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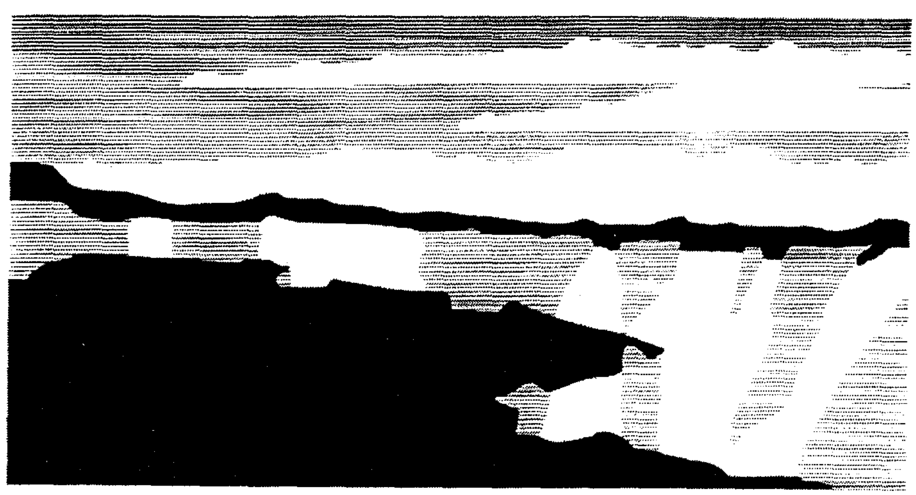

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Accelerator-Driven Transmutation of High-Level Waste from the Defense and Commercial Sectors

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Abstract

This is the final report of a three-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). The major goal has been to develop accelerator transmutation of waste (ATW) system designs that will thoroughly and rapidly transmute nuclear waste, including plutonium from dismantled weapons and spent reactor fuel, while generating useful electrical power and without producing a long-lived radioactive waste stream. We have identified and quantified the unique qualities of subcritical nuclear systems and their capabilities in bringing about the complete destruction of plutonium. Although the subcritical systems involved in our most effective designs radically depart from traditional nuclear reactor concepts, they are based on extrapolations of existing technologies. Overall, care was taken to retain the highly desired features that nuclear technology has developed over the years within a conservative design envelope. We believe that the ATW systems designed in this project will enable almost complete destruction of nuclear waste (conversion to stable species) at a faster rate and without many of the safety concerns associated with the possible reactor approaches.

1. Background and Research Objectives

Nuclear systems under study in the Los Alamos Accelerator-Driven Transmutation Technology (ADTT) Program have been conceived for destruction of nuclear spent fuel and

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weapons-return plutonium, as well as for production of nuclear energy from the thorium cycle, without a long-lived radioactive waste stream. The subcritical systems proposed represent a radical departure from traditional nuclear concepts (reactors), yet the actual implementation of ADTT systems is based on extrapolations of existing technology. These systems strive to keep the best that the nuclear technology has developed over the years, within a sensible conservative design envelope, and eventually to offer a safer and more environmentally sound approach to nuclear power.

Concerns about waste from the defense and commercial nuclear sectors has grown to such an extent in recent years that it now dominates the nuclear enterprise. The emphasis in the nuclear technology field has moved from its earlier reactor-design focus into clean-up of defense production sites and a resolution of the commercial nuclear waste problem. The development of cleaner and safer systems for nuclear energy generation is almost at a standstill because of growing international concerns about the waste issues. The predominant approach to this problem for the past thirty years has been the geologic storage of waste, whether it be from the defense or the commercial sector. Geologic storage offers the prospect of confining nuclear waste by the confinement features of a stable geologic structure rather than relying on long-term containment of the waste in man-made containers. In addition the waste is made much less accessible by its placement deep underground. Therefore, many countries are providing significant funding for the development and siting of geologic waste-storage facilities. While a number of sites might be under study in a given country, the intent is to provide a single site capable of confining the high level waste.

It has become increasingly difficult to convince a community to become host to a nation's single site for storage of waste that many consider to be the nation's most dangerous. The fact that the waste remains dangerous for many tens of thousands of years exacerbates these concerns. The concern that such repositories can become mines for plutonium has become of even greater concern as the United States has made it known that dangerous nuclear weapons can be made from commercial plutonium. The natural transformation of commercial plutonium into excellent weapons plutonium by radioactive decay means that eventually many thousands of tons of weapons plutonium will be stored at many sites around the world. Some are becoming concerned about the possibility of natural or induced super criticality of fissile material stored underground. As a consequence of these and other concerns remaining to be resolved about geologic storage, no nation is expected to begin emplacement of high-level waste in a geologic repository before the year 2010, and the ultimate viability of the geologic storage concept remains to be demonstrated.

In the United States, commercial nuclear waste is accumulating at reactor sites, and the defense site clean-up effort is struggling to understand what will happen to the plutonium and other high-level waste which will be gathered together after the clean-up has been completed. The Los Alamos National Laboratory, along with a rapidly developing national and international community, has been studying ADTT as a possible means of destruction of this nuclear waste and of generating nuclear power by systems which do not generate the most dangerous components of this waste and which concurrently destroy their own waste. If the full capability of the ADTT systems can be realized at acceptable cost, large-scale geologic storage of defense and commercial waste would not be required.

2. Importance to LANL's Science and Technology Base and National R&D Needs

This project supports the Laboratory's core competencies in nuclear science, plasmas, and beams as well as analysis and assessment. It enhances LANL's ability to respond to initiatives involving accelerator-driven transmutation technologies.

3. Scientific Approach and Results to Date

The general features of our proposed ADTT system are shown in Figure 1. When an intense medium-energy proton beam (10-20 mA at 500-1000 MeV) interacts with an appropriate target of flowing lead-bismuth eutectic, neutrons are produced that are thermalized and multiply in a surrounding moderating blanket containing fissile and fertile material in liquid (molten salt) form. A neutron production of over 30 neutrons per incident particle per GeV is possible from such targets, plus further multiplication of these neutrons by a factor of 20 to 30 in the blanket. Electricity is generated from the heat released by the fission processes, and a part (between 10 and 15 percent) is used to drive the accelerator. The use of molten salt (at 650-720 °C) in the moderated blanket allows high thermal-to-electrical efficiencies, exceeding those of current water-cooled reactors. As a result, a large excess of electric power will be available to the electric power grid at rates that could be competitive with standard nuclear power costs.

The base-design system is driven by a 15-mA beam of 800-MeV protons and operates at 500 MW with an effective neutron multiplication factor of 0.95. The subcritical margin is sufficient to insure subcriticality in all conditions. The low-power density of the system allows for an inherently safe design, where the after-heat cooling can be handled by purely passive means in case of emergencies. By completely consuming the nuclear fuel and the most troublesome fission products, this system would obviate the need for transport and geological storage of the long-lived actinide species and fission product waste.

Operation of the system in a driven subcritical mode eliminates the possibility of a criticality accident and provides quick and absolute shutdown by simply turning off the accelerator. On a more fundamental level, the accelerator combines the traditional reactor control functions of control rods, burnable poison and chemical shim in one streamlined mechanism which is flux- and burnup-independent. Coupled with the features of a liquid fuel system, this translates in the full utilization of the fuel without actinide extraction and excess breeding. Through the use of the accelerator drive and a fuel cleanup cycle, the fuel can be utilized completely. Additionally, the extra neutrons provided by the accelerator can be applied to the transmutation of long-lived fission products. The low-level and short-lived radioactive waste remnants would be placed in near-surface managed storage at the power plant site itself.

To review, a system for destroying weapons Pu and commercial Pu (spent fuel) has been conceptualized. It integrates the basic concept of the Pu subcritical burner (continuous Pu feed, long cycles before fission product removal) with the front-end (fuel preparation) and the back-end (fuel cleanup) developed by a parallel LDRD effort in separations. In the context of the accelerator transmutation of waste/accelerator-based conversion (ATW/ABC) system, we have defined four basic design choices in the target/blanket area.

3.1. Liquid Fuel (Molten Salt)

The molten salt is a LiF-BeF_2 eutectic that melts at about 450 °C and operates at between 650 and 720 °C. Almost all elements react as fluorides and can be dissolved in small but adequate amounts into the carrier salt for the transmutation and fission requirements. It also has a low vapor pressure at high temperature, which is a major safety advantage allowing operation without a pressure vessel, which would be required for a higher vapor pressure medium such as water. Also, the salt is non reactive with air, nitrogen, and concrete, in contrast with the liquid sodium coolant on which fast reactor technology is now based.

Molten salts as reactor fuels and as coolants have been under study and development for over 40 years, and their chemical, physics, and irradiation properties are excellent. The Molten-Salt Reactor Experiment (MSRE) at the Oak Ridge National Laboratory, which was shut down in 1970 after about five years of very successful operation, contributed significantly to molten-salt reactor technology.

The high operating temperature allows a thermal-to-electric efficiency that might be as high as 35 to 40 percent. Safety concerns also require the use of liquid fuels that can reach high operating temperatures at low pressure. Molten fluoride salts can operate at temperatures well above 600 °C at near-atmospheric pressure. This allows for the economical construction of rather large, low-pressure vessels, thereby reducing to extreme unlikelihood the event of a salt spill.

The substantially more centrally peaked flux of accelerator-driven systems, in contrast to that present in nuclear reactors, also favors the use of liquid fuels, where the fuel is uniform and all potentially unstable build ups of poisons or excess fissile material in special locations are completely eliminated.

The use of homogeneous molten-salt fuel, as opposed to solid fuels or liquid slurries, allows the adoption of straightforward on-line fuel preparation (front-end) and cleanup (back-end) processes. Because no fuel partitioning and refabrication is involved with molten salt use, these processes, under development at Los Alamos, do not introduce extraneous waste streams and have substantial proliferation and diversion barriers.

Molten-salt fuels also allow the attainment of substantially more powerful "inherently safe" units than possible with solid fuels. The power level per unit is driven by the criterion that the unit should be passively safe in the event of a coolant failure, without need for salt drainage and removal. Contrary to solid-fuel systems, molten-salt systems in fact have a heat source that is distributed throughout the large-volume vessel. In ADTT molten-salt systems, two-thirds of the fuel is in heat exchangers and peripheral ducts and plenums at any given time and therefore any residual decay heat can be removed a lot faster than the case of solid-fuel systems, where the heat source is concentrated at the center of the core, where it is most difficult to remove when active cooling systems fail. The natural convection of the liquid fuel and the on-line removal of the fission products throughout operation also help considerably. The 500 MW level per base unit is a very conservative figure that might be the size of a first demonstration-type unit. Passively safe modules sized at 1000 MW were shown to be practical. A much larger unit (3000 MW) would require some means of draining the salt out of the blanket into a special passively cooled reservoir in the event of a coolant failure. Drainage in this case could still be obtained in a passive way through the use of melt plugs. Such a unit would have a power rating of 1200 MW.

3.2. Graphite Moderator/Reflector

Graphite was chosen as the moderator because of its high-temperature compatibility with the liquid fuel chosen, which is molten fluoride salt. In order for the graphite to last throughout the life of the plant, the power density in the blanket is limited to 5-10 W/cm^3 in the moderated region. Because the fuel is kept clean by on-line processes, even at such low power densities the neutron flux is anticipated to be in the range 10^{14} to 10^{15} n/cm^{2}s, depending on whether a thorium-bearing or a pure actinide fuel is used. The amount of graphite present in the system is kept to a minimum to limit the waste stream and to achieve an optimum neutron spectrum for the destruction of plutonium.

3.3. High-Current Linear Accelerators

The high-power accelerator technology required for ADTT has been under continuous development for the past three decades at Los Alamos. The average power needed for the very largest of systems we propose (6 x 500 MW), requires an accelerator average power of some tens of megawatts (100 mA beam). This can only be achieved with a linear accelerator (linac). However, the world's highest-power operational linear proton accelerator at LANSCE, the Los Alamos Neutron Science Center, operates at around one megawatt (1.25 mA at 800 MeV). At first this seems like a very large extrapolation, but it is not nearly so bad as it appears. With the LANSCE LINAC, only every fourth bucket is filled with beam, and so filling every bucket (if we use funneling) immediately gives a factor of four in average power. Also, the LANSCE LINAC is a pulsed machine operating at 10 percent duty factor, and going to 100 percent duty factor gives an additional factor of 10. The charge in each microbunch can be increased by about a factor of four and still stay well within the stable space-charge regime. Therefore, an improvement by a factor of 160 (4x10x4) is possible by the simple extension of proven technologies, so that up to 150 mA, 1000 MeV LINACs can be built based on current technology.

Experience with the use of an accelerator in an ADTT application can be readily gained using the existing LANSCE accelerator. With its present beam capabilities, it is capable of reliably providing up to 1 mA of 800-MeV proton beam to a scaled-down target blanket similar to the one described here. Thus, LANSCE could drive a deeply subcritical experimental facility with a neutron multiplication of five to generate a fission power level of about 5 MW, a sufficient level for a thorough testing of the ADTT concept.

3.4. Liquid-Lead Target Technology

The function of a target in ADTT systems is the conversion of high-energy (greater than 500 MeV) protons to lower energy (less than 20 MeV) neutrons and the delivery of these neutrons to the blanket in a useful manner. The conversion is achieved through the process of spallation, in which neutrons and other charged particles are stripped off the target nuclei. Eventually these neutrons will escape and propagate into the surrounding multiplying blanket. The ideal target should be simple in construction and operation, able to produce neutrons efficiently, and to leak them to the surrounding blanket with minimal absorption losses.

At the high power densities generated by the beam currents proposed for ADTT systems, liquid-metal targets offer important advantages in practical implementations over solid targets. Besides the obvious ease of heat removal from liquid targets, there are no structural damage limits to the life of the target material, and the use of liquid targets would allow the primary target material to be reused beyond the structural life of the container material. This minimizes residual waste and maintenance time. High atomic weight, low-melting-point metals or alloys are needed. Among the low-melting-point heavy metals, lead and bismuth have very low neutron-absorption cross sections and therefore can be arranged in simple monolithic configurations. The lead-bismuth eutectic (LBE) has a lower melting point and operating temperature than either lead or bismuth and therefore is favored in terms of structural considerations.

The liquid lead (or LBE) target used for the ADTT design is inserted vertically into the blanket from the top. The reasons for this choice are simple. A horizontal target would require very heavy shielding against forward directed high energy neutrons. It would also introduce the risk of a radioactive molten salt spill from a side penetration into the blanket. Vertical insertion of the target from the bottom would also introduce penetrations in the bottom part of the blanket or the presence of permanent metal structures in the center of the blanket where neutron damage is most severe. In the vertical insertion from the top, a window is placed upstream of the target along the beam path and it is of traditional design. Past the window, the proton beam enters a hydrostatic helium plenum which keeps the lead surface down in the bottom of the target. There would be no hot window between the beam and the lead. This configuration avoids the most serious cooling problems of a hot window at the center of the blanket. The liquid lead flows down through an annulus into the target volume receiving the beam directly on its free surface and then flows up on a concentric annulus.

The target is contained within a totally enclosed structure, bolted on the lid of the blanket and capable of being rapidly inserted and extracted from the blanket. Due to radiation damage, it is anticipated that the structure of the target most exposed to the neutron flux will have to be replaced as often as every one to two years. The liquid lead will last through the life of the plant and can be recycled beyond into the next ADTT system.

3.5. Conclusion

We have developed the technical approach and designed ADTT systems that should be capable of more than 99 percent destruction of weapons Pu and spent reactor fuel. We have pointed out the unique qualities of subcritical systems and their capabilities in bringing about the complete destruction of plutonium. The design process has included selection of the main components of the ADTT systems. The proposed subcritical systems represent a radical departure from traditional nuclear concepts (reactors), yet the actual implementation of ADTT systems is based on modest extrapolations of existing technology. Overall, care has been given to retain the best features that the nuclear technology has developed over the years, within a sensible conservative design envelope, and eventually to offer a safer and more environmentally sound approach to nuclear power.

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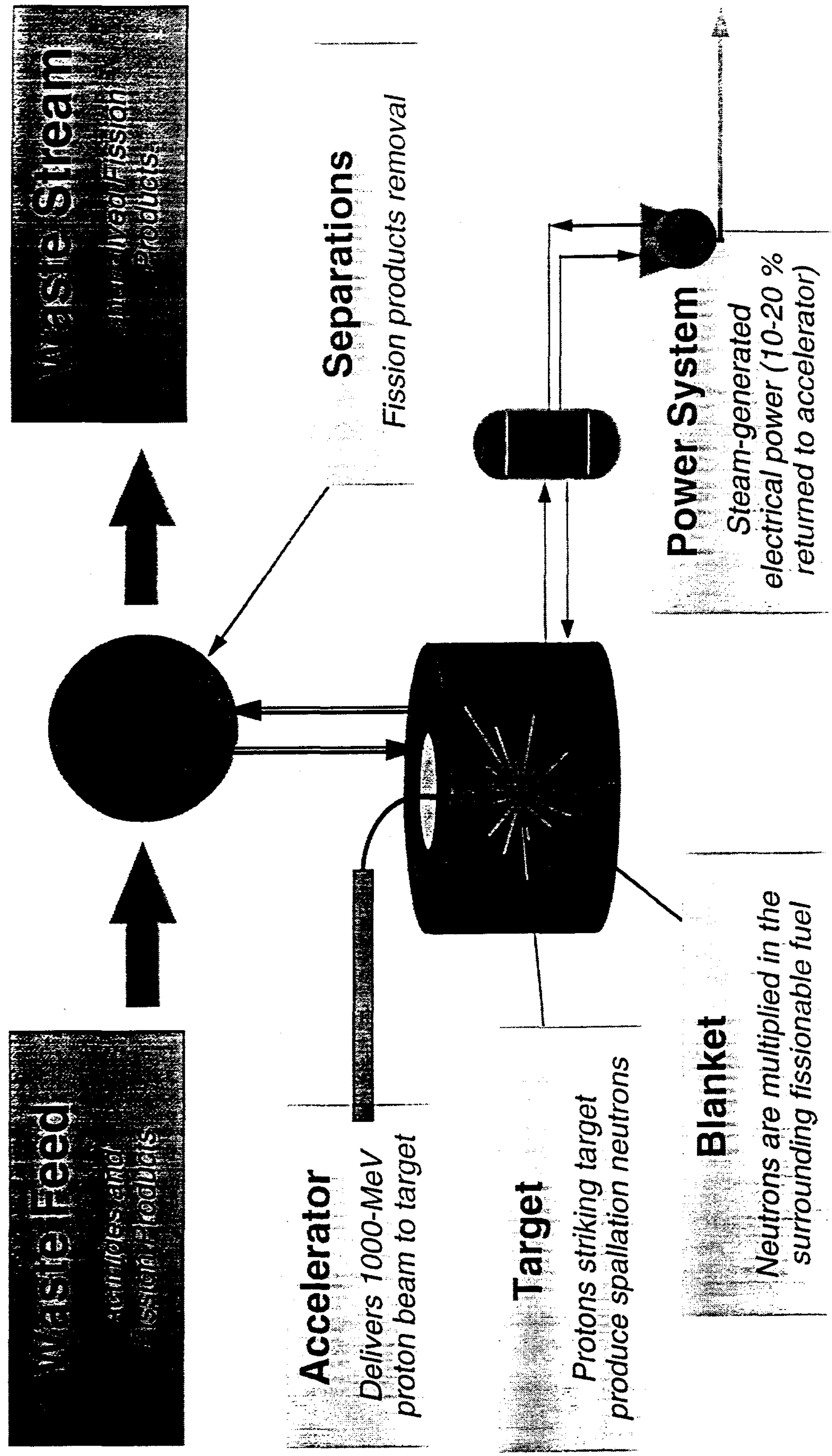

Figure 1. Principal components of the Los Alamos design for an accelerator-driven transmutation system for burning high-level nuclear waste while generating useful nuclear power.