THE ROLE OF ACCELERATORS IN THE ENERGY PROBLEM

R. L. Sheffield# and E. J. Pitcher, LANL, Los Alamos, NM 87545, U.S.A.

Abstract

Nearly all risks to future generations arising from long-term disposal of used LWR nuclear fuel are attributable to the transuranic elements and long-lived fission products, about 2% of its content. The transuranic elements of concern are plutonium, neptunium, americium, and curium. Long-lived (>100,000-year half-life) isotopes of iodine and technetium are also created by nuclear fission of uranium. If we can reduce or otherwise securely handle this 2% of the used fuel, the toxic nature of the remaining used fuel after a few centuries of cooling is below that of the natural uranium ore that was originally mined for nuclear fuel. Only a small fraction of the available energy in the fuel is extracted on a single pass and the majority of the 'problem wastes' could be burned in fast-neutron spectrum reactors or sub-critical accelerator driven transmuters. The goals of accelerator transmutation are some or all of the following: 1) to significantly reduce the impacts due to the minor actinides on the packing density and long-term radiotoxicity in the repository design, 2) preserve/use the energy-rich component of used nuclear fuel, and 3) reduce proliferation risk.

INTRODUCTION

A key roadblock to development of additional nuclear power capacity is the concern over management of nuclear waste. Nuclear waste is predominantly comprised of used fuel discharged from operating nuclear reactors. The 104 operating US light water reactors (LWRs), that currently produce about 20% of the US electricity or more than 70% of the U.S. emission-free electricity, and, given the life extension of present plants, will create about 120,000 tons of such used fuel over the course of their lifetimes. Worldwide, more than 250,000 tons of used fuel from reactors currently operating will require disposal. The toxicity of the used fuel, mainly due to ionizing radiation, will affect future generations for long into the future. The large quantity and its long-lived toxicity present significant challenges in waste management.

The U.S. currently employs a "once-through" nuclear fuel cycle. In the 1970's, apprehension about an ever expanding "Plutonium Economy" and the associated proliferation concerns in other countries led the Carter administration to cancel the U.S. Department of Energy's Clinch River Breeder Reactor project and place a ban on reprocessing commercial used nuclear fuel. Also, the low price for uranium ore over the last several decades has made "once-through" cycle economical. However, the long term nuclear waste disposal still needs to be addressed. Under any scenario, at some point in time either short-term or long-term geologic repository(ies), Figure 1, must be made available to receive the reactor waste. In the U.S., used nuclear fuel is presently stored in buildings on reactor sites, with the expectation that it was to be sent to the proposed Yucca Mountain repository once it received regulatory approval to accept high-level waste. Recently the present U.S. administration decided to terminate the Yucca Mountain project and withdraw its license application to the Nuclear Regulatory Commission, which has revived the question of what is the best course of action for used nuclear fuel. Thus a credible long-term solution beyond implementing short-term on-site storage must be advanced to ensure political acceptance of continuing and expanding nuclear power in the U.S.

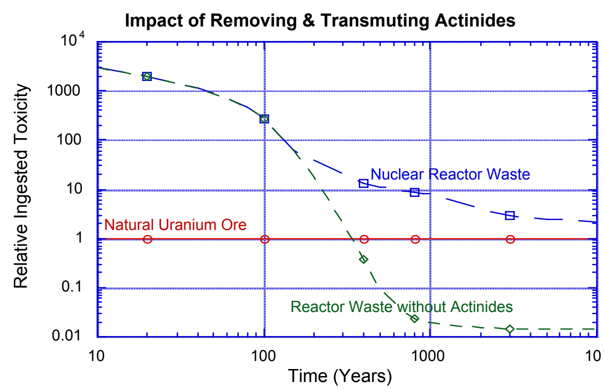

Figure 1: Unprocessed spent fuel containing materials that need isolation from environment for greater than 10,000 years requires a geologic repository. This type of repository uses geologic characteristics to isolate wastes after containers and barriers fail. For geologic repositories, the ground water transport is a key issue and climate change and population shifts add uncertainty to the long term isolation design basis. However, if the plutonium and minor actinides are removed, the requirements change in that the toxicity falls below natural uranium ore within a few centuries. Current man-made containers are capable of providing more than 300 years of isolation.

Nuclear fuel seems ideally suited for recycling. Only a small fraction of the available energy in the fuel is extracted on a single pass and the majority of the "problem wastes" could be burned in fast-neutron spectrum reactors. Most of the remaining wastes have half-lives of a few hundred years and can be safely stored in man-made containment structures (casks or glass). The very small amount of remaining long-lived waste could be safely stored in a small geologic repository. The problem for the next 100 years is that it is highly unlikely that a sufficient number of fast reactors will be built by industry to burn their own waste and the LWR waste from existing and new reactors. So an interim solution is required to address the ever-increasing inventory of used nuclear fuel. Proposed solutions should consider all

sheff@lanl.gov

aspects of the nuclear fuel cycle, including costs and complexity of reprocessing, methods for fabrication of fuel from recycled material and the performance of this fuel (reliability and lifetime as measured by burn-up), cost of the infrastructure needed to utilize this fuel (can existing LWR's use it without major modifications, or will a fleet of new reactors be needed?), and benefit to geologic repositories resulting from recycling.

One interim solution is to dispose used fuel using a combination of approaches depending on the lifetime of the radioactive isotope. Long-lived fissile isotopes like Pu-239 and U-235 can be stored with U-238 and Np-237 for fabrication into nuclear fuel at a future date. The short-lived fission products can be stored in man-made containers until they safely decay to low radiotoxicity and decay-heat levels. The long-lived fission products can be vitrified and sent to a HLW repository. A number of projects have looked at transmuters with only minor actinide loading. [1] From a repository perspective, near-term packing density of HLW is limited by the heat from radioactive decay in the short-term (<500 year), which is dominated by fission products. Long-term storage is limited by characteristics of the geologic medium in which the HLW is placed and the potential spread of radiotoxic isotopes. Isotopic contributions to the decay heat are shown in Figure 2.

Americium can be most efficiently eliminated through nuclear transmutation using fast neutrons. One method for fast-neutron production uses a high-energy proton beam hitting a heavy-metal target, generating spallation neutrons. These spallation neutrons can drive a subcritical core to transmute the long-lived Am isotopes to shorter-lived fission products, and these products are disposed in short-term repositories. The Am feedstock is assumed to be from used fuel that has set for 50 years after removal from the reactor. At 50 years, 97% of the Pu-241 has decayed to Am-241. The remaining un-decayed Pu-241 can be sent for long-term storage with the other Pu isotopes without significantly impacting the overall properties (internal heating, neutron source, etc.) of the stored material.

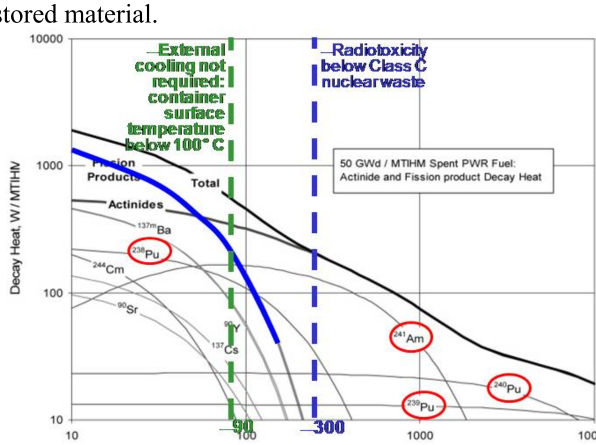

Figure 2: Dominant decay heat contributors in spent PWR fuel irradiated to 50 GWd/MTHM.[**2**] Goal is to eliminate components of the nuclear waste stream that account for the majority of the heat load and toxicity over the 300 to 10,000 year time frame. The isotopes circled in red are the major contributors to the decay heat in this time frame. If these isotopes are removed then: the solid blue line shows the decay heat of the remaining waste; the green dashed line shows the time at which the surface temperature of the waste container is below the boiling point of water; and the blue dashed line gives the time at which the waste radiotoxicity is below Class C nuclear waste.

ACCELERATOR BASED TRANSMUTATION

Fast-neutron based transmutation has three major technology elements: separations, fuels and waste forms, and a fast neutron source coupled with a transmuter. A well designed accelerator-driven transmuter would operate in a sub-critical mode, and with limited excess reactivity such that the transmuter cannot reach criticality under any design basis accident.[3] For this type of transmuter, the fission rate (and hence total power generated by the transmuter) is directly proportional to the source neutron production rate. The flexibility enabled by subcritical operation has several advantages:

- Can use fuel with low fissile content (Th or M.A.) or high burden of non-fissile materials
- Unlike critical reactors, can safely operate with fuel having a relatively low delayed neutron fraction
- Can compensate for large uncertainties in initial reactivity or burnup reactivity swings by varying the source rate, which for an accelerator-driven system is proportional to the beam current.

Accelerator Technology

The power of the accelerator is determined by the design of the subcritical multiplier. For example, for a subcritical blanket fission power of 3 GW and a reactivity k_{eff} in a range of 0.95 to 0.98, the proton beam power ranges from 55 MW to 21 MW, which corresponds to a beam current swing of 37 mA to 14 mA, assuming a beam energy of 1.5 GeV. Either starting out with a lower k_{eff} or going to deeper burn, again resulting in a lower k_{eff}, would require an increase in the accelerator current. Given fixed beam energy, the accelerator capital cost is determined in large part by the average current. Designing an accelerator for a large current swing requires a very high beam current that is used for only part of the transmutation cycle. This application is best served by a continuous wave machine, either linac or cyclotron. Cyclotrons could potentially deliver up to 10 MW of beam power (10 mA at 1 GeV). Linacs are limited to about 100 mA per front end system, with funneling used to double the current. Either type could serve to drive a subcritical transmuter.

Since this transmuter system will be a production system, a factor of 1.5 to 2 overhead margin is typically built into the performance specification to assure high operational reliability and long life. So the maximum operational currents are 5 to 8 mA for cyclotrons and 50 to 75 mA for linacs. In this paper we are looking at accelerator systems that could drive plants of several GW thermal power and have currents up to 40 mA, and so the accelerator technology covered in this article will be limited to linac systems. If designing lower power transmuters, say less than 800 MW thermal, then MW-class cyclotrons should also be considered.

Economy of scale generally favors going to the highest average power from a single accelerator. Note that the beam may impinge on a single target in a core, be split into separate targets in a single core, or be directed to multiple cores. Of course, with the consideration of multiple targets, multiple accelerators may provide system redundancy and improved reliability, but at added cost. Beam parameters and components consistent with the above operating numbers were demonstrated to be feasible under the Accelerator Production of Tritium (APT)[4,5] program (Figure 3). Under the APT program, a prototypical CW 100 mA, 6.7 MeV (0.67 MW of average power) RFQ and MW RF power couplers were successfully demonstrated. Also, the existence of significant beam halo growth in MW proton beams and the means to mitigate said halo were demonstrated.

The linac requirements follow from other sub-system requirements, but more thorough studies are required to determine the full sets of requirements. For example, beam interrupts longer than one second might negatively impact the subcritical multiplier. The engineering challenges need to be fully scoped out for the safe, controlled coupling of an accelerator to a subcritical reactor through a spallation target. System control and safe operation will demand the understanding and resolution of the potentially complex behavior of this coupled accelerator/target/reactor system.

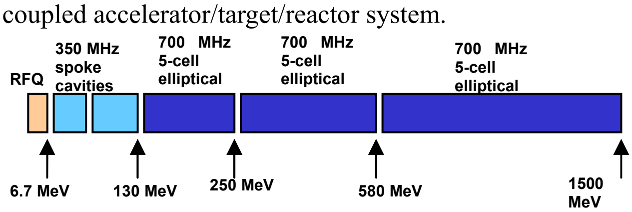

Figure 3. The accelerator preliminary design is based on the technologies developed for the APT program. The superconducting linac reduces cost and improves performance and reliability (i.e. beam continuity).

A SCRF linac is likely the best choice for the linac because, compared to linacs using traditional room-temperature (RT) copper technology, SCRF linacs are more power efficient and expected to have higher reliability. The SCRF linac will employ independently controlled RF modules with redundancy, allowing the less than 300 ms adjustment of RF phases and amplitudes of RF modules to compensate for faults of individual cavities, klystrons, or focusing magnets. The basic concept of using adjacent cavities to compensate for the loss of a cavity has been demonstrated at the Spallation Neutron Source.[6] The SCRF cavities will have larger bore radius that relaxes alignment and steering tolerances, as well as reducing beam loss. Thermal transient has been a major cause of out-of-lock trips in RT linacs. Operating at a stable cryogenic temperature, SCRF linacs are expected to have significantly reduced number of such trips.

The expected cost of an accelerator based transmuter system compared to a reactor is expected to be <30%. As will be seen later, we estimate that just 3 high-power linacs can transmute the Am generated by the entire existing fleet of US LWRs. So if the incremental cost to the present electrical rate is based on the addition of three transmuter plants (that don't produce any electricity), the incremental operating-cost of the 104 LWRs should be just 4 to 6 percent, not including reprocessing costs. Experience in France has shown that reprocessing costs are fully covered by the fuel produced by reprocessing. Since operation of the transmuter is expected to generate an excess of 8 GWth/year, converting that power into a useable energy source is highly advantageous to help recover the facility capital and operating costs.

Consideration should still be given to converting the generated energy to another form useful for national consumption. One option is to drive electrical generators and sell the excess power to the grid. Power storage devices, such as flywheels or compressed gas storage, could provide the electricity to run through faults if they can store enough energy to enable providing steady power to the grid through the longest of expected interruptions. The practicality of running through the range of possible interruptions requires a more detailed design effort.

Another option is to convert the power into another energy form. Charles Forsberg has proposed that biomass can be converted to greenhouse-gas-neutral liquid fuels.[7] The conversion of biomass-to-liquid fuels is energy intensive but the transmuter can produce the significant amount of heat, electricity, and hydrogen required for the processing of biomass-to-liquid fuels. The overall process has a comparable efficiency to electrical production, but the end result can be carried away in tankers. If the accelerator operation is deemed too unreliable for the electrical grid, then converting biomass into fuel for a net-zero carbon-footprint would seem to be not only a good option, but the preferred option.

Other than intermittent operation affecting the quality of the power produced by the transmuter, transients will affect the lifetime of components in the transmuter assembly. Effect of transients on materials and fuels was evaluated at LANL for the proposed Material Test Station.[8] The studies show no significant deleterious effects for core clad or structural materials for the expected accelerator interruptions. Similar studies also show no concern for fuels.

Accelerator Technology

To understand and develop a path forward to manage LWR used fuel, LANL put together a team of experts on fuels, proliferation, actinide chemistry, repositories, reactor design and neutronics, and accelerators. The result of this team's effort was a concept called SMART (Subcritical Minor Actinide Reduction through

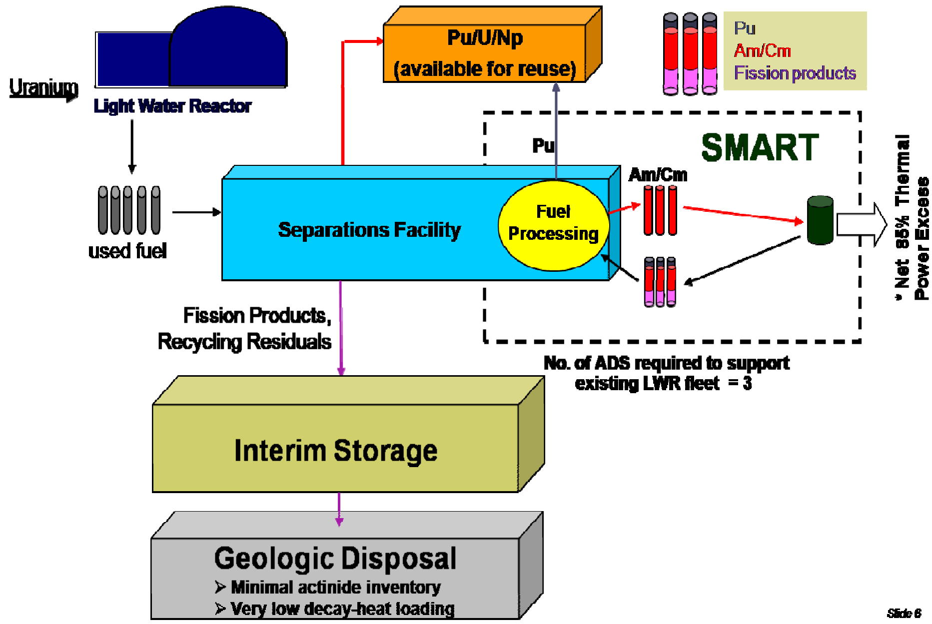

Figure 4. The SMART concept is shown above. Spent LWR fuel is sent to a reprocessing center where the short-lived fission products are vitrified and sent to storage. The Pu, U, and Np are sent to another storage facility for the possible fabrication into fuel. After transmuter start-up with an initial feed of Am and Pu, the only feedstock is Am. In equilibrium, an excess of Pu is generated from the Am transmutation chain. This Pu supplies the required fertile fuel component for maintaining k_{eff} and the excess goes back into the separations facility and stored with the Pu, U, Np.

Transmutation), the goal being to support the existing U.S. LWR economy, preserve the energy rich component of nuclear waste as a future energy resource, and provide a long-term strategy enabling the continuation and growth of nuclear power in the U.S.

The basic concept of SMART was discussed in the Introduction of this paper. SMART is to extract and store the Pu, U, and Np together in interim storage facilities for future fabrication into fuel, vitrify and store the short-lived fission products in interim storage facilities, vitrify and store the long-lived fission products in small geologic repositories along with small amounts of other residual high-level waste, and burn the Am in an accelerator driven transmuter.

The reason for burning only the Am is economic and technical. From an economic stand-point, no incentive exists for the large scale deployment of accelerator-based facilities by private industry, and so any facility whose primary function is to deal with burning nuclear waste will probably be government owned and operated by a government contractor. This implies a scenario that uses the minimal number of facilities to support the waste mission. The number of facilities depends in large part on what mixture of actinides the facility is to burn. A reactor of fixed size is ultimately limited by the thermal heat generated from burning its nuclear fuel. A 3 GW thermal (GWth) reactor burns about 1 metric ton (MT) of fuel each year. A sub-critical transmuter also burns fuel, but the composition of the fuel is not limited by the same safety considerations as a critical reactor. The equilibrium feed for a subcritical burner can be 100% Am, whereas critical reactors cannot operate on a pure Am feed stream. SMART, Figure 4, is focussed on transmuting the one element that has the greatest impact on nuclear waste management.

The major technical reasons, the repository decay-heat, radiotoxicity, and long half-life, for concentrating on Am was given in the Introduction, but other related aspects give concern for storing Am as a future component of LWR or fast reactor fuel follow:

- Am and its daughter nuclides are difficult to handle because of their high radioactivity, making fuel fabrication much more expensive.
- The vapor pressure of americium limits fuel pellet fabrication temperatures.
- In fast reactor accident scenarios the americium could boil out of the fuel and thus present a more difficult safety case.

Separating out and storing the Np, U and Pu for future use has several advantages.

- Neptunium has similar chemical properties to uranium and plutonium and shows good compatibility not only in nitride fuel but also in oxide fuel.
- Processing criticality issues are mitigated since the Pu is not separated out.
- U/Pu/Np ratios are 98.7:1.2:0.1 making this a very unattractive material for diversion since the fissile isotopes are heavily diluted.

Plutonium-240 (0.3% of U, Np, Pu at 50 years) has an easily detectable neutron signature adding in diversion detection. [9]

Flow sheets for this transmuter can be based on advances to PUREX reprocessing or newer concepts such as modification to the DUPIC process. The initial separation of an Am/Cm product stream from reprocessing does not appear to be the major technical challenge from a chemistry/materials view point. However, the following could be problem areas:

- Target manufacture if Cm is a component of that fuel target.
- Dissolution of that target after irradiation for recycle of actinides.
- Recycle options that minimize chemical steps, such as reliance on mechanical or thermal separations, are most desirable as they will likely reduce recycling waste streams.

A fuel form optimized for the proposed Am transmuter needs to be developed. Although some effort has been undertaken for heavily Am-loaded fuels, [10] each different type of transmuter requires its own fuel form depending on the isotopic composition of its radionuclides and the reprocessing flow sheets.

More effort is needed to fully develop the SMART concept, specifically:

- Design an optimized sub-critical reactor core and associated refueling methodology
- Develop a tailored flow-sheet for Am, U, Pu, and Np separation
- Determine the optimal fuel form for an Am burner
- Develop fair cost comparison to other fuel cycles.

SUMMARY

A significant impact on nuclear waste treatment can be made by conversion of the 0.24% non-fissile fraction of the commercial spent fuel that requires long-term isolation into materials that are primarily stable or short-lived. The use of an accelerator adds flexibility in burning these "difficult" fuels and is the missing link in integrated waste transmutation systems. Most likely the LWR waste will be the government's problem – this is consistent with an accelerator collocated with a government reprocessing facility with the following objectives:

- Reducing isolation requirements to fit the lifetime of man-made containers and barriers.
- Reducing incentives and consequences of intrusions into repositories.
- Improving prospects for waste storage and nuclear technologies.
- Improving fuel utilization.
- Reducing proliferation risk.

The major challenges in accelerator driven transmuters are related to fuel forms and separations; the accelerator is based on demonstrated technologies.

Production of electricity or converting the energy to other forms is optional but would pay for the facility and associated operational costs. Since only two to three transmuters are required for the present U.S. LWR fleet then the incremental electrical cost should be a few percent, neglecting the other components needed to close the nuclear fuel cycle (e.g., reprocessing and hot fuel fabrication). Over the next three decades a reasonable expectation is that:

Accelerator faults can be reduced to an acceptable level through technology improvements. To help address the remaining interrupts, high-capacity energy storage systems will see significant improvements driven by alternative energy sources, such as solar and wind.

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