Advanced Nuclear Fuel Cycles – Main Challenges and Strategic Choices

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REPORT SUMMARY

This report presents the results of a critical review of the technological challenges to the growth of nuclear energy, emerging advanced technologies that would have to be deployed, and fuel cycle strategies that could conceivably involve interim storage, plutonium recycling in thermal and fast reactors, reprocessed uranium recycling, and transmutation of minor actinide elements and fission products before eventual disposal of residual wastes.

Background

Current civil uses of nuclear power in the United States are based on a once-through fuel cycle involving the irradiation of low-enriched uranium fuel in light-water reactors and the subsequent storage and eventual disposal of the spent fuel. Past and more recent findings published by the Electric Power Research Institute (EPRI), the U.S. electric utility industry's Advanced Reactor Corporation, the National Academy of Sciences, and the Massachusetts Institute of Technology have been in general agreement with regard to support for the present U.S. policy that relies on the once-through fuel cycle because of its simplicity, economic advantages, and nonproliferation benefits. However, there is also broad agreement that research and demonstration should be conducted on selected topics to support the safe and cost-effective implementation of a rational fuel cycle policy involving the recycling of fissile materials in fast neutron spectrum reactors.

Objectives

- To provide a systematic review of the technological challenges associated with advanced nuclear fuel cycles
- To evaluate promising and viable paths forward for implementation in the twenty-first century

Approach

The research team first reviewed the main technological challenges to the growth of nuclear energy that would be required for significantly contributing to the supply of carbon-free energy. These challenges include: (a) sustainability of natural resources, (b) economic competitiveness, (c) waste management, and (d) nonproliferation. Second, the research team briefly reviewed the principal technologies that are at the core of advanced fuel cycles: (a) fast neutron spectrum reactors, (b) spent fuel treatment options (reprocessing), and (c) waste disposal options. Third, the fuel cycles in France and in the United States were examined in order to document the impact of different national policy and security contexts that have guided their implementation. Finally, the research team evaluated several paths forward that appear to be industrially feasible for stepwise deployment in the twenty-first century.

Results

New strategies may be required to balance the needs for 1) sustainability—particularly the shift to a plutonium economy and reduction in high level waste (HLW) burden on permanent geologic repositories, 2) operational efficiencies, and 3) diversion resistance of plutonium-based fuel cycles. Such strategies may rely on interim storage of spent fuel as well as on partitioning and transmutation of plutonium and minor actinides before final HLW disposal in a permanent geologic repository. Many options can be envisioned; however, many of those represent dramatic changes compared to the current situation and are not likely to lend themselves to industrial-scale deployment. An evolutionary and progressive pathway is likely to be more realistic than a revolutionary pathway that attempts to simultaneously solve all real or perceived fuel cycle issues with advanced technologies. The externalities of nuclear energy, such as waste generation and proliferation risks, have to be addressed in a safe, but reasonable, way. Thus, advocating transmutation of all the transuranics and fission products or making nuclear materials so unattractive that they are practically unusable in the fuel cycle itself do not represent realistic options.

EPRI Perspective

Continued and expanded use of nuclear power may be predicated on improved economics and sustainability, especially when it is assumed that applications of nuclear technology may expand beyond production of electricity, such as production of hydrogen for industrial and transportation applications. These developments may require evolution of the present nuclear fuel cycle. A rational fuel cycle strategy must provide a flexible framework that can adapt to changes in technology and national policy and integrate, in due time, more advanced technologies. The technical and economic conditions for the breakthrough of these advanced technologies are challenging because they encompass not only reactors, but also dedicated reprocessing, fuel fabrication, and waste disposal facilities. These elements are closely interdependent, and their performance will have to be consistent. Their competitiveness may be anticipated on paper, but it will have to be proven by experience.

Keywords

Fast reactor Nuclear fuel cycle Recycling Reprocessing Waste disposal

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1 THE CHALLENGES OF A SUSTAINABLE NUCLEAR FUEL CYCLE

With the growth of energy consumption and the challenge of global warming, nuclear energy is likely to expand. But this growth must be sustainable, which raises four main challenges: 1) sustainability of natural resources, 2) economic competitiveness, 3) waste management, and 4) non-proliferation. Safety is also a major concern - certainly the most important for the nuclear industry - but is one that must be addressed regardless of the specific fuel cycle. Therefore, safety should be considered as an intrinsic requirement common to all options.



Figure 1-1

The Four Main Challenges of a Sustainable Nuclear Fuel Cycle

1.1 Nuclear Natural Resource Sustainability

Fissile nuclides are necessary to generate nuclear power, and in nature, only U-235 is present in significant amounts (representing 0.71% of the natural uranium resources). Two others fissile nuclides, Pu-239 and U-233, can be created through neutron capture by the fertile nuclides U-238 and Th-232, which make up most the uranium (99.28%) and thorium (100%) natural

The Challenges of a Sustainable Nuclear Fuel Cycle

resources. As a consequence, U-235, U-238 and Th-232 constitute the natural resources available for any nuclear fuel cycle.

Today, almost all the commercial reactors operate with a thermal neutron spectrum (Heavy Water Reactors, Pressurized Light Water Reactors, and Boiling Light Water Reactors). Consequently, they use the energy content of U-235, and to a very limited extent, the energy content of U-238, mostly through the creation of Pu-239 that is generated during reactor operation. Therefore, the current nuclear power sector depends on the availability of U-235 contained in uranium that can cost-effectively be recovered from the earth's natural resources.

1.1.1 Reserves of Natural Uranium

In 2006, the worldwide nuclear fleet of 370 GWe generated 2,675 TWhe of electricity (which implies a capacity factor of 83%) and consumed 66,500 tU¹⁻². So, on average, one ton of uranium provides 40 GWhe of electricity. This translates to approximately 180 tU/year required for a 1-GWe reactor with a capacity factor of 83%. Because nuclear power usually operates in a baseload mode, the relation between installed capacity and uranium requirement is relatively constant, which simplifies and improves the reliability of forecasting uranium sustainability.

Every two years, the IAEA and NEA compile and publish data and estimates on international uranium resources, production and demand in a report commonly referred to as the "Red Book." The resources are classified as Identified Resources (sub-divided into Reasonably Assured Resources and Inferred Resources) and Undiscovered Resources (divided into Prognosticated Resources and Speculative Resources). The results used in this report are from the 2008 edition³, as presented below in Tables 1-1 and 1-2 in ktU (original units) and years of current consumption, respectively. These estimates indicate sufficient uranium resources available at reasonable prices to ensure fueling of nuclear power plants in the short and medium terms, i.e., for the next 50 – 100 years.

The real concern in this timeframe is the production capacity of natural uranium. In 2006, world uranium production (36,603 tU) met only 60% of world reactor requirements. Secondary sources, such as excess inventories, down-blending of Highly Enriched Uranium (HEU) from the dismantling of nuclear warheads, re-enrichment of depleted uranium or use of reprocessed uranium, provided the other 40%. But these secondary sources are expected to decline in the future⁴. Therefore, uranium mine production will have to increase significantly, especially in

¹ "Uranium 2007: Resources, Production and Demand," A Joint Report by the OECD Nuclear Energy Agency and the International Atomic Energy Agency, OECD 2008, NEA No. 6345, also referred to as the "Red Book"

² The units are metric tons of uranium, abbreviated as tU or MTU

³ The 2010 edition "Uranium 2009: Resources, Production and Demand," A Joint Report by the OECD Nuclear Energy Agency and the International Atomic Energy Agency, OECD 2010, NEA No. 6891 was published in July 2010

⁴ Decline of secondary sources are expected after 2013 according to the 2008 "Red Book" (page 86)

light of continued global expansion of nuclear power. As a consequence, significant volatility in uranium prices, resulting from market forces, is likely to continue (Figure 1-2).⁵

	Identified Resources		Undiscovered Resources	
Cost/Category	Reasonably Assured	Inferred	Prognosticated	Speculative
<\$40/kgU	>1,766	1,204	/	/
<\$80/kgU	2,598	>1,858	1,946	/
<\$130/kgU	3,338	2,130	2,769	4,798
No cost range	/		/	2,973
Sub-total	5,469		10,540 (7,567 at <\$130/kgU)	
Total	16,009 (13,036 at <\$130/kgU)			
Unconventional (phosphates)	~22,000			
U in Seawater	~4,000,000			

 Table 1-1

 Identified and Undiscovered Resources of Natural Uranium (in ktU)⁶

Table 1-2 Estimated Resources of Natural Uranium (in Years of Current Consumption Rate)

	Identified Resources		Undiscovered Resources	
Cost/Category	Reasonably Assured	Inferred	Prognosticated	Speculative
<\$40/kgU	27	18	/	/
<\$80/kgU	39	28	29	/
<\$130/kgU	50	32	42	72
Sub-total	/		/	45
Total	82		158 (114 at <\$130/kgU)	
	241 (196 at <\$130/kgU)			
Unconventional (phosphates)	~330			
U in Seawater	60,150			

⁵ Source: *TradeTech*, <u>http://www.uranium.info</u>: NUEXCO Exchange Value (monthly spot price determined as of the last day of the month indicated); price given in \$/lb of U_3O_8 ; price in \$/kg of U (or \$/kgU) is obtained by applying a 2.6-multiplier to the price in \$/lb of U_3O_8

⁶ Kilo (metric) tons of uranium, or ktU

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Figure 1-2 Evolution of Natural Uranium Spot Price

A simple relationship can be used to calculate the number of years (N) uranium will be available at a reasonable price, given a recoverable uranium stockpile (S) and a yearly growth rate (G). The basic equation among these three parameters is:

$$S = \sum_{i=0}^{N-1} 66,500 \times (1+G)^i$$
 Equation 1-1

Rearranging for N yields:

$$N = \frac{\ln\left(1 + \frac{S \times G}{66,500}\right)}{\ln(1+G)}$$
 Equation 1-2

For illustration, several hypothetical uranium reserve scenarios are considered, assuming that the existing mix of reactor technologies is not substantially altered:

- Identified Resources: S = 5.5 MtU
- Conventional (Identified + Undiscovered) Resources: S = 16 MtU
- Conventional + Unconventional (phosphates) Resources: S = 38 MtU
- Seawater: S = 4,000 MtU

In addition, two hypothetical uranium reserve scenarios are considered for which it is assumed that advanced reactor systems (such as fast reactors that can efficiently use U-238) are relied upon:

- U-238 in Conventional + Depleted Uranium Resources: S = 900 equivalent MtU⁷
- U-238 + Th-232 Resources: S = 3,600 equivalent MtU⁸

Figures 1-3 and 1-4 show that the assumed growth of nuclear generation has a huge impact on natural resources sustainability.



Figure 1-3 Natural Resources Sustainability (in Years) as a Function of Annual Growth of Nuclear Generation (All Scenarios)

⁷ The followings are assumed: (1) 16 MtU of Conventional Resources + 2 MtU of existing depleted uranium stockpile; and (2) reliance on advanced reactor systems increases recovery of the energy content of uranium by a factor equal to 50

⁸ Assuming that thorium is three times more abundant than uranium ["Thorium fuel cycle – Potential benefits and challenges," IAEA-TECDOC-1450 (2005)]



Figure 1-4 Natural Resources Sustainability (in Years) as a Function of Annual Growth of Nuclear Generation

A "high scenario"⁹ defined by the World Energy Council consists of an annual growth of 3.5% between 2006 and 2050, and of 1.8% between 2050 and 2100. It corresponds to an average annual growth of 2.9% in the coming century. A "low scenario"¹⁰ consists of an annual growth of 3.2% until 2050 and of 1.4% between 2050 and 2100. It corresponds to an average annual growth of 2.2% in the coming century. For reference, the average annual growth of nuclear power in the world at the end of the latest expansion (1985-1990) was 5.6%.

Assuming a significant role for nuclear energy (annual growth of ~2.5%) in the coming decades, the "Identified Resources" would be exhausted in ~50 years and the "Conventional Resources" in ~75 years. Taking uranium from phosphates into account, resources would be exhausted in ~100 years. Present and future thermal reactors are likely to operate for up to 60 years, and even possibly longer. Therefore, advanced reactor systems together with their supporting nuclear fuel cycle infrastructure (i.e., fast reactors that are capable of utilizing all the energy content of uranium and thorium) must be developed, except if new and large sources of uranium (such as uranium from seawater) recoverable at competitive costs become available on a timely basis. Therefore, development efforts towards defining and developing the most cost-effective advanced reactor systems and associated fuel cycle facilities, on a schedule consistent with their expected significant role in meeting future energy requirements, are very important.

⁹ Scenario WEC-A3

¹⁰S. Massara et al., "Fast Breeder Reactor Scenarios of Nuclear Energy Evolution over the World Scale," 15th International Conference on Nuclear Engineering, Nagoya, Japan, April 22-26, 2007

1.1.2 Potential Natural Uranium Savings

In the interval between the current situation (mostly once-through thermal reactors) and the deployment of advanced reactor systems (assumed to be fast reactors), four approaches can be pursued to decrease the demand for <u>N</u>atural <u>U</u>ranium (NU).

Down-blending of Additional Highly-Enriched Uranium (HEU) from Military Stockpiles

Beyond the existing commitments the U.S. and Russia already made regarding down-blending of some of their surplus military HEU, each country will have more than 550 tons of HEU remaining.¹¹ This inventory, with an average enrichment around 80%, is equivalent to ~126,000 tons of NU, i.e., approximately two years at current worldwide consumption rate. In addition, non-military stockpiles provide about three years of worldwide uranium requirements.¹²

Enrichment of Depleted Uranium Tails

Not all of the U-235 contained in uranium ore is used during the enrichment process. As a result, some U-235 remains in the depleted uranium tails. The recovery ratio of U-235 depends on the relative prices of NU and Separative Work Unit (SWU).

Let N be equal to the U-235 weight fraction in natural uranium (NU), H (high) the weight fraction of U-235 in Enriched Uranium (EU) and L (low) the weight fraction of U-235 in Depleted Uranium (DU).

Then, 1 ton of NU results in
$$\frac{N-L}{H-L}$$
 ton of EU and $\frac{H-N}{H-L}$ ton of DU.

Thus, the ratio of U-235 recovery is $\frac{H}{N} \times \frac{N-L}{H-L}$ (which is equal to 1 if L=0).

H is determined by the nuclear fuel enrichment requirements to sustain the nuclear reaction over a specified length of time, and N is equal to 0.71%; therefore, only L is an unspecified parameter. The relative ratio (in percent) of U-235 recovery as a function of tail assay is shown in Figure 1-5, assuming a product assay of 4.5%.

¹¹Union of Concerned Scientists. Preventing Nuclear Terrorism Project Factsheet: Enormous Military Stockpiles of Highly Enriched Uranium (HEU) in Russia and the United States. 13 April 2004, http://www.ucsusa.org/assets/documents/nwgs/nuclear_terrorism-stockpiles.pdf> accessed 6 August 2010.

¹² Average over the major nuclear countries (from 2007 edition of the Red Book)



Figure 1-5 Percentage of U-235 Recovery as a Function of Tail Assay (for a Product Assay of 4.5%)

The choice of L depends on the relative price of NU and SWU. The number of SWU required is given by the following formula:

$$SWU = EU \times \Phi(H) + DU \times \Phi(L) - NU \times \Phi(N)$$
 Equation 1-3
where $\Phi(X) = (2X - 1) \times \ln\left(\frac{X}{1 - X}\right)$

For a given quantity of EU at an enrichment of H, the total cost is $C = P_{NU} \times NU + P_{SWU} \times SWU$.

Combining these two formulas, it is possible to express the cost by unit of EU as a function of H, L, N, $P_{_{NU}}$ and $P_{_{SWU}}$:

$$C/EU = P_{NU} \times \frac{H-L}{N-L} + P_{SWU} \left(\Phi(H) + \frac{H-N}{N-L} \Phi(L) - \frac{H-L}{N-L} \Phi(N) \right)$$
 Equation 1-4

The optimal L is obtained by applying the Lagrangian method on this expression. The result does not depend on H, the product assay. The optimal tail assay is the solution of the following implicit equation:

$$\frac{P_{NU}}{P_{SWU}} + (N - L)\Phi'(L) + \Phi(L) - \Phi(N) = 0$$
 Equation 1-5

So, the optimal tail assay only depends on the price ratio P_{NU}/P_{SWU} (P_{NU} in kgU and P_{SWU} in SSWU) (see Figure 1-6).





In the current market, the price ratio is around 0.8,¹³ and the corresponding optimal tail assay is 0.25 %. If $P_{_{NU}}$ increases, L is likely to decrease and the recovery ratio of U-235 increases correspondingly; this results in a reduced demand for natural uranium as shown in Figure 1-7. Using $P_{_{NU}} = $48/lbU_3O_8$, $P_{_{SWU}} = $155/SWU$, and a tail assay L = 0.25 wt% in U-235 as the reference case, the increased uranium price leads to a lower optimal tail assay, assuming a fixed enrichment price of \$155/SWU, which results in the natural uranium savings shown in Figure 1-7.



Figure 1-7 Natural Uranium Savings as a Function of Natural Uranium Price, P_{NU} (in \$/lbU₃O₈)

¹³ This ratio is based on a natural uranium price of $48/lbU_3O_8$ (125/kgU) and an enrichment price of 155 per SWU.

So, savings of 15% in natural uranium resource are expected when $P_{_{NU}}$ is three times $P_{_{SWU}}$. In addition, if the existing tails (1,600,000 tons with an average U-235 content of 0.3%¹⁴) would be re-enriched at 4.5% with a new tail assay of 0.15%, then 55,000 tons of EU corresponding to 440,000 tons of NU (6.5 years of current consumption), would be saved.

Use of MOX Fuel

Reprocessing of spent UOX and mono-recycling of plutonium in LWRs will be discussed in Section 4 of this report. Concerning uranium resources, it is generally recognized that use of MOX fuel allows savings in natural uranium around 12.5%. Once again, the attractiveness of mono-recycling of Pu depends greatly on P_{NU} . In addition, if the existing 200,000 MTHM of spent fuel were to be reprocessed, 236,000 tons of equivalent NU (3.5 years of current consumption) would be saved by the use of MOX. However, this figure is not realistic because the actual reprocessing capacities are far too limited.

Use of Reprocessed Uranium

As will be discussed in Section 4, the use of reprocessed uranium (RepU) raises some issues because of its U-232 and U-236 contents. So, RepU has to be purified and re-enriched at a higher level than NU (because of the neutron absorbing properties of U-236), which raises the cost of ERU (Enriched Reprocessed Uranium). This operation is economical when P_{NU} is high. If all uranium in spent UOX were to be converted into ERU, the additional savings in natural resources would be around 9.5%. In addition, the existing 45,000 tons of RepU correspond approximately to 38,000 tons of NU (0.5 year of current consumption). If the existing 200,000 MTHM of spent fuel were to be reprocessed, 163,000 tons of NU equivalent (2.5 years of current consumption) would be saved by the use of RepU.

Conclusions Regarding Potential Natural Uranium Savings

If further processing of uranium tails, reprocessing, and use of RepU become economically competitive, a 33% saving in natural resources would be possible (Figure 1-8). In addition, around 12 years at current consumption rates (18 years if all the existing spent fuel is reprocessed) could be saved with the existing stockpiles of fissile materials: military HEU, RepU or depleted uranium (Figure 1-9). These figures are all but negligible in the short term, and do not significantly change the long term outlook in terms of sustainability.

¹⁴ 2007 Red Book



Figure 1-8 Potential Future Natural Uranium ("Unat") Savings (Relative to Current Once-through Cycle)





1.2 Economic Competitiveness

The cost of nuclear power can be divided as shown in Figure 1-10.



Figure 1-10 Breakdown of Nuclear Generation Electricity Cost

Depending on the fuel cycle chosen and on different assumptions made for the different unit costs, reactor costs represent between 80 and 90% of electricity costs,¹⁵ reflecting the high capital cost of constructing nuclear power plants that, alone, can represent 60% or more of the nuclear electricity costs. As a result, the fuel cycle choice has a relatively small impact on the overall economics of nuclear power. However, it is important to note that once the plants are in operation, recurring fuel cycle costs become much more important, as do operation and maintenance (O&M) costs. This is especially true for plants whose capital costs have been largely amortized.

1.2.1 Analyses of Once-Through and MOX Mono-Recycling Fuel Cycles

To illustrate the impact on fuel cycle costs of reprocessing and recycling in the existing fleet of reactors, two different fuel cycles are considered: a once-through cycle (with spent UOX sent to a repository for disposal) and mono-recycling of Pu in LWRs (with spent MOX+HLW sent to a repository for disposal).¹⁶

Figure 1-11 shows, as expected, that recycling Pu reduces the front-end and the waste management costs, but increases the overall back-end costs. Currently, the once-through fuel

¹⁵ Source: Advanced Fuel Cycle Economic Analysis of Symbiotic Light-Water Reactor and Fast Burner Reactor Systems, Idaho National Laboratory, Idaho Falls: 2009. INL/EXT-09-15254

¹⁶ Source: *Nuclear Fuel Cycle Cost Comparison Between Once-Through and Plutonium Single-Recycling in PWR*, EPRI, Palo Alto, CA: 2009. 1018575

cycle is less costly than reprocessing but is also more sensitive to the price of natural uranium and enrichment services. Figure 1-12 shows under what conditions reprocessing could become economically competitive compared to the once-through cycle.



Figure 1-11 Fuel Cycle Costs (\$/MWhe)

The economic competitiveness of reprocessing improves for scenarios where PUREX costs decrease and other fuel cycle costs increase significantly (by 100% or more). However, as already previously discussed, the differences in fuel cycle costs for different options are small relative to the overall cost of nuclear electricity, and in the recycling scenario examined, disposition of spent MOX fuel is by disposal in a geologic repository. Further recycling of spent MOX fuel in fast reactors would substantially improve the attractiveness of reprocessing spent UOX fuel, as discussed in greater detail in Section 4.

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Percentage Increase in U and U+SWU+Waste Costs Required for Break-even as a Function of Reprocessing Costs (\$/kgHM)

1.2.2 Analyses of More Advanced Fuel Cycles

More advanced fuel cycles, based on LWR and Fast Reactor (FR) technologies, are now considered using the results of the INL study entitled "Advanced Fuel Cycle Economic Analysis of Symbiotic Light-Water Reactor and Fast Burner Reactor Systems."¹⁵ In this study, three cycles are considered: Once-Through Cycle, 1-Tier (LWR-UOX + FR) and 2-Tier (LWR-UOX + LWR-MOX + FR). The cost breakdowns are shown in Figure 1-13. Again, it can be seen that the cost breakdown does not depend significantly on the fuel cycle choice. Reactor capital costs represent the major part of the total electricity costs in every scenario.

An informative comparison can also made with other generation types. Figure 1-14 shows that even if the once-through cycle currently represents the lowest cost option for nuclear power, the differences between the fuel cycles are very small compared to the other generation types, especially if a carbon tax is implemented. Consequently, it appears unlikely that the economic competitiveness of nuclear energy with respect to other forms of electricity generation would be driven by the choice of the fuel cycle. Economics does not appear to be the dominant issue as far as the fuel cycle choice is concerned, especially when considering deployment of new units. The future competitiveness of advanced fuel cycles will mostly depend on the capital cost of advanced reactor systems, assuming that the technology to build and operate these systems and supporting infrastructure is available and highly reliable.



Cost Composition of Systems

Figure 1-13 Cost Breakdown for Different Fuel Cycles (Source: INL)¹⁵





1.3 Waste Management

Irradiation of nuclear fuel generates several types of radioactive by-products that require management as either waste or potentially as feedstock for further energy generation.¹⁷ Important classes of these by-products include:

- Fission Products (FP). Two FP nuclei are typically generated per fission of uranium or plutonium
- Actinides. The two major actinides are U and Pu; the minor actinides (MAs) of greater interest are neptunium, americium and curium
- Activation Products. The main long-lived activation products are C-14 and Cl-36.

Fission products and actinides represent a small fraction of the total amount of the waste generated by the nuclear industry, but they concentrate more than 95% of the overall radioactivity. Moreover, those with half-lives on the order of thousands of years or more require long-term isolation or removal from the biosphere, either by disposal in a suitable repository or through transmutation into shorter-lived nuclides. Although there is a technical consensus today among the scientific community that deep geologic disposal provides a reliable and safe solution, there is presently no geologic repository for spent nuclear fuel (SNF) or high-level waste (HLW) in operation. Therefore, interim storage over increasing periods of time of SNF and HLW is a necessary waste management activity. It is, however, a beneficial activity with regard to allowing a reduction of the thermal load generated by the SNF and HLW.

Figure 1-15 presents the main ongoing technical concerns associated with nuclear waste management.



Figure 1-15 Waste Management Main Issues

¹⁷ More details about characteristics of HLW nuclides are presented in the report *Advanced Fuel Cycles Impact on High-Level Waste Disposal*, EPRI, Palo Alto, CA: 2007. 1015129

For fuel cycles currently in use worldwide, two kinds of spent fuel have to be considered: spent UOX and spent MOX resulting from recycling Pu in the existing fleet of reactors. The chemical and isotopic compositions of spent UOX and MOX fuels differ significantly; this leads to different decay heat and radiotoxicity levels. In particular, ten years after discharge from the reactor, spent MOX releases about three times more heat than spent UOX, and this ratio increases with cooling time.

1.3.1 Interim Storage

Once irradiated, nuclear fuel is very radioactive, as shown in Figure 1-16. Spent fuel requires some period of cooling in wet storage, typically in pools adjacent to the reactor, prior to (i) transfer to an on-site dry storage facility, or (ii) transportation to an off-site storage facility. The latter could be a centralized storage facility, a reprocessing plant, or a subsurface facility associated with a permanent geologic repository.



Figure 1-16 Radioactivity of Spent Nuclear Fuel after Discharge Compared to Other Radioactive Materials (ILW = Intermediate Level Waste) (Source: ANDRA)¹⁸

On-site storage is necessary regardless of the fuel cycle. Wet storage in spent fuel pools is still the primary option for storage of irradiated fuel today. However, spent fuel pools were designed at a time when transportation of the discharged spent fuel to a reprocessing plant was anticipated after a relatively short wet storage duration. Therefore, in increasing numbers, pools no longer

¹⁸ Adapted from « Référentiel de connaissance et modèle d'inventaire des colis de déchets à haute activité et à vie longue », ANDRA, June 2005.

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have sufficient capacity to accommodate the increasing inventory of spent fuel, even after pool re-racking(s) to allow higher storage density.

In the 1980's, two technologies were tested extensively in the U.S.: (i) spent fuel consolidation for continued storage in pools, and (ii) dry storage under inert atmosphere in heavy-wall casks or vaults. Spent fuel consolidation involves extracting the fuel rods from their assemblies or bundles and placing the individual rods in consolidated fuel storage boxes. Because of the significant operational burden, spent fuel consolidation was not selected by utilities, as dry storage technology was successfully demonstrated over the same time frame.

Dry storage has been licensed in the United States since 1986.¹⁹ As of May 2010, close to 50,000 spent fuel assemblies have been transferred to ~1,250 dry storage canister/cask systems in the U.S.²⁰ The extensive experience with dry storage in the U.S. and internationally underscores the technological maturity of this option.

Dry storage vaults are an alternative to individual cask systems and typically consist of a modular or fixed concrete civil structure in which large numbers of fuel assemblies are stored. Cooling is provided by either natural or forced air convection.

The initial (fixed) investment cost for wet storage in pools is intermediate between the two dry storage alternatives, i.e., higher than dry cask storage and lower than dry vault storage. The operation and maintenance (variable) costs are higher than those for dry storage, but are less than the total cost of cask procurement required for dry storage in cask systems. In principle, wet storage in pools or dry storage in either individual casks or collective vaults is scalable and can be adapted for use in large centralized (or regional) interim storage facilities.

When spent fuel is reprocessed, the reprocessing plant plays the role of a centralized interim storage: spent fuel is stored in pools, then reprocessed, and finally the resulting high-level waste and long-lived intermediate-level waste are stored at the reprocessing plant, before eventually being sent to a geologic repository. Spent MOX fuel presents additional challenges for dry storage due to significantly higher heat loads and increased transuranic²¹ inventories. This requires much longer cooling periods in wet storage before spent MOX fuel can qualify for dry storage in large capacity casks. As a result, spent MOX fuel is almost exclusively managed by wet storage in pools at-reactor sites or at the reprocessing plant.

On-site storage of spent fuel in dry casks is a safe and practical option as long as the power plant is in operation. But once a power plant site is decommissioned, it would be highly desirable to move the spent fuel off-site in order to return the site to unrestricted use. A centralized storage facility that would accept the spent fuel from decommissioned reactors could demonstrate not

¹⁹ NRC, 2007. Dry Cask Storage. 13 February 2007. http://www.nrc.gov/waste/spent-fuel-storage/dry-cask-storage.html. Accessed 10 November 2009.

²⁰ Store Fuel and Decommissioning Report, Vol. 12, No. 142 (June 2010), Ux Consulting

²¹ Transuranic elements are elements that have an atomic number higher than that of uranium; they include neptunium, plutonium, americium, curium, berkelium, californium, etc.

only the industry capability to totally clean former reactor sites, but also the institutional ability to find acceptable medium-term solutions to the management of nuclear wastes independently of power plant location.

1.3.2 Geologic Disposal

Decay Heat

Geologic disposal is strongly influenced by waste characteristics, which can vary widely based on the associated nuclear fuel cycle. Decay heat is a critical waste characteristic that impacts many important aspects of geologic disposal. For the universally accepted reference concept of a mined deep-geologic repository, consideration of waste heat figures heavily in site selection (based on physical properties of candidate geologic media), waste package, and repository design, which ultimately determine repository performance. Of particular significance, the size of the repository is mostly determined by the decay heat of the waste. Accordingly, it is instructive to look at the contributions of the different constituents of spent UOX and spent MOX (both at a nominal burnup of 50 GWd/MTHM) in terms of decay heat, as shown in Figures 1-17 and 1-18. The factor-of-three difference in the vertical axis limits associated with decay heat (W/MTHM) for the two fuel types should first be noted.



Figure 1-17 Decay Heat of Spent UOX (50 GWd/MTHM) (Source: EDF)²²

²² EDF R&D.



Figure 1-18 Decay Heat of Spent MOX (50 GWd/MTHM) (Source: EDF)²²

MOX spent fuel generates more decay heat than UOX spent fuel (three times more after 10 years, 5 times more after 100 years) due to the higher inventory in transuranic elements present in irradiated MOX fuel. Specifically, actinides contribute four times more heat than fission products (FPs) after 10 years in spent MOX fuel, and 20 times more heat after 100 years. On the other hand, spent UOX decay heat is dominated by fission products during the first 60 years following discharge from the reactor, after which time actinide elements account for most of the decay heat (2.2 times more than FPs after 100 years). As far as fission products are concerned, after 10 years, only two parent/daughter pairs need to be considered, Sr-90 and its daughter Y-90, and Cs-137 and its daughter Ba-137m, as shown in Figures 1-17 and 1-18.

Overall, the total decay heat generated by fission products is more important in spent UOX than in spent MOX, because slightly more energy is released by the fission of Pu than U, and also because the fission yield of Sr-90 is higher for uranium-235 compared to Pu-239. For MOX, Cs-137 contributes ~70% of the total FP decay heat versus ~50% for spent UOX (Figure 1-19). As far as the actinides are concerned, only Pu, Am and Cm isotopes contribute significantly to decay heat as other actinide elements typically comprise less than 0.1% of the total actinide decay heat.


Figure 1-19 Contribution of Cs and Sr in Fission Products' Decay Heat in Spent UOX Fuel

For spent UOX (Figure 1-20), Pu (mostly Pu-238) is the largest decay heat contributor for up to 40 years. Afterwards, Am (mostly Am-241 produced from the decay of Pu-241) becomes the dominant heat generating nuclide, contributing greater than 50% of the total actinide decay heat. It is worth noting that Am-241 continues to accumulate after irradiation due to the decay of Pu-241 with a half-life of 14.4 years. Cm isotopes are less significant contributors to actinide decay heat due to the relatively short half-life (~18 years) of its main heat generating member, Cm-244.

Spent MOX decay heat profiles differ from those for spent UOX in that Cm plays a more significant role during the first 50 years (Figure 1-21). As mentioned above, the decay of Pu-241 results in continued accumulation of Am-241 post-irradiation. Consequently, separation of Pu from spent fuel decreases the thermal loading from Am-241 in the resulting HLW.



Figure 1-20 Contribution of Pu, Am and Cm to Actinides' Decay Heat in Spent UOX

In general, disposal of fission products (FPs) is preferable to transmutation. Most are short-lived relative to relevant timeframes for disposal technologies. The two most significant FPs for decay heat, ⁹⁰Sr and ¹³⁷Cs, have half-lives on the order of 30 years. Accordingly, the thermal burden of FPs can be decreased substantially by decay during interim storage for a period equivalent to two to three half lives (60 to 90 years). Separation and transmutation of FPs are technologically very difficult, costly, and do not result in any energy production, unlike the recycling of actinides in reactors. The management of fission products in spent fuel and HLW is addressed in greater detail in Section 4.2.5.

Given the unattractiveness of FP recycle, the major distinctions among fuel cycles are centered on which transuranics (TRUs) are recycled and in what manner. A number of options can be considered; Table 1-3 provides a concise list of potential TRU recycling options along with the primary benefits and challenges associated with each.



Figure 1-21 Contribution of Pu, Am and Cm to Actinides' Decay Heat in Spent MOX

Table 1-3 Impact of TRU Recycling on HLW Decay Heat

Option	Description	Benefit	Drawback
1	Once-Through Cycles	Does not require reprocessing	Generates the highest decay heat in the disposal
2	Disposal of Am and Cm Recycling of Pu	Does not require advanced reprocessing	Generates a high decay heat in the geologic disposal
3	Disposal of Cm Recycling of Pu and Am	Allows elimination of the most significant long-term decay heat contributors	Requires separation of Cm and Am from each other
4a	Interim storage of Cm Recycling of Pu and Am	Cm-244 decays in Pu-240 (half-life: 18 years), which can be recycled	Requires separation of Cm and of Am
4b	Recycling of Pu, Am and Cm	Provides most significant reduction in decay heat for the geologic disposal	Cm is highly radioactive and difficult to handle in the fuel cycle

Recycling or storage of separated Cm does not represent an attractive option for spent UOX fuel because it comprises only 7% of the total decay heat after 25 years of cooling. In contrast, Cm represents 30% of the total decay heat for spent MOX after 25 years, which makes separation of Cm more attractive in that case; however, an extended decay period could also reduce the Cm heat load before emplacement of waste in a repository.

Long-Term Radiotoxicity

The ultimate goal of geologic disposal is to isolate spent fuel and/or high-level radioactive waste from the biosphere in order to minimize effects on humans and the environment over unprecedented timeframes, i.e., tens of thousands to hundreds of thousands of years. Repository performance over this timeframe depends on multiple attributes of the waste inventory itself and the repository environment (geology, hydrology, geochemistry) and repository design.

Waste radiotoxicity is a figure-of-merit that is often invoked to illustrate the relative hazards of waste constituents over time (e.g., Figure 1-22).



Figure 1-22 Radiotoxicity (Expressed as Radioactivity/Unit Mass) of Spent UOX Fuel Over Time

Based on this metric, Pu is the most important element in terms of radiotoxicity, especially between 100 and 100,000 years after discharge. So, Pu recycling can dramatically decrease the radiological inventory of waste. But this measure of radiotoxicity is strictly a function of the waste inventory itself and does not take into account engineered and natural aspects of the whole repository system, which ultimately drive repository performance. As such, waste radiotoxicity alone is of secondary significance compared to dose performance assessments that take into account all key factors. For this purpose, it is worth looking at the Yucca Mountain (U.S.A.) (Figure 1-23) and Bure (France) (Figure 1-24) system performance assessment results. EPRI total system performance assessment modeling of a geologic repository for commercial spent fuel at Yucca Mountain indicates that the primary isotopes of concern for dose are Tc-99, I-129 and Np-237. For a geologic repository at the Bure site in France, the isotopes I-129, CI-36 and Se-79 have been found to pose the primary radiological concerns in terms of public dose.



Figure 1-23 Radiological Impact of Different Radionuclides in the Case of Yucca Mountain on a Log-Log Scale (Source: EPRI)²³



Figure 1-24 Radiological Impact of Different Radionuclides in the Case of Bure, France (Source: ANDRA)²⁴

²³ Source: *EPRI Yucca Mountain Total System Performance Assessment Code (IMARC) Version 10 – Model Description and Analyses*, EPRI, Palo Alto, CA, 2009 : 1018712.

²⁴ Dossier 2005: Argile. Tome: Evaluation of the Feasibility of a Geological Repository in an Argillaceous Formation, ANDRA 2005

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In these two examples of dose performance assessments for HLW repositories, the maximum dose after several thousands of years remains well below regulatory limits. For Yucca Mountain and Bure, the nuclides producing the highest radiological impact are fission products, except for Np-237. So the only question raised by long-term performance assessment results on the choice of the nuclear fuel cycle is whether Np should be transmuted or not (and only in the case of Yucca Mountain). Because of the decay of Pu-241 into Am-241 and the decay of Am-241 into Np-237, efficient and effective transmutation of Np would have to be accompanied by recycling of Pu (destruction of Pu-241 by fissioning) and transmutation of Am-241. However, because of the low radiological impacts estimated from total system performance assessments, transmutation of Np and fission products would be deemed to be unnecessary.

Waste Volumes

As mentioned earlier, the decay heat of the waste ultimately determines the physical extent or footprint of the geologic repository needed to provide adequate heat dissipation into the host rock/media. However, it is also instructive to compare the volumes of waste resulting from fuel cycles with and without reprocessing (Table 1-4). In the "once-through cycle" scenario, spent UOX fuel is stored temporarily and then sent directly to a geologic repository for disposal. In the "reprocessing" scenario, spent UOX is reprocessed, Pu and RepU are recovered and only the fission products and the minor actinides (FP+MA) are vitrified and sent to a geologic repository. Long-lived intermediate level wastes (ILW-LL) are also generated through this process and require disposal, as well.

	Once-Through	Reprocessing		
	Spent Fuel	ILW-LL	HLW	Total
Primary Waste ²⁵	0.41	0.19	0.16	0.35
Yucca Mountain Disposal ²⁶	1.3		_	_
Bure-like Disposal ²⁷	3.4 (waste package)	1.4	0.41	1.81

Table 1-4 Comparison in Waste Volumes in m³/MTHM between Once-through and Reprocessing

²⁵ Basis: Spent PWR fuel with burnup of 45 GWd/MTHM. Initial uranium mass is 0.46 MTHM/assembly and the volume is 0.188 m³/assembly. Based on proven glass formulation technology at La Hague, reprocessing of 1 MTHM of used fuel results in 0.85 canister of vitrified HLW and ~1 canister of compacted ILW-LL. The volume of the vitrified HLW canister and of the compacted ILW-LL canister is identical: 0.194 m³/canister. Reprocessing data are from "Référentiel de connaissance et modèle d'inventaire des déchets à haute activité et à vie longue," p. 85, ANDRA (2005)

²⁶ Author's estimate based on Yucca Mountain's 21-PWR waste package

²⁷ Author's estimate based on Bure clay's 4-PWR waste package design

In Table 1-4, it can be seen that the volumes of primary waste are quite similar between oncethrough or reprocessing. Reprocessing of used fuel with higher discharge burnup will increase the HLW volume,²⁸ while improved glass formulation technology will reduce the HLW volume.²⁹ The ILW-LL volume reflects the considerable improvements made by the La Hague facility to reduce these volumes.

The comparison between the waste storage packages depends on the chosen waste package design. For the Bure clay, disposal of spent fuel would require twice as much repository volume as storing waste packages from reprocessing. For Yucca Mountain, there are no data about storing ILW-LL or HLW from reprocessing. Such comparisons must be considered cautiously because they depend greatly on the geologic disposal sites and concepts involved. Moreover, the size of the repository is primarily determined by the decay heat of the waste –which is almost the same between the two scenarios considered here– and not by the waste package volumes.

1.3.3 Reduction of TRU inventory

Given the time required to deploy or phase out a nuclear fuel cycle, it is necessary to look not only at the reduction of TRU going to geologic repository, but also at the entire TRU inventory in the fuel cycle as a whole.³⁰ In fact, during a possible phase-out of an advanced fuel cycle technology, the actinide inventory could become an important contributor to the waste legacy, as shown in Table 1-5.

Recycling of Pu in LWRs does result in a reduction of Am and Cm going into the repository, so TRU burning in fast reactors is the only way to reduce the TRU inventories going to waste disposal using fission reactor technology.³¹ However, such reductions in the TRU content of HLW streams are counter-balanced by large increases in the total fuel cycle inventory of TRUs. For example, as far as Am is concerned, the comparison between once-through fuel cycle and TRU burning in Fast Burner Reactor shows a reduction of the inventory going to waste by a factor larger than 100. But it would take more than 108 years of operation³² at equilibrium to compensate for the higher Am inventory in the fuel cycle.

²⁸ For 55 GWd/MTU used fuel, a factor of 1.2 canister/MTHM is applicable versus 0.85 for proven glass formulation technology at La Hague.

²⁹ For 45 GWd/MTU and 55 GWd/MTU, factors of 0.74 and 0.9 are expected to be demonstrated with improved glass formulation technology recently implemented at La Hague.

³⁰ More information about this issue can be found in EPRI report 1015129 "Advanced Fuel Cycles Impact on High-Level Waste Disposal"

³¹ Aside from partitioning and transmutation schemes employing fast reactors, the use of accelerator-driven fuel cycle schemes (ADS) have been proposed for destruction of minor actinides. [Source: NEA No. 5990, OECD 2006]

³² Obtained by dividing the difference in Am inventories (701 kg - 22 kg = 679 kg) by the difference in Am disposal (6.35 kg/year – 0.055 kg/year = 6.295 kg/year)

The Challenges of a Sustainable Nuclear Fuel Cycle

These results indicate that in order to obtain significant reductions in total inventories of Am or other TRU, advanced nuclear fuel cycles would have to be maintained for a very long period time – on the order of several hundred years.

Fuel cycle \rightarrow	Once-through	Mono-recycling of Pu in PWRs	Multi-recycling of Pu in PWRs	TRU Burning in Fast Breeder Reactors	
TRU content going in kg/year (amount	to HLW repository (s normalized to 8.76	assuming 0.1% loss TWhe/year)	in separation proce	esses)	
Pu	230	153	0.37	1.25	
Np	16.2	16.6	14.45	0.0066	
Am	6.35	16.2	39.4	0.055	
Cm	3.3	8.1	19.7	0.013	
TRU inventory in the in kg (amounts nor	TRU inventory in the fuel cycle (reactors+fabrication+reprocessing) in kg (amounts normalized to 8.76 TWhe/year)				
Pu	767	3,285	4,818	17,520	
Np	53	131	116	88	
Am	22	88	307	701	
Cm	11	44	158	175	

Table 1-5	
TRU Inventories for Different Fuel Cycles (Source: CEA) 3	3

1.3.4 Conclusions on Waste Management

High-level waste management is a long-term concern, given the long half-lives of some radionuclides and the associated period of performance for a repository that spans tens of thousands to hundreds of thousands of years. The management of decay heat represents a more useful and objective figure-of-merit compared to waste radiotoxicity, because it more directly impacts the size, design, and performance of the geological repository. Interim storage of spent fuel and vitrified wastes is a necessary and important fuel cycle activity that should be integrated in the context of managing spent fuel/HLW for either geologic disposal or recycling. Waste management is also an important consideration in terms of public acceptance.

1.4 Nonproliferation

Nonproliferation is a complex topic spanning the scientific, technical and political arenas. Accordingly, a full treatment of non-proliferation is beyond the scope of this report. The aim of

³³ Adapted from a CEA report on "Synthèses des Résultats des Recherches sur les Axes 1 et 3", 2005

the following discussion is to address key technical aspects of nonproliferation as it relates to nuclear fuel cycle options for a declared weapon state, such as France or the U.S.

Figure 1-25 distinguishes between the concepts of proliferation resistance and physical protection as they relate to the general topic of non-proliferation. Proliferation resistance is associated with the acquisition of nuclear weaponry by a non-weapon state, while physical protection relates to defeating the threat of theft of fissile material by a sub-national group.



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Figure 1-25
Impact of Nuclear Fuel Cycle Choice on Two Key Proliferation Threats
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The potential threats associated with non-weapon states and sub-national groups are therefore different. A non-weapon state presumably would be interested in acquisition of nuclear weapons in order to gain a strategic advantage; therefore, the reliability (and safety/stability) of a nuclear device is of paramount importance. Sub-national groups, on the other hand, may obtain their desired objectives through the psychological effects of confirmed possession or demonstration of a nuclear device (nuclear weapon, improvised nuclear device, radiological dispersion device, radiological exposure device) regardless of its effectiveness.

So, a relevant question for this review is how the choice of a nuclear fuel cycle in a weapon state influences these two different threats?

Three main issues can be considered:

- Effective international safeguards of nuclear facilities applied to non-weapon and weapon states (based on proliferation resistance methodology)
- High-performing physical protection for all nuclear facilities

The Challenges of a Sustainable Nuclear Fuel Cycle

• Limiting or degrading material attractiveness

Only the third issue, material attractiveness, is typically explicitly addressed in the context of various fuel cycle schemes. The four well established standards are:

- Threshold for low enriched uranium (i.e., ²³⁵U enrichment less than 20%)
- Radioisotope thermoelectric generator plutonium (i.e., ²³⁸Pu enrichment greater than 80%)
- Self-protecting dose rate (i.e., 500 rad/h at 1 m)
- Spontaneous fission neutron rate of reactor-grade plutonium (i.e., ²⁴⁰Pu content \geq 20%).

Two nuclear materials are particularly attractive for building nuclear devices: High Enrichment Uranium (HEU) and Plutonium (Pu). HEU is no longer physically present in commercial nuclear fuel cycles³⁴, so the material attractiveness issue boils down to a Pu attractiveness issue that includes considerations related to critical mass, decay heat, radiological exposure, and spontaneous neutron emission rate.

Works by B. Pellaud, G. Kessler, M. Saito, and more recently by C. G. Bathke et al. address potential Pu attractiveness as a function of its isotopic make-up and point to high burn-up as a most practical means to reduce Pu attractiveness for spent LWR fuels.

1.4.1 Plutonium Attractiveness

The main, relevant characteristics of plutonium isotopes are listed in Table 1-6.³⁴

Isotope	Bare Critical Mass (kg)	Neutron Emission (neutrons/s-kg)	Decay Heat (W/kg)	Dose at 1 meter (Sv/hr-kg)
Pu-238	10	2.6 10 ⁶	560	
Pu-239	10	22	1.9	0.85 10 ^{-₅}
Pu-240	40	0.91 10 ⁶	6.8	for separated Pu
Pu-241	10	49	4.2	spent fuel
Pu-242	100	1.7 10 ⁶	0.11	

Table 1-6Main Characteristics of Plutonium Isotopes

As shown in Table 1-6, the even Pu isotopes are unattractive for use in a nuclear device due to their high spontaneous neutron emission and high decay heat (particularly for Pu-238), whereas the odd isotopes are particularly attractive, especially Pu-239. Pu-241 is also a fissile isotope, but

³⁴ However, HEU remains in use in some research reactors (as fuel) around the world – currently a US priority for conversion of these facilities. Also, HEU targets remain in use for production of Mo-99 for generation of Tc-99 for nuclear medicine/imaging.

it has a relatively short half-life, decaying into Am-241; its attractiveness decreases with time as the fissile content decreases and emission of penetrating radiation [X-rays from de-excitation of Am-241 daughter nucleus (Np-237)] increases. The mass fraction of odd isotopes provides a practical first order metric for Pu attractiveness with regard to proliferation risk.

Research has been conducted to provide more sophisticated measures of Pu attractiveness. B. Pellaud³⁵ focuses on spontaneous neutron emission that could dramatically decrease the yield and the reliability of a nuclear device. He defines five different grades of Pu corresponding to five different levels of weapon-usability.

G. Kessler³⁶ focuses on decay heat that could negatively impact the reliability of a nuclear device. He concludes that separated Pu from spent PWR fuel with a burnup of 50 GWd/MTHM (having 2.8% of Pu-238) could be used for a nuclear device only if advanced technology would be employed; material with Pu-238 content higher than 6% would be practically unusable for use in a nuclear device, according to Kessler.

An approach from M. Saito³⁷ attempts to account for both spontaneous neutron emission and decay heat with a single attractiveness function:

$$ATTR = \frac{\frac{\alpha}{\alpha^{239}}}{\frac{DH}{DH^{238}} + \frac{SN}{SN^{238}}}$$
Equation 1-6

where α is the alpha-Rossi coefficient (ratio of super-criticality to prompt neutron lifetime). This formula is a ratio between the potential yield of nuclear device (which depends on α) and the technical difficulties to construct the device. The formula is convenient in that it only depends on the isotopic composition of Pu and defines Pu-attractiveness as a function of Pu-238 and Pu-240 contents.

Bathke, et al.³⁸ delineate a set of figures-of-merit (FOM) that are intended to explain the attractiveness or preferences for a range of nuclear materials across a span of credible nuclear adversaries. The basic concept of the FOM is to relate candidate nuclear material to the four standards described in the previous section. The expression for the FOM is:

³⁵ B. Pellaud, *Proliferation Aspects of Plutonium Recycling*, Journal of Nuclear Materials Management, Vol. XXXI, No. 1 (2002)

³⁶ G. Kessler, *Plutonium Denaturing by* ²³⁸*Pu*, Nuclear Science and Engineering, **155**, 53-73 (2007)

³⁷ M. Saito, *Development of Methodology for Plutonium Categorization*, Reactor Physics, Vol. 98, p. 669 (2008)

³⁸ C. G. Bathke et al., *The Attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios*, Proceedings of Global 2009, Paris, France, September 6-11, 2009, Paper #9544

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$$FOM_{2} = 1 - \log_{10} \left(\frac{\frac{M}{800} + \frac{Mh}{4500} + \frac{MS}{6.8(10)^{6}}}{+ \frac{M}{50} \left[\frac{D}{500}\right]^{\frac{1}{\log_{10} 2}}} \right)$$

Equation 1-7

where *M* is the bare critical mass of the metal in kg, *h* is the heat content in W/kg, *S* is the spontaneous-fission neutron production rate in n/(s-kg), and *D* is the dose rate of 0.2*M* evaluated at 1 m from the surface in rad/h.

According to the authors, the applicable FOM for a sub-national group, for most of the less advanced proliferant nations, or for a technically advanced proliferant state should not contain the term $MS/6.8(10)^6$. The latter term needs to be included only when dealing with those very few relatively unadvanced proliferant nations that desire a reliably high yield, and for which pre-initiation is an issue; therefore, the material attractiveness for such a nuclear device must necessarily be reduced for materials with a high spontaneous neutron generation rate.

An observation resulting from the evaluation of available proliferation based categorizations for Pu is irradiation of nuclear fuel to higher burnup provides for a practical approach for reducing Pu-attractiveness without adding a huge burden on the nuclear industry. Figure 1-32 illustrates this graphically: heat, spontaneous neutron emissions, and Pu-238/Pu-239 ratios all increase with increasing burnup.³⁹ However, the primary conclusion of the study by Bathke et al. is that all fissile material needs to be rigorously safeguarded to detect diversion by a state and provided with the highest levels of physical protection to prevent theft by sub-national groups. No "silver bullet" was found, that would permit the relaxation of current international safeguards or national physical security protection levels.

³⁹ E. Moniz, MIT Nuclear Fuel Cycle Study: Proliferation Chapter, Presentation made on June 9, 2009





Figure 1-26 Heat vs. Neutron Emission from Plutonium in Irradiated Reactor Fuels Compared to Weapon-grade Plutonium³⁸

In the past decades, proliferation risks were mostly associated with the back-end of the fuel cycle, and enrichment technology was frequently seen as offering a substantial barrier to weapons proliferation due to the size of the facilities (especially gaseous diffusion) required, which are generally detectable and difficult for a non-State entity to develop independently or access without the assistance of a State entity. Moreover, the amount of uranium required to obtain a significant quantity of HEU via gaseous diffusion enrichment is prohibitively large. However, the continued evolution and improvement of centrifuge technology allow for a smaller footprint and requires far less power and feed material to operate. In the coming years, laser isotope separation technology could lower the barrier to HEU production further. As a result, front-end fuel cycle technologies have become a dominant source of concern, resulting in a more balanced view about the proliferation risks of the back-end vs. the front-end of a nuclear fuel cycle.

2 POTENTIAL TECHNOLOGIES FOR ADVANCED FUEL CYCLES

This section provides a brief introduction and discussion of the key technologies and options for use in the deployment of an advanced nuclear fuel cycle. Primary fuel cycle elements include advanced nuclear reactors, reprocessing technologies, and waste disposal approaches.

2.1 Advanced Reactors

In current LWRs, the neutrons responsible for the fission process are mostly in thermal equilibrium with the surrounding water that acts both as coolant and moderator. This essentially implies reliance on the only naturally occurring fissile material, U-235, which represents only 0.71% of uranium. In contrast, most fuel cycles employing advanced reactor technology include deployment of reactors in which the neutrons responsible for the fission process have kinetic energy several orders of magnitude greater than thermal neutrons. Figure 2-1 shows the number of neutrons emitted per neutron absorbed by a Pu-239 nucleus, or η , as a function of the energy of the incident neutron (i.e., neutron being absorbed). For Pu, it can be seen that η increases significantly and consistently when the incident neutron energy is greater than ~10 keV. When $\eta > 2$, the potential exists for "breeding." Taking into account that one neutron is necessary for keeping the nuclear reaction going,⁴⁰ one or more neutrons are then available to be captured by a U-238 nucleus to form U-239 that subsequently decays into Pu-239. Therefore, the original Pu-239 nucleus that was consumed is regenerated at the expense of a U-238 nucleus. These fast reactors maximize natural energy resource utilization through breeding of fuel from relatively abundant U-238 (or alternatively, Th-232).

The breeding capacity of a fast reactor is characterized by the Conversion Ratio, or CR. CR is the number of fissile nuclei generated by the fission of one fissile nucleus. If CR <1, the fast reactor is a burner, which means that it decreases the fissile inventory. If CR = 1, the reactor is self-sustainable. If CR >1, the fast reactor is a breeder. Conventional PWRs have a typical CR of 0.3 - 0.5.⁴¹ Fast reactors are also capable of transmuting and burning minor actinides more efficiently than thermal reactors because of lower capture/fission ratios (Figure 2-2).⁴²

⁴⁰ The average number of neutron for keeping the reaction going is actually greater than one, given that not all neutron absorptions result in fission

⁴¹ 2008, CEA "PWR with hardened spectrum for improved conversion ratio"

⁴² Data from CEA

Potential Technologies for Advanced Fuel Cycles

A number of promising advanced reactor concepts are being considered by the Generation IV International Forum (GIF), with one-half comprising fast reactor designs and the other half comprising innovative thermal reactors concepts. Table 2-1 summarizes the main characteristics of these advanced GIF designs.



Figure 2-1 Number of Neutrons Available for Breeding in Thermal or Fast Neutron Spectrum





Name	Neutron Spectrum	Fuel Cycle	Applications Other than Electric Power Generation
Gas-Cooled Fast Reactor (GFR)	Fast	Closed	Actinide management Hydrogen
Lead-alloy Fast Reactor (LFR)	Fast	Closed	Actinide management Hydrogen
Sodium Fast Reactor (SFR)	Fast	Closed	Actinide management
Very High Temperature Gas Reactor (VHTR)	Thermal	Open	Hydrogen Process heat
Supercritical Water Reactor (SCWR)	Thermal/Fast	Open/Closed	None identified
Molten Salt Reactor (MSR)	Thermal	Closed	Actinide management Hydrogen

Table 2-1 GEN IV Advanced Reactor Concepts

The sodium-cooled fast reactor (SFR) is the only GEN IV concept that has been demonstrated on an industrial scale, for examples SuperPhenix in France and BN-600 in Russia. As a consequence, the SFR can be considered as the leading concept to provide the most deployable technology in the near future.

In addition to these advanced reactor designs, other advanced nuclear concepts are under consideration worldwide. Many have trans-uranium element (or transuranic or TRU) inventory reduction as a primary design objective. Some prominent examples include:⁴³

- Accelerator-Driven Systems (ADS) Accelerator-generated high energy protons are impacted onto a spallation target for the purpose of generating neutrons to drive fissioning of actinides in a subcritical core. This technique would allow a high concentration of actinides in the core (given the core's subcritical nature).
- Fission-Fusion Hybrids (FFH) Neutrons emitted by fusion of deuterium and tritium nuclei are harnessed for fissioning of actinides (and possibly breeding) in one or several blankets surrounding the fusion chamber.

2.2 Advanced Reprocessing

Except for the once-through fuel cycle, most nuclear fuel cycles rely on reprocessing in order to separate spent fuel into different waste and re-usable fuel material streams. Reprocessing can be

⁴³ Research Needs of Fusion-Fission Hybrid Systems, U.S. Department of Energy, Report of the Research Needs Workshop (ReNeW) Gaithersburg, Maryland Sept 30 – Oct 2, 2009

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characterized based on the different output streams generated. Currently, only the PUREX⁴⁴ process is available at a proven, commercial scale technology with three major operational facilities worldwide: La Hague, France (responsible for more than two-thirds of the commercial LWR fuel reprocessed to date), Sellafield, UK (which has historically reprocessed all of the UK's metallic MAGNOX spent fuel inventory as well as a small quantity of LWR oxide fuel), and Mayak, Russia. A fourth major facility, Rokkasho-Mura, Japan, which is largely based on the La Hague design, is not yet fully operational, although construction and substantial portions of testing have been completed. The well-established PUREX process separates spent fuel into three primary streams: reprocessed Uranium (RepU), Plutonium, and fission products (FPs) + minor actinides (MAs). The FP+MA fraction comprises the bulk of waste material destined for permanent disposal.

In spite of the historical reliance on the PUREX process, many innovative concepts of reprocessing have been developed, which offer different options in terms of major actinide, MA, and FP separations. These variations impact the number and character of nuclear materials suitable for recycling and for disposal as waste. To simplify the analysis, only actinide (U, Np, Pu, Am, and Cm) separation will be considered; consideration of FP separation and inventory reduction will be addressed later in the report.

The first six options in Table 2-2 are based on aqueous processes, which are well suited for reprocessing of oxide fuels. The seventh and last option, pyroprocessing, is designed to reprocess metal fuel, but can also be applied to oxide fuels by incorporating an initial reduction step. There is a significant economy of scale for aqueous reprocessing facilities; this feature favors large centralized facilities. Pyroprocessing is feasible at smaller scales and could, in principle, be implemented on a more distributed basis.

Co-extraction of U and Pu (COEX, UREX+2, NUEX) has been proposed as a means to improve fuel cycle proliferation resistance, given that separated Pu is considered to be highly attractive for use in an improvised nuclear device. However, this proliferation resistance is temporary if the two co-extracted U and Pu are subsequently separated using PUREX or some other process.

Separation of Cm and Am from each other is very difficult because these two species have very similar chemical properties. However, recycling of Am apart from Cm is very desirable, since Am represents a major source of decay heat in the context of waste disposal, while shorter-lived Cm isotopes greatly complicate handling in industrial facilities due to their high radioactivity and associated penetrating radiation.

These different reprocessing options typically allow for separation of some fission products, mainly Tc, I, Cs and Sr. Transmutation of fission products is possible, but presents technical challenges and questionable returns in terms of efficiency and risk reduction as opposed to disposal either as a segregated waste stream or as a co-disposed waste form. These challenges are discussed in greater detail in Section 4.

⁴⁴ PUREX = <u>P</u>lutonium and <u>UR</u>anium <u>EX</u>traction

Option	Description	Recycled Streams	Waste Streams
1	Once-Through	None	U+Np+Pu+Am+Cm+FP ^a
2	PUREX	U, Pu	Np+Am+Cm+FP
3	Evolutionary PUREX (COEX, UREX+2, NUEX)	U, U+Pu(+Np)	(Np+)Am+Cm+FP
4	Selective MA Separation (DIAMEX-SANEX, UREX+3, TOGDA)	U, U+Pu(+Np), Am+Cm	FP
5	Grouped MA Separation (GANEX, NEXT, UREX+1)	U, U+Np+Pu+Am+Cm	FP
6	Separation of Cm and Am from Each Other (UREX+4)	U, U+Pu(+Np), Am	FP, Cm (storage)
7	Pyroprocessing	U, U+Np+Pu+Am+Cm	FP

Table 2-2Main Reprocessing Options

^aAs spent nuclear fuel.

2.3 Waste Disposal

Several options, as illustrated in Table 2-3, have been considered for long-term management of used nuclear fuel and HLW and are covered in detail elsewhere.⁴⁵ Of the many options that in principle could meet the needs for long-term isolation of radionuclides from the accessible environment needed to protect human and environmental health, some have fallen from favor, others are either deemed too risky or costly, while others do not appear to be feasible from an international policy or law perspective. As a result of five decades of scientific consideration and debate, an international consensus exists today within the scientific community that geologic disposal is a safe and effective way to isolate used nuclear fuel and nuclear wastes over the requisite thousands to tens of thousands of years.^{46,47,48} All nations seriously pursuing a disposal program for used fuel or high-level radioactive waste are focused on mined geologic repositories as the technology of choice. International programs are listed in Table 2-4.

⁴⁵ Disposition of High-Level Waste and Spent Nuclear Fuel, National Academy of Sciences, National Research Council, National Academy Press, Washington, D.C., 2001.

⁴⁶ The Disposal of Radioactive Waste on Land. National Research Council, Publication 519, National Academy Press, Washington, D.C., 1957.

⁴⁷ Managing the Nation's Commercial High-Level Radioactive Waste. U.S. Congress, Office of Technology Assessment, OTA-O-171. Washington, D.C., 1985.

⁴⁸ Scientific Basis for the Geological Disposal of Radioactive Waste. Technical Report Series no. 413, International Atomic Energy Agency, Vienna, 2003.

Potential Technologies for Advanced Fuel Cycles

Table 2-3Long-term Storage and Permanent Disposition Options for Spent Fuel and High-levelRadioactive Waste**

Approach	Description	Advantages and/or Disadvantages
Surface storage	Long term storage provided by emplacement of waste into a suitable waste package, canister, cask, vault system under dry conditions	Requires monitoring and maintenance over entire storage period
	Currently practiced in U.S. and other countries on a short-term/interim basis	
Geologic disposal (mined repository)	Emplacement of packaged waste into mined repository at large depths in a suitable geological formation and environment (i.e., 100's of meters below surface)	Reference permanent disposal concept for most HLW management programs
Deep borehole disposal	Emplacement of packaged solid waste in boreholes drilled deep into crust far below groundwater influence	Retrieval of waste may not be feasible (can also be considered a benefit)
		Most feasible for small volumes (e.g., small inventories such as separated minor actinides)
Sub-seabed disposal	Emplacement of packaged solid waste in geologically stable deep ocean sediments	Retrieval of waste may not be feasible (can also be considered a benefit)
	or in sub-seabed rock formations	Likely to conflict with international policy and law
Deep well injection	Direct injection of liquid wastes into appropriate geological	For liquids wastes only Phased out in favor of other
	Tormation Used historically for injection of low-level wastes in U.S. and for intermediate level wastes in former Soviet Union	geologic disposal methods

⁴⁹NAS/NRC, 2001.

Approach	Description	Advantages and/or Disadvantages
Partitioning and transmutation	ioning and transmutation Exposure of very long-lived radionuclides, e.g., plutonium and minor actinides, to neutron fluxes resulting in transmutation to shorter lived radionuclides	
		achieve significant waste reduction benefits
Extraterrestrial disposal	Physical removal of waste from the earth through launch of waste form into space	Excessive risk due to probability of launch failure and number of launches required

Table 2-3 (continued)Long-term Storage and Permanent Disposition Options for Spent Fuel and High-levelRadioactive Waste

Of course, each precise concept depends on the site geology and requires very detailed studies. For the time being, only one geologic repository for radioactive wastes is in operation, the Waste Isolation Pilot Plant (WIPP) (near Carlsbad, New Mexico, U.S.A.) for disposal of defense-related transuranic wastes. WIPP is located in a deep salt deposit chosen for its hydrologically isolation and very low permeability. The bedded salt formation itself is known to have been stable since its deposition with the evaporation of an ancient ocean during the Permian Age some 250 million years ago. The fact that the salt deposit exists today is evidence that flowing groundwater, which would have dissolved the salt, has not been present over this period and will likely not be present for the time period required for decay of the transuranic wastes.⁵⁰

Table 2-4

Candidate Geology, Hydrology, and Host Country for Several High-level Radioactive W	Vaste
Repository Programs⁵¹	

Geology	Hydrology	Countries
Crystalline rock (e.g., granite, gneiss)	Saturated	Sweden, Finland, Japan
Agrillaceous rock (e.g., clay)	Saturated	France, Switzerland, Belgium
Salt	Isolated	U.S.A. (WIPP), Germany
Volcanic tuff	Unsaturated	U.S.A. (Yucca Mountain)

⁵⁰ http://www.wipp.energy.gov/fctshts/salt.pdf; accessed 20 October 2009

⁵¹ Adapted from Table I., Technical Reports Series no. 413, Scientific and Technical Basis for the Geological Disposal of Radioactive Wastes, International Atomic Energy Agency, Vienna, 2003.

Potential Technologies for Advanced Fuel Cycles

There is a priori no single best geology or design for a mined deep geologic repository; many different geological environments could prove to be suitable for hosting a repository, as indicated by the diversity in candidate sites among international programs, as shown in Table 2-4.⁵² Moreover, the ultimate performance of a repository will be driven by both the intrinsic properties of the geology and environment and by the features of the engineered barrier system (EBS), which can augment, supplement, and complement those of the natural system. Therefore, it is important to evaluate a potential host site in light of an appropriately matched repository design and components by focusing on site characteristics, engineering design, wasteform properties, maintaining "defense in depth," and keeping a prudent eye on the overall performance of the total system versus individual system components. Figure 2-3 illustrates the proposed disposal concept for a repository in unsaturated volcanic tuff at Yucca Mountain, NV, U.S.A.⁵³

Another geologic disposal concept for HLW is deep boreholes disposal, consisting of deep drilled bore holes on the order of several kilometers (typically 3 - 5 km deep). The waste packages are emplaced at the bottom of these holes below the influence of groundwater and covered by grout and backfill material (Figure 2-4). The advantage of this approach is that it can be implemented on a modular basis (e.g., one borehole at a time) and therefore could be altered or stopped at any time with little loss of investment. However, volume capacity is limited per borehole; accordingly, this option is particularly well suited for low volume applications such as the disposal of minor-actinide-bearing wasteforms as a complement to mined geologic disposal or other technologies.

⁵² See, for example, IAEA (2003) Technical Report Series no. 413 and Disposition of High-Level Waste and Spent Nuclear Fuel, National Research Council, National Academy Press, Washington, D.C., 2001.

⁵³ The Yucca Mountain program faces termination following intense political opposition and withdrawal of federal support



Potential Technologies for Advanced Fuel Cycles

Figure 2-3 Yucca Mountain Concept (Source: U.S. DOE/OCRWM)⁵⁴

⁵⁴ U.S. Department of Energy, Office of Radioactive Waste Management.



Figure 2-4 Deep Borehole Disposal Concept (Source: Swift et al., 2009)⁵⁵

⁵⁵ P. Swift et al., "Deep Borehole Disposal Concepts: Preliminary Assessment for the Disposal of Used Fuel Assemblies," DOE Fusion-Fission Hybrid Workshop, Gaithersburg, MD (October 1, 2009).

3 U.S. AND FRENCH NUCLEAR FUEL CYCLE STRATEGIES

After presenting the main challenges of a nuclear fuel cycle, describing the potential advanced technologies to be used, and before discussing the different strategic choices about nuclear fuel cycles, it is worth understanding how different energy contexts can explain different nuclear strategies. For this purpose, fuel cycles in France and the United States are considered. Indeed, these countries have the largest nuclear power generation, but have adopted fundamentally different approaches to the nuclear fuel cycle due to different national policy and security contexts. Japan, third in terms of nuclear power generation, has adopted a nuclear fuel cycle strategy similar to France's.

3.1 Energy Context of France

Knowledge of France's energy, political, and economic contexts is essential for understanding the French strategy for, and approach to, its pursuit of an advanced nuclear fuel cycle. In terms of energy supply and domestic energy security, France's reliance on nuclear generation for three-quarters of electricity supply (Figure 3-1: 76% in 2008⁵⁶) is the highest of any nation (Figure 3-2). This situation results from the execution of a deliberate national energy policy to promote energy independence and security in the wake of the global fallout from the Arab oil embargo of 1973.

3.1.1 Natural Resources

The French reliance on nuclear energy to generate electricity is driven to a large extent by the scarcity of domestic fossil resources: coal, gas, and oil. These resources are almost exhausted; the last operational coal mine in France was closed in 2004 in Lorraine.

Domestic uranium resources are also limited. In 1954, France began uranium production in Bessines, and in the 1970s, a total of five plants provided the majority of natural uranium requirements of operating nuclear power plants. However, since then, the gap between consumption and production of uranium has continued to widen, as shown in Figure 3-3.⁵⁷

⁵⁶ Source: DGEC (French administration)

⁵⁷ Source: 2007, NEA Red Book

U.S. and French Nuclear Fuel Cycle Strategies



Figure 3-1 Power Generation in France in 2008⁵⁶



Figure 3-2 Nuclear Share of Electricity Generation in 2008 by Country (Source: IAEA)⁵⁸

⁵⁸ Adapted from IAEA, Power Reactor Information System,

http://www.allcountries.org/rankings/nuclear_share_electricity_generation_by_country_2009.html



Figure 3-3 Uranium Consumption and Production in France in tU/year (Source: OECD/NEA)⁵⁹

Today, France has no reasonably assured resources of uranium as defined by the OECD/NEA Red Book, and inferred resources are estimated at 11,740 tU, which represents only 1.3 years of French requirements.

3.1.2 Nuclear Fleet

The make-up of the French fleet is shown in Figure 3-4. The first French nuclear reactors were Gas-Cooled Reactors designed by the CEA (Commissariat à l'Energie Atomique) in the mid-1970s. The French state-owned utility, EDF, licensed Westinghouse PWR design technology and subsequently built a fleet of 58 PWRs from 1977 to 1999. One commercial sodium-cooled fast reactor (Superphenix) was also built in 1986, but was shutdown in 1998 following chronic operational and maintenance challenges; the reactor is currently being decommissioned.

One new 1,650 MW Generation III+ reactor of the AREVA Evolutionary Pressurized Reactor (EPR) design is under construction in Flamanville, Normandy. This is the second of its kind globally (the first being constructed in Finland). A second French EPR is planned for construction in Penly, Normandy, starting in 2012.

To illustrate the demand for replacement nuclear power plants in the future, the lifetime for each operating reactor in the French fleet is assumed to be a uniform random variable between 40 and 80 years.

Hence, based on this projection, decommissioning of existing plants (Figure 3-5) and construction of new plants will become a major concern for the French nuclear sector around

⁵⁹ Adapted from OECD/NEA "Forty Years of Uranium Resources, Production and Demand in Perspective," OECD 2006.

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2025 and will last for many decades - beyond the 2050 timeframe. At that time, uranium resources may become a major concern, especially in light of the fact that any new reactors should be expected to operate for at least 60 years. Consequently, there is a significant incentive for France to deploy a limited number of Fast Breeder Reactors during this period.



Figure 3-4 Composition of the French Nuclear Power Plant Fleet



Figure 3-5

Expected Number of Nuclear Power Plants to Be Replaced in France (Number Averaged over a Five-Year Period of Time)

3.1.3 CO₂ Emissions

Because of the large share of nuclear energy in France's energy mix, French CO₂ emissions are significantly lower than other European countries. For example, France's electric power sector

emits just 14% of the CO_2 emitted by Germany per unit of GDP. This low carbon footprint for the electricity sector translates into an overall position of strength in terms of complying with European Union's targets, meeting international commitments, and providing a favorable economic position for industry in a carbon-constrained marketplace.

3.1.4 Outcomes of French Nuclear Energy Policy

The stability of the French energy policy is the result of a relative consensus among the French government on this issue and by the absence of significant policy changes in the administration after each election. Along with this stability, France has built a powerful nuclear industry dominated by two state-controlled corporations: Electricité de France (EDF), the sole owner/operator for all nuclear plants in France; AREVA, a leading global nuclear services provider and technology vendor. The size and integration of its industry allows for a high degree of standardization nationally and for significant influence on the global nuclear industry through export of French nuclear technology. The heavy reliance on nuclear generation also means that France releases much lower quantities of carbon dioxide than other industrialized nations. Therefore, nuclear plays a major role in French energy, industrial and environmental policies. One tangible result is that France is the world's largest net exporter of electricity.⁶⁰

3.2 French Strategy on Advanced Nuclear Fuel Cycles

Uranium sustainability has always been a major driver for the French strategy on the nuclear fuel cycle. Initially, this interest was motivated by the scarcity (and eventual exhaustion) of domestic uranium resources and the widespread belief that global uranium resources were limited and would threaten the reliability of the global nuclear fuel supply. This view of the global uranium market proved to be overly pessimistic, as evidenced by an extended period of low uranium spot prices. However, energy, economic, and environmental concerns have continued to evolve since the early days of commercial nuclear power, and the French strategy has also evolved over the past 40 years in response to these concerns.

3.2.1 Key Players in the French Nuclear Sector

In addition to the central roles played by the ministerial and parliamentary setting and execution of French nuclear policy, five major actors play key roles in the definition and implementation of the national nuclear policy in France: the Nuclear Safety Authority (ASN), the Commission of Atomic and Alternate Energy (CEA)⁶¹, the National Radioactive Waste Management Agency (ANDRA), AREVA, and Electricité de France (EDF).⁶² These organizations are described in Table 3-1.

⁶⁰ WNA, 2010. Nuclear Power in France. World Nuclear Association. < http://www.world-nuclear.org/info/inf40.html > February 2010. Accessed 24 February 2010.

⁶¹ Formerly the French Atomic Energy Commission

⁶² WNA, 2010. Nuclear Power in France. World Nuclear Association. < http://www.world-nuclear.org/info/inf40.html > February 2010. Accessed 24 February 2010.

The descriptions in Table 3-1 indicate the high level of state involvement in every element of the French nuclear sector. However, recent pressure within the European Union for greater liberalization of the energy sector has led to the emergence of new private actors on the nuclear scene. Notable in France among new actors is the utility GDF-Suez, which is also partially state-owned (35%).

Organization	Description	Function	U.S. Analog
ASN	Independent nuclear regulatory authority	Provide regulatory oversight of nuclear industry in France to ensure protection of public and environment through safe use of nuclear technology, including nuclear power plants and other fuel cycle facilities	Nuclear Regulatory Commission
CEA	French government-funded public research organization	Research on advanced reactor technologies, advanced reprocessing and waste management	Department of Energy
AREVA	French nuclear technology and services vendor, predominantly State-owned	Uranium mining and milling, enrichment, fuel fabrication, LWR design and construction and reprocessing	Commercial vendors
EDF	French electric power utility, predominantly State-owned	Construction, operation and maintenance of nuclear power plants – exclusive owner-operator of nuclear plants in France	Investor-owned, publicly-owned, consumer-owned, and Federal electric utilities
ANDRA	French government agency	Management of nuclear waste	Department of Energy

Table 3-1				
Key Organizations I	nvolved in	the French	Nuclear	Sector

3.2.2 Open or Closed Fuel Cycle?

Originally, the strategy of France, as with every country pursuing nuclear power in the early days of the technology, was to close the fuel cycle. This strategy consists of irradiation of low-enriched uranium in LWRs, separation of Pu from the spent LWR fuel, and subsequent use of this Pu to feed a fleet of fast breeder reactors (FBRs). FBRs use ²³⁸U (~99.3% of natural uranium) to produce more fissile material than they consume. This technology allows for greater utilization of the full energy content of uranium resources instead of being mostly restricted to the fissile content of natural uranium – 0.711% of ²³⁵U – as is the case with thermal reactor technologies.

Two important factors have led to changes in this original strategy.

- First, uranium has proven to be much more abundant than originally expected. For example, in 1996 there was less than 1.5 MtU of Reasonably Assured Resources (RAR) below \$130/kgU; this figure more than doubled to >3 MtU of RAR below \$130/kgU in 2002.
- Second, development of the requisite fast reactor technology did not keep pace with the deployment of reprocessing technology. The success in reprocessing LWR fuel, but with no concurrent commercial deployment of a fleet of FBRs, resulted in the buildup of an increasingly large stockpile of separated Pu.

France began reprocessing for its nuclear weapons program in 1958 with the construction and startup of the UP1 plant in Marcoule. CEA was then directed by the French government in 1961 to construct a second reprocessing plant in La Hague to provide redundant capacity for strategic Pu separation from irradiated UOX fuel. Support for the La Hague facility waned in the late 1960's as military Pu stockpiles increased. The first oil crisis reinvigorated interest in La Hague, this time for a civilian mission for Pu recovery from spent PWR fuel for production of mixed-oxide (MOX) fuel for electricity generation, and in 1974, the French government approved construction of the UP2-400 facility at La Hague. COGEMA took ownership of this facility from CEA in 1976 and expanded reprocessing capacity with the startup of the UP2-800 and UP3 plants. To date, more than two-thirds of all LWR fuel reprocessing worldwide has occurred at La Hague.

In light of the lagging development of FBR technology and maturation of reprocessing and Pu recycling technologies, France changed its near-term emphasis from deployment of a closed fuel cycle to the full-scale implementation of a fuel cycle in which Pu is recycled once as MOX fuel in LWRs to yield a 10-15% saving in natural uranium. However, the ultimate goal of the French strategy remains to reprocess spent MOX (and spent UOX that has not been reprocessed) and recycle plutonium, and possibly other actinides, in a fast breeder reactor fleet in order to reap the full energy benefits of the technology.

To achieve this longer-term goal, the French government announced in 2006 that the CEA would construct a fourth generation (GEN IV) fast reactor prototype; the project is to be completed in 2020. This prototype will likely be an evolution of the Superphenix sodium-cooled fast reactor. Initial introduction of a commercial-scale fast reactor is expected in the 2040 timeframe.

Concerning reprocessing, three main R&D lines (not necessary exclusive from each other) are kept open by the CEA: evolution of the PUREX process to co-extract U and Pu and possibly Np (COEX process), selective separation of minor actinides (DIAMEX-SANEX process), and group extraction of all actinides for homogeneous recycling in fast reactors (GANEX).

3.2.3 What about the Waste?

ANDRA, the French National Agency for Waste Management, was separated from CEA in 1979 and is responsible for the management of nuclear wastes.

In 1991, a law ("Loi Bataille") was passed by the French Parliament to define three areas of waste management research. The first, conducted by CEA, is the separation and transmutation of long-lived radioactive materials contained in the waste (minor actinides and some fission

products). The second area, conducted by ANDRA, concerns the final disposal of nuclear waste in a geologic repository. The third area, conducted by CEA, involves long-term (up to 300 years) interim storage of nuclear waste. The Bataille law specified a 15-year period after which the French Parliament was directed to pass follow-up legislation to clarify the path forward for the French waste management program. During this time period, ANDRA opened an underground research laboratory in a clay formation in Bure (Meuse), France. The 2006 enactment of clarifying legislation has directed French nuclear waste management research to:

- Complete by 2012 an evaluation of the industrial prospects for partition and transmutation (P&T) of the minor actinides in fast neutron spectrum reactors;
- Complete construction of a sodium-cooled fast reactor prototype by 2020;
- Receive approval for and operate a permanent geologic by 2015 and 2025, respectively; and
- Complete studies on interim storage before 2015 to allow for timely development of new or modification of existing storage facilities.

3.3 Conclusions on French Nuclear Energy Strategy

France has deliberately focused its research on advanced fuel cycles to address uranium sustainability concerns and waste management challenges. In one proposed scenario⁶³ for the 21st century, the current LWRs and new Evolutionary Pressurized Reactors (EPRs) (capable of operating with UOX and MOX fuels) are envisioned to continue to dominate the French fleet through 2040. Deployment of a fast reactor fleet would start during the decade staring in 2040 eventually resulting in reliance on a fleet composed exclusively of fast reactors by the end of the century. This scenario, if realized, would make France almost entirely energy independent with respect to electric power generation. This outcome is conceivable if the continuity and the perseverance of the French nuclear policy over the last 50 years – spanning six presidential administrations – can continue for another multi-decade period, given that such a continuity in national policy is likely to be required to develop and deploy a closed nuclear fuel cycle.

3.4 Energy Context of the United States

The political, economic, and social contexts for the U.S. nuclear industry are significantly different from those of the French nuclear sector and largely explain the striking differences between the nuclear fuel cycle policies currently implemented in the two countries as well as the differences in approach with regard to advanced nuclear fuel cycles.

3.4.1 Nuclear Share in Electricity Generation

The U.S. operates the largest nuclear fleet in the world, with 104 units in operation, but nuclear power produces only one-fifth of the total electric generation (19% in 2008), as shown in Figure 3-6. This energy mix is the reverse of that in France with respect to reliance on nuclear.

⁶³ Source: EDF/CEA/AREVA "Potential contribution of fast reactors and new reactor concepts to fissile materials and minor actinides management"



Figure 3-6 Electricity Generation in the U.S. in 2008 (Source: U.S. EIA)⁶⁴

3.4.2 Natural Resources

The predominant reliance of the U.S. electricity sector on fossil fuels (and continuing inertia in shifting away from this dependence) can to some degree be explained by the presence of substantial domestic fossil resources (coal, gas, and oil). In fact, the U.S. is currently reported to possess the largest coal resources in the world (273 billions tons, about 273 years of current U.S. consumption),⁶⁵ the sixth most important natural gas resources, and the tenth most important oil resources.

The U.S. also possesses domestic uranium resources. The U.S. began industrial scale uranium production as early as 1947 for defense programs. This production steadily increased throughout the 1970's to meet the civilian reactor demand. Over the last fifteen years, a deficit between domestic consumption and production has emerged and widened (Figure 3-7). This is largely because of the availability of inexpensive imported ore, such as that available from Canada.⁶⁶

⁶⁴ Data from U.S. Energy Information Administration.

⁶⁵ U.S. EIA, 2010. Coal Reserves Data for 2008. 17 February 2010,

<http://www.eia.doe.gov/cneaf/coal/reserves/reserves.html> accessed 6 April 2010.





As of 2007, the U.S. has 342,000 tU of reasonably assured resources, which corresponds to 15 years at current domestic consumption rate, and an estimated 3,452,000 tU of undiscovered resources, which represents 150 years at current U.S. consumption rate. Hence, uranium is relatively abundant and could become available again, if uranium prices were to rise substantially. At current prices, most of the U.S. uranium resources remain largely untapped.

3.4.3 Nuclear Fleet

The U.S. nuclear fleet is currently composed of 104 nuclear power plants, 35 boiling water reactors (BWRs) and 69 pressurized water reactors (PWRs), which began operating between 1969 and 1996, as shown in Figure 3-8.

U.S. and French Nuclear Fuel Cycle Strategies





No new plants have been brought online since 1996 following the cancellation or mothballing of a large number of projects due to increasing capital costs and a precipitous drop in public support for nuclear energy following the accidents at Three Mile Island Unit 2 in the U.S. and at Chernobyl Unit 4 in what is now Ukraine. As with the French fleet, Figure 3-9 illustrates the anticipated demand for replacement of aging nuclear power plants in the future, assuming the operating lifetime to be a uniformly random variable between 40 and 80 years.



Figure 3-9

Expected Number of Nuclear Power Plants to Be Replaced in the U.S. (Number Averaged over a Five-Year Period of Time)

Hence, decommissioning of existing plants and construction of new ones are expected to become major activities from 2015 on, lasting until around 2060. In the 2015-2040 timeframe, it will be necessary to replace more than 60% of the existing fleet (compared to less than 50% of the existing French fleet over the same timeframe). In addition, the new reactors are expected to conform exclusively to evolutionary or passive LWR designs, generically designated as Advanced Light Water Reactors (ALWRs). Accordingly, the prospects for commercial deployment of fast reactor technology appear much less likely than in France.

3.4.4 CO₂ Emissions

Because of its large dependence on fossil generation, particularly domestic coal, the carbon footprint of the U.S. electricity sector has grown to be the largest emitter of CO_2 among all U.S. sectors. As electricity demand continues to grow in the U.S., so will the CO_2 emissions without a substantial shift to low carbon energy sources or the introduction of adequate levels of carbon sequestration capacity. EPRI has published a number of studies exploring feasible energy technology mixes that could achieve target reductions in CO_2 emissions under a range of scenarios.⁶⁷. By extension, the U.S. electricity sector is a major reason for the U.S. top ranking in per capita CO_2 emissions in the world.

3.4.5 Outcomes of U.S. Energy Policy

The U.S. approach to nuclear power and the resulting nuclear sector as it exists today represent the antithesis of the situation in France. After a significant rise in the 1970s and the 1980s, nuclear power growth has been flat since the 1990s. U.S. utilities have forgone new plant construction and instead have focused on increasing plant availability and capacity (through power uprates). As a result of power uprates alone, more than 5,700 MWe of nuclear capacity has been added since 1977 – the equivalent generation from 5 or 6 new nuclear plants.⁶⁸

In the interim, domestic U.S. reactor technology vendors have disappeared, diminished in stature, been sold to foreign interests, or have formed joint ventures with foreign entities in order to survive the stagnation in nuclear growth in the U.S. Meanwhile, government support for expansion of nuclear power has been inconsistent over the past four decades, resulting in a lack of vision, direction, and of the overall leadership that has defined the French program, especially since the 1991 Bataille Law.

The cessation of commercial reprocessing of spent nuclear fuel in the U.S. in the late 1970's due to proliferation concerns effectively locked U.S. policy and industry into a once-through nuclear fuel cycle. Although the administrative ban on reprocessing was reversed in the 1980's, U.S. government promotion of recycling technology and broader encouragement of reprocessing technologies, albeit ones emphasizing proliferation resistance, would not be revisited by the U.S.

⁶⁷ EPRI Prism/MERGE Analyses – for example: The Power to Reduce CO2 Emissions: The Full Portfolio – 2009 Technical Report. EPRI, Palo Alto, CA: 2009. 1020389.

⁶⁸ U.S. Nuclear Power Plants: Resources and Statistics. Nuclear Energy Institute, June 2010.

http://www.nei.org/resourcesandstats/nuclear_statistics/usnuclearpowerplants/ accessed 6 August 2010.
for two more decades. The past decade showed a potential change in the U.S. government policies, as evidenced by the following developments:

- The 2000 U.S.-Russia Plutonium Management and Disposition Agreement at the origin of the program to fabricate LWR MOX fuel from weapons grade Pu;
- The now defunct domestic element of the Global Nuclear Energy Partnership (GNEP) launched in 2006; and
- The 2009 announcement and 2010 formation of a cabinet-level Blue Ribbon Commission to re-examine fuel cycle options following the announcement of the termination of the Yucca Mountain program.

The Energy Policy Act of 2005 laid the groundwork for government-backed loan guarantees for the first new builds, and in 2010 the first of these loan guarantees was awarded to Southern Company for construction of Vogtle Units 3 and 4 near Augusta, Georgia.

On one hand, the economic competitiveness of coal and natural gas to produce electricity and the substantial domestic fossil resources limit the economic and political incentives for assuming the large risk and capital costs associated with construction of new LWRs, not to mention other nuclear fuel cycle facilities such as reprocessing plants. On the other hand, increasing awareness of, and concern about, climate change among the U.S. public and political leaders, as well as concerns about energy security, has changed the social and political landscape regarding acceptance of nuclear as a low carbon alternative for base-load generation of electricity.

In all cases, optimistic outlooks on uranium availability and the high reliability and profitability of the current LWR fleet favors a "more of the same" approach for the near term deployment of new nuclear plants and argues against any aggressive pursuit of advanced nuclear fuel cycle technologies, especially given the decentralized nature of the U.S. electric power industry, comprising many utility companies with fairly modest capitalizations compared to a national utility such as EDF. From a national security policy perspective, nuclear proliferation concerns remain a formidable obstacle to rapid U.S. deployment of commercial scale reprocessing.

3.5 U.S. Strategy on Advanced Nuclear Fuel Cycles

The U.S. strategy on the development and deployment of advanced nuclear fuel cycle technology has experienced many changes in the last 50 years, including a number of disruptive course changes due primarily to public and political pressures. As with France and other nations that have seriously pursued nuclear power for electricity generation, the initial goal was to maximize existing (and supposedly dwindling) natural uranium resources. However, with the lessening of uranium sustainability concerns, the primary focus of the U.S. program fell on resolving non-proliferation and waste management issues.

3.5.1 Key Players in the U.S. Nuclear Sector

In addition to the broad, central roles played by the U.S. Congress and Executive Branch in defining and executing nuclear energy policy, four major actors/groups play important roles in the definition and implementation of the national nuclear policy in the U.S.: the Nuclear

Regulatory Commission (NRC), the Department of Energy (DOE)⁶⁹, the nuclear technology and service vendors, and the utilities. These organizations and their roles are described in Table 3-2.

Private interests are much more represented in the U.S. than in the French system. However, major decisions on nuclear fuel cycle policy, research, and strategy remain a function of the U.S. government either due to statutory authority, the business risks involved, or the level of capital investment required.

3.5.2 The Department of Energy as a Focus of U.S. Advanced Fuel Cycle Strategy

The U.S. DOE plays a multifaceted role in the pursuit of advanced nuclear technology.

The Fuel Cycle Research and Development program (formerly known as the Advanced Fuel Cycle Initiative) aims at closing the Nuclear Fuel Cycle by focusing on safety, reduction in the long-term radiotoxicity of nuclear waste, and non-proliferation. This long-term research program seeks to reduce the nuclear waste burden from hundreds of thousands years to centuries by recycling the minor actinides.

The Generation IV (or GEN IV) International Forum (GIF) is an international collaboration on advanced nuclear reactors in which the U.S. is involved. One of the main concerns of this program is resource availability and sustainability. Half of the GEN IV concepts are fast reactors that could potentially breed Pu. The U.S. is interested in five of the six GEN IV technologies, two thermal reactor concepts [Very High-Temperature Reactor (VHTR) and Super-Critical Water Reactor] and three fast reactor concepts [Gas-Cooled Fast Reactor (GFR), Lead-Alloy Fast Reactor (LFR) and Sodium Fast Reactor (SFR)], with a high priority given to VHTR because it could produce electricity and hydrogen, and SFR because it is the most mature fast reactor concept.

⁶⁹ Originally, the functions of the NRC and the DOE fell under the U.S. Atomic Energy Commission. However, the regulatory function was separated from the energy research and the development of nuclear technology and weapons with the passage of the Energy Reorganization Act of 1974. The Act led to the 1975 establishment of an independent nuclear regulator, the NRC, and a separate Energy Research and Development Agency (ERDA). ERDA was subsequently reorganized into the twelfth cabinet office, the Department of Energy, in 1977.

Organization	Description	Function
NRC	Independent nuclear regulatory authority	Provide regulatory oversight of commercial nuclear industry in U.S. to ensure protection of public and environment through safe use of nuclear technology, including nuclear power plants and other fuel cycle facilities
DOE	Department of Energy and contractor-operated national laboratory system	Research and development of advanced reactor technologies (GEN IV), advanced fuel cycle technologies, and waste management.
		Receipt and disposal of used nuclear fuel from commercial power industry and high-level waste from weapons production programs through siting, licensing, and operation of a geologic repository (per the Nuclear Waste Policy Act of 1982, as amended).
		Environmental management and clean up of contaminated sites associated with former weapons production and energy research programs.
		Also retains some self-regulatory authority for non- commercial nuclear activities from Atomic Energy Act of 1954, as amended.
AREVA GE-Hitachi Toshiba- Westinghouse, et al.	Commercial nuclear reactor technology and service companies	Reactor design, nuclear fuel fabrication, and other nuclear services
Electric Power Utilities	Investor-owned, publicly- owned, consumer-owned, and Federal electric utilities	Construction, operation, maintenance, and decommissioning of nuclear power plants

Table 3-2Key Organizations Involved in the U.S. Nuclear Sector

3.5.3 Moving from a Once-Through Fuel Cycle to a Closed Fuel Cycle

The U.S. was the first country to use nuclear energy for military purpose and also the first to apply nuclear power for civilian generation of electricity. The PUREX process, the foundation of modern aqueous reprocessing technology, was developed at Oak Ridge National Laboratory in 1949, and in 1954 the first industrial scale reprocessing plant using PUREX, the "F Canyon", began operating at the Savannah River Plant in South Carolina. In 1953, in his famous "Atoms for Peace" speech, U.S. President Eisenhower defended peaceful use of nuclear technology and the development of civilian nuclear power in the world. For this purpose, the International Atomic Energy Agency (IAEA) was created four years later to promote the peaceful uses of nuclear technology and the development of purpose.

At that time, the U.S. nuclear policy was geared to promote a significant growth of nuclear energy and to rely on reprocessing to accumulate Pu and then start a fast reactor fleet, given the generally accepted scarcity of adequate uranium supplies worldwide. As with France, improving prospects for natural uranium resources and setbacks in the development of fast reactor technology began to shift the U.S. fuel cycle policy and strategy in the early 1970s. The 1974 test of a nuclear device by India using western technology obtained under the auspices of the Atoms for Peace program resulted in a dramatic shift of U.S. policy away from commercial reprocessing with the intent of discouraging further proliferation of technologies applicable for weapons production. This watershed event motivated the Ford and Carter administrations to end U.S. support for commercial reprocessing in the U.S. and moved U.S. foreign policy to discourage foreign countries from pursuing similar technologies. This major policy shift effectively institutionalized the commercial once-through fuel cycle, in which irradiation of UOX assemblies would be followed by permanent disposal. From a commercial standpoint, pursuit of an exclusive once-through fuel cycle was accepted on the basis of simplicity and favorable economics, given the abundance, availability, and low price of natural uranium.

In 2006, the Bush administration initiated a dramatic change in non-proliferation policy with the launch of the Global Nuclear Energy Partnership (GNEP), whose domestic agenda evolved into a plan to close the fuel cycle that was predicated on proliferation resistance and reduction in nuclear waste. The initiative eventually included plans to construct and operate a demonstration scale reprocessing plant in the U.S. However, following the 2009 change in administration, the domestic element of GNEP was terminated.

3.5.4 What about Yucca Mountain?

One missing, major piece of the once-through cycle is the disposition pathway providing either direct permanent disposal, or isolation of the spent fuel generated by the U.S. fleet of LWRs. As waste disposal represents an essential element of all nuclear fuel cycles, open or closed, the U.S. had been evaluating concepts as early as the 1950's and performing field investigations with the intent of siting a geologic repository in the 1970's and 1980's. In 1982, the Congress enacted the Nuclear Waste Policy Act (NWPA), making DOE responsible for siting, construction, licensing, and operating a geologic repository for commercial spent nuclear fuel and used fuel and high-level radioactive waste from defense programs. Amendment of the NWPA in 1987 summarily ended the site selection process by designating Yucca Mountain, Nevada, as the sole subject of DOE's site evaluation program.

The NWPA required electric utilities (through their customers) to pay 1/10th of a cent per kWhr of nuclear power generated into a Nuclear Waste Fund to cover the cost of the repository program. As of 2010, cumulative contributions to the Fund and interest exceed \$33 billion.⁷⁰ For its part, the Federal government agreed to begin removing used nuclear fuel from commercial reactor sites beginning in 1998 – a contractual timeline explicitly incorporated in a formal arrangement between the government and nuclear utilities known as the "Standard Contract" for disposal of commercial spent nuclear fuel.

⁷⁰ NEI, 2010. Key Issues: Repository Development. Nuclear Energy Institute.

<http://www.nei.org/keyissues/nuclearwastedisposal/yuccamountain/> Accessed 25 February 2010.

Congress and the Bush Administration formally approved Yucca Mountain in 2002 as the first national repository site following DOE confirmation of the site suitability. DOE submitted a license application for construction of the repository to the U.S. Nuclear Regulatory Commission (NRC) in June 2008. In early 2009, the Obama administration indicated that "nuclear waste storage at Yucca Mountain is not an option"⁷¹ and accompanying policy shifts have effectively terminated the Yucca Mountain program, although the licensing process has continued. This major shift in U.S. waste policy has been accompanied by the formation of a "Blue Ribbon Commission" charged with re-evaluating the options and alternatives for managing the U.S. inventory of commercial used nuclear fuel.

3.6 Conclusions on U.S. Nuclear Energy Strategy

Whatever choices are made about advanced nuclear fuel cycles, it is highly likely that the U.S. utilities will continue to rely on LWR technology in the coming decades. While fuel cycle closure promises enhanced natural resource utilization on a scale sufficient to address both increasing energy demands and environmental constraints, U.S. energy policy and strategic interests remain in flux due to short term oscillations in political support and leadership and insufficient economic incentives needed to put the U.S. energy sector on a path toward long-term investment in R&D and nuclear infrastructure, which would be needed to seriously pursue Pu recycle and fuel cycle closure.

⁷¹ February 26, 2009 release of administration's draft budget reveals severe cuts to Yucca Mountain program; DOE press secretary announces "nuclear waste storage at Yucca Mountain is not an option." March 5, 2009 – Energy Secretary Chu's remarks at senate hearings confirm the "not an option" position and suggest "blue ribbon commission" formation.

4 ADVANCED NUCLEAR FUEL CYCLE STRATEGIC CHOICES

In Sections 1 and 2, the challenges of pursuing advanced fuel cycles and available or foreseeable technologies were presented to define and bound the problem at hand. Section 3 provided two real world examples and lessons by examining the U.S. and French programs resulting from national policies leading to distinct technology tracks toward either a once through or a closed fuel cycle.⁷² In light of the challenges, technology, and history of nuclear energy in the industrialized world as well as the coming energy demands, resource limitations, and external pressures (e.g., from climate change concerns), this section evaluates a number of paths forward that appear most promising and viable for implementation in the 21st century.

4.1 General Presentation of Advanced Nuclear Fuel Cycles

The aim of advanced fuel cycles is to improve the sustainability of nuclear energy by enhancing the effectiveness of natural uranium resource utilization and by mitigating waste disposal issues, while keeping the costs of energy products, in particular electricity, economically viable. In addition, this aim has to be achieved under conditions that minimize the risks of diversion of separated fissile materials and their possible misuse for non-peaceful ends.

The term "nuclear fuel cycle" can encompass a diverse collection of strategies and technologies, ranging in complexity from a once-through cycle to a fully-closed cycle incorporating fast breeder reactors and recycling of Pu and minor actinides for maximum energy recovery from natural uranium resources. The terms *open* and *closed* fuel cycles are often associated with different understanding by different authors. In this report, a *closed* cycle exists if no plutonium is intentionally sent for permanent disposition (disposal). *Partially closed* cycles feature cycles that are fully closed for plutonium, but neptunium is always transferred to waste, and treatments of americium and curium are variable. *Fully closed* cycles recycle all actinides continuously until they fission, and only actinide processing losses are transferred to waste. The once-through fuel cycle and Pu single-recycling in LWRs followed by disposal of the spent MOX fuel are *open* cycles.

The following schematic (Figure 4-1) has been developed to capture the present, anticipated, and potential (future) nuclear fuel cycle elements in a single, consistent framework.⁷³

⁷² It is worth noting that this characterization applies only to the current status and the situation is clearly subject to change, particularly in the U.S. where fuel cycle options are being re-examined following major changes to the U.S. HLW management program and the apparent resurgence in support for new nuclear plant construction.

⁷³ Schematic developed by EPRI

Advanced Nuclear Fuel Cycle Strategic Choices





Present (LWR Power Block and Managed Storage), Anticipated (Geologic Repository), and Potential (FBR Power Block) Nuclear Fuel Cycle Elements

4.1.1 LWR Power Block

More than 85% of the installed nuclear capacity consists of pressurized and boiling water reactors (BWRs). The head-end infrastructure (uranium mining and milling, conversion, enrichment, and fuel fabrication) is well established. The LWR technology makes very limited use of the potential energy content of natural uranium resources by using less than 1% of the mined uranium.

Some general features, challenges, and considerations associated with the implementation of an open fuel cycle, as depicted in Figure 4-2, are:

- Overall simplicity, industrial maturity, and widespread commercial implementation
- High nonproliferation credentials, but increasing concerns about enrichment technology
- Potentially constrained by uranium availability, given its poor uranium resource utilization, with less than 1% of potential energy from natural uranium resources recovered



Figure 4-2 Illustration of Open Nuclear Fuel Cycle Options

4.1.2 Managed Storage

The fuel discharged from LWRs is either placed in interim storage for several decades (as in the United States, Sweden, Finland, South Korea, Taiwan, Canada, Brazil, Argentina, and many other countries), or reprocessed (as in France and Japan). Interim storage of used LWR fuel has been implemented in centralized facilities (Sweden) or at the reactor sites (United States, Germany). Reprocessing employs the plutonium and uranium extraction (PUREX) process and results in three main products: reprocessed uranium, reactor-grade plutonium, and wastes. The reprocessed uranium and plutonium can be recycled in existing LWRs, resulting in potential

natural uranium savings of up to about 25%. The used fuel derived from using recycled uranium or plutonium is then placed in interim storage. Among the different waste streams, vitrified HLW, containing the fission products and the minor actinides neptunium, americium, and curium dispersed into a glass matrix, is also placed in interim storage. The end result for both options is thus interim storage. The technology and facilities to implement interim storage, reprocessing and fuel refabrication have been deployed at commercial scale. The managed storage block is generic to all fuel cycles as it does not presuppose (or preclude) any endpoint for used fuel, whether UOX or MOX in origin.

4.1.3 FBR Power Block

By recovering the plutonium available in used fuel, use of U-238 can be fully enabled in fast reactors: the plutonium is consumed and regenerated from the U-238. The leading design is the sodium-cooled fast reactor operating in the near-breeder or breeder mode. Reprocessing of used FBR fuel and fuel re-fabrication are required. Depending on the reprocessing scheme, separation and transmutation of some long-lived fission products and minor actinides can be contemplated. The largest operating fast reactor is presently the Russian BN-600 (1470 MWth), fueled with enriched uranium and operating since 1980. An advanced design, the BN-800 is scheduled for operation in 2016 and will be fueled with mixed uranium and plutonium oxide. First criticality of the 65-MWth China Experimental Reactor (CEFR) was achieved in July 2010. Operation of the 714-MWth Monju reactor in Japan was re-initiated in May 2010. Initial criticality of a 500-MWe prototype fast breeder reactor (PFBR) in India is scheduled by the end of 2011.

Advanced reprocessing technologies based on the PUREX process are being developed in several countries. The two main options being pursued are selective separation of minor actinides for heterogeneous⁷⁴ recycling in fast reactors and group actinide separation intended for homogeneous⁷⁵ recycling in fast reactors. Also, innovative methods based on electro-chemistry are being developed as integral parts of the refueling/waste management system of specific types of fast reactors. These methods allow for the treatment of different types of highly radioactive fuels with high plutonium content. Commercial deployment of these technologies is not likely for several decades.

Some general features, challenges, and considerations associated with the implementation of a closed fuel cycle, as depicted in Figure 4-3, are:

- Fast reactor fleet required
- Complex technical challenges yet to be addressed
- Much higher utilization of natural uranium resources
- Non-proliferation concerns
- Potential for reducing long-lived radioactive waste burden, but repository still required

⁷⁴ Meaning that the minor actinides and nuclear fuel are packaged separately

⁷⁵ Meaning that the minor actinides are incorporated into the nuclear fuel



Figure 4-3 Illustration of Multiple Closed Nuclear Fuel Cycle Options

Other advanced concepts, such as accelerator-driven systems, traveling wave reactors, and fission-fusion hybrids, could play pivotal roles in the benefits and sustainability of future nuclear fuel cycles as game changing technologies; however, their deployment on a commercial scale is unlikely in the coming decades. Therefore, this report does not consider them in its analysis.

4.1.4 Geologic Repository

All options require a geologic repository. There is broad agreement among the technical community that deep geological disposal constitutes a safe option for the relatively small volumes of HLW (including used fuel) generated by the nuclear power plants. The safety case for an HLW repository requires extensive R&D (regarding site suitability and waste packaging, for example), because the final selection of a site and disposal concept will be challenged from every possible angle. However, technical issues are generally not the limiting timing factors. Societal and political acceptance of these systems is currently the limiting factor for implementation in most countries.

4.2 Discussion of Five Nuclear Fuel Cycle Choices

Strategic decisions on nuclear fuel cycles are essentially limited to a small number of choices by external factors and intrinsic characteristics. In this report, these choices have been narrowed to five for the sake of simplicity, as shown in Figure 4-4. These five options are embedded in Figure 4-1.



Figure 4-4 Five Major Strategic Choices for a Nuclear Fuel Cycle

4.2.1 Managed Storage of Spent UOX Fuel

As illustrated in Figure 4-5 below, irradiated LWR UOX fuel with a burnup of ~50 GWd/MTHM comprises approximately 94% uranium, 1% plutonium, 0.1% minor actinides and 4-5% fission products. Roughly 2% of the isotopic inventory of spent UOX fuel is fissile (²³⁵U, ²³⁹Pu, and ²⁴¹Pu) and 93% is fertile (²³⁸U). Hence, used UOX fuel can be considered as a potentially valuable energy resource, especially for reactors capable of making full use of ²³⁸U.



Figure 4-5 Illustrative Composition of Spent Uranium Oxide Fuel (nominally ~50 GWd/MTU)

In the context of the first strategic choice (Strategic Choice #1 in Figure 4-4), the issue is to assess the value of re-using the fissile content of the used fuel discharged from the operating fleet of LWRs.

This assessment requires consideration of Strategic Choice #3 (Open or Closed Fuel Cycle).

Open Fuel Cycles – Comparison between Interim Storage of Spent UOX Fuel and Mono-Recycling of Plutonium

In the context of open fuel cycles operating exclusively with thermal LWRs, only the fissile fraction of spent fuel matters. On one hand, used UOX fuel contains a higher net fissile content than natural uranium (approximately the same amount of ²³⁵U plus 1% of fissile Pu), but on the other hand, it requires chemical separation and purification (i.e., reprocessing).

The two options considered under the first strategic choice shown in Figure 4-4 are (1) interim storage of used UOX fuel, and (2) reprocessing of used UOX fuel for Pu recycling in LWRs as MOX. In the first case, used UOX fuel is stored (in wet or dry storage systems) and then sent to a geologic repository. In the second case, used UOX fuel is reprocessed; the fission products and minor actinides are vitrified and sent to a suitable geologic repository; Pu is used to fabricate MOX fuel; after irradiation, the spent MOX fuel is stored and then sent to a geologic repository. Reprocessed Uranium (RepU) recycling is not considered here, but is addressed in the next section.

From an economic perspective, the comparison of these two options has already been made in Section 1.2. For the selected parameters, this comparison indicates that the once-through cycle is less costly than reprocessing. However, the difference in terms of overall power generation costs is relatively small (on the order of a few percents) and this gap could disappear if natural uranium and/or SWU prices increase significantly and reprocessing costs decrease.

The main benefit of mono-recycling of Pu in LWRs is the modest savings in natural U resources through the use of MOX fuel. Given that the typical fissile Pu content (²³⁹⁺²⁴¹Pu) in spent UOX is between 0.8 and 0.9% (depending on the burn-up and the cooling time), Pu from five used UOX assemblies contains the equivalent fissile content as one fresh UOX assembly (with 4.5% initial ²³⁵U enrichment). However, in reality, seven, not five, used UOX assemblies are required for the fabrication of one "UOX-equivalent" MOX assembly due to (i) the presence of strong neutron absorbers in reprocessed Pu, particularly ²⁴⁰Pu and other even Pu isotopes; and (ii) the reduced fission cross section of ²³⁹Pu in an epithermal neutron spectrum typical for MOX fuel utilization versus the fission cross section of ²³⁵U fission in a thermal spectrum (Table 4-1).⁷⁶

lastana	Cross Section [barns]		
Neutron Spectrum	σ _r Fission	σ _。 Capture	σ٫/(σ٫+σ _。) Ratio
²³⁵ U Thermal	38.8	8.7	82%
²³⁹ Pu Epithermal	21.7	12.2	64%

Table 4-1Neutronic Data Comparison between U and Pu

So, the amount of energy generated by eight UOX assemblies in the once-through fuel cycle can be generated by seven UOX assemblies plus one MOX assembly with mono-recycling of Pu. This represents a net 12.5% savings in natural uranium resources.

A major issue associated with reprocessing is the potential for increased proliferation risks due to the separation of pure Pu. This issue is partially addressed in Section 1.4. Proliferation concerns can be reduced or mitigated via implementation of stringent physical protection measures at the

⁷⁶ Data from CEA and EDF R&D

reprocessing plant and the co-location of the reprocessing and MOX fuel fabrication facilities. To further enhance the proliferation resistance of reprocessing, the co-extraction of Pu with some U and possibly Np has been proposed through some modifications of the current PUREX process. It can also be argued that high burn-ups of the used UOX fuel feedstock for reprocessing also significantly reduce the attractiveness of the separated fissile materials.

From a waste management perspective, a comparison has also been presented in Section 1.3 showing that used UOX fuel and the cumulative waste streams from reprocessing (ILW-LL and HLW) have approximately the same primary volume. Since most geologic repository concepts and designs are principally heat limited, not volume limited, waste-volume-based arguments and metrics do not appear to be of primary importance. In terms of waste management, reprocessing does offer the possibility of tailored waste forms and reduced heat loads through vitrification and actinide separation, respectively. The impact on waste heat load resulting from the removal of the plutonium isotopes is illustrated in Figure 4-6.





As Figure 4-3 illustrates, HLW heat loads can potentially be reduced by a factor of 2 after 80 years of cooling. Even greater reductions in heat loads can be realized if some fission products, such as cesium and strontium, are also removed from HLW and dispositioned separately. Reprocessing has also been promoted on the basis of overall waste inventory radiotoxicity reductions (generally expressed in terms of total activity in Bq), but this metric is not generally indicative of or related to repository performance, is particularly subject to misinterpretation and

⁷⁷ Data from EDF R&D

misrepresentation, and does not provide a good technical basis for communicating potential long-term radiological hazards from spent fuel and HLW disposal.

The major drawback of plutonium mono-recycling from a waste management point of view is the generation of spent MOX fuel, which is characterized by elevated decay heat loads (on the order of three or four times those of spent UOX fuel). Accordingly, in the context of open fuel cycles, when the spent MOX fuel is disposed of in a geologic repository, there is no advantage in terms of heat loading with Pu recycling, as shown in Figure 4-7.





This comparison is not entirely consistent because HLW and spent MOX are not generated at the same time; nevertheless, it illustrates what an equilibrium condition would look like.

The comparison of once-through and reprocessing followed by mono-recycling of Pu does not yield an overwhelming winner in terms of key metrics such as cost, resource utilization, etc. Relative to the once-through fuel cycle, Pu recycling offers modest improvements in uranium resource utilization but requires significant capital investment, is expensive when natural uranium and SWU costs are low, presents a number of proliferation concerns, and provides marginal waste management benefits, if any, when used MOX is sent to a geologic repository for direct disposal. These offsetting advantages and disadvantages suggest that the reprocessing benefit for a nation is situational. For a country with no existing infrastructure and no sunk costs, such as the U.S., there is no compelling case to be made for adoption of reprocessing with Pu recycle in a fleet comprised exclusively of LWRs. Likewise, for a country that has made a substantial investment in the infrastructure for reprocessing and Pu recycle, such as France, there

is not a compelling reason to forego reprocessing. Instead, managed storage of spent UOX, either through interim storage or Pu recycling, represents a strategic issue in the context of long-term energy security as an intermediate step on the path to more advanced partially or fully closed fuel cycles.

Closed Fuel Cycles – Comparison between Interim Storage of Spent UOX Fuel and Mono-Recycling of Plutonium

In this comparison, used LWR fuel is considered as an energy source, especially in light of its fissile plutonium content. Indeed, the availability of a sufficiently large inventory of fissile Pu (in lieu of enriched ²³⁵U) is a de facto prerequisite for starting up a self-sustained fleet of fast reactors.

As in the previous section, the two options resulting from Strategic Choice #1 will be compared from the perspective of a "Pu economy." For this purpose, it is necessary to precisely define the timing for both scenarios. In both cases, the time between spent UOX irradiation and onset of advanced reprocessing of used UOX or MOX fuel for fabrication of fast reactor fuel is 50 years. In the first scenario, the used UOX is stored over this 50-year timeframe. In the second scenario, the used UOX is cooled for five years, reprocessed, fabricated into MOX over a two-year time period, irradiated for four years, and the used MOX fuel is stored over the remaining 39-year time period (Figure 4-8).



Figure 4-8 Comparison of Timelines for Fuel Cycle Activities Associated with Interim Storage and Single-Recycling of Pu over 50 Years

There is the option of promptly reprocessing used UOX fuel as soon as possible, but without any Pu recycling in LWRs for the purpose of stockpiling Pu for a fast breeder cycle. However, this approach would result in a double penalty of increasing separated Pu stockpiles and of eventually

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requiring a second reprocessing step to remove nuisance ²⁴¹Am, which continues to accumulate through ²⁴¹Pu decay. Once Pu is separated, it is best to recycle it as soon as possible in order to fully utilize the ²⁴¹Pu as a fissile resource and to minimize in-growth of nuisance nuclides.

In the second scenario, spent fuel is reprocessed; MOX is fabricated; and irradiated over the first eleven years. This approach results in costs on one hand, but, on the other hand, electricity is produced by MOX irradiation, and this generates income. In the previous section related to open fuel cycles, it was shown Pu mono-recycling in LWRs is not too far from economic viability. So, there is almost a balance between cost and the income. This observation allows focusing only on the characteristics of nuclide inventories available after 50 years in both scenarios.

These materials differ by three parameters:

- Pu quantity (expressed in Pu-239 equivalent quantity in fast spectrum⁷⁸, which takes into account the isotopic quality of Pu) generated per MTHM of initial spent UOX fuel. In fact, in both scenarios, this quantity decreases over time because of Pu-241 decays into Am-241. Moreover, some Pu is destroyed during MOX irradiation.
- Reprocessing capacity (in MTHM) necessary to fabricate one fast reactor core assembly⁷⁹. This figure of merit depends on the isotopic quality of Pu, but also on the Pu concentration in spent MOX assemblies that is higher than in spent UOX assemblies.
- Spent Fuel (UOX or MOX) decay heat, which has an impact on the ease to reprocess this fuel. In the present comparison, spent MOX is far hotter than spent UOX because it contains more minor actinides and it is cooled over a shorter time.

Indicators After 50 Years	Scenario 1: Interim Storage of UOX	Scenario 2: Mono- Recycling of Pu
Pu-239 Equivalent Quantity (in kg/MTHM of Initial Spent UOX)	7.74	4.96
Reprocessing Capacity (per MTHM of FR-MOX) in MTHM of Spent Fuel	14.3	3.12
Spent Fuel Decay Heat (in W/MTHM)	852	3,566

Table 4-2Pu Comparison after 50 Years in Both Scenarios

This table shows that mono-recycling of Pu decreases the equivalent quantity of Pu-239 for fast reactor fuel fabrication by 35%, but it also significantly reduces the required advanced reprocessing capacity. It also generates four times more decay heat, which makes reprocessing

⁷⁸ Equivalent Pu-239 is obtained by using the following weighting factors: 0.7506 for Pu-238; 1 for Pu-239; 0.3158 for Pu-240; 1.1118 for Pu-241; and 0.0360 for Pu-242 (source: EDF R&D)

⁷⁹ Considering that Pu represents 15% of the core mass if it comes from spent UOX.

more difficult, but given the fact that advanced reprocessing is supposed to handle spent fuel from fast reactors (with a high decay heat), this is not considered a critical factor.

These figures demonstrate that even in the Pu mono-recycling scenario, it is probably valuable to not reprocess all spent UOX, in order to mix spent UOX and spent MOX and, hence, increase the quantity and the quality of Pu necessary to start a fast reactor fleet. Another advantage of mixing spent UOX and spent MOX is to use current reprocessing plants to reprocess spent MOX (today, this is possible with the proportions 85%/15% at the La Hague facility).

In fact, the real advantage of reprocessing and recycling in LWRs must be considered from an industrial point of view. Reprocessing is a highly complex industrial process even though its chemical principles are relatively simple and well-known. A considerable know-how is required to operate a reprocessing plant reliably. Only one LWR fuel reprocessing plant (La Hague, France, operated by AREVA) has been consistently operated successfully during the past decades. Without this industrial success, it is likely that reprocessing would not be considered as a reasonable industrial/commercial enterprise at this time. Hence, reprocessing and recycling of Pu in LWRs has been promoted as a necessary intermediate step toward a closed fuel cycle policy. Interim storage, alone, implies a "revolutionary approach" with the direct construction of an advanced reprocessing plant, whereas mono-recycling of Pu in LWRs supports an "evolutionary approach" that may be more conducive to a successful industrial deployment.

Closed or Open Fuel Cycles - Multi-Recycling of Plutonium in LWRs

After discussing the relative attributes of interim storage or mono-recycling of Pu in LWRs, one of the options embedded in Strategic Choice #1 is whether or not to recycle Pu in LWRs for a second or third time.

A previous study from NEA⁸⁰ uses the APOLLO code developed by CEA to answer this question. In this study, the burnup is 51 GWd/MTHM for every kind of fuel (UOX or MOX), which is relatively high compared to the current burnup of MOX fuel. As a consequence, the total Pu content of first generation MOX fuel (MOX1) is 10.15% instead of the 8.65% used today. After irradiation of MOX1 during 4 years and cooling time of five years, MOX1 is reprocessed together with spent UOX (3 spent UOX assemblies for 1 spent MOX1 assembly) and MOX2 is fabricated within two years. This dilution of spent MOX1 with spent UOX is necessary both for technical (criticality, decay heat...) and economic reasons. Then MOX2 is irradiated, cooled, reprocessed together with three used UOX assemblies in order to fabricate MOX3, and so on.

⁸⁰ 2002, NEA "Physics of Plutonium Recycling: Multiple Pu recycling in PWR"



Figure 4-9 Timeline for Fuel Cycle Activities Associated with Multi-Recycling of Pu in LWRs

Each cycle lasts 11 years. At each recycling, the isotopic quality of MOX decreases, even though Pu from new spent UOX is added. Therefore, the total Pu content of MOX has to increase to achieve the same burnup.

Pu content is about 10.15% for spent MOX1, 1.24% for spent UOX and has to be at 13.6% for fresh MOX2. The resulting Pu mass balance leads to:

8.2	$UOX^{Spent} \rightarrow 1 MOX1^{Fresh}$	Faultion 4.4
1.35	$MOX1^{Spent} + 4.05 \ UOX^{Spent} \rightarrow 1 \ MOX2^{Fresh}$	Equation 4-1

Whereas MOX1 allows savings in natural uranium resources of 1/(8.2+1) = 11%, MOX2 allows additional savings of 2.5%. A similar reasoning leads to less than 1% additional savings by the use of MOX3. So, because of the net consumption of Pu and the decrease of its quality during MOX irradiation, the potential uranium resource savings decrease after each MOX generation. This makes multiple recycles less and less attractive.

Moreover, spent MOX reprocessing and MOX fabrication become more and more difficult after each generation. Because of the decrease in Pu quality, higher and higher Pu contents are required (e.g., 10.15% for MOX1, 13.6% for MOX2, 16% for MOX3, and 17.8% for MOX4). This demand for greater Pu content raises safety problems for LWRs. Pu contents exceeding 12% in LWR fuel can result in a positive void coefficient. So, the Pu content limit is reached as soon as the second generation, especially if the burnup is high (a burnup target of 45 GWd/MTHM would lead to a Pu content of MOX2 less than 12%). Pu content is lower for MOX-EU (MOX that uses an enriched uranium matrix instead of a depleted uranium matrix), but this option would further reduce the natural uranium savings.

Multi-recycling of Pu in LWRs results in a build-up of even numbered Pu isotopes, especially Pu-238 that leads to a high decay heat due its relatively short half-life. The build-up of higher Pu isotopes (Pu-240 and Pu-242) also leads to significant in-growth of minor actinides (Am, Cm

and even Cf), with important implications for decay heat, neutron emission, and waste radiotoxicity.

Some of these drawbacks, especially the high Pu content, could be mitigated by the use of highly moderated LWR (new design with a higher moderator/fuel volume ratio). In that case, a lower Pu content would be required: 6.7% for MOX1, 9.6% for MOX2, 12.1% for MOX3. However, the costs associated with deploying the required new LWR core designs would greatly outweigh the benefits realized from multi-recycling of Pu in LWRs.



Figure 4-10 Pu Content Required for Fresh MOX Fuel (Burnup of 51 GWd/MTHM)

Multi-recycling of Pu in LWRs does not represent an attractive option technically. In the context of a closed fuel cycle strategy, multi-recycling decreases the quantity and the quality of Pu, making it more difficult to start a fast reactor fleet. In the context of an open fuel cycle strategy, the savings in natural uranium resources are not really significant. So, the only advantage of multi-recycling of Pu, assuming that the impact of high Pu contents could be overcome, would be to reduce the inventory and the quality of the Pu sent to the geologic repository. But it would also result in the build-up of very undesirable species such as Pu-238, Am, Cm and higher minor actinides.

All these drawbacks also show that infinite recycling of Pu in LWRs (discussed in Section 4.3 as a possible closed fuel cycle) would require an enriched uranium matrix.

Synthesis

The following table summarizes the major advantages and disadvantages of the three options of managed storage considered here:

Perspective		Interim storage of spent UOX	Mono-recycling of Pu in LWRs	Multi-recycling of Pu in LWRs
Open Cycle	Advantages	Most economical option	~12% savings in natural U resources	- Reduction of Pu inventory - Less Pu sent to final disposal
	Disadvantages	- No natural U resource savings - Large volume of spent fuel sent to final disposal	- Generate spent MOX - Requires a reprocessing industry	- May not be technically feasible - Generates undesirable species
Closed Cycle	Advantages	Provide the largest amount of Pu with the highest quality	- Provide technological know- how about reprocessing - Concentrate Pu in spent MOX	None
	Disadvantages	Require introduction of advanced reprocessing technologies without benefit of prior experience	Decrease the quantity and the quality of Pu	Decrease even more the quantity and the quality of Pu

 Table 4-3

 Synthesis about Managed Storage of Spent UOX Fuel

4.2.2 Use of Reprocessed Uranium

The second Strategic Choice (#2 in Figure 4-4) concerns the option of re-using reprocessed uranium (RepU) to fabricate Enriched Reprocessed Uranium (ERU) assemblies. This option is obviously dependent on Strategic Choice #1, because availability of RepU pre-supposes that spent UOX has been reprocessed. But, at the same time, this second choice may influence the first one, by making reprocessing more economically competitive.

The U-235 content of RepU is close to that of natural uranium. Because it is a (free) by-product of reprocessing, it is an interesting secondary source of uranium. The RepU isotopic composition is slightly different from natural uranium. This requires additional treatments for its use leading to added costs. Hence, it is not obvious whether recycling of RepU is an efficient option or not.⁸¹

⁸¹ "Management of Reprocessed Uranium" published in 2007 by IAEA provides a very detailed discussion of these issues.

Physics of Reprocessed Uranium

Natural uranium comprises three isotopes:

- U-238, which represents 99.28% of natural uranium and only half of its radiological activity.
- U-235, which represents 0.71% of natural uranium and is fissile.
- U-234, which represents only 0.0053% of natural uranium but half of its radiological activity. It is a neutron absorber. It is co-enriched with U-235 around 0.05% and then is partially transmuted during irradiation by neutron capture.

During irradiation of UOX fuel, four other isotopes of uranium are formed and constitute the telltale isotopic signature for RepU:

- U-232, even in very small concentration (some parts per billion), is radiologically the most significant uranium isotope. This is due mainly to a few short-lived decay products: Pb-212, Bi-212 and Tl-208. The latter has a very strong gamma-ray radiation around 3.4 MeV. The maximum activity of U-232 and its daughters occurs ~10 years after discharge. U-232 is created by several different and complex ways, the dominant one being α-decay of Pu-236. U-232, once present, can absorb a neutron and become U-233 which is fissile.
- U-233 is generated in small amount during irradiation, mostly by α -decay of Np-237 followed by β -decay of Pa-233. It is fissile and does not represent a significant radiological hazard.
- U-236 is a strong neutron absorber, mostly created by neutron capture of U-235. As a consequence, the amount of U-236 increases with the initial U-235 content and with burnup.
- U-237 has almost no impact on RepU because of its negligible concentration.

Table 4-4 presents the typical isotopic composition of RepU (with a burnup of 48 GWd/MTHM and after 5 years of cooling):

Table 4-4

RepU Isotopic Composition after Five Years of Cooling (Burnup of 48 GWd/MTHM, Initial Enrichment of 4.5%)

Isotope	Grams per metric ton
U-232	2.86 E-03
U-233	3.81 E-03
U-234	2.19 E+02
U-235	1.03 E+04
U-236	5.83 E+03
U-237	4.38 E-05
U-238	9.21 E+05

In fact, among those seven isotopes, only three really matters for RepU: U-235, which determines the fissile content; U-236 (and U-234, but to a lesser extent), which acts as a neutron poison (absorber); and U-232, which generates radiological hazards. This means that enriched reprocessed uranium (ERU) needs to be over-enriched to provide the equivalent performance as enriched natural uranium (ENU). Figure 4-11 presents the required ERU enrichment as a function of ENU/ERU discharge burnup for four different ENU enrichments. For a burnup of 50 GWd/MTHM, an over-enrichment of 0.5% is a good approximation.

Figure 4-12 illustrates the evolving radiological hazard from U-232 over time in spent nuclear fuel. Accordingly, there is an incentive to reprocess spent UOX as soon as possible, to avoid the build-up of U-232, an isotope arising from alpha decay of Pu-236, which has a short half-life of 2.9 years. Another incentive of early reprocessing is to avoid the decay of Pu-241 into Am-241. RepU also may require purification at different steps of the recycling process, in order to get rid of the troublesome daughters of U-232.

Consequently, over-enrichment and purification of RepU are two major drawbacks that incur additional costs.



Figure 4-11 Over-enrichment Required for Reprocessed Uranium



Figure 4-12 U-232 Concentration Evolution in Spent UOX Fuel

Economics of Recycling Reprocessed Uranium⁸²

There are two ways to recycle RepU and to fabricate new fuel assemblies. The first one consists of blending RepU with Moderately Enriched Uranium (MEU) or Highly Enriched Uranium (HEU) and the second one consists of re-enriching RepU by ultra-centrifugation or gaseous diffusion.

Down-blending of MEU

In this case, RepU is mixed with MEU or HEU in order to provide ERU, i.e., uranium enriched at the target level (4.5% in this example). MEU or HEU can come from existing military stockpiles (from the Russian Federation or the U.S.A), but the quantities potentially available are very small compared to what would be necessary to recycle all RepU⁸³.

Natural uranium can also be enriched, on purpose, into MEU in order to be subsequently mixed with RepU. This is the case analyzed here.

For 1 MT of RepU with a residual enrichment of R, mixed with X MT of MEU with an enrichment of Y, to yield (1+X) MT of ERU at 4.5%, the following equation applies:

⁸² Also refer to *Parametric Study of Front-End Nuclear Fuel Cycle Costs Using Reprocessed Uranium*, EPRI, Palo Alto, CA: 2009. 1020659

⁸³ 2006, NUKEM "RepU's second chance?"

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 $(1 \times R) + (X \times Y) = (1 + X) \times 0.045$, Equation 4-2 (no U-236 penalty is taken into account here).

This leads to the required quantity of natural uranium, or NU:

$$NU = X \frac{Y - 0.0025}{0.0071 - 0.0025}$$
, (assuming a 0.25% tails assay) Equation 4-3

and to the required quantity of Separative Work Units:

$$SWU = X \left(\Phi(Y) + \frac{Y - 0.0071}{0.0071 - 0.0025} \Phi(0.25\%) - \frac{Y - 0.0025}{0.0071 - 0.0025} \Phi(0.71\%) \right)$$
Equation 4-4

where $\Phi(x) = (2x-1) \times \ln\left(\frac{x}{1-x}\right)$, (as mentioned in Section 1.1).

So, it is possible to compare SWU/EU and NU/EU, where EU = 1 + X, in the case of mixing of MEU with RepU with direct enrichment of natural uranium (Figure 4-13). Mixing of RepU with MEU enriched in the 5 to 20% range (horizontal axis) requires more and more additional SWU and less natural uranium as MEU enrichment increases⁸⁴.



Figure 4-13 Comparison between Down-blending of MEU and Direct Enrichment of Natural Uranium

⁸⁴ The result depends on R, the residual enrichment of RepU

So, the total additional cost of down-blending depends on the relative price of SWU, or P_{SWU} , and natural uranium (expressed in kgU), expressed as P_{NU} . Figure 4-14 shows the result with $P_{SWU}/P_{NU} = 1.3^{85}$: It can be seen that down-blending of enriched ²³⁵U is not economically interesting, whatever the MEU enrichment is. In addition, this calculation does not take into account the cost of mixing itself, which is generally assumed to be relatively small.

In fact, the economic interest of down-blending depends on two parameters: R and P_{swu}/P_{nu} . The break-even P_{swu}/P_{nu} ratio is displayed in Figure 4-15.



Figure 4-14 Additional Cost of MEU Down-blending Compared to Direct Enrichment of NU

⁸⁵ This ratio corresponds to recent market prices: 165/SWU and $48/lbU_3O_8$ (125/kgU)



Figure 4-15 Break-even P_{swu}/P_{NU} Ratio as a Function of RepU Residual Enrichment

Current typical P_{swu}/P_{NU} ratios (P_{NU} expressed in \$/kgU) are between 1 and 1.5 and typical RepU residual enrichments are around 0.7 for high discharge burnup levels⁸⁶. So recycling of RepU by down-blending with fresh MEU is not economically interesting, and even less so if the issues of ²³²U (which requires purification), ²³⁶U (which requires a higher enrichment) and licensing of reactors are taken into account. Moreover, enrichments above 20% would raise proliferation concerns. Most important, mixing of MEU and RepU would dramatically increase the quantity of fuel contaminated by ²³²U and its daughters. So, this option would involve the use of a greater number of nuclear power plants to irradiate this fuel, instead of concentrating the use of RepU-based fuel in a relatively small fraction of the NPP fleet.

These drawbacks reduce the attractiveness of RepU recycling through the down-blending of fresh MEU. As a result, this option appears most feasible for the reduction of existing stockpiles of defense-related HEU, for which the incentives extend beyond economics.

Direct Enrichment of RepU

Another way to recycle RepU in LWRs is via direct enrichment using centrifuge technology. In this case, RepU experiences essentially the same process as natural uranium feedstock, albeit with some purification as well. RepU is converted into UF_6 and then sent as soon as possible to the enrichment plant to avoid the buildup of ²³²U daughters. An over-enrichment of 0.5% is

⁸⁶ In fact, the correlation between burnup and residual enrichment is not obvious, because of the higher initial enrichment of UOX fuel required to achieve higher burnup.

required to counteract the presence of ²³⁶U after enrichment. Finally, ERU fuel is fabricated. The feasibility of RepU enrichment versus natural uranium enrichment can be made using the nominal parameters⁸⁷ in Table 4-5.

Assuming the nominal values in Table 4-5, use of ERU appears to be ~13% cheaper than exclusive reliance on ENU (Figure 4-16). The competitiveness of ERU increases as the price of natural uranium goes up and as the residual enrichment of RepU increases, as shown in Figure 4-17.

Parameter	Value	
NU price	\$48/lbU₃O₅ (\$125/kgU)	
RepU residual enrichment	0.6%	
RepU price	0 (by-product)	
NU conversion	\$7/kgU	
RepU conversion	\$21/kgU (x3)	
Over-enrichment for ERU	0.5%	
Enrichment of NU	\$165/SWU	
Enrichment of RepU	\$190/SWU (+15%)	
ENU fuel fabrication	\$200/kgHM	
ERU fuel fabrication	\$220/kgHM (+10%)	

Table 4-5 Comparison between Enrichment of NU and RepU (Nominal Values)

⁸⁷ Data taken from EPRI Report "Parametric Study of Front-End Nuclear Fuel Cycle Costs Using RepU" (2009) and from the 2006 Nukem study "RepU's second chance?"



Figure 4-16 ENU and ERU Cost Breakdown



Figure 4-17 Break-Even NU Price as a Function of Residual Enrichment of RepU

As a by-product of reprocessing, RepU may be worth recycling by direct enrichment based on the nominal values considered here. However, the pursuit of higher discharge burnup could restrict the feasibility of ERU and RepU recycling, because the requirement for over-enrichment of ERU and the current licensing limit of 5% for ²³⁵U in LWR fuel processing. In addition, use of ERU further complicates fuel cycle operations relative to the use of fresh ENU fuel due to the introduction of additional radioisotopes into the process, which among other things requires additional purification and increases the radiation protection burden throughout the front end of the fuel cycle. Accordingly, demonstrating the economic advantages of re-using RepU is critical before committing to its implementation.

Impact of Reprocessed Uranium Recycling on Reprocessing Competitiveness

In the previous section, RepU was considered as a by-product of reprocessing. But once a positive economical value can be attributed to RepU, it is necessary to re-evaluate the whole economic competitiveness of reprocessing and Pu recycling. Figure 4-18 presents a side-by-side comparison of the key features of interest for the Pu recycling and interim storage options, including quantities of used fuel and natural uranium feedstock required for yielding an equivalent quantity of LWR fuel.⁸⁸



Figure 4-18 Material Flows with or without Plutonium and Reprocessed Uranium Recycling

The four costs used for evaluating the costs of these two options are summarized in Table 4-6. The ENU and ERU costs are from Figure 4-16; the reprocessing cost is the same as in Table 4-5; the MOX fuel fabrication cost is the nominal cost used in EPRI Report 1018575.⁸⁹

⁸⁸ Assumptions: initial enrichment of 4.5% in UOX (ENU) and 5% in ERU. Spent UOX contains 1.2% of Pu and 94% of U. MOX fuel has 8.6% in Pu content. Tails assay of 0.25%.

⁸⁹ *Nuclear Fuel Cycle Cost Comparison Between Once-Through and Plutonium Single-Recycling in PWRs*, EPRI, Palo Alto, CA: 2009. 1018575.

Cost item	Value
Reprocessing (in k\$/MTHM of spent fuel)	1,000
MOX Fuel (in k\$/MTHM of product)	1,250
ERU Fuel (in k\$/MTHM of product)	2,224
ENU Fuel (in k\$/MTHM of product)	2,555

Table 4-6Cost Comparison between Once-through and Recycling

If only MOX is fabricated after reprocessing, this yields ~0.14 MTHM of MOX fuel per MTHM of spent UOX (Figure 4-18), and the associated cost is the cost of reprocessing $(1 \times \$1,000k) +$ the cost of MOX fuel (0.14 x \$1,250k). The cost is therefore \$1,175k per 0.14 MTHM of MOX, or \$8,393k per MTHM of MOX.

If both MOX and ERU are fabricated after reprocessing, this yields ~0.24 MTHM of fuel (same fissile equivalence) per MTHM of spent UOX (Figure 4-18), and the associated cost is the cost of reprocessing ($1 \times 1,000$ k) + the cost of MOX fuel ($0.14 \times 1,250$ k) + the cost of ERU fuel ($0.10 \times 2,224$ k). The cost is therefore \$1,397.4k per 0.24 MTHM of MOX/ERU, or \$5,823k per MTHM of MOX/ERU.

None of these two options are currently economically competitive compared to the cost of ENU (at \$2,555k per MTHM of ENU), but a cost comparison of the two options indicate that MOX + ERU is more attractive than MOX alone.

A sensitivity study on natural uranium price and reprocessing cost is presented in Figure 4-19. Under the nominal cost assumptions considered in Table 4-6, recycling of RepU lowers the natural uranium break-even price by approximately 36%.

Multi-Recycling of Reprocessed Uranium - Other Uses of Reprocessed Uranium

As was done for multiple recycling of plutonium in Section 4.2.1, it is informative to study if multiple recycles of RepU make sense from a technical and economical point of view.

Figure 4-20 compares the concentration of the different uranium isotopes in used ENU fuel and in used ERU fuel. As shown in Figure 4-20, spent ERU contains ~four times more ²³²U and ~three times more ²³⁶U than spent ENU, which dramatically decreases its quality. Accordingly, RepU from used ERU presents greater radiological hazard and it has a more consequential neutron absorber to fissile content ratio, as shown in Figure 4-21.



Figure 4-19 Natural Uranium Break-even Price as a Function of Reprocessing Cost



Figure 4-20 U Isotope Composition in Used ERU Relative to Used ENU



Figure 4-21 U-235 and U-236 Concentrations in Spent ENU and Spent ERU

As a consequence, multiple recycle of RepU appears to be unattractive for application in LWRs.

Other potential fuel cycle applications for RepU include:

- Using RepU instead of depleted uranium in MOX fuel. Because of the fissile content of RepU, this could result in a lower required concentration of Pu in MOX fuel, which could mitigate some concerns associated with MOX fuel irradiation to higher burnup
- Using RepU in heavy-water reactors, as proposed in the DUPIC (<u>Direct Use of spent PWR fuel In CANDU</u>) process.⁹⁰ A residual fissile content of RepU exceeding 0.71% would allow for higher HWR fuel burnup; and
- Using RepU as a blanket material in fast reactors. The presence of ²³⁶U in RepU (especially RepU from spent ERU) would result in production of ²³⁷Np and ²³⁸Pu in the blanket via successive neutron captures. The presence of ²³⁸Pu would reduce the material attractiveness of Pu produced in the blanket, and so would contribute to its proliferation resistance. However, it is unlikely that the ²³⁶U content of RepU would yield significant quantities of ²³⁸Pu.

⁹⁰ The Evolution of CANDU Fuel Cycles and their Potential Contribution to World Peace by J. J. Whitlock, "International Youth Nuclear Congress 2000", Bratislava, Slovakia, April 9-14, 2000

4.2.3 Open or Closed Fuel Cycle

The third strategic choice (Figure 4-4, Choice #3) faced in pursuing a fuel cycle is strategically the most important one, as it addresses the question of whether or not to pursue closure of the nuclear fuel cycle. Only Pu is considered here; the minor actinides are addressed in the next section. As mentioned earlier, the decision of pursuing an open or a closed fuel cycle has a strong influence on decisions related to the managed storage of used fuel. In an open fuel cycle, used LWR fuel is considered as waste; whereas in a closed fuel cycle, used fuel represents an important energy resource for starting and deploying a fleet of fast reactors.

As discussed in Section 4.1, the fuel cycle options are delineated on the basis of the material sent for permanent disposal in lieu of recycling. For the purposes of the ensuing discussions, open, partially-closed, and fully-closed fuel cycles are defined as follows:

- An open fuel cycle is a fuel cycle in which Pu is sent to a geologic repository for permanent disposal;
- A partially closed cycle is a fuel cycle in which only the minor actinides are sent to a geologic repository for permanent disposal; and
- A fully closed cycle is a fuel cycle in which no Pu or minor actinides are sent to a geologic repository for permanent disposal.

These definitions are consistent with the open and closed fuel cycle depictions in Figures 4-2 and 4-3. In the strictest sense, because material losses will inevitably occur with reprocessing and other steps, no cycle can be considered as perfectly closed.

From this perspective, first considering only Pu management, five general classes of fuel cycles are considered, two open and three partially closed cycles (Figure 4-22).⁹¹

⁹¹ The main characteristics of these fuel cycles are detailed in the 2006 OECD/NEA study "Advanced Nuclear Fuel Cycles and Radioactive Waste Management," Nuclear Energy Agency, OECD 2006, NEA No. 5990



Figure 4-22 Five General Fuel Cycles for Pu Management

To evaluate the relative merits of these five fuel cycles, as discussed in Section 1, it is worth starting from the four main challenges deployment of any nuclear fuel cycle faces, and inferring some figures of merit.

- Uranium sustainability translates easily into natural uranium consumption, expressed as a percentage of once-through cycle consumption, without taking into account recycling of RepU.
- Economic competitiveness is mainly determined by the price of natural uranium and the capital cost of the fuel cycle facilities. This is captured in two key figures of merit: (1) natural uranium consumption, and (2) technological difficulty, the latter being expressed as the fraction of advanced, not-yet-available facilities required for fuel cycle operation.
- Waste management concerns tend to track the quantity of Pu considered for permanent disposal. However, Pu is not very mobile in a geologic repository, so its potential radiological impact is in fact very low. The main impact of Pu is heat generation from the Pu isotopes and their decay products (such as Am-241, daughter of Pu-241), which impacts the size of the repository.
- Non-Proliferation can be translated into two figures of merit: quantity of separated Pu in the fuel cycle at equilibrium and relative Pu attractiveness, as discussed in Section 1. For simplicity, the traditional suitability of a plutonium mixture for explosive devices will be used as follows: Category 1: "Practically unusable"; Category 2: "Conceivably usable"; Category 3: "Practically usable"; Category 4: "Standard material"; and Category 5: "Best quality"⁹²

⁹² Table I in B. Pellaud, *Proliferation Aspects of Plutonium Recycling*, Journal of Nuclear Materials Management, Fall 2002, Volume XXXI, No. 1
Once-Through Cycle (OTC)

This fuel cycle consists simply of irradiating slightly enriched UOX fuel in LWRs and then sending the spent fuel to a permanent repository, anticipated to be a mined geologic repository. No advanced facilities are required in this scenario. Assuming a burnup of 60 GWd/MTHM and an electrical efficiency of 34.1%, it leads to 26 kg of Pu per TWhe sent to a geologic repository. No Pu is separated in the fuel cycle and the Pu isotopic composition after several decades of cooling is 3% of Pu-238, 57% of Pu-239, 28% of Pu-240, 2% of Pu-241 and 10% of Pu-242. So this material is almost unusable to build a nuclear weapon, albeit slightly below Pellaud's "practically unusable" limit of 30% of Pu-240⁹³ and clearly below the Kessler limit of 6% of Pu-238⁹⁴ (Section 1-4). Because of the presence of very radioactive fission products and minor actinides, this material can however be considered as Category 1. This option is summarized in Column I of Table 4-7 below.

Plutonium Mono-Recycling in Light Water Reactors

This fuel cycle provides ~11% savings in natural resources compared to once-through cycle,⁹⁵ and represents the current French approach. Pu is separated after UOX irradiation to fabricate MOX fuel, at a rate of 23 kgPu/TWhe. About 8 kgPu/TWhe is consumed by recycling MOX in LWRs, while ~15 kgPu/ThWhe remains in the spent MOX fuel, which is assumed to be sent to a permanent disposal facility. Therefore, for a nominal burnup of 60 GWd/MTHM and an electrical efficiency of 34.1%, approximately 15 kg of Pu per TWhe is slated for disposal. In terms of proliferation concerns, material attractiveness is higher than that of the OTC due to the separation of Pu during reprocessing of used UOX fuel, corresponding to a Category 2 versus the Category 1 for OTC. However, the spent MOX is even less attractive than spent UOX from the OTC due to substantially greater neutron emissions and much greater decay heat from long-lived minor actinides. This fuel cycle option is summarized in Column II of Table 4-7.

Plutonium Multi-Recycling in Light Water Reactors

In this case, used fuel (UOX or MOX) is always reprocessed, and the recovered Pu is indefinitely recycled in an LWR fleet. Because of the inevitable (and rapid) degradation of the Pu isotopic quality and the safety limit of 12% of Pu content in LWR fuel, the MOX fuel produced in this fuel cycle requires significant input of fissile ²³⁵U into the process. The result is a MOX fuel with an enriched uranium matrix designated as MOX-EU. As a consequence, more natural uranium feedstock is required for unlimited Pu recycling, and therefore, the natural uranium savings realized is similar to that for mono-recycling, i.e., 13% instead of 11%. In this scenario, advanced LWR reactors with a higher moderator/fuel ratio are required, representing 28% of the

⁹³ B. Pellaud, *Proliferation Aspects of Plutonium Recycling*, Journal of Nuclear Materials Management, Vol. XXXI, No.1 (2002)

⁹⁴ G. Kessler, Plutonium Denaturing by Pu-238, Nuclear Science and Engineering, 155, 53-133

⁹⁵ Note: This figure is less than the 12.5% saving stated in Section 4.2.1 under "Open Fuel Cycles – Comparison between Interim Storage of Spent UOX Fuel and Mono-recycling of Plutonium" due to the assumption of a higher burnup, 60 GWd/MTHM, instead of the 50 GWd/MTHM assumed earlier

LWR fleet. Current reprocessing technology would conceivably be sufficient for use. As a result of this modest reliance on new LWR designs but little reliance on new reprocessing technology, the fraction of advanced facilities in this scenario is around 14%,(assuming a weighting factor of 50% for the reactors and 50% for reprocessing facilities). While theoretically no Pu is sent for permanent disposal, there are losses during reprocessing, which leads to 0.07 kgPu/TWhe in one or more waste streams. The cumulative quantity of separated Pu, 69 kg/TWhe, is 3 times that for the mono-recycling scenario. In terms of proliferation, the material attractiveness of the separated Pu from spent UOX is the same as for mono-recycling: Category 2, whereas the severely degraded isotopic quality of Pu from used MOX (Pu-240 content above 35%) in a multi-pass recycling scenario provides for a Category 1 classification. This fuel cycle option is summarized in Column III of Table 4-7.

Plutonium Burning in Fast Reactors (FRs)

In this scenario, UOX is irradiated in LWRs, and then reprocessed. The recovered Pu is then recycled indefinitely in fast neutron spectrum reactors with a conversion ratio less than unity, i.e., in fast burner reactors. For the purpose of this evaluation, a conversion ratio of 0.85 is assumed. Under this scenario, 15% of the Pu used to fuel the FR fleet is derived from used UOX and 85% comes from MOX irradiated previously in a fast reactor, MOX-FR. Assuming that the electric efficiencies of LWRs and FRs are 34% and 40%, the LWR fleet provides 44% of the electric power and the FR fleet provides 56%. Under this burner scenario, some 56% savings in natural resource use are realized because the FRs only use materials generated in or cycled through LWRs: Pu or depleted uranium. In terms of technology, the LWR fleet could either be any generation of reactors, while the FRs represent introduction of advanced nuclear technology. In addition to this 56% share of advanced reactor technology, advanced reprocessing technology is required. So, assuming the same weighting factor for reactors and reprocessing plants, the overall minimum fraction of facilities involving advanced technology is 78% for this scenario. As a result of the inevitable losses incurred during reprocessing, 0.08 kgPu/TWhe is designated as waste for disposal. The cumulative amount of separated Pu in this scenario is estimated to be 80 kgPu/TWhe, 11 kgPu/TWhe derived from used UOX and 69 kgPu/TWhe derived used spent MOX-FR. Since all the Pu separated at the reprocessing plant comes from used UOX and used MOX-FR which have similar attractiveness rankings (i.e., less than 60% of Pu-239, and less than 30% of Pu-240, and minor quantities of Pu-238), this material can be considered as Category 2 in terms of proliferation ranking. This fuel cycle option appears in Column IV of Table 4-7.

Plutonium Breeding in Fast Reactors

A fast breeder reactor is any fast spectrum neutron reactor having a conversion ratio greater than unity. In theory, a 100% fast breeder reactor fleet would not consume any new natural uranium resources, as it would utilize existing depleted uranium stockpiles and produce, at minimum, all of the Pu consumed for power production. However, for a realistic steady-state scenario, natural uranium is necessary to provide some fertile U-238 for conversion into Pu-239. However, this quantity of natural uranium consumption would be exceedingly small, on the order of 1 or 2% of the natural uranium required for the once-through cycle. For this scenario, 100% of the technology (reactors and reprocessing plants) is advanced and does not yet exist. Losses incurred during reprocessing would result in 0.14 kgPu/TWhe of the total 143 kgPu/TWhe handled at the reprocessing plant being lost to one or more waste streams. Overall material attractiveness

depends on the nature of Pu reprocessing. For the case of group extraction of actinides, Pu breeding in FRs can be considered very unattractive (Category 1) in terms of proliferation ranking. However, if a more conventional separation technology, i.e., PUREX-based, is used for selective extraction of actinides, proliferation resistance is degraded, meriting a Category 2 ranking. In addition, if fertile U-238 blankets are used to increase the conversion ratio of the reactor, the overall Pu-239 isotopic composition in the reactor could exceed 95%, making it a very attractive material (Category 4) and rendering this approach less favorable in terms of proliferation resistance. To mitigate proliferation concerns, the reactor blanket could conceivably be denatured through the addition of U-236 (from RepU), Np-237, Am or Cm, which would among other things result in the build-up of Pu-238. Research is also underway on self-sustaining core designs that would provide for a conversion ratio greater than one without the need for blankets. This fuel cycle option appears in Column V of Table 4-7.

	FUEL CYCLE				
Figure of Merit (FOM)	l Once- through cycle (OTC)	II Pu mono- recycling in LWRs	III Pu multi- recycling in LWRs	IV Pu burning in FRs	V Pu breeding in FRs
Natural Uranium consumption (compared to OTC)	100%	89%	87%	44%	<1%
Fraction of advanced facilities	0%	0%	14% 28% of advanced light-water reactors and 0% of advanced reprocessing	78% 56% of advanced fast burner reactors and 100% of advanced reprocessing	100% 100% of advanced fast breeder reactors and 100% of advanced reprocessing
Pu sent to geologic repository (kg/TWhe)	26	15.2	0.07	0.08	0.14
Separated Pu in the cycle (kg/TWhe)	0	23	69	80	143
Material attractiveness	1 (spent UOX)	2 2 (Pu from spent UOX) 1 (spent MOX)	2 2 (Pu from spent UOX) 1 (Pu from spent MOX)	2 (Pu from spent UOX and MOX- FR)	Up to 4 (Depends on the FBR design)

Table 4-7Summary of the Different Options for Pu Management

Fuel Cycle Comparison

Of the five scenarios describe above and summarized in Table 4-7, infinite Pu multi-recycling in LWRs does not appear to be very attractive or feasible in many regards. It does not provide substantial benefits above and beyond those realizable in Pu mono-recycling in LWRs, except with respect to the reduced quantity of Pu sent for disposal. But even for this secondary benefit, the major waste disposal burden results from the minor actinides, which continue to build-up throughout repeated recycling of Pu in LWRs and results in the production of ever greater quantities of the higher actinides such as Cf-252 compared with single pass recycling. Consequently, the infinite multi-recycling of Pu in LWRs fails to adequately address any of the four key challenges faced in pursuing a sustainable nuclear fuel cycle: 1) use of natural resources, 2) economic competitiveness, 3) waste management and 4) non-proliferation.

Similarly, Pu burning in FRs does not look like a feasible option when compared to a fast breeder cycle. A burner cycle requires very advanced technology without ultimately resolving any of the four key challenges to a meaningful extent. While the use of a fast burner cycle does provide natural uranium savings, it yields a modest reduction by a factor of two, whereas a fast breeder cycle can provide U resource savings on the order of 50 times or more relative to a once through cycle. Ultimately, the fast burner option suffers from two major flaws: (1) it does not provide transformational improvements in natural resource sustainability and utilization over that of the reference once-through fuel cycle, and (2) it requires the large scale deployment of advanced technology in the form of reactors and reprocessing facilities in a manner similar to the much more resource-friendly breeder cycle.

For the remaining open fuel cycles, the once-through cycle and Pu mono-recycling in LWRs can be regarded as providing comparable long-term benefits, while differing in two largely offsetting areas. Specifically, the once-through fuel cycle offers greater proliferation resistance, whereas Pu mono-recycling provides modest gains in uranium resource savings.

In light of the above fuel cycle analysis and down selection provided above, the real strategic choice appears to lie between open fuel cycle (once-through or Pu mono-recycling) and Pu breeding in fast reactors. Both present important advantages and disadvantages for consideration. Table 4-8 summarizes major long-term challenges faced by the fuel cycles highlighted and potential solutions to those drawbacks.

Plutonium breeding in FRs implies deployment of advanced technology and therefore involves a significant level of technological risk, especially if minor actinides are to be recycled as well. Table 4-8 indicates that even the pursuit of a more conventional open fuel cycle strategy could entail the application of advanced technologies, such as accelerator-driven systems for transmutation of problematic nuclides, if disposal pathways or breakthroughs in uranium recovery from low grade sources fail to materialize within the requisite timeframe.

Fuel Cycle	Challenge	Potential Solution
Open cycles	Unsustainable uranium resource utilization	Recovery of uranium from seawater
	Accumulation of transuranics in used fuel or reprocessing waste	Transmutation in non-reactor based systems (e.g., accelerator-driven systems)
Pu breeding (FRs) Accumulation of minor actinides		Transmutation in FR systems
	Pu attractiveness in FR blankets	Self-sustaining FR core without any blanket Denaturing of blankets via addition of Np-237 or minor
		actinides

Table 4-8 Comparison between Open Cycle and Pu Breeding in Fast Reactor

4.2.4 Management of Minor Actinides

The minor actinides (MAs) are produced in nuclear fuel under irradiation in either thermal or fast neutron spectra. Once generated, MAs can be eventually recycled in advanced fuel cycles. However, waste management decisions do impact this option. In CEA studies,⁹⁶ the recovery of MAs from vitrified glass for subsequent transmutation was found to be technically challenging and economically prohibitive.

As discussed in Chapter 1, fast spectrum reactors are more efficient for fissioning minor actinides than thermal reactors. As a result, open fuel cycles would require the deployment of dedicated minor actinides burners, such as accelerator-driven systems, which require development of advanced technologies at least as challenging as those for closed fuel cycles. Transmutation of minor actinides also does not yield significant improvements with respect to long-term radiotoxicity if the Pu is segregated and recycled. Consequently, active management of minor actinides has been studied mostly in the framework of closed fuel cycles.

In addition to the considerations above, multi-recycling of MAs in LWRs results in the build up of significant inventories of Cm isotopes and ²⁵²Cf.⁹⁷ Many of these isotopes, especially ²⁵²Cf, are very intense neutron emitters that would render reprocessing or fuel fabrication effectively impossible, given that the neutron source term for multi-pass recycling of MAs in LWRs is more than 2,000 times higher than that for fast reactors. Together, these limitations point to management of MAs exclusively in fast reactor systems; and multi-recycling of MAs in LWRs is not considered further here.

⁹⁶ 2005, CEA "Dossier final: loi du 30 décembre 1991"

⁹⁷ J.-L. Carbonnier, *Merits of Fast Reactors for an efficient use of uranium ore and reduction of ultimate waste*, MIT Symposium: Rethinking the Nuclear Fuel Cycle, Cambridge, MA (October 30-31, 2006)

Management of minor actinides consists of two steps, partitioning and transmutation (P&T), and can be evaluated in terms of three criteria:

- Reprocessing technology difficulty;
- Transmutation efficiency; and
- Impact on the fuel cycle.

As already discussed in Section 1.3.2 under "Long-term Radiotoxicity," waste radiotoxicity is a figure-of-merit that is often invoked to illustrate the relative radiological hazards based on the strength of the radioactive materials source term. The Pu isotopes are by far the most important constituents of spent fuel in terms of radiotoxicity up to a few million years (Figure 1-22). Next are the americium isotopes from ~100 years to ~100,000 years. Next are the curium isotopes between ~300 years and ~30,000 years. The uranium and neptunium contribute minimal amounts to radiotoxicity, given their very low specific activity. Based on radiotoxicity considerations, three recycling strategies for minor actinide management are typically considered: recycling of only Pu, recycling of Pu+Am, and recycling of Pu+Am+Cm.

Given that long-term repository performance is the direct result of the geologic environment, which tends to effectively immobilize the transuranic elements (especially under reducing conditions), P&T allowing for recycling of some or all of the minor actinides offers only modest reductions in radiological hazards. In addition, for countries that already possess significant quantities of vitrified wastes from reprocessing of used LWR fuel, any existing inventories of Am and Cm sent to a geologic repository for disposal will dominate the long-term radiotoxicity even if new inventories of Am and Cm are successfully transmuted.

As an alternative figure of merit, decay heat is generally considered superior to radiotoxicity or volume of waste. And since used fuel or HLW can be cooled in interim storage before being sent to a repository for permanent disposal, waste decay heat after 100 years represents a useful figure-of-merit.

Minor Actinide Partitioning

There are many different incarnations of advanced reprocessing technologies. These range from established PUREX process to pyroprocessing. Most fall under the broader category of aqueous reprocessing. One useful metric for comparing reprocessing technologies is on the basis of their TRU output streams, as illustrated in Table 4-9.

These reprocessing concepts are classified based on their technical difficulty as defined by their departure from the conventional and established PUREX process. Separation of Np together with Pu only requires minor changes to the current PUREX process and could be implemented in a new reprocessing facility based on the currently operating reprocessing plant at La Hague. Another potential adaptation of this process, not mentioned in the table above, is the co-extraction of Pu (and Np) with some U, for enhanced proliferation resistance.

Reprocessing Concept	Stream 1	Stream 2	Stream 3	Waste Stream
PUREX	Pu			Np+Am+Cm+FP
Advanced PUREX	Pu+Np			Am+Cm+FP
Selective extraction of MAs	Pu+Np	Am+Cm		FP
Advanced selective extraction of MAs	Pu+Np	Am	Cm	FP
Grouped extraction of TRU (hydro or pyro processes)	Pu+Np+Am+Cm			FP

Table 4-9Advanced Reprocessing Options

A second process can be added to PUREX process for separating the remaining actinides and lanthanides (heaviest fission products) from the other fission products, followed by a third process to separate the minor actinides (Am+Cm) from the lanthanides. These two additional processes allow selective extraction of MAs, but they significantly complicate the PUREX process. To separate Cm from Am, it would necessary to add a fourth process, which is challenging due to the very similar chemical properties of Am and Cm. Research is also being conducted to achieve direct extraction of Am from the MA+FP stream of PUREX.

Finally, grouped extraction of TRU relies on aqueous or pyrochemical processes completely different from PUREX process (especially pyroprocessing which is the most advanced and the less mature technology presented here).

These processes have proven high level of performance in laboratory, but many technical challenges will have to be overcome in order to make them available on an industrial scale. The advantage of the selective extraction pathway, compared to grouped extraction, is that it is a progressive and incremental evolution of PUREX process, not a revolutionary approach, and therefore represents a risk-limiting approach by taking advantage of the knowledge and experience base accumulated over the past several decades of LWR fuel reprocessing.

Minor Actinide Transmutation

In fast reactors, two modes of MA transmutation are conceivable: homogeneous and heterogeneous. In the first mode, MAs are mixed homogenously with Pu in the fuel, whereas in the second mode, MAs are introduced into the reactor separately from the Pu fuel. In both cases, MAs are subjected to an intense neutron flux in the reactor and undergo a series of neutron capture and/or fission reactions. From a neutronic point of view, capture reactions result in a net consumption of neutrons whereas fission reactions result in a net production of neutrons. The D-value quantifies the net consumption of neutrons for fissioning an actinide and production of all daughters via successive neutron captures or radioactive decay as a function of the neutron energy spectrum. A negative D-value implies a production of neutrons. In LWR spectra,

D-values are globally negative for Pu isotopes but positive for MAs, except for Cm-244, which yields the highly fissile isotope Cm-245 following neutron capture. This neutronic character of the MAs indicates that efficient transmutation requires an external source of neutrons, typically via over-enrichment of U-235 in the fuel. In a fast neutron spectrum, D-values are negative for all actinides; this indicates that efficient transmutation of MAs is feasible in FRs from a neutronic point of view without the need for an external source of neutrons.

• Homogeneous recycle of MA

In this mode, MAs extracted by advanced reprocessing using either selective or grouped extraction technology are recycled together with Pu in FR fuel. One major drawback of this approach is that it results in the contamination of the entire FR fuel cycle with MAs. However, this disadvantage is mitigated by the fact that this dilution avoids MA concentration in any one process or material form.

MAs have an influence on reactor operation by degrading safety coefficients (void, Doppler, reactivity or boron coefficients). CEA calculations indicate that MA fuel content should remain below 2.5% for sodium-cooled fast reactors and 5% for gas-cooled fast reactors.⁹⁸

For a nominal 1,700-EFPD⁹⁹period of irradiation in FRs, CEA calculations¹⁰⁰ indicate that around 25% of the initial MA inventory undergoes fission (24% for Np-237 and Am-241, 15% for Am-243 and 27% for Cm-244), whereas 39% of Np-237, 45% of Am-241, 48% of Am-243 and 23% of Cm-244 underwent successive neutron captures without fission. Accordingly, after a single pass in a fast reactor, approximately one-third of the initial MA inventory is fissioned, one-third is transmuted into higher actinides, and one-third remains in its initial form. As a result, MA transmutation in a homogeneous mode requires multi-recycling of MAs and generates a significant inventory of higher actinides, many of which are intense neutron emitters. Hence, FR fuel at equilibrium in a homogeneous recycle scenario emits 2,000 times more neutrons than LWR MOX fuel. However, this buildup of neutron emitting actinides is relatively minor compared to the buildup of higher actinides for multi-recycling of MAs in LWRs.

The actual numbers will depend on what actinides are effectively recycled. Transmutation of MAs (Np, Np+Am, or Np+Am+Cm) does not instantaneously eliminate them, but instead shifts the management and radiation exposure burden from the backend to another portion of the fuel cycle, greatly complicating fresh fuel fabrication, reprocess spent fuel reprocessing, reactor operation. To evaluate this burden fairly, tracking of the total TRU inventory in the fuel cycle

⁹⁸ CEA « La faisabilité scientifique de la transmutation des déchets à vie longue, » by A. Zaetta et al., RT-SPRC/04-178 (2005)

⁹⁹ EFPD = Effective Full Power Days

¹⁰⁰ CEA « La faisabilité scientifique de la transmutation des déchets à vie longue, » by A. Zaetta et al., p. 13, RT-SPRC/04-178 (2005)

provides a useful figure of merit, which can then be converted into neutron emission, γ dose or decay heat, as shown in Table 4-10.¹⁰¹

Actinides content of FR fuel → (comparison with reference MOX- FR fuel)		2.5% Np	2.5% Am	2.5% Cm
	Decay heat	x 1	x 4	x 12
Fuel Fabrication	γ dose	x 4	x 80	x 500
	Neutron source	x 1	x 2	x 1,700
	Decay heat	x 2	х З	x 6
Reprocessing	γ dose	x 1	x 1	x 1
	Neutron source	x 1	x 4	x 8

Table 4-10 Impact of Recycling Minor Actinides on Fuel Fabrication and Reprocessing

The multipliers in Table 4-10 for key fuel cycle components and MA burden show the relative impact of different MA components. Recycling of Np does not present serious concerns for fuel cycle operations and management. Recycling of Am seems to present greater difficulty due the strong γ emission from Np-239 (daughter of Am-243), but these challenges still appear to be manageable, provided the fuel can be fabricated in hot cells with use of automation and remote handling systems. However, Cm recycling substantially increases the difficulty of fuel fabrication due to the high energy γ emissions from Cm-243 and Cm-244 and the neutron emissions from Cm-244.

The impacts on reprocessing are much less significant due to the invariable presence of fission products for all scenarios, which dominate radiation exposure concerns due to their high specific activities and ubiquitous penetrating γ radiation associated with their decay.

• Heterogeneous recycle of MA

For heterogeneous transmutation, selective extraction of MA is necessary to concentrate the desired minor actinides in dedicated targets. This method reduces the impact of handling MA during reactor operation through segregation, but introduces additional fuel cycle burdens associated with target fabrication and reprocessing of highly radioactive targets.

Single-pass irradiation of these targets avoids reprocessing concerns and the buildup of higher actinides; therefore, research on MA targets tends to emphasize high fission rates (90% fission rates instead of the 25% typical for homogeneous modes). To achieve this performance, MAs are incorporated in an inert matrix, thereby avoiding the in-growth of higher actinides following ²³⁸U

¹⁰¹CEA « La faisabilité scientifique de la transmutation des déchets à vie longue, » by A. Zaetta et al., p. 31, RT-SPRC/04-178 (2005)

neutron capture reactions, and the resulting targets require close proximity to an appropriate neutron moderator for maximizing fission cross-sections in the reactor core.

The in-core inventory of targets is limited by the availability of excess neutrons, which restricts MA fuel content to a level comparable with that of homogeneous modes, i.e., on the order of 2.5% of the total heavy nuclides. In addition, because of helium formation, the targets are damaged during irradiation and the objective of a 90% fission rate is very difficult to achieve. The behavior of the core is also modified by the introduction of the targets: neutron moderation by the targets could lead to increased fissioning of Pu in the core resulting in some core instability and requiring some mitigation to trap moderated neutrons within the targets. Also, the fabrication of these targets with sufficiently high MA content has not been demonstrated on an industrial scale.

As a result of these technological difficulties and challenges, the use of in-core inert matrix targets for heterogeneous transmutation of MA has fallen out of favor.¹⁰² In its place, heterogeneous multi-recycling of MAs in fast reactor blankets has gained in popularity. In this approach, MAs are added to blanket material composed of a ²³⁸U matrix and placed outside of the core's perimeter. This approach has the advantage of not perturbing in-core neutronics and eliminates the need for development of inert matrix targets with high MA loadings and compensating for localized neutron moderation.

One ancillary benefit of heterogeneous MA transmutation in breeder blanket material is the improved proliferation resistance of the blanket material itself. This improvement comes with the considerable decrease in the isotopic quality of the Pu produced in the blanket due to the generation of Pu-238 and Pu-240 from Am and Cm, as shown in Table 4-11.¹⁰³

Isotopic composition of Pu in the blankets after irradiation	Blankets with 10% of MAs	Blankets with 40% of MAs
Pu-238	23%	46%
Pu-239	65%	39%
Pu-240	12%	15%

 Table 4-11

 Isotopic Composition of Pu in Blankets with Minor Actinides

Fabricating, reprocessing, and recycling of this blanket material represent complex technical activities in their own rights and will require additional development and demonstration prior to industrial scale deployment. Table 4-12 below illustrates the technical challenges associated with

¹⁰² Source: "Etude detaillée des scénarios de transmutation faisant appel aux technologies actuelles pour les réacteurs," by F. Varaine, CEA, Rapport Technique DPRGD/2003/2 (2003)

¹⁰³ Source: "Study of minor actinides transmutation in Sodium fast reactor depleted uranium radial blankets," F. Varaine et al., Global 2007, Boise, Idaho, September 9-13, 2007

the increased thermal power and neutron emissions associated with blanket material from homogeneous MA recycling.

In all these scenarios, decay heat and neutron emissions are dominated by the isotope Cm-244, which has a half-life of 18.1 years. As in the case of homogeneous mode, recycling of Cm in heterogeneous mode would complicate the process without providing overall substantial benefits. Consequently, many proposals limit MA transmutation to Am. Because of its overall minimal impact on the fuel cycle relative to other MA elements, neptunium can be flexibly dealt with, either through homogeneous recycling with Pu in the core or through heterogeneous recycling with Am in FR blanket material.

 Table 4-12

 Comparison between MA Homogeneous Recycling and MAs in Blankets¹⁰⁴

Data for Fresh Fuel or Fresh Blankets (Relative Values)	Homogeneous Recycling of MAs (U/Pu + 0.7% MA) [Reference]	Blanket with 10% of MAs	Blanket with 40% of MAs
Relative thermal Power	1	x 8	x 31
Relative neutron source	1	x 11	x 47

Conclusion

Minor actinide management is technically a very complex issue. Four different options can be considered:

- Reprocessing via grouped or selective MA extraction;
- Selection of minor actinides for recycling;
- Selection of homogeneous or heterogeneous recycling modes; and
- For heterogeneous recycling, selection of mono- or multi-recycling of MA options.

The different combinations of these options comprise a population of more than 20 different MA management strategies available for use in fast reactor systems. Screening of this option set based on the overall merits of individual options or elements can quickly and transparently reduce the number to a more manageable subset.

The first logical screening step considers the fate of neptunium in the fuel cycle independently of all other MAs. Np is relatively easy to separate at the reprocessing plant, does not add any significant burden to the fuel cycle either for fuel fabrication or reactor operation, and its transmutation does not lead to significant improvements in backend fuel cycle management (except for geologic disposal in an oxidizing environment for a location such as Yucca

¹⁰⁴ Adapted from Table 5 in "Study of minor actinides transmutation in Sodium fast reactor depleted uranium radial blankets," F. Varaine et al., Global 2007, Boise, Idaho, September 9-13, 2007

Mountain, as discussed in Section 1.3.2). Based on this balanced set of marginal potential gains and drawbacks, Np transmutation does not appear to be a critical component of MA management.

Accordingly, excluding neptunium from the decision matrix leaves nine scenarios for further consideration, as shown Figure 4-23.



Figure 4-23 Minor Actinides Management in Fast Reactors

These remaining scenarios are summarized in Table 4-13 in terms of three key criteria: partitioning difficulty, transmutation efficiency, and overall impact on the fuel cycle.

Scenario	Partitioning Difficulty	Transmutation Efficiency	Impact on the Fuel Cycle
1. Pu recycling	Technology already available (PUREX process)	No MA is transmuted, which leads to a high decay heat of waste	No specific impact
2. Pu and Am homogeneous recycling	Requires advanced selective extraction of actinides to separate Am from Cm. Very complex technology	Transmutation of Am reduces significantly waste decay heat, especially after a few decades of cooling (allows for Cm-244, Sr-90 and Cs-137 decay)	Increases thermal power of fuel at the fabrication and the reprocessing plants
3. Pu, Am and Cm homogeneous recycling	Requires advanced extraction of actinides (separation of Am+Cm from FP). Complex technology	Am and Cm are transmuted, only FP really contribute to waste decay heat	Dramatically increases thermal power and neutron emission of fuel at the fabrication and the reprocessing plants
4. Single irradiation of Am targets	Requires advanced selective extraction of actinides to separate Am from Cm. Very complex technology	Most Am is transmuted but a part still goes to waste with Cm. Moderate impact on waste decay heat reduction	-Targets are very difficult to fabricate because of their high content in MAs -Perturbation of the neutronics in the core -Important damage on the targets during irradiation
5. Am in blankets	Requires advanced selective extraction of actinides to separate Am from Cm. Very complex technology	Transmutation of Am significantly reduces waste decay heat, especially after a few decades of cooling (allows for Cm-244, Sr-90 and Cs-137 decay)	High thermal power at blanket fabrication and reprocessing
6. Single irradiation of Am+Cm targets	Requires advanced extraction of actinides (separation of Am+Cm from FP). Complex technology	Most Am and Cm are transmuted but a part still goes to waste. Significant impact on waste decay heat reduction	-Very difficult to fabricate these targets because of their high content in MA (especially Cm) -Perturbation of the neutronics in the core -Important damages on the targets during irradiation
7. Am+Cm in blankets	Requires advanced extraction of actinides (separation of Am+Cm from FP). Complex technology	Am and Cm are transmuted, only FP really contribute to waste decay heat	High thermal power and neutron emission at blanket fabrication and reprocessing

Table 4-13Comparison of the Minor Actinide Management Options

Scenario	Partitioning Difficulty	Transmutation Efficiency	Impact on the Fuel Cycle
8. All TRU recycling after hydro chemical process	Requires hydrochemical group extraction of actinides. Complex technology	All TRU are transmuted, only FP contribute to waste decay heat	Dramatically increases thermal power and neutron emission of fuel at the fabrication and the reprocessing plants
9. All TRU recycling after pyrochemical process	Requires pyrochemical group extraction. Very complex technology	All TRU are transmuted, only FP contribute to waste decay heat	Dramatically increases thermal power and neutron emission of fuel at the fabrication and the reprocessing plants

Table 4-13 (continued)Comparison of the Minor Actinide Management Options

Each scenario exhibits at least one significant drawback, which indicates that there is no obvious or simple solution to the MA management issue. Any final decision will require weighing of benefits and costs within the technology, policy, and economic contexts.

The least attractive options include Scenarios 8 and 9. In fact, group extractions of actinides remain technologically challenging tasks and are based on a concept fundamentally different from the established, conventional PUREX process. Scenarios 8 and 9 also allow for very little flexibility for MA transmutation (i.e., via homogeneous recycling only).

Scenarios 4 and 6, based on MA targets, also provide relatively unattractive options for MA management. Fabrication of Am targets is feasible but requires the technically challenging separation of Am from Cm. Fabrication of Am+Cm targets on an industrial scale appears to be a non-feasible option due to the high thermal power and neutron emissions associated with the Cm isotopes.

If separation of Am from Cm can be demonstrated on an industrial scale, Scenario 5 (Am heterogeneously recycled in blanket) appears to be preferable to Scenario 2 (Am homogeneously recycled), because it does not lead to widespread contamination of the fuel cycle with Am. The segregation of Am from the fuel cycle is clearly preferable from an industrial point of view (i.e., maintenance, operation, occupational exposure), but excessive build-up of Cm in irradiated Am blankets could lead to very long cooling periods before transportation and reprocessing due to the associated heat loads.

If efficient Am and Cm separation is not realized, fabrication of blankets with Am and Cm make for a very complicated industrial scenario due to the high thermal power and strong neutron emissions. Accordingly, if Am and Cm separation cannot be demonstrated at scale, Scenario 3 (homogenous recycling of Am and Cm) becomes the more favorable approach over Scenario 7 (Am and Cm in blankets).

Scenario 1 (Pu recycling) represents an option that relies exclusively on disposal for MA management. In this regard, no advanced reprocessing technology is required and there are no

deleterious impacts on the fuel cycle. This option could be pursued as an interim step until Scenario 3 (homogeneous recycling of Am and Cm) or Scenario 5 (Am in blankets) could be implemented.

4.2.5 Management of Fission Products

Compared to the case of minor actinides, fission product management is far less amenable to separation and transmutation techniques. Fundamentally, fission products do not undergo further fission in thermal or fast reactor spectra and are therefore only transmutable by neutron capture reactions. Secondly, among the hundreds of different fission products produced in reactors, only a small subset are really troublesome for justifying additional manipulation for waste management purpose. Most have either (1) very short half-lives allowing decay to effectively remove them from inventories over relatively short timeframe, or (2) extremely long half-lives, which means very low specific activities.

The issue of fission product management can be narrowed to the consideration of only five key isotopes: ¹³⁷Cs, ¹³⁵Cs, ¹²⁹I, ⁹⁰Sr, and ⁹⁹Tc. The isotopes Cs-137 and Sr-90 (with their respective daughters Ba-137 and Y-90) are responsible for more than 90% of FP decay heat alone. Tc-99, I-129, and Cs-135 represent the three most important long-lived fission products; they are less radiotoxic than the long-lived minor actinides, but are generally much more mobile in the environment. Accordingly, these three fission products tend to dominate the long-term performance of geologic disposal systems such as mined repositories.¹⁰⁵

Fission Product	Half-Life (Years)	Decay Heat at Discharge (W/MT of Spent UOX)
Cs-137 (Ba-137)	30.17 (2.5 minutes)	561
Sr-90 (Y-90)	28.6 (64.1 hours)	565
Tc-99	210,000	Negligible
I-129	15,700,000	Negligible
Cs-135	2,300,000	Negligible

Table 4-14Main Characteristics of Several Fission Products

In contrast to the minor actinides, fission products are not candidates for recycle as they do not provide energy benefits (or neutron generation) via fission reactions. Their production is also an inevitable outcome of fission whether in thermal or in fast spectrum reactors. Accordingly, the only fuel cycle choices associated with fission product management are to send them for permanent disposal or to transmute them. As the *disposal* of fission products is reasonably straightforward and well understood, with the most important fission products, ¹³⁷Cs and ⁹⁰Sr,

¹⁰⁵ The isotopes Cl-36 and Se-79, which are generated in small quantities in nuclear reactors, can also have disproportionate impacts on the long-term performance of disposal systems.

decaying in a few hundred years, this section focuses exclusively on fission product *partitioning and transmutation* of long-lived fission products.

The principle of fission product transmutation consists of irradiating them under conditions favoring neutron capture in order to create more desirable radioelements, i.e., stable nuclides or short-lived nuclides that decay into stable ones. Because fission products do not participate in fission reactions, this approach is a net consumer of neutrons for both thermal and fast spectrum reactors.

Short-Lived Fission Products

Because of their short half-lives, Cs and Sr do not represent a long-term radiological concern for geologic disposal over timeframes on the order of thousands to hundreds of thousands of years. However, they do contribute significantly to the decay heat of used fuel - especially for used UOX fuel that contains a lower inventory of heat generating minor actinides than used MOX (Figure 4-22).¹⁰⁶



Figure 4-24 Decay Heat Contribution of Cs and Sr to Spent UOX and Spent MOX with Burnup of ~50 GWd/MTHM

Separation of Cs-137 and Sr-90 would obviously decrease the decay heat of the waste. After separation, they could then be placed in an interim storage facility before being sent to a geologic repository. But the same result could be achieved by cooling intact spent fuel assemblies or

¹⁰⁶ Source: EDF R&D

HLW before sending them to the final disposal. As a consequence, the potential waste management benefits provided by Cs and Sr separation, such as reduced heat generation loading in a disposal facility, do not justify the technical and operational complexity associated with deployment of Cs and Sr separation technology at an industrial scale.

Long-Lived Fission Products

Partitioning specific long-lived fission products substantially complicates reprocessing. CEA research does, however, show that their extraction is technically feasible with modification of the PUREX process, albeit with a number of important limitations and considerations:¹⁰⁷

- 99% of iodine is released as a gas during the dissolution of spent fuel at the beginning of the PUREX process. 97% is recovered in treatment effluents and 2% is trapped in iodine filters. Today, effluents are released into the sea in order to efficiently dilute I-129. Alternatively, it is conceivable to put iodine in a chemical stable form in order to transmute it in reactors.
- Technetium could also be recovered with some adaptations to the current PUREX process or with the UREX+ process. However, a substantial fraction of Tc is present in a form that is very hard to dissolve. Tc would then have to be converted into a carbide or metal form to be transmuted in reactors.
- Cesium cannot be easily extracted by the current PUREX process. It is easier to separate it from the Fission Product stream after selective extraction of actinides. This stream contains most of the FP except for the lanthanides. Cs could also be extracted at the second step of the UREX+ process, along with Sr. In any case, Cs partitioning requires advanced technology.

The three long-lived fission products, ¹³⁵Cs, ¹²⁹I, and ⁹⁹Tc, could in principle be transmuted in LWRs or in FRs via neutron capture, but their respective cross sections are very small in both neutron spectra: less than 1 barn in a fast spectrum and a few barns in a thermal spectrum. As a result, the expected transmutation times of these isotopes could be very long. In addition, other isotopes of I and Cs are present in the fission products that could complicate the process (Table 4-15).

UOX Fuel With a Burnup of 60 GWd/MTHM	Production of the Element (g/MTHM)	Production of the Isotope (g/MTHM)
Cs-135	4,600	769 <i>(17%)</i>
I-129	379	308 <i>(81%)</i>
Tc-99	1,410	1,410 <i>(100%)</i>

Table 4-15 Cs, I, and Tc Production in Spent Fuel

Cs-135 represents only 17% of Cs in spent fuel compared to 41% of Cs-133 (stable form) and 42% of Cs-137 (short-lived with high decay heat). As a result, irradiation of Cs could result into

¹⁰⁷ Source : CEA « La faisabilité scientifique de la transmutation des déchets à vie longue, » A. Zaetta et al., RT-SPRC/04-178 (2005)

two consecutive neutron capture of Cs-133 that would increase the Cs-135 inventory. So efficient transmutation of Cs-135 would require isotopic separation before irradiation, which would be very expensive. Combined with the advanced reprocessing technology necessary to extract Cs, it can be concluded that P&T of this element is not a realistic option.

I-129 represents 81% of Iodine in spent fuel; the other 19% are I-127, the stable form of this element. As a result, the situation is somewhat similar to Cs, except that the isotope that must be transmuted is the most abundant one, so there is less I-129 produced by double neutron capture of I-127 than effectively transmuted I-129. However, studies from CEA show than if iodine could be theoretically transmuted, its chemical form would not be stable under irradiation in a real reactor. So, P&T of I-129 does not appear to be an attractive option for the current technology.

Tc-99 is the only isotope of Tc present in fission products. In addition, it is the most abundant and represents 96% of the activity of the three major long-lived FP considered. Tc-99 can be transmuted into the stable Ru-100 via neutron capture. It is possible to build targets of pure metallic Tc-99. Transmutation rates around 15% have been demonstrated: 25% is projected. So, efficient transmutation of Tc-99 would require several passes in the reactor. Even in the most favorable case, hundreds of years would be necessary to reduce the Tc-99 inventory by a factor of 10.

While partitioning of long-lived fission products may ultimately prove to be feasible, such an approach will at best be restricted to the most problematic fission product constituents.

Summary

The partitioning and/or transmutation of fission products, while theoretically possible, do not appear to be realistic for deployment on an industrial scale. Release of I-129 into seawater during reprocessing, natural decay of relatively short-lived Cs-137 and Sr-90 and geologic storage of Cs-135, Tc-99 (and also Se-79) currently offer acceptable and relatively simple management of the fission product burden. In the long-term, only transmutation of Tc-99 (which is the most active long-lived fission product) appears to be feasible.

5 CONCLUSION: ATTRIBUTES OF A SUSTAINABLE NUCLEAR FUEL CYCLE

A sustainable nuclear fuel cycle should achieve three major objectives.

First, it has to rely on the energy content of U-238, which represents more than 99% of the natural uranium. A partially closed fuel cycle with fast reactors, in which fertile U-238 is converted into fissile Pu-239, is presently considered the most attractive advanced option. Another possibility is the thorium fuel cycle, in which fertile Th-232 is converted into fissile U-233. However, much less work has been conducted on the thorium fuel cycle, and its supporting infrastructure is still in its infancy. Cost-effective recovery of uranium from seawater could make the once-through fuel cycle practically sustainable with regard to natural uranium supply, but the required technology is likely to remain prohibitively expensive.

Second, a sustainable nuclear fuel cycle has to be as simple as possible. Many different options are on the table; most of them represent dramatic changes compared to the current situation. What works on paper does not necessarily lend itself to industrial scale deployment. To put it differently, a nuclear fuel cycle has to be "industrially sustainable". An evolutionary and progressive pathway appears to be more realistic than a revolutionary approach that attempts to solve all the fuel cycle issues with extremely advanced technologies. The separation and the identification of the different fuel cycle choices, described in Section 4, help to elaborate this evolutionary pathway:

- 1. Once-through cycle.
 - 1a. Option: reprocessing of the used LWR fuel and single-recycling of the extracted plutonium and reprocessed uranium into LWRs. This option is most suitable as long as there is no near-term pathway to recycle the extracted plutonium in a fast reactor, while at the same time mastering the industrial deployment of reprocessing technology (likely to be PUREX, or an evolution of the PUREX process).
- 2. Interim storage of spent UOX and spent MOX.
- 3. Partial closure of the fuel cycle with multi-recycling of plutonium in fast reactors (FRs) requiring advanced reprocessing of both LWR and FR fuels (likely to be based on the experience accumulated with the PUREX or PUREX-like process).

3a. Option: recycling of the neptunium together with the plutonium.

4. Full closure of the fuel cycle with homogeneous multi-recycling of plutonium and minor actinides requiring group separation of the transuranic elements.

- 4a. Option: Full closure of the fuel cycle with heterogeneous recycling of americium in the form of americium targets and storage of curium to allow decay into lower actinides.
- 5. In all cases, disposal of fission products and of remaining actinides in a permanent geologic repository.

Of course, other pathways are conceivable, depending on the evolution of current technologies and the emergence of new ones (boreholes, accelerator-driven systems, fission-fusion hybrids, etc.). A fuel cycle strategy must provide a general framework, but flexible enough to (i) adapt to changes in technologies, and (ii) integrate in due time separation and transmutation technologies that may prove helpful in facilitating public acceptance related to the management of radioactive wastes.

Third, with nuclear energy's contributions to satisfying worldwide energy demand expected to increase in the decades to come, the nuclear fuel cycle has to remain focused on efficient power generation. The externalities of nuclear energy, such as waste generation and proliferation risks, have to be addressed in a safe, but reasonable way. Thus, advocating transmutation of all the transuranics and fission products, or making nuclear materials so unattractive that they are practically unusable in the fuel cycle itself, do not represent realistic options.

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