

ACCELERATOR-DRIVEN TRANSMUTATION TECHNOLOGY: A HIGH-TECH SOLUTION TO SOME NUCLEAR WASTE PROBLEMS

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ABSTRACT

This paper discusses current technical and non-technical issues regarding the innovative concept of using accelerator-driven transmutation processes for nuclear waste management. Two complex and related issues are addressed. First, the evolution of the current U.S. conceptual design is identified to indicate that there has been sufficient technological advancement with regard to a 1991 scientific peer review to warrant the advent of a large-scale national research and development program. Second, the economics and politics of the transmutation system are examined to identify non-technical barriers to the implementation of the program. Although a number of key challenges are identified in this paper, the benefits of the research and development effort and the potential paradigm shift in attitude toward resource stewardship could greatly enhance public confidence in nuclear waste management that will have rapid positive repercussions on nuclear technology research and commercial applications.

INTRODUCTION

The resolution of the nuclear waste problem is crucial to the continued or expanded use of nuclear electricity generation and other nuclear technologies. Any proposal for alternate approaches can easily capture the attention of scientific and public constituencies. In the United States, no significant progress has been made to establish broad-based political support for either a civilian high-level nuclear waste repository or a monitored retrievable storage facility. As an alternative to the stalemate over nuclear waste disposal, the development and implementation of transmutation technology could (1) make the contents of a repository more benign, and (2) significantly reduce the timeframe of concern thereby making it easier to provide confidence in the repository's ultimate safety.

Transmutation of waste has been historically viewed by nuclear engineers as a pipe dream technology that is too good to be true and probably too expensive to be feasible. However, both the technology and economics of accelerator-driven transmutation technology (ADTT) are showing signs of promise. The concept discussed in the present paper uses neutrons (that result from protons accelerated into spallation targets) to transmute the major very long-lived hazardous materials such as the radioactive isotopes of technetium, iodine, neptunium, plutonium, americium, and curium (i.e., the radioisotopes identified by the U.S. Department of Energy (DOE, 1998) as responsible for the highest long-term risk to people from a repository at Yucca Mountain, Nevada). Although not a new concept, ADTT led by a team at Los Alamos National Laboratory (LANL) has made some significant advances that are discussed in the present paper.

The major attributes of the ADTT concept are: (1) it addresses issues of waste management storage capacity by efficiently destroying transuranics (TRU), transmuted long-lived fission products to more benign or stable isotopes, and partitioning of all fission products for optimal disposition, (2) it is reactor-like in scale and function and has the potential to be economically viable by producing usable energy by destroying hazardous components of spent nuclear fuel, (3) its components are based on proven technology, and (4) the radiotoxicity of the residual material from a transmutation facility after 300 years is lower than direct-disposal of spent nuclear fuel after 100,000 years, thus impacting the local population's concern over long-term repository performance.

Transmutation is not a new technology, in fact, scientific knowledge of transmutation has been around since 1919 when Ernest Rutherford bombarded nitrogen with alpha particles and converted it to hydrogen and oxygen. In 1980, the DOE began to look at strategies for high-level radioactive waste management, including transmutation of waste to a more benign form, but selected mined geologic disposal as the approach for waste isolation in the U.S. (presuming reprocessing of spent nuclear fuel and waste form optimization would occur first).

STATS Committee Report

In June 1991, a 19-member interdisciplinary Separations Technology and Transmutation Systems (STATS) committee of the U.S. National Research Council was formed at the request of the U.S. Secretary of

Energy to evaluate the state of the art of separations and transmutation concepts. The committee's peer-reviewed findings and recommendations (STATS, 1996) were not encouraging. In essence, the committee felt that any transmutation process would not eliminate the need for a high-level radioactive waste repository and would be less economically attractive than the current once-through fuel cycle. STATS (1996) discusses the committee's concerns and their recommendation that any research undertaken by the U.S. should be modest.

In defense of the committee, they acknowledged that accelerator-driven transmutation systems were in a far less developed state than those of the light-water and liquid-metal fission reactors they evaluated. In fact, the system they reviewed was changed so significantly that the accelerator-driven transmutation concept was reviewed again in 1998 by the Nuclear Engineering Department of the Massachusetts Institute of Technology (MIT) that included some of the original STATS committee reviewers.

The ADTT project reviewed by the STATS committee had the aim of reducing TRU waste to such an extent that all waste containing residual TRU would be suitable for shallow land burial. The concepts they reviewed used a thermal neutron spectrum, fluid fuels or molten salt with dissolved radionuclides, and only aqueous separations technology. The current design has

evolved significantly with particular attention to the STATS (1996) concerns and recommendations. For example, the committee felt that long-lived fission products may represent an equally significant concern as that of TRU and that advances in reprocessing may allow highly efficient separation operations. Technical improvements to the ADTT conceptual design discussed in this paper include a fast neutron spectrum that allows destruction of long-lived fission products, liquid lead-bismuth eutectic (LBE) coolant and spallation target, solid fuel, and pyrochemical processing.

The current U.S. ADTT concept has evolved to the point that it is now based on maturing engineered designs as opposed to its 1991 predecessor. As currently envisioned, the facility would consist of three major components as illustrated in Figure 1: a high-power proton accelerator, a spent fuel partitioning and treatment system, and a subcritical reactor containing the target and blanket system. There is optimism among eight national laboratories and many at research universities that the interfacing of the ADTT's complex engineered components is achievable. Excerpts, taken by Schriber (1998), from the 1998 MIT review are summarized as follows:

- No insurmountable issues or show stoppers were identified. "While the proposed technologies are in several instances extrapolations of existing

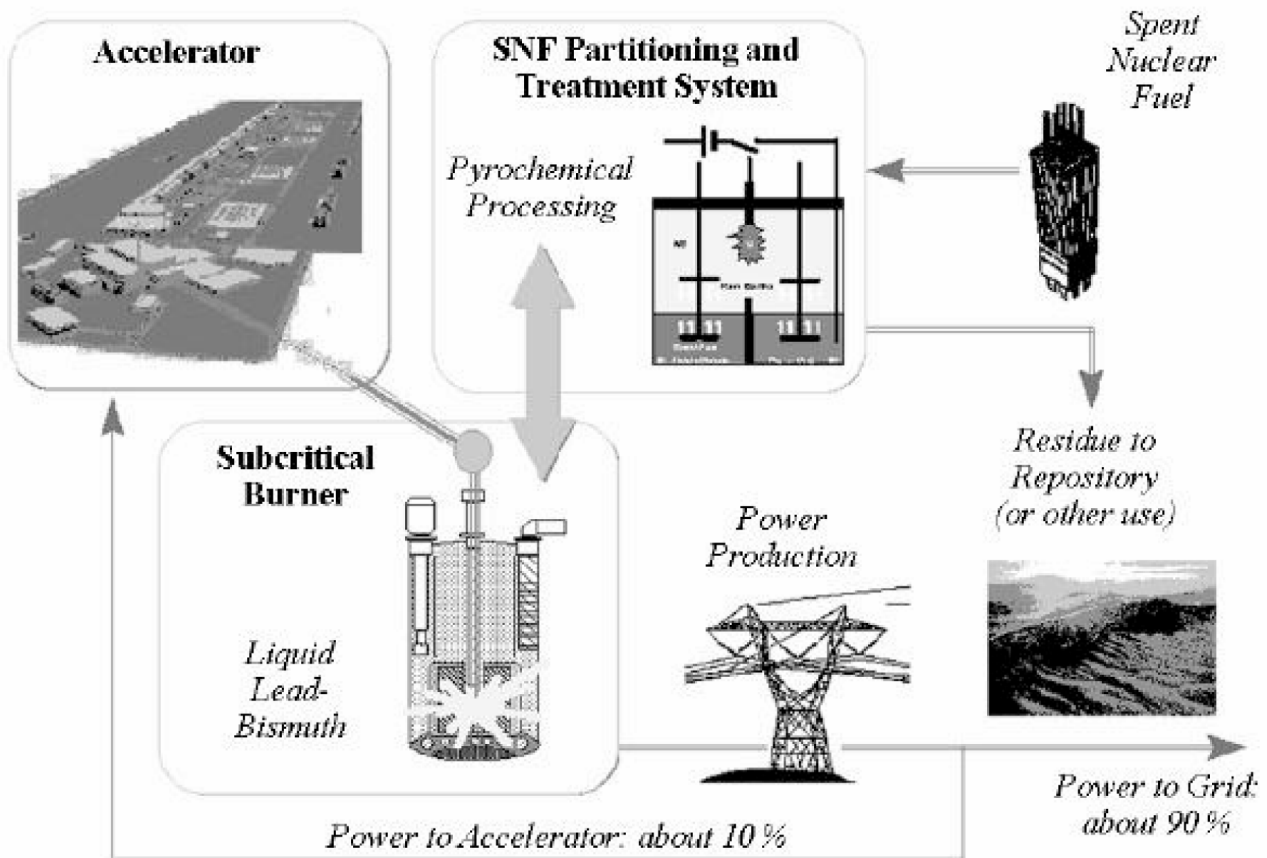


FIGURE 1. Major Components of ADTT System

experience to untested conditions, they represent reasonable targets for development over the next 5 to 10 years.”

- “While we do not judge the merits of the [ADTT project] as an option to improve the management of nuclear wastes, we acknowledge that it has the potential to provide added flexibility to the design of the high level waste repository and to reduce the uncertainties about its performance.”
- The main technologies to be developed are all worthwhile technologies for other applications beside the transmutation of wastes. “We see the spin-offs from the development efforts as equally important reasons for the undertaking of the proposed development program in the next few years.”
- There are several commendable attributes, but also questions and caveats to be addressed. “The R&D [Research and Development] program is well designed to address these concerns.”

The U.S. Congress did not provide the necessary funding to support the required research in 1998 nor 1999.

A NEW ROADMAP FOR DEVELOPMENT

In 1998, the U.S. Congress mandated that a roadmap for the development of accelerator transmutation of waste be prepared. Specifically, the U.S. Department of Energy convened a steering committee to address (1) the scope of the congressional mandate to identify the technical issues that must be resolved, (2) a proposed time schedule and program to resolve these issues, and (3) the estimated cost of such a program. In addition, the report was to include an assessment of the institutional challenges of this program, the impact this technology could have on the civilian spent nuclear fuel program, areas of development which could have benefits to other ongoing programs, and the estimated capital and operational life cycle costs to treat civilian spent nuclear fuel.

The steering committee’s report (DOE, 1999) was published in October 1999 in which they estimate a six-year R&D program with a total cost of about \$281 million would be necessary to complete a research, development, and demonstration plan. A near full-scale prototype to demonstrate the ability to deploy an ADTT system would require approximately 20 years and cost about \$10 billion. Furthermore, because the issues addressed by ADTT are global (such as current and future energy needs and options, weapons nonproliferation, and management of radioactive waste), the committee recommended international collaboration.

The principal technical issues to be resolved were identified in DOE (1999) as:

- Lifetimes of proposed materials and components in the radiation, thermal, and chemical environments of an ADTT system
- Operational reliability and availability of an ADTT system

- Operational safety of an ADTT system consistent with regulatory requirements
- Degree of partitioning separation achievable for uranium, transuranics, and long-lived fission product elements from discharged commercial reactor fuel and spent ADTT system fuel assemblies
- Quantification of long-lived radioactivity generated by an ADTT system including spallation products, and the implications for waste streams and waste forms.

The following subsections summarize the findings of the DOE (1999) roadmap that includes four Technical Working Groups established for Accelerator Technology, Target-Blanket Technology, Separations and Waste Forms Technology, and Systems Scenarios and Integration. In addition, a report evaluating the impact of ADTT on the Performance Assessment of a geological repository and a report on the estimated cost of the technology comprise the supporting documents of the roadmap. The integrated schedule for a rapid deployment scenario is shown in Figure 2.

Accelerator Technology

The design requirements developed by the Accelerator Technical Working Group (Lawrence et al., 1999) assumed eight Subcritical Burners driven by two high-power proton linear accelerators. The plant would produce 2555 M_we of electricity of which 380 M_we would be used by the plant and about 2175 M_we would be exported electric power.

During the six-year R&D program, DOE (1999) would focus accelerator work on:

- Addressing the beam reliability issue through system analysis and component testing
- Developing and testing key components expected to have a high impact on the plant and demonstration machine designs and that address performance requirements
- Determining a pre-conceptual design for a demonstration accelerator.

Target-Blanket Technology

According to the Target and Blanket Technical Working Group (Venneri et al., 1999), the target and blanket system consists of a spallation target producing the high intensity neutron source, and a subcritical blanket (analogous to a reactor core) surrounding the target and containing the TRU and long-lived fission products to be transmuted. However, unlike reactor systems, a subcritical system relies on an external source of neutrons meaning, in principle, that any isotope can be destroyed with little regard for their neutronic behavior.

DOE (1999) identifies ten R&D issues for this component of the program:

- Upgrade or development of computer simulation tools

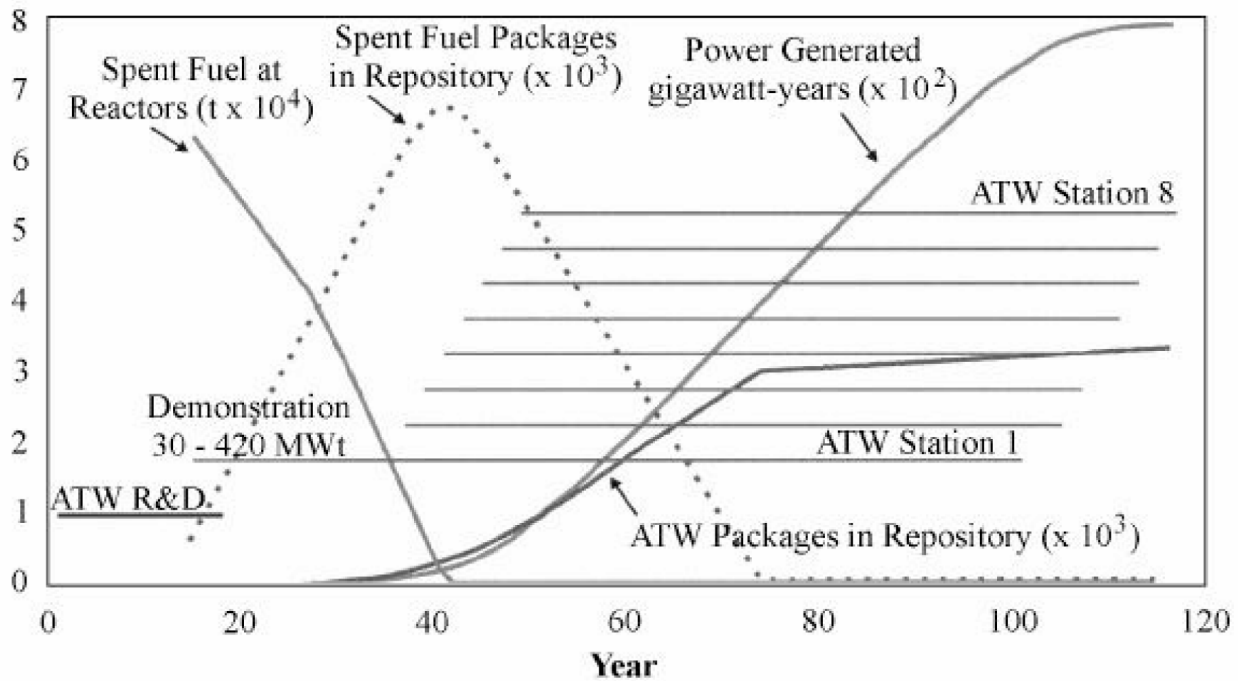


FIGURE 2. Integrated Schedule for Rapid Deployment of a U.S. ADTT Concept

- Calculations to determine blanket performance
- Assessment of the feasibility of processing Tc and I into transmutation elements
- Development of system controls: (1) control of the degree of subcriticality, (2) control of burnup reactivity, (3) control of system dynamic behavior, (4) formulation of monitoring and control, and (5) core design database and simulation validation
- System optimization for safety and economics
- Evaluation of target window and structure design concepts
- Study of lead-bismuth eutectic chemistry control techniques, materials compatibility, and heat transfer and flow characteristics
- Evaluation of preliminary LBE-cooled system designs
- Development of fuel fabrication and determination of fuel properties
- Demonstration of irradiated fuel behavior and performance.

Separations and Waste Forms Technology

According to the Separations Technologies and Waste Forms Technical Working Group (Laidler et al., 1999), removal of uranium from commercial spent nuclear fuel should be done in such a way that the uranium could be disposed as a non-TRU, Class C low-level waste. A goal of better than 99.9 percent recovery of uranium in spent fuel has been set. In addition, because of the importance of TRU elements to nonproliferation

objectives and repository performance, Laidler et al. (1999) also set a recovery goal of better than 99.9 percent for TRU as well. Finally, the recovery goal for Tc and I has been set at greater than 95 percent.

The baseline concept combines aqueous and pyrochemical processes for efficient separation of U, Tc, I, and TRU from light-water reactor fuel. Although alternative options need to be addressed, the following issues are focused on the current baseline concept.

The key technical issues identified by DOE (1999) with the initial PUREX-based aqueous processing step are:

- High recovery (99.9%) of the actinide elements
- Selection of a suitable Pu reductant that does not significantly alter the composition of the actinide product stream
- Separation and recovery of Np in high yield (99.9%)
- Separation and recovery of Tc and I
- Demonstration that a Class C low-level uranium waste can be produced at high yield
- Ability to calcine the actinides in the aqueous product stream that permits their transfer to the electrometallurgical (EM) process
- (In case of an aqueous TRUEX-based step), identification of the optimum combination of stripping and wash reagents in order to minimize the generation of secondary waste.

Most of the technology required for the subsequent electrometallurgical steps has already been demonstrated. The major technical challenge is to scale up the process. The key technical issues identified by DOE (1999) are:

- Production of a non-TRU uranium stream

- Separation of Tc from zircaloy cladding hulls
- Separation and isolation of I
- Scale-up of all process steps and equipment.

The key technical issues for the processing of spent ADTT fuel are similar to the above for spent nuclear fuel, but also include:

- Spent fuel chopping/grinding system reliability at high throughput
- Optimization of the interface between the off-gas and chloride volatility systems
- Chemical and metallurgical behavior of Tc and the actinides in the chloride volatility systems
- Am, Cm, and Tc electrochemical behavior in the actinide electrowinning process
- The development of pilot- to demonstration-scale iodine separation processes
- The compatibility of process residues with the desired waste forms.

The key technical issues with the final ceramic waste form identified by DOE (1999) are:

- The definition of waste stream composition
- The development of full-scale processing methods
- The development of waste minimization and salt recycling technology.

The key technical issues with the final metal waste form identified by DOE (1999) are:

- The definition of waste stream composition
- The characterization and qualification of zirconium-8 stainless steel waste form
- The development of full-scale processing methods for salt removal and casting
- The definition of process residuals that must go to the waste form, such as residual Tc.

Systems Scenarios and Integration

The Systems Scenarios and Integration Technical Working Group (Hill et al., 1999) was responsible for evaluating the overall role of the ADTT concept in energy and waste reduction scenarios and the work of the above three technical working groups to ensure consistency of technical assumptions, schedules, and key system parameters. The group acknowledges that several technology options in ADTT exist; however, the reference design discussed in the present paper was established to ensure a consistent discussion of technical and life cycle cost issues. A key recommendation is to conduct initial studies to confirm technology choices and optimize the reference design. Hill et al. (1999) recommend that initial R&D establish the performance capabilities of key technologies such as lead-bismuth eutectic, acquire necessary material properties, nuclear and thermal-hydraulic data, and validate tools such as simulation codes that will be used to optimize and establish the safety envelope for the design.

Impact on the Performance Assessment of a Geological Repository

Duguid et al. (1999) conducted a performance assessment of the potential impact the ADTT waste form may have on a geological repository in the U.S. They used the DOE (1998) model for the Yucca Mountain, Nevada, Viability Assessment to establish a consistent reference line.

The assumed ADTT efficiencies were 99.9 percent destruction of transuranics, 99.9 percent recovery of uranium, and 95 percent transmutation of technetium-99 and iodine-131. With these assumptions, the assessment indicates that partitioning and transmutation of spent fuel could reduce its contribution to the peak effective dose from a repository at Yucca Mountain, Nevada, by a factor of about 10,000 for the first 10,000 years (a calculated dose of around 10^{-11} Sv per year). The peak dose over a 1 million year period would be reduced from on the order of 10^{-3} Sv per year to less than on the order of 10^{-6} Sv per year. The inventory of plutonium and other transuranics chemically separated from spent fuel would be reduced by a factor of 1000 to less than 1 ton at the end of ADTT operations.

The vastly reduced inventory will also mitigate the effects of inadvertent intrusion scenarios assuming reduced concentration and optimization of final waste form.

Estimated Cost

Smith et al. (1999) estimated the life-cycle costs associated with the implementation of a national ADTT development and deployment program. They estimate the initial R&D or six-year effort to cost between \$281 million (see Table 1) and \$723 million based on the aggressiveness of the effort, i.e., a modest science-based program versus a rapid deployment-based program. The estimated total cost under a rapid deployment scenario (see Table 2) is \$2 billion over about a 20-year period for R&D leading to the design of a demonstration facility. The demonstration facility is estimated to cost \$9 billion. (Note: all dollar amounts are given in constant 1999 dollars.) The current deployment scenario consists of eight ADTT installations, each generating approximately 2200 MW of net electricity. Capital costs for each installation would be from \$6 to 7 billion. Annual operating costs including debt servicing would be about \$500 million. Assuming electricity revenue rate of \$0.043 per kW-hour produces an annual revenue of about \$800 million. This indicates that most if not all of the capital, operational, and decommissioning costs may be offset by the sale of electricity.

These estimates show that not only could an ADTT system greatly reduce the hazard of a nuclear waste repository destined for Yucca Mountain, Nevada, it could also more than pay for itself and provide a valuable source

of regional power. In other words, ADTT may prove itself worthy to the skeptical nuclear engineering community.

TABLE 1. Estimated Cost for a Six-Year Science-based R&D Program (DOE, 1999)

Program Component	Six-Year R&D Cost
System Studies	18.0
Accelerator Development	58.0
Separations and Waste Forms	55.5
Target and Blanket	123.5
Program Management	26.0
Total	281.0

TABLE 2. Estimated Cost for a Rapid Deployment R&D Program and Demonstration Facility (DOE, 1999)

Program Element	R&D	Demonstration
Accelerators	0.17	3.0
Transmuters	1.0	2.0
Separations	0.5	2.0
ATW Fuel Fabrication	0.0	0.6
Site Support	0.0	1.0
Retrieval/Transportation/Disposal	0.0	0.1
Integration	0.07	1.0
Total	2.0	9.0

NON-TECHNICAL BARRIERS

The Systems Scenarios and Integration Technical Working Group (Hill et al., 1999) identified three broad categories of non-technical barriers: institutional capabilities, public acceptance, and regulatory issues. Institutional capabilities refers to the ability of the federal government to provide the organizational and financial resources required to carry out the complex technical program over a period of several decades.

Implementation of any radioactive waste management option will face difficult public and political issues. ADTT systems will likely face many of the same risk perception and communication issues plaguing the nuclear industry, such as moral relevance, risk equity, and radiation phobia. The implementation of an ADTT research, development, and deployment program in the U.S. should work closely with community groups and local representatives to ensure grass roots support is solid. For example, the problems created by siting the Yucca Mountain project in a hostile public environment should be avoided with the siting of any ADTT R&D and commercial facilities.

Funding for the R&D and demonstration components of the project were estimated by Smith et al. (1999) to cost about \$10 billion over about 30 years with the potential commercial application showing profit capability. It appears that the U.S. Nuclear Waste Fund (estimated at about \$7 billion) established by the Nuclear Waste Policy Act of 1982 (P.L. 97-425, 1983) does not

allow for its use toward research and development activities not directly tied to the development of the geologic repository. Therefore, appropriations for ADTT R&D and demonstration activities must be lobbied annually against many other federal programs.

The regulatory requirements imposed on the ADTT demonstration facility and commercial systems may pose a significant challenge similar to any nuclear facility in the U.S. These will include requirements of the U.S. Department of Energy, the U.S. Nuclear Regulatory Commission, and the U.S. Environmental Protection Agency. For example, under the U.S. National Environmental Policy Act, an environmental impact statement with public involvement will have to be produced for the demonstration facility.

BENEFITS

Although the previous sections highlight a number of challenges to the development and deployment of an ADTT system, the potential benefits are vast. Hill et al. (1999) and DOE (1999) name the following direct benefits:

- Improved isolation safety due to smaller inventory
- Improved confidence in repository performance due to a reduced time frame and optimized waste form
- Reduced waste volume reduces waste capacity requirement
- Elimination of the criticality issue in a repository
- Improved repository design flexibility
- Proliferation and material diversion risk reduction
- Improved resource stewardship from energy production.

Potential indirect benefits include:

- U.S. nuclear science and technology leadership
- New frontiers of science will be explored
- Medical, research, and industrial source of isotope production
- Back-up tritium production capability
- Intense neutron source for basic research needs
- International collaboration
- Maintenance of core competency in nuclear technology.

CONCLUSIONS

The United States may be embarking on a national program to develop accelerator-driven transmutation technology at its national laboratories as a component of radioactive waste management. The success of such an endeavor will have a greater impact on the State of Nevada than on any other state because it will greatly mitigate the hazards of the proposed Yucca Mountain repository. Nevadans with diverse backgrounds and viewpoints have shown a remarkable interest in this

technology. Although a number of key challenges are identified, the benefits of the research and development effort and the potential paradigm shift in attitude toward resource stewardship could greatly enhance public confidence in nuclear waste management that will have rapid positive repercussions on nuclear technology research and commercial applications.

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