REFERENCE THORIUM FUEL CYCLE

F. E. Driggers

August 1978

Approved by
L. H. Meyer, Program Manager

E. I. DU PONT DE NEMOURS AND COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, SOUTH CAROLINA 29801

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT AT(07-2)-1

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CONTENTS

REFERENCE THORIUM FUEL CYCLE

INTRODUCTION

Development of processing technology for spent thorium fuel began October 1, 1977 as part of the Thorium Fuel Cycle Technology Program (TFCT Program). The scope of the program includes broad evaluations to identify viable thorium/uranium recycle strategies; research and development programs to confirm the feasibility of the selected fuel cycle or cycles; a design integration study to identify development areas and safeguards and proliferation aspects; and the development and testing of key systems, equipment, and components.

In September 1977, a preliminary thorium fuel cycle was defined to serve as a common basis for development work at participating DOE laboratories, universities, and commercial facilities. This fuel cycle has been revised in several minor ways. The revised fuel cycle, as described in this report, should still be considered a preliminary one because proliferation criteria have not yet been completely developed and because economic comparisons of alternative thorium fuel cycles are not complete.

SUMMARY

Characteristics of the reference fuel cycle for the TFCT Program are:

- Fissile uranium will be denatured by mixing with ^{238}U.

- Chemical processing plant design will be based on the assumption that plants are located in secure areas.

- Plutonium will be recycled within these secure areas.

- Thorium will be recycled with recovered uranium and plutonium.

- The head end of the chemical processing plant will handle
  a variety of core and blanket fuel assembly designs for
  light water reactors (LWRs) and heavy water reactors (HWRs).

- The fuel form may be a homogeneous mixture either of uranium
  and thorium oxide powders pressed into pellets or sol-gel
  microspheres compacted using vibratory techniques.

- Fuel cladding will be *Zircaloy.**

- MgO may be added to the fuel to improve the thorium
  dissolving characteristics.

Thorium is being considered as the fertile component of
nuclear reactor fuel in the expectation that its use will
provide increased proliferation resistance compared to the use
of ^{238}U as the fertile material in reactor fuel. The reason is
that ^{235}U (which constitutes the initial fissile content) and
^{233}U (which is produced when thorium is irradiated) can be
denatured, or made unusable for feasible explosive devices, by
mixing with ^{238}U, whereas plutonium cannot be denatured. How-
ever, the use of ^{238}U results in the production of plutonium
during irradiation at a rate of about one-third that in LWR
uranium fuel cycles. It is thought that a reprocessing plant
cannot be designed to prevent the recovery of this plutonium
by those in control of a functioning plant, and it is therefore
assumed that the plant will be located in a secure area. Within
this area, it is assumed that plutonium can be fabricated into
fuel assemblies in co-located facilities and recycled in co-
located reactors. It is also assumed that spent uranium fuel
can be processed if it is economically attractive to do so. A
block diagram of the proposed fuel cycle is included (Figure 2).

Spent uranium recovered from thorium based fuels must be
re-enriched before recycle to prevent a very rapid buildup of
^{238}U at the expense of thorium as recycle proceeds. If the
spent uranium is re-enriched with denatured ^{235}U, ^{238}U will
build up as recycle proceeds at a slower but still unacceptable
rate. Thus, it is assumed that the recovered uranium will be
re-enriched with highly enriched ^{235}U. In addition to elimi-
nating the buildup of ^{238}U with recycle, the use of highly
enriched uranium also results in decreased ^{238}U feed and
separative work requirements.

---

* Registered trademark of Westinghouse Electric Corp.,
  Blairsville, Pa.

Stainless steel will continue to be considered as a backup to *Zircaloy* in case the use of zirconium proves to be incompatible with commercial-scale aqueous dissolution. The storage of recovered irradiated thorium will be considered as a backup to its use in the recycle of recovered plutonium and uranium in case this turns out to be economically desirable. Finally, the use of additives to the pre-fabrication oxides will be considered as needed. The main example of such additives is the use of MgO to improve dissolution characteristics.

Estimates have been made of the time required to introduce the thorium fuel cycle into the current and developing LWR power industry. Given adequate motivation for switching to thorium, a modest penetration by the thorium fuel cycle could be achieved in about 15 years and complete penetration might occur 5 to 10 years later.

It is likely, if the thorium fuel cycle is implemented, that LWRs will continue using the once-through uranium cycle for the intervening years and that uranium fuel exposures will be increased from about 30,000 MWD/MT to about 50,000 MWD/MT. There may then be considerable interest in developing a thorium fuel cycle that has similarly high exposures. The reprocessing plant should be designed with the possibility in mind that thorium fuel with an average exposure of about 50,000 MWD/MT might be received at the reprocessing plant.

DETAILS

Proliferation Resistance

Proliferation criteria have not yet been completely established; hence, some assumptions have been made concerning these criteria. It is understood that the selected fuel cycle may require some modifications when proliferation criteria are completely formulated.

Assumptions related to proliferation criteria that were used in selecting the thorium fuel cycle described in this document are:

● Unrestricted recycle of separated plutonium is unacceptable.

● Fissile uranium must be denatured with ^{238}U.

● Chemical processing plants must be located in secure areas.

● Plutonium may be recycled through Pu/Th fabrication plants to light water or breeder reactors within these secure recycle areas.

- Spent uranium fuel may be reprocessed within the secure recycle areas.

- Uranium with a higher fissile content than the denaturing limit is available either in secure areas containing enrichment plants or in the secure recycle areas.

Reasons for, and consequences of, some of these assumed proliferation criteria are discussed in subsequent sections.

Uranium Denaturing and Re-enrichment

The amount of fissile material required to make any particular type of nuclear explosive device increases with the amount of ^{238}U assumed to be mixed with the fissile material. A proliferation resistance threshold for each fissile uranium isotope can be defined as the concentration of that isotope in a ^{238}U mixture at which and below which the explosive device is considered to be impractical or impossible without isotopic enrichment. The fissile isotope is then said to be denatured. Current estimates are that concentration limits of 20% for ^{235}U and 12% for ^{233}U are acceptable. These estimates will be refined during the course of the TFCT Program. Plutonium cannot be denatured.

Typical thorium fuel compositions that meet this denaturing requirement are given in Table 1. The denaturing limit for combinations of ^{233}U and ^{235}U is assumed to be a linear combination of the limits for each isotope separately. Thus, the fraction of ^{238}U required to denature the fissile uranium listed for the spent fuel in Table 1 is 0.135 compared to 0.177 actually present, and uranium recovered from spent fuel is inherently denatured.

Spent fuel does not have sufficient fissile content to be used directly in a uniform fuel loading. Either the thorium content must be reduced or the fissile content must be increased, for example, by blending with enriched uranium. The consequences of re-enriching the recycled fuel by blending with uranium of various enrichments are summarized in Table 2. It is assumed that 1 kg of spent fuel with the composition given in Table 1 is mixed with x kg of ^{235}U and k x kg of ^{238}U where x is determined by requiring that the ratio of the sum of $\nu\Sigma_f$ to the sum of Σ_a in the recycle fuel be equal to the ratio for fresh fuel, and k is the ratio of ^{238}U to ^{235}U. At the composition of spent fuel in an LWR, the following ratios are calculated.

Isotope	$\sigma_\alpha^i/\sigma_\alpha^{th}$	$(\nu\sigma_f)^i/\sigma_\alpha^{th}$
^{232}Th	1.0	0.07
^{238}U	1.8	0.30
^{235}U	41	79
^{233}U	53	117

Significant parameters for comparing the use of various makeup concentrations are the increase in total ^{238}U in the reactor, at the expense of thorium, and the feed and separative work required to produce a fixed amount of fuel. Data for these parameters are derived from Table 2 and are given in Table 3. The results are also plotted in Figure 1. Clearly, the makeup uranium should contain the highest ^{235}U level available. Since the ^{235}U in the makeup U exceeds the denaturing limit, either the makeup U can be securely shipped (if acceptable) to the recycle secure area or the recycle material can be shipped to the enrichment secure area for blending. The resulting blend is below the denaturing limit.

Disposition of Irradiated Thorium

Irradiated thorium is radioactive and its use in fabrication plants will require remote operation. Because recycled uranium is also radioactive, it is proposed that irradiated thorium and uranium be recycled together. Fresh fuel can be fabricated using new thorium either in existing fabrication plants or in similar new plants. Recycled fuel will be fabricated in plants designed for remote operation. In this way, thorium mining requirements are reduced from the total lifetime throughput (if new thorium is always used) to the amount required to fill the pipeline (which is proportional to the number of reactors) plus enough to make up for minor burnup and processing losses. In any event, thorium mining requirements will be much less than uranium mining requirements since uranium is mined primarily for its fissile content.

Disposition of Irradiated Plutonium

Since fissile uranium must be denatured with ^{238}U, plutonium will be produced during irradiation. As indicated in Table 1, plutonium comprises a little over 20% of the fissile isotopes produced during the irradiation and remaining after an average exposure of 33,000 MWD per metric ton. In a typical 1500 MT/year chemical processing plant, the fissile plutonium throughput will

be 10 to 15 kg per operating day. It is assumed that it will not be possible to design a thorium cycle chemical processing plant that will make obtaining plutonium appreciably harder or more time-consuming, given control of a functioning plant, than is the case in a processing plant designed for a uranium fuel cycle. It is, therefore, assumed that the chemical processing plants for the denatured thorium cycle must be located in secure areas.

It has been calculated that, averaged over an assumed 30-year reactor life, uranium feed requirements for a thorium cycle with storage of plutonium are about equal to those of the uranium cycle with recycle of uranium and plutonium and are 32% less than for the uranium stowaway cycle. Thus, a given amount of uranium feed will fuel 47% more reactors using the thorium (Pu storage) cycle than using the uranium stowaway cycle. If plutonium is also recycled in the thorium cycle, the given amount of uranium will fuel 10% more reactors than in the plutonium storage case. Thus, although plutonium recycle is not absolutely essential, it makes a significant contribution.

Because plutonium-bearing spent fuel must be processed in a secure area, it seems reasonable to burn it as Pu/Th fuel in the secure area. Not only does it allow operation of additional reactors in the secure area without an increase in uranium resource requirements, but it also provides additional ^{233}U for recycle in dispersed reactors. Finally, burning the separated plutonium may be safer than storing it.

Fabrication Plants

This section will not discuss the design of fabrication plants but only their location in the fuel cycle. It is expected that plants designed to fabricate uranium fuel can be used to fabricate non-recycled Th/U fuel. When fuel is recycled, plants designed for remote operation will be required. Two types of remotely operated fabrication plants are required although they are not necessarily completely independent. Pu/Th fuel must be fabricated inside the secure area in which it is recovered and irradiated. Recycled U/Th fabrication plants may be built anywhere since the fissile content is denatured. They may be combined with Pu/Th plants in secure areas if this is economically advantageous.

Reactors

Current light water reactors can be loaded with unrecycled thorium fuel. Receiving and handling facilities would have to be modified somewhat for recycled fuel. Reactors using Pu/Th fuel would have to be located in secure areas. Full Pu/Th loads are probably feasible, but a supply of U/Th fuel designed to be compatible with the Pu/Th fuel design should be available for use when there is insufficient plutonium for refueling a reactor.

As time proceeds, the use of HWRs loaded with thorium might become attractive and the fuel fabrication and reprocessing industry should be adaptable to the changes required.

Chemical Processing Plants

As described earlier, chemical processing is assumed to occur only in secure areas. The most important product would be the uranium stream which contains most of the fissile isotopes. Because the uranium is denatured, it does not matter, for proliferation resistance, whether it is coprocessed with some or all of the thorium or not. The uranium and/or thorium can be shipped to dispersed fabrication plant sites if desired.

The remainder of the fissile content of the spent fuel is plutonium. Because plutonium is protected by keeping it in a secure area, it may be coprocessed with some of the thorium or not depending on the relative feasibility of the two flow designs. In any event the plutonium will be blended with thorium before fabrication.

Recycling of Spent Uranium Fuel

Although the unrestricted recycle of plutonium is unacceptable, it has been assumed that plutonium will be recycled within secure areas in the thorium cycle. If chemical processing plants are designed to handle spent uranium fuel, it is feasible from a technical and proliferation resistance point of view to reprocess the stockpile of spent uranium and to recycle the plutonium (mixed with thorium) within the secure area. The uranium would be sent to enrichment plants. The amount of excess processing capacity provided for this purpose would have to be chosen on the basis of an economic study. Because it is desirable not to require too large a fraction of reactor capacity within secure areas (for plutonium recycle), it is assumed that the rate of reprocessing spent uranium would remain small, and thus, that the spent uranium stockpile would last a number of years.

Complete Fuel Cycle

Figure 2 is a schematic diagram of a fuel cycle incorporating the concepts described in previous sections. The top row of blocks represents the current status of the industry. If a decision is made to convert to the thorium fuel cycle, existing uranium-based fuel fabrication plants and light water reactors would be converted.

Spent thorium-based fuel would be stored until chemical processing capacity becomes available. Chemical processing plants, located in secure areas, would recover spent uranium, would re-enrich it with highly enriched uranium, and would ship it, together with recovered thorium, to fabrication plants designed for remote operation. Plutonium would also be recovered and transferred, mixed with recovered thorium, to fabrication plants within the secure area. Pu/Th fuel would then be irradiated in reactors within the secure area. Recycled U/Th would be used in these reactors in the absence of enough Pu/Th fuel. To the extent that excess chemical processing capacity is available, spent uranium-based fuel would also be processed. Recovered plutonium would be used in co-located facilities and recovered uranium would be re-enriched in the enrichment plants.

Fuel Alternatives and Choice of Reference Design

A list of alternatives chosen to characterize the fuel in the thorium cycle is given in Table 4. Some of the implications of the various alternatives are discussed in this section.

Most uranium-based reactor and assembly loadings are uniform except for variations in enrichment. There may be an advantage in the thorium cycle in introducing several types of fuel assemblies (core and blanket, for example) in a single reactor charge and in using assemblies with different types of fuel columns (seed and blanket design, for example). The primary implication of such mixed loadings for chemical processing is in the flexibility required of the head end (the shear design, for example). It appears to be worthwhile to provide this flexibility so that a range of LWR or HWR thorium fuel assembly designs can be reprocessed.

Candidate cladding materials are *Zircaloy* and stainless steel. Thorium is more difficult to dissolve than uranium, and the fluorine that is probably required as a catalyst will probably complex with the *Zircaloy*, thus increasing the volumes of waste and complicating thorium dissolution. Stainless steel would be expected to dissolve to a lesser extent. However, if the thorium cycle is adopted partly because of its long range uranium resource savings, it is important to achieve a high conversion ratio during reactor operation and stainless steel absorbs many more neutrons than *Zircaloy*. A high conversion ratio would be reflected in a lower initial enrichment requirement with *Zircaloy* as opposed to stainless steel and thus as an immediate uranium savings. For this reason, *Zircaloy* was selected as the fuel cladding for the reference fuel cycle. Stainless steel cladding will be carried as a backup.

Possible fuel forms are oxides, carbides, nitrides, or metal alloys of uranium and thorium. Oxide is selected as a conservative choice because more irradiation experience with oxide fuel forms is available.

The product sent from the reprocessing plant to the fabrication plants could be oxide powder or some small refractory form such as microspheres obtained from the sol-gel process developed at Oak Ridge. Because the fissile uranium is denatured, little proliferation resistance advantage exists for coprocessing the uranium with thorium, and it is assumed that the uranium and thorium are recovered separately. Because ^{233}U will contain ^{232}U which is radioactive, it is assumed that recycled fuel will also use irradiated thorium rather than storing the thorium to allow decay. Oxide powder or sol-gel microspheres, produced either at the processing plant or the fabrication plant, are the chosen fuel forms. It is possible that pressed pellets will be used for fresh fuel fabricated in current plants and microspheres will be used for remote fabrication of recycled fuel.

It is assumed that plutonium is recycled within the secure recycle area and that it will be recovered separately to minimize the capital investment required. The recovered plutonium will be mixed with irradiated thorium either in the reprocessing plant or in the co-located fabrication plant.

Additives to the fuel could be considered to increase reactor exposure limits or to make the irradiated fuel easier to dissolve. There is some evidence that MgO makes thorium easier to dissolve and the effect of its use on reactor performance and on dissolvability will be investigated.

TABLE 1

Typical Fuel Compositions, weight fractions

| Isotope | Initial Fuel | | Limit of Fresh Recycle Comp.[b] |
	Fresh	Spent[a]	
Th	0.7787	0.7861	0.708
^{235}U	0.0438	0.0161	-
^{238}U	0.1775	0.1769	0.257
^{233}U	-	0.0119	0.035
Fissile Pu	-	0.0028	-

---

a. At an assumed exposure of 33,000 MWD/MT.

b. Assuming ^{233}U available from thorium blanket irradiation. Recycle material will be intermediate in composition between fresh fuel and this limit.

TABLE 2

Blending of 1 kg of Spent Fuel and Makeup U

| | ^{235}U in Makeup U | | | | Fresh Fuel[b] |
	0.2	0.3	0.5	0.9	
k	4	2.33	1.0	0.091	-
^{233}U, g	10.8	10.8	10.8	10.85	-
^{235}U, g	28.2	26.8	26.0	25.47	44.3
^{238}U, g	218.3	198.0	185.4	178.05	177.2
^{232}Th, g	793.8	793.8	793.8	793.82	778.5
Total HM, g	1051.2	1029.5	1016.0	1008.19	1000.0
Fresh fuel, g[a]	0	21.7	35.2	43.0	51.2
Total ^{233}U, g	10.8	10.8	10.8	10.8	-
Total ^{235}U, g	28.2	27.8	27.5	27.4	46.6
Total ^{238}U, g	218.3	201.8	191.6	185.6	186.3
Total ^{232}Th, g	793.8	810.7	821.2	827.3	833.7

---

a. The amount of heavy metal is largest when a makeup uranium value of 0.2 is used; fresh fuel is added to the other cases for the purpose of comparison.

b. Included for comparison with recycle fuel.

TABLE 3

Comparison of Blended and Fresh Fuels per 1.051 kg Total Fuel

	^{235}U in Makeup U				Fresh Fuel
	0.2	0.3	0.5	0.9	
Increase in ^{238}U, %	17.2	8.4	2.9	0	0
Total U in Makeup, g	51.2	29.5	16.0	8.2	-
Total U in fresh fuel, g[a]	0	4.8	7.8	9.5	232.8
U feed required, kg[b]	1.98	1.91	1.86	1.81	9.02
SW required, kg SWU[b]	2.34	2.32	2.31	2.30	10.65

a. All at 20% ^{235}U.

b. For comparison, the feed and SW contents of 1.051 kg of uranium fuel (3.3% ^{235}U) are 6.38 kg and 5.22 kg SWU, respectively.

TABLE 4

Selected Alternatives for the Reference Thorium Fuel Cycle

Parameter	Selected Alternative
Reactor Loading and Assembly Design	Variety of blanket and core designs for LWR and HWR
Cladding	Zircaloy[a]
Product from Reprocessing	Mixed oxide powders or sol-gel microspheres
Product from Fabrication	Pressed powder pellets or vibratory compacted microspheres
Thorium Disposal	Recycled with ^{233}U and Pu
Additives	MgO

a. Stainless steel will be considered a backup candidate if chemical processing problems with Zircaloy are too great.
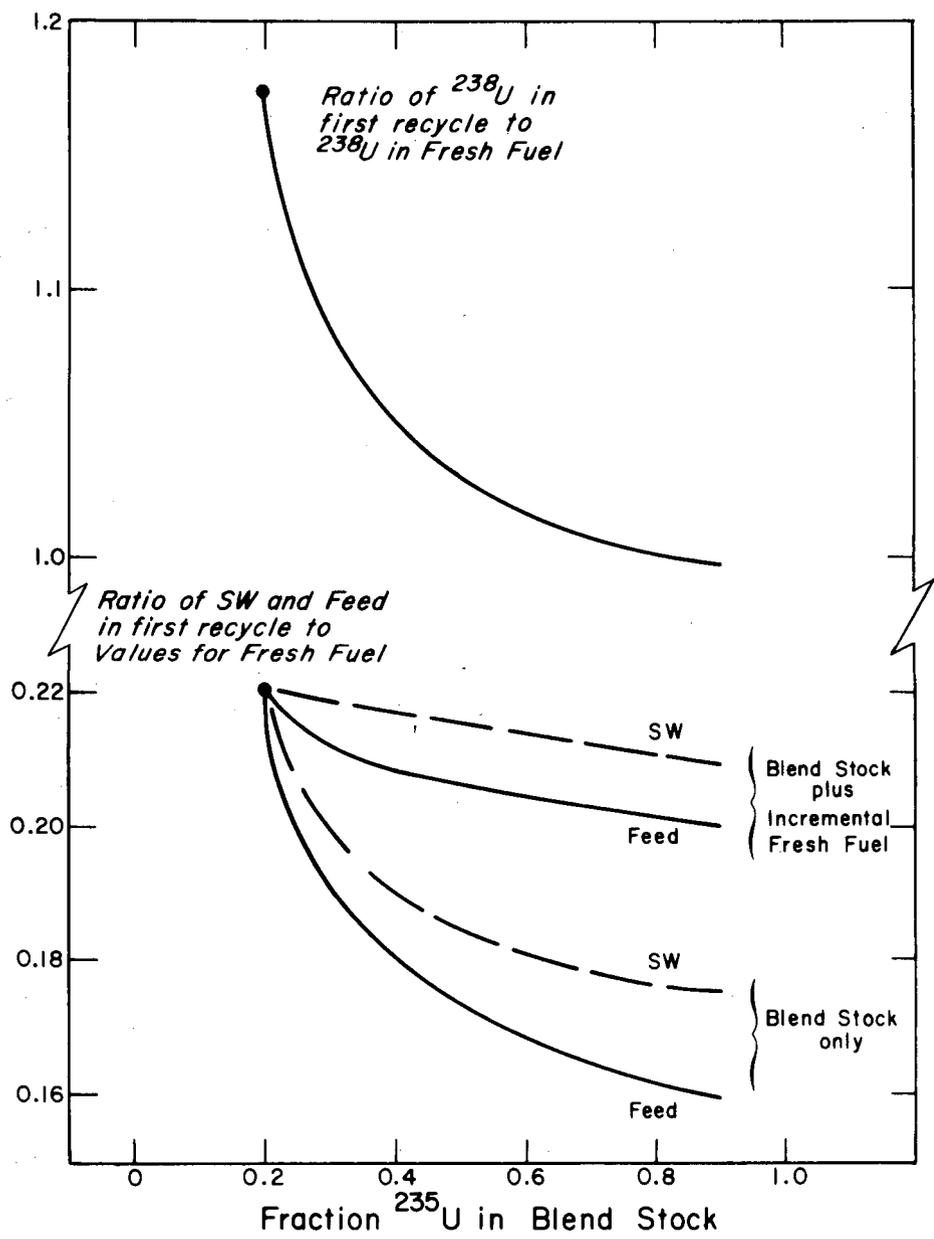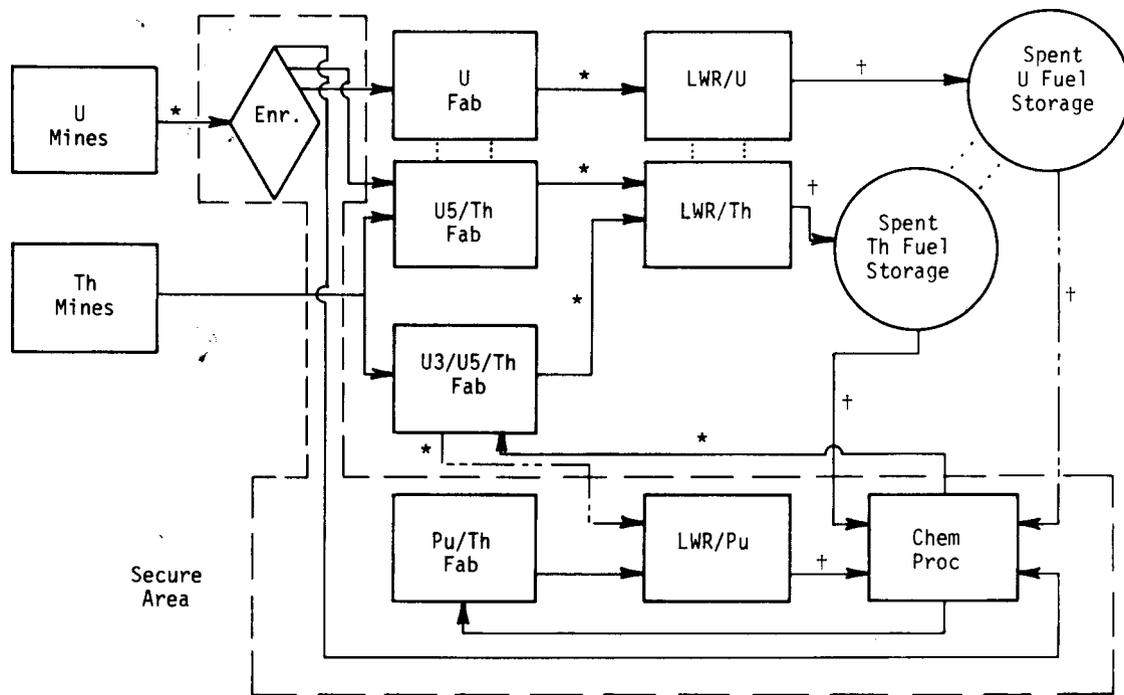
FIGURE 1. Effects of Blend Stock Enrichment

Fissile Content Protected by: *   Denaturing
                                †   Fission Products

FIGURE 2.   Proposed Thorium Fuel Cycle