

A Practical Approach to a Closed Nuclear Fuel Cycle and Sustained Nuclear Energy-12383*

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ABSTRACT

Recent systems analysis studies at Oak Ridge National Laboratory (ORNL) have shown that sufficient information is available from previous research and development (R&D), industrial experience, and current studies to make rational decisions on a practical approach to a closed nuclear fuel cycle in the United States. These studies show that a near-term decision is needed to recycle used nuclear fuel (UNF) in the United States, to encourage public recognition that a practical solution to disposal of nuclear energy wastes, primarily UNF, is achievable, and to ensure a focus on essential near-term actions and future R&D. Recognition of the importance of time factors is essential, including the multi-decade time period required to implement industrial-scale fuel recycle at the capacity needed, and the effects of radioactive decay on proliferation resistance, recycling complexity, radioactive emissions, and high-level-waste storage, disposal form development, and eventual emplacement in a geologic repository. Analysis of time factors led to identification of the benefits of processing older fuel and an “optimum decay storage time.” Further benefits of focused R&D can ensure more complete recycling of UNF components and minimize wastes requiring disposal. Analysis of recycling costs and nonproliferation requirements, which are often cited as reasons for delaying a decision to recycle, shows that (1) the differences in costs of nuclear energy with open or closed fuel cycles are insignificant and (2) nonproliferation requirements can be met by a combination of “safeguards-by-design” colocation of back-end fuel cycle facilities, and applied engineered safeguards and monitoring. The study shows why different methods of separating and recycling used fuel components do not have a significant effect on nonproliferation requirements and can be selected on other bases, such as process efficiency, maturity, and cost-effectiveness. Finally, the study concludes that continued storage of UNF without a decision to recycle is not a solution to the problem of nuclear waste disposal, but can be a deterrent to public confidence in nuclear energy.

INTRODUCTION

Recent systems studies have been made at Oak Ridge National Laboratory on how to achieve a practical solution to used nuclear fuel (UNF) management and disposal with the overall goal of enabling sustained nuclear energy. Unlike other renowned and recent studies, such as those of the Blue Ribbon Commission on America’s Nuclear Future [1] and the Massachusetts Institute of Technology report on *The Future of the Nuclear Fuel Cycle* [2], our conclusion is that a near-term decision to recycle UNF in the United States is needed, primarily to encourage public

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recognition that a practical solution is achievable and to ensure a focus on essential near-term actions and future research and development [3].

Our conclusion, again unlike those of the more prominent studies, recognizes that continued storage is not a practical solution to the problem of UNF disposal. Moreover, continued storage for an indefinitely long time period can lead to problems with future fuel handling and transportation, to increased nonproliferation security and safeguards due to decay of the “self-protective” radiation barrier, and to the continued buildup and accumulation at multiple sites of UNF and depleted uranium (DU) from enrichment plants, which are regarded currently as “nuclear wastes.” As a result, public confidence in nuclear energy will be degraded, if that option is taken.

Findings of the Blue Ribbon Commission studies showed that full deployment of a centralized interim storage option will cost billions of dollars. In addition, finding an acceptable away-from-reactor location for a centralized interim storage facility would likely encounter similar social and political barriers as the proposed repository at Yucca Mountain, Nevada. However, if a near-term decision to recycle UNF is made, a centralized away-from-reactor interim storage facility would be established at the Recycle Plant location, at less overall costs and with more offers of job availability and other economic incentives to make the location more socially and politically acceptable.

IMPORTANCE OF TIME FACTORS

Our study recognized the importance of several time factors as reasons for a near-term decision to implement UNF component recycling in the United States [4]. Worldwide use of nuclear energy is continuing, even after the accident at Fukushima, and rapid growth is occurring in China, India, and other countries. It is only logical to recognize that at some future time, the availability of low-cost natural uranium will decline and there will be a future need for breeder reactors and industrial-scale UNF component recycling to utilize the tremendous potential energy in fertile materials.

INVENTORY GROWTH

Figure 1 illustrates the generic growth of UNF and fissile plutonium with increasing time and shows that the growth will not be curtailed until industrial-scale fuel recycle is implemented with an annual capacity equal to the annual UNF generation rate. Currently, the U.S. commercial nuclear industry inventory includes ~69,000 MT of UNF heavy metal with a growth rate of ~2200 MT/year and ~500 MT of plutonium with growth rate of ~20 MT/year. In addition, the inventory includes ~600,000 MT of DU from enrichment operations with a growth rate of ~20,000 MT/year.

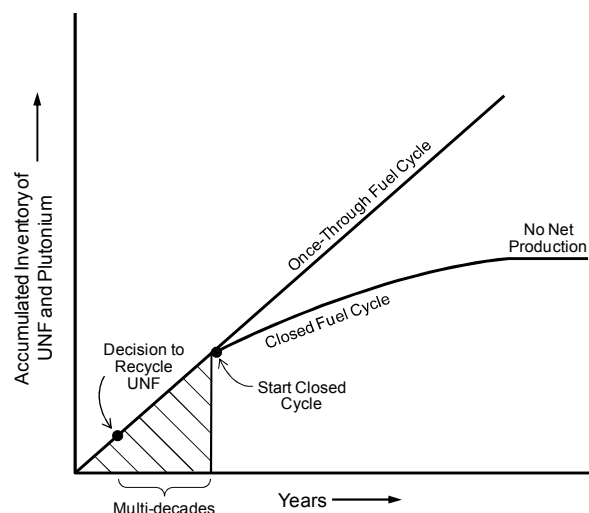


Fig. 1. Generic diagram showing accumulation of UNF and plutonium with time.

TIME TO IMPLEMENT INDUSTRIAL-SCALE UNF RECYCLING CAPACITY

A very important time factor must be recognized when considering when to begin UNF treatment. Based on worldwide experience, deployment of industrial-scale recycling is a multi-decade process and the first plant will take 15 to 20 years for design and construction. Furthermore, subsequent expansion of plant capacity to equal the annual rate of generation of used fuels will likely take another 20 or more years to accomplish. While this is occurring, the inventory of used fuels requiring storage will continue to grow until the treatment capacity equals the rate of generation. To provide perspective, a conservative growth scenario in nuclear power generation is shown in Table I.

Table I. Scenario for Generation and Treatment of Used Fuel.

	2012	2020	2030	2040	2050	2060
Number of operating reactors	104	108	116	124	132	136
Event	Decision to recycle UNF		1st plant begins operation	2nd plant begins operation	3rd plant begins operation (fully closed fuel cycle)	
Treatment capacity (MT/year)	0	0	1,000	2,000	3,000	3,000
UNF generation rate (MT/year)	2,200	2,250	2,300	2,700	2,900	3,000
UNF Storage capacity required (MT)	69,000	87,000	110,000	126,000	134,000	134,000

After a decision is made to recycle UNF components, at least four decades of time, and likely longer, will be required to deploy annual recycle capacity equal to the annual UNF generation rate, and thus, to curtail the continuation of increasing inventory and storage requirements.

RADIOACTIVE TIME FACTORS

Radioactive decay of key components of the UNF during continued storage can have significant time-dependent effects on (1) proliferation resistance, (2) recycling complexity and radioactive emissions (if a decision is made to recycle UNF components), (3) storage and waste form development, and (4) eventual repository emplacement of the high-level waste (HLW).

Proliferation resistance time factors

Many people consider the radiation emanating from UNF to be the primary safeguard for the once-through, direct disposal method. However, as illustrated in Fig. 2(a) by the plot of radiation emission from a pressurized-water-reactor UNF assembly versus time, the so-called “radiation barrier” decays at an exponential rate, thus making used fuel older than several decades more vulnerable to diversion and theft. Figure 2(b) shows that for up to 5–10 years after reactor discharge, radiation barrier contributors are short-lived fission products, intermediate-lived cesium–strontium (half-lives: ~30 years), and long-lived transuranium element actinide elements (TRU). From 5 to 70 years, the major contributor to the radiation barrier is the gamma radiation from cesium. After 70 to 100 years and beyond, the long-lived TRU elements are predominant, but they do not provide a sufficient radiation barrier, as indicated in the Fig. 2 plots for periods longer than 70–100 years.

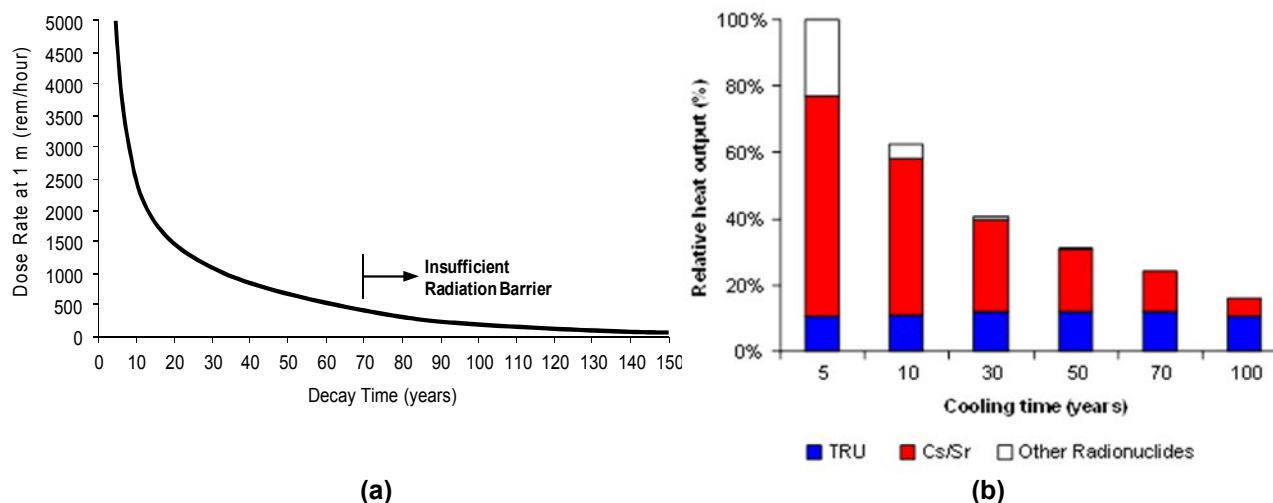


Fig. 2(a). Exponential decay of radiation barrier and (b) radiation barrier contributors.

The vulnerability of an insufficient radiation barrier can be eliminated if fuel recycle is begun before the radiation barrier has decreased to a susceptible level, because re-irradiation of the recycled plutonium will restore the effective radiation barrier. From a nonproliferation safeguards standpoint alone, recycling UNF at a relatively short time period after discharge of the UNF from reactors (~2 to 10 years) would provide the largest radiation barrier before

separation of the components of UNF during reprocessing operations. However, our studies have determined that substantial advantages to the overall recycling and re-irradiation process (i.e., partitioning and transmutation of TRU actinide elements) can be gained by waiting for at least 30 years after reactor discharge to begin the recycling process. Thus, the conclusion was made that the “optimum time” for recycling UNF is between 30 and 70 years after discharge from reactors.

In many of the “partial separation” reprocessing methods being proposed, such as the “DUPIC (Direct Use of Spent PWR fuel in CANDU)” process developed in Korea and Canada or other similar “modified open cycle” processes, the primary radiation source, radiocesium, can be easily separated from the plutonium because their chemical valences are widely different (cesium is monovalent, while plutonium is either tri-, tetra-, or hexavalent) and because cesium can be removed by volatilization at temperatures above ~800°C. Radiocesium could be added back to recycle fuel to provide a radioactive barrier during fuel fabrication and transportation, but that action would cause these operations to be more difficult. Considering the relative ease of separation of cesium and the operational difficulties that would be added, other means of protection are better choices.

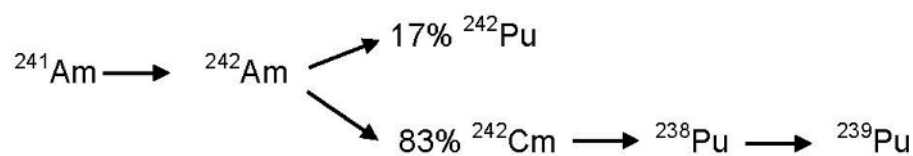
UNF recycle time factors—reduced complexity and radioactive emissions

In contrast to the increasing proliferation risks with long storage time, our systems analysis studies have shown that major benefits can be obtained by allowing the UNF assemblies to remain in safe storage for >30 years before treatment, followed by treatment of the oldest fuels first. One of the major benefits is that the 30+ year storage period allows a substantial decay of ^{241}Pu (14.3 year half-life) to ^{241}Am and thereby, for either thermal or fast spectrum irradiations, a significant alteration of the actinide transmutation pathway to produce predominantly the lighter isotopes of plutonium (^{238}Pu , ^{239}Pu) instead of the heavier actinides (^{243}Am , ^{244}Cm , etc.) which require shielded facilities and remote handling. The alteration of the neutron capture pathway

From



to



enables transmutation of long-lived transuranium element (TRU) actinides to be accomplished in existing thermal spectrum reactors without excessive buildup of heavier actinides and without waiting on the development, licensing, and deployment of future fast reactors.

Also during the 30+ year storage period prior to reprocessing separations, the radioactivity and decay heat generation of several key fission product and actinide radionuclides decrease

substantially (Fig. 2), such that some gaseous waste components (^{85}Kr and possibly tritium) can be released at levels below regulatory limits, and the solid fission product wastes containing ^{137}Cs and ^{90}Sr require decay storage for a shorter additional time period after separation and before emplacement in a geologic repository.

In addition, the separations process can be simplified and less costly. For example, the need for extra purification cycles for the uranium and uranium-plutonium-(neptunium) products is greatly diminished by decay of fission product isotopes, primarily ^{106}Ru and ^{154}Eu . Also, the need for separating curium from americium is lessened by decay of ^{244}Cm and in-growth of ^{241}Am from decay of ^{241}Pu . Furthermore, after the reprocessing separations have been completed, the further in-growth of ^{241}Am from decay of ^{241}Pu is reduced, and therefore, the radiation dose to recycle fuel fabrication operations is lower.

Optimum decay storage time

Overall, as indicated in Fig. 3(a), there appears to be an "optimum decay storage time" of 30 to 70 years before processing to recycle UNF components. This optimum time is necessary to maximize benefits in safety, environmental effects, and cost reduction while maintaining acceptable proliferation resistance, taking into consideration that, after ~ 70 years, the "radiation barrier" will become insufficient for self-protection. Re-irradiation of the actinides during fuel recycle, whether accomplished in current thermal-spectrum reactors, predominantly light-water reactors, or in future fast reactors, restores the fission product components and the effective radiation barrier that enables safe storage for another ~ 70 years.

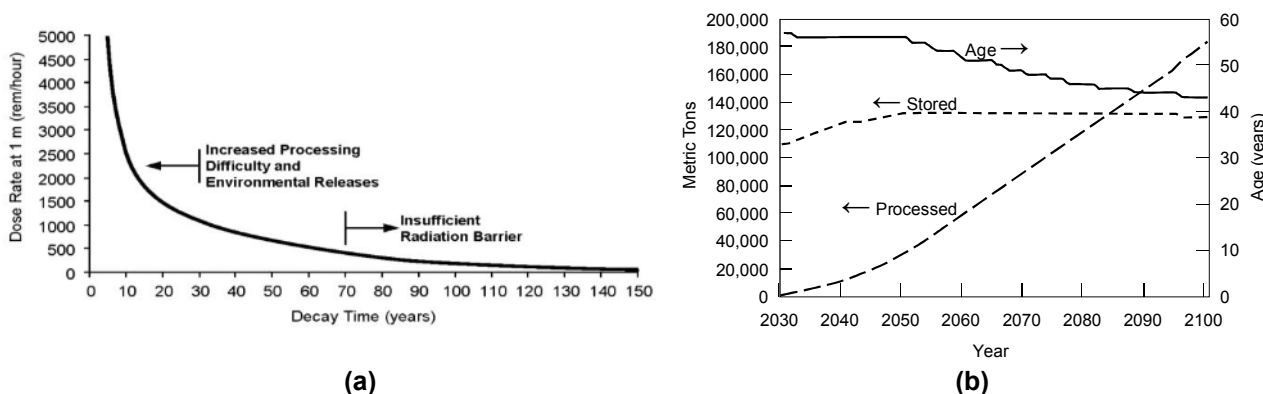


Fig. 3(a). Westinghouse PWR fuel assembly dose rate and (b) scenario for treatment of used fuel.

Based on the advantages identified, treatment of all used fuels in the U.S. inventory can be planned using a basis of treating the oldest fuel first. Currently, the United States has accumulated an inventory of used fuels that includes the earliest discharged fuel, now ~ 40 years old. Even with a near-term decision to recycle UNF, initial industrial-scale implementation will require 15–20 years, and the oldest fuel will be 55–60 years old, still within the "optimum decay storage time." Direct disposal of the older UNF to a geologic repository would not be beneficial and is not necessary. Reprocessing and recycle of UNF will allow the residual HLW to be properly packaged for disposal, and the geologic repository can be designed without the need for future retrieval.

Figure 3(b) illustrates, graphically, the storage and processing data of the conservative growth scenario described in Table I and also shows the age of the UNF processed, which never drops below 40 years old throughout the rest of the century. As indicated in Table I, a decision to initiate the multi-decade effort to design and build fuel recycle facilities must begin in the next few years to enable the schedule shown in the scenario. Any delay to beginning the deployment of recycling or any increase in the rate of expansion of reactor capacity will increase the average age of the UNF that is recycled.

If future UNF recycle treatment is done within the “optimum period of 30 to 70 years,” surface storage of the HLW for another 30 to 70 years (a total of ~100 years decay storage of the fission products) can provide benefits toward more specific advanced waste form development and eventual waste disposal benefits. Long-term storage of HLW canisters containing vitrified waste currently is being implemented at U.S. defense and former commercial sites, as well as in foreign commercial reprocessing plants. Surface storage of such wastes may be required for ~50 years or longer, and no problems are anticipated.

Development of an advanced compact HLW storage form without added diluents, such as glass formers that add mass and heat limitations may be beneficial. The optimum storage form may be a calcined solid waste, such as is often produced as the intermediate to glass waste, or a cermet waste form that provides enhanced heat transfer. Following the surface storage period, further treatment of the HLW to remove in-grown stable decay daughters prior to encapsulation of the HLW in a disposal form can provide further mass reduction and eliminate deleterious chemical effects on the waste disposal form.

MORE COMPLETE RECYCLING OF UNF COMPONENTS

More complete recycling of UNF components can be implemented in future advanced fuel cycle treatment facilities than is done in current industrial practice. Recovery and recycle of over 90% of the components—the uranium, the zirconium contained in the cladding, and most or all of the TRU actinides—could be accomplished with focused development and application of currently known technologies.

The UNF components also include other potentially valuable chemical elements that might be recovered and reused within the nuclear industry if R&D is directed toward those components. The potentially recoverable components include the xenon gas, the highly valuable noble metals, and some of the lanthanide elements.

COMPARATIVE COST OF FUEL CYCLES

A cost analysis was made and, in summary, the results (Table II) show comparative nuclear electricity generation cost components for open (direct disposal) and closed (recycle) fuel cycles. As shown, the high capital and financing costs of reactors dominate all other cost effects. This is true even though the capital and financing costs of fuel cycle facilities are higher than those of a single reactor, because the fuel-cycle-facility life cycle costs are spread over a large fleet of reactors. Thus, the fuel cycle costs are <15% of the overall costs for any option shown, and the

comparative cost differences of direct disposal and advanced recycle are insignificant in terms of the total cost of nuclear energy.

Future needs for breeding fissile materials from depleted uranium wastes and thorium resources will require more expensive advanced reactor and fuel designs. This indicates that focused research, development, and demonstration are needed to meet these necessities.

NONPROLIFERATION UNCERTAINTIES—A MAJOR BARRIER TO FUEL RECYCLE

To clarify uncertainty issues, our approach was to identify what components of UNF are nonproliferation concerns [5]. Figure 4 shows the mass composition of the average inventory of used fuels in the United States, as of July 2002. The average burn up is 33 GWd/MT (71% from pressurized water reactors and 29% from boiling water reactors), and the calculated decay period is 40 years. Currently produced higher-burnup UNF contains about 1% TRU elements and 5% fission products.

Table II. Fuel Cycle Cost Contributions to the Levelized Unit Electricity Cost.

Fuel cycle type	UOX LWR direct disposal	UOX/MOX LWR current recycle (only)	LWR advanced recycle (U, TRUs, Zr, and some fission products)	Advanced reactors breeder recycle (U, Pu) drivers DU blankets
Percent of used fuel assembly mass in waste	100	99	5	5–10
Comparable levelized costs, mills/kWh ^{a,b}				
U ore/U enrichment/UOX fabrication/UOX credits ^c	4.3	3.9	3.5	0.1
Reactors ^d	49.5	49.5	49.5	59.0
Used fuel dry storage	0.3	0.0	0.0	0.0
Recycling ^e	0.0	3.4	3.9	5.0
Waste disposal	1.6	1.0	0.3	1.5
Total	55.7	57.8	57.2	65.6
Fuel cycle component of above costs	6.2	8.3	7.7	6.6

^aUnits of mills/kWhr are equivalent to \$/MWhr.

^bFuel transportation and safeguards costs are similar for all fuel cycles.

^cCredit for UOX assemblies displaced by MOX or RU-derived UOX (RUOX) taken in this row. Costs to prepare the MOX or RUOX assemblies are in the “recycling” row (U₃O₈ to UF₆ conversion also included).

^dAssumes that capital cost for *n*th-of-a-kind (NOAK) sodium-cooled fast reactor is 20% higher than for NOAK LWR.

^eRecycle cost includes (U, Pu) refabrication cost.

Material balance for LWR based on 51,000 MW(th)-day/MTHM PWR utilizing ~4.3% ²³⁵U LEU (equilibrium reloads).

Material balance for sodium fast reactor (SFR) “breeder” based on Japanese data. Unit fuel cycle costs from INL/EXT-09-15254 [6].

Used fuel assembly mass in the first row includes heavy metal (fuel meat) plus cladding and hardware. SFR fuel is assumed to be stainless steel clad.

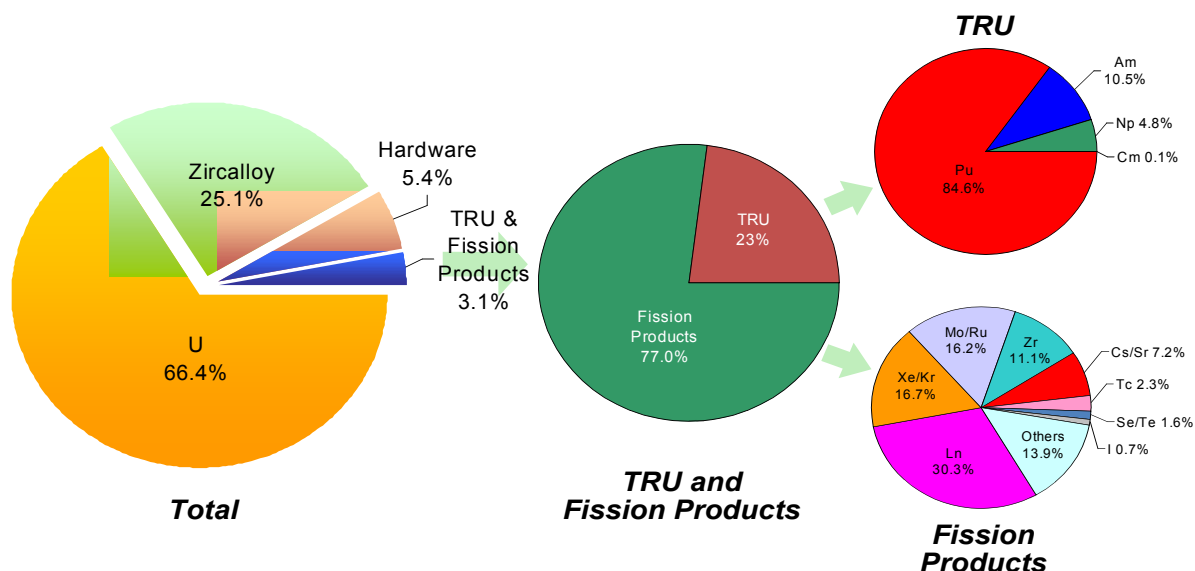


Fig. 4. Mass composition of used fuel components.

Three fissile isotopes are of concern to nonproliferation: ^{235}U , ^{239}Pu , and ^{241}Pu . Even though the mass of ^{235}U is about the same as the sum of ^{239}Pu and ^{241}Pu , the ^{235}U concentration in the uranium element is sufficiently low and would require isotopic enrichment to become a nonproliferation and safeguards concern. However, the fissile plutonium isotopes are in the range of 50 to 80%, depending on burnup, *making the plutonium element a nonproliferation safeguards concern* because chemical separation processes, which are relatively less difficult than isotopic enrichment processes, can be used to produce separated plutonium.

A variety of well-known separation methods used by the chemical and hydrometallurgical industries could be used to separate plutonium. These separation methods include fluoride volatility, solvent extraction, ion exchange, and precipitation processes, or combinations of these. Furthermore, the scale of operation is much larger for commercial UNF reprocessing operations than for a smaller-scale separations process that could and should be of greater concern for undeclared weapons production activities. In any case, the conclusion was that plutonium has no intrinsic safeguards. Even though unseparated or partially separated plutonium in UNF may have a lower “attractiveness level,” the plutonium remains to be separable. So, the further conclusion, which is similar to that of the “attractiveness level” studies by Bathke et al.[7], is that physical protection and other proliferation resistance methods are necessary to prevent diversion.

Perhaps the most significant finding with regard to nonproliferation uncertainties is that physical protection and proliferation resistance methods are necessary for unseparated UNF or partially separated plutonium as it is for separated plutonium. Thus, continued storage or direct disposal of UNF must be protected because recovery and separation of the plutonium will always be possible.

For commercial reprocessing and UNF component recycle facility, engineered safeguards and physical protection can be applied to provide a “defense-in-depth” to nonproliferation requirements. The safeguards-by-design concept can be used to co-locate and integrate the used

fuel treatment facilities within the same “recycle plant” (Fig. 5). This method can ensure that (1) the used fuel storage, component separation (reprocessing), and recycle fuel fabrication facilities are co-located within the physically protected recycle plant; (2) the fissile SNM (i.e., plutonium) enters and leaves the recycle plant only in the form of large, heavy, easily accountable fuel assemblies; (3) effective continuous monitoring and surveillance of waste shipments and personnel exiting the plant are maintained; and (4) use of near-real-time monitoring and accounting of the plutonium location and movement are utilized to the extent technologically possible.

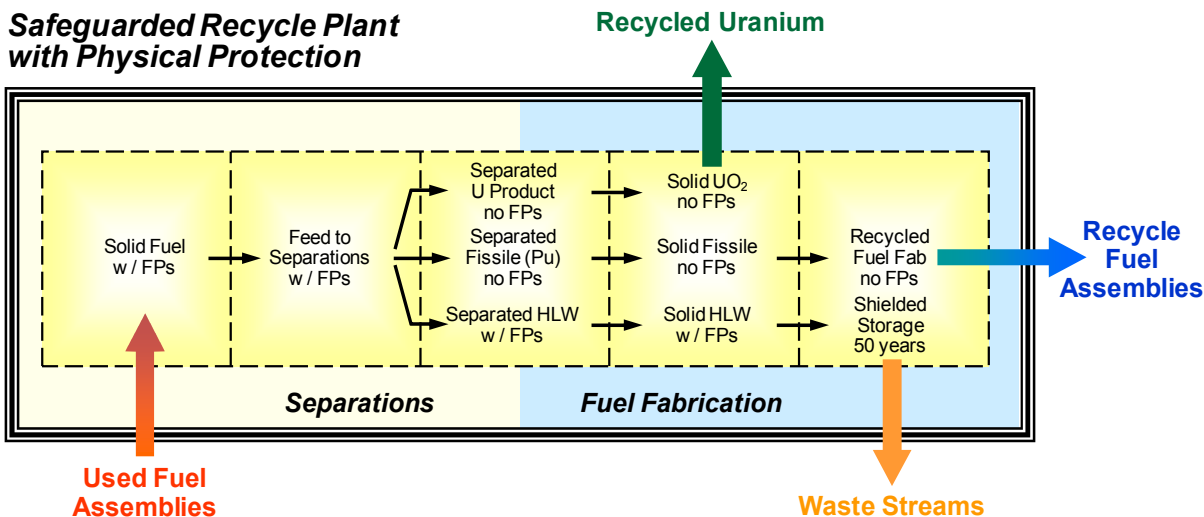


Fig. 5. Safeguards-By-Design—colocation and integration of used fuel treatment facilities.

By having the facilities co-located and integrated, the inventory of plutonium “in-process” can be minimized and no large quantity of partially separated plutonium will be accumulated within or transported outside the recycle plant, except in the form of recycle fuel assemblies.

Defense-in-depth engineered safeguards include (1) limiting receipts and shipments of plutonium-containing nuclear fuel to large, heavy, easily accountable fuel assemblies; (2) locating separations and recycle fuel production equipment in inaccessible, contained, and monitored areas within the physically protected recycle plant; and (3) applying continuous effective portal monitoring of waste shipments and operating personnel leaving the recycle plant. When such safeguards are in place, the vulnerability to theft and diversion by “insiders” or subnational groups can be virtually eliminated.

Partially separated plutonium is acceptable for use in recycle fuel, as long as the neutron-poison fission products (heavy lanthanides) have been removed. Since “attractiveness” studies have shown that part of the uranium component in UNF can be left with the plutonium to lower the attractiveness level, that method of separation can be utilized. Process equipment can be designed to produce the desired concentrations of recycled uranium and plutonium and can be designed and placed in relatively inaccessible locations within the recycle plant to prevent modification.

A variety of chemical separations processes can be chosen to provide the necessary “co-processing” and “co-conversion” of uranium and plutonium for recycle fuels. Therefore, the processes chosen can be selected on the basis of process efficiency, maturity, and cost-effectiveness, and not on a basis of “greater proliferation resistance.”

By these means, the engineered safeguards and practical safeguards-by-design decisions can provide adequate proliferation resistance to meet nonproliferation requirements. Thus, the nonproliferation uncertainties barrier to a decision to recycle commercial UNF in the United States can be resolved.

SUMMARY

In summary, our studies have shown, in contrast to findings of the more prominent studies, that today we do have sufficient knowledge to make informed choices for the values and essential methods of UNF recycling, based on previous research, industrial experience, and current analyses. We have shown the significant importance of time factors, including the benefits of an optimum decay storage time on deploying effective nonproliferation safeguards, enabling reduced recycling complexity and environmental emissions, and optimizing waste management and disposal. Together with the multi-decade time required to implement industrial-scale UNF recycle at the capacity needed to match generation rate, our conclusion is that a near-term decision to recycle as many UNF components as possible is vitally needed.

Further indecision and procrastination can lead to a loss of public confidence and favorable perception of nuclear energy. With no near-term decision, the path forward for UNF disposal will remain uncertain, with many diverse technologies being considered and no possible focus on a practical solution to the problem. However, a near-term decision to recycle UNF fuel and to take advantage of processing UNF and surface storing HLW, together with development and incorporation of more-complete recycling of UNF components, can provide the focus needed for a practical solution to the problem of nuclear waste disposal.

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