

**THE ADVANCED FUEL CYCLE INITIATIVE OF THE U.S. DEPARTMENT OF ENERGY:
DEVELOPMENT OF SEPARATIONS TECHNOLOGIES**

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ABSTRACT

The Advanced Fuel Cycle Initiative of the U.S. Department of Energy is intended to address the issue of high-level nuclear waste disposal in the future, by providing a closed fuel cycle technology that can support the current fleet of commercial power reactors as well as those reactors that are deployed in the future. It is estimated that the capacity of the Yucca Mountain repository could be increased by a factor of 40-60 times by such processing, through a reduction in the volume and heat load of the waste to be emplaced. This would ensure the sustainability of an expanded nuclear energy supply system in the United States and could delay the need for a second repository until well into the next century.

INTRODUCTION

Because of its environmental advantages and abundant fuel resource base, nuclear power is expected to be an important source of energy in the United States in the future. Projections for the growth of nuclear power in the U.S. vary over a wide range, with generating capacity in 2050 between 175 and 500 GWe (cf. about 100 GWe today). The legislated capacity of the Yucca Mountain geologic repository (70,000 tons) will be reached in 2015 as the spent fuel inventory grows from the operation of the current fleet of commercial power reactors. If we define the 70,000-ton figure as a "Yucca Mountain Equivalent" (YME), then the various growth scenarios would require disposal capacity of 2.4 – 4.3 YMEs by 2050, if the U.S. continues with the once-through LWR fuel cycle. Absent legislative relief, the difficult and costly process of siting, licensing and constructing a geologic repository for high-level nuclear waste in the U.S. would be repeated a number of times over the next 40-50 years. Even if the actual repository capacity is increased by further exploration, the once-through fuel cycle would require at least one additional repository with three or four decades.

The Advanced Fuel Cycle Initiative of the U.S. Department of Energy is intended to provide an alternative approach to high-level nuclear waste disposal in the future, by providing a closed fuel cycle technology that can support the current fleet of commercial power reactors as well as those reactors that are deployed in the future. Chemical separations technology development is an important part of this program, and the development effort is directed toward separations processes that facilitate the removal of those constituents of spent fuel that contribute most to the heat load and waste volume imposed on the disposal of high-level waste in the repository. Processes are being developed that will (1) remove over 90% of the uranium in sufficiently pure form that it can be disposed as a low-level waste or re-enriched for recycle to LWRs; (2) remove over 99% of the cesium and strontium present in spent fuel, thereby eliminating the short-term heat load; and (3) separate the transuranic elements (plutonium, neptunium, americium and curium) for storage or for recycle to LWRs or future advanced reactors for fissioning, thereby eliminating the long-term heat load. It is estimated that the capacity of the Yucca Mountain repository could be increased, in terms of equivalent tons of spent nuclear fuel, by a factor of 40-60 times by such processing. This would ensure the sustainability of an expanded nuclear energy supply system in the United States and delay the need for a second repository until well into the next century.

Both advanced aqueous and pyrochemical processing methods are being developed under the scope of the Advanced Fuel Cycle Initiative. The aqueous process, known as UREX+, is at an advanced stage of technological maturity and could conceivably be deployed in the 2020-2025 time period. It represents a minor but significant departure from the processes presently utilized in commercial reprocessing plants in France and the United Kingdom. The pyrochemical processing methods are directed principally toward the treatment of spent fuels arising from the operation of third- and fourth-generation reactor plants, and their development benefits greatly from the experience being gained in the processing of spent fuel from the EBR-II fast reactor.

LONG-RANGE STRATEGY

Projections of the long-range future of nuclear power in the United States are complicated by the existence of many unquantifiable variables, and range from a nuclear phase-out to no growth through 2025 [1] to sustained growth to 300 GWe [2] or to 500 GWE [3] by 2050. Taking a moderate growth scenario for the purpose of developing a strategy upon which to base technology development decisions, we conclude that light water reactors and advanced light water reactors will constitute the bulk of the U.S. nuclear generating capacity for at least another 50 years. Separations for the purpose of waste management will be important until it becomes practical to recycle separated plutonium (and perhaps neptunium) as mixed oxide fuel in thermal spectrum reactors. Subsequently, it may be possible to reduce the long-term heat load imposed on the repository by burning separated minor actinides (americium and curium) in dedicated fast-spectrum burner reactors. Finally, a transition to Generation IV reactor systems will occur, with full closure of the nuclear fuel cycle. These distinct periods in the evolution of the U.S. advanced fuel cycle strategy can be categorized as a series of phases. Phase 0 is the current once-through cycle with deployed commercial light water reactors. Phase 1 is a transitional waste management phase, in which commercial spent fuel would be processed so as to facilitate the disposal of high-level nuclear wastes in a manner that extends the effective capacity of the geologic repository. Phase 2 would see the recycle of fissile materials in advanced light water reactors as mixed oxide fuel. Phase 3 involves the use of dedicated fast-spectrum burner reactors to destroy the minor actinide elements, Am and Cm, for the purpose of reducing the long-term heat load on the repository. Phase 4 represents the transition to advanced Generation IV reactors with closed fuel cycles. An illustration of this strategy is shown in Fig. 1.

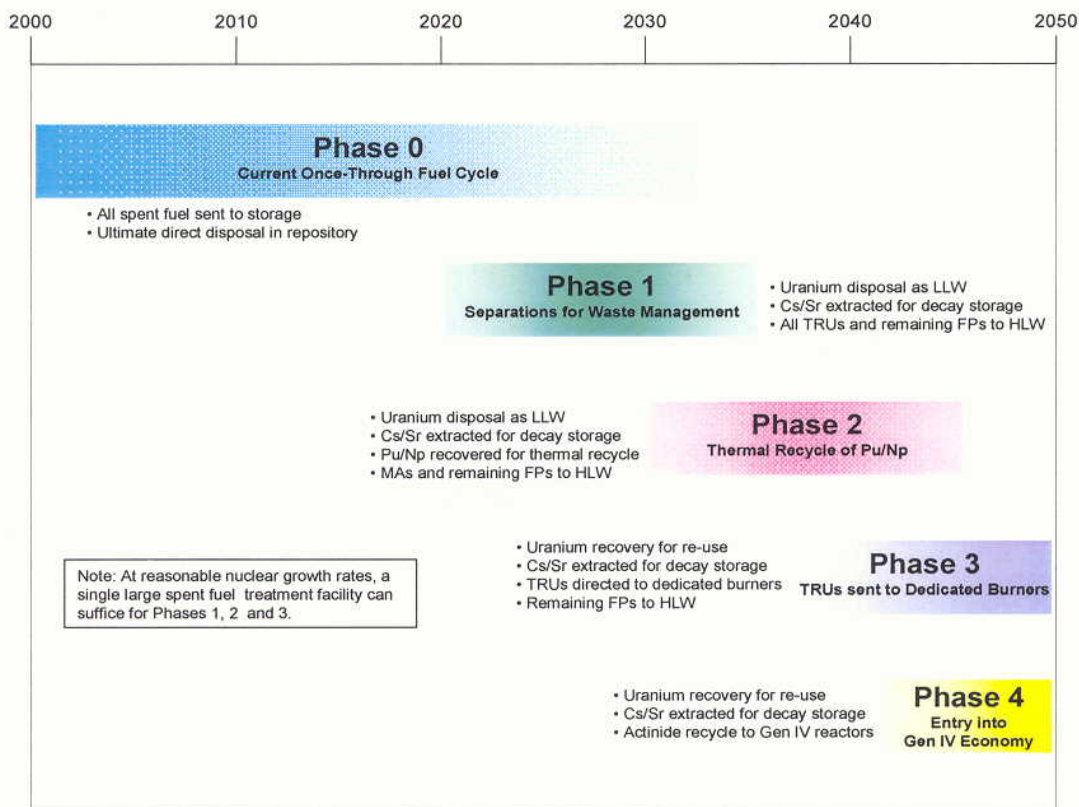


Fig. 1. Phased strategy for the implementation of advanced nuclear fuel cycles in the United States. Times are only approximate and durations of the phases, or the extent of their overlap, are subject to change.

This phased strategy is considered to be consistent with the realities of a transition to advanced nuclear power systems. Utilities in the U.S. are likely to place new orders for advanced LWRs in the near term, capitalizing on the great strides made in increased operational efficiency and productivity with the current fleet of commercial reactors. If there are to be no new orders of any reactor type, then the need for an advanced fuel cycle technology is greatly diminished; but even if the future nuclear electricity generating capacity were only to remain constant at the current level of about 100 GWe through 2050, there is still likely to be a benefit to nuclear waste management from spent fuel treatment. Repository benefits, of course, are open to debate and certainly subject to changes in repository performance analyses and criteria. Sufficient time is available, however, to resolve those issues before it becomes necessary to commit to construction of a large spent fuel treatment plant.

ADVANCED AQUEOUS PROCESSING

Aqueous reprocessing of light water reactor (LWR) spent fuel is currently practiced in France, the United Kingdom, and Russia. Japan will soon begin operation of a commercial facility, the Rokkasho Reprocessing Plant. The scale of these processing plants is on the order of that which would be required to accommodate the current rate of generation of spent LWR fuel in the United States, and the technologies employed are technologically mature. These factors were instrumental in the selection of advanced aqueous processing as the reference method for development as part of the Advanced Fuel Cycle Initiative. Aqueous processing also affords the flexibility in process configuration required by the multi-phase strategy: the same plant that is used for Phase 1 can be easily reconfigured to support Phases 2, 3 and possibly 4.

Development of advanced aqueous processing methods for the treatment of spent LWR fuel is proceeding on a schedule that would see the selection of process flowsheets for Phases 1-3 by the end of U.S. fiscal year 2008. Process development is being guided by the preliminary separations criteria shown in Table I.

Table I Preliminary Separations Criteria for Use in Process Evaluation and Selection

Criterion	Thermal Recycle, Fertile Fuel	Fast Reactor Recycle of U and All TRUs
Recovery Efficiency		
Uranium	90%	90%
Pu/Np	99%	99%
Am/Cm	99.5%	99.5%
Cs/Sr	97%	97%
Tc, I	95%	95%
Purification Requirements		
Uranium	99.9%, 99.97%*	99%
Pu/Np	99.5%	97%
Am/Cm	TBD	97%
Cs/Sr	<100 nCi TRU/g **	<100 nCi TRU/g **

*Higher purity requirement is for the option of re-enrichment of the uranium stream

** Purification as necessary to meet 10CFR61.55 requirements for Class C waste

Phase 1 Separations

The Phase 1 strategy is based on spent fuel processing for waste management purposes. Therefore, the process requirements are for separation of pure uranium (which could be disposed as a low-level waste), extraction of cesium and strontium in pure form (for decay storage and eventual disposal as low-level waste), efficient recovery of technetium and iodine (for incorporation into durable waste forms), and recovery of transuranic elements together with lanthanide fission products for repository disposal as a self-protecting waste form. Because the TRU waste is a long-term decay heat generator, consideration is being given to storage of this material in retrievable form so that it could be recovered before repository ventilation is terminated and further processed to recover the transuranics for recycle as fuel for future fast spectrum reactors. A schematic flow diagram for Phase 1 processing is shown in Fig. 2. After conventional dissolution of the spent fuel in nitric acid, the clarified dissolver solution is sent to a solvent extraction process called UREX. The UREX process uses tri-butyl phosphate (TBP) as the extractant, with acetohydroxamic acid added in the scrub stage to reduce plutonium to the unextractable Pu(III) state. Uranium and technetium are co-extracted, and the technetium is then stripped at high acidity to yield a pure uranium stream and a pure technetium stream. The transuranics and the remaining fission products are in the UREX raffinate, which is then directed to the Cs/Sr extraction step. The present reference process for recovery of cesium and strontium is the CCD/PEG process (chlorinated cobalt dicarbollide/polyethylene glycol); the use of alternate extractants such as calixarenes is also being studied. After removal of the cesium and strontium, the raffinate is denitrated to produce a mixture of transuranic oxides and fission product oxides. Alternatively, the raffinate could be processed by the TRUEX process to recover the transuranics together with the lanthanide fission products, with the remaining fission products going to the waste stream. In either case, the transuranics and accompanying fission products are to be encapsulated and stored in the repository. The nature of the storage form is yet to be decided; it could take the form of a cermet fuel rod, utilizing part of the zircaloy cladding hull stream to provide the matrix material, for eventual retrieval and use as a fuel or target in a fast reactor. The TRU product could also be encapsulated directly, with the proviso that further processing would be necessary before the

fissionable material would be recycled. Regardless of the path taken, the Phase 1 processing scheme appears to have significant benefit to repository operation. The effective increase in repository capacity, if all LWR spent fuel were to be processed prior to waste emplacement, could be as much as a factor of 60.

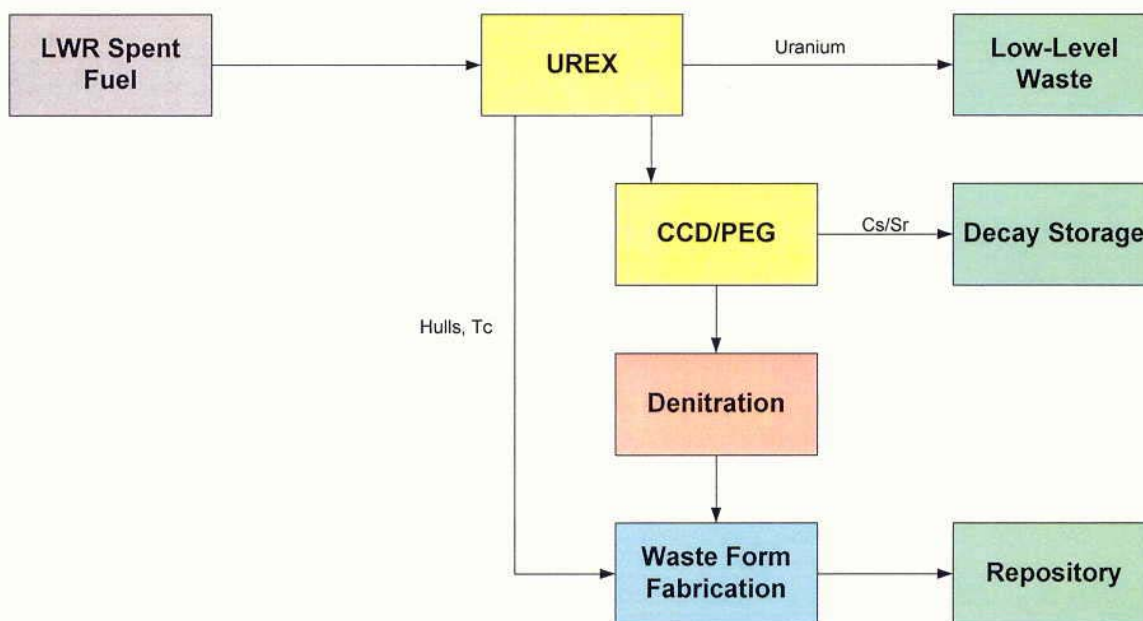


Fig. 2 Phase 1 processing of LWR spent fuel. Iodine recovery is not shown for purposes of simplification.

The elements of the Phase 1 process flowsheet have been demonstrated at laboratory scale with actual spent LWR fuel, using the CCD/PEG process for cesium and strontium extraction [4]. All process criteria were satisfied, in most cases exceeded by a wide margin. An engineering-scale demonstration is planned for the near future.

Phase 2 Separations

Phase 2 adds the extraction of plutonium (and perhaps neptunium) for recycle to thermal reactors. At least 30 of the existing 103 commercial nuclear power plants in the U.S. are capable of burning mixed-oxide fuel, and a transition period is anticipated over which time the recycle fuel could be qualified and incentives established for utilities to accept such fuel. Modification of the Phase 1 separations processes to achieve the Pu/Np extraction step are rather simple, with one possibility shown in Fig. 3. Here, a TBP-based extraction step would be inserted after the Cs/Sr extraction step. An alternative to this process is a co-decontamination process [5], in which the initial separation would be of (1) uranium, (2) technetium, and (3) plutonium/neptunium. The subsequent extraction of Cs/Sr could be with either the CCD/PEG process or an alternative such as a calixarene-based process. Once again, all elements of both process options have been successfully demonstrated at laboratory scale.

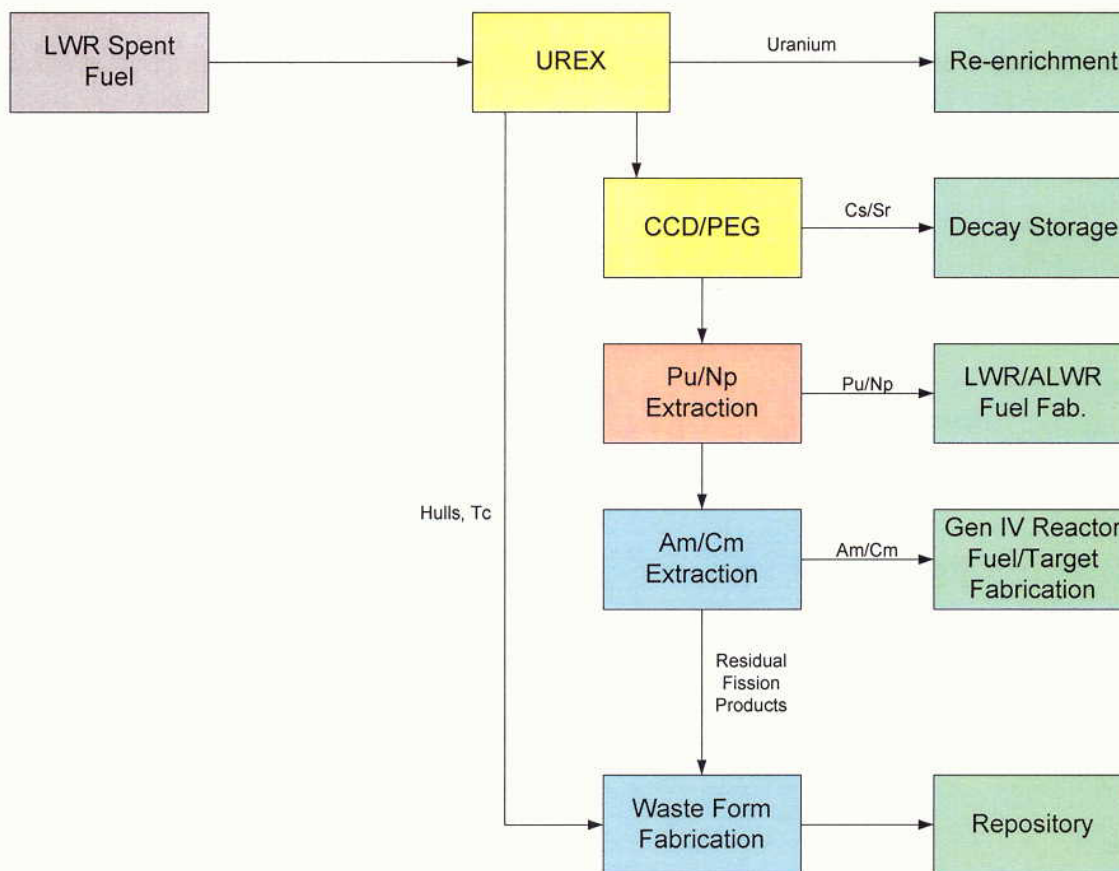


Fig. 3 Phase 2 processing of LWR spent fuel. Iodine recovery is not shown for purposes of simplification.

The minor actinides remaining in the waste stream are imagined to be incorporated in temporary storage forms as is the case in Phase 1. Given the possibility that the storage of these materials may in fact become permanent, it is most likely that the choice of encapsulation method will tend toward the more durable form.

Phase 3 Separations

In Phase 3 of the advanced fuel cycle, minor actinides are recovered for burning in fast spectrum reactor systems. The schematic flow diagram for this phase is shown in Fig. 4. In this case, considerable development work remains to optimize the process for separation of the americium and curium from fission products, especially the +3 lanthanides that are difficult to separate from the +3 minor actinides. An extensive testing program is underway in both the U.S. and Europe, with the aim of developing the best process for Am/Cm separation. Two-step processes (separation of Am/Cm/lanthanides from the balance of the fission products, followed by separation of Am/Cm from the lanthanides) and one-step processes are being evaluated. The DIAMEX-SANEX process has been tested with some success in various laboratories, and recent U.S. tests of the CYANEX-301[®] process with spent fuel have yielded excellent results. [4]

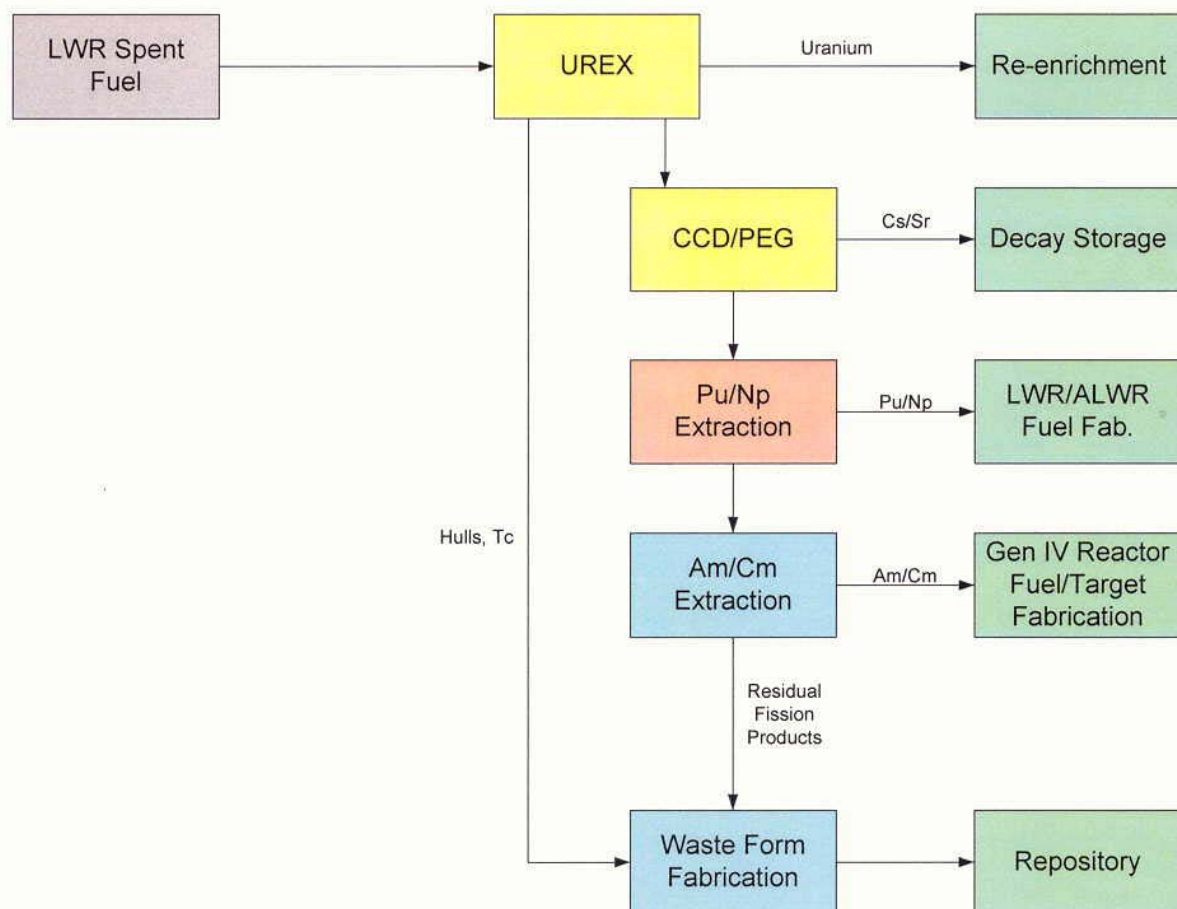


Fig. 4 Phase 3 processing of LWR spent fuel. Iodine recovery is not shown for purposes of simplification.

The residual fission products from Phase 3 processing would be placed in a ceramic waste form for repository disposal. Because the remaining fission products are comparatively benign (the only heat generator being Eu-154, with a half-life of 8.6y), the fission product loading of this waste form can be quite high, leading to a very small volume of high-level waste for disposal.

PYROCHEMICAL PROCESSING

Fuel systems for Generation IV reactors, with the possible exception of the Supercritical Water Reactor, represent a significant departure from the commercial LWR oxide fuel. Many of the fuel types that are foreseen for these reactors are intuitively not compatible with aqueous processing of the sort discussed above, and include coated-particle fuels, inert matrix fuels (ceramic-ceramic and ceramic-metal), metal alloy fuels, mixed nitride fuels (e.g., AnN/ZrN, where An is actinide), and carbide fuels. Phase 4 separations processes thus must consider the application of processing technologies other than aqueous. The general class of pyrochemical processes [6] offers some distinct advantages in treating the variety of Generation IV fuels.

Electrorefining has been used for over four years for the conditioning of spent fuel from the EBR-II reactor. In this process, the irradiated metallic fuel is chopped and anodically dissolved in molten LiCl-KCl salt. Uranium is electrotransported to a metallic cathode, and the transuranics are left in the salt together with the active metal fission products for eventual incorporation in a ceramic waste form. Noble

metal fission products (including Tc) are melted together with the stainless steel cladding hulls to produce a metallic waste form. The addition of a transuranic recovery step to this process would make it applicable to the Phase 4 processing of metallic and nitride fuels. Tests of TRU recovery methods are currently in progress in the U.S. and elsewhere. Addition of an electrochemical reduction head-end step would make the process applicable to oxide fuels, both dispersion type and inert matrix fuels.

Development of pyrochemical processing technologies in the U.S. program has been limited to date to work on metal alloy and nitride fuels for fast reactors. Process concept development for the pyrochemical processing of coated-particle fuels is now underway, and work with carbide fuel will begin soon. The conduct of extensive experimental programs, other than that involving the conditioning of EBR-II fuel, is being deferred until the Generation IV fuel types are better defined.

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