# UTILIZATION OF ACCELERATORS FOR TRANSMUTATION AND ENERGY PRODUCTION

R. L. Sheffield, LANL, Los Alamos, NM 87545, U.S.A.

## *Abstract*

Given the increased concern over reliable, emission-free power, nuclear power has experienced a resurgence of interest. A sub-critical accelerator driven system (ADS) can drive systems that have either safety constraints (waste transmutation) or reduced fissile content (thorium reactor). The goals of ADS are some or all of the following: 1) to significantly reduce the generation or 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.

ADS systems have been actively studied in Europe and Asia over the past two decades and renewed interest is occurring in the U.S. This talk will cover some of the history, possible applicable fuel cycle scenarios, and general issues to be considered in implementing ADS systems.

## 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. Worldwide, more than 250,000 tons of spent fuel from reactors currently operating will require disposal. The toxicity of the spent 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.

Nuclear fuel seems ideally suited for recycling. However, the low price for uranium ore over the last several decades has made the "once-through" cycle economical. Under any scenario, at some point in time a combination of short-term and long-term geologic repositories must be made available to receive the reactor waste.

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 reactors. Fast-reactors have a hard neutron spectrum relative to thermal 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 a sufficient number of fast reactors will not 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 transition to a fast reactor economy.

One interim solution is to dispose spent fuel using a combination of approaches depending on the lifetime of the radioactive isotope. The short-lived fission products can be stored in man-made containers until they safely decay to low radiotoxicity levels. 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 long-lived fission products can be vitrified and buried.

Repository design is significantly impacted by the radioactive decay heat for at least 10,000 years. Long term storage is also limited by container failure and the potential spread of radiotoxic isotopes. Isotopic contributions to the decay heat are shown in Fig. 1. Note that Am-241 is the major source of decay heat at times longer than the lifetime of engineered barriers.

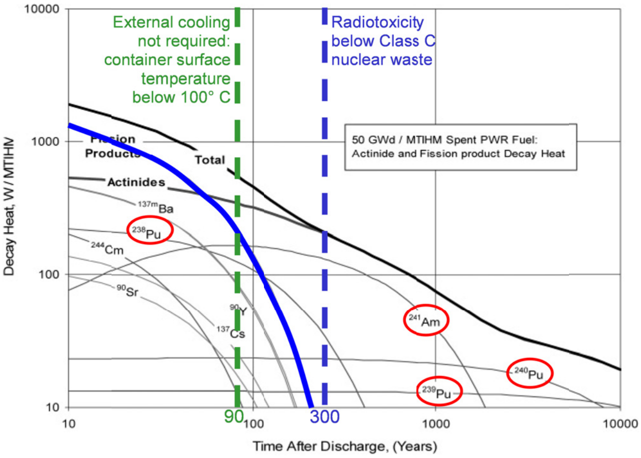

Figure 1: Dominant decay heat contributors in spent PWR fuel irradiated to 50 GWd/MTHM. [1] Goal is to eliminate components of the nuclear waste stream that account for the majority of the heat load and toxicity over the 300 to 10,000 year time frame. The isotopes circled in red are the major contributors 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 DRIVEN SYSTEMS

Accelerator Driven Systems (ADS) operate in a sub-critical reactor mode. This mode offers two significant advantages over critical reactors: greater flexibility with respect to fuel composition, and potentially enhanced safety. Accelerator driven systems are ideally suited to burning fuels which are problematic from the standpoint of critical reactor operation, namely, fuels that would degrade neutronic characteristics of the critical core to unacceptable levels due to small delayed neutron fractions and short neutron lifetimes, such as U-233 and minor actinide fuel. Additionally, ADS allows the use of non-fissile fuels (e.g. Th) without the incorporation of U or Pu into fresh fuel. The enhanced safety of ADS is due to the fact that once the accelerator is turned off, the system shuts down. If the margin to critical is sufficiently large, reactivity-induced transients can never result in a super-critical accident with potentially severe consequences. Power control in accelerator-driven systems is achieved through the control of the beam current, a feature which can compensate for reactivity loss due to fuel burn-up.

### *Waste Transmutation*

To date no country employs a fuel cycle that destroys the minor actinides (MA) present in used LWR fuel. Waste transmutation of the minor actinides requires a significant number of neutrons with energies greater than 1 MeV. A fast-neutron spectrum can be produced by a high-energy proton beam generating spallation neutrons. These spallation neutrons can then drive a subcritical core to transmute the minor actinides. Unlike critical fast reactors which generally incorporate uranium or thorium in the fuel for safe operation, ADS can potentially operate on a pure MA feed stream, meaning a smaller number of ADS can be deployed to burn a fixed amount of minor actinides. ADS can recycle the MA multiple times until it is completely fissioned. The only actinide waste stream from these systems would derive from the recycling residuals, which could yield a significant reduction (by a factor of hundreds) in the amount of actinide waste per kW-hr of electricity generated, as compared to a once-through fuel cycle. Because accelerator driven systems do not require fuels containing uranium or thorium, they are more efficient at destroying MA waste than critical reactors, based on grams of minor actinides fissioned per MW-hr of energy generated.

As indicated in the introduction, transmuting one minor actinide in particular, americium, can significantly decrease the amount of decay heat in a repository, thus decreasing the overall costs. Transmuting the long-lived Am isotopes to shorter-lived fission products enables the end-products to be disposed in short-term repositories. The Am feedstock is assumed to be from spent fuel that has set for 50 years after removal from the reactor. Accelerator-driven systems can probably operate on a pure Am feed stream in the equilibrium cycle. At 50 years, 97% of the Pu-241 has decayed to Am-241. The remaining un-decayed 3% of 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.

For example, the current US LWR fleet generates about 3 MT/yr Am after 40 yrs cooling (~1 MT/yr after 7 years). Burning 3 MT/yr Am generates 8 GW of fission heat (about 3% of US nuclear fleet size). After 40 year cooling, three high-powered accelerators can burn the Am generated by the current US fleet. If younger fuel is processed then after 7 year cooling only 1 system is required. We have proposed an approach, sub-critical minor actinide burner, SMART, built on an Am burner approach, shown in Fig. 2.

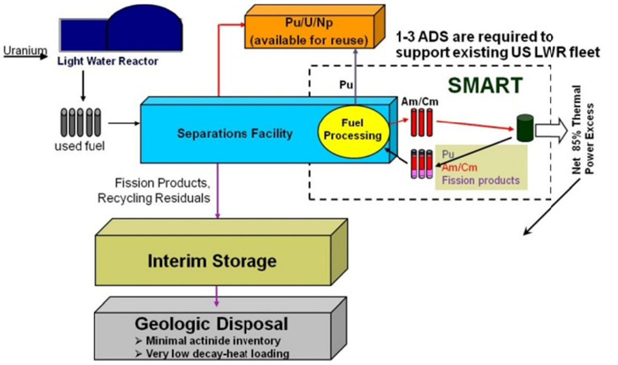

Figure 2: SMART supports LWR economy and preserves U, Pu, & Np as a future energy resource. Generally this thermal power can be used to offset the facility capital and operational costs. If the process heat is used for unbuffered electrical production for the grid, then the required accelerator reliability would substantially increase accelerator cost. Efficient use of this energy would benefit from advanced research based in biomass conversion or $H_2$ advanced cycles.

A facility for transmutation of waste would also generate substantial power; the process heat could be utilized to produce another form of energy (e.g. biofuels) or could be used to generate electrical power. The expected cost of an accelerator based transmuter system compared to a reactor is expected to be <30%. Under the worst case scenario of not using the process heat, the incremental cost to the present electrical rate based on the additional non-power production transmuter plants is 2 to 5 percent not including reprocessing costs. Not including reprocessing costs is fair if the fuel cycle going forward will be a closed cycle and reprocessing will be an inherent feature.

### *Power Production*

Many proposed ADS concepts with the goal of power production [2] utilize thorium-based fuel to take advantage of some of Th benefits of greater natural abundance (3-4 times greater than uranium), proliferation resistance, and significantly reduced production of transuranics that are a major source of radiotoxicity and decay heat relative to uranium-based fuel. Both liquid and solid fuel blankets have been proposed. An ADS system based on Th fuel would not require incorporation of fissile material into fresh fuel, and could operate almost indefinitely in a closed fuel cycle.

A limited number of critical reactor concepts based on thorium have been designed and operated (e.g., the Molten Salt Reactor at ORNL, and the Light Water Breeder Reactor at Shippingport). Expanded use of thorium-based fuels is actively pursued in some countries with large reserves of thorium, principally India, Norway and China. These programs are investigating whether ADS can speed up the deployment of the U-233/Th fuel cycle by breeding U-233, which does not exist in nature.

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 is directly proportional to the source neutron production rate. The flexibility enabled by subcritical operation has several advantages:

» can drive systems 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, and

» 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.

### *Process Heat Utilization*

Converting the fission power into a useable energy source is highly advantageous for transmuters to help recover the facility capital and operating costs and essential for a facility designed for power production.

One option is to sell the excess power to the grid. Based on recent experience with superconducting accelerator technology, the design of highly fault-tolerant accelerators is a reasonable expectation. [4] Storing power with the use of power storage devices could provide the electricity to run through faults if they can store enough electricity 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. [5] 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.

## ACCELERATOR TECHNOLOGY

### *Accelerator Design*

Accelerator-based transmutation includes four major technology elements: accelerators, transmuters, and separations, fuels and waste forms, Shown in Fig. 3. This paper only covers the accelerator systems.

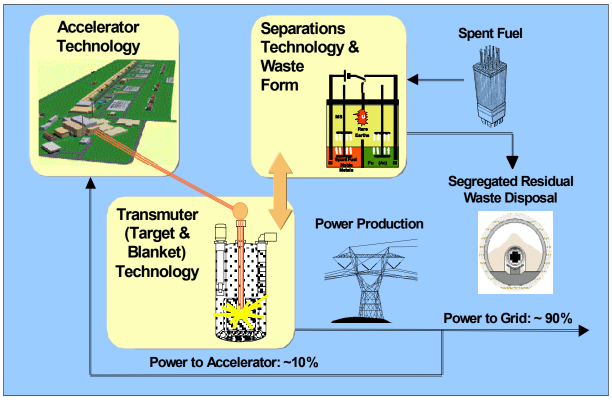

Figure 3: Major sub-systems of an ADS facility.

The power of the accelerator is determined by the design of the subcritical multiplier. For example, for a subcritical blanket fission power of 1 GW and with the multiplier $k_{eff}$ in a range of 0.95 to 0.98( $k_{eff}$ gives the neutron multiplication factor in a reactor; this factor is $k_{eff}/(1- k_{eff})$) will have a proton beam power ranging from 18 MW to 7 MW and a beam current swing of 12 mA to 5 mA, assuming a beam energy of 1.5 GeV. Either starting out with a lower $k_{eff}$ for safety or going to deeper burn and resulting in a lower $k_{eff}$ at cycle end, requires an increase in the accelerator current to maintain a constant neutron flux in the reactor. 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 resulting in cost inefficiency.

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 1000 MeV). 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. Based on present research, the maximum operational currents are 5 to 8 mA for cyclotrons and 50 to 75 mA for linacs. We are looking at accelerator systems that could drive several GW thermal power plants and have currents up to 40 mA. The accelerator technology covered in this article will be limited to linac systems.

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. With the consideration of multiple targets, multiple accelerators may provide system redundancy and improved reliability, but at added cost. Beam parameters consistent with the above operating numbers were demonstrated to be feasible under the Accelerator

Production of Tritium (APT) [6] program, as shown in Fig. 4.

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 1 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.

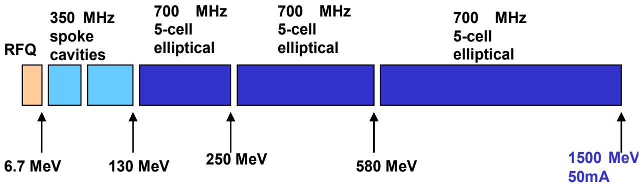

Figure 4: The accelerator preliminary design is based on the technologies developed for the APT program. The superconducting linac reduces cost and improves performance and reliability (i.e. beam continuity).

A superconducting-radiofrequency (SCRF) linac is typically chosen 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 SCRF cavities will have larger bore radius that relaxes alignment and steering tolerances, as well as reducing beam loss.

Alternative approaches to high proton beam power include synchrotron technology, which has the capability of achieving powers in excess of 1 MW, but is limited to pulsed operation at relatively low duty factor, and Fixed-Field Alternating Gradient (FFAG) accelerators that are actively studied at laboratories throughout the world. Synchrotrons and FFAGs have some similar intrinsic features, but the repetition rate for FFAGs can be much higher (albeit without the capability for true CW operation). While promising, FFAGs have yet to demonstrate high beam-power capability.

### *Accelerator Issues*

The major ADS related issues are:

» Multiplying assembly design
  - Neutronics analysis
  - Thermal-hydraulic analysis
  - Safety analysis
  - Fuels
  - Structural materials

» Coolant technologies (i.e. - lead-bismuth-eutectic, etc. )
  - Corrosion studies / oxygen control
  - Erosion studies
  - Safety assessment / polonium release

» Spallation target technologies
  - Window vs. windowless targets
  - Target material and coolant options beyond LBE

» Accelerator systems
  - Effect of transients on materials
  - Effect of transients on fuels
  - Quality of electrical power delivered to the grid
  - Periodic maintenance
  - High-power accelerator design
  - Reliability-Availability-Maintainability-Inspectability (RAMI) assessments

This paper only covers the accelerator beam trip requirements that follow from thermo-mechanical considerations of transients on the spallation target and subcritical assembly and, for power production applications, reliable electrical power delivery to the grid. The maximum number of allowed beam trips of a given duration depends on the design details, including the coolant parameters and characteristics, the coolant system design, the materials used, and the average power densities in the different ADS components.

In the last several years, more thorough and detailed beam trip requirement analyses have been performed based on transient analyses of ADS reactor system components. Three analyses in particular show reasonable agreement on the transient response and resulting beam trip requirements. A JAEA study [7] considered an 800 MWth subcritical reactor driven by a 30 MW proton beam. The analysis considered thermal shock and cycling on the beam window, reactor vessel, inner barrel and turbine system. The resulting beam trip rate limits are 25,000/yr for short beam interruptions (< 5 sec), 2500/yr for interruptions greater than 5 and less than 10 seconds, 250 per year for interruptions greater than 10 seconds and less than 5 minutes, and 50/year for interruptions greater than 5 minutes. A recent MYRRHA study [8] found similar results, yielding beam trip limits of 2500 trips/year for interruptions greater than 1 second and less than 10 seconds, 2500 trips/year for interruptions between 10 seconds and 5 minutes, and less than 25/year for interruptions greater than 5 minutes. These results include a factor of 10 safety margin. A U.S. study performed in 2001 [10] yielded beam trip limits of 1000 trips/year for interruptions longer than 0.3 sec but shorter than 100 sec, and 30 trips/year for interruptions longer than 100 seconds. It is worth emphasizing that these beam trip limits, derived from transient analyses of subcritical reactor components, are two orders of magnitude less stringent than typical values published previously [9]. For power generation applications, the beam trip rate requirements are more stringent, limited to only a few long unscheduled interruptions per year in order to meet reliability requirements set by the demands of commercial power production.

Additional safety-related requirements include safety-class beam shutdown capability, limitations on maximum beam current/power, rate of change of beam current, automatic closed-loop control of the current and the capability of controlled ramping up (or down) the beam power over seconds to minutes.

The ADS application has more stringent trip rate requirements than the historical high power proton accelerator experience base. However, it should be emphasized that present high power accelerators were not designed with a low trip rate requirement. In particular, accelerator facilities to serve a scientific research function do not typically invest in redundant hardware systems that would be required to achieve the high reliability performance expected for an industrial-scale installation. Nevertheless, experience at these facilities provides important guidance on the systems that require improvement in future ADS applications. Beam trip rates for the present operating high power proton accelerators (LANSCE, SNS, ISIS and PSI) are shown in Fig. 5 [10]. Total annual trip counts of order $10^4$ are typical, most of which last less than one minute. Present day trip frequencies with outages less than about 10 minutes are approaching recent ADS requirements. But factors of 10 to 100 reductions in the frequency of longer interruptions are needed to meet the latest ADS requirements.

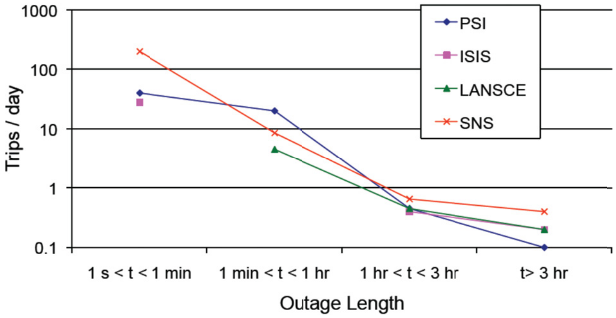

Figure 5: Beam trip frequency for operating high-power proton accelerators [10].

Detailed reliability analyses utilizing modern reliability engineering approaches have been performed [11]. The result of these studies suggest that reliability goals can be met with appropriately chosen redundancy, with adequate engineering margin, and with the incorporation of rapid fault-recovery algorithms made possible with an independently-phased superconducting linac architecture. The superconducting linac approach to production of high power beams has an inherent operational reliability advantage. Acceleration is provided by many independently-powered cavities, each of which provides only a small fraction of the total beam power. Failure of a single cavity (including its RF drive components) can be quickly "tuned around" by bringing on-line spares into operation (or adjusting already operating cavities), as has been demonstrated in practice in routine operation of the SNS [12]. The technique for SC cavity fault recovery at SNS is amenable to rapid (< 1 sec) implementation with specially designed control systems.

Extremely high-reliability has been achieved in large accelerator systems. The European Synchrotron Radiation Facility routinely achieves Mean-Time-Between-Failure of many days, and has recently operated for an entire month without a beam trip. The Advanced Photon Source completed 2009 with 63 beam trips recorded that year.

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