Report of
ADVANCED NUCLEAR TRANSFORMATION TECHNOLOGY SUBCOMMITTEE
of the
NUCLEAR ENERGY RESEARCH ADVISORY COMMITTEE

26-27 February 2004

BURTON RICHTER, CHAIR
DARLEANE C. HOFFMAN
SEKAZI K. MTINGWA
RONALD P. OMBERG
SYLVIE PILLON
JOY L. REMPE

I. SUMMARY, FINDINGS AND RECOMMENDATIONS

The ANTT Subcommittee of NERAC met February 26th and 27th (S. Pillon absent) to begin a review of the potential role of transmutation technologies in increasing the capacity of the geological repository for spent reactor fuel. This work is in support of the recommendation required from the Secretary of Energy later in this decade on the need for a second repository. Since repository issues were under discussion, representatives of the Office of Civilian Radioactive Waste Management (RW) were in attendance.

The focus of the transmutation program up to now has been on the potential to reduce the required time of isolation of spent fuel from the biosphere from hundreds of thousands of years to on the order of a thousand years. What has evolved from the program is the potential of a system involving a combination of spent-fuel partitioning, recycling of actinides and other long-lived radioactive components in thermal-spectrum reactors, followed finally by a treatment in a fast-spectrum facility (one fast spectrum facility for every five to ten thermal reactors). If such a system were to be developed and deployed not only would the required isolation time be reduced, but the capacity of a geological repository such as Yucca Mountain would be increased by a factor of roughly 50, more than enough to handle all the spent fuels generated in this century from any conceivable nuclear energy deployment scenario. The increase in capacity comes from the destruction of the actinides that generate the heat which limits the capacity of the repository (see Section II).

If the issue to be addressed is only the capacity of the repository, the allowable amount of actinides and other long-lived elements in the material going to the repository can be significantly increased above that allowed in the reduced isolation time scenario. The Committee therefore asked that the laboratories review transmutation scenarios looking at the potential only for an increase in capacity. Further, we asked that thermal-spectrum-only systems be studied as well as a combination of thermal- and fast-spectrum systems. For the next several decades, only thermal-spectrum light-water reactors (LWRs) will be available in the U.S. for any such application. This report is mainly devoted to reviewing the results of that study. It should be noted that the analysis is preliminary, as it was only requested about three months prior to the Committee review and, therefore, the scenarios are by no means optimized.

The question posed was what could be done with some number of cycles of spent-fuel partitioning and transmutation where after the last cycle all of the remainder was to be sent to the repository. This approach is described in more detail in Section II. The results are summarized in Table 1.

Table 1. Repository Requirements in the U.S. by 2100*

Nuclear Futures	Legal Limit	Extended License for Current Reactors	Continued Constant Energy Generation	Constant Market Share	Growing Market Share
Total Discharged Fuel by 2100, MTHM	63,000	120,000	240,000	600,000	1,300,000
Repositories needed with current approach	1	2	4	9	21
Repository with expanded capacity		1	2	5	11
With thermal recycle only			1	2	5
With thermal and fast					1

*Adapted from Reference 2.

Five different nuclear scenarios were assumed giving amounts of spent-reactor fuel [in units of metric tons of heavy metal (MTHM)] ranging over a factor of twenty. At the lower end is the presently legislated limit on Yucca Mountain capacity. The next case is the result of extending the license of all currently operating nuclear reactors. The third case assumes that nuclear-power generation continues at its present level through the rest of the 21st century. The last two cases are two different scenarios for expanding nuclear energy (the assumptions of the recent MIT study lie between these).

The results show that if the currently legislated limit were applied to all future repositories, up to 20 such repositories might be needed by the year 2100.

The Office of Civilian Radioactive Waste Management (RW) has said that if the legislated limit on Yucca Mountain were removed, its particular geology would allow somewhat more than a doubling of the amount of spent fuel that could be stored therein and, in this case, the number of similar repositories required would range up to eleven.

Multiple recycling in thermal-spectrum reactors results in a further factor of two increase in capacity in this preliminary analysis. Up to five repositories would be required, depending on the scenario, and a decision on the second repository could clearly be deferred for quite some time.

Finally, as indicated earlier, with a combination of thermal- and fast-spectrum reactors only one repository would be required and that one could handle even more spent fuel than in the most aggressive nuclear scenario.

Scenarios have been studied using fuels containing plutonium (Pu), americium (Am) and neptunium (Np) in the LWR recycle schemes. Options using inert matrix fuels (IMF) and mixed oxide fuels (MOX) have been evaluated. It appears that a roughly factor of two increase in capacity can be achieved with either. While there is less experience with IMF, the potential gain in repository capacity is achieved with fewer recycles. The effect of storage time of spent-reactor fuel between its removal from the reactor and its first recycle is currently being evaluated. The IMF option may have greater proliferation resistance than MOX, and this is under study.

There are indications that it **may** be possible to bring an LWR recycle scheme into equilibrium where the long-lived component generated from fresh fuel in the reactor is about the same as that consumed from the recycled fuel in the same reactor. If practical, this would allow a kind of continuous recycle and a considerable increase in capacity beyond the factor of two indicated so far. It is too early to say if this scheme will work.

Research and development on separations technology continues to advance (Sections III). RW is interested, even in the absence of transmutation, because it may be possible to develop more stable and less costly storage forms for the repository by packaging different components of the spent fuels separately. While NE is short of funds for moving separation R&D to a larger scale, RW interest and support may make it possible to advance some parts of the technology development. In particular, an experiment using the French reprocessing plant (Sections III and V) is under active discussion. Using the French facility may get the answers RW requires on their required time scale, but it will, however, not give the benefit of the experience gained at this scale to U.S. scientists and engineers, or demonstrate all of the steps required for transmutation. (Also, there seems to be some uncertainty in the French schedule.)

The transmutation schemes under study raise important issues in fuel development (Section IV). There is little experience with IMF and none with MOX containing Am and Np along with Pu and U. The main issue seems to be fabrication of fuels containing Am. Information today is insufficient to help in making the choice between IMF and MOX.

The availability of a fast-spectrum system would simplify the entire process. Even at the present stage of R&D, it is highly likely that repository capacity could be greatly increased. It is also probable that the required isolation time of spent fuel could be greatly decreased. If there is to be a great expansion of nuclear energy use worldwide, fast-spectrum systems that could burn transuranics as well as U-235 would be very beneficial. However, if the thermal-spectrum multi-recycle schemes work out as they may, repository capacity will be adequate for many decades, even without a fast-spectrum system.

There are many international collaborations ongoing (Section V). Of particular relevance for this report is the work on IMF and the possible tests of the UREX+ system using the COGEMA facility in France mentioned earlier.

There is broad interest in transmutation. All countries interested in nuclear energy must face the issue of treatment and disposal of spent-reactor fuel. There are opportunities for still broader international collaborations of great benefit to the U.S. program.

Findings on Repository Capacity

1. Without alteration of the current legislated limit on the capacity of the Yucca Mountain repository, it cannot hold all of the spent fuel that will be produced by the present fleet of nuclear power reactors.
2. If the legislated limit is removed, all of the spent fuel from currently operating reactors can be stored, including that produced by life extensions. However,

the repository cannot store all the fuel produced through this century with a continuation of power production at current levels.
3. It is highly likely that transmutation in LWRs alone can increase the capacity of a repository about two fold.
4. It is possible that continuous recycling in LWRs can increase capacity by a larger factor.
5. It is highly likely that transmutation in a combination of LWRs and fast-spectrum reactors can increase repository capacity by a large amount and decrease the required isolation time from the biosphere by a large amount.

Recommendations on Repository Capacity
1. Continue the study of LWR recycling retaining for now both the IMF and MOX options.
2. Determine the feasibility of continuous recycling under realistic conditions.
3. Potential repository benefits indicate the priority of a fast-spectrum system in GEN-IV should be high.

Findings on Partitioning
The COGEMA experiment now under discussion may answer some of RWs questions in a timely fashion. However, U.S. technical personnel will not gain desired experience, and the full AFCI process will not be demonstrated.

Recommendation on Partitioning
Review the possibility of doing a cost-effective equivalent experiment in the U.S.

Findings on Program Issues
The AFCI program is under funded. The decrease in funding proposed for FY2005 only makes worse the “options overload” that we noted in our last report.

Recommendation on Program Issues
Nuclear Energy needs to better match its road map to its resources.

II. CAPACITY OF THE YUCCA MOUNTAIN REPOSITORY

II-1. Introduction

The capacity of Yucca Mountain is controlled by several limits -- legislated and technical. If held to its legislated limit, the capacity of Yucca Mountain is insufficient for the fuel that will be generated from existing commercial U.S. nuclear power plants. However, if one considers only technical limits, calculations

suggest that there is adequate capacity to accommodate the fuel from existing plants (even with power upgrades and life extension). If separation and reprocessing schemes are implemented, calculations suggest that there is sufficient capacity to accommodate existing plants and a range of currently-proposed U.S. nuclear expansion scenarios.

II-2. Legislative Limit

The Nuclear Waste Policy Act (NWPA) of 1982 (Ref. 1) limits the amount of spent nuclear fuel and high-level radioactive waste that can be emplaced in the first U.S. geologic repository to 70,000 MTHM for its first phase of operation (until a second repository is in operation). As stated in Ref. 2, the materials that may be disposed at Yucca Mountain under current restrictions include about 63,000 MTHM of commercial spent nuclear fuel; about 2,333 MTHM of DOE spent nuclear fuel; and about 4,667 MTHM of DOE high-level radioactive waste.

To put this limit in perspective, consider the Spent Nuclear Fuel (SNF) from commercial nuclear power plants in the US. Reference 3 indicates that as of December 31, 1998, there is nearly 40,000 MTHM from SNF in storage pools at US nuclear power plants. If existing plants continue operating until the end of their existing licenses, it is estimated that SNF from commercial nuclear power plants will amount to nearly 90,000 MTHM. (Ref. 4) With power upgrades and life extension for all existing plants, it is estimated that SNF from commercial plants will result in nearly 120,000 MTHM. It is clear that the legislated capacity limit for Yucca Mountain is insufficient to contain all of this SNF.

II-3. Technical Limits

For the Yucca Mountain repository, compliance with US Nuclear Regulatory Commission (U.S. NRC) requirements leads to specifications for waste package materials and limits for the repository's loading and operating conditions. At Yucca Mountain, these considerations set temperature limits for various parts of the repository. These temperature limits are met by a combination of specifications (e.g., the maximum decay heat of each waste package at the time of placement, the maximum average linear heat rate for waste packages placed in a repository drift tunnel, the duration and rate of forced ventilation in the tunnels, the spacing between emplacement drifts in the tunnels, etc.).

For the current high-temperature operating mode (HTOM) of the Yucca Mountain repository, limits of interest include:

- the rock temperature midway between the emplacement drifts must remain below 96ºC to allow continuous groundwater drainage, and present buildup above the drifts.
- the rock temperature at tunnel walls must remain below 200ºC to prevent alteration of the mountain rock crystalline structure.

In order to meet the above limits, the current Yucca Mountain design is to have the emplacement drifts 81 m apart with average drift linear heat loads of about 1.45 kW/m at the time of waste placement (assuming an average age of about 25 years for spent fuel, this corresponds to a drift spent fuel average linear loading of 1.1 MTHM/m) and to have the repository actively cooled by forced ventilation for at least 50 years.

To evaluate temperature limits, it is useful to consider the decay heat generated by a typical PWR fuel element (assumed to have a 50 GWd/MTHM burn-up) and the reference case used for Yucca Mountain evaluations (defined in Ref. 2). As shown in Figure 1 (from Ref. 5), decay heat from this typical PWR fuel element drops rapidly after the first 200 years. For the first 60 years, decay heat is primarily generated by fission products such as barium and yttrium that are decay products of cesium and strontium. After 60 years, the decay heat is primarily associated with actinide elements such as plutonium and americium. Beyond 200 years, decay heat is dominated by plutonium and americium and in particular, Am-241, which is due to decay of Pu-241. Other contributors are Pu-238, Pu-239, and Pu-240.

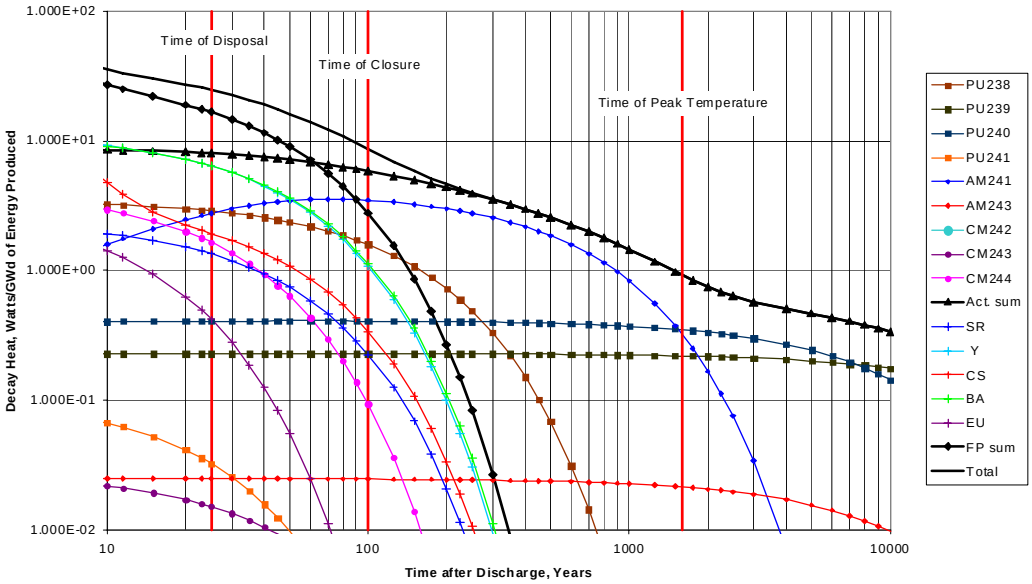

Figure 1. Decay heat generated by PWR fuel irradiated to 50 GWd/MTHM (Ref. 5).

Figure 2 shows transient temperatures predicted for the Yucca Mountain reference case (Ref. 5). In this case, ventilation is stopped 75 years after waste placement. For an average drift loading of 1.1 MTHM/m and the decay heat shown in Figure 1, the temperature midway between adjacent drifts remains just below its limit of 96°C. Drift wall temperatures remain below 140°C, which ensures that mountain rock temperatures remain below their 200°C limit. Hence, calculations suggest that the controlling limit is associated with the temperature between adjacent drifts, and peak temperatures occur at between 1000-2000 years after waste placement. Because ventilation was stopped after the first 75 years, elements having the highest integrated decay heat from the time after ventilation ceases have the most impact on the temperatures midway between adjacent drifts.

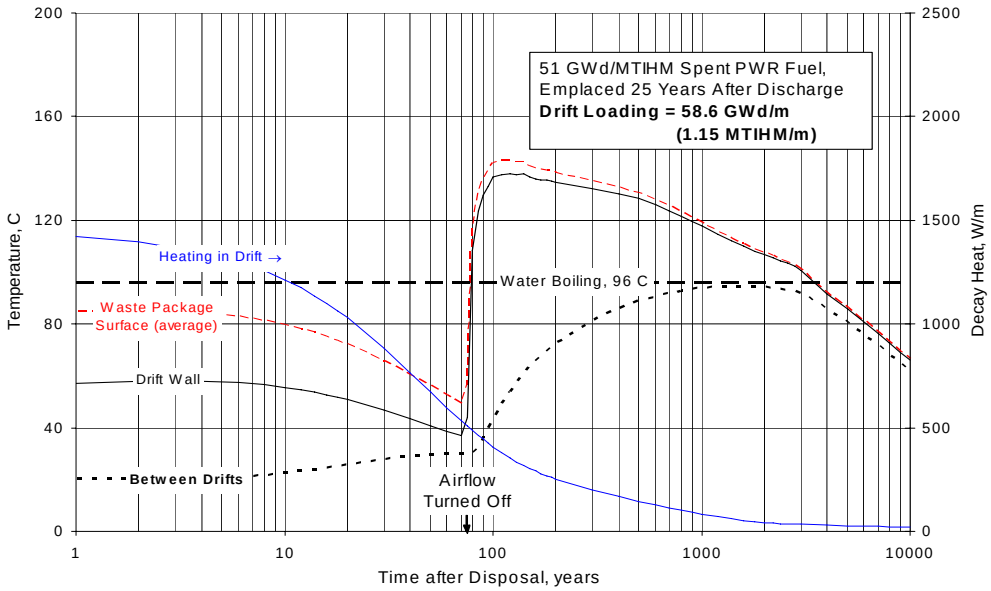

Figure 2. Transient thermal response of a repository at Yucca Mountain for reference loading conditions of SNF and 75 years of forced ventilation (Ref. 5).

Reference 5 evaluates potential increases in repository capacity by separating isotopes that are dominant contributors to decay heat. Cesium and strontium are stored separately. Removal of all plutonium and americium could increase the drift loading of the repository by at least a factor of four. If neptunium and curium were also removed, the drift loading in the repository could be increased by a factor of 40. However, a more thorough evaluation of actual separation and reprocessing schemes is required in order to assess the feasibility of such increases.

II-4. Potential Benefit of AFCI Separation and Reprocessing Scheme

In response to a request from this subcommittee, new calculations were completed to compare the benefit of various separation and recycling schemes being investigated in the AFCI program. These calculations used a set of simplifying assumptions to give a rough estimate of potential benefits. An optimization has not yet been attempted. Key assumptions and guidelines for these calculations include:

- Calculations considered PWR fuel that had been removed from the reactor for five years
- Plutonium and americium are separated from the spent PWR fuel with an efficiency of 99.9%. Neptunium is also separated from the spent PWR for non-proliferation and radionuclide inventory concerns. Plutonium, americium, and neptunium are then recycled for further irradiation in an LWR.
- The number of recyclings of plutonium, americium, and neptunium is treated as a parameter. After the last recycling, all materials are sent to the repository, including both processing waste and the spent fuel assemblies from the last irradiation.
- Curium is not recycled, but is sent directly to the repository in the process waste stream.
- Cesium and strontium are separated from the processing waste, and are stored separately, either in dry storage external to the repository (a policy change) or in a separate area of the repository. Because decay heat from these elements only persists for about 200-300 years, they could be placed in the repository after this period without impacting the repository's thermal limits.
- The assemblies fabricated using recycled material are, in some cases, assumed to be irradiated in reactor cores of identical assemblies, e.g., homogeneous reactor cores. In some cases, this assumption leads to impractical or non-feasible reactor cores, but this does allow parametric studies.
- Assemblies fabricated using recycled materials may be substituted for standard PWR assemblies and produce the same integrated energy per assembly.

The following three LWR recycling schemes were evaluated:

- Mixed Oxide Fuel (MOX) – In this approach, the separated plutonium, americium, and neptunium are used to fabricate new fuel assemblies, in a fuel matrix of uranium, (all elements are present as oxides).
- CORAIL-PNA – This concept uses heterogeneous assemblies with some of the fuel pins (approximately 1/3rd) fabricated from separated plutonium, americium, and neptunium and the remainder of the fuel pins (about 2/3rd) fabricated from new enriched uranium (all elements are present as oxides).
- Inert Matrix Fuel (IMF) – This approach is similar to MOX, but the fuel matrix is an inert material, zirconium dioxide, rather than uranium dioxide.

For each of these schemes, the detailed fuel fabrication history, separation, and irradiation histories were calculated for a range of the number of recyclings. The resulting isotopic compositions of the spent fuel assemblies and the processing waste were collected into the appropriate groupings to preserve total integrated energy produced and compared with the reference case of direct disposal of spent PWR fuel. Figure 3 compares potential increases in repository drift loadings.

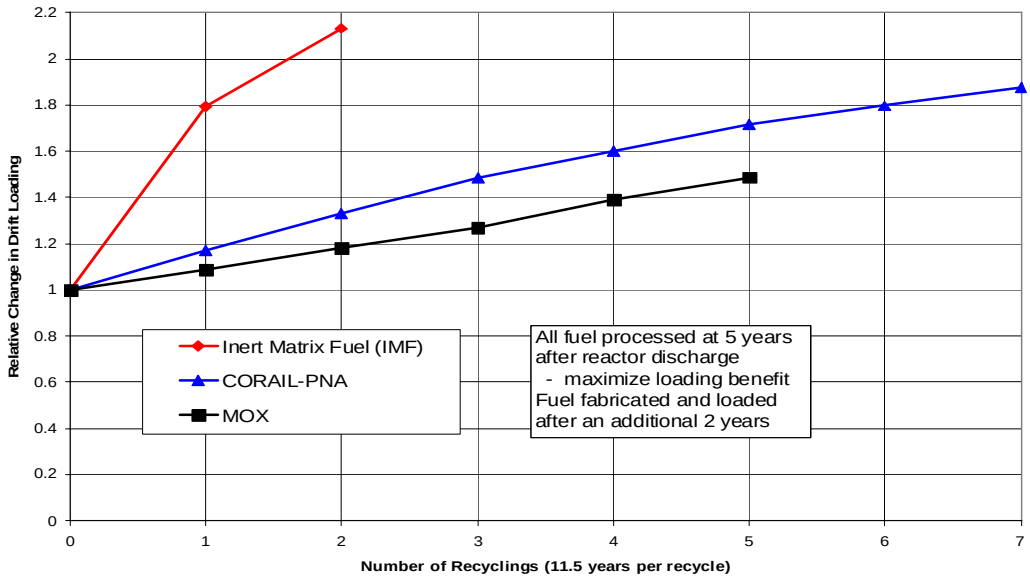

Figure 3. Comparison of the potential benefit of various recycling schemes.

As indicated in this figure, the use of MOX allows a steady increase in drift loading with each recycle of plutonium, americium, and neptunium, reaching a factor of 1.5 after five recyclings of MOX fuel. However, as noted above, the calculations assumed a homogenous reactor core with fuel of the same generation of MOX, which may not be viable after a few generations due to safety concerns. Although the use of a heterogeneous core with assemblies of all generations may alleviate this problem, Ref. 5 did not evaluate such options.

The use of CORAIL-PNA also allows a steady increase in drift loading with each recycle at a faster rate than MOX, reaching a factor of two after seven recycles. Furthermore, the CORAIL-PNA scheme appears to be tending toward an equilibrium state, where the charge and discharge amounts of plutonium, americium, and neptunium are equal and only process losses go to the repository.

The use of IMF provides a factor of 2.1 increase after two recycles. Further recycle of IMF is hindered by the rapid depletion of fissile material with each subsequent irradiation (especially for Pu-239). It should be noted that recycling of

IMF requires new processing technology, and the benefit of IMF (compared to other schemes) may be much lower if calculations assume older fuel. Additional analysis is needed.

The results from this study show that the potential increase to the repository capacity from all schemes is limited to around a factor of two if only a limited number of recycles are employed. Although in each case, the process waste can be very densely loaded in the repository drift (about a factor 40 to 50 greater than spent PWR fuel), direct disposal of the assemblies in the last recycle requires most of the repository space. Continuous recycling is needed if one wishes to obtain large increases in drift loading in the repository. This may be possible in a thermal spectrum using the CORAIL concept or advanced MOX, IMF, or target strategies. Increased uranium enrichment may be needed.

It should be noted that a combination of thermal and fast-spectrum reactors can achieve the full factor of 40 to 50 capacity increase. This combination has been evaluated previously as a method to decrease the required time that spent fuel need be isolated from the biosphere to on the order of 1000 years. It appears that one fast-spectrum system for each five to ten thermal spectrum systems is adequate.

II-5. Conclusions

Table 1 gives the number of Yucca Mountain equivalent repositories required with various nuclear energy scenarios. These are a restriction in spent fuel to the present legislated limit; life extension for all current reactors; a continuation of nuclear energy production at today's rate through the end of the century; and two scenarios with increasing nuclear energy production. (The recent MIT study scenario would lie between the last two scenarios.)

As indicated in Table 1, the most restrictive limit on Yucca Mountain capacity is the 70,000 MTHM specified in the NWPA. Because there will be 70,000 MTHM associated with operating commercial nuclear power plants until the end of there initial 40-year scenarios, the capacity of Yucca Mountain will not be sufficient if it is limited by language in the NWPA.

According to RW, if limits imposed by technical considerations and geology rather than legislation were adopted, the capacity of Yucca Mountain could be doubled. As indicated in Table 1, this factor of two increase in the capacity of Yucca Mountain would accommodate all of the SNF generated from currently operating plants in the U.S., even assuming that all of these plants are granted power upgrades and life extension.

If thermal recycling schemes such as MOX, CORAIL-PNA, or IMF are pursued, the capacity of Yucca Mountain could be increased by another factor of two. This increase requires that fission products, such as cesium and strontium are separated and stored separately for around 200 years. If a fast spectrum reactor is employed, the capacity of Yucca Mountain can be increased by factors of 40 or higher. Such an increase would yield a repository capacity that could accommodate the SNF generated from any nuclear expansion scenario that is currently proposed for the US for this century.

III. SEPARATIONS TECHNOLOGY

III-1. Spent Fuel Recycling Scenarios

The potentially significant benefit to the repository resulting from partitioning of spent fuel followed by subsequent transformation of the transuranics has been previously emphasized by Jim Laidler, National Technical Director for AFCI Separations Technology Development. This position is strengthened by the comprehensive study, "Repository Impact of Spent Fuel Recycling Using Water-Moderated Thermal Spectrum Reactors" conducted by Wigeland et al. (Ref. 6) in response to the request from ANTT Subcommittee Chair Richter's request "to analyze the model reprocessing system set forth" to assess the impact of partitioning, and recycling of actinides, on the capacity of a repository at Yucca Mountain. Repository benefit was defined as the allowable increase in drift loading consistent with satisfying all repository design limits.

A number of recycling schemes using existing light-water reactors were analyzed and the potential repository benefits were quantified. As discussed in the previous section, MOX, CORAIL-PNA, and IMF were examined, mainly in scenarios using spent fuel that had been out of the reactor for only five years.

A study of the "Continued Advantages of Processing Older Fuel" (Ref. 7) was presented at the AFCI semiannual review. In this study it was asserted that there may be key advantages in processing 30- to 40-year old fuel as opposed to only 5- to 10-year old fuel. We did not review this work. Its conclusions on repository capacity are not very different from those of Ref. 6. Studies of alternatives will be part of future optimization exercises.

III.2. Partitioning/Processing

RW is interested in partitioning even without transmutation in order to make more efficient use of the repository. In the long-range strategy (Ref. 8) presented at our February 27, 2004, subcommittee meeting, three phases including RW's needs were discussed that include RW's interests.

Phase 1: Separations for Waste Management

LWR spent oxide fuel can be treated by the UREX+ process to separate the uranium for disposal as low-level waste (LLW) which would greatly reduce the volume of waste to be sent to the repository. Technetium and iodine would be separated and incorporated in a suitable waste form for storage as high-level waste (HLW). The short-term heat load would be reduced by removal of cesium and strontium for separate decay storage and later disposal as LLW. The transuranics together with lanthanide fission products would be "self-protecting" for up to ~50 years and could be temporarily stored in inexpensive packaging in the repository until needed for use as fuel when there is a U.S. capability for doing so. This scenario would significantly reduce the waste volume and heat load of the material to be stored in the Yucca Mountain repository.

Phase 2: Thermal Recycle of Pu or Pu/Np or Pu/Np/Am

Phase 2 processing would be introduced when MOX containing Pu, Pu/Np or Pu/Np/Am is qualified for commercial LWR use. The LWR spent oxide fuel can be treated by the UREX+ process as for Phase 1, but the transuranics would be separated as oxides for mono- or multi-recycle. Development of a process for Am/Cm separation and of a storage form for Cm would be needed.

Recycle as inert matrix fuel might present a problem for aqueous processing if, for example, yttria-stabilized zirconia is used although magnesia does not present a problem. A hybrid aqueous-pyro process or an all-pyro process might be appropriate for the zirconia case, but considerable process development would be required.

Phase 2 processing might reduce fuel cycle costs due to reduced disposal costs and reduced on-site storage requirements. The volume of waste and the associated heat load of waste to be sent to the repository would also be reduced. The Am/Cm or separated Cm only product would go to temporary retrievable storage for decay of the Cm.

Phase 3: TRU Burning in Dedicated Fast Reactors

Again, the UREX+ process would be used but the transuranic oxides separated for recycle would be converted to metal or nitride forms, but Am and Cm would not be separated from each other. Pyroprocessing would be used to recycle the fast reactor fuel. Obviously, this phase is dependent on the introduction of dedicated fast-spectrum burner reactors, but could proceed in parallel with Phase 2. Then at

some point there would be no additional need for temporary retrievable storage of separated products.

III-3. Status of Laboratory Demonstrations

Progress in both aqueous- and pyro-processing development for the AFCI was presented (Ref. 9) during the semi-annual review meeting and some of the highlights are given below.

Aqueous process development:
UREX and UREX+ demonstrations to date include:

- Successful separation of pure uranium from irradiated fuel at SRTC on the laboratory scale in 2002.
- UREX+ flow-sheet test using simulant solution completed with 24 stage 2-cm contactors in 2003. Flow-sheets developed using Argonne Model for Universal Solvent Extraction (AMUSE) code.
- First-time demo of complete UREX+2 flow sheet for Phase 2 processing, consisting of the UREX process for uranium extraction with subsequent separations of Pu/Np, Cs/Sr, and Am/Cm, with Big Rock Point high-burnup fuel attempted at ANL in August-September 2003. Conducted laboratory-scale dissolution of 1 kg spent fuel and UREX test using 24-stage centrifugal contactors in shielded hot cell with subsequent processes for Cs/Sr extraction (CCD-PEG), Np/Pu (NPEX), removal of non-lanthanide fissions products (TRUEX) and Am/Cm extraction (CYANEX-301). AFCI separations criteria for product recoveries were met but operational “mishaps” caused some problems including Pu contamination of the U product. Lighter lanthanides carried with Am/Cm and some 20% of original Pu remained in the sludge. Valuable lessons were learned and additional complete Phase 2 “hot” demos are needed. The preferred flow-sheet now includes a co-decontamination process at the front end, and this flow-sheet will be tested in the balance of FY2004.
- The following activities were carried out to optimize the various processes needed in the complete Phase 2 processing (UREX+2):
    - INEEL conducted laboratory-scale investigations with the CCD/PEG process to optimize parameters for use in the UREX+ hot demo.
    - Two tests of UREX+ co-decontamination (Pu+Np) process flowsheets without Cs/Sr extraction conducted at ORNL helped determine best conditions for co-extraction of Np with Pu using various reagents such as AHA and nitrite ion. Recoveries of >99.99% for U and Pu and 97.5% for Np were achieved, but 10% of Tc was in the Pu/Np product. Am/Cm separations with simulant solutions and several different extractants are also being investigated at ORNL.

Pyrochemical process development:
As pointed out under the section on Phase 2, recycle with inert matrix fuel might be a problem for aqueous processing and a hybrid aqueous/pyro-process or an all-pyro-process might be envisioned. It is too early to review these Phase 2 processes, but electrolytic reduction of oxide-type fuels has been achieved in lab-scale cell studies in which oxides of U and Pu were reduced to metal. Modeling to produce a scalable design is in progress, and an advanced electro-refiner with high throughput and high efficiency that can be integrated with subsequent pyro-process operations is under development.

The AFCI separations 10-year development plan (Ref. 8) presented at our February 27, 2004 meeting is given in Fig. 4.

Much has been accomplished in the past year and participation of all the laboratories (ANL, INEEL, LANL, ORNL, WSRC) in the development program is outstanding. However, the momentum needs to be maintained and even accelerated if an engineering-scale demo of the Phase 1 separations process is to begin in FY-2008.

All of the thermal recycle scenarios require spent fuel processing and appropriate steps need to be taken in a timely fashion if a large spent fuel treatment plant is to begin operation in 2025. It appears that NE is unable to fund a demo of the Phase 1 separations process in the U.S. in the near term, but together with RW is seriously considering a COGEMA proposal to perform an engineering scale demo of parts of the UREX+ flowsheet at La Hague, France. This would be facilitated in the form of an Implementing arrangement appended to the September 2000 DOE-CEA Cooperative Agreement. ***If*** the demo starts at the beginning of FY-2005 it could be completed, including written reports, by the end of FY-2007.

A major portion of the UREX+2 process flow-sheet would be matched to the extent possible to the La Hague commercial reprocessing plant of COGEMA using 80 metric tons of French fuel. Performance goals include: >99% recovery of uranium from spent fuel feed and purity from the second extraction cycle that meets criteria for Class C low-level waste; recovery of >99.5% of the plutonium; minimization of the neptunium content and less than 5% loss of soluble technetium in the raffinate of the first U/Pu/Np extraction cycle. Mixed U/Pu/Np oxide is to be produced and recovery of Tc and Np is to be maximized. If the demo were completed by the end of FY2007 the results could be considered in preparation of the Secretary of Energy's recommendation to Congress on the need for a second repository.

A demonstration at the 50 to 100 tons of spent fuel per year scale would still be needed in the U.S. for obtaining additional economic data and is considered a necessary precursor to Congressional approval of line item construction of a large

AFCI Separations 10-Year Development Plan

James J. Laidler Presentation, Slide 16, February 27, 2004

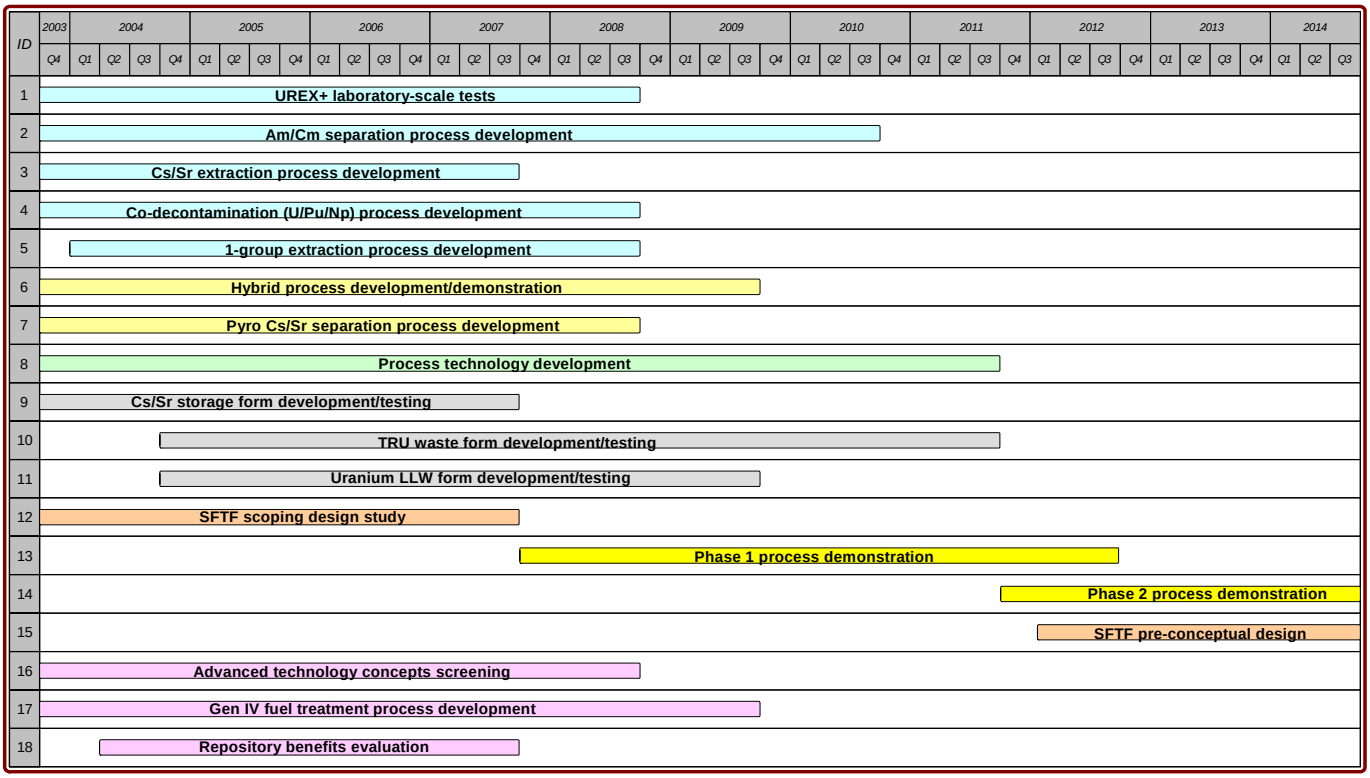

Figure 4.

project. Preparation for the engineering-scale demonstration for a large spent fuel treatment plant would need to begin in 2012 in order to become operational by 2025.

FUEL AND BURNER DEVELOPMENT

A recent systems study (Ref. 6) indicates that Inert Matrix Fuel, when used in Light Water Reactors, has the potential to increase the drift loading in the geologic repository by as much as a factor of two in two recycles. This is the maximum repository benefit and is obtained when plutonium along with americium and neptunium are recycled. This result can be obtained while keeping the temperature between the drifts below the boiling temperature, which was one of the criteria employed in this study. To complete the Inert Matrix Fuel story, two systems questions should be answered and the results documented. They are: (a) how many Light Water Reactors must be loaded with this fuel and when must this loading begin, and (b) how much Inert Matrix Fuel must be loaded into each reactor and how is this loading distributed over the core? Answers to these questions will provide the information necessary to make a comparison of the relative benefits of burning americium and neptunium in Mixed Oxide Fuel versus doing the same with Inert Matrix Fuel. Obtaining answers to these questions are high-priority steps for moving forward in the systems area as they will determine the strategic usefulness of the Inert Matrix Fuel concept.

In contrast to systems issues, the principal technical issue to be addressed with either Mixed Oxide or Inert Matrix Fuel is the ability to fabricate such fuel with americium. Recent fabrication development efforts (Ref. 10) have shown that fabricating fuel with americium may pose some difficultly. But the experiments were not conclusive because they attempted to fabricate fuel rods with nitride pellets containing americium with a sodium bond between the pellet and the cladding. Since this is unlikely to be the reference Inert Matrix Fuel for Light Water Reactors, the results should not be taken as demonstrating the infeasibility of either Inert Matrix Fuel or burning americium and neptunium in Light Water Reactors. Rather plans should be made to explore the feasibility of fabricating both Mixed Oxide Fuel and Inert Matrix Fuel with americium and neptunium. With respect to Inert Matrix Fuel, the inert material should probably be zirconium oxide.

It is difficult to choose technically between Mixed Oxide Fuel and Inert Matrix Fuel, both burning neptunium and americium so as to provide maximum benefit to the geologic repository, with the information currently at hand. But the technical criteria will be better understood when the work outlined in the two proceeding paragraphs is completed. At that time in the future, the relative ease or difficultly associated with fabrication will be known and the nuclear system architecture required to support such a burner system will likewise be known. When this information is combined with the relative drift loading, which is now known, three of

the four parameters needed to choose between the two pathways on a first principles basis will be available. There may be proliferation related considerations that may influence the final choice.

The fourth parameter required to define the pathway is some definition of the need for a fast spectrum reactor. If the pathway of choice is to burn plutonium, americium, and neptunium in either Mixed Oxide Fuel or Inert Matrix Fuel in Light Water Reactors, the role of the fast spectrum reactor as either a steady-state reference burner or an end-state burner has yet to be determined. This is again a system study product, and both the timing and the relative number of fast spectrum reactors should be determined for both the fuel systems above, when used in Light Water Reactors. Knowing the required date and the number of fast spectrum reactors will provide the fourth decision parameter with respect to the choice between Mixed Oxide Fuel and Inert Matrix Fuel. Experimental fuel development and systems analysis activities should be directed toward providing information related to these four decision parameters.

V. INTERNATIONAL COLLABORATIONS

The formation of international collaborations has been an important element of the U.S. next generation nuclear energy R&D Program. For the GEN-IV Program, perhaps the most important collaboration is the Generation IV International Forum (GIF), a partnership of Euratom and the following ten countries: Argentina, Brazil, Canada, France, Japan, Republic of South Africa, Republic of Korea, Switzerland, United Kingdom, and the United States. To date, there is no comparable large-scale international collaboration devoted to the partitioning and transmuting of radioactive wastes. However, there have been a number of smaller efforts that have resulted in DOE's being able to obtain important analytical and experimental data that would have cost in excess of $100 million had the work been performed in the U.S.

Previously, the ANTT Subcommittee has described a number of AFCI international collaborations. Most notable have been the following:

TRADE:	Coupled cyclotron to TRIGA reactor in Italy.
MUSE:	Coupled external sources to fast reactor criticality facility (CEA-Cadarache).
PROFIL:	Small sample irradiation experiments at the PHENIX fast reactor.
CEA-Saclay:	Advanced Cavity Development.
JPARC:	Target Test Station and low-power Subcritical Multiplier in accelerator complex.
MEGAPIE:	Megawatt scale lead/bismuth spallation source at PSI.

An important component of the AFCI Program will be the testing of fuels and reactor materials in a fast neutron flux environment. With the permanent shutdown of the Fast Flux Test Reactor and the probable shutdown of France's PHENIX fast reactor around 2008, few fast spectrum research reactors will exist in the world. Thus, there is a need to collaborate with Russia on experiments in its 60 MW BOR-60 fast reactor in Dimitrovgrad, and/or with the Japanese on their JOYO and Monju fast reactors. Hopefully, relations between the U.S. and Russia will improve in the near future to make that collaboration a real possibility.

In the area of fuels development, the ANTT Subcommittee was impressed by the recent work of Roald Wigeland and his colleagues at Argonne National Laboratory. Wigeland's team ran analyses that showed that MOX and inert matrix fuels (IMF) could achieve significant enhancements in repository loading when compared to once-through spent fuels, particularly if there is a final burn of the minor actinides in a fast reactor. The IMF approach is similar to MOX, with the difference being that the fuel matrix is an inert material, like zirconium oxide, instead of uranium oxide. There is little known worldwide about the fabrication of MOX and IMF fuels containing both Pu and the minor actinides; hence, there is fertile ground here for international collaborations.

Inert Matrix Fuels (IMF)

For IMF, most of the international interest over the last decade has been the development of fuels for once-through plutonium disposition followed by direct disposal, a concept called OTTO, for "Once Through, Then Out." An important benefit of IMF is the faster Pu burn due to the lack of fertile uranium. To facilitate the progress of international collaborations on IMF, there have been yearly meetings in Europe and Japan. Out of these collaborations, under the OTTO initiative, have come irradiation tests of potential fuel matrices and comparisons of the performance of various fuel types, such as spinel versus zirconia matrices, macro versus micro dispersions, and more generally IMF versus MOX fuels. These collaborations are aiming to resolve a variety of IMF issues, such as the effect on fuels of introducing Americium in significant quantities. Two notable effects are the increased generation of Helium gas and the decrease in the fuels' thermal conductivity. The introduction of neptunium into such fuels presents fewer problems, such as a much lower rate of helium gas generation.

IMF-related activities are in progress in Canada, where they are studying ion beam (72 MeV iodine) irradiation of a variety of matrices, including $ZrSiO_4$, $MgAl_2O_4$, CeO_2, $CePO_4$, ZrO_2, Er_2O_3, Y_2O_3, and SiC. Other IMF work is in progress in Russia, where they are studying Pu burning via the irradiation of $MgO\text{-}40PuO_2$ in its BOR-60 fast spectrum reactor. Still other IMF work is taking place in Japan, where they are studying Rock-like Oxide (ROX) fuel at JAERI (Japan Atomic Energy Research Institute) and several universities. They are also performing

scoping studies on a variety of matrices, such as yttria stabilized zirconia, and they are leach testing irradiated fuels.

Hence, with so much worldwide activity centered around advanced fuels development and testing, and in particular the promising inert matrix fuels, DOE should take full advantage of mutually beneficial collaborations with other nations.

COGEMA

The collaboration on the UREX+ partitioning with French collaborators at the La Hague reprocessing plant of COGEMA is discussed elsewhere in this report. Here, we only note that the French do not have a repository and have based their reprocessing plant design on the vitrification of high-level wastes containing Cs, Sr, Am, and Cm. Thus, Cs/Sr and Am/Cm extraction, which are part of the AFCI R&D Program, would not be demonstrated in the La Hague experiment. However, following a report that is due in 2006, the French law may require future processing of the minor actinides so that they are not sent to geologic disposal; hence, in the future, there may be more convergence of the French and AFCI Programs on minor actinide reprocessing.

Lead Bismuth Eutectic (LBE) Corrosion Studies

The work in this area is centered on the DELTA Loop corrosion studies at Los Alamos National Laboratory, with participation by a number of universities. They have completed some 1000 hours corrosion tests of over twenty (20) materials in DELTA. Testing was done at temperatures around 400°C, at LBE flow velocity of 2 m/s, and for various oxygen concentrations. Post-exposure studies of the materials are in progress, and future tests will obtain more systematic measurements of corrosion resistance of selected materials.

On the international front, the team is exchanging data with CEA; submitting an INERI proposal with the Korea Atomic Energy Research Institute (KAERI) and Seoul National University to test materials under varying conditions, improve oxygen sensors, and improve modeling analyses; and forming the OECD/Nuclear Energy Agency LBE Expert Group to prepare a review of the fundamental issues of LBE corrosion and a summary of DELTA operating experience.

General Comments

Some of the main challenges that could be overcome at least partly by international collaborations are the following:

(1) Separating plutonium and americium at 99.9% efficiency (as assumed in the Argonne analyses)
(2) Demonstrating the UREX+ process on an engineering scale

(3) Recycling the matrix material for IMF fuels
(4) Developing water-compatible matrix material for IMF fuels
(5) Fabricating minor actinide-containing fuels remotely
(6) Irradiating promising fuel types in fast neutron spectra.

The ANTT Subcommittee was pleased to learn that DOE officials met with a French team during March 2004 to discuss collaborating on reprocessing issues. The French have excellent expertise in reprocessing; thus, joint experiments with them, such as the proposed UREX+ engineering scale demonstration at La Hague, will be most important for the AFCI Program.

Conclusions

The ANTT Subcommittee believes that the AFCI Program do the following:

(1) Perform further evaluations and validation of the important Argonne work on the relative benefits to repository loading from various reprocessed fuel types
(2) When the political climate permits, test reactor components and the most promising minor actinide-containing fuel types in the Russian BOR-60 fast reactor or Japanese JOYO or Monju fast reactors
(3) Continue with the ongoing collaborations like TRADE that are enumerated above.

Finally, the ANTT Subcommittee notes that aqueous reprocessing of LWR spent fuel is currently practiced in France, the United Kingdom, and Russia. Moreover, Japan soon will begin operation of a commercial facility, the Rokkasho Reprocessing Plant. Thus, there will be many opportunities for international collaborations of which the AFCI should take full advantage.

REFERENCES

1) *Nuclear Waste Policy Act of 1982*. Public Law No. 97-425. 96 Stat. 2201 (it became law in January 1983 and was amended in December 1987).

2) *Yucca Mountain Science and Engineering Report – Technical Information Supporting Site Recommendation Consideration*, Rev. 1, DOE/RW-539-1, February 2002.

3) Energy Information Administration, *Spent Nuclear Fuel Data, Detailed United States, as of December 31, 1998*, www.eid.doe.gov, accessed March 19, 2004.

4) Herring, S. T., *Interactions of Generation IV Systems and the Advanced Fuel Cycle*, Advanced Fuel Cycle Initiative Semi-annual Review Meting, Sandia Livermore, February 26, 2004.

5) R. A. Wigeland, et al., *Spent Nuclear Fuel Separations and Transmutation Criteria for Benefit to a Geologic Repository*, Waste Management 2004 (WM'04), Tucson, AZ, February 29-March 4, 2004.

6) R. A. Wigeland, et al., *Repository Impact of Spent Fuel Recycling using Water-Moderated Thermal Spectrum Reactors*, ANL Report Transmitted to ANTT Subcommittee by F. Goldner, DOE-NE, February 18, 2004.

7) E. D. Collins, J.-P. Renier, B. Del Cul, B. B. Spencer, and D. T. Ingersoll, "Continued Advantages of Processing Older Fuel: Partitioning/ Transmutation in LWRs," AFCI Semiannual Review, February 24, 2004, Sandia National Laboratory, Livermore, CA.

8) J. J. Laidler, "Advanced Fuel Cycle Initiative: Separations Technology Development Strategy," presented to ANTT Subcommittee of NERAC, February 27, 2004, Sandia National Laboratory, Livermore, CA.

9) J. J. Laidler, "Advanced Fuel Cycle Initiative: Progress in Aqueous and Pyrochemical Process Development," AFCI Semi-Annual Rview, February 27, 2004, Sandia National Laboratory, Livermore, CA.

10) Willson, et al., "Actinide Nitride Fuel Development," presented to ANTT Subcommittee of NERAC, February 27, 2004, Sandia National Laboratory, Livermore, CA.