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REPROCESSING AND FUEL FABRICATION SYSTEMS

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DISCLAIMER

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ABSTRACT

The study of alternative fuel cycles was initiated to identify a fuel cycle with inherent technical resistance to proliferation; however, other key features such as resource use, cost, and development status are major elements in a sound fuel cycle strategy if there is no significant difference in proliferation resistance. Special fuel reprocessing techniques such as coprocessing or spiking provide limited resistance to diversion. The nuclear fuel cycle system that will be most effective may be more dependent on the institutional agreements that can be implemented to supplement the technical controls of fuel cycle materials.

a. The information contained in this article was developed during the course of work under Contract No. AT(07-2)-1 with the U. S. Department of energy.

BACKGROUND

In April 1977, President Carter announced a nuclear policy that contained the following major new features:

- The reprocessing of spent nuclear fuel was deferred indefinitely in the U. S.
- The pace of developing the liquid metal fast breeder development was reduced.
- Programs of nuclear development were redirected to emphasize alternative fuel cycles that reduce the risk of proliferation; fuel cycles were to feature reduced access to material directly usable in nuclear weapons.

The Department of Energy (DOE) responded to the announcement by establishing a Nonproliferation Alternative Systems Assessment Program (NASAP) to identify nuclear systems with added resistance to proliferation as well as other beneficial features of nuclear power systems:

- Favorable economics so that nuclear power costs are competitive with nonnuclear sources.
- Ample resources so that the power source is reliable for the years needed to recover the investment of time and people in a new power source.
- Reasonable implementation or timing of commercialization so that the system can be considered in energy planning schedules.

Some of the other data for NASAP include unique environmental features or safety characteristics that would require special technical effort or revised regulatory considerations to implement the fuel cycle. Thus, the scope of the fuel cycle evaluations involve more parameters than proliferation resistance; but this new area requires special study[1] to define parameters of interest.

This paper will focus on features of fuel cycle facilities associated with some of the alternative fuel cycle systems included in the scope of the NASAP study. A generalized fuel cycle diagram is shown in Fig. 1. Two processes in the fuel cycle are the sensitive technologies for proliferation: enrichment and reprocessing.

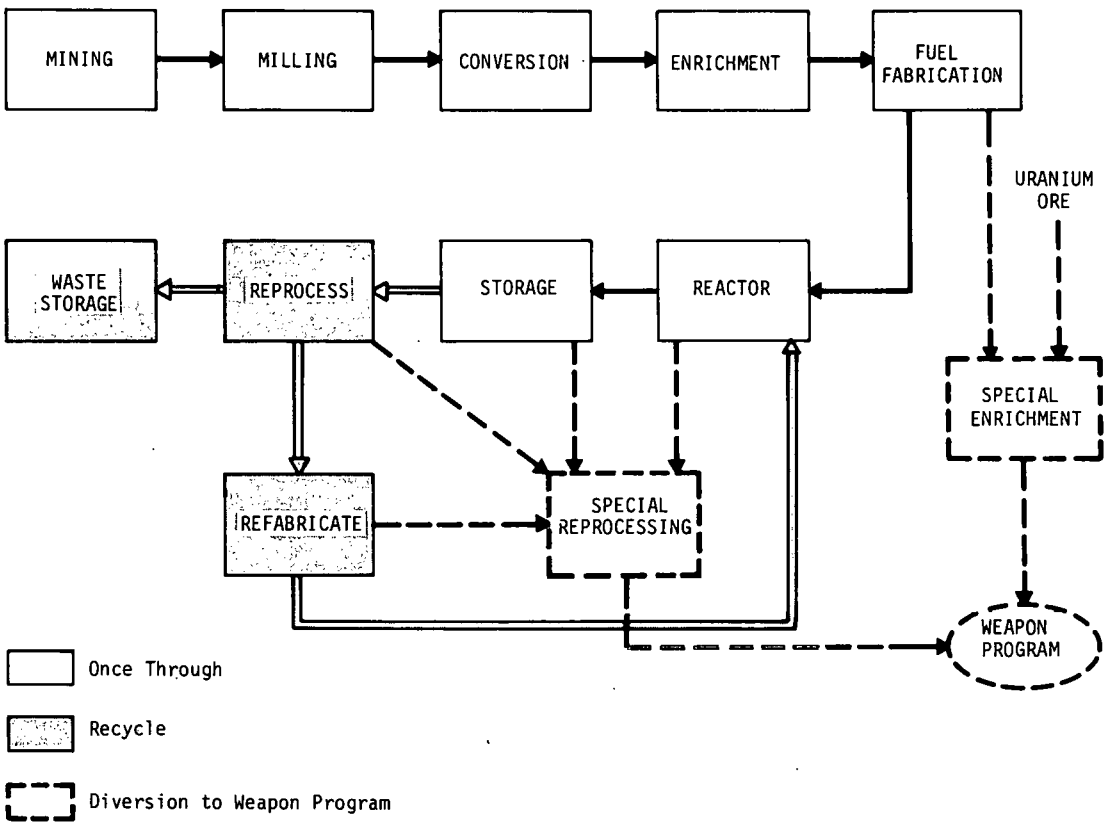

Fig. 1. The nuclear fuel cycle.

PROLIFERATION RESISTANCE

Four steps are required for proliferation -

- Preparation
- Material Acquisition
- Material Processing
- Weapons Fabrication and Testing

The proliferation resistance features in a fuel cycle include a combination of technical and nontechnical features associated with the first three tasks. The comparison of fuel cycles is intended to define features that make the diversion of fuel cycle materials for improper use more time consuming, more costly, more difficult, more visible, or more susceptible to sanctions. The features identified for comparison of fuel cycles are listed in Table I. A major emphasis is placed upon the timing features because of the belief that the U. S. and other countries can discourage a program to acquire nuclear weapons if enough time is available to develop proper deterrence. Other features such as cost and difficulty can be defined by technical comparisons of fuel cycles. The method[1] of study developed in the program is intended to be useful when analyzing proliferation by both fuel cycle and nonfuel cycle paths.

Follow-up studies are being made to determine the major factors in the decision to acquire nuclear weapons in the six weapons states. The interaction between technical barriers, institutional barriers, and political barriers is also under study. These studies are helpful in defining the scenario or strategy to be assumed. Some general assumptions must be made concerning the strategy used by a country in acquiring nuclear weapons material from the nuclear fuel cycle. Two general strategies are assumed - covert and covert/overt.

In the covert strategy, the acquisition of fissile material from the fuel cycle facilities for processing in a covert plant is attempted without assistance from the operator or use of the equipment in the recycle facilities. The covert strategy could be used either by national or subnational groups. Techniques developed under U. S. safeguards programs to prevent subnational groups from acquiring fissile materials would be effective against the covert strategy.

TABLE I.

Proliferation Resistance Attributes

Attribute	Comment
Time to acquire sufficient fissile material for purpose intended.	The time interval is measured from when the intent of the action is known.
Cost to convert fissile material to an appropriate form.	Cost is considered secondary in importance to timing consideration.
Inherent difficulty of the processes used.	Both manpower skills and technology are to be considered.
Detectability of the preparation.	Special features should be described.
Vulnerability to response by other countries	Is the country energy or economically dependent.

The covert-overt strategy, however, consists of a concerted action by the operators of the recycle plant to prepare fissile material in a form suitable for weapon use. The national decision for an action to revoke international agreements and inspection could represent a change in governmental policy even though the recycle plants were built when the country had no interest in a nuclear weapons program.

Proliferation resistance evaluations of fuel cycles are dependent upon judgment and logical choices that may not be applicable to a particular national plan to acquire nuclear weapons. Means to proliferate independent of fuel cycles (research reactors, dedicated facilities, and enrichment) will remain. All fuel cycles are to some degree susceptible to proliferation. Thus, part of the problem after defining differences in proliferation resistance will be to determine if the differences are significant, relevant, or worth the added cost.

Part of the technical barrier to proliferation is the technology to transform fissile materials used in nuclear power fuel cycles to a form useful in weapons. Weapon fabrication is not included in this discussion of facilities for alternative fuel cycles. The NASAP studies use unclassified information as needed to relate materials in fuel cycles to weapon programs. If step 4, the technical barrier of weapons technology, is sufficiently large, any difference in difficulty in the previous steps to acquire weapons materials will not be a major factor in the total four-step proliferation program.

METHOD

The NASAP program is partly complete, and no final conclusions about fuel cycles are available. But the analysis method being used can be described. A simplified diagram of the method of analysis being used in NASAP is shown in Fig. 2. Relevant data are submitted from a variety of sources to the DOE Fuel Cycle Evaluation Task Force. Separate analysis groups develop methods to evaluate the information and to define important features. Review of preliminary results both within the program management and by advisory groups leads to changes in the type of data needed or in different program emphasis.

- TECHNICAL SYSTEM OPTIONS
- INSTITUTIONAL AND NONTECHNICAL FRAMEWORKS
- STRATEGIES
- U.S. INPUT TO INFCE

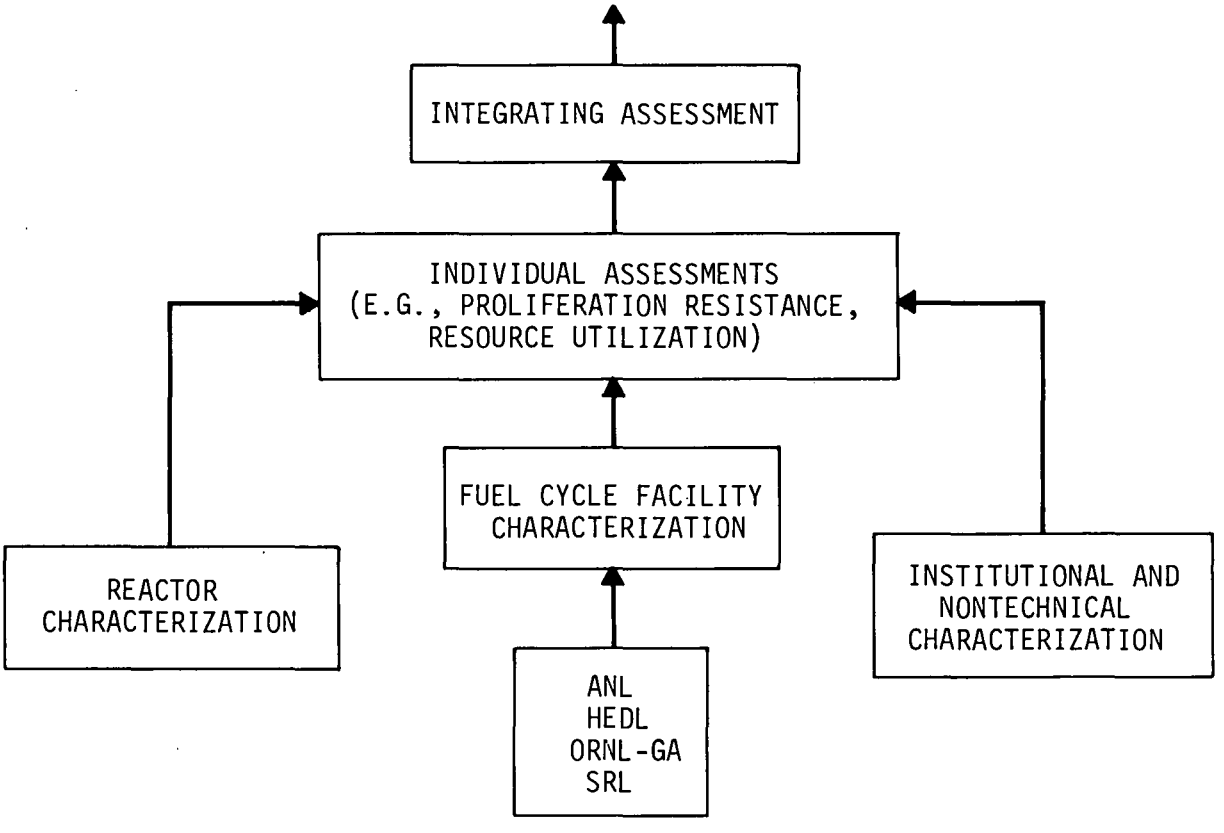

Fig. 2. NASAP work flow.

One part of the data input on alternative fuel cycles is associated with fuel cycle facilities. The necessary facilities vary with the fuel cycle, but, in general, include fuel fabrication processes, enrichment, spent fuel storage, fuel reprocessing, and waste storage. The data on fuel cycle facilities are provided by Argonne National Laboratory, Hanford Engineering Development Laboratory, Oak Ridge National Laboratory, General Atomic Corporation, and Savannah River Laboratory. Reactor design data are also specified for the fuel cycles.

The scope of the NASAP program also includes identifying institutional arrangements that contribute to increasing proliferation resistance and analyzing the feasibility of international acceptance of those arrangements. Examples of such arrangements are inspection systems, accountability systems, fuel cycle centers serving many nations, and fuel supply - spent fuel return contracts.

The NASAP also provides technical information requested by the working groups in the International Fuel Cycle Evaluation (INFCE) program. This effort is an international study of the many features of alternative fuel cycles, fuel supply, and reduced proliferation risk.

As shown in Fig. 2, the study of the fuel cycles is being done by assembling a large data base of technical information on reactor systems and on fuel cycle systems. Material quantities and costs associated with 50 GWe nuclear capacity are used for either reactor or fuel cycle facilities. Some of the fuel cycle facilities and some of the less developed reactor systems are conceptual. Costs and performance data developed for these facilities will have more uncertainty. This data base of information is necessary to make technical estimates of system performance and susceptibility to diversion of fissile material.

In addition to the technical data base, other aspects of NASAP and INFCE studies are to describe institutional and nontechnical features that could supplement the technical features that add proliferation resistance to a fuel cycle system. Two examples of institutional systems are: 1) multinational fuel cycle centers with onsite inspectors and 2) conditions in contracts by fuel supplier nations. Technical input is needed to develop effective institutional controls. A third portion of proliferation resistance outside the scope of this paper is the political constraints that can be developed to discourage proliferation.

The preliminary scoping of fuel cycle systems in NASAP included about 70 fuel cycles in the technical data base. The large number was caused by the many types of reactor concepts (PWR, HWR, LMFBR, etc.) combined with several types of fueling (^{235}U, ^{239}Pu, ^{235}U-thoria, ^{233}U, etc.), several modifications of fuel reprocessing, and energy park alternatives. Fuel cycle processes for most of these fuel cycles were identified; technology status and attractiveness of fissile material in the process were also evaluated.[2] A reduced list of about 40 systems now being evaluated in more detail is summarized in Table II. The preliminary state of development of accelearator breeders, gas core reactors, and fusion reactors precludes near-term analysis of these systems. From the standpoint of spent fuel processing and fuel fabrication, the fuel cycles can be grouped as once-through fuel cycles, converter reactors with recycle, and breeder reactors. Uranium-plutonium fueling applies to all groups. ^{233}U-thoria-^{235}U systems in the once-through mode are not feasible. The analysis of single systems must be supplemented by studies that consider interactions between current systems when introducing future systems. Finally, the recycle plant systems can be analyzed with different configurations and controls - colocated systems, single national systems with IAEA controls, multinational systems with special controls, or energy parks for onsite fuel recycle to reactors and export from the fuel recycle plant.

The studies can be simplified by considering several broad classes of fuel cycle systems and identifying unique characteristics of reactor fuels that alter process requirements within that set. One group of process steps can be used to characterize all once-through systems. Fuel recycle systems can be described as Purex systems for reprocessing uranium-plutonium fuels or Thorex systems for reprocessing thoria-uranium plutonium fuels. Some reactor features cause variations such as concentration differences in fuels (e.g., breeder or LWR fuel) or process differences in the fuel fabrication. The timing of recycle systems will affect the fuel cycle. Recycle in converters may precede and thus provide an experience base for the essential recycle portion of breeder reactor systems. Another strategy would defer reprocessing until reprocessing LWR fuel was required to initiate and expand a breeder reactor system.

TABLE II.

Systems of Interest

	Once-Through Cycles	Denatured U_{233}-Th	Pu Burners and U-233 Converters	U-Pu Recycle
LWR	X	X	X	X
SSCR	X	X	X	
HWR	X	X	X	
HTGR/Pebble Bed	X	X	X	
LWBR/HWBR		X	X	X
LMFBR/GCFR		X	X	X
MSBR		X		
Tandem Electronuclear Fuel and Power Producers (Accelerator Breeders)				
Gaseous Core Reactor				
Fusion-Fission Hybrid				

ONCE-THROUGH FUEL CYCLES

Once-through fuel cycles with terminal storage of fuel assemblies have been judged to contain desirable proliferation resistance attributes because no reprocessing facilities are included in the fuel cycle. The unirradiated reactor fuel, 3% ^{235}U, requires isotope separation facilities to be converted to weapon material. The spent reactor fuel contains approximately 0.8% ^{235}U and approximately 1% plutonium, but the fission products make recovery and separation a significant technical effort. A small recovery and separation plant is considered one possible way to obtain the plutonium; the detectability of the plant is enhanced if no commercial fuel reprocessing industry is present to mask the gaseous releases from such secret recovery facilities. Spent fuel could be returned to a supplier country. If not returned, accounting for the spent fuel assemblies at reactor sites or at terminal storage sites will depend upon long-term piece counting records and other inspection methods. While suitable in the near term, one drawback of once-through cycles is associated with resource utilization; the finite amount of uranium ore means that the size or duration of the nuclear power industry is related to the availability of ^{235}U. The distribution of uranium ore is also a factor that raises questions about the acceptance of once-through fuel cycles in all countries of the world.

To reduce ore consumption, once-through fuel cycles in LWR's could be redesigned to provide a 10 to 15% savings in uranium ore requirements if the fuel exposure can be increased from 33,000 MWD to about 46,000 MWD. A similar fuel exposure is achieved in the tandem LWR-HWR fuel cycle. One effect of the increased fuel cycle exposure on decay heat is shown in Fig. 3. If a decay period of five years is required for the nominal exposure case before encapsulation and terminal geologic storage of fuel, an additional five years would be required for the higher exposure fuel. The interim fuel storage requirement would be increased by 20% by the effects of higher exposure even though the annual fuel discharge would be reduced 40%.

The decay curve for 20,000 MWD is associated with an optimized once-through HWR system (slightly enriched). Although the fuel decay period is reduced to about 3½ years to match the specific power of current nominal LWR fuel, the higher fuel throughput in a HWR system causes the quantity of fuel to be stored prior to terminal storage to increase about

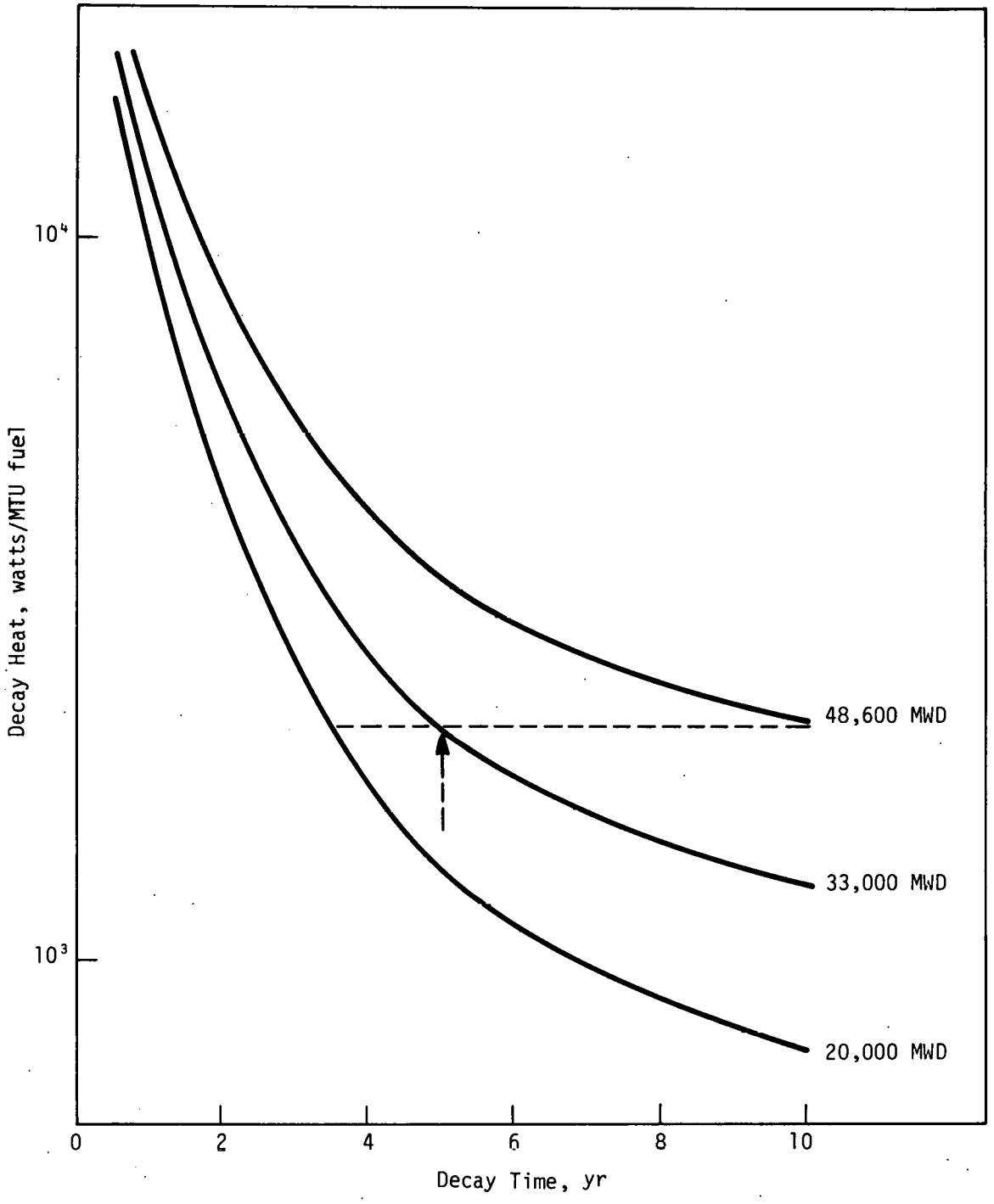

Fig. 3. Power decay of LWR-UO_2 fuel.

10%. Other papers at this meeting have addressed the current shortage of spent fuel storage. This example is cited to demonstrate that fuel cycle changes to accomplish one objective (resource utilization) or to gain a small reduction in fuel cycle cost often conflict with other nuclear issues such as waste disposal, operating margins, etc. In addition, some fuel cycle changes involve licensing hearings or added risks of fuel failure and may not be cost effective.

RECYCLE SYSTEMS

A small number of process options are required to provide recycle fuels for converter or breeder reactors. The systems that are essentially commercially developed are the Purex process for spent fuel processing and a pressed powder pellet fabrication method of preparing mixed oxide fuel. A schematic diagram of the Purex process system is shown in Fig. 4. Some technical features still under development for a Purex reprocessing system are the retention of radioactive off-gases, such as ^{85}Kr and tritium, means to implement full-scope safeguards and plutonium accountability, and optimum means of high-level waste solidification. In NASAP reviews, the product streams and inventories of plutonium solution or plutonium oxide are considered a constant temptation to proliferation. Thus, several technical methods have been proposed to increase the proliferation resistance of the Purex process - denaturing, chemical dilution or coprocessing, and enhanced radiation or spiking.

Denaturing

Denaturing implies isotopic dilution of a fissile material so that the complex technology of isotope separation would be required to produce material usable in a weapon. Denaturing is effective with uranium fuel; LWR fuel with 3% ^{235}U or ^{233}U fuels containing nonfissile ^{238}U require isotope separation. The proliferation characteristics associated with various techniques of isotope separations (gaseous diffusion, centrifuge, nozzle, laser) have been recognized as sensitive technology prior to NASAP studies. However, there is no isotope of plutonium comparable to ^{238}U.

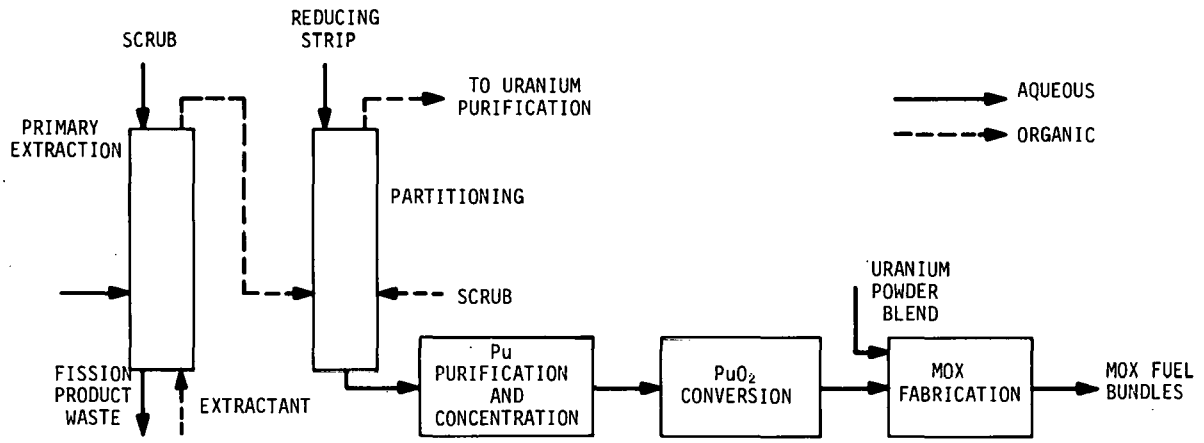

Fig. 4. Purex flow sheet.

Coprocessing

It is feasible to modify the Purex process (coprocessing) to produce a chemical mixture of uranium and plutonium and thereby provide an incremental reduction in the risk of diversion. A schematic for coprocessing by partial separation of uranium from LWR spent fuel is shown in Fig. 5. The example shown produces about 12% Pu - 88% U, so additional uranium is added to the fuel fabrication plant to produce fuel for LWR recycle. The impact on the reprocessing plant is primarily associated with the conversion process. The heavy metal throughput would be increased by a factor of 3 to 10 for the process equipment, and some developmental work will be needed for an industrial-scale operation. Accountability performance is not expected to be adversely affected by dilution with uranium. Diversion of eight times as much material would be required to give an equal amount of plutonium. However, the technical difficulty of chemically separating uranium and plutonium is not formidable on a national scale. Coprocessing could be implemented in first-generation reprocessing plants because of the advanced state of development and because of the small input on proven fuel fabrication techniques or other issues of recycle plants (waste treatment, occupational exposure). The cost impact of coprocessing is judged to be minor. The coprocessed fuel is expected to have improved dissolvability characteristics that would be beneficial in plant scrap recycle or further reprocessing after reactor service.

Spiking

Providing a source of intense radiation in plutonium fuels has been analyzed as a method of safeguarding plutonium from terrorist diversion. With sufficient radiation, the recycle fuels become more similar to spent fuel and thus are presumed to offer proliferation resistance that is comparable to once-through fuel cycles.

A survey of the many alternatives for spiking plutonium fuels is summarized in Table III. The effectiveness of alternative spiking methods can be evaluated after the objective or criteria are specified. The proposed criteria used in NASAP is to provide a sufficient radiation field to ensure that any attempts to purify the fissile material must be performed remotely. For significant amounts of fuel such as one assembly, a radiation field in the range of 1000 R/hr seems appropriate;

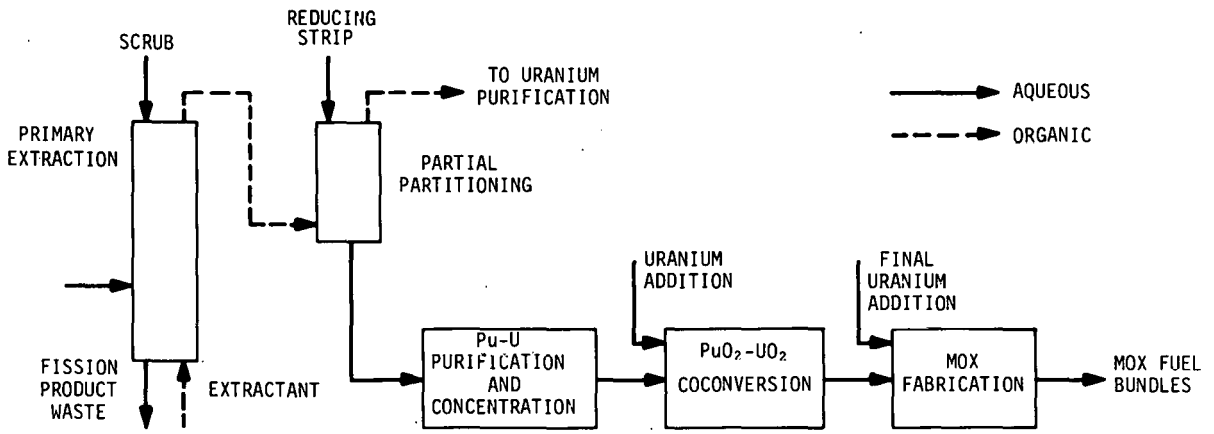

Fig. 5. Coprocessing flow sheet.

TABLE III.

Methods of Spiking Plutonium

Method	Spiking Affects Fuel Fabrication	Means to Defeat	Radiation Level
With Fission Products			
Incomplete Removal	Yes	Recycle, adjust process chemistry.	Can be high.
Selective Partition	Yes	Avoid add-back.	High - depends on isotope.
Irradiate Fuel After Fabrication	No	Stop irradiation.	Adjustable, but short term will decay faster.
Add Kr to Fuel Rods	Minor	Stop addition.	Low - not a deterrent, but detectable.
With ^{60}Co			
Mix with Pu Fuel	Yes	Separate	Adjustable but long term (5-year half life).
Attach Source to Fuel Assembly at Pu Fabrication Plant	No	Remove source	
With ^{238}Pu or ^{236}Pu	No	Isotope separation.	Minor (~few R/hr).

for smaller masses, a radiation rate of 100 R/hr per kg seems consistent. The radiation intensity should persist for at least one year and preferably two years to allow for shipping and storage at a reactor before use. Monitoring of process parameters will be needed to ensure the continued presence of radiation for either partial removal of fission products or adding a stable source of activity.

The significant features of candidates for spiking, fission product isotopes and cobalt, are compared in Table IV. While ruthenium and zirconium are the most straightforward fission products to coextract with plutonium, only ruthenium has an appropriate decay characteristic. Reprocessing plants can probably be developed to mix fission products with plutonium fuels, but the characteristics of many fission product species are not compatible with subsequent fuel fabrication processes. Ruthenium characteristics complicate any coconversion step because the volatile oxides formed would not survive subsequent fuel fabrication steps. Cesium has an attractive half-life for spiking, but separating a pure cesium fraction involves complex processes. Cerium is most compatible with fuel fabrication but does not provide sufficient radiation. Adding 1 to 2% of mixed fission products by entrainment would meet the radiation requirement but would need development to ensure that the method was dependable and not easily rendered ineffective. Cobalt provides a more reliable source of radiation than fission products and may be more compatible with developed fuel fabrication processes. All spiking techniques seriously complicate fuel fabrication technology, fuel inspection, and accountability. Remote fabrication methods or new lower temperature processes to retard evolution of activity such as sol-gel sphere-pak technology could probably be developed for recycle fuels, but years of reactor tests of fuels would be required to qualify the fuel for industrial use. The economics of once-through versus recycle of spiked fuel in LWR might not justify the effort until ore prices increased significantly.

To avoid the interaction with fuel fabrication methods, two different spiking methods are under study. Preirradiation of fuel assemblies has a minimum effect on fuel fabrication, but as shown in Fig. 6, even an exposure of 10 full power days (fuel burnup of about 1%) barely meets the radiation criteria for one year. Cobalt sources attached to MOX fuel rods can be provided to meet the radiation criteria. The addition of radiokrypton to MOX fuel rods is a different spiking technique; the concept is to discourage diversion by increasing

TABLE IV.

Candidates for Fission-Product Spiking of Plutonium

Isotope	Radiation from Isotope in MOX Assembly R/hr @ 3 Ft		Recovery Fraction, %	Half Life, Yr	Volatility in MOX Pellet Fabrication
	1 Year Decay	1½ Year Decay			
^{95}Zr	5,000	750	~100	0.2	No.
^{106}Ru	40,000	28,000	10	1	Probably yes.
^{134}Cs } ^{137}Cs }	50,000	40,000	5	2 } 30 }	Yes.
^{144}Ce	2,000	1,200	~90	0.8	No.
Mixed Fission Products (1%)	2,000	1,400	1	-	Some.
^{60}Co	-	-	-	5	Probably no.

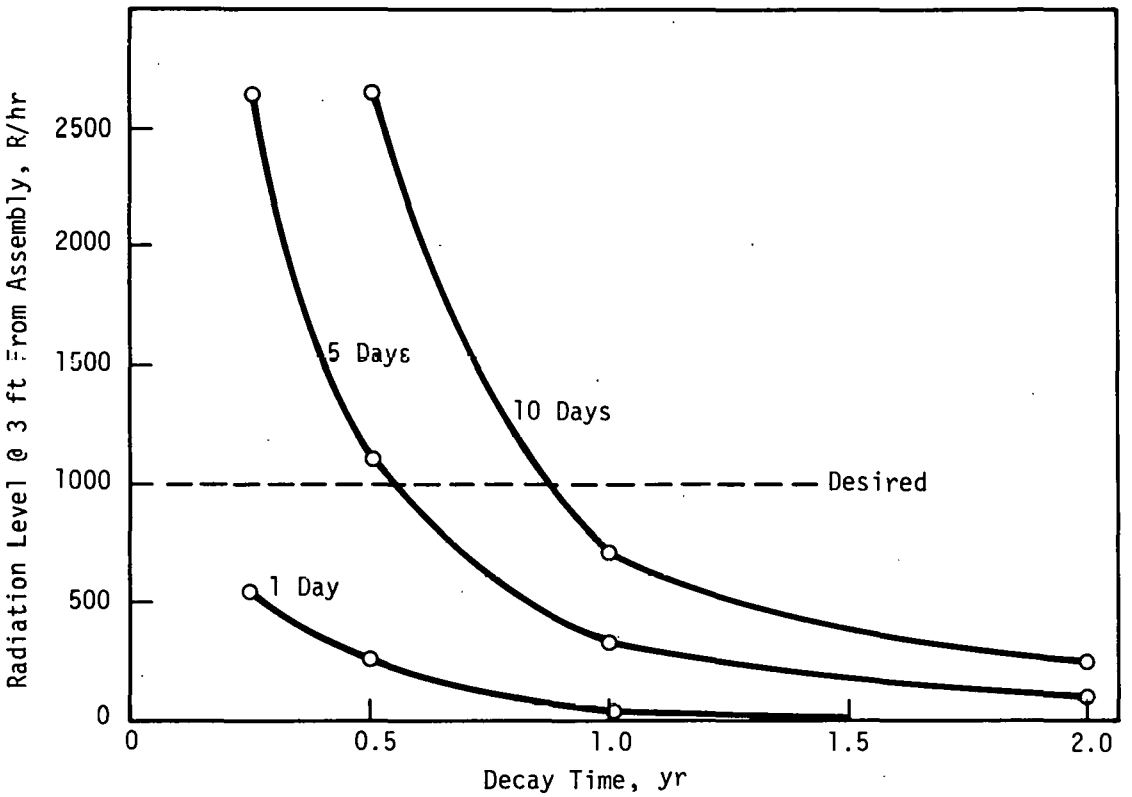

Fig. 6. Irradiation of LWR-MOX assemblies.

the detectability of an attempt to reprocess fuel and separate the plutonium after it leaves the recycle plant. Preirradiation, cobalt sources, or krypton addition rely on international control of desired actions in the recycle plant.

OTHER PROCESSES

Reprocessing of spent thorium-uranium fuel will probably require a modified Thorex process as indicated in Fig. 7. In the diagram, the ^{233}U product is assumed not to exceed 12%. Some of the ^{238}U added to denatured ^{233}U will produce plutonium[a] that can be routed to waste in the first cycle. The plutonium can also be recovered for producing more ^{233}U for export from a secure energy center. A major development program is needed for these unique features of reprocessing thorium fuels:

- Dissolution of thoria fuel requires fluoride ion; therefore, materials of construction must resist corrosion from fluoride. The fluoride dissolution also dissolves some of the Zircaloy hulls and this could inhibit thoria dissolution; the added solids complicate the waste disposal technology.
- The waste solidification systems under development are not compatible with fluoride ion.
- The basic chemistry flowsheet to separate the three products needs development of optimum conditions.
- The off-gas from dissolution includes radon; techniques to entrap radon require development. Tritium release from thorium fuels may require different technology than from uranium fuels.
- The conversion of the ^{233}U stream (and perhaps the thorium stream) to oxide for recycle requires shielded remote equipment. The sol-gel sphere-pak process, a leading candidate for fabricating thorium-uranium fuels, can replace the conversion step.
- The lower solubility characteristics of thorium require larger volumes of solvent, larger equipment to reprocess fuel at the same rate as for uranium fuel.

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a. The amount of plutonium produced in LWR with denatured ^{233}U fuel is about one-third the plutonium produced with 3% ^{235}U fuel.

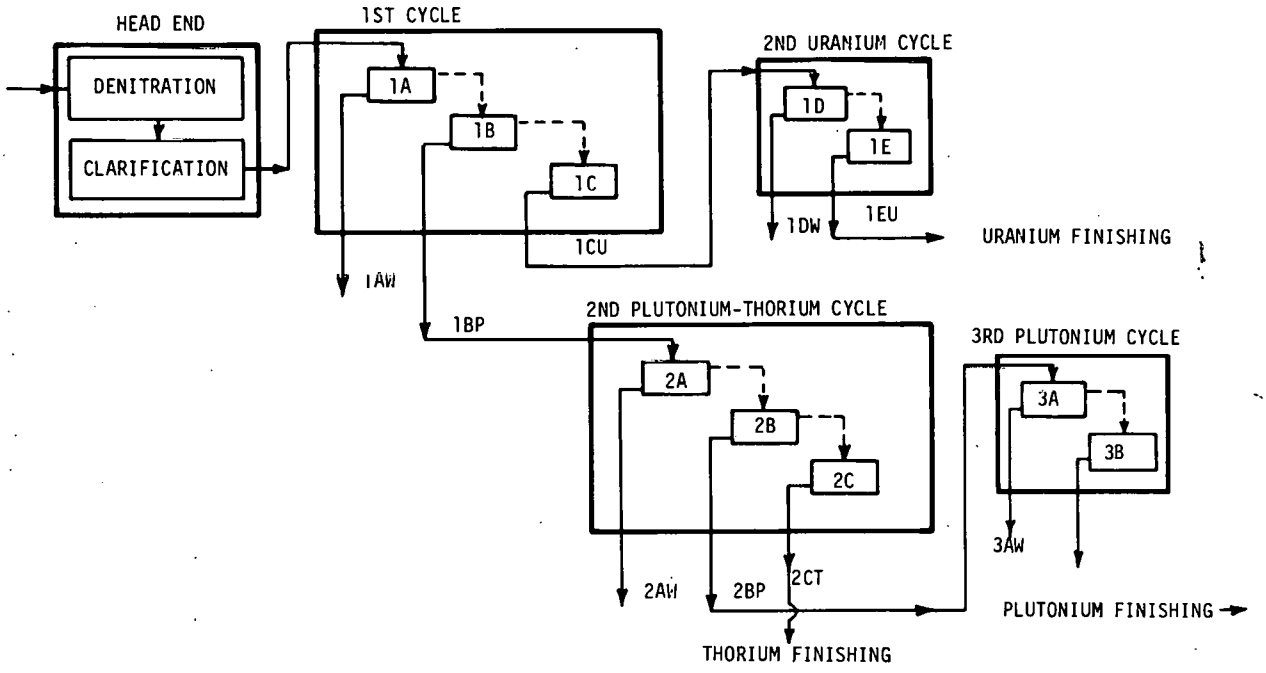

Fig. 7. Thorex flowsheet.

Nonaqueous processing of uranium-plutonium fuels is also included in the NASAP because the inherent inflexibility and lower decontamination achievable is judged to be preferable to the aqueous processes. Long-term development is needed for such systems.

INSTITUTIONAL SYSTEMS

The proliferation resistance introduced by technical revisions can be supplemented by institutional systems. International monitoring and/or control of regional fuel cycle centers described in Reference 3 would be a major supplement to any fuel cycle strategy. Onsite inspection or other instrumentation techniques to monitor recycle facilities will be a major means to implement proliferation resistant technologies. The control of processing in a fuel cycle center might be sufficiently well developed that remote monitoring could substitute for some onsite inspection and accountability. Processes suitably controlled within such a center can focus on diversion resistance against outsiders rather than inherently inflexible processes intended not to attract host country takeover. The economies of scale provide benefits to those participating in the recycle center.

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