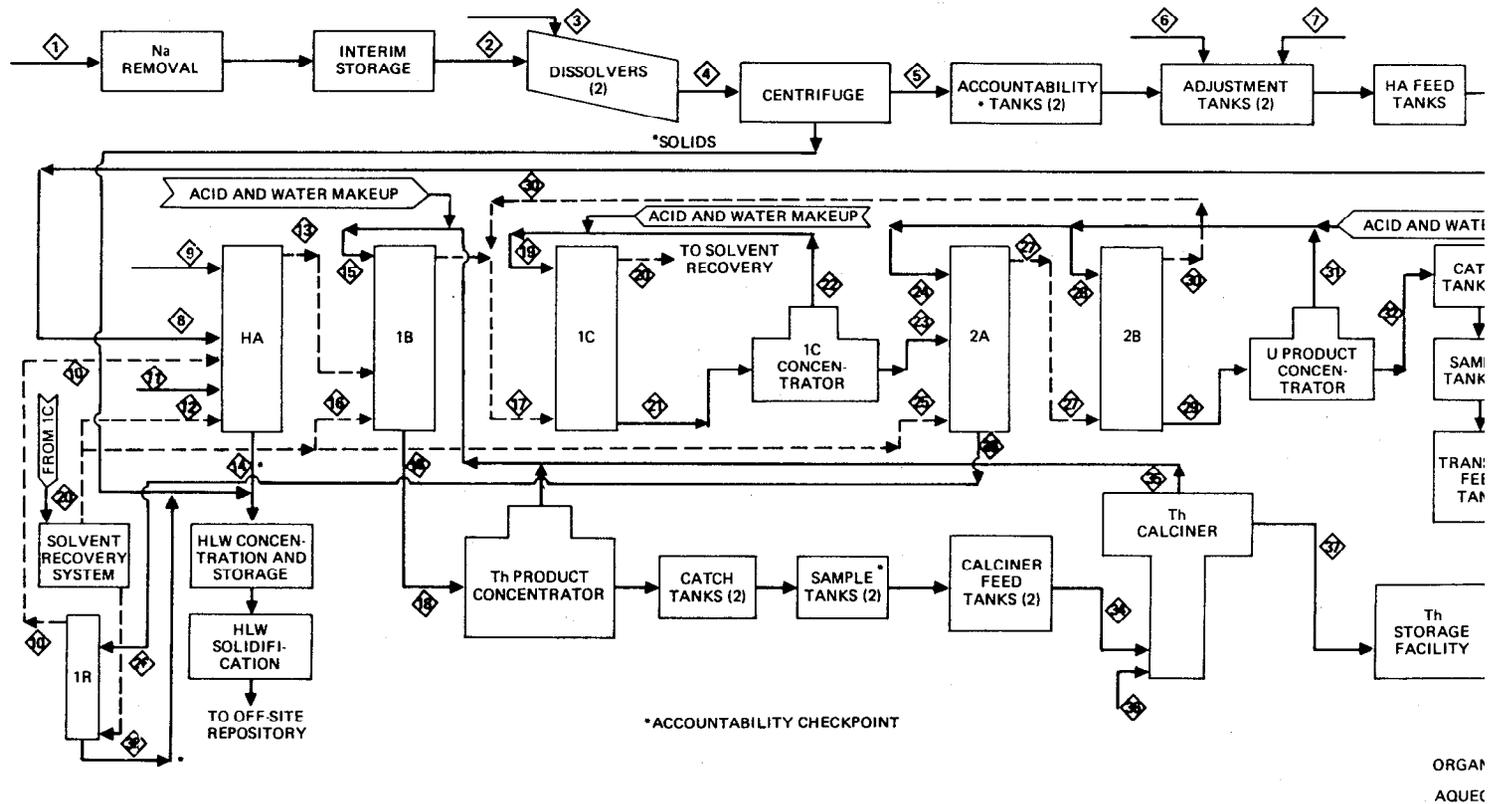


FIGURE VII.B-2. Process flowsheet.

TABLE VII.B-2. Process stream description.

| Stream Number <sup>a</sup> | Identification                       | Description                      |
|----------------------------|--------------------------------------|----------------------------------|
| 1                          | Th sphere transfer                   | Unclean, Na contaminated spheres |
| 2                          | Dissolver feed                       | Cleaned Th spheres               |
| 3                          | Dissolver chemicals                  | ~12M nitric acid with 0.001M HF  |
| 4                          | Dissolver solution                   | _____                            |
| 5                          | Clarified dissolver solution         | _____                            |
| 6                          | Chemical adjustment lines            | Acid and water                   |
| 8                          | HA feed                              | U, Th, FPs in ~ 1M nitric acid   |
| 9                          | Nitric acid scrub                    | _____                            |
| 10                         | Organic solvent from recovery column | 30% TPB in hydrocarbon diluent   |
| 11                         | Acid Butt                            | ~12M nitric acid                 |
| 12                         | Clean organic solvent                | 30% TPB in hydrocarbon diluent   |
| 13                         | Loaded organic (U+Th)                | _____                            |
| 14                         | High-level waste                     | FPs in nitric acid               |
| 15                         | Nitric acid strip                    | ~0.1M                            |
| 16                         | Clean organic solvent                | 30% TPB in hydrocarbon diluent   |
| 17                         | Loaded organic (U only)              | _____                            |
| 18                         | Aqueous Th-nitric acid stream        | _____                            |
| 19                         | Nitric acid strip                    | 0.1M                             |
| 20                         | Organic raffinate                    | _____                            |
| 21                         | Aqueous U-nitric-acid stream         | _____                            |
| 22                         | 1C concentrator overheads            | _____                            |
| 23                         | Concentrated U bearing acid stream   | 4-5 gU/l                         |

<sup>a</sup> From Figure VII.B-2.

TABLE VII.B-2. (Continued.)

| Stream Number <sup>a</sup> | Identification                       | Description                                    |
|----------------------------|--------------------------------------|--|
| 24                         | Nitric acid scrub                    | _____  |
| 25                         | Clean organic solvent                | 30% TPB in hydro-carbon diluent                |
| 26                         | 2A column waste (to recycle)         | _____  |
| 27                         | Loaded organic (U only)              | _____  |
| 28                         | Nitric acid strip                    | ~0.1M  |
| 29                         | Aqueous U-nitric acid stream         | _____  |
| 30                         | Recycle organic                      | _____  |
| 31                         | U product concentrator overheads     | _____  |
| 32                         | U product                            | 300 gU/l                                       |
| 33                         | U product                            | 300 gU/l                                       |
| 34                         | Concentrated Th-nitric acid solution | 300 gTh/l                                      |
| 35                         | Th calciner overheads                | _____  |
| 36                         | Steam                                | _____  |
| 37                         | Calcined ThO <sub>2</sub>            | 2-5 gm/cm <sup>3</sup> ThO <sub>2</sub> powder |
| 38                         | IR recovery column waste             | _____  |

<sup>a</sup> From Figure VII.B-2.

Thorium metal spheres discharged from a suppressed fission hybrid blanket are loaded from the reactor storage compartments into cylindrical tubes and transferred (1)\* to the sodium removal cell. All operations are carried out in an inert atmosphere to prevent sodium fires. In the sodium

\*Numbers refer to process lines on Figure VII.B-2.

removal cell, the transfer tube is placed in a concentric cylindrical unit where a mixture of water vapor and nitrogen gas is pumped through the sphere bed. Reaction with the water vapor affects the sodium removal. A more detailed discussion of the process (as applied to LMFBR fuel assemblies) can be found in Reference 11. Following sodium removal, the transfer tubes are removed, end fittings are screwed-on, and the tubes are temporarily stored in a water basin. The basin capacity is one month's throughput capacity (~100 MTHM), but is normally kept half-full.

After removal from the interim storage basin, the spheres are loaded into a dissolver feed hopper (2) where they are continuously fed to the rotary kiln dissolver. In the dissolver, the spheres are immersed in a 13M nitric acid solution (3) containing a small amount of fluoride catalyst (~0.001 to 0.005M HF). Upon leaving the dissolver (4), the thorium-uranium bearing nitric acid solution is centrifuged and passed (5) to the accountability tanks. From the accountability tanks, the solution is transferred to adjustment tanks where the chemical content is adjusted to approximately 1.0M nitric acid, 1.07M thorium, and 1.8 gU/ℓ.

The clarified and chemically adjusted solution (8) is fed to the HA pulse column where the uranium and thorium are extracted into an organic phase (13) (30% tri-n-butyl phosphate). The majority of the fission products (FPs) and other impurities remain in the aqueous raffinate stream (14). The thorium is then stripped from the organic phase (13) in the 1B column. The U remains in the organic stream (17). Criticality is prevented by concentration control and neutron poisons (pulse column plates contain a strong neutron absorber).

The aqueous stream (18) containing the thorium is then concentrated, sampled, calcined to oxide (34), and placed in storage for 10-15 years (37). This storage period allows the decay of <sup>228</sup>Th and its daughter products to a level where hands-on chemical conversion to metal (i.e., a "bomb" process) can be performed. If the discharge fuel were thorium oxide rather than thorium metal, the 10-15 year storage might be eliminated and the calcined oxide might be remotely refrabricated back into the blanket fuel form. This operation would be performed on-site.

The organic stream (containing the U) exiting the 1B column is combined with another organic stream (30) which is backcycled from the uranium purification columns. The combined organic stream (17) enters the 1C column where the U is back extracted to the aqueous phase (21). After concentration (23), the uranium is extracted into the organic phase in the 2A pulse column (27) and stripped back to an aqueous phase in the 2B column (29). Criticality control is by concentration and poison plates in the 1C column and by geometry (critically safe column diameters) in the 2A and 2B columns. After concentration in the U product concentrator (32), the uranium solution is sampled and transferred to the LWR fuel fabricator (33). The aqueous raffinate (26) from the 2A column is routed to a recovery column (1R) where any remaining uranium is extracted and backcycled to the HA column (10).

In addition to the processes described above, the plant will also include systems for off-gas treatment, solvent recovery, acid and water recovery, high level liquid waste concentration, storage, and solidification, building ventilation, process cooling and heating, and thorium storage. A detail description of these systems is not provided since they are similar (with the exception of the thorium storage) to the systems in previously design fuel reprocessing plants.<sup>7,8,9,11</sup> However, due to the decreased burnup of the thorium spheres in comparison to LWR fuel, the throughput of all these auxiliary systems will be greatly reduced. The thorium dioxide storage facility is a set of fifteen, buried, carbon steel tanks, approximately 12 m in diameter and 5 meters in depth.

Process Equipment and Facilities. The equipment required for the reprocessing flowsheet described in the previous section is similar to that of the Barnwell Nuclear Fuel Plant (BNFP).<sup>7,8</sup>

The correspondence is:

| <u>BNFP Equipment Designation</u>          | <u>Figure VII.B-2 Designation</u> |
|--|-----------------------------------|
| HS   | HA                                |
| 1BX  | 1B                                |
| 1C   | 1C                                |
| 2A   | 2A                                |
| 2B   | 2B                                |
| 1S   | 1R (somewhat smaller than the 1S) |
| 2E Concentrator                            | Th Concentrator                   |
| 3P Concentrator                            | 1C and U Product Concentrator     |
| U Calciner (in BNFP UF <sub>6</sub> Plant) | Th Calciner                       |

The equipment will be fabricated from 304 stainless steel with the possible exception of the dissolver, associated tanks, HA column, and parts of the waste treatment system. Other alloys, more resistant to attack by fluoride, could be used for this equipment. Equipment in the auxiliary systems is similar to that described for the corresponding system in references 7 and 8. A brief description of the components in each major system of the reprocessing plant is provided in Table VII.B-3.

The layout of the blanket reprocessing facility is shown in Figure VII.B-3. The materials are basically reinforced concrete with stainless steel liners on the inside of each cell.

The Na removal cell houses the transfer crane, the transfer airlock, the Na removal unit, and the associated chemical feed tanks and equipment. The remote process cell contains the interim water storage tank, dissolvers, crushers, burners, solids handling equipment and high level waste concentrator. All operations and maintenance in the cell are performed remotely.

The high level cell contains the dissolver solution transfer, solution surge, accountability, feed adjustment and HA feed tanks, the centrifuge, the HA column, and their associated piping and auxiliary equipment. The cell is designed for remote operation and contact maintenance after process fluid removal and decontamination of the equipment.

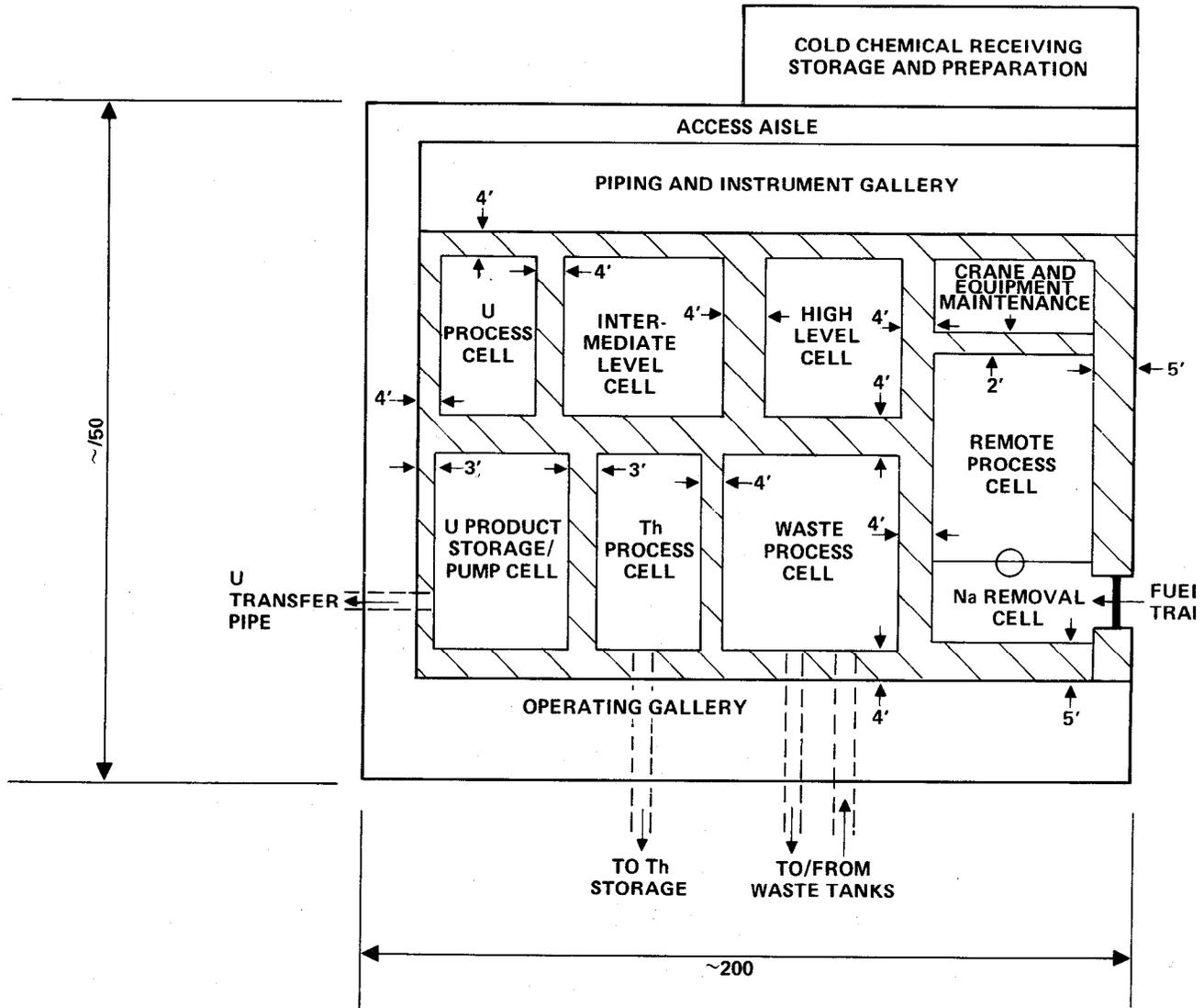


Figure VII.B-3. Facility layout

TABLE VII.B-3. Reprocessing plant components.

| Component                               | Description   |
|---|---|
| 1. Head End                             |   |
| Fuel receiving and sodium removal       | Batch operation: cylindrical container with screw-on end fittings   |
| Interim storage                         | Water basin capacity for 1 mo. operation  |
| Accountability                          | Weight measurements, gamma scan, neutron measurements   |
| 2. Feed Preparation                     |   |
| Dissolution                             | Rotary kiln dissolver   |
| Centrifuge                              | Vertical, solid bowl  |
| Accountability                          | Tank with weight, gamma, and neutron metering instruments   |
| Feed adjustment                         | Tank  |
| 3. Solvent Extraction                   |   |
| Pulse columns and associated equipment  | (1) large co-extraction column<br>(1) large Th stripping column<br>(1) medium U stripping column<br>(2) small U purification columns<br>(1) medium recovery column<br>(1) intercycle concentrator |
| 4. U Product Preparation                |   |
| Concentration and Sampling              | U product concentrator<br>U product catch tank<br>U product sample tank<br>U product storage tanks<br>U product pumping system  |
| 5. Th Product Preparation               |   |
| Concentration, Sampling and Calcination | Th product concentrator<br>Th product catch tank<br>Th product sample tank<br>Th fluidized bed calciner<br>ThO <sub>2</sub> storage transfer system   |
| 6. Th Storage                           |   |
| Storage and Recovery                    | ThO <sub>2</sub> storage tank<br>ThO <sub>2</sub> slurring and pumping system   |

TABLE VII.B-3. (Continued.)

| Component                                 | Description  |
|---|--|
| 7. Off-Gas Treatment                      |  |
| Dissolver Off-Gas (DOG)                   | Particulate, ruthenium, iodine and nitrogen oxides removal                                 |
| Vessel Off-Gas (VOG)                      | Particulate, ruthenium, iodine and nitrogen oxides removal                                 |
| 8. Acid Recovery                          |  |
| Capture, Treatment and make-up            | Concentrator overhead catch tanks, chemical adjustment system                              |
| 9. Solvent Recovery                       |  |
| Treatment and make-up                     | Filtration, aqueous wash column, ion exchange column, catch and feed tanks                 |
| 10. High-Level Waste Treatment            |  |
| Concentration, storage and solidification | Waste feed tanks, concentrator, underground storage tanks, spray calciner and glass melter |
| 11. Low-Level Waste Treatment             |  |
| Collection and Transfer                   | Transfer cask, 55 gal. drums, boxes, etc.  |

The intermediate level cell houses the 1B, 1C, and 1R columns, the 1C concentrator, and their associated piping and auxiliary equipment. This cell also contains the off-gas treatment and solvent recovery equipment. The cell is remotely operated and contact maintained.

The U process cell is also remotely operated and contact maintained and houses the 2A and 2B columns and the U product concentrator. The U product storage/pump cell contains the catch, sample storage and transfer tanks and the U solution pumps for transfer to the LWR fuel fabricator.

The Th process cell houses the Th product concentrator, the catch, sample and calciner feed tanks, and the Th calciner. This cell also utilizes contact maintenance and remote operation.

The waste process cell contains the waste calciner, the glass melter, the transfer equipment (liquid to and from storage and solid to storage and shipment) and a water-cooled storage basin. The cell is remotely operated and maintained.

VII.B.1.b.(3). Process Cost. The unit cost (\$/KgHM processed) was determined by estimating the capital and operating cost of the facility. These capital and operating costs were estimated on a modular basis in a manner similar to that described in reference 3. The economic assumptions used are:

- 1980 dollars (end of second quarter)\*
- 15.2% fixed charge rate on capital (typical utility financing)\*

Table VII.B-4 provides a breakdown on the capital and operating cost and the resulting unit price for three different size reprocessing facilities.

TABLE VII.B-4. Cost summary for thorium metal reprocessing plant.

| Plant Throughput (MTHM/YR)                                   | 600 | 1200 | 1800 |
|--|-----|------|------|
| Head End, \$/M <sup>a</sup>                                  | 81  | 137  | 185  |
| Solvent Extraction, \$/M                                     | 19  | 26   | 32   |
| Waste Treatment (including Th calcination and storage), \$/M | 26  | 37   | 45   |
| Balance of Plant, \$/M                                       | 68  | 103  | 131  |
| Total Equipment and Facility Direct Cost, \$/M               | 194 | 303  | 393  |
| Field Indirect Cost, \$/M                                    | 39  | 61   | 79   |
| Home Office Cost, \$/M                                       | 47  | 73   | 94   |
| Contingency, \$/M  | 56  | 87   | 113  |
| Owners Cost, \$/M  | 23  | 37   | 48   |
| Time Value of Money During Construction, \$/M                | 48  | 75   | 97   |
| Total Indirect Cost, \$/M                                    | 213 | 333  | 431  |

<sup>a</sup> 1980 Dollars.

\* Both compatible with Chapter IX assumptions.

TABLE VII.B-4. (Continued.)

| Plant Throughput (MTHM/YR)                         | 600 | 1200 | 1800 |
|--|-----|------|------|
| Total Capital Investment, \$/M                     | 407 | 636  | 824  |
| Levelized Annual Capital Cost, \$/M <sup>a</sup>   | 62  | 97   | 125  |
| Levelized Annual Operating Cost, \$/M <sup>b</sup> | 43  | 57   | 67   |
| Levelized Total Annual Cost, \$/M                  | 105 | 153  | 192  |
| Levelized Cost Per Kilogram Thorium, \$/Kg         | 175 | 127  | 107  |

<sup>a</sup> Based upon 15.2%/yr capital charge (typical of utility financing).

<sup>b</sup> Operating cost levelizing factor = 2.038.

Useful scaling relationships can be generated using the information in Table VII.B-4.

Referring to Table VII.B-4, the total capital investment scales with throughput as follows:

$$I = 407 \cdot 10^6 (T/611)^{0.653} \quad (1)$$

where T (MT/yr) is the annual heavy metal throughput. The levelized annual capital cost for a 15.2% annual change rate (typical of utility financing) is then given by

$$A = 62.0 \cdot 10^6 (T/611)^{0.653} \quad (2)$$

Similarly, the levelized annual operation and maintenance cost scales with throughput as follows:

$$B = 4.28 \cdot 10^6 (T/611)^{0.411} \quad (3)$$

Concerning the levelized cost method, operating and maintenance costs escalate with time due to inflation and other factors. As a result, a factor (greater than unity) is included in Table VII.B-4 and equation 3 to adjust the first year operating cost estimates upwards to reflect such increases over the operating life of the plant<sup>12</sup>. The levelizing factor represents a disconnected average contribution for each year and is given by

$$\ell = \frac{\sum_{n=1}^N \left( \frac{1+e}{1+d} \right)^n}{\sum_{n=1}^N \left( \frac{1}{1+d} \right)^n} \quad (4)$$

where

N = financial life of the plant (yr)

e = average cost escalation

d = discount rate (cost of financing equity and debt).

If we again assume typical utility financing with a 30 year plant life and an 11.35%/year discount rate, then  $\ell = 2.038$ .

Therefore, the total levelized annual cost of reprocessing plant operation, as a function of throughput, is given by

$$L = [62.0 (T/611)^{0.653} + 42.8 (T/611)^{0.411}] \cdot 10^6 \quad (5)$$

The levelized cost per kilogram of thorium is then given by

$$K = \frac{L}{1000T} \quad (6)$$

and the levelized cost per gram of  $^{233}\text{U}$  is given by

$$G = \frac{K}{1000 a_{23}} = \frac{L}{10^6 T a_{23}} \quad (7)$$

Where  $a_{23}$  is the discharge assay of  $^{233}\text{U}$  (including  $^{233}\text{Pa}$ , if any) in thorium. For example, for  $T = 1200 \text{ MT/yr}$  and  $a_{23} = 0.0055$  (0.55%) the above equations give  $L = 153 \cdot 10^6 \text{ \$/yr}$ ,  $K = 127 \text{ \$/KgHM}$ , and  $G = 23.18 \text{ \$/g}$ .

As discussed earlier, these unit prices are considerably lower than most reference LWR reprocessing plants due to the numerous differences between the conceptual design outlined in this section and standard LWR reprocessing facilities. Nevertheless, there is considerable uncertainty

associated with the cost due to the thorium metal fuel form. If greater levels of fluoride are required to dissolve the thorium metal and more extensive off-gas treatment is required, the cost could increase substantially. However, if the thorium metal dissolves easily in nitric acid, and no fluoride is required, the cost could decrease. An experimental program to better understand the dissolution rate will be required.

VII.B.1.c. Considerations for Thorium Oxide TMHR Fuel Reprocessing. A conceptual design and cost study of a reprocessing plant for low burnup thorium oxide fuels of discharge concentration similar to the thorium metal fuels discharged by the beryllium/thorium oxide TMHR is not available. Nevertheless, a recent Exxon study<sup>3</sup> did compare reprocessing plants and costs for ThO<sub>2</sub> versus UO<sub>2</sub> fuels discharged by LWRs. Their principal conclusion relating to this comparison was as follows:

"The increased costs of processing thorium-based fuels result primarily from the relatively refractory nature of thoria which necessitates the use of more dissolvers (16 vs 4 hours dissolution time); more evaporation load because of the dilute flowsheets; more frequent equipment replacement because of corrosion from the fluoride catalyst used to accelerate dissolution and from the equipment erosion caused by small particles of undissolved thoria; the increased volume of high-level wastes resulting from the presence of fluoride ion and its metallic complexants."

These increased costs were expressed in their work as relative scaling factors. In particular, Exxon concluded that for LWR fuels, the capital cost of a ThO<sub>2</sub> reprocessing plant would be 1.9 times that of a UO<sub>2</sub> reprocessing plant. Similarly, they conclude that the operation and maintenance cost ratio would be 1.7.

The Bechtel thorium metal reprocessing plant design is expected to be very similar to a similar plant design for unclad, low burnup UO<sub>2</sub> fuel under similar discharge conditions. This results because the thorium metal with only minimal amounts of fluoride added is expected to dissolve as easily as uranium oxide. Therefore, a crude estimate of the reprocessing for low burnup thorium oxide fuels discharged by the TMHR may be deduced based upon the Bechtel plant costs<sup>6</sup> and the Exxon scaling factors.<sup>3</sup>

Such an estimate may be conservative for three reasons:

- The  $\text{ThO}_2$  proposed for use in the TMHR may dissolve more easily than LWR  $\text{ThO}_2$ . This could result from either smaller  $\text{ThO}_2$  particle size or the use of an uncalcined  $\text{ThO}_2$  fuel form.<sup>13</sup>
- The possible use of KF instead of HF to aid dissolution.<sup>13</sup>
- NASAP results<sup>5</sup> indicate that thorium oxide is only about 20% more costly to reprocess than uranium oxide.

In contrast, such an estimate could be optimistic. In the case of the Bechtel process, sodium coolant is stripped from the thorium metal by use of a straightforward process involving contact with steam.<sup>11</sup> If the TMHR thorium oxide is mixed with Pb-Li, both lead contamination and tritium could complicate the process. The following cost estimate is, therefore, inadequate and will be only indicative of thorium oxide reprocessing costs.

Applying the Exxon cost factors directly to Eq. (5), we obtain

$$L = [118 (T/611)^{.653} + 73 (T/611)^{0.411}] \cdot 10^6 \quad (8)$$

where T is the heavy metal throughput and the quantities K and G are calculated as before. Using the same example of  $T = 1200$  MT/yr and  $a_{23} = 0.0055$  we obtain the following:  $L = 280 \cdot 10^6$  \$/yr,  $K = 233$  \$/Kg, and  $G = 42.42$  \$/g. This conservative model is compared with a less conservative model of the thorium oxide reprocessing costs based upon NASAP data in Table VII.B-5. As expected, the conservative model results in unit processing costs which are about 50% higher than those predicted using the less conservative model. However, the high cost and large uncertainties associated with both models strongly suggest the need to perform further analysis with regards to  $\text{ThO}_2$  reprocessing if this option is retained.

TABLE VII.B-5. Estimates of thorium oxide reprocessing costs.<sup>a</sup>

| Plant Throughput (MTHM/YR)  | 600     | 1200    | 1800    |
|---|---------|---------|---------|
| Conservative projections based upon Exxon/Bechtel Work<br>(1.9 X Capital, 1.7 X Operating)              |         |         |         |
| Levelized Annual Capital Cost, \$/M   | 116     | 183     | 239     |
| Levelized Annual Operating Cost, \$/M   | 73      | 97      | 114     |
| Levelized Total Annual Cost, \$/M   | 189     | 280     | 353     |
| Levelized Cost Per Kilogram Thorium,<br>\$/KgHM   | 315     | 233     | 196     |
| -----   |         |         |         |
| Less conservative projections based upon final NASAP report<br>(1.2 X uranium oxide reprocessing costs) |         |         |         |
| Levelized Cost Per Kilogram Thorium,<br>\$/KgHM   | 210     | 152     | 128     |
| -----   |         |         |         |
| Range of Levelized Cost Per Kilogram<br>Thorium, \$/KgHM  | 210-315 | 152-233 | 128-196 |

<sup>a</sup>See Table VII.B-4 notes.

## VII.B.2 Application of Molten Salt Reprocessing for TMHR Fuel Reprocessing

VII.B.2.1 Overview. Many aspects of molten salt technology were developed, or were under study, over a more than 20-year period at the Oak Ridge National Laboratory.<sup>14,15</sup> The object of that development program was to have been a Molten Salt Breeder Reactor (MSBR) to produce an appreciable excess of  $^{233}\text{U}$ , through the intermediate  $^{233}\text{Pa}$ , from  $^{232}\text{Th}$ . The proposed MSBR was to have been a high-temperature reactor constructed of modified Hastelloy N; the fuel was to have been a molten mixture of  $^7\text{LiF}$ ,  $\text{BeF}_2$ ,  $\text{ThF}_4$ , and  $^{233}\text{UF}_4$  (71.7, 16, 12, and 0.3 mole %, respectively) pumped through a core containing unclad graphite as the neutron moderator and through an external heat exchanger for removal of the 2250  $\text{MW}_{\text{th}}$  produced by fission of  $^{233}\text{U}$ .<sup>16</sup>

Results from this large, though still incomplete, development effort suggest that a molten fluoride blanket for a TMHR could be maintained and that continuous reprocessing of such a blanket could be developed and demonstrated. The following subsections present (1) characteristics of the TMHR blanket system, (2) the chemistry of salt processing and salt maintenance, (3) descriptions of two alternative processes for the TMHR blanket, (4) estimates of capital and operating costs for the processes and (5) a description of the development program required to demonstrate such reprocessing for TMHR applications.

VII.B.2.2. Characteristics of the Molten Salt TMHR Blanket. The reference molten salt TMHR blanket (described more fully in Chapter IV) is to contain a molten mixture of  $\text{LiF}$ ,  $\text{BeF}_2$ , and  $\text{ThF}_4$  with the composition and physical properties<sup>17</sup> shown in Table VII.B-6 below. This salt, for which the  $\text{LiF}$  is to be prepared from 99.8%  $^7\text{Li}$ , contains 44% Th by weight. It is estimated that a total of 1152  $\text{m}^3$  (3860 metric tons) of this salt is required.

TABLE VII.B-6  
COMPOSITION AND PROPERTIES OF TMHR BLANKET SALT

|                                |                  |            |
|--------------------------------|------------------|------------|
| Composition (mole %)           | LiF              | 72         |
|                                | BeF <sub>2</sub> | 16         |
|                                | ThF <sub>4</sub> | 12         |
| Liquidus (°C)                  |                  | 500        |
| Properties at 600°C            |                  |            |
| Density (g/cm <sup>3</sup> )   |                  | 3.35       |
| Heat capacity (cal/g°C)        |                  | 0.33       |
| Viscosity (centipoise)         |                  | 12         |
| Vapor pressure (torr)          |                  | Negligible |
| Thermal conductivity (W/°C cm) |                  | 0.011      |

Such a blanket, with a fusion driver of 3000 MW<sub>th</sub>, is estimated to produce 0.202 grams of <sup>233</sup>Th per second and to produce <sup>3</sup>H at a rate of 2.654 x 10<sup>-4</sup> grams per second. The blanket is assumed to be processed at a rate such that the concentration of <sup>233</sup>U is maintained at 1.1 x 10<sup>-3</sup> times the concentration of Th. Such a processing rate corresponds to an inventory of 1870 kg <sup>233</sup>U in the blanket salt, with 4.845 x 10<sup>-4</sup> grams <sup>233</sup>U per gram of salt and with 0.202 grams <sup>233</sup>U (or equivalent) generated and removed each second. Under such conditions the fission rate in the blanket is estimated to be about 800 MW<sub>t</sub>. This fission heat along with decay heat from the <sup>233</sup>Th and <sup>233</sup>Pa, is removed by circulating the salt at a rate of 3.75 x 10<sup>4</sup> metric tons per hour (186.6 m<sup>3</sup>/min) through external heat exchangers of Hastelloy N where the heat is transferred to metallic sodium.

Molten fluoride mixtures for MSRE and for many chemical and engineering experiments have been prepared by treating batches of the mixed materials in the molten state at about 600°C with anhydrous H<sub>2</sub>-HF mixtures and, subsequently, with H<sub>2</sub>.<sup>18-20</sup> Such treatment should certainly suffice for initial preparation of the TMHR blanket salt, though countercurrent (continuous) procedures would seem necessary for preparation of quantities as large as those required. Costs of the materials and of the necessary conversion and purification steps to provide the initial blanket salt are shown in Table VII.B-7 below. The initial charge of TMHR salt (1152 m<sup>3</sup>) would entail a cost of about \$192 x 10<sup>6</sup>; this cost represents a relatively small fraction of that of the total system, but advantages can clearly accrue if a smaller volume of blanket salt can suffice.

Table VII.B-7  
COST (PER CUBIC METER) OF INITIAL CHARGE OF TMR BLANKET SALT\*

| Initial blanket salt   | Cost, dollars/m <sup>3</sup> |
|--|------------------------------|
| <b>Materials</b>   |                              |
| <sup>7</sup> Li (267.13 kg at \$160/kg)                        | 42,745                       |
| Th (1679.3 kg ThO <sub>2</sub> at \$33/kg)                     | 55,415                       |
| BeF <sub>2</sub> (398.6 kg at \$33/kg)                         | 13,155                       |
| <b>Conversion and Purification</b>                             |                              |
| <sup>7</sup> Li (992.2 kg LiF at \$2.20/kg)                    | 2,183                        |
| Th(1959.1 kg ThF <sub>4</sub> at \$4.40/kg)                    | 8,620                        |
| LiF-BeF <sub>2</sub> -ThF <sub>4</sub> (3350 kg at \$13.25/kg) | 44,388                       |
| <b>Total</b>   | <b>166,500</b>               |

\*The <sup>6</sup>Li depleted lithium (99.8% <sup>7</sup>Li) cost of 160 \$/kg is based upon work performed by McGrath and assumes a 20 \$/kg cost of natural lithium with a 50 \$/kg separative work cost for lithium isotope separation. Other costs from ref. 21, page 124.

#### VII.B.2.C Chemistry of Salt Maintenance and Processing

VII.B.2.C.(1) Fission Product Behavior. Since the MSBR was not built and operated, most of the (still incomplete) understanding of fission product behavior in molten fluoride systems was obtained during operation of the Molten Salt Reactor Experiment (MSRE). The observed behavior has been described in detail;<sup>22</sup> a few pertinent points are noted below.

Fission products that originate in a well mixed molten salt (as in MSRE or TMR) must come to a steady state as commonly encountered chemical entities. The valence states that they assume in the liquid are presumably defined by the requirements that cation-anion equivalence be maintained and that redox equilibria be established among the components of the melt and between the melt and the surface layers of the container metal.<sup>17,23</sup> Early assessments<sup>19,24</sup> suggested that fission product cations would be equivalent to the fission product anions plus the fluoride ions released by fission only if some of the cations assumed oxidation states corrosive to Hastelloy N. Inclusion, and

maintenance, of a small quantity of  $UF_3$  in the MSRE fuel was adopted to avoid the possibility of corrosion from this source.

The fission product gases Kr and Xe form no compounds under conditions existing in a molten salt reactor or in TMHR, and these gases are only sparingly soluble in molten fluorides.<sup>25,26</sup> Neither of these gases diffuse appreciably through metals, and they can be removed from molten fluorides by sparging with He. Such sparging was done for MSRE,<sup>27</sup> and was to be done for MSBR.<sup>16,28,29</sup>

The fission products Rb, Cs, Sr, Ba, yttrium, the rare earths, and Zr all form quite stable fluorides that are soluble in the molten fuels of MSRE, MSBR, and in the very similar salt to be used as the TMHR blanket. Consideration of solubility relationships among the actinide and rare earth trifluorides for an unprocessed version<sup>21</sup> of a large molten salt reactor, indicates that 30 years operation of TMHR should not result in precipitation of any of the fluorides of these elements.

The fission products Ge, As, Nb, Mo, Ru, Rh, Pd, Ag, Cd, Sn, Sb, Se, and Te were expected to exist in MSRE largely, if not entirely, in the elemental state and to precipitate within the reactor circuit.<sup>17,19</sup> This behavior seems to have been confirmed during MSRE operation,<sup>22</sup> but additional study will be required before details of their behavior can be predicted with confidence for TMHR (or for MSBR). Precipitation of these species within MSBR was adjudged to be tolerable,<sup>16,29,30</sup> and problems posed by these species should prove manageable in TMHR.

However, operation of MSRE led to embrittlement of the grain boundaries of Hastelloy N exposed to the molten fuel; shallow cracks were observed in the heat exchanger. Early studies<sup>31</sup> suggested, and subsequent work<sup>32</sup> has shown, that fission product Te is responsible for this embrittlement. Hastelloy N modified by addition of 1 to 2% Nb appears markedly more resistant than unmodified or Ti-modified Hastelloy.<sup>32</sup> However, a reasonably small change in fuel chemistry has been shown to be even more markedly beneficial.<sup>32-34</sup> Careful experiments have shown that crack frequency and crack depth both decrease markedly in Hastelloy specimens exposed to tellurium-bearing salts as the  $UF_3/UF_4$  ratio is increased (that is, as the melt is made more reducing). Additional study will be required, but it seems likely that use of a moderate  $UF_3/UF_4$  ratio in the fuel, perhaps combined with use of the Nb-modified Hastelloy-N, will alleviate the Te problem for TMHR.

In brief, the ability of molten fluorides to withstand irradiation, fission, and grow-in of fission products appears to be demonstrated. As indicated above, not all details of fission product behavior are understood, and further study is necessary. It is not, however, considered likely that deleterious effects of fission products would prejudice operation of a molten-salt blanket for TMHR.

VII.B.2.C.(2). Chemistry of Molten Fluoride Processing. The proposed MSBR required processing of about 190 grams of fuel salt per second to pass its entire inventory (49 m<sup>3</sup>) through its processing plant each ten days. It was necessary<sup>35,36</sup> that the process remove soluble fission product fluorides (notably those of the rare earths) that are important neutronic poisons and remove the <sup>233</sup>Pa from the fuel for decay to <sup>233</sup>U outside the neutron flux. Removal of these species required prior removal of <sup>233</sup>U from the fuel, and the process had to include steps to return most of this <sup>233</sup>U to the purified fuel stream. In addition, the MSBR process was required to perform several fuel maintenance functions (see VII.B.2.C.(3), below). Several of these operations will be required in management and processing of the TMHR blanket salt.

Key features of the proposed MSBR process were studied for several LiF-BeF<sub>2</sub>-ThF<sub>4</sub>-UF<sub>4</sub> compositions, but most of the studies, and essentially all of the conceptual designs, assumed use of LiF, BeF<sub>2</sub>, ThF<sub>4</sub>, and UF<sub>4</sub> (71.7, 16, 12, 0.3 mole %, respectively). Accordingly, most of what is known about MSBR processing is directly applicable to the salt for the reference design THMR. However, it must be noted that fuel processing for MSBR was far from a demonstrated reality at the termination of that effort. All of the key chemical separations had been repeatedly demonstrated on a small scale, and the process chemistry was well understood. However, much work (especially on suitable materials of construction and upon engineering development) remains to be done before overall feasibility can be assured.<sup>14,15</sup>

Uranium can be recovered from solutions in molten fluorides by fluorination to produce volatile UF<sub>6</sub>. Batch fluorinations can reduce the uranium concentration of the molten fluoride to less than one part per million by weight.<sup>36,37</sup> Such batch fluorinations were proposed<sup>36</sup> for recovery of uranium from salt to be discarded (as waste) from the <sup>233</sup>Pa decay system of the MSBR

processing plant. A continuous fluorinator<sup>14,15,36</sup> was to be used to remove most of the uranium from the MSBR fuel. Such fluorination was conservatively assumed to remove 95 to 99% of the uranium from a feed stream containing 0.3 mole % UF<sub>4</sub>.<sup>14,23</sup>

Fluorination also serves to remove fission product halogens (as volatile BrF<sub>5</sub> and IF<sub>5</sub>) from the molten fluoride and removes the oxide ion, which might be present as an inadvertent contaminant, by volatilization as O<sub>2</sub>. Any tritium present in the molten fluoride would be converted to <sup>3</sup>HF and partially removed with the UF<sub>6</sub> and excess F<sub>2</sub>. The small quantities of Kr and Xe would also be removed in that gas stream. In addition, several fission product species (including Se, Mo, Tc, Ru, Sb, and Te) would be partially converted to volatile fluorides and be removed with the uranium;<sup>23</sup> however, these species largely precipitate within the reactor circuit (see VII.B.2.C.(1), above) and their concentrations in the molten salt to be processed should be very small. Fluorination will not remove protactinium, nor will it remove transuranic elements, such as Pu, or nonvolatile fission product compounds, such as the rare earth fluorides, from the molten fluoride stream.

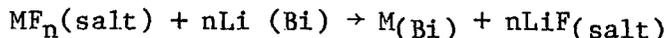
After fluorination, the molten fluoride would contain a small quantity of fluorine, some dissolved UF<sub>5</sub> and oxidized states of some fission product species. This salt stream would require treatment with gaseous H<sub>2</sub> for reduction of these oxidized and oxidizing species before return to the active circuit or before passage to other processing steps.

The fluorinator and the H<sub>2</sub> purge column could be constructed of Ni or a high Ni alloy but these pieces of equipment would require protection by a layer of frozen salt on all wetted surfaces of the units.<sup>36</sup> Development of continuous fluorination equipment was underway in the MSBR program,<sup>14,15</sup> but was still incomplete at the termination of that effort.

Recovery of protactinium and of uranium remaining in salt streams after fluorination can be accomplished by reductive extraction into alloys of bismuth containing lithium and thorium metals. Fission product Zr can also be removed, and (with more difficulty) fission products such as the rare earths and the alkaline earths can also be removed.<sup>14,15,36</sup> Early studies<sup>17,38,39</sup> showed the versatility of such reductive extractions, but most of the definitive studies of selective reductions from fluoride mixtures, however, are due to Ferris et al.<sup>40-46</sup>

Bismuth is a low-melting (271°C) metal that is essentially immiscible with molten halides. Its vapor pressure is negligible over the required temperature interval (500–700°C), and it can dissolve adequate quantities of Li, Th, U, Pa, and the fission product elements of interest. Fortunately, Be is essentially insoluble in Bi and is not appreciably extracted.

Reductive extraction equilibria between materials in salt and metal phases can be represented by the reaction:



in which the fluoride of metal M in the salt phase reacts with lithium from the bismuth phase to produce metal M dissolved in the bismuth and n moles of LiF in the salt. It has been found<sup>14,43</sup> that the distribution coefficient (D) for metal M depends on the mole fraction of lithium ( $X_{\text{Li}}$ ) in the metal phase:

$$\log D = n \log X_{\text{Li}} + \log K_M$$

where  $X_{\text{Li}}$  is the concentration (in mole fraction) of Li in the Bi after equilibrium is established. The quantity  $K_M$  for each element depends only upon the temperature and D (the distribution coefficient) is defined by the relationship

$$D = \frac{\text{mole fraction of M in metal phase}}{\text{mole fraction of } MX_n \text{ in salt phase}}$$

The separation factor for any pair of elements is the ratio of their distribution coefficients.

Table VII.B-8 shows values of n (the effective valence of fluorides involved in the reduction) and  $K_M$  for a number of pertinent species at 600°C along with the distribution coefficient for each at that temperature for a typical lithium concentration ( $X_{\text{Li}} = 1.22 \times 10^{-3}$ ). Such reductive extraction affords an excellent separation of U (and fission product Zr) from Th and a good separation of Pa from Th. For example, extraction with three equilibrium stages and flows such that 5 moles of salt are equilibrated with 1 mole of Bi alloy results in removal of 99.5% of the <sup>233</sup>Pa and essentially 100% of the <sup>233</sup>U and Zr from the salt. Good recovery (and separation from Th) of the transuranics can be obtained but such recovery is of little consequence to TMHR. However, the lanthanide and the alkaline earth elements are poorly extracted from the molten fluoride and are poorly separated from thorium by such extraction.

Table VII.B-8. VALUES FOR n, log K, AND D FOR REDUCTIVE EXTRACTION  
FROM 72LiF-16BeF<sub>2</sub>-12ThF<sub>4</sub> AT 600°C\*

| Specie | n   | log K  | D <sup>†</sup> |
|--------|-----|--------|----------------|
| U      | 3   | 11.056 | 561            |
| Pa     | 4   | 12.467 | 24.5           |
| Th     | 4   | 9.082  | 0.0101         |
| Zr     | 4   | 14.683 | 4045           |
| Np     | 3   | 10.514 | 161            |
| Pu     | 3   | 9.979  | 47.0           |
| La     | 3   | 6.628  | 0.021          |
| Nb     | 3   | 6.801  | 0.024          |
| Pm     | 3   | 6.575  | 0.019          |
| Sm     | 2.6 | 5.734  | 0.034          |
| Eu     | 2   | 3.740  | 0.016          |
| Ba     | 2   | 4.049  | 0.032          |

\*Values for n and log K from references 34 and 38.

†Values for D at  $X_{Li} = 1.22 \times 10^{-3}$ .

All of the dissolved metals (including the Li and Th) can be completely and rapidly transferred from the Bi phase to a molten fluoride by treatment of the Bi alloy with anhydrous HF in the presence of the salt.

The fraction of lanthanides and alkaline earths that are extracted can be improved by use of large volumes of Bi alloy with small volumes of salt though the factors for separation of these species from Th are not affected. Smith and Ferris<sup>44</sup> observed that the lanthanides are well separated from Th when Bi alloys are equilibrated with molten LiCl. With this information McNeese<sup>14,47,48</sup> was able to devise a reductive extraction, metal transfer process for removal of lanthanides and other fission product species from molten fluorides along with the <sup>233</sup>Pa and <sup>233</sup>U. The proposed processing plant for MSBR was quite small; processing at about 0.9 gallons/minute (0.205 m<sup>3</sup>/hr) served to pass the entire fuel volume through the plant each 10 days. A conceptual design and a preliminary cost estimate for such a reductive extraction, metal transfer plant was prepared by Carter and Nicholson.<sup>36</sup>

It should be possible to develop and adapt the reductive extraction, metal transfer process for removal of rare earth and other fission product species to the TMHR blanket system, and it is possible that such processing (which could effect some improvement in blanket safety problems) should be considered in further studies. For the present study, however, the reductive extraction of  $^{233}\text{Pa}$ ,  $^{233}\text{U}$ , and zirconium only has been considered.

Elimination of the metal-transfer steps considerably simplifies the proposed<sup>36</sup> process. It needs to be emphasized, however, that much research and development is still necessary (see VII.B.2.f, below) before the reductive extraction process can be engineered and its overall feasibility assured.<sup>14,15</sup> Even for the process as simplified for TMHR, three key problem areas are evident. The first lies in the scarcity of materials capable of containing both molten salts and bismuth alloys as required by the extractor assembly and by the hydrofluorinator which removes  $^{233}\text{Pa}$ ,  $^{233}\text{U}$ , etc. from the Bi alloy and transfers it to the  $^{233}\text{Pa}$  decay system salt. Graphite, molybdenum, and tungsten appear to be capable of long-term service in such environments,<sup>14,15</sup> but all these are very difficult to fabricate into complex assemblies. Tantalum and some of its alloys are easily fabricated and appear to be capable of withstanding the Bi alloys, but their resistance to the salt is questionable. It seems likely that recent advances in coating technology (i.e., plasma spraying, etc.) would permit coating of alloys, such as stainless steel or carbon steel, with adherent, stable coatings of the resistant metals.

A second, though less fundamental, problem area lies in the lack of engineering-scale testing of the several unit processes and, especially, in the lack of operation of such processes as a closely-coupled integrated system. Such studies were barely beginning when the MSBR program was terminated.<sup>15</sup>

Waste disposal may represent a third area of difficulty. Whether the solidified fluoride mixture with contained fission products will be an acceptable waste form is unknown (and, at present, probably unknowable). No studies of conversion of the fluoride mixture into another form, if required, have been attempted. For this study it is assumed that such a solid (and relatively insoluble) material can be considered an acceptable waste form.

VII.B.2.C.(3) Molten Salt Maintenance Functions. Several maintenance functions were required for the MSBR fuel and will be required for the TMHR blanket salt. Two of these, which can be performed in the fuel reprocessing plant and which are described (in VII.B.2.d, below) as part of the process alternatives, are:

- Adjustment of the salt composition to compensate for changes due to burnup, to transmutation, and to addition of reagents required by the process.
- Removal of oxide ion, should any be inadvertently introduced into the molten fluoride, to avoid precipitation of very sparingly soluble  $\text{ThO}_2$ ,  $\text{PuO}_2$ , and  $\text{UO}_2$ .<sup>24</sup> Fluorination of the blanket salt, proposed in both process alternatives, should effectively perform this maintenance function.

However there are two additional maintenance functions, related to fuel processing, that cannot be performed in the processing plant. These functions, described below, must be performed by equipment added as part of the system for removal of heat from the circulating blanket salt.

Maintenance of Required  $\text{UF}_3/\text{UF}_4$  Ratio. The deleterious effect of fission product Te on Hastelloy N appears to be markedly diminished if an appreciable fraction (perhaps 5%) of the uranium is maintained as  $\text{UF}_3$  (see VII.B.2.C.(2), above). This  $\text{UF}_3$  must be replenished at intervals since the fission of U and Th as tetrafluorides is oxidizing<sup>19,20</sup> as is the generation of  $^3\text{HF}$  from transmutation of LiF. Fission (see VII.B.2.b, above) at a level of about 800  $\text{MW}_T$  would be responsible for oxidation of about 3 moles of  $\text{UF}_3$  per day while generation of  $1.95 \times 10^{-3}$  grams of  $^3\text{HF}$  per second would oxidize an additional 7 moles of  $\text{UF}_3$  per day. Maintenance of the desired concentration of  $\text{UF}_3$  in MSRE was readily accomplished by reaction of metallic Be with dissolved  $\text{UF}_4$ ; solid rods of metallic Be in perforated baskets of Ni were lowered through the sampler system into the flowing fuel in the pump bowl.<sup>17</sup> For TMHR, addition of about 45 grams of Be per day would suffice to maintain the  $\text{UF}_3$  concentration after steady state conditions (and reduction of 5% of the uranium inventory) are reached. Though initial startup of the assembly will be more complex (and might necessitate addition of 25 kg of  $^{233}\text{U}$  to the blanket), maintenance of the required  $\text{UF}_3/\text{UF}_4$  ratio in TMHR seems to be a relatively trivial problem.

Removal of Tritium, Kr, and Xe from Blanket Salt. A purge system using He gas was to be used to remove fission product Kr and Xe from the MSBR fuel.<sup>16,28,29</sup> About 1% by volume of He was to be maintained as small bubbles in the circulating fuel, and 10% of the fuel flowing through the heat exchangers was passed through an assembly that included a gas separator and a He bubble generator. Helium was removed and added at a rate of 4.7 liters per second (10 cfm) to the 350 liters per second of molten salt; average residence time of a He bubble in the fuel was near 140 seconds. This system was adjudged adequate to keep the  $^{135}\text{Xe}$  poisoning in MSBR to an acceptable level,<sup>28</sup> and a similar system operating on a modest fraction of the total heat exchanger flow should prove adequate for Kr and Xe removal from TMHR.

Removal and recovery of tritium from TMHR (and MSBR) is more difficult. Tritium is, in principle, generated from LiF as TF, but this compound is largely reduced to the elemental form by the  $\text{UF}_3$  necessarily maintained in the salt. If a generation (and removal) rate of  $2.654 \times 10^{-4}$  grams/sec (22.93 grams per day) is assumed, more than 90% of the tritium atoms will be present in elemental form in salt with 5% of the uranium present as  $\text{UF}_3$ . Elemental tritium diffuses readily through hot metals; it was estimated that nearly 2000 Ci per day of tritium might diffuse (at an effective pressure of  $1.28 \times 10^{-6}$  atmospheres) through the MSBR heat exchangers<sup>49</sup> with the purge system described. It is clear that TMHR will require a more rapid purge rate than that proposed for MSBR if losses of tritium to the sodium coolant system are to be kept to tolerable limits.

The flow of salt to the proposed TMHR heat exchangers totals about 3.1  $\text{m}^3$  per second. It should prove possible (though demonstration would be required) to maintain 1% by volume of He bubbles in the blanket salt and to remove and add this gas (31 liters per second at  $600^\circ\text{C}$ ; equivalent to about 22.5 standard cubic feet per minute) from the total flow prior to passage of the heat exchangers. The mean residence time for He bubbles in the blanket system would be near 367 seconds. Equipment for removal and addition of the bubbles would be similar (though some nine-fold larger) than that described for MSBR.<sup>28,29</sup>

Operation of such a system under steady state conditions would lead to an inventory of about 0.12 grams of  $T_2$  and 0.04 grams of TF in the blanket. The salt plus gas bubbles fed to the stripper each hour would contain 1.2 grams of  $T_2$  and 0.4 grams of TF; of this, some 80% of the  $T_2$  and very little of the TF would be stripped. With tritium-free He reinjected prior to passage of the heat exchanger, the effective pressure of  $T_2$  in the salt within the heat exchanger would be near  $2 \times 10^{-5}$  atmospheres. Tritium diffuses at a rate dependent upon the square root of its effective pressure, and the TMHR heat exchanger is smaller and its top temperature is lower than that of MSBR. It seems likely that less than 4000 Ci per day would diffuse through the TMHR heat exchanger and contaminate the sodium coolant. Escape of 4000 Ci/day to the environment may not be tolerable. Escape of this small quantity could presumably be minimized (at some cost) by cold trapping in the sodium system or, possibly, by processing of the sodium.

If further study should show that an intolerable quantity would escape, the sodium coolant could be replaced by the molten salt coolant mixture (sodium fluoride-sodium fluoborate) proposed<sup>14-16</sup> for use in MSBR and shown experimentally<sup>50</sup> to be capable of trapping diffused tritium for recovery.

Use of such a large purge system would ensure that Kr and Xe isotopes were essentially completely removed from the blanket salt at each pass.

The contained tritium would have to be recovered from the He purge gas and the short-lived isotopes of Kr and Xe would have to be held up for decay. Modifications of the system proposed<sup>28</sup> (but not designed in detail) for MSBR could presumably accomplish these functions. In addition, the long-lived isotopes (largely  $^{89}\text{Kr}$ ) might have to be retained and bottled for disposal. Cryogenic systems or fluorocarbon processing could certainly be adapted if necessary.

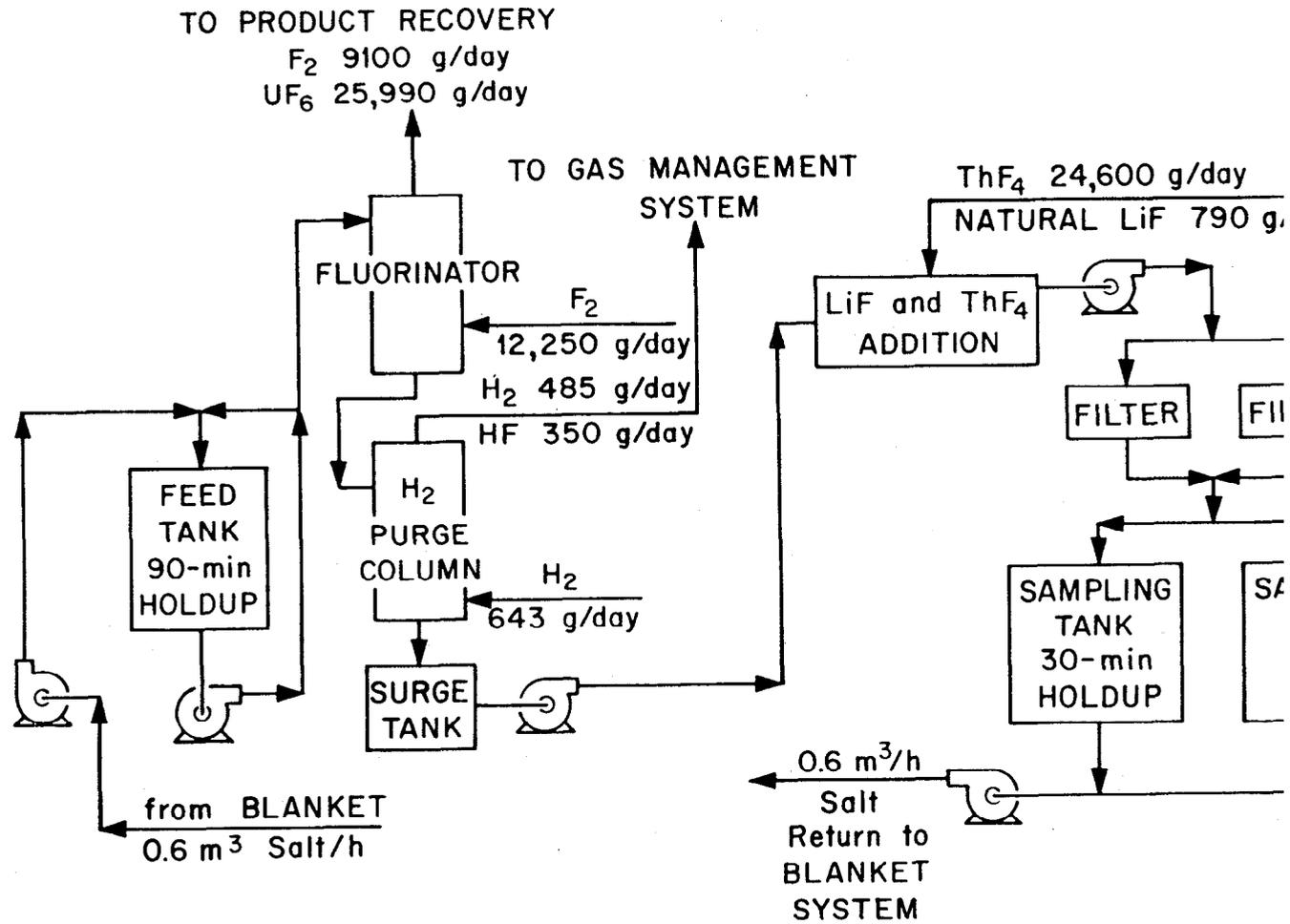
VII.B.2.d Alternative Processes for TMHR Blanket. The TMHR blanket is to be processed at a rate that will maintain the  $^{233}\text{U}$  concentration at  $1.1 \times 10^{-3}$  times the Th concentration. Such processing will lead to an inventory of 1870 kg of  $^{233}\text{U}$  within the  $1152 \text{ m}^3$  of blanket salt ( $4.846 \times 10^{-4}$  gram  $^{233}\text{U}$  per gram of salt), and will require removal of 0.202 grams per second of  $^{233}\text{U}$  or  $^{233}\text{U}$  plus  $^{233}\text{Pa}$  from the salt. Two process alternatives are presented. The simpler process, chosen as the reference process, uses fluorination alone to

recover  $^{233}\text{U}$  from the blanket salt by volatilization as  $\text{UF}_6$ . The alternative process uses a similar fluorination combined with reductive extraction to remove both  $^{233}\text{U}$  and  $^{233}\text{Pa}$  from the processed salt. Both of the processes will receive salt from the external heat exchanger system of the TMHR; such salt will have passed the helium purge system described above. Both of the processes are adapted from those proposed for MSBR fuel and use equipment similar to that described for processing that fuel.<sup>36</sup>

VII.B.2.d.(1). Processing by Fluorination Only. Fluorination readily removes uranium from molten fluoride solutions but is incapable of removal of Pa and most of the fission product species (see VII.B.2.C.(2), above). If complete removal of the contained  $^{233}\text{U}$  were attained, the required 0.202 grams  $^{233}\text{U}$  per second could be obtained by processing at a rate of 417 grams of salt per second. Continuous fluorination in equipment of reasonable size cannot, however, be expected to approach complete removal of  $^{233}\text{U}$  from solutions so dilute as these. If, as seems reasonable, 75% removal can be realized, the required processing rate is about 560 grams of salt per second, corresponding to 0.6 m<sup>3</sup> per hour or to 2.63 gallons per minute. Such a processing rate, which is about three times that proposed<sup>36</sup> for MSBR fuel, would pass the entire blanket inventory through the process each 80 days. The required processing rates are further discussed in Section VII.A.3.6.(1).

The salt received by this process will contain (in grams per cubic meter) about 0.334 of  $^{233}\text{Th}$ , 590 of  $^{233}\text{Pa}$ ,  $3 \times 10^{-5}$  of tritium, and concentrations of stable fission product fluorides (see VII.B.2.C.(1) above) that will increase during the lifetime of TMHR. Since the efficiency of the He purge system for stripping Kr and Xe isotopes is very high, only trivial quantities of these gases will be received by the process. Initial heat generation levels would be near 160 KW/m<sup>3</sup> corresponding to an adiabatic temperature rise of about 2.1°C/minute; of this heat generation about 42, 91, and 27 KW/m<sup>3</sup> would be due to  $^{233}\text{Pa}$ ,  $^{233}\text{Th}$ , and fission products, respectively.

As shown in Figure VII.B-4, the salt is received in a recirculating feed tank capable of effective holdup of the salt for 90 minutes to drop the heat load on the salt to less than 60 KW/m<sup>3</sup>. The salt is then pumped to the primary fluorinator where it descends countercurrent to a stream of fluorine added at a rate of 5 liters (stp) per minute. The salt leaving the fluorinator passes directly to the H<sub>2</sub> purge column where it descends countercurrent to a stream of



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Figure VII.B-4. Salt Flow System for Reference (Fluorination Only) Process.  
 (Reagent usage in grams per full-power day.)

H<sub>2</sub> fed at 5 liters (stp) per minute. Treatment of the gases from the fluorinator and from the H<sub>2</sub> purge column is briefly described below.

Salt from the purge column passes to equipment where solid reagents are added for adjustment of the salt composition. Maintenance of the desired UF<sub>3</sub> concentration (see VII.B.2.C.(3), above) requires addition of 45 grams of Be each full power day. This is equivalent to generation of 235 grams of BeF<sub>2</sub> per day and compensation for this (if necessary) would require addition of 585 grams of LiF and 1.16 kg of ThF<sub>4</sub> each full power day. In addition, transmutation of Li isotopes requires addition of about 200 grams of LiF, and transmutation and fission of <sup>232</sup>Th requires addition of about 23.5 kg of ThF<sub>4</sub> per full power day. Addition of 4.1 kg of ThF<sub>4</sub> at four hour intervals and of about 393 grams of LiF each 12 hours would certainly keep the composition in balance. The additional salt volume produced through compensation of added Be would total 4.4 m<sup>3</sup> in 30 years of operation at 70% plant factor; this quantity should be readily accommodated within the TMHR blanket drain tanks. The LiF additions might be prepared from Li enriched in <sup>6</sup>Li if the isotopic composition within the blanket is to be kept at 99.8% <sup>7</sup>LiF. It seems likely, however, that addition of LiF prepared from normal Li will suffice.

The salt leaving the composition adjustment equipment is filtered and fed to a tank where it is held for 30 minutes for sampling before its return to the TMHR blanket circuit.

All of the process vessels and piping described above are constructed of Hastelloy N, but the fluorinator and the H<sub>2</sub> purge column require protection of all wetted surfaces by a film of frozen salt.<sup>36</sup> Heat removal from each of the assemblies is accomplished (as proposed for MSBR) by circulating liquid NaK. The feed tank heat removal system is of tube and shell construction, while the fluorinator and the purge column are cooled by NaK circulating between the vessel and a complete jacket of Hastelloy N.

Management of the gaseous streams, containing the product <sup>233</sup>UF<sub>6</sub> and fission products volatilized by fluorination, will be accomplished by modifications of the processes used to recover uranium from spent MSRE fuel<sup>37</sup> and of that proposed<sup>36</sup> for MSBR. As shown in Figure VII.B-5, the gas stream from the fluorinator (containing about 9100 g F<sub>2</sub> and 25,990 grams <sup>233</sup>UF<sub>6</sub> per full power day) would pass NaF traps at 425°C for retention of any volatile ruthenium, niobium and antimony fluorides and then through a series of five NaF traps maintained at 100°C for complete sorption of the product <sup>233</sup>UF<sub>6</sub>.

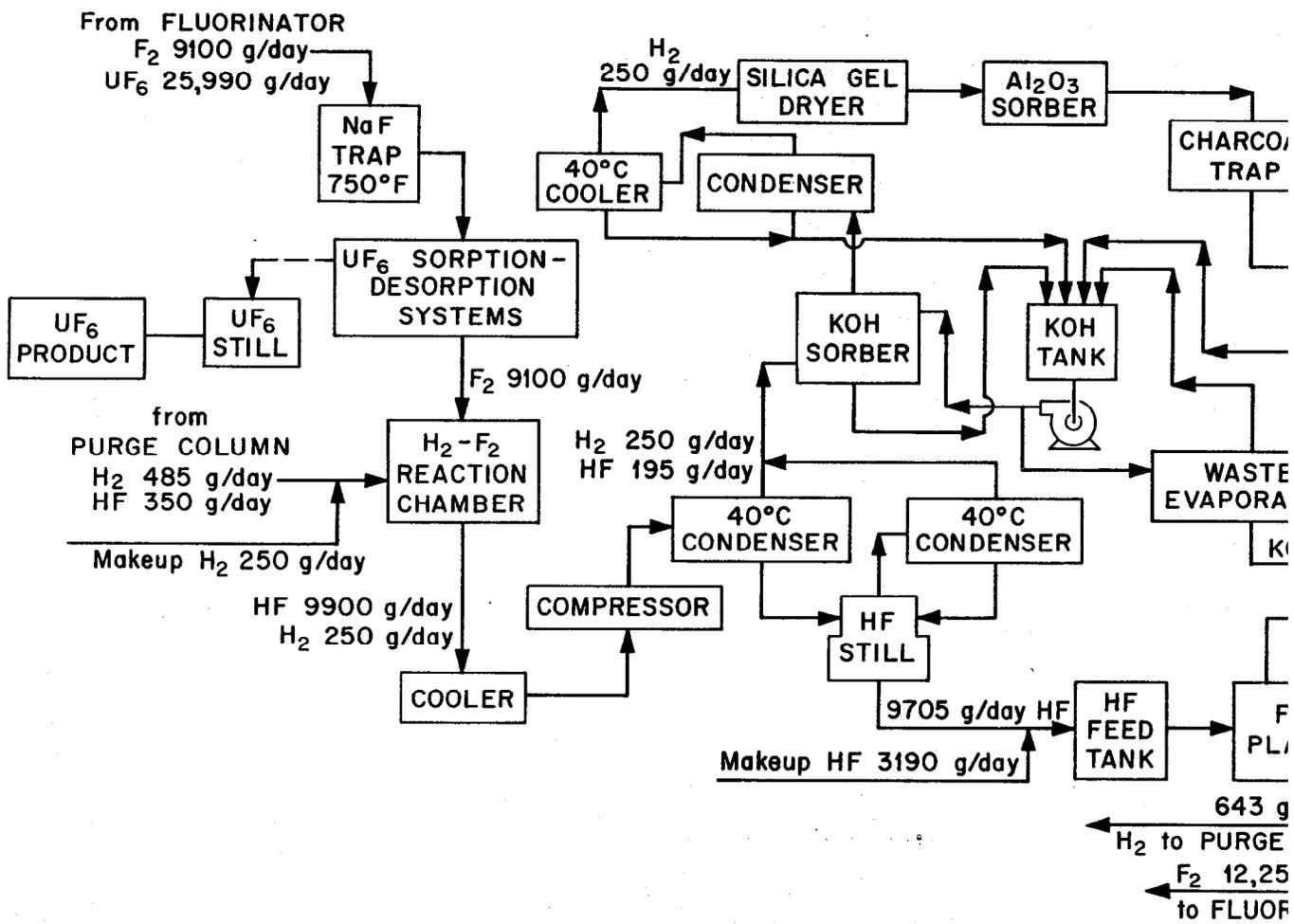


Figure VII.B-5. Gas and Product System for Reference (Fluorination Only) Process.  
 (Reagent usage in grams per full-power day.)

The F<sub>2</sub>, along with fission product fluorides such as IF<sub>5</sub>, TeF<sub>6</sub> and most of the MoF<sub>6</sub>, pass through these traps. The gas from the H<sub>2</sub> purge system (containing about 485 grams H<sub>2</sub> and 350 grams HF per full power day) plus an additional 250 grams of H<sub>2</sub> per day is reacted with the F<sub>2</sub> stream to produce a gas containing about 9900 g HF and 250 grams H<sub>2</sub>/day. This gas is chilled, compressed to 2 atmospheres pressure, and sent to a HF distillation column. The still bottoms (9705 gram HF/day) are sent along with 3190 grams of makeup HF to produce the 12,250 grams per day of F<sub>2</sub> and the 643 grams per day of H<sub>2</sub> required as feed for the fluorinator and H<sub>2</sub> purge system. The HF distillation overheads (250 g H<sub>2</sub> and 195 grams HF per day) are neutralized with 10 molar KOH solution and the H<sub>2</sub> (about 0.07 scfm) is vented and burned after passage through traps of silica gel, Al<sub>2</sub>O<sub>3</sub>, and activated charcoal. Such venting would release about 6 Ci of tritium and trivial quantities of Kr and Xe per full power day.

The product <sup>233</sup>UF<sub>6</sub> is desorbed from the 100°C NaF traps by heating. Fission product contamination (largely from traces of MoF<sub>6</sub>) seems unlikely to be such as to require additional purification, but purification by distillation could be performed if required. The desorbed UF<sub>6</sub> is transferred to product cylinders for shipment to fuel fabrication plants.

Such a process would produce small quantities of waste contaminated with fission products. Evaporation of the 10 M KOH neutralizer solution would produce about 0.5 m<sup>3</sup> of solid waste per year. The condensate from the evaporation would be used to prepare the 10 M KOH solution for the neutralizer. Additional solid wastes would result from occasional replacement of the 425°C NaF trap, of the NaHF<sub>2</sub> electrolyte from the small F<sub>2</sub> production cell, and of the silica gel and Al<sub>2</sub>O<sub>3</sub> sorbers. These are estimated to total about 5 m<sup>3</sup> of contaminated solid per year.

VII.B.2.d.(2). Processing by Fluorination and Reductive Extraction. Reductive extraction with alloys of Li and Th in Bi is capable (see VII.B.2.C.(2), above) of removing Pa and Zr (and, with more difficulty) other fission products along with U from molten fluorides. With essentially complete removal of <sup>233</sup>U and <sup>233</sup>Pa from the salt, processing at a rate of 325 grams of salt per second (0.349 m<sup>3</sup>/hr or 1.535 gallons per minute) would be required to maintain the <sup>233</sup>U at the desired concentration of 4.846 x 10<sup>-4</sup> grams per gram of salt. Such a processing rate (about 1.75 times that proposed for MSBR) would pass the TMHR blanket salt through the plant each 138 days. The <sup>233</sup>U inventory within the

blanket system would again be 1870 kg while the maximum  $^{233}\text{Pa}$  inventory would be 530.1 kg if the TMHR plant operates for several months at full power. The heat generation rate in the fuel received by the plant would be near  $150 \text{ kW/m}^3$ , and as for the fluorination only plant, trivial quantities of tritium (3.2 Ci/day) and Kr and Xe isotopes would be present in the salt for processing.

The salt would be received in a recirculating feed tank and would be sent to a primary fluorinator and a  $\text{H}_2$  purge column very similar to, but somewhat smaller than, those described for the fluorination only process above. Fluorine feed of 3 liters (stp) per minute is again assumed to remove 75% of the uranium, and a similar  $\text{H}_2$  feed rate should suffice for the purge. Blanket salt from the purge column, containing (in grams/ $\text{m}^3$ ) 405.8 of  $^{233}\text{U}$  and 466.2 of  $^{233}\text{Pa}$  passes a small surge tank and is pumped to the bottom of the primary extractor where it passes countercurrent to an alloy of Li and Th in Bi fed at  $0.0794 \text{ m}^3$  per hour. This extractor is scaled from that proposed for MSBR,<sup>36</sup> but we have assumed only 3 (instead of 5) equilibrium stages capable of extracting 99.5% of the  $^{233}\text{Pa}$  along with essentially all of the  $^{233}\text{U}$ . Estimated concentrations of the entering and exit streams are shown in Table VII.B-9.

The TMHR blanket salt from the extractor is passed through a large bed packed with Ni wool for removal of traces of Bi and through a filter system for removal of possible particulate materials. Finally the salt passes, as in the fluorination process above, equipment for adjustment of its composition and through a 30-minute holdup tank for sampling prior to its return to the TMHR blanket system.

The quantities of  $\text{LiF}$  and  $\text{ThF}_4$  that must be added per full power day to compensate for transmutation, for fission, and for the necessary Be addition are, of course, the same as those described for the fluorination process above. However, the reductive extraction process adds 63 grams of lithium and 7130 grams of Th per day to the blanket. As a consequence, addition of 275 grams of  $\text{LiF}$  each 12 hours and 2.33 kg of  $\text{ThF}_4$  each 4 hours should keep the blanket at the desired composition. Again, it would seem reasonable to add the  $\text{LiF}$  (as well as the  $\text{Li}^0$  in the Bi) as normal lithium.

TABLE VII.B-9. QUANTITIES OF PERTINENT SPECIES IN ENTERING AND EXIT STREAMS OF PRIMARY EXTRACTOR (MOLES PER HOUR)

| Specie            | Salt streams |        | Bi streams |       |
|-------------------|--------------|--------|------------|-------|
|                   | Entering     | Exit   | Entering   | Exit  |
| Salt              | 18,500       | 18,500 | 3700       | 3700  |
| <sup>233</sup> U  | 0.608        | 0      | 0          | 0.608 |
| <sup>233</sup> Pa | 0.689        | 0.003  | 0          | 0.686 |
| Zr                | 0.045        | 0      | 0          | 0.045 |
| Sr and Ba*        | 2.13         | 2.13   | 0          | 0.005 |
| Lanthanides*      | 10           | 9.96   | 0          | 0.04  |
| Li <sup>o</sup>   |              | 0.38†  | 4.90       | 4.51  |
| Th <sup>o</sup>   |              | 1.28†  | 5.79       | 4.48  |

\*Estimated equilibrium values obtained only after many years operation.  
 †Moles/hr of Li and Th transferred to the salt (as LiF and ThF<sub>4</sub>).

The Bi alloy from the primary extractor, containing (see Table VII.B-9) the extracted <sup>233</sup>U and <sup>233</sup>Pa, passes a surge tank and is pumped at 600°C through an assembly similar to that proposed for MSBR<sup>36</sup> in which it passes countercurrent to captive salt and to a stream of anhydrous HF fed (2-fold excess) at 21 liters (stp) per minute. The captive salt, in a circuit isolated from the TMHR blanket, has a composition (in mole %) near 71 LiF-29 ThF<sub>4</sub>, is prepared from LiF containing natural Li, and flows at 0.25 m<sup>3</sup> per hr through the hydrofluorinator. In this assembly, all of the active metals (including U, Pa, Zr, Li and Th) are stripped completely into the captive salt. The essentially pure Bi is pumped to a recirculating pump-tank system where the Bi is brought to the required concentration (73 mole per m<sup>3</sup> of Th and 61.7 moles per m<sup>3</sup>) of Li for return to the primary extractor.

The captive salt receives the active metals from the Bi alloy in the hydrofluorinator and flows in a separate circuit to permit decay of <sup>233</sup>Pa and recovery of the <sup>233</sup>U produced. This salt passes from the hydrofluorinator through a secondary fluorinator, fed with F<sub>2</sub> at 2 (stp) liters per minute, where 75% of the uranium is assumed to be removed and through a purge column where it flows countercurrent to 2 (stp) liters of H<sub>2</sub> per minute. Captive salt from the purge column enters the <sup>233</sup>Pa decay tank from which it is pumped to the hydrofluorinator.

The captive salt increases in volume from 10 m<sup>3</sup> to 11.9 m<sup>3</sup> over a 220-day cycle due largely to accumulation of LiF and ThF<sub>4</sub> from the hydrofluorinator and to addition of 4 kg of normal LiF per day to maintain a usefully low melting salt composition. The inventories of <sup>233</sup>U and <sup>233</sup>Pa near the end of each 220-day cycle are about 12.4 kg and 149.4 kg, respectively. On the average, about 301 grams of <sup>233</sup>U per hour are recovered from the secondary fluorinator. At the end of each cycle, 1.9 m<sup>3</sup> of the captive salt are transferred to a secondary (static) decay system where the <sup>233</sup>Pa is allowed to decay for an additional 220 days. This 1.9 m<sup>3</sup> of salt initially contains about 1.98 kg of <sup>233</sup>U and 23.86 kg of <sup>233</sup>Pa. After 220 days all except 84 grams of the <sup>233</sup>Pa have decayed and the 25.76 kg of <sup>233</sup>U are recovered by fluorination in a batch fluorinator and the salt containing the residual <sup>233</sup>Pa and about 10 grams of <sup>233</sup>U is transferred to waste storage. Since the salt is to be stored on site for some years, a very large fraction (90%) of the discarded 95 grams of <sup>233</sup>U could be recovered by batch fluorination if desired.

All of the equipment and piping intended to handle only salt is, as for MSBR,<sup>36</sup> to be fabricated of Hastelloy N, but as described previously, the fluorinators and the H<sub>2</sub>-purge columns require protection by a film of frozen salt. Equipment intended to handle Bi alloys only (such as the Li and Th makeup tanks) may be constructed of tantalum alloys jacketed with Hastelloy or steel. Equipment such as the primary extractor and hydrofluorinator that must handle both Bi alloys and salts must be of special construction. For this study it is assumed that suitably protective coatings of Mo or W can be emplaced on steel or Hastelloy assemblies. The large (15 m<sup>3</sup>) <sup>233</sup>Pa decay tank is assumed to be of Hastelloy N. It will require equipment for addition of about 4 kg per day of normal LiF. The heat load on this decay tank will be near 10.50 MW<sub>t</sub> from <sup>233</sup>Pa decay and perhaps an additional 5 MW<sub>t</sub> from fission products. It is, accordingly, assumed to be passively cooled by a system in which NaK is moved by thermal convection to an air-cooled radiator in a simple natural convection stack; a generally similar system was proposed for cooling the MSBR drain tank.<sup>16,29</sup> All of the other systems are to be cooled by forced circulation of NaK through tube and shell heat exchangers or vessel jackets.

Management of the gas streams, containing the product <sup>233</sup>UF<sub>6</sub> and volatile fission products, would be accomplished by processes identical to those of the reference fluorination plant described above. The necessary incorporation of the

hydrofluorinator in the reductive extraction process slightly increases the size of some of the equipment. Material balances for the gaseous  $F_2$ ,  $H_2$ , and HF indicate that 465 grams of HF must be neutralized by KOH, 825 grams of  $H_2$  must be vented, and 13,150 grams of makeup HF must be purchased for each full power day of plant operation. Venting of the  $H_2$  at about 6.4 liters (0.23 scf) per minute would result in release of about 4 Ci of tritium and trivial releases of Kr and Xe isotopes per full-power day.

This process, like the reference fluorination only process above, would produce small quantities of solid waste contaminated with fission products. Evaporation of the neutralizer solution would yield about  $1.2 \text{ m}^3$  of waste per year and replacement of traps and adsorbers would add about  $5 \text{ m}^3$  per year. In addition, the discard ( $1.9 \text{ m}^3$  per 220 days) from the captive salt circuit would add about  $3.2 \text{ m}^3$  per year of solid waste for eventual discard to a repository.

#### VII.B.2.e. Estimates of Capital and Operating Costs of Alternative Processes.

Estimation of cost for the MSBR<sup>16,29</sup> and for its complicated processing plant<sup>36</sup> were performed many years ago, were presented in 1969 dollars, and were based upon conceptual rather than detailed and final designs. Several features of the salt maintenance systems and of the processing plants for TMHR differ substantially from those proposed for MSBR. For purposes of this study, the pertinent estimates made for the MSBR systems were accepted; they were updated to 1981 dollars by assuming 6% inflation for five years and 10% inflation for seven years. In addition, they were scaled to the TMHR capacity by assuming that equipment costs scale as the 0.5 power of the throughput. The estimates presented here in 1981 dollars, represent only the direct cost of the system, and include no contingency.

VII.B.2.e.(1) Costs for Salt Maintenance Functions. The operations required for maintenance of the required  $UF_3/UF_4$  ratio in the blanket salt and for removal and recovery of  $^3H$ , Kr, and Xe must be performed by systems associated with the primary heat exchanger system and outside the processing plant.

Maintenance of the required  $UF_3/UF_4$  ratio (VII.B.2.C.(3), above) is most important but is easily accomplished by periodic addition of metallic Be. For this purpose, one of the TMHR blanket salt pumps would require addition of an assembly to permit lowering of a small basket containing Be into the circulating salt in the pump bowl. This relatively small addition to the pump is assumed to

involve a direct capital cost of \$225,000; its operation, requiring the part time service of one operator, is assumed to cost \$25,000 per year. The (trivial) cost of the added Be is included in the reagent cost for the processing plants.

Removal and retention of  $^3\text{H}$ , Kr, and Xe (VII.B.2.C.(3), above) is a much more difficult operation. The similar operations for MSBR<sup>16,28,29</sup> were performed on six-fold smaller salt flow and involved more Kr and Xe but far less  $^3\text{H}$ . Moreover, the MSBR system, though it proposed<sup>14</sup> to retain the  $^{89}\text{Kr}$  cryogenically, did not recover the tritium separately. The MSBR system,<sup>14</sup> after adjustment for inflation and twelve-fold scaleup to TMHR size, would entail a direct capital cost of  $7 \times 10^6$ . It is not, at present, clear how this system would be modified to permit recovery of a large fraction of the tritium. Such a system, capable of recovery of a large fraction of the 20+ grams of  $^3\text{H}$  per day and of bottling the noble gases for disposal, might entail a direct capital cost of  $18 \times 10^6$  and an annual operating cost of \$200,000.

VII.B.2.e.(2). Costs for the Reference Fluorination Only Process. The process for recovery of  $^{233}\text{U}$  from the TMHB blanket by fluorination alone is to operate upon  $0.6 \text{ m}^3$  of salt per hour (VII.B.2.d.(1), above). This reference process is considerably simpler than the MSBR process, though the proposed throughput of salt is three-fold higher, and all equipment in the blanket salt circuits can be of Hastelloy N. The TMHR process is required to handle less  $^{233}\text{UF}_6$  than did that for MSBR, but it must purify a much larger quantity of  $\text{UF}_6$  as product. Direct capital costs (no contingency) have been estimated (after detailed listing of the equipment required) by scaling, wherever possible, from the costs estimated by Nicholson and Carter.<sup>36</sup> Costs for auxiliary equipment, piping, thermal insulation, process instrumentation, etc. were then estimated by the methods used for the MSBR process.<sup>36</sup> Results of this estimate of the direct capital cost of the reference processing system are shown in Table VII.B-10. To the total direct capital cost (with no contingency) of \$17,025,000 must be added the direct capital costs (\$18,225,000) for the salt maintenance functions described in VII.B.2.e.(1) above. On this basis the direct capital costs associated with maintenance of the  $\text{UF}_3/\text{UF}_4$  ratio, removal and recovery of  $^3\text{H}$ , Kr and Xe, and the total direct cost of the reference process would be about \$35,250,000.

At the end of its life, the TMHR will contain about 1160 m<sup>3</sup> of blanket salt contaminated with fission products. It is possible that this still valuable salt can be transferred, at that time, to another TMHR at the same site. However, at some time this large volume of salt must be discarded, presumably as high-level waste. About 8,850 cannisters (10 inches in diameter, 10 feet in length, filled to a depth of 8.5 feet) would be required for disposal if the solidified salt can be considered an acceptable waste form. Such disposal (at \$9000 per cannister) plus transportation would be expected to cost about \$85 x 10<sup>6</sup>. It should be noted, however, that a THOREX plant processing 1500 MgHM per year at 3 ft<sup>3</sup> of high-level waste per MgHM would produce more than three times as much waste for disposal.

VII.B.2.e.(3). Costs for the Fluorination and Reductive Extraction Process.

The process using fluorination and reductive extraction (VII.B.2.d.(2)) is to operate upon 0.349 m<sup>3</sup> of blanket salt per hour. This rate is 1.75 times that proposed for MSBR,<sup>36</sup> but the process is simpler since no provisions are made for removal of alkaline earth and rare earth fission products. However, this alternative process is required, as was the reference process, to purify a much larger quantity of <sup>233</sup>UF<sub>6</sub> product than did the MSBR process. In addition, this alternative process will require special materials or special coatings to protect the equipment that must handle both Bi alloys and molten salts.

As before, the direct capital cost estimates have used the methods of, and have been scaled from the results of, Carter and Nicholson.<sup>36</sup> While it seems likely that ultra-modern techniques can apply suitably resistant coatings of (for example) molybdenum to base metals, we have assumed that costs of such equipment (in 1969 dollars) would be those estimated<sup>36</sup> for fabrication from molybdenum. Results of this estimate of direct capital cost (with no contingency) are shown in Table VII.B-12. As for the reference process, the direct capital costs (\$18,225,000) for the salt maintenance functions must be added to the total from the table. The combined direct costs of the processing plant plus the maintenance operations is, therefore, \$74,703,000.

Table VII.B-10. DIRECT CAPITAL COST OF REFERENCE PROCESSING PLANT FOR FLUORINATION OF 0.6 m<sup>3</sup> OF SALT PER HOUR

| Item                              | Cost (\$1000) |
|-----------------------------------|---------------|
| Hastelloy and stainless equipment | 5,648         |
| Auxiliary equipment               | 2,441         |
| Hastelloy and stainless piping    | 2,591         |
| Thermal insulation                | 582           |
| Fluorine plant                    | 395           |
| Process instrumentation           | 2,397         |
| Cell electric connections         | 255           |
| Sampling stations                 | 1,148         |
| Radiation monitoring              | 318           |
| Waste handling and storage        | 1,250         |
| Total direct cost                 | 17,025        |

Table VII.B-11 shows the estimated annual operating cost of the reference processing plant. Reagents used in the blanket salt maintenance functions previously described are included in this total, but other operating costs are not. As a consequence, to the total of \$1,386,500 shown in the table must be added some \$225,000 for the UF<sub>3</sub>/UF<sub>4</sub> ratio maintenance and the <sup>3</sup>H, Kr and Xe removal functions. Operating costs, assuming 70% plant factor, accordingly, total \$1,611,500 per year.

TABLE VII.B-11. ESTIMATED ANNUAL OPERATING COSTS FOR REFERENCE PROCESSING PLANT FOR FLUORINATION OF 0.6 m<sup>3</sup> OF SALT PER HOUR

| Item                               | Cost (\$ per year) |
|------------------------------------|--------------------|
| Reagent use*                       |                    |
| ThF <sub>4</sub> (6.3 metric tons) | 205,500            |
| Be (11.5 kg)                       | 2,000              |
| LiF (normal; 201 kg)               | 8,800              |
| H <sub>2</sub> (64 kg)             | 1,250              |
| HF (816 kg)                        | 3,000              |
| KOH (200 kg)                       | 1,000              |
| Total                              | 221,550            |
| Plant payroll†                     | 700,000            |
| Waste disposal*                    | 325,000            |
| Total operating cost               | 1,386,550          |

\*Calculated for 70% plant factor.

†Does not include blanket salt maintenance operations.

TABLE VII.B-12. DIRECT CAPITAL COST OF PLANT FOR PROCESSING BY  
 FLUORINATION AND REDUCTIVE EXTRACTION (0.349 m<sup>3</sup> SALT PER HOUR)

| Item                              | Cost (\$1000) |
|-----------------------------------|---------------|
| Hastelloy and stainless equipment | 15,486        |
| "Molybdenum" equipment            | 5,901         |
| Auxilliary equipment              | 4,860         |
| Hastelloy stainless piping        | 9,527         |
| "Molybdenum" piping               | 1,770         |
| Thermal insulation                | 1,397         |
| Fluorine plant                    | 410           |
| Process instrumentation           | 8,962         |
| Cell electric connections         | 750           |
| Sampling stations                 | 2,870         |
| Radiation monitoring              | 795           |
| Waste handling and storage        | 3,750         |
| Total direct cost                 | 56,478        |

Table VII.B-13 shows the estimated operating costs, assuming a 70% plant factor, for the alternative processing plant. Reagents used in the salt maintenance functions are included, but other costs of operating the maintenance systems are not. Some \$225,000 must be added for those systems leading to a total of \$2,279,000 as the operating cost of the processing and maintenance functions.

As for the reference process, the blanket salt inventory (about 1160 m<sup>3</sup>) of the TMHR at its end of life must - possibly after re-use in subsequent TMHR's - be disposed of as high-level waste. Costs for this disposal would be very similar to those presented above for the reference process.

TABLE VII.B-13. ESTIMATED ANNUAL OPERATING COSTS FOR ALTERNATIVE PROCESSING PLANT (0.349 m<sup>3</sup> SALT PER HOUR)

| Item                                 | Cost (\$ per year) |
|--------------------------------------|--------------------|
| Reagent use*                         |                    |
| Li (natural, 16.1 kg)                | 2,700              |
| <sup>232</sup> Th (1.82 metric tons) | 99,000             |
| Be (11.5 kg)                         | 2,000              |
| LiF (1.2 metric tons)                | 52,500             |
| ThF <sub>4</sub> (3.0 metric tons)   | 128,000            |
| HF (3.36 metric tons)                | 12,400             |
| KOH (480 kg)                         | 2,400              |
| Total                                | 299,050            |
| Plant payroll†                       | 1,200,000          |
| Waste disposal*                      | 555,000            |
| Total operating cost                 | 2,054,000          |

\*Calculated for 70% plant factor.

†Does not include salt maintenance operations.

VII.B.2.f. Special Development Needs in Molten Salt Technology. The staff of the MSBR program at the Oak Ridge National Laboratory prepared in 1974 a detailed program plan for development of Molten Salt Breeder Reactors.<sup>15</sup> That document described the gaps and uncertainties in the data base, as did a previous study,<sup>14</sup> presented in detail an 11-year program for the required development effort, and estimated the costs of each individual research and development item. Costs of the required MSBR technology effort, in 1975 dollars, were presented for each of the 11 years. The operating fund requirements (excluding reactor design and analysis) averaged about \$6.6 x 10<sup>6</sup> per year for the first five years and about \$13.9 x 10<sup>6</sup> per year for the final six years. Capital equipment funds to support that effort averaged \$1.6 x 10<sup>6</sup> per year over that period. In addition, a total of 104 x 10<sup>6</sup> of capital project funds (not including a test reactor) were identified as necessary during that period. It was not presumed that MSBR technology development would be completed at the end of the 11-year period, but it was believed that such expenditures would permit a MSBR test reactor to be authorized at the end of the fifth year and to be adequately supported during its design and construction phases.

The MSBR program included some items (such as graphite development) that are not germane to the present TMHR. [It should be noted, however, that subsequent TMHRs may include graphite.] However, most of the general classes of needed MSBR development items will be required for the TMHR. A brief description of key items that concern the molten salt portion of TMHR is presented and estimates of the operating and capital equipment costs (scaled from the MSBR estimates<sup>15</sup>) are tabulated in the following.

- Hastelloy-N and its modifications seem likely to prove adequate, but these alloys are not yet approved reactor materials, and expensive development is needed. Some of the MSBR program concerned interaction with the sodium fluoborate coolant (which might be adopted by TMHR), but, in any event, testing of Hastelloy-N with the chosen coolant will be required.
- Basic chemistry of molten  $\text{LiF-BeF}_2\text{-ThF}_4$  mixtures is relatively well known, but better physical property measurements, better understanding of  $^3\text{H}_2$  solubility and behavior, and, especially, better understanding of fission product behavior is needed.
- A few on-line instruments for chemical analysis are well along in development, but several others are needed both for the TMHR blanket and for its processing plant.
- The development requirements in reactor safety (hot-spot analysis, consequences of flow blockage in fuel channels, leaks in heat exchangers, and consequences of primary system leakage may be less serious for TMHR, but they are not trivial.
- Graphite development, including development of less permeable graphites with increased radiation stability, will be required for TMHR if graphite is to be used in contact with molten salts.
- Molten salt reactor technology needs for TMHR, as for MSBR, include development of improved pump and heat exchanger designs, and of improved flow, level, and other monitoring equipment. TMHR may not require development of salt-steam heat exchangers but it must develop and demonstrate stripping, recovery, and control of large quantities of tritium.

- Development of improved instruments and control systems is probably of less consequence to TMHR than to MSBR, but, again, the problems are not trivial.
- If the reference TMHR process (fluorination only) proves satisfactory, a smaller effort in processing technology than that required for MSBR will suffice. Key items for the reference process include the degree of protection afforded the fluorinator by frozen salt films and the efficiency of continuous fluorination in removal of  $^{233}\text{U}$  from very dilute molten salt solutions. An especial need is for demonstration of, and improvement on, the system for recovery, purification, and handling of large quantities of  $^{233}\text{UF}_6$  and for management of the appreciable quantities of fission products volatilized in the fluorinator. If the alternative process is to be preferred, a program of the magnitude of that proposed for MSBR is necessary. In any event, a relatively expensive demonstration facility for the integrated process will be required.
- The program of materials development for the processing plant depends markedly upon whether the reference process or some variant of the alternative process is chosen. If it is the latter, a difficult, and necessarily rapid, development program is necessary to provide and demonstrate compatibility of a metal or coating with combined Bi alloys and salts.

Table VII.B-13 presents an estimate of the total operating fund and capital equipment fund requirements (in 1981 dollars) required over a ten-year period to provide adequate support to a molten-salt blanketed TMHR. As the table shows, a development effort averaging about  $\$15 \times 10^6$  per year and with capital equipment funds of about  $\$2.7 \times 10^6$  per year would be required for a ten-year period to provide support for a test of a TMHR using a molten salt blanket. [It should be noted that the capital equipment requirement is likely to be much higher than above, since the MSBR estimates<sup>15</sup> presumed, as is no longer the case, that ORNL's 1974 pool of test stands and equipment were available.] Research and development on molten salt technology for TMHR would certainly be required beyond this ten-year period, as detailed problem areas indicated by the test design and subsequent designs are identified. Indeed it

seems certain that expenditures beyond this ten-year period would increase substantially (in constant dollars). A recent examination of MSBRs<sup>21</sup> suggests that with development beginning in 1980, a molten salt fission reactor could be made commercial in 30 years and that the research and development costs (including several expensive facilities but not the intermediate scale reactors) would be near  $\$700 \times 10^6$ . It is not apparent that a molten salt version of a TMHR could be built in a shorter period or with development costs markedly less than could a molten salt fission reactor.

TABLE VII.B-13. ESTIMATED OPERATING FUND AND CAPITAL EQUIPMENT FUND REQUIREMENTS FOR DEVELOPMENT OF MOLTEN SALT BLANKETS FOR TMHR (1981 DOLLARS)

|                                     | Operating Fund Requirements<br>( $10^6$ dollars) | Capital Equipment Requirement<br>( $10^6$ dollars) |
|-------------------------------------|--|--|
| Development of Structural Materials | 24.6   | 6.7  |
| Chemical Research and Development   | 16.9   | 3.2  |
| Analytical Chemistry Development    | 8.0  | 1.7  |
| Reactor Safety                      | 9.6  | -  |
| Graphite Development                | 16.2*  | 4.2*   |
| Reactor Technology                  | 44.6   | 6.1  |
| Maintenance                         | 4.4  | 1.1  |
| Instrumentation and Controls        | 2.9  | 0.3  |
| Blanket Processing                  | 20.1†  | 2.8†   |
| Processing Material Development     | 1.2**  | 0.5**  |
| Total                               | 148.5  | 26.6   |

\*If graphite is to be used in contact with molten salts in TMHR.

†If the alternative process is chosen, these numbers become 31.6 and 4.8, respectively.

\*\*If the alternative process is chosen, these numbers become 4.0 and 1.7, respectively.

## VII.C ISOTOPIC CONSIDERATIONS

### VII.C.1 Overview

The generation and depletion of actinide and fission product isotopes in suppressed fission blankets is an important concern which impacts the overall economics of three areas:

- The reprocessing of discharge fuel from the TMHR
- The recovery and re-use in the TMHR of fertile thorium discharged from the blanket
- Shielding requirements for fabrication of the bred  $^{233}\text{U}$  for use in an LWR

Concerning the first of these, minimizing the fission product and actinide generation also minimizes the hazard of high level wastes. The second and third areas are most applicable to thorium metal and thorium oxide fuels and are impacted by the generation of  $^{232}\text{U}$  ( $T_{1/2} = 72 \text{ y}$ ) which decays to  $^{228}\text{Th}$  ( $T_{1/2} = 1.9 \text{ y}$ ).  $^{228}\text{Th}$  ultimately decays to  $^{208}\text{Tl}$  with the resultant production of a 2.6 MeV gamma for 36% of all  $^{228}\text{Th}$  decays.

Although fully remote fuel reprocessing operations are required, savings in shielding thicknesses for various other processes, including thorium and thorium oxide fuel fabrication, may be possible by minimizing the generation of  $^{232}\text{U}$ ,  $^{228}\text{Th}$ , and the resulting 2.6 MeV gamma activity. For example, the inventory of thorium required for the lifetime of the hybrid will be determined by how long it will be stored to allow the  $^{228}\text{Th}$  content, produced by the decay of  $^{232}\text{U}$  prior to the separation of uranium at thorium in reprocessing, to decay to acceptable levels for economically reforming it into hybrid fuel. If hands-on processing of the thorium after a 10-15 year cooling period is more economical than immediate remote processing for re-introduction into the hybrid, a 10 to 15 year supply of thorium and storage facilities will be required.

Isotopic generation and depletion during and following irradiation in the TMHR has been estimated from an ORIGIN-like calculation<sup>51</sup> which follows both the fission rate and the production of  $^{232}\text{U}$  and other isotopes as a function of time. The computer program, ISOGEN, developed for this application,

uses the same matrix exponential method as ORIGIN for the production and loss of the isotopes of interest. Figure VII.C-1 shows the isotopes of interest and their production and loss mechanisms.  $^{233}\text{U}$  is produced when a neutron is captured in  $^{232}\text{Th}$  and there are two successive beta decays:  $^{233}\text{Th}$  to  $^{233}\text{Pa}$ , and then to  $^{233}\text{U}$ .  $^{232}\text{U}$  can be produced from (n, 2n) reactions in the bred  $^{233}\text{U}$ . A second important path to  $^{232}\text{U}$  is as follows: A  $^{232}\text{Th}$  (n, 2n) reaction produces  $^{231}\text{Th}$ ,  $^{231}\text{Th}$  decays to  $^{231}\text{Pa}$ ,  $^{231}\text{Pa}$  captures a neutron becoming  $^{232}\text{Pa}$ ,  $^{232}\text{Pa}$  then decays to  $^{232}\text{U}$ . Both production mechanisms require two neutron induced reactions and both have high neutron energy thresholds, so moderating the neutron spectrum before entering the fertile fuel reduces the  $^{232}\text{U}$  production as well as the fission rate.

ISOGEN computes the time-dependent concentrations of  $^{232}\text{Th}$ ,  $^{233}\text{U}$ ,  $^{232}\text{U}$ ,  $^{231}\text{Pa}$ ,  $^{233}\text{Pa}$ ,  $^{228}\text{Th}$ , and fission products. Other intermediate isotopes (e.g.,  $^{231}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{232}\text{Pa}$ ) have half-lives which are short compared with the irradiation time, so they are assumed to be in equilibrium with their parents and go directly to their longer-lived daughters in the calculation. Averaged isotopic transfer probabilities are input for relevant transfer mechanisms including nuclear reactions ( $\langle\sigma\phi\rangle \text{ s}^{-1}$ ), nuclear decays ( $(\lambda)\text{ s}^{-1}$ ) and continuous chemical removal of selected elements ( $(F\eta/I)\text{ s}^{-1}$ ) by the molten salt processing loop, as applicable. The reaction probabilities are computed from the average flux and activation rates in the blanket as calculated using the ANISN and TARTNP codes for neutron transport calculations.<sup>52,53</sup> The chemical processing transfer rates for the molten salt blanket are based upon the molten salt plant operating point. (See section VII.A.3.6)

### VII.C.2 Isotopic Considerations for the Reference Beryllium/Thorium Oxide Blanket

The time-dependent concentrations of several important isotopes were computed for the reference beryllium/thorium oxide blanket. In this analysis, a 1.8 month irradiation (at 70% capacity factor) was required to build up a fissile enrichment of 0.62 (54%  $^{233}\text{Pa}$ ). A 6 month delay was then allowed before reprocessing to permit most of the  $^{233}\text{Pa}$  present when irradiation was discontinued to decay to  $^{233}\text{U}$  (final  $^{233}\text{Pa}$  concentration of 0.003%). At the time of discharge from the reprocessing plant, uranium

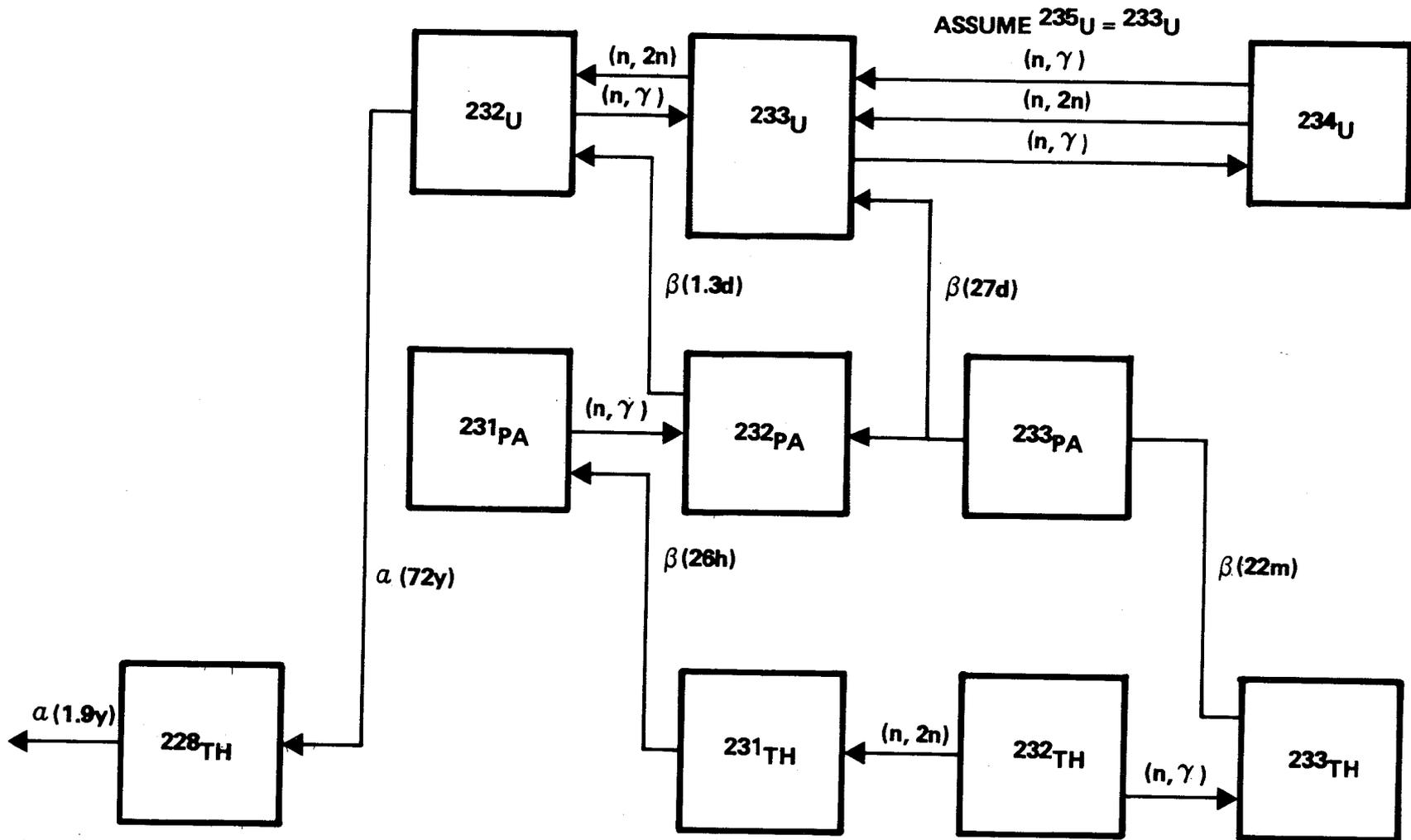


Figure VII.C-1. Production and decay chain for thorium fuel hybrid blankets.

isotopes and thorium isotopes were assumed to be partitioned from each other and the waste stream. Figure VII.C-2 shows the calculated buildup of various isotopes and fission products during the irradiation and 6 month decay period prior to reprocessing.

After reprocessing, the uranium fuel is assumed to be fabricated into LWR fuel. At the time of separation from the thorium isotopes, the uranium is relatively free of penetrating gamma activity and, therefore, relatively easily handled. As the  $^{228}\text{Th}$  buildup from the decay of  $^{232}\text{U}$ , there is a corresponding buildup of 2.6 MeV gamma activity which makes handling necessarily remote and well shielded. At the time of separation from the thorium, the uranium bred in the suppressed fission hybrid is composed of 99.98%  $^{233}\text{U}$  and 0.017%  $^{232}\text{U}$  (173 appm). Table VII.C-1 shows the buildup of  $^{228}\text{Th}$  in the uranium fuel as a function of time and the activity of the 2.6 MeV gamma in the fuel. The activity builds roughly linearly to 40  $\mu\text{Ci/g}$  of uranium in one month. An LWR fuel assembly contains about 12-14 kg of fissile fuel, so the 2.6 MeV gamma activity of a fuel assembly builds to about 0.5 Ci in the one month. Routine handling of this quantity of radioactivity requires remote techniques for shielding.

TABLE VII.C-1. Density of  $^{228}\text{Th}$  and associated 2.6 MeV gamma activity in bred  $^{233}\text{U}$  fuel.

| Weeks From Separation | Density of $^{228}\text{Th}$ (appm) | 2.6 MeV- $\gamma$ Activity ( $\mu\text{Ci/g-U}$ ) |
|-----------------------|-------------------------------------|---|
| 0.25                  | 0.0080                              | 2.31  |
| 0.5                   | 0.0161                              | 4.66  |
| 0.75                  | 0.0241                              | 6.97  |
| 1.0                   | 0.0321                              | 9.28  |
| 2.0                   | 0.0642                              | 18.6  |
| 4.0                   | 0.1284                              | 37.1  |
| 6.0                   | 0.1926                              | 55.7  |

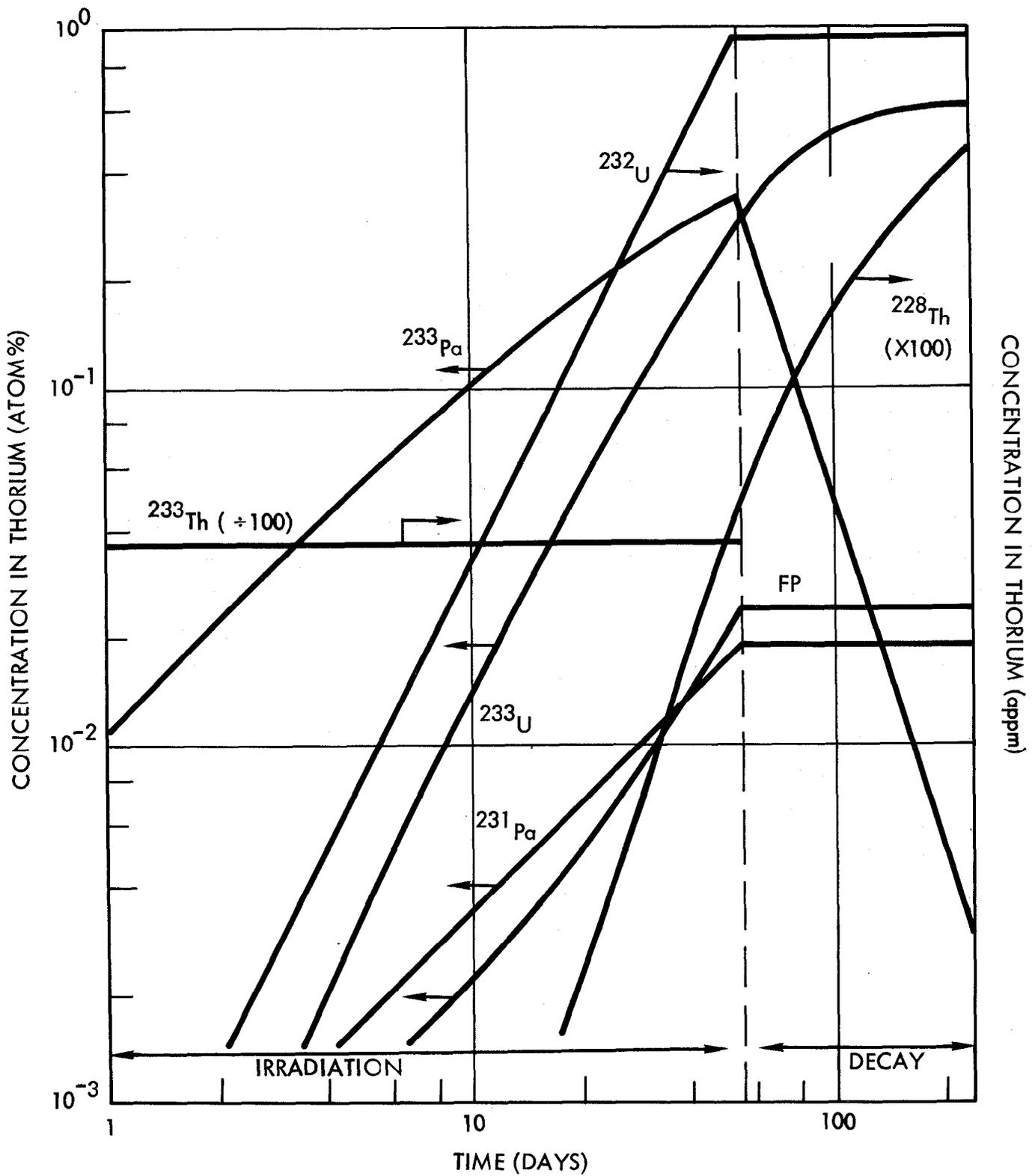


FIGURE VII.C-2. Concentration buildup of important isotopes and fission products during irradiation and six month cooling period for beryllium/thorium oxide blanket.

After reprocessing, the recovered thorium oxide may be stored to allow the  $^{228}\text{Th}$  and associated gamma activity to decay to levels where "hands-on" techniques for refabrication into TMHR fuel are possible. However, more detailed analysis is required to determine if such a procedure would be more economical than the use of remote handling or the simple disposal of the thorium. Table VII.C-2 shows the  $^{228}\text{Th}$  concentration and associated 2.6 MeV gamma activity in the thorium from the time of discharge from the reprocessing stream. The table gives only the  $^{228}\text{Th}$  bred in the hybrid, and not the natural occurrence of  $^{228}\text{Th}$  and other gamma activities associated with the natural decay chain of the fertile  $^{232}\text{Th}$  in the blanket.

TABLE VII.C-2. Radioactivity due to artificially produced  $^{228}\text{Th}$  in reprocessed thorium.

| Time From Separation (Yrs) | Density of $^{228}\text{Th}$ (appm) | 2.6 MeV- $\gamma$ Activity ( $\mu\text{Ci/g-Th}$ ) |
|----------------------------|-------------------------------------|--|
| 0                          | 0.00464                             | 1.334  |
| 0.25                       | 0.00424                             | 1.219  |
| 0.5                        | 0.00387                             | 1.113  |
| 0.75                       | 0.00353                             | 1.015  |
| 1.0                        | 0.00322                             | 0.926  |
| 2.0                        | 0.00224                             | 0.644  |
| 5.0                        | 0.00075                             | 0.216  |
| 10.0                       | 0.00012                             | 0.035  |
| 15.0                       | 0.00002                             | 0.006  |
| Natural Thorium            | 0.000136                            | 0.039  |

Table VII.C-3 gives the natural decay chain of  $^{232}\text{Th}$ . Besides having a constant 2.6 MeV gamma activity, other penetrating gammas are present due to the decay of  $^{228}\text{Ac}$  which is not present in the decay chain after  $^{228}\text{Th}$ . Computing the relative doses from the natural decay chain of  $^{232}\text{Th}$  and the

TABLE VII.C-3. Decay chain for natural thorium showing significant gamma lines and entry point from  $^{232}\text{U}$  decay.

| HEAVY NUCLIDE DECAY CHAIN<br>THORIUM SERIES (4n)                 |                |                         |                               |   |
|--|----------------|-------------------------|-------------------------------|---|
| NUCLIDE  | ELEMENT NAME   | HALF-LIFE               | $\lambda$ ( $\text{s}^{-1}$ ) | MAJOR GAMMA ENERGIES (MeV) AND INTENSITIES                |
| $^{232}_{90}\text{Th}$   | THORIUM - 232  | $1.41 \times 10^{10}$ y | $1.56 \times 10^{-18}$        | -----   |
| $^{228}_{88}\text{Ra}$   | RADIUM - 228   | 6.7y                    | $3.29 \times 10^{-9}$         | -----   |
| $^{228}_{89}\text{Ac}$   | ACTINIUM - 228 | 6.13h                   | $3.14 \times 10^{-5}$         | 0.34 (15%)<br>0.908 (25%)<br>0.96 (20%)                   |
| $^{232}_{92}\text{U} \rightarrow ^{228}_{90}\text{Th}$           | THORIUM - 228  | 1.9107                  | $1.15 \times 10^{-8}$         | 0.084 (1.6%)<br>0.214 (0.3%)                              |
| $^{224}_{88}\text{Ra}$   | RADIUM - 224   | 3.64d                   | $2.20 \times 10^{-6}$         | 0.241 (3.7%)  |
| $^{220}_{86}\text{Rn}$   | RADON - 220    | 55s                     | $1.26 \times 10^{-2}$         | 0.55 (0.07%)  |
| $^{216}_{64}\text{Po}$   | POLONIUM - 216 | 0.15s                   | 4.62                          | -----   |
| $^{212}_{82}\text{Pb}$   | LEAD - 212     | 10.64h                  | $1.81 \times 10^{-5}$         | 0.239 (47%)<br>0.300 (3.2%)                               |
| $^{212}_{83}\text{Bi}$   | BISMUTH - 212  | 60.6m                   | $1.91 \times 10^{-4}$         | 0.040 (2%)<br>0.727 (7%)<br>1.620 (1.8%)                  |
| $^{212}_{84}\text{Po}$ (64.0%)<br>$^{208}_{81}\text{Tl}$ (36.0%) | POLONIUM - 212 | 304ns                   | $2.28 \times 10^3$            | -----   |
| $^{208}_{81}\text{Tl}$   | THALLIUM - 208 | 3.10m                   | $3.73 \times 10^{-3}$         | 0.511 (23%)<br>0.583 (86%)<br>0.860 (12%)<br>2.614 (100%) |
| $^{208}_{82}\text{Pb}$   | LEAD - 208     | STABLE                  |                               | -----   |

hybrid produced  $^{228}\text{Th}$ , it appears the hybrid contributed dose is about equal to the natural dose after only about 10 years. This calculation is conservative since, in equilibrium, the natural chain has a 60% higher dose due to the  $^{228}\text{Ac}$  gammas. Therefore, storage of the reprocessed thorium for about 10-15 years would probably be satisfactory to allow use of whatever handling procedures were used initially for the natural thorium.

Also shown in Figure VII.C-2 is the equilibrium concentration of  $^{233}\text{Th}$  in the blanket. Since this isotope has a 22 m half-life, its concentration will quickly increase and decrease with the reactor operating power level. Therefore, while all other isotopic concentrations are calculated using the average plant capacity factor of 70%, the  $^{233}\text{Th}$  level is a conservative estimate based upon full power operation. This isotope is particularly important as it dominates the short term afterheat level in the blanket. The  $^{233}\text{Pa}$  ( $T_{1/2} = 27$  d) level is similarly important as it dominates the long term afterheat level.

Typical isotopic afterheat contributions for a beryllium/thorium oxide suppressed fission blanket (but not the reference blanket) are shown in Figure VII.C-3. As shown,  $^{233}\text{Th}$  dominates the afterheat for 0.7 hours,  $^{233}\text{Pa}$  dominates for the next 0.35 years, and fission products can be expected to dominate the afterheat only for longer periods. A more detailed discussion of afterheat safety considerations may be found in Chapter VI.

### VII.C.3 Isotopic Calculations for Reference Lithium/Molten Salt Blanket

Isotopic generation and depletion calculations for the fluorination only and full molten salt reprocessing modes were performed using the ISOGEN code. In both cases, the isotopic content of the blanket and primary loop was tracked over the 30 year operating life of the plant. The continuous removal of uranium and protactinium (as applicable) via reprocessing was also modeled. For this purpose, the process flow rates discussed in Section VII.A.3.b were used.

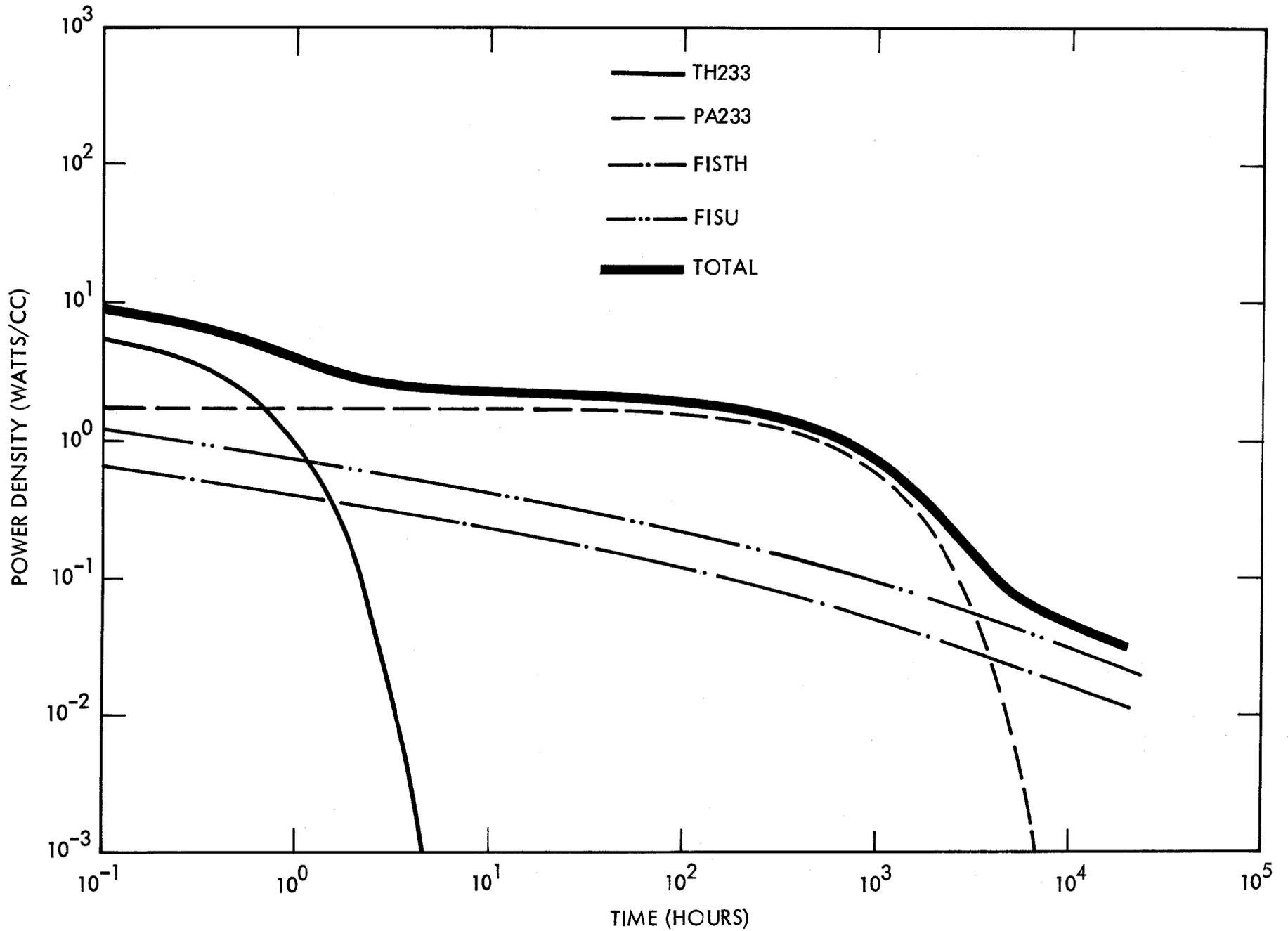


FIGURE VII.C-3. Typical suppressed fission blanket afterheat sources.

VII.C.3.a. Isotopics for Fluorination Only Reprocessing Mode. Figure VII.C-4 shows the calculated buildup of various isotopes and fission products during the 30 year plant lifetime. In this mode,  $^{233}\text{Pa}$  is not removed, but reaches its equilibrium concentration of 0.028% in thorium in only about 0.5 years. The  $^{233}\text{U}$  is removed, and its removal causes it to reach a process equilibrium concentration of about 0.11% in thorium in about 2 years. Concerning  $^{232}\text{U}$ , an equilibrium is almost attained in the 2-6 year period, but for later periods the  $^{232}\text{U}$  level continues to increase due to the continued buildup of  $^{231}\text{Pa}$  (see Figure VII.C-1) which is not removed. The  $^{232}\text{U}$  level after 30 years is  $3.28 \cdot 10^{-4}\%$  in thorium (3065 appm in  $^{233}\text{U}$ ). This level of  $^{232}\text{U}$  results in a  $^{228}\text{Th}$  level of 0.0823 appm in thorium. A maximum  $^{233}\text{Th}$  level of 0.23 appm in thorium was calculated based upon the assumption of short term operation (~several hours) at the full rated capacity of the plant (i.e., 100% plant factor).

Finally, a lumped estimate of fission product generation was made based upon the rate of  $^{232}\text{Th}$  and  $^{233}\text{U}$  fissions in the blanket. For the fluorination only process, the fission products accumulate linearly to a level of 0.24% in thorium in 30 years. However, it should be noted that this calculation does not account for losses of gaseous fission products which are not retained by the fuel. Also, since these fission products accumulate over the life of the plant (rather than during a short period as in an LWR) the associated radiological hazard is expected to be greatly reduced relative to that of the same quantity of "fresh" fission products. The particular makeup of these species is, however, unknown and this topic requires further investigation.

Since thorium recycle is continuously accomplished in the molten salt system, the principal isotopic issue associated with this blanket is the  $^{232}\text{U}$  level in the bred  $^{233}\text{U}$  discharged from the molten salt reprocessing plant. As shown in Table VII.C-4, the  $^{232}\text{U}$  discharge level increases during the plant lifetime to a level of 2400 appm after 30 years. The principal cause of this increase is an increasing  $^{232}\text{U}$  level in the molten salt primary loop (see Figure VII.C-4) which, in turn, results from a continuous buildup of  $^{231}\text{Pa}$  in the system.

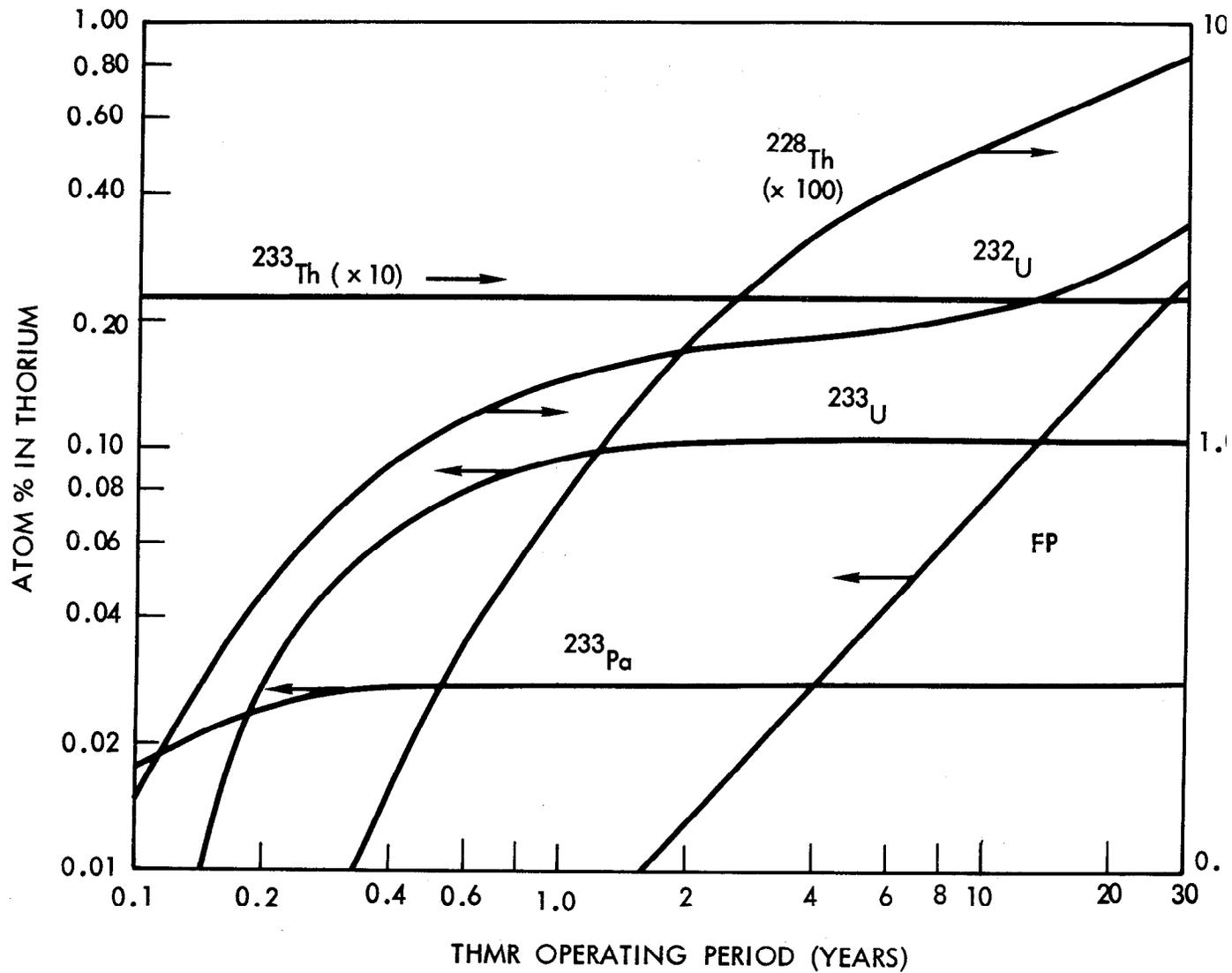


FIGURE VII.C-4. Isotopic generation in the molten salt blanket TMHR for fluorination only

TABLE VII.C-4.  $^{232}\text{U}$  discharge concentrations in  $^{233}\text{U}$  for the molten salt reprocessing plant.

| <u>Year</u> | <u>Concentration (appm)</u> |                        |
|-------------|-----------------------------|------------------------|
|             | <u>Fluorination Only</u>    | <u>Full Processing</u> |
| 5           | 1600                        | 1250                   |
| 10          | 1770                        | 1250                   |
| 15          | 1950                        | 1250                   |
| 20          | 2120                        | 1250                   |
| 25          | 2270                        | 1250                   |
| 30          | 2400                        | 1250                   |

Since the  $^{232}\text{U}$  level above is considerably higher than the  $^{232}\text{U}$  level discharged from the beryllium/thorium oxide system (175 appm in  $^{233}\text{U}$ ), the concerns related to shielding requirements for LWR fuel fabrication will be magnified. In particular, the results given in Table VII.C-1 would be multiplied by a factor of 13.9 for the molten salt blanket with fluorination only.

VII.C.3.b. Isotopics For Full Reprocessing Mode. Figure VII.C-5 shows the calculated buildup of various isotopes and fission products during the 30 year plant lifetime. In comparison with the fluorination only mode of fuel processing, the full processing option can remove protactinium and (possibly) some fission products from the primary molten salt loop. This results in the following features relating to isotopic generation:

- The equilibrium  $^{233}\text{Pa}$  level in the primary molten salt loop is about 20% lower.
- $^{231}\text{Pa}$  equilibrates in about 3 years to a 0.002% level in thorium. This results in lower  $^{232}\text{U}$  and  $^{228}\text{Th}$  generation.

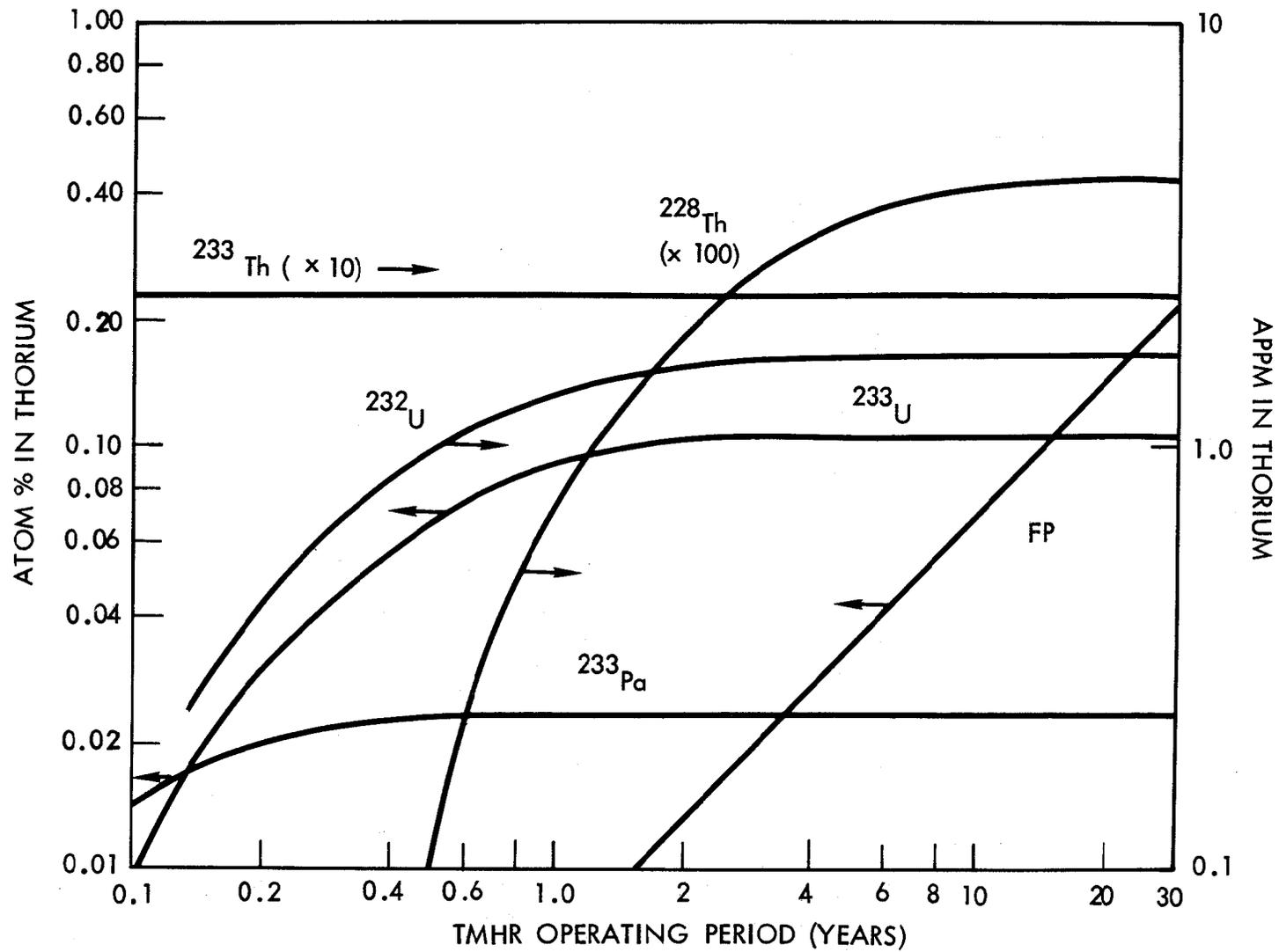


FIGURE VII.C-5. Isotopic generation in the molten salt blanket for full reprocessing mode.

• Fission product concentrations in the primary loop can, at most, be reduced about 10%. This results because the low rare earth fission product extraction efficiency (assumed to be 0.004) as well as the fission rate are both quite low. Since fission product removal requires a more sophisticated molten salt process with more required development (see section VII.B.2.C), this option has not been pursued.

As a result of  $^{231}\text{Pa}$  control for this process,  $^{232}\text{U}$  concentrations is the bred  $^{233}\text{U}$  are expected to be reduced up to 50% over the life of the plant (see Table VII.C-4). However, the calculated  $^{232}\text{U}$  levels are still about 7 times higher than those of the beryllium/thorium oxide blanket. Further reductions might be achieved by increasing the molten salt processing rate to further reduce the levels of  $^{231}\text{Pa}$  and  $^{233}\text{U}$  (both are parents of  $^{232}\text{U}$ ), by reducing the molten salt inventory in the primary loop by the use of reflector materials (e.g., graphite), and by the use of a thicker multiplier region to further soften the neutron energy spectrum. Further studies are required to determine realistic goals with respect to tolerable levels of  $^{232}\text{U}$  which reflect practical considerations regarding remote and shielded fuel fabrication activities.

## VII.D. CONCLUSIONS

### VII.D.1. Fuel Reprocessing Costs

Commercial introduction of the TMHR will certainly be paced by the price of bred fissile fuel as compared with the price of natural uranium ores. Since the bred fuel cost is substantially influenced by the cost of extracting the fissile fuel from the hybrid fertile fuel, reprocessing costs can be a major determinant in the viability of the suppressed fission hybrid. Our results indicate that acceptable fuel cycle costs (consistent with the timeframe for the development of commercial fusion applications and the depletion of inexpensive sources of natural uranium) appear possible.

Table VII.D-1 compares reprocessing costs for the thorium metal, oxide, and salt cases. The unit costs to reprocess thorium oxide fuels using the THOREX process are expected to be most expensive (28-42 \$/gm) while the cost to reprocess thorium metal fuels is expected to be significantly less (23 \$/gm) and the expected cost of molten salt reprocessing (2-4 \$/gm) is greatly reduced when compared with the other options. For the reference lithium/molten salt and beryllium/thorium oxide blankets, the fraction of the overall levelized bred fuel cost (see Chapter IX) attributable to reprocessing is 1.15% and 30% (worst case), respectively. The low cost of molten salt reprocessing is a significant economic advantage, but molten salt technology is less developed than the THOREX-based options and has a greater impact upon the blanket design itself (i.e., materials issues and design fixes to circumvent such issues).

### VII.D.2 Isotopic Considerations

The quality of bred fuel and the effect upon fuel cycle costs depends on the concentrations of contaminant isotopes. A previous study of the quality of hybrid produced fuel suggested that  $^{233}\text{U}$  production was unattractive because of the high  $^{232}\text{U}$  concentrations.<sup>54</sup> Figure VII.D-1 compares the  $^{232}\text{U}$  concentrations in hybrid bred  $^{233}\text{U}$  between the fast fission and suppressed fission cases. As shown, each of the suppressed

TABLE VII.D-1. Comparative results for various fuel forms and reprocessing technologies.

|   | Fuel Type/Reprocessing Option |               |                                    |                              |
|---|-------------------------------|---------------|------------------------------------|------------------------------|
|   | Thorium Metal                 | Thorium Oxide | Reference Molten Salt <sup>b</sup> | Molten Salt with Pa Recovery |
| Total fissile discharge assay in thorium <sup>a</sup> | 0.55%                         | 0.55%         | 0.14%                              | 0.13%                        |
| Uranium discharge assay in thorium                    | 0.25%                         | 0.25%         | 0.11%                              | 0.11%                        |
| Process rate, MT/yr                                   | 1200                          | 1200          | 5440                               | 3160                         |
| Levelized cost per Kg thorium                         | \$127                         | \$152-233     | \$1.59                             | \$5.06                       |
| Levelized cost per gm uranium                         | \$23                          | \$28-42       | \$1.93                             | \$3.90                       |

<sup>a</sup>Uranium plus protactinium.

<sup>b</sup>For this system only uranium is recovered and recovery efficiency is 75%. So cost per gram =  $1.59 / (1.1 \cdot 0.75) = 1.93$  \$/gm.

fission cases results in a lower <sup>232</sup>U concentration than the fast fission case of equivalent fissile discharge enrichment - a factor of 45 lower for the reference beryllium/thorium oxide blanket.

Reference 54 suggests that for <sup>232</sup>U concentration below ~ 400 appm, shielding and remote handling requirements may be reduced, and that up to 1200 appm the technology developed for HTGR reprocessing applies. If this assertion is correct, the discharge fuel from the reference beryllium/thorium oxide blanket would not require fully shielded fabrication facilities with remote handling and the discharge fuel from a lithium/molten salt blanket with a molten salt process to remove both uranium and protactinium could utilize already developed HTGR fuel fabrication technology.

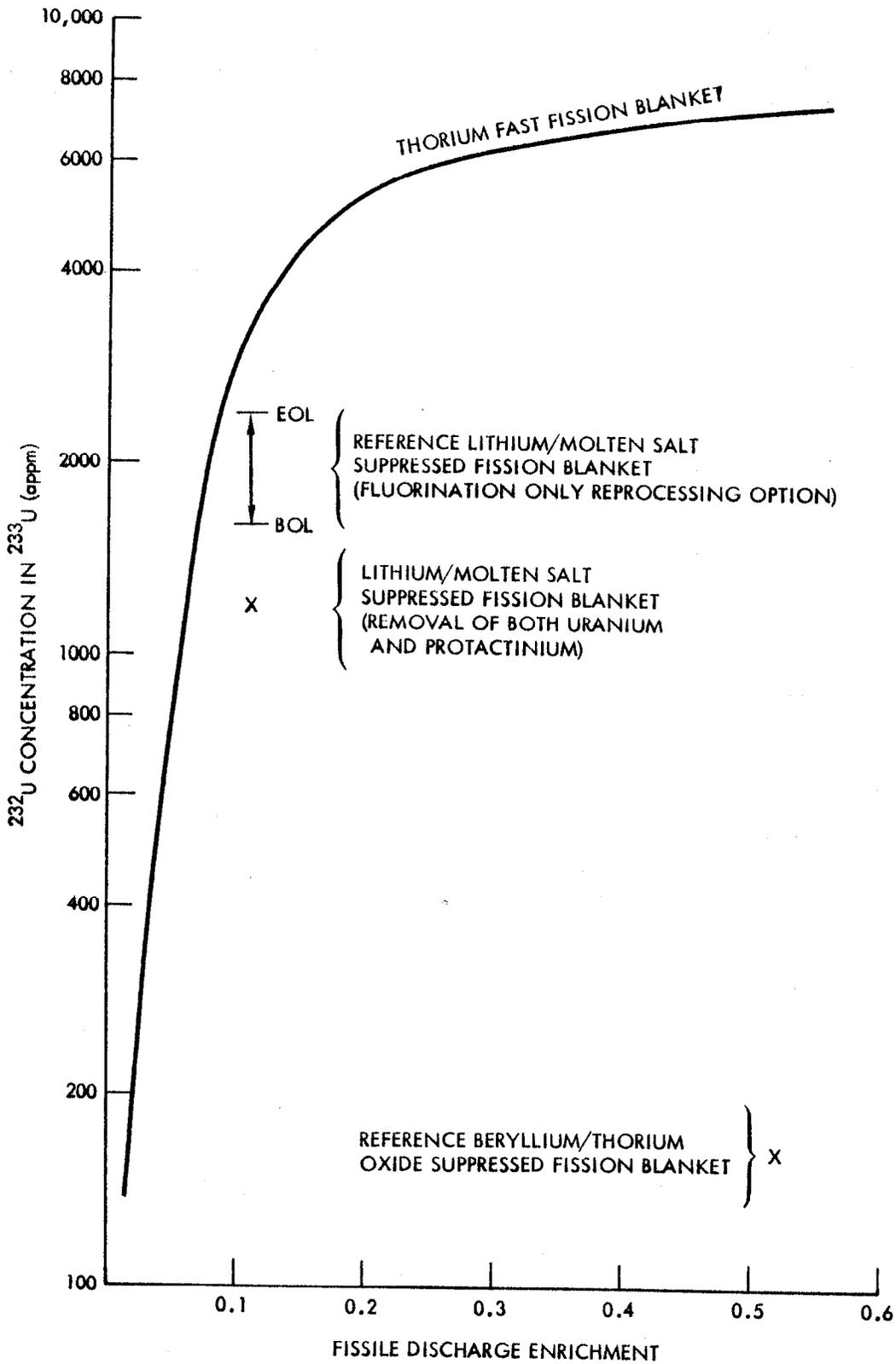


FIGURE VII.D-1. Comparison of  $^{232}\text{U}$  generation for fast and suppressed fission blankets.

In contrast, for the molten salt blanket with uranium recovery only (the reference reprocessing option for this blanket), the expected  $^{232}\text{U}$  levels are higher than 1200 appm (i.e., 1600-2400 appm) an increase over the life of the plant due to the accumulation of  $^{231}\text{Pa}$  in the blanket (see Section VII.C.3.a). The impact of these  $^{232}\text{U}$  generation levels is unknown and further studies are required.

Concerning the buildup of  $^{228}\text{Th}$  activity in fertile thorium to be refabricated and recycled back to the blanket following reprocessing and recovery of the bred fissile fuel, we have shown (in Section VII.C.2) that a 10-15 year cooling period will suffice to reduce the radioactivity level below the natural background level associated with unirradiated thorium. This concern is irrelevant to the molten salt blanket since these systems avoid fuel fabrication steps. Its impact on fuel fabrication for the thorium oxide blanket is unknown, but the worst case (i.e., 15 years cooling), is not prohibitive.

### VII.D.3. Proliferation Resistance

Diversion or proliferation resistance is an issue of some interest and the hybrid offers unique features in this respect. For example, the 2.6 MeV gamma associated with the  $^{232}\text{U}$  decay chain is always present in some quantity and we can consider some amount to be beneficial. Hopefully, the suppressed fission hybrid will have enough penetrating gamma to afford some degree of proliferation resistance without requiring excessive shielding. The presence of penetrating radioactivity in the fissile fuel makes the diversion of the fuel difficult, provided the activity is high enough to require remote handling while moving the fuel assemblies and removing the fissile material. Diversion resistance due to high radioactivity is not considered to render the fuel proliferation-proof, so isotopic denaturing and institutional safeguards are still desirable for proliferation-resistance.  $^{232}\text{U}$  can be isotopically denatured by the addition of  $^{238}\text{U}$  in the fabricated fuel. If the uranium contains less than 20% fissile isotopes, it is considered to be isotopically denatured.<sup>4,55</sup> Fabricated fuel for use in LWRs could contain 3%  $^{233}\text{U}$  and a minimum of 18%  $^{238}\text{U}$  and be considered denatured. The remaining 79% of the fuel could be either  $^{238}\text{U}$  or  $^{232}\text{Th}$ .

Fuel containing 79% thorium might be desirable because it would convert thorium to  $^{233}\text{U}$ , which would be isotopically denatured in the spent fuel. The  $^{238}\text{U}$  will convert to  $^{239}\text{Pu}$  which is not isotopically denatured in the spent fuel. While some  $^{239}\text{Pu}$  will be bred in both fuel types, about seven times as much will be bred in the 97%  $^{238}\text{U}$  fuel. If  $^{239}\text{Pu}$  can be burned only in safeguarded reactors, then it may be advantageous to breed the minimum amount of  $^{239}\text{Pu}$ .

One advantage of the 3%  $^{233}\text{U}$ , 97%  $^{238}\text{U}$  fuel is that the reprocessing of  $\text{UO}_2$  is easier, less expensive, and a better established technology than reprocessing  $\text{ThO}_2$ . Thorium oxide is difficult to dissolve and requires the addition of corrosive fluorine which increases the cost of reprocessing (see Section VII.B). Also, this denatured fuel cycle option provides an additional margin of denaturing for the fresh fuel charged to the LWR. Fusion fuel factories, distributed LWRs, and safeguarded plutonium burners will require tradeoff analyses of reprocessing economics and proliferation-resistance.

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