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ABSTRACT

Given the range of fuel cycle goals and criteria, and the wide range of fuel cycle options, how can the set of options eventually be narrowed in a transparent and justifiable fashion? It is impractical to develop all options. We suggest an approach that starts by considering a range of goals for the Advanced Fuel Cycle Initiative (AFCI) and then posits seven questions, such as whether Cs and Sr isotopes should be separated from spent fuel and, if so, what should be done with them. For each question, we consider which of the goals *may be* relevant to eventually providing answers.

The AFCI program has both "outcome" and "process" goals because it must address both waste already accumulating as well as completing the fuel cycle in connection with advanced nuclear power plant concepts. The outcome objectives are waste geologic repository capacity and cost, energy security and sustainability, proliferation resistance, fuel cycle economics, and safety. The process objectives are readiness to proceed and adaptability and robustness in the face of uncertainties.

We pose the following seven questions:

I. How do we address proliferation resistance?
II. Which potential energy futures do we plan for? What are the ramifications regarding the number of geologic repositories and use of U resources?
III. Should we separate uranium? If we separate uranium, should we recycle it, store it or dispose of it?
IV. Which transuranic (TRU) elements should be separated and transmuted?
V. Of those TRU separated, which should be transmuted together?
VI. Should we separate and/or transmute Cs and Sr isotopes that dominate near-term repository heating?
VII. Should we separate and/or transmute very long-lived Tc and I isotopes?

For each question, we summarize relevant issues associated with the range of AFCI goals. *We emphasize that this analysis is a "work in progress" and specifically invite comments and suggestions so that this on-going work is more complete, correct, and comprehensive.* The real message of this paper is not in the details, which are still evolving, but that this is a complex issue involving many priorities (sometimes conflicting) and significant unknowns. The "solution" is still unclear.

AFCI GOALS

The draft FY2004 Comparison Report to Congress from the AFCI program states…

> "The AFCI program addresses critical national needs associated with past and future use of nuclear energy. First, the AFCI provides alternatives to building multiple geologic repositories for disposal of past and future commercial spent nuclear fuel, while supporting an expanding role for nuclear energy. Second, the AFCI provides fuel cycles that recover most of the energy content in spent nuclear fuel, in conjunction with the complementary Generation IV Nuclear Energy Systems

> Initiative (Generation IV). Third, the AFCI provides nuclear fuel cycles that improve proliferation resistance via advanced separations and fuels technologies, by reducing the inventory of weapons-usable material, and by eventually reducing the need for uranium enrichment. While accomplishing these goals, the AFCI program also will ensure competitive economics and excellent safety for the entire nuclear fuel cycle."

This text motivates seven goals, which are listed in Table 1. The Table goes further by listing potential criteria associated with each goal. Taken together, the goals and criteria are inclusive of past public justifications for the program and appear sufficient to inform key future program decisions without producing unbalanced selections.

Table 1. AFCI Goals and Associated Criteria

<table>
<tr><th colspan="4">Process Goals and Criteria</th></tr>
<tr><td colspan="2">1. Consider Readiness to Proceed
• Technical Knowledge is Sufficient to Proceed
• Legal and Policy Constraints are Identified and Managed
• Infrastructure Constraints are Identified and Managed</td><td colspan="2">2. Maximize Adaptability and Robustness
• Scenario is Adaptable to Nuclear Futures and External Events
• Fuel Cycle Operations are Flexible and Robustness
• Exit Options are Maintained</td></tr>
<tr><th colspan="4">Outcome Goals and Criteria</th></tr>
<tr><td colspan="2">3. Improve Waste Management, especially geologic repository ramifications
• Geologic Repository Post-Closure Radiotoxicity is Managed
• Geologic Repository Post-Closure Heat Load is Managed
• Geologic Repository Packaging Requirements are Reduced
• Production of Other Wastes is Managed</td><td colspan="2">4. Improve Proliferation Resistance and Physical Protection
• Technology Protection is Considered
• Weapons Usable Material Protection is Considered
• Radiological Source Material Protection is Considered</td></tr>
<tr><td>5. Recover Energy from spent fuel
• Natural Resources are Utilized Effectively</td><td colspan="2">6. Ensure Competitive Economics (as part of the entire fuel cycle)
• Fuel Costs are Managed
• Electricity Production is Enhanced
• Non-Electric Production is Enabled
• Elemental Separations Costs are Managed
• Waste Disposition Costs are Managed</td><td>7. Ensure Safety (as part of the entire fuel cycle)
• Raw Material Processing Safety is Enhanced
• Fuel Processing Safety is Enhanced
• Energy Generation Safety is Maintained
• Transportation Safety is Maintained</td></tr>
</table>

The first two goals are "process," how we get from here to there, how ready are we to proceed? The other five are "outcome", which are consistent with, but distinct from, Gen IV goals.

Regarding the process goals, consider that successful programs are often those that can address a key problem in reasonable time when the need becomes apparent. For example, if asked, "can AFCI help move spent nuclear fuel (SNF) from current nuclear power plant (NPP) sites faster than is currently planned, to reduce safety and terrorist threats," how could we respond? We could answer that processing SNF in *parallel* to shipping unprocessed SNF to the Yucca Mountain Project (YMP) would reduce the NPP SNF inventory faster. If asked "how soon?" We could answer that, if this were a national priority, this could start in a decade or so. This is similar to the time horizon suggested by the 8-year time lag between the 2002 Congressional override to planned YMP operation in 2010.

These goals and criteria for AFCI emphasize the balance of the fuel cycle with only limited consideration for the NPP portion of the total fuel cycle, they are therefore complementary with Generation IV NPP goals.[GIF2002] Engineered reliability is ultimately to be addressed as part of economics. Economics also includes a non-electric production criterion to provide linkage with the Nuclear Hydrogen Initiative.

QUESTIONS TO ANSWER

Given the range of goals and criteria, and the wide range of fuel cycle options, how can the set of options eventually be narrowed in a transparent and justifiable fashion? We suggest an approach that posits seven questions. We produced this list by considering the key issues being analyzed and discussed within the program. For each question, Table 2 indicates which of the seven goals *may be* relevant to eventually providing an answer.

Table 2. Which Goals May Be Relevant to Each Question?

Goals Questions	Consider readiness to proceed	Maximize adaptability and robustness	Improve waste management (geologic repository)	Improve proliferation resistance and physical protection	Recover energy from SNF	Ensure competitive economics (as part of the entire fuel cycle)	Ensure safety (as part of the entire fuel cycle)
I. How do we address proliferation resistance?	X	X		X			
II. Which potential energy futures do we plan for? What are the ramifications regarding the number of geologic repositories and use of U resources?		X	X		X		
III. Should we separate uranium? If separate uranium, should we recycle it, store it, or dispose of it?	X	X	X	X	X	X	X
IV. Which TRU should be separated and transmuted?	X	X	X	X	X	X	X
V. Of those TRU separated, which should be transmuted together?	X	X		X		X	X
VI. Should we separate and/or transmute Cs and Sr isotopes that dominate near-term repository heating?	X	X	X	X		X	X
VII. Should we separate and/or transmute very long-lived Tc and I isotopes?	X	X	X			X	

At the current time, rather than attempt to weight goals, we suggest (a) determining for each decision whether each option offers improvement for that goal and (b) attempting to create as many win-win decisions (attractive or neutral to most goals) as possible. Goals relevant to a given decision will vary as to their importance; they are unlikely to have equal weight. The relative importance of goals will vary among decisions, as a consequence of other decisions being taken, and Energy Future. The concept of Energy Future is discussed under Question 2.

QUESTION I.
How do we address proliferation resistance?

There are (at least) three possible ways to analyze proliferation resistance; we need to consider all three:

- Determine the inventory of weapons usable (WU) material.
- Measure intrinsic proliferation resistance, such as the so-called "spent fuel standard"
- Perform a comprehensive measurement of proliferation resistance including both intrinsic and extrinsic (safe guard technologies) measures.

We should never imply that a fuel cycle that involves WU material or does not fully meet the "spent fuel standard" lacks proliferation resistance; just as we would never imply that a NPP that was not "passively safe" lacks safety. In fission reactor safety, comprehensive probabilistic risk assessments can (and do) show that a given reactor design is extremely safe by taking credit for active safety systems. That is suitable for technically-aware and regulatory-minded experts. It is not fully satisfactory for much of the public. Dependence on complicated extrinsic and intrinsic proliferation resistance would seem to be analogous to complex probabilistic risk assessment. Indeed, much of the methodology is the same. Both probabilistic risk assessment and comprehensive proliferation assessments *are necessary*; they are sufficient in a technical sense in protecting people; but, they may not be sufficient for advancement of nuclear power. Thus, NPP designers now seek "passive" safety, also known as intrinsic safety, or walk-away safety. This gives extra emphasis to characteristics that are always available to protect the public (natural circulation for example), even if all power is lost and all operators walk away. The analogous proliferation resistance protection would seem to be meeting the spent fuel standard, or something similar. The analogy in Table 3 might be helpful.

Table 3. Analogy Among Power Plant Safety, Proliferation Resistance, and Geologic Disposal

	Reactor safety	Proliferation resistance	Geologic disposal
Current fuel cycles and reactors	Use human operation, active safety systems, passive features, and inventory control, to provide complete protection of the public. Analyze via complex probabilistic risk assessment	Use human protectors, safeguard technologies, intrinsic characteristics, and inventory control to provide proliferation resistance. Analyze with comprehensive assessments	Use institutional control, engineered features, natural geologic features, and inventory control to provide protection. Analyze with complex engineering-subsurface assessments.
Potential objectives for advanced fuel cycles and reactors	Use passive safety and inventory control to ensure public protection against major accidents, avoid need for emergency evacuation planning. Augment as needed with active systems to provide operationally safe plants.	Use intrinsic characteristics to provide as much protection as possible (e.g. spent fuel standard), augment with extrinsic protection.	Use geologic features and inventory control to provide long-term protection, e.g., reduce radiotoxicity below that of uranium ore in X years. Augment with engineered features to provide nearer-term additional protection.

Issues associated with this question

Issues associated with the goals relevant to this question will now be discussed.(Goals 1, 2, 4 in Table 2.)

Goal 1. Consider readiness to proceed

Ii is inconceivable that any new nuclear technology will be deployed unless there is a strong case that proliferation resistance is maintained, preferably improved. In any time period exceeding 12 years (e.g. President George W. Bush elected in 2000 through the next three elections in 2004, 2008, 2012), history suggests that there is an approximately 84% probability a change of political party will occur in the Executive Branch. Thus, the proliferation resistance case must have some degree of bipartisan support. Until there is a clear consensus on what constitutes "proliferation-resistance", there can be no unambiguous assessment of any fuel cycle. Accordingly, the best near-term efforts would be those directed at establishing what constitutes "proliferation resistance" in both technically defensible and internationally acceptable terms – presumably a mixture of intrinsic and extrinsic protection.

Goal 2. Maximize adaptability and robustness

The best approach is to consider how to improve fuel cycle options as much as possible from *both* material (intrinsic) and technology diversion (extrinsic) perspectives, while considering the corresponding disadvantages (if any) from the perspectives of waste management, sustainability, safety and reliability, and economics.

Goal 4. Improve proliferation resistance and physical protection

It would be very advantageous if a fuel cycle might possibly be developed which avoids **"weapons usable"** material. Such an advanced fuel cycle might prove highly desirable over the current once-through approach.

The IAEA's definition of "weapons usable" includes two factors - could a sphere of the pure metal go super prompt critical and can the material be kept together long enough to construct a weapon? The first criterion eliminates Th and low grade U as being WU. The second factor focuses on whether the material would generate so much heat that construction of a weapon is impractical; it eliminates Pu over 80% Pu-238. It appears that it would eliminate AFCI-relevant pure Cm compositions (because of Cm-244 heat generation). Am+Cm mixtures would be borderline (Cm-244 and Am-241). Pure Am, even pure Am-241, does not generate enough heat to match the heat generation rate of 80% Pu-238 and thus Am would appear to be weapons usable. No U-Pu fuel cycle appears to avoid the "weapons usable" designation.

A pure Th cycle will also not avoid "weapons usable" designation, because it generates U-233 exceeding 12% of the U content. The only conceivable fuel cycle that would not involve weapons usable material per IAEA definition would be a hypothetical Th-U-Pu mixture that had sufficient U to denature U-233 and sufficient high-heat-emitters (like Pu-238 and Cm-244) that it met the Pu-238 style heat generation criterion. It is unlikely that such a fuel cycle exists, but we are not aware of a definitive study on this point. If such a fuel cycle did exist, it would be very difficult to handle and fabricate associated fuels.

Even if (as seems likely) any fuel cycle involves weapons usable material, the inventory of weapons usable material is clearly an important metric, one easily defined and calculated. This is more defendable than simply discussing Pu-239 or Pu-total inventories.

Another line of query would be to determine which fuel cycles meet the "spent fuel standard," defined as the fuel cycle being as proliferation resistant as SNF. We focus on the spent fuel standard because it is the most recognized way of characterizing intrinsic resistance. The simplistic interpretation of the "spent fuel standard" is simply gamma field, X rem/hour. However, the more inclusive interpretation of the "spent fuel standard" would definitely give credit for chemical and physical characteristics of waste/storage forms relative to spent fuel's characteristics.[NAS2000] Maximizing achievement of the "spent fuel standard" would seem to be a worthy objective. Using the more inclusive definition of [NAS2000] increases the chance of success and increasing the technical defendability of the analysis.

Ultimately, we must **combine both extrinsic and intrinsic protection**. Advanced fuel cycles should concentrate on designing easy to safeguard facilities without adding excessive costs, administrative or operational burdens on the nations in which the facility is present. Although one possibility should be the development of better technologies to monitor process streams, a holistic approach of integrating such safeguards and security features during early design and construction phases should be investigated thoroughly. A useful starting point might be to assess the current reasoning regarding proliferation resistance to ascertain at what point is SNF cool enough to not meet the Spent Fuel Standard, and our "Stand Alone" storage facilities require additional protection. Next would be to consider whether proliferation resistance must be applied at a specified certain baseline level throughout the fuel cycle or rather can various optimum measures be applied to different stages along the chain requiring greater protection. That is, the best case would be if all portions of the fuel cycle met the "spent fuel standard;" the next best would be minimize the "exposure" from portions that do not meet the "spent fuel standard".

Nonetheless, we should always ensure that a fully comprehensive analysis - using intrinsic and extrinsic protection - shows excellent proliferation resistance. The search for simpler metrics (inventory of weapons usable material, intrinsic protection characteristics) should not downgrade the importance of comprehensive protection.

QUESTION II.

Which potential energy futures do we plan for? What are the ramifications regarding the number of geologic repositories and use of U resources?

The success of energy supply and technology forecasts have been poor, see for example [Bezdek and Wendling2002]. Yet, because decisions today have ramifications for decades in the future, there is no alternative but to forecast a range of futures, and plan for a reasonable set of them. Indeed, the estimated attractiveness of a given future (what it takes to get there) influences whether actions are taken that maximize the chance of achieving that "energy future."

Table 4 defines six Energy Futures. We believe that AFCI plans and technology R&D should accommodate (to the extent practical) the entire range, especially those on the right side of the table. Future 1 – Legislative Limit for the first geologic repository - is the only future in which the current statutory capacity of YMP (70,000 MT) is adequate. The U.S. is already in Future 2, Existing License Completion. Indeed, as additional license extensions are granted, we are moving steadily toward Future 3, Extended License Completion, which would result in about 120,000 MT of SNF cumulative by the year 2100. While the statutory limit for YMP is 70,000 MT, the 2001 YMP Science and Engineering Report explores options that would accommodate 119,000 MT. The value of 119,000 MT is as high a value for the capacity as we have seen published by YMP, implying the technical capacity is at least his high; the actual value is uncertain.

Table 4. Potential Energy Futures (See Dixon and Piet, Impact of Nuclear Energy Futures on Advanced Fuel Cycle Options, this conference)

Future	Total Discharged Fuel
1. Legislative Limit	70,000 MT = Based on the legal capacity of the first repository per the Nuclear Waste Policy Act (63,000 MTiHM commercial, 7,000 MT defense)
2. Existing License Completion	100,000 MT = Based on existing spent fuel inventories plus a plant-by-plant extrapolation of future discharges developed using current discharge rates until the end of each operating license, including known license extensions as of 10/03 – result rounded.
3. Extended License Completion	120,000 MT = Based on existing spent fuel inventories plus a plant-by-plant extrapolation of future discharges assuming on all operating plants having one 20 year extension, result rounded.
4. Continuing Level Energy Generation	250,000 MT = Based on extension of the current average annual SNF discharge rate of 2100 MT/yr through the year 2100.
5. Continuing Market Share Generation	600,000 MT = Extension of the current average annual SNF discharge rate through 2100 with 1.8% compounded market growth starting in 2004.
6. Growing Market Share Generation	1,500,000 MT = Extension of current average annual SNF discharge through 2100 with 3.2% growth in nuclear power.

Issues associated with this question

Issues associated with the goals relevant to this question will now be discussed.(Goals 2, 3, 5 in Table 2.)

Goal 2. Maximize adaptability and robustness
Goal 3. Improve waste management, especially geologic repository ramifications

How well could the nation adapt to the higher energy futures? An obvious question is whether a single geologic repository (assumed to have the characteristics of YMP) would be adequate for the SNF generated this century. We consider three broad types of fuel management strategies, starting with the once-through fuel cycle (baseline, all SNF goes to geologic repository). The second case is limited recycle in thermal reactors. The calculations have been based on light water reactor performance (LWR). The third case is repeated recycle in thermal and/or fast reactors.

Wigeland et al [Wigeland2004a], [Wigeland2004b] have analyzed how much the repository capacity can be effectively increased for different fuel cycles, given that capacity is first limited by long-term heat generation of emplaced waste in the current US repository design. A repository factor capacity factor of **1.0** represents once-through; all SNF is emplaced.

Recycling long-term heat-dominate isotopes (especially Pu-241 and Am-241) a few times in thermal reactors can achieve a repository factor increase of perhaps **1.7x**. That is, about 1.7 times more GWe-year of nuclear power can be generated for the same repository capacity; there is less long-term heat generated per GWe-year because many of the key isotopes are transmuted. In thermal reactors, a repository factor increase of 1.7 is relatively easier and quicker to achieve with so-called inert matrix fuel than with mixed oxide fuel, because the former produces no new TRU from U-238 capture. Indeed, a single recycle with mixed oxide fuel may only achieve an increase of about 1.2. Wigeland's work also shows it is best from this perspective to recycle the youngest fuel first, because long-term heat load from Pu-241/Am-241 is more efficiently reduced if Pu-241 is transmuted before decay to Am-241.

With repeated recycles, the door opens for higher repository capacity increases. The way to achieve a high number of recycles of material that has received the most attention and is probably the most credible is to include fast reactors in the mix. Fast reactors better transmute a broader range of TRU than do thermal reactors. This means that a mix of isotopes can be recycled more times with lower penalty to maintaining criticality in the reactor. Fast reactors can recycle fuel almost indefinitely, with natural uranium added each cycle. In contrast, thermal reactors with inert matrix fuel (i.e. no new U-235 added) can probably only recycle material one or two times; the fuel lacks sufficient "punch" to keep going. By more effectively reducing TRU and their associated long-term heat load, repository capacity increases up to **5x** can be achieved. To go further requires also addressing shorter-term heat load which is dominated by Cs and Sr, see question VI. Then, fuel cycles with fast reactors (either totally fast reactors or a mix of thermal and fast reactors) can probably achieve repository capacity factor increases of **~50x** based on Wigeland's work, with repeated recycle of material to transmute TRU and remove Cs and Sr.

So, with three representative fuel management strategies, we can address three issues for each Future.

- How many geologic repositories are needed to accommodate SNF discharged by 2100? (Table 5).
- For different energy futures, how much of repository capacity must be reserved for processed waste (versus unprocessed waste) as a function of energy future, repository capacity, and fuel management approach? (Table 6)
- How much of the fuel discharged by 2100 would have to be processed so that the processed+unprocessed waste fit into one repository? (Table 6)

Table 5. Number of Geologic Repositories Needed by 2100

	Nuclear Futures	Legislative Limit	Existing License Completion	Extended License Completion	Continuing Level Energy Generation	Continuing Market Share Generation	Growing Market Share Generation
	Cumulative discharged fuel in 2100 (MTiHM)	63,000	100,000	120,000	250,000	600,000	1,500,000
Fuel Management Approach	Repository Capacity (MT)	**Number of Geologic Repositories Needed to Accommodate Fuel Discharged by 2100**					
No recycle	70,000	1.0	1.6	1.9	3.7	8.7	21.6
	119,000	0.6	0.9	1.1 (a)	2.2	5.2	12.7
Limited thermal recycle (capacity multiplication 1.7x)	70,000	0.7	1.0	1.2 (b)	2.3	5.2	12.8
	119,000	0.4	0.6	0.7	1.3 (c)	3.1	7.5
Repeated thermal+fast recycle (capacity multiplication of 50x)	70,000	0.2	0.2	0.2	0.2	0.3	0.6
	119,000	0.1	0.1	0.1	0.2	0.2	0.4

Repository capacity dominated by temperature limits, hence decay heat density.

a. Borderline case, can be reduced to 1 repository with further repository capacity increase from 119,000 to 127,000 MT or a 10% increase in fuel burnup.

b. Borderline case, can be reduced to 1 repository with 20% increase in fuel burnup.

c. Borderline case, can be reduced to 1 repository with further repository capacity increase from 119,000 to 154,000 MT or a 30% increase in fuel burnup.

Table 6. How much of geologic repository must hold processed waste, how much fuel must be processed?

	Nuclear Futures	Legislative Limit	Existing License Completion	Extended License Completion	Continuing Level Energy Generation	Continuing Market Share Generation	Growing Market Share Generation
	Cumulative discharged fuel in 2100 (MTiHM)	63,000	100,000	120,000	250,000	600,000	1,500,000
Fuel Management Approach	YMP Capacity	**What % of repository must hold processed waste? What % of fuel generated by 2100 must be processed?**					
No recycling	70,000	Capacity sufficient	🟨	🟨	🟨	🟨	🟨
	119,000		Capacity sufficient	🟨	🟨	🟨	🟨
Limited thermal recycle	70,000		84% of YMP 90% by 2100	🟨	🟨	🟨	🟨
	119,000		Processing not needed	10% of YMP 16% by 2100	🟨	🟨	🟨
Repeated thermal+fast recycle	70,000		1% of YMP 38% by 2100	2% of YMP 48% by 2100	6% of YMP 76% by 2100	17% of YMP 91% by 2100	47% of YMP 98% by 2100
	119,000		Processing not needed	<1% of YMP 7% by 2100	3% of YMP 56% by 2100	9% of YMP 83% by 2100	25% of YMP 94% by 2100

Table 5 shows our estimate of the number of geologic repositories needed for 2100. Shaded (yellow) cells require more than one repository; green cells require only one repository. By definition, if the repository capacity is 70,000 MT (63,000 of which is for commercial SNF), in only the "Legislative Limit" future is one repository adequate in the absence of recycling. The nation is already implementing "Existing License Completion" meaning that (a) the repository capacity must be increased to at least 120,000 MT (plus allowance for defense wastes), (b) a second repository must be built, or (c) recycle of at least some SNF must be done. By 2100, without recycling of some type, there could be 22 repositories worth of SNF generated from the decay heat perspective.

Table 6 takes a deeper look at those cases where one repository appears adequate. How much of the SNF must be recycled to "fit" the waste into the repository? How much of the repository must be used to store processed versus unprocessed fuel?

By definition, if discharged SNF is limited to 63,000 MT, then a repository capacity of 70,000 (or 119,000) is adequate. In this case, 100% of YMP can hold unprocessed waste and 0% of the fuel must be recycled. This situation (shaded green in Table 6) is also true if the repository capacity is 119,000 and the accumulated fuel is 100,000.

Between the cases where no recycling is needed (shaded green) and cases where one repository is not adequate (yellow) are the intermediate cases (shaded white) where the repository would contain a mix of unprocessed and processed fuel. A specific example may help explain Table 6. Consider the cell "Continuing Energy Level Generation" + "Repeated thermal+fast recycle" + repository capacity of 70,000 MT. To avoid exceeding 70,000 (with 7,000 reserved for DOE waste), 76% of the 250,000 MT to be generated by 2100 must be processed with a factor of 50, so that the 250,000 partitions into 191,000 processed + 59,000 unprocessed. This means that 6% of the 63,000 MT assigned to commercial waste would contain processed waste (191,000 MT effectively reduced to 4,000) and 94% of the repository's 63,000 MT would be unprocessed waste (59,000 MT).

As one moves right in Table 6, more SNF must be processed to stay within one geologic repository. A higher fraction of the repository must eventually be used to house processed (versus unprocessed) waste.

The percentages in Table 6 are the minimums, if more waste is recycled (and a higher fraction of the repository stores processed waste) then the repository can continue to accept waste past the year 2100.

Goal 5. Recover energy

Table 7 summarizes estimates of world uranium resources. Table 8 shows the estimated fraction of world U resources used by each Energy Future. For illustrative purposes, we used energy multiplication of 1 (no recycle), 1.3 (limited thermal recycle), and 75x (repeated fast recycle). Obtaining 1.3x for thermal recycle would require consuming most of the TRU generated (e.g. via inert matrix fuel). Obtaining 75x requires fast reactors to convert most of the U-238 in original ore to fissile isotopes and consuming them.

Table 7. World-Wide Potential Uranium Resources

Uranium Resource Category	Reference	Tonne U
Pessimistic, known recoverable resources		**3.1e6**
known recoverable resources of uranium	Herring2004	3.1e6
known conventional resources at <$80/kg-U (<$31/lb U308)	Herring2004	3.0e6
known conventional resources at <$30/lb-U3O8 (<$78 kg/U)	Steyn2003	3.1e6
Realistic, estimated conventional resources		**1.6e7**
Undiscovered conventional resources (total) + known conventional resource at < $130/kg-U ($50/lb-U3O8)	Herring2004	1.5e7
Estimated undiscovered+known conventional resources at <$50/lb-U3O8 ($130/kg-U)	Steyn2003	1.6e7
Optimistic, including unconventional resources		**4.2e9**
Uranium in sandstone deposits	Herring2004	1.8e8
Uranium in volcanic deposits	Herring2004	2.0e9
Uranium from seawater	Herring2004	4.2e9
Uranium in phosphate deposits	Herring2004	8.0e1

Table 8. Percent of World Uranium Resources Used by U.S. by 2100 (Preliminary Analysis)

	Nuclear Futures	Legislative Limit	Existing License Completion	Extended License Completion	Continuing Level Energy Generation	Continuing Market Share Generation	Growing Market Share Generation
	Cumulative discharged fuel in 2100 (MTiHM)	63,000	100,000	120,000	250,000	600,000	1,500,000
Fuel Management Approach	World resources (MT)	Percent of World U Resources Used by U.S. by 2100					
No recycle	Known (4.0e6)	16%	26%	31%	65%	160%	390%
	+Estimated conventional (1.6e7)	3%	4%	5%	10%	24%	60%
	+Estimated unconventional (4.2e9)	0.0%	0.0%	0.0%	0.0%	0.1%	0.3%
Limited thermal recycle (energy multiplication of 1.3x)	Known (4.0e6)	13%	20%	24%	50%	120%	300%
	+Estimated conventional (1.6e7)	2%	3%	4%	8%	18%	46%
	+Estimated unconventional (4.2e9)	0.0%	0.0%	0.0%	0.0%	0.1%	0.2%
Repeated fast recycle (energy multiplication of 75x)	Known (4.0e6)	0.2%	0.3%	0.4%	0.9%	2%	5%
	+Estimated conventional (1.6e7)	0.0%	0.1%	0.1%	0.1%	0.3%	0.8%
	+Estimated unconventional (4.2e9)	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%

Table 8 does not include the amount of uranium needed for a given nuclear power plant further into the future, only that which is actually consumed by 2100. Note also that for illustration purposes, we used 4.2e9 MT for estimated unconventional resources, i.e., the estimate for uranium from seawater. The table shows that such a resource makes uranium a non-issue. Phosphate or volcanic resources would produce similar or better results.

QUESTION III.
Should we separate uranium?
If we separate uranium, should we recycle it, store it, or dispose of it?

As baseline, assume we desire to separate U with sufficient purity so it would qualify for near-surface burial per U.S. regulation 10CFR61. This preserves maximum flexibility. If separated, do we store the uranium above ground or shallow below ground for easy accessibility. There is relatively low value in separating U without sufficient purity for 10CFR61. As separated U is a relatively low proliferation risk and relatively low radiotoxicity, there is relatively little benefit from storing it in a geologic repository.

Issues associated with question

Issues associated with the goals relevant to this question will now be discussed.(Goals 1-7 in Table 2.)

Goal 1. Consider readiness to proceed

The Nuclear Waste Policy Act (NWPA) would have to be changed. It currently indicates that the repository capacity is determined by initial tonnes of heavy metal; thus, even when uranium is removed, the original mass (including uranium) would be used in calculating compliance with the repository limit.

Goal 2. Maximize adaptability and robustness

If any SNF separation is done, there would seem to be little down-side to separating uranium (other than the sunk cost involved). There is always the option of mixing uranium back with the TRU if for some unanticipated reason this was desirable.

There are possible up-sides to separating uranium. Used uranium can be re-enriched and returned to thermal reactors. It is unclear whether this is advantageous versus using natural uranium ore, but having the uranium in separated form creates that possibility. For thermal reactors, using used uranium versus uranium ore would seem to be relatively more advantageous as (a) the price of uranium ore increases, (b) high efficiency isotope-specific separation techniques like laser isotope separation become available, and/or (c) the environmental hazards of uranium mining and milling are viewed as significant detriments for nuclear power. For fast reactors, of course, “used” uranium would be a natural feedstock; separating uranium now stockpiles the material for potential future use.

Goal 3. Improve waste management, especially geologic repository ramifications

Uranium separation does not impact repository capacity, as determined by short-term heat load, long-term heat load, or hypothetical dose if repository canisters fail. Uranium isotopes do not dominate any of those metrics. However, uranium may dominate the volume of SNF and thus, potentially, dominate the volume of waste. Separation of uranium then offers the potential to reduce the volume of material that must go into geologic repositories. If so, one would expect that the number of packages and the number of operations involving packages (welding, inspection, etc.) could be reduced accordingly. This is not automatic as heat load tends to be a dominant packaging design parameter. Further analysis of the potential advantages to packaging volume, mass, and number is needed.

Goal 4. Improve proliferation resistance and physical protection

The uranium content of SNF does not prevent it from being considered WU material; there would seem to be little or no proliferation resistance advantage from keeping uranium with the TRU material separated from SNF. Indeed, uranium separation reduces the volume involved, so that the remaining gamma emitters, neutron emitters, and heat generation isotopes would be more concentrated, tending to increase the intrinsic measures of proliferation resistance. There is a mild proliferation resistance penalty from volume reduction (less mass to be diverted); if this metric were deemed significant, then non-uranium filler material could be added to the TRU product stream so that uranium separation did not reduce the net volume and/or mass.

Goal 5. Recover energy

By design, fuel cycles with fast reactors are able to extract energy from “used” uranium, hence separating uranium from SNF would be advantageous. As noted above, there is relatively less motivation to use “used” uranium (versus natural ore) in thermal reactors. However, there are trends (uranium ore cost, environmental restrictions on mining/milling, etc.) that could make it advantageous to recover energy from “used” uranium in thermal reactors.

Goal 6. Ensure competitive economics (as part of the entire fuel cycle)

Definitive analyses need to compare the cost of geologic disposal (capital, packages, and packaging operations) with and without uranium in the material to be disposed. The resulting advantage (if any) should be compared to the cost of uranium separation.

Goal 7. Ensure safety (as part of the entire fuel cycle)

U ore mining, purification, and enrichment appear significant to overall fuel cycle safety. This would be a mild additional argument in favor of U separation.

QUESTION IV.

Which transuranic elements should be separated and transmuted?

Past work indicates that Pu and Am must be separated and recycled to achieve fundamental program goals of waste management and proliferation resistance. Np does not appear a major factor; it is mostly a "separation for convenience" case. On balance, it should probably be kept with Pu.

Cm is the key unknown. As transmutation calculations have shown, in thermal spectrum reactors, higher TRU will accumulate. Equilibrium values for Pu+Np+Am+Cm (with the CORAIL concept) would increase decay heat (3.4x), neutrons (1680x) and gamma (3.0x) relative to only Pu+Np+Am.[Salvatores2003][Salvatores2004] The Cm penalty for fast reactors is modest. We therefore suggest keeping the following Cm options open.

- Thermal with limited number of recycles – Cm and no-Cm are both options, it may be best to use oldest fuel first and accept the minor penalty of Cm/Cf accumulation for at least 1-2 cycles. How bad are those penalties?
- Thermal with extended recycle – no-Cm as baseline, Cm as alternative. We need to understand the implications of extended recycle with and without Cm.
- Fast recycle – Cm as baseline – we need to better understand how each TRU inventory is adjustable via conversion ratio.

To resolve the Cm issue, we need clarification regarding the difficulty of making such hot fuel, examination of NPP accident source impacts, calculations of how Cm accumulates in the first several cycles, additional clarifications of which TRU can be reasonably separated together and system assessments of trade-offs and timing, specifically impact of cooling time. For example, to the extent that the ratios cited above are dominated by Cm-244 (an oversimplification), the ratios are reduced to 1.0 with cooling times of 32 years, 194 years, 29 years respectively. Thus, if fuel fabrication is dominated by decay heat and gamma problems, recycling Cm after ~30 years may be sufficient, at which time neutron source (a proliferation benefit) would still be substantial. If we start recycling with the oldest SNF, some of the delay will have already occurred. Collins and Renier calculate various benefits from Cm decay.[Collins2004]

Issues associated with this question

Issues associated with the goals relevant to this question will now be discussed.(Goals 1-7 in Table 2.)

Goal 1. Consider readiness to proceed

To obtain credit for reducing material counted against the NWPA limit, the method of calculating the YMP capacity limit would have to change to preclude material that is recycled, for any of the options here.

U.S. industry, NRC, and DOE have addressed the issues with Pu recycle, most recently in the context of burning down weapons-grade Pu via weapons-grade mixed oxide fuel. There has not been comparable U.S. effort regarding issues associated with recycling Np, Am, and Cm. Thus, recycling of any TRU other than Pu would probably require a significant industrial and regulatory effort. As an example, NRC regulations associated with NPP accident source term would have to be re-examined. The NRC required a weapons-grade Pu mixed oxide fuel re-qualification effort; it is quite plausible that such an effort would be needed for each additional TRU addition.

Goal 2. Maximize adaptability and robustness

In a simplistic sense, the most adaptable situation would be separation of all four major TRU (Np, Pu, Am, and Cm) with later decisions on what to do with each. This, of course, would run afoul of the anti-proliferation policy of avoiding (and accumulating) separated Pu.

It also runs afoul of decay by various TRU isotopes that change the elemental composition. Consider two examples. (1) Pu-241 (14.35 years) decays to Am-241 (432 years) to Np-237; this is unfortunate as Np-237 is a key long-term repository dose issue and Am-241 is a key long-term heat source. Other things being equal, one would like to recycle and burn Pu-241 before decay. And, if one is planning to use a Pu stream in some fashion without Am, then one must control the time between Pu separation and Pu use. (2) Cm-244 (18.11 years) decays to Pu-244 (8.2e7 years), which is good in thermal reactors because if Cm-244 is recycled to thermal reactors (before decay), successive (n,gamma) reactions will produce very high gamma and neutron emitting isotopes.

Another consideration is that SNF coming into a fuel processing and fabrication plant will have varying compositions. Recycled fuel forms going out to NPPs may then have varying compositions. Are the NPPs tolerant of slightly different amounts of Pu, Np, Am, Cm, etc. in recycled fuel due to safety restrictions, fuel handling equipment, planned burn up, etc? If not, keeping recycled fuel forms with constant composition would require a "wine cellar" of fuel component elements.

The basic point is that separation of individual TRU may look more adaptable, but the actual value (or disadvantage) requires a deeper examination. In contrast, fuel form, fuel fabrication, and an NPP safety case that could handle any (reasonable) mixture of Pu-Np-Am-Cm would certainly be more robust against minor composition changes.

Goal 3. Improve waste management, especially geologic repository ramifications

As discussed above, to reduce long-term repository heat load, both Pu and Am must be transmuted.

Goal 4. Improve proliferation resistance and physical protection

Separation and transmutation to reduce the inventory of Pu are the first and easiest steps; it is easy to transmute Pu-239 as it is fissile in thermal reactors. A more complete approach to WU inventory reduction would be transmuting Pu, Np, and Am – each appears to be WU. Other than adjusting for Significant Quantities of TRU, there is no known difference in WU classification among Pu, Np, Am, Pu+Np, Pu+Am, Np+Am. Since no AFCI option has >80% Pu-238, any of these mixtures are WU. Thus, from a high-level perspective, if we are going to argue the benefits of AFCI reducing Pu, it would be a more robust case to show that we are reducing Pu, Np, and Am.

The situation with Cm is less clear; its high neutron emission rate may avoid the WU label. Even better (from the proliferation resistance standpoint) would be how much "credit" one would get for including

Cm with Pu+Np+Am. It is probably hopeless to avoid the WU label, as even U-238-rich U mixed with Pu does not avoid the WU label.

Some AFCI options – those with repeated recycle - provide the means for stabilizing the TRU inventory. In contrast, once-through "cycles" and limited recycle approaches cannot. Along these lines, we need to understand how parameters such as conversion ratio impact the stabilization levels.

Goal 5. Recover energy

Pu must be recycled to recover energy content.

Goal 6. Ensure competitive economics (as part of the entire fuel cycle)

Much more needs to be done on economics.

Goal 7. Ensure safety (as part of the entire fuel cycle)

Perhaps the dominant issue is potential impact to NPP safety – either reactor physics or accident source term. It is already clear within the program that reactor physics issues such as void coefficient have to be considered carefully. The accident source term issue appears to have received less attention.

Lyman has attacked the concept of recycling Pu in mixed oxide fuel in light-water reactors on the grounds that this would increase the accident source term.[Lyman2000] Indeed, by design, the inventory of TRU in LWR with recycled TRU would increase. In contrast, most analysts do not find that TRU inventories dominate NPP accident source terms (for example Table 9), which are instead dominated by isotopes of more volatile elements, such as the noble gases.

Table 9. Grouping of Elements for NPP Source Term [NUREG-1465]

Group	Elements in Group
Noble Gases	Xe, Kr
Halogens	**I**, Br
Alkali Metals	**Cs**, Rb
Tellurium Group	Te, Sb, Se
Barium, Strontium	Ba, **Sr**
Noble Metals	Ru, Rh, Pd, Mo, **Tc**, Co
Lanthanides	La, Zr, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am
Cerium Group	Ce, Pu, Np

QUESTION V.
Of TRU separated, which are transmuted together or separately?

Here, we assume that Pu and Am are recycled, we are not sure about Cm (at least for thermal reactors), and Np is "separate and transmute" if convenient.

- For aqueous processing, each of the following has problems and therefore down-selection would be premature.
    - Pu+Np (Am+Cm separate) – This apparently is the natural chemical separation approach for UREX+. But, repository benefits are low unless Am is recycled. So, this approach tends to lead to either (a) subsequently separating the Am from the Am+Cm to recycle it or (b) storing the Am+Cm until Cm-244 decays (18 years) or until at least one fast reactor/ADS becomes available.

  - Pu+Np+Am (Cm separate) – This may be difficult to do chemically.
  - Pu+Np+Am+Cm – This would seem possible to do, but has penalties of higher actinide buildup in thermal reactors.
- For pyroprocessing and fast reactors, combining Pu+Np+Am+Cm is natural.

A possible "policy" would be to have recycled fuel as "hot" as we can safely and economically handle it, thereby maximizing proliferation resistance. Resolving this requires more work on fuel fabrication practicalities, NPP source term, and proliferation resistance – in parallel to the implications to separation technologies.

Issues associated with this question

Issues associated with the goals relevant to this question will now be discussed.(Goals 1, 2, 4, 6, 7 in Table 2.)

Goal 1. Consider readiness to proceed

Naturally, Pu separation (PUREX) and Pu-only recycle fuel forms are more ready to proceed than are separation processes and fuel forms for Pu+Np, Pu+Np+Am, or Pu+Np+Am+Cm. See a companion paper at this conference, "On-Going Comparison of Advanced Fuel Cycle Options," information on relative technology readiness levels.

Goal 2. Maximize adaptability and robustness

Robustness may prove to be a deciding factor because fuel composition can change:

- Before separation. Pu241 will decay to Am241.
- During reactor irradiation. Approaches that depend on very precise compositions would seem to have practical difficulties.

Goal 4. Improve proliferation resistance and physical protection

Regarding intrinsic proliferation resistance and minimizing WU inventories, keeping Pu+Np+Am together would seem to have modest advantages versus Pu or Pu+Np. The Am tends to increase decay heat, neutron source, and especially gamma source during fabrication. The major breakpoint would come with inclusion of Cm.

Table 10. Intrinsic Proliferation Resistance Parameters as Function of TRU Included for Equilibrium Thermal Recycle

Equilibrium state parameters for CORAIL, adapted from [Salavatores2004]		Pu (taken as reference)	Pu+Np	Pu+Np+Am	Pu+Np+Am+Cm
Decay heat	Fabrication	1	1.9	5.9	20
	After 5 years cooling	1	1.1	2.2	3
Neutron source	Fabrication	1	1.5	3.8	6375
	After 5 years cooling	1	0.94	2.4	395
Gamma source	Fabrication	1	1.9	13.7	41
	After 5 years cooling	1	0.99	0.95	0.95

Goal 6. Ensure competitive economics (as part of the entire fuel cycle)

More work needs to be done.

Goal 7. Ensure safety (as part of the entire fuel cycle)

More work needs to be done on these topics.
- Number of operations – the more processing steps – the more things that can go wrong.
- Relative transportation risk.
- "Hot" fuel fabrication – if a major difficulty, this would argue for keeping as many TRU separate as proliferation concerns allow.
- Impact to NPP source term.

QUESTION VI.
Should we separate and/or transmute Cs and Sr isotopes that dominate near-term repository heating?

Discard the option to transmute Cs and Sr, i.e., keep them out of products to be transmuted. As baseline, separate Cs and Sr for long-term interim storage. Long-term storage could be near-surface or in a geologic repository; in either case after a few hundred years, the material should qualify for "near-surface burial" per 10CFR61; our preliminary assessment suggests that Cs-135 does not appear to be a barrier to the 10CFR61 strategy, but this needs to be confirmed.

However, if a given separation technology has difficulty in separating Cs and Sr and there is little chemical and handling benefit from subsequent separation steps to retaining Cs and Sr in the stream headed for HLW, non-separation of Cs and Sr could be kept as an alternative. In this case, Cs and Sr would be disposed with residual HLW. Thus, if there are separation technologies with difficulty in separating Cs and Sr, they may remain viable candidates.

Issues associated with this question

Issues associated with the goals relevant to this question will now be discussed.(Goals 1-4, 6, 7 in Table 2.)

Goal 1. Consider readiness to proceed

Any disposition path other than direct geologic disposal would require a change in regulations. At discharge, Cs and Sr do not qualify for near-surface burial per 10CFR61. Long-term storage for the 100-300 years necessary to decay to 10CFR61 levels is not an option already identified in regulations. It is likely that a change in NRC regulations would be required. And, long-term storage probably would require an EPA RCRA permit.

Goal 2. Maximize adaptability and robustness

Separating Cs and Sr preserves the flexibility of doing subsequent things with them later. Removing the material from other product streams makes them less difficult to handle and less complicated chemically.

Goal 3. Improve waste management, especially geologic repository ramifications

Salvatores [Salvatores1998] shows that transmutation of Cs and Sr is slow compared to their decay rate and thus transmutation is ineffective. Cs and Sr are 0.2% to 0.3% of SNF mass; thus packaging volume would not be a major factor.

Wigeland [Wigeland2004a, Wigeland2004b] shows that they dominate the repository heat load for the first several decades. However, the advantage for separation of Cs and Sr is not dramatic for limited thermal recycle because of heat from residual TRU. However, for thermal+fast or fast recycle, where TRU are more effectively reduced, separation of Cs and Sr would allow an increase of repository capacity from 5x to 50x. (See Question II for more discussion on repository capacity.)

Thus, the question is whether it is easier to remove heat from Cs and Sr stored (a) separately near-surface, (b) separately elsewhere in the repository, or (c) with HLW. The YMP baseline is forced ventilation for 50 years to remove Cs and Sr (and other short-lived decay heat). The recent Spent Fuel Treatment Facility (SFTF) design team showed that passive heat rejection to the atmosphere from near-surface storage (option "a") was practical. We know of no calculations that show what happens in option "b"; presumably this would be practical as the ventilation rate for the entire repository would not increase relative to the YMP baseline; however, the ventilation rate in the specific Cs-Sr drift tunnels would presumably increase. Still, the rock temperature limitation could perhaps be relaxed for those tunnels as they would not be expected to provide protection for millennia. Wigeland's calculations show that forced ventilation of Cs and Sr stored with HLW (option "c") is practical. The heat flux in these tunnels is intermediate between the baseline and option "b". Thus, there is reason to believe that any of these schemes can be made to work. It is less obvious which is best.

The issue has arisen as to whether Cs-135 would prevent Cs-Sr from qualifying under 10CFR61. Cs-135 is not included in 10CFR61. We are aware of two calculations that extend 10CFR61 methodology to Cs-135.

- NUREG-0945 (1982) and NUREG/CR-4370 (1986). As referenced in D. C. Kocher and A. G. Croff, A Proposed Classification System for High-Level and Other Radioactive Wastes, ORNL/TM-10289, June 1987; Table ES-2 gives a class-C equivalent as 800 Ci/m^3 for Cs-135 versus 4600 Ci/m^3 for Cs-137.
- S. Fetter et al, Fusion Energy and Design, 2(6), April 1988. His calculations indicate that Cs-135 would not be an issue because even fully dense Cs-135 would not exceed limits.

At 34 GW-day/MT burnup, J. S. Herring (personal communication) provided values of 0.3 Ci Cs-135/tonne-fuel and 2800 g-total-Cs/tonne-fuel, which gives 1.07e-4 Ci Cs-135/g-total-Cs. The SFTF team's documents give the density of the waste form as 4.65 g/cc (Cs_2O) or 2.90 g/cc (Cs-Al-Si). Taking the higher density as worse case, we obtain 500 Ci of Cs-135/m^3-waste form. This is close, but **below** the referenced limit of 800 Ci/m^3. If one uses Fetter's results, then the situation is yet better.

These numbers will change as a function of burn up (higher Cs-135 content), decay (Cs-137 decaying to Ba), waste form, etc. For example, at higher burn up, the total Cs mass goes to 3670 g-Cs/tonne (1430 Cs-133, 202 Cs-134, 418 Cs-135, 0.9 Cs-136, 1620 Cs-137). As Cs-137 decays to Ba, there is a significant elemental composition change. Assuming, however, that the permanent waste form is made at 5-year fuel age at SFTF, then the mass number above (2800 g-total-Cs/tonne) are still correct even though some of the Cs will later change to Ba. (Is the proposed Cs waste form stable to such an elemental composition change?)

Tentative conclusion: Cs-135 does **not** prohibit Cs waste form being stored as class C waste, but it is close and should be checked further. Subject to Cs-135, we thus concur that after several halflives of decay, Cs/Sr would qualify for near-surface disposal per 10CFR61. However, at the time that Cs/Sr is separated and solidified, it would logically be considered a waste that (at that time) would not qualify for near-surface burial. The regulatory authority for storing it for several hundred years prior to permanent disposal is not clear; not obvious that either 10CFR70 or RCRA could provide a basis for doing so. We have a grey area in the regulations on that score that should be highlighted for future analysis and resolution.

Goal 4. Improve proliferation resistance and physical protection

Although short-lived fission products are not WU, they are certainly concerns for radiological dispersion devices. So much so that they are even considered as "templates" for preparing public information regarding radiological dispersion devices.[ANS2003] Thus, by separating Cs and Sr, we would be creating obvious targets for theft. Such sources would obviously have to be carefully controlled. Nonetheless, this factor does not appear a major argument against separation as the material from which Cs and Sr would be separated from (TRU stream) must itself be carefully protected.

Goal 6. Ensure competitive economics (as part of the entire fuel cycle)

Cs and Sr are about 0.2-0.3% of SNF mass.[ANL] At discharge from LWR, they are about 7% of negative reactivity[Christian1999] and thus separation helps performance of recycled fuel. SFTF designers saw advantages to removing Cs and Sr early in the process flow because that simplified the chemistry and shielding for subsequent steps. For UREX+, therefore, it seems a reasonable assumption that removal of Cs and Sr is desirable. If there are separate schemes without obvious Cs and Sr removal advantages (from the chemical process point of view), the following should be estimated.

- Change in processing cost (with, without Cs+Sr separation)
- Change in subsequent recycle fuel fabrication cost
- Interim storage cost (if separate Cs+Sr)
- Subsequent waste form fabrication cost (if separate Cs+Sr)
- LLW disposal cost (if separate Cs+Sr)
- Change in HLW disposal cost (with, without Cs+Sr separation) – capital cost for packages, packaging/handling cost, ventilation costs, etc.

Goal 7. Ensure safety (as part of the entire fuel cycle)

Cs and Sr isotopes are definitely important to reactor accident safety source term, most notably Cs-137 and Sr-90. Unfortunately, key Cs and Sr isotopes are sufficiently long-lived to be waste management concerns ("short-term" heat load to geologic repository) and short-lived to be major contributors to the accident source term. Such key isotopes do not reach equilibrium during a single pass through the reactor, indeed the inventories continue to increase with fluence. Thus, recycling Cs and Sr with the TRU stream would progressively increase the accident source term, which would be undesirable. If the elemental content of Cs and Sr in oxide fuel is substantially increased, there may be issues regarding the chemical behavior of such fuels in accident conditions.

QUESTION VII.

Should we separate and/or transmute very long-lived Tc and I isotopes?

Note: obtaining >80% separation of I "for free" in some separation technologies is different than deliberately separating it with an explicit separation specification. Thus, we only address deliberate, specified separation of these elements rather than "separation for convenience" or separation for "free."

We suggest keeping three options open.

- Separate and transmute Tc and I – whether with TRU or as separate targets.
- Separate Tc and I to put into a more durable waste form than the bulk of HLW, do not transmute.
- Neither separate nor transmute Tc and I, direct disposal.

Transmutation of I-129 and Tc-99 is slow but of course faster than waiting for decay. If "sustainability" is particularly important for nuclear power or long-term radiotoxicity is critical to NWPA changes and the cost is modest, separation and subsequent transmutation of Tc and I would be logical. In the absence of further clarification of these factors, it would be premature to dismiss Tc/I transmutation. So, there may be significant long-term benefit from recycling with only minor penalties. This is more likely true for Tc than for volatile Iodine.

We do not expect recycle of Tc-99 nor I-129 to impact accident source terms nor gamma fields or heat load during fuel fabrication, as they are very long-lived. Iodine isotopes important to accident source terms have sufficiently short lives (days) that they decay before material would be recycled.

Issues associated with this question

Issues associated with the goals relevant to this question will now be discussed.(Goals 1-3, 6 in Table 2.)

Goal 1. Consider readiness to proceed

Each time a separate waste stream is created, there must be a waste form created and an applicable regulation to handle it. Tc and I have only one long-term disposal option – geologic disposal. Of course, if Tc and I are to be transmuted, there must be a "fuel" form – either incorporation into the TRU fuel or separate targets. Neither have been adequately explored.

Goal 2. Maximize adaptability and robustness

If Tc and I are separated (whether for transmutation or not), there is more flexibility to decide later what to do with them.

Goal 3. Improve waste management, especially geologic repository ramifications

Tc-99 and I-129 are not important to heat generation, either short-term or long-term. Nor do they significant impact mass or volume of HLW. Thus, to the extent that heat, mass, or volume dominate the cost, safety, and environmental impact of waste management, then Tc-99 and I-129 are irrelevant.

Separation and transmutation of Tc-99 and I-129 may, however, be significant to hypothetical dose calculations or radiotoxicity. *Radiotoxicity* refers to the hazard of disposed material, with no adjustment or credit for transportation through groundwater or holdup by waste packages. *Dose* calculations include the entire assessment of waste material performance, waste package performance, transport through the unsaturated vadose zone around the repository, transport through the saturated groundwater zone (aquifer), and eventually water consumption by humans. In current US repository dose calculations (versus the 10,000-year dose performance standard), neither Tc-99 nor I-129 are important. Even at longer times, dose from these isotopes does not appear to exceed the current 15 mrem/year dose limit. So, it would appear that separation and transmutation (destruction) of these isotopes is not required for dose limits.

Indeed, Yang et al have concluded that, "The current Yucca Mountain release evaluations do not indicate a compelling need to transmute Tc-99 and I-129 because the resulting dose rates fall well below current regulatory limits. However, elimination of [this long lived fission product (LLFP)] inventory could allow significant relaxation of the waste form and container performance criteria, with associated economic benefits. Therefore, some development of either specialized waste form or transmutation target for the LLFP is prudent, especially considering the potential accumulation of large LLFP inventory with sustained use of nuclear energy into the future."[Yang2004]

If many reactor-years worth of SNF are going to be processed and recycled, the relative contribution of Tc-99 and I-129 to dose/radiotoxicity of residual HLW will increase. They could become sufficiently important as to warrant their separation and transmutation. Work is proceeding to clarify and quantify this possibility.

Salvatores [Salvatores1998] calculates slow transmutation rates, slower in fast reactors (per neutron) than in thermal as the dominant destruction mode is (n,gamma). However, as fast reactors are more neutron rich, he concludes that fast reactors may be a superior transmutation platform. So, if waste management plans and metrics evolve such that Tc-99/I-129 dose or radiotoxicity are significant contributors to HLW limitations, transmutation remains a feasible option.

Goal 4. Improve proliferation resistance and physical protection

Long-lived isotopes of Tc and I are obviously not WU, neither are they obvious concerns for radiological dispersion devices. Perhaps short-lived iodine isotopes would be an issue, but they are not the subject of this decision.

Goal 6. Ensure competitive economics (as part of the entire fuel cycle)

Long-lived Tc and I isotopes are about 0.1% of the mass of SNF. At discharge, Tc and I are about 5% of the negative reactivity in LWR discharged fuel.[Christian1999] Both the mass fraction and reactivity worth fraction are lower than Cs+Sr. To be sure of the trade-off (separate/transmute or not), we would need to know these factors.

- Cost of separation
- Cost of waste form fabrication (if separated)
- Cost of disposal (if separated)
- Cost impact on subsequent fuel fabrication (if not separated)
- Cost impact on reactor operation (if not separated)
- Cost impact on subsequent used fuel disposal (if not separate)

Goal 7. Ensure safety (as part of the entire fuel cycle)

We do not expect recycle of Tc-99 nor I-129 to significantly impact accident source terms or gamma fields during fuel fabrication, as they are very long-lived – however this must be checked. Iodine and Tc isotopes important to accident source terms, e.g., Tc-99 and I-131, have sufficiently short lives (days) that they decay before material would be recycled. Notably, Lyman in attacking Pu recycling from the NPP safety perspective, did not even bother to calculate Tc-99 and I-129.[Lyman2000]

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