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DISPOSITION OF NUCLEAR WASTE USING SUBCRITICAL ACCELERATOR-DRIVEN SYSTEMS

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INTRODUCTION

Spent fuel from nuclear power plants contains large quantities of Pu, other actinides, and fission products (FP). This creates challenges for permanent disposal because of the long half-lives of some isotopes and the potential for diversion of the fissile material. If worldwide nuclear power generation continues at present levels, more than 250,000 tons of spent fuel will exist by 2015, containing over 2000 tons of Pu. While there is agreement on using geologic repositories for ultimate disposal of high-level nuclear waste, different strategies for dealing with spent reactor fuel are being followed by various countries. Current US policy is for unreprocessed spent fuel to be placed directly in the repository, while other countries are opting for treatment prior to storage, including partial utilization of the fissile material.

Two issues of concern for the US repository concept are: (1) long-term radiological risk peaking tens-of-thousands of years in the future; and (2) short-term thermal loading (decay heat) that limits capacity. The long-term radiological risk is from the long-lived transuranics and the fission products Tc-99 and I-129, while thermal loading concerns arise mainly from the short-lived fission products Sr-90 and Cs-137 [1,2]. Beyond these performance concerns, it is clear that the first repository, designed for 70,000 tons of commercial spent fuel and other high-level waste, will be at capacity by 2015. If nuclear energy remains a viable option for electricity generation, there will eventually be a need for a second US high-level waste repository.

An accelerator-driven neutron source can destroy actinides through fission, and can convert long-lived fission products to shorter-lived or stable isotopes. Studies over the past decade have established that accelerator transmutation of waste (ATW) can have a major beneficial impact on the nuclear waste problem. Specifically, the ATW concept we are evaluating:

- ***Destroys over 99.9% of the actinides.*** This eliminates concern over their release to ground water and the environment, and possible diversion from spent fuel for weapons use. Their elimination also greatly reduces long-term heat loading in the geologic repository.
- ***Destroys over 99.9% of the Tc and I.*** By transmuting these fission products, two of the major long-term radiotoxicity release hazards are eliminated.
- ***Separates Sr-90 and Cs-137.*** These short-lived isotopes dominate the repository short-term heat loading. They are separated from the waste for short-term storage.
- ***Separates uranium from the spent fuel.*** The separated uranium is stored or reenriched for further use.
- ***Produces electric power.*** Energy is released during actinide destruction (by fission) and converted into electric power. A fraction (10-15%) of this power is used to run the accelerator and plant, and the rest is sold to the grid to offset ATW facility costs.

ATW SYSTEM AND PROCESS DESCRIPTION

An ATW facility consists of three major elements: (1) a high-power proton linear accelerator; (2) a pyrochemical spent-fuel-treatment & waste-cleanup system; (3) a liquid lead-bismuth cooled burner that produces and utilizes an intense source-driven neutron flux for transmutation in a heterogeneous (solid fuel) core (Fig. 1). The concept is the result of many years of development at LANL [3] as well as other research centers [4]. In the ATW concept, spent reactor fuel would be shipped to a dedicated site where the Pu, other transuranics and selected long-lived FPs would be destroyed by fission or transmutation in a single pass through the facility. This approach contrasts with reprocessing practices in Europe and Japan, in which high purity Pu is produced and fabricated into fresh mixed-oxide (MOX) fuel that is shipped off-site for use

DISCLAIMER

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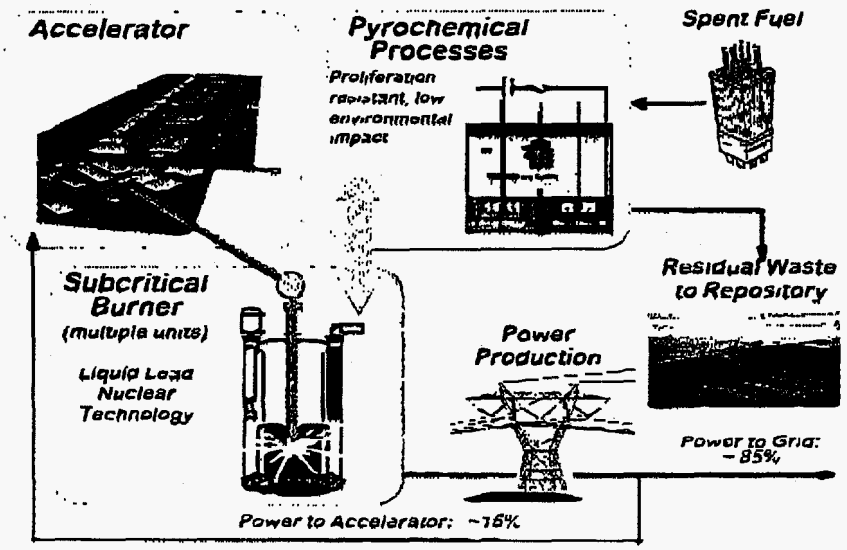

Fig. 1. ATW process schematic.

ACCELERATOR

The 1000-MeV 40-mA CW high-power accelerator design is derived from the design of a 1700-MeV, 100-mA CW proton linac being developed for another nuclear spallation application [5]. As shown in Fig.2, it is an integrated normal-conducting (NC), superconducting (SC) high-power linac that is optimized to balance several important design factors. These include compact length, low beam losses, high conversion efficiency of electric power to beam power, reliable operation and high availability, insensitivity to errors in alignment and parameter settings, and cost minimization.

Copper NC accelerating structures are used in the low-energy linac, providing strong focusing and smooth transitions in phase space, which results in low emittance growth and minimization of beam halo. In the high-energy linac, niobium SC cavities are used to provide high efficiency acceleration, very large beam apertures, and a broad velocity acceptance. The latter feature allows for retuning to continue operation, following component failures.

A 40-mA continuous proton beam produced in a microwave-driven injector is accelerated to 6.7 MeV in a 350-MHz radio frequency quadrupole (RFQ); a prototype injector has been demonstrated at LANL with performance parameters exceeding ATW requirements. Following the RFQ is a 700-MHz coupled-cavity drift-tube linac (CCDTL) that accelerates to beam to 21.2 MeV. Combining the best features of the proven Alvarez linac and the coupled-cavity linac, this hybrid structure is made up of short DTL sections that are resonantly coupled and embedded in a 9-$\beta\lambda$ focusing lattice. The focusing elements (quadrupoles) are external to the accelerating structures. The beam from the CCDTL is fed into the SC linac, which is divided into four sections, each containing niobium cavities whose shape and configuration is optimized for a particular velocity (β) range. The first section, which takes the energy up to 100 MeV, consists of spoke-type 1/2-wave resonators. These have been demonstrated, but will require further development to form integrated accelerating/focusing units (cryomodules). The last stage of acceleration is carried out in two sections of SC linac made up of cryomodules containing elliptically-shaped 5-cell cavities optimized at $\beta = 0.48$ and 0.71. Electron-accelerator ($\beta = 1$) versions of these cavities have shown the capability of reaching the high gradient and Q performance regime desired for ATW. The elliptical cavities are grouped into cryomodules containing 2 to 4 cavities each, and 3 to 5 SC quadrupoles each (in a FODO lattice), to provide

focusing. RF power for both the NC and SC sections is supplied by 1-MW CW 700-MHz klystrons, except for the RFQ, which is driven by two 350-MHz 1.2-MW klystrons.

With the design parameters shown in Fig. 2, the ATW linac length is 355 m. The RF power requirement is 42.3 MW, which is provided by 56 1-MW klystrons. The figure shows the average accelerating gradient in each section, the transition energies, and the β values at which the cavities are optimized. The accelerator is designed to produce a 40 MW beam in order to drive up to a total 2000 MWt fission power in the burner(s), which could be distributed in two or more modules. The first (Demo) ATW is projected to be a 500-1000 MWt system.

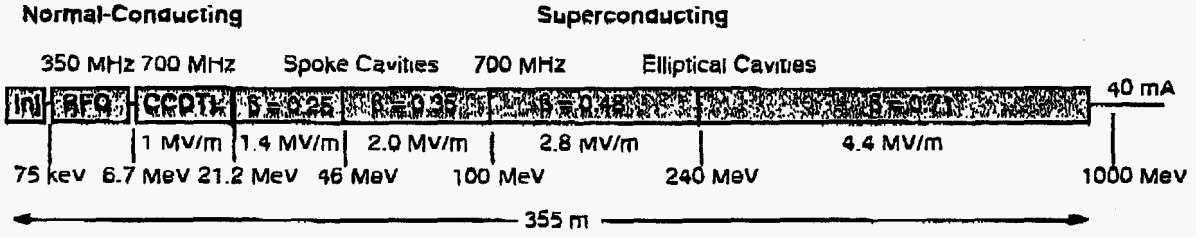

Fig. 2. ATW high-power linear accelerator concept; a NC linac injecting into a SC linac.

FUEL-CYCLE AND SPENT-FUEL TREATMENT

In the spent fuel treatment system, uranium and a majority of the FPs are separated from the transuranics and the targeted long-lived FPs by pyrochemical (non-aqueous) processes. A requirement is the removal of enough uranium (99%) so that no significant Pu or other actinides are produced during transmutation. FP extraction is not explicitly sought but comes out naturally from the process.

The flow of the spent fuel in the treatment system is broken down into three basic streams. One contains the spent-fuel cladding, the majority of the spent fuel fission products, and the remaining fission products from the transmuted waste, all of which are prepared for permanent disposal. Following electrochemical extraction of the uranium, the second stream consists of actinides and some cladding zirconium, which is cast into solid metallic fuel elements (*transmutation assemblies*) to be introduced into the ATW burner for irradiation. The third stream consists of uranium sent out and stored for possible recycle.

In our reference ATW concept, one-third of the burner core is extracted and processed every year. Eventually all the FP in the irradiated waste is partitioned into three forms: active metals, noble metals and lanthanides. This remnant waste is prepared for permanent storage as: (1) oxides in engineered containers for the active metals (including strontium and cesium); (2) oxides for the lanthanides; and (3) metal ingots and oxides for the noble metals, including zirconium. An average of 50 kg of FP per ton of spent fuel is discharged as waste after transmutation (including the FP originally present in the spent fuel), contaminated with less than 100 ppm of transuranics (mostly metal-oxides). Most of the radioactivity in these discharges will decay in less than three hundred years, with only weak residual activity (of negligible environmental impact) remaining afterwards.

Spent fuel treatment technology is derived from pyrochemical methods developed for plutonium processing at Los Alamos [6] and the Integral Fast Reactor program at ANL [7]. Pyrochemical processes are chosen over conventional aqueous processes because they are proliferation resistant - group separations are used instead of single species separations. These allow the processing media, molten salts and liquid metals, to be recycled multiple times, thus reducing secondary waste, and allow for short turnaround times for waste treatment. Radiolysis and decay heat are not significant issues [8]. In addition, the product from the electrochemical processes is easily fabricated into fuel for the ATW system.

The central issue for process chemistry is to establish scaling information by fabricating and testing various separation systems, using the information to develop a detailed material

balance for the fuel treatment and process plant parameters. An ATW fuel treatment facility would be similar to the fuel-cycle facility proposed for the Advanced Liquid Metal Reactor (ALMR) Program [9].

WASTE BURNER (TARGET/BLANKET ASSEMBLY)

The ATW waste burner consists of a liquid lead-bismuth eutectic (LBE) target that generates spallation neutrons from reactions induced by the 1-GeV proton beam. These are multiplied by neutrons from fission reactions induced in the surrounding subcritical core, which contains the transmutation assemblies. Since significant heat production occurs from the fissioning of the actinides in these assemblies, adequate heat removal must be provided, analogous to that in critical reactors of similar power level. A major advantage of LBE is that it can be used both as a spallation neutron source and a nuclear coolant. The LBE technology, successfully developed in Russia for nuclear submarine propulsion, has only recently become accessible and appears ideally suited to the ATW application. LBE's low melting point (123.5°C), high boiling point (1670°C), and very low vapor pressure allow a wide operating temperature range, eliminates coolant boiling, and enhances cooling circuit safety. Its high density, combined with the wide temperature range, offers extraordinary natural convection cooling capability for enhanced passive safety. In addition to its use as a coolant and spallation target, LBE provides a negative coolant void and temperature reactivity coefficient [10]. A schematic of the ATW burner (target/blanket) is shown in Figure 3.

The subcritical liquid LBE ATW concept operates with a fast neutron spectrum to ensure optimal actinide destruction efficiency and high neutron availability for FP transmutation. Very low end-of-life inventories are achieved by burn-down strategies involving gradual thermalization of the spectrum to exploit the large capture cross sections in the nuclear resonance region.

While the subcritical operation of the ATW burner does not make it intrinsically *safer* than critical reactors, it facilitates burnup tasks that would be very difficult or inefficient in conventional critical nuclear systems. Subcritical systems do not rely on delayed neutrons for control and power change; they are driven by the external neutron source (i.e. by the protons from the accelerator). Control rods and reactivity feedback have low importance, since these systems are neutronically (but not thermally) decoupled from the neutron source. Subcriticality allows the ATW burner to work with any composition of fuel or waste and to greatly relax the required separation in the waste-treat-ment steps. This permits the destruction of any isotopes (actinides or FPs or mixture of both) with little concern for their neutronic behavior. Fertile materials are not needed to compensate for neutronic uncertainties or undesirable reac-tivity responses of the fuel, and extended burn-up is achieved by increasing the power of the accelerator drive to compensate for reactivity decreases.

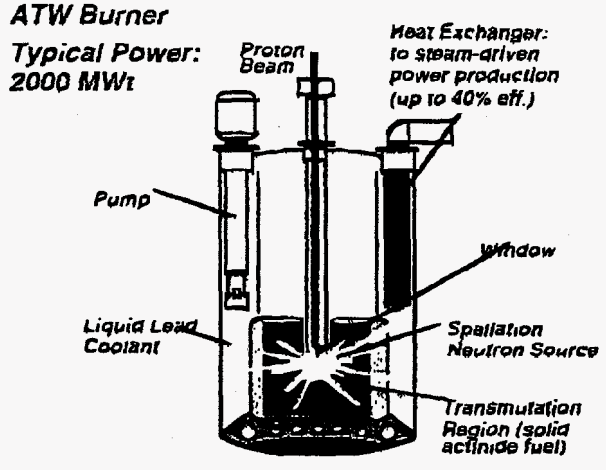

Fig. 3. ATW burner (LBE target/blanket)

Because of the above features, ATW is ideally suited as an incinerator of material that: 1) is not well characterized; 2) burns very poorly or not at all in reactors; 3) has potentially unstable and hazardous reactivity responses. This includes higher actinides such as Np (the worst contributor to oxidizing long-term performance uncertainties in a repository), Am and Cm, all isotopes of Pu, and some long-lived FPs. In addition, the neutron-poor Th-U fuel cycle, never successfully implemented in critical reactors, could be used rather straightforwardly in accelerator-driven subcritical systems.

Although LBE can be rather corrosive and can be contaminated by solid admixtures due to interaction with construction materials and oxygen, the Russian [11] heavy-metal technology has mitigated these effects by selecting proper materials and actively controlling oxygen thermodynamic activity in the coolant. The essence of this technology is to adjust the oxygen level in the LBE so that a self-healing protective oxide film can grow on the surface of the structural materials, inhibiting corrosion, while at the same time no excessive oxygen is available to form solid admixtures (mostly lead oxide).

TRANSMUTING THE SPENT FUEL BACKLOG IN 65 YEARS; A SCENARIO

The design flexibility afforded by subcriticality provides many possibilities for implementing ATW in effective waste destruction scenarios. Sketched below is an implementation scenario for transmuting the spent nuclear fuel backlog in 65 years. Many others can be developed [9].

By 2015, there will be 70,000 tons of spent fuel in the US containing about 600 tons of Pu and higher actinides. The ATW objective is to treat this spent fuel backlog, destroy the transuranics and selected FPs, and prepare the remaining waste for permanent disposal in a geologic repository within a relatively short time. To accomplish this objective, 21 ATW systems are brought on line over 65 years. Each ATW system consists of a 1-GeV accelerator with a current variable from 20-40 mA, a subcritical 2000 MWt LBE burner, and a pyrochemical plant having 50 tons/year throughput per burner. In the scenario, there are three large facilities, with up to seven ATW systems located together in one facility. The parameters for the burners are as shown in the table below.

Parameter	Value	Parameter	Value
Fission power	2000 MWt	Thermal-electric efficiency	40%
Power to grid	675 MWe	Recirculated power	125 MWe
Operational TRU inventory	3000 + 1000 kg	Conversion ratio	0.0
Process losses	1/1000	TRU burn rate	up to 650 kg/yr
TRU burn efficiency	16.25%		

Of particular relevance is the fact that each ATW burner can destroy up to 650 kg of actinides (Pu andTRU) per year, and that 675 MWe would be available for distribution to the grid from each burner after powering the driving accelerator and the plant. In the 3-facility scenario, a new ATW system is brought online at each site every 4 years. As the burners reach the end of their operational life, they are decommissioned, and their fuel is sent to feed other still operational burners. Eventually, the last ATW system in operation at each site will receive all the remnant waste and destroy it down to less than 1 ton over a protracted (5 years) inventory burn-down period (Figs. 4,5).

In general the following key points can be made about the cost of such an ATW system: 1) the cost of the particle accelerator does not dominate the economics; 2) the pyrochemical waste treatment processes are less expensive than traditional aqueous chemistry processes [8]; 3) the cost of subcritical ATW burners based upon lead/bismuth coolant technology should be comparable to or lower than the cost of sodium-cooled reactors; and 4) electricity produced by the ATW plant will offset operating costs and produce revenue. A detailed cost estimate and balance sheet remains to be done.

Taking credit for the probable elimination of the cost of a second US geologic repository, it is reasonable to conclude that the economic prospects for ATW are encouraging, possibly providing an economic gain along with the other benefits.

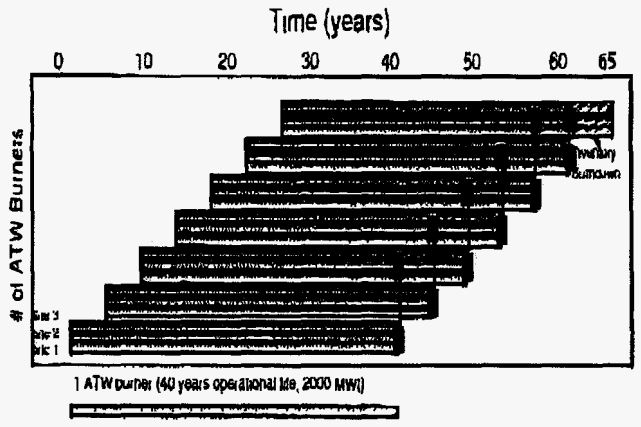

Fig. 4. ATW backlog burndown scenario.

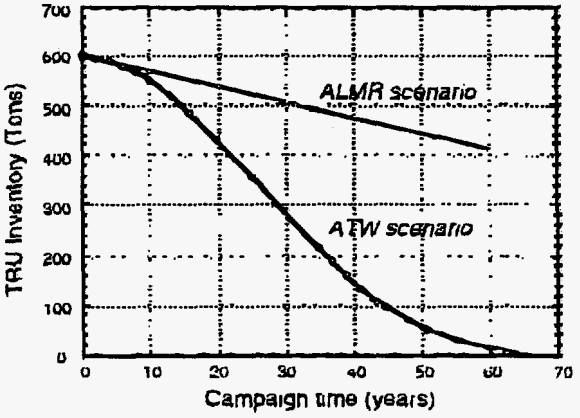

Fig. 5. ATW burndown vs ALMR.

CONCLUSION AND ACKNOWLEDGEMENTS

ATW systems can destroy virtually all the Pu and higher actinides without reprocessing the spent fuel in a way that could lead to weapons material diversion. Once demonstrated and developed, ATW could be an essential part of a global non-proliferation strategy for countries building up large quantities of plutonium from their commercial reactor waste. ATW technology, initially proposed in the US, has received wide and rapidly increasing attention around the world, especially in Europe and Asia, with major programs now being planned, organized and funded. Substantial convergence presently exists on the technology choices among the programs, opening the possibility of a strong and effective international collaboration on phased development of the technology.

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