LA-UR- 98-1789

Approved for public release; distribution is unlimited.

Title: DISPOSITION OF NUCLEAR WASTE USING SUBCRITICAL ACCELERATOR-DRIVEN SYSTEMS

CONF-980659--

Author(s): DOOLEN, Gary D., T-13
VENNERI, Francesco, LANSCE-3
LI, Ning, MST-10
WILLIAMSON, Mark A., MST-11
HOUTS, Michael, TSA-10
LAWRENCE, George, APT-TPO

Submitted to: Presentation at:

9th International Conference on Emerging Nuclear Energy Systems
Tel-Avis, Israel
June 27-July 2, 1998
&
Tel-Aviv University
June 25, 1998

Los Alamos
NATIONAL LABORATORY

DISCLAIMER

DISPOSITION OF NUCLEAR WASTE USING SUBCRITICAL ACCELERATOR-DRIVEN SYSTEMS

Francesco Venneri, Ning Li, Mark Williamson, Michael Houts, George Lawrence
presented by Gary Doolen
Los Alamos National Laboratory
Los Alamos, NM 87545, USA

1. INTRODUCTION

Spent reactor fuel from commercial power plants contains significant quantities of plutonium, other fissionable actinides and fission products, all of which create challenges for permanent disposal because of the very long half-lives of some isotopes and because of the potential for diversion.

While there is agreement on using geologic repositories for the ultimate disposal of high-level nuclear waste, different strategies for dealing with spent nuclear fuel are being followed by various countries, reflecting their views on nuclear power, reprocessing and non-proliferation. Current US policy is to store unreprocessed spent fuel in a geologic repository. Other countries are opting for treatment of spent fuel, including partial utilization of the fissile materials contained in the spent fuel prior to geologic storage.

Key issues for the current US repository concept fall into two categories, (1) a long-term radiological risk with the peak risk projected tens of thousands of years after repository closing and (2) a short-term thermal loading (decay heat) that limits the capacity of the repository. While not identified as an issue, it is clear that the repository, designed for 70,000 tons of commercial spent fuel and other government-generated high-level waste (mostly from defense-related activities), will be fully occupied by the spent fuel produced through the year 2015. If the nuclear energy remains to be a viable option for electricity generation in the future, ultimately there will be a need for a second high-level waste repository.

Studies have shown [1,2] that the repository long-term radiological risk is from the long-lived transuranics and the fission products Tc-99 and I-129, thermal loading concerns arise mainly from the short-lived fission products Sr-90 and Cs-137.

In relation to the disposition of nuclear waste, ATW is expected to accomplish the following: (1) Destroy over 99.9% of the actinides. (2) Destroy over 99.9% of the Tc and I. (3) Separate Sr and Cs (short half-life isotopes). (4) Separate uranium. (5) Produce electricity.

In the ATW concept, spent fuel would be shipped to a ATW site where the plutonium, other transuranics and selected long-lived fission products would be destroyed by fission or transmutation in their only pass through the facility. This approach contrasts with the present-day reprocessing practices in Europe and Japan, during which high purity plutonium is produced and used in the fabrication of fresh mixed-oxide fuel (MOX) that is shipped off-site for use in light water reactors.

2. ATW SYSTEM 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. The concept is the result of many years of development at LANL [3] as well as other major international research centers [4].

The high-power accelerator for ATW would be based on the APT (Accelerator Production of Tritium) accelerator (1.7Gev, 100mA, 170MW proton beam). An accelerator, similar to but smaller than the one now being designed for tritium production would serve as the driver (40 MW) to a subcritical burner, where transuranics and selected fission products are fissioned or transmuted.

In the spent fuel treatment system, uranium and a majority of the fission products are separated from the transuranics and the targeted long-lived fission products by pyrochemical (non-aqueous) processes. The only requirement is the separation of enough uranium (99%) so that no significant new plutonium or other actinides are produced during transmutation. Fission product extraction is not explicitly sought but comes out naturally from the process.

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

In one reference design concept, a third of the core is extracted and processed every year. In the ATW waste cleanup process, eventually all the fission products in the irradiated waste are 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 kilograms of fission products, per ton of spent fuel, are discharged as waste after transmutation (including the fission products originally present in the spent fuel), contaminated with less than 100 ppm of transuranics (mostly in the metal oxide waste form). Most of the radioactivity in the discharges would decay before three hundred years, with only weak residual activity of negligible environmental impact remaining afterwards.

The waste burner consists of a heavy metal target (liquid lead-bismuth eutectic (LBE)) producing the high intensity neutron source and the surrounding subcritical core containing the transmutation assemblies. Since significant neutron multiplication and heat production occurs from the fissioning of the waste actinides contained in the

surrounding transmutation assemblies, adequate means for heat removal must be present, analogous to critical reactors of similar power level. ATW takes advantage of the exceptional properties of liquid LBE, both as nuclear coolant and as spallation neutron source, for use in the subcritical waste burner. The technology, successfully developed and used in Russia for nuclear submarine propulsion of very fast, deep diving vessels, is becoming accessible to western researchers and engineers.

The subcritical liquid LBE systems presently being developed at Los Alamos operate in the fast neutron spectrum, to ensure optimal destruction efficiency for the actinides and large neutron availability for transmutation of the targeted fission products. Very low end-of-life inventories are rapidly achieved by burn-down strategies involving gradual thermalization of the spectrum to exploit the large capture cross sections of resonances.

Subcriticality does not make ATW by definition "safer" than critical reactors. Rather, subcriticality facilitates tasks that would be exceedingly difficult or inefficient in critical systems. Subcritical systems do not rely on delayed neutrons for control and power change, they are driven only by the externally generated neutron source (i.e. by the ion beam coming from the accelerator). Control rods and reactivity feedback have very low importance: these systems are neutronically (but not thermally) decoupled from their neutron source. Subcriticality therefore allows the ATW system to work with any composition of fuel (or waste) and to greatly relax the required separation in the waste treatment steps. This makes possible, in principle, the destruction of any isotopes (actinides or fission products or mixture of both) with little concern for their neutronic behavior. Fertile materials are not needed to compensate for the neutronic uncertainties or undesirable reactivity responses of the fuel, and extended burnup is achieved by increasing the power of the accelerator drive to compensate the reactivity decrease.

Because of its subcritical mode of operation, ATW will be ideally suited as "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; and (4) should not for whatever reason be isolated and placed in reactors. This includes higher actinides such as neptunium (the worst contributor to an oxidizing repository long-term performance uncertainties), americium and curium, all isotopes of plutonium and some long-lived fission products. In addition, the neutron-poor thorium-uranium fuel cycle, never successfully implemented in critical reactors, can be used rather straightforwardly in accelerator-driven subcritical systems.

3. LINAC

The 1000 MeV reference linac design for ATW is based on consideration of a number of important issues. These include low beam losses, high efficiency of electrical power to beam power conversion, reliable operations, insensitivity to errors in alignment and settings, and cost optimization. The design uses demonstrated components to transmit the beam through the different energy regimes. Initially the

beam is accelerated to 6.7 MeV in a RFQ (Radio Frequency Quadrupole) based on a well-defined beam emerging from a reliable injector. A suitable injector has been working for months at LANL with currents in excess of a factor of two for what ATW needs and with beam parameters better than the ATW requirements to ensure low beam loss. In addition, the CRITS RFQ has been accelerating a proton beam with good transmission and beam parameters exceeding the ATW requirements.

Following the RFQ is a CCDTL (Coupled Cavity Drift Tube Linac) that will be demonstrated in the APT LEDA program. This structure has revolutionized the ability to transport high quality beams from a RFQ to following structures. The 21.2 MeV beam from the CCDTL is then fed into a set of superconducting cavities that take the beam up to 1000 MeV. The first type of cavity that accelerates the beam to 100 MeV is based on a "spoke" resonator design that will require some testing before it is fully qualified for this program. All indications are that there should be no problems with this geometry and with the required field levels in the cavities.

The last stage of acceleration will be with elliptical shaped cavities: these have been demonstrated to be able to meet the required performance regimes necessary for ATW. The ATW linac length is 355 m with an rf power need of 42.3 MW for a 40 MW beam. The accelerator was designed for 40 MW in order to be able to drive up to a total 2000 MWt fission power, which could be distributed within one or more modules. The first Demo ATW is projected to be a 500-1000 MWt system.

4. FUEL CYCLE TECHNOLOGY

Spent fuel treatment technology is derived from pyrochemical processes developed for plutonium production at Los Alamos [5] and the Integral Fast Reactor program at Argonne [6]. Pyrochemical processes were chosen over the conventional aqueous processes because they are proliferation resistant - group separations are used instead of single species separations; 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 [7]. In addition, the product from the electrochemical processes is easily fabricated into fuel for the system. An ATW fuel treatment facility would be similar to the fuel cycle facility proposed for the Advanced Liquid Metal Reactor (ALMR) Program [8].

Process technologies are based on modifications of existing technologies so as to achieve the ATW process requirements (see section 1). A brief description of the flow sheet using as an example the conversion of spent uranium oxide fuel to ATW fuel and the recycle of ATW fuel follows.

Spent fuel chopping and decladding prepares the fuel rods into small sections and allows for the separation of the spent fuel, uranium oxide, from the clad matrix, zircalloy. This process is based on technology used at the major reprocessing plants in Europe. Separation of the oxide fuel from clad material is desired so that the clad material is not carried into the chemical processes. The clad material could be used as

the inert matrix in ATW fuel. Fission product gases, primarily xenon and krypton, released during the decladding process are collected by cryogenic methods, or in getter-beds, and sent to storage. The gas collection system is based on technologies used in Europe and those studied and proposed for use in the US.

Spent oxide fuel is converted to metal by the direct oxide reduction process. This process involves the reaction of calcium metal with the oxide fuel to produce calcium oxide and heavy metal (i.e., U, Np, Pu, Am, Cm). It is completed in a calcium chloride molten salt flux maintained at approximately 1025 K. Some fission product partitioning takes place during the oxide reduction process. Fission gases are released from the matrix of the oxide fuel and are recovered by the same methods described for the decladding system. Active metals, such as cesium, strontium, and barium, are partitioned to the molten salt and are periodically removed from the salt during the direct oxide reduction salt recycle process, placed in engineered storage containers, and sent to the repository. Iodine is also partitioned to the molten salt and recovered from the salt during the salt recycle process. It is collected by cryodistillation methods, fabricated into targets, and transmuted in the ATW system. The heavy metal produced in the oxide reduction process is sent to the electrorefining system.

Electrorefining is used to partition the uranium, transuranics (TRU), and fission products. The system uses electrochemical methods to electrotransport the U from the anode to a solid cathode. The U-bearing cathode is removed from the system and either sent to storage or recycled. An eutectic mixture of NaCl-KCl molten salt, at approximately 1000 K, is used as the transport medium. Noble metal fission products (i.e., Zr, Mo, Ru, etc.) remain at the anode heel in the cell. The anode heel is subjected to a second electrorefining process to further reduce the amount of TRU present in the matrix. It is then sent to the technetium recovery process before being discharged to the repository. TRU's and rare earth fission products remain in the molten salt. This salt is treated by the electrowinning process.

Electrowinning is an electrochemical process used to electrodeposit the TRU's from the NaCl-KCl molten salt. The TRU's, present in the molten salt as complex chlorides, are reduced at the cathode of the cell. A sacrificial anode is used to react with the free chloride produced by the reduction of the TRU's. The TRU's are transferred to the vacuum casting fuel fabrication system. Fuel for the system is 85% Zr, or zircalloy, - 15% TRU clad in steel. The molten salt is recycled to the electrorefining system after the rare earth fission products are removed from the salt by a reductive extraction process. After the extraction process, the rare earths are collected, oxidized, packaged, and sent to the repository.

Technetium is removed from the electrorefining anode heels and sent to the ATW system for transmutation. The Tc metal is alloyed with Mo or Ru, fabricated into targets, and irradiated in ATW. The remaining transition metal oxides are packaged and sent to the repository.

The back-end of the fuel cycle uses processes similar to those used at the front-end. Spent ATW fuel is chopped and decladded by standard techniques. It is transferred to

an electrorefining system where the TRU's are partitioned from the active metal, noble metal, and rare earth fission products. These TRU's are sent to the vacuum casting system where fresh ATW fuel is fabricated. Fission product gases released during the electrorefining processes are collected by the aforementioned methods. Noble metals are removed from the electrorefining cell, transferred to the Tc recovery process, and ultimately sent to the repository. Periodically the TRU content in the transport molten salt is decreased by using the aforementioned electrowinning process. Also periodically, the rare earth fission products are removed from the electrorefining salt by reductive extraction techniques and are collected, oxidized, packaged, and sent to the repository.

4.1 Metallic Fuel. Existing technology is used wherever possible in the ATW nuclear subsystem. The primary exception is the ATW fuel. The need to eliminate uranium from the waste, the desire to use LWR clad (zircalloy) as the inert fuel matrix, and the desire to make processing as simple and waste-free as possible drives the fuel form to a zirconium-based metal matrix with an initial transuranics loading of about 15%. The fuel is a high melting alloy (> 1900 K) and at the operating temperature of the transmutation system is a solid solution of TRU in alpha zirconium. Metallic fuels have long been proposed for use in ALMR's and have been studied in experimental reactor facilities.

5. LEAD-BISMUTH NUCLEAR COOLANT AND SPALLATION TARGET

Lead-Bismuth eutectic (LBE) possesses some unique physico-chemical properties, making it an excellent nuclear coolant and spallation neutron source. LBE's (44.5wt% Pb - 55.5wt% Bi) low melting point (123.5°C), high boiling point (1670°C) and very low vapor pressure allow for a wide operating temperature range, eliminates coolant boiling and enhances circuit safety. The high density of LBE combined with wide permissible temperature range offers extraordinary natural convection cooling capability for enhanced passive safety. LBE's low chemical activity inhibits violent reactions (fire and explosion) with air and water. The sealed vessels and circuits readily prevent air-borne lead contamination from exceeding established industrial standards ($0.01 mg/m^3$ in Russia, $0.03 mg/m^3$ in US). The choice of LBE coolant for the ATW system is based primarily on two factors. First, the LBE can be used as both the coolant and the spallation target. Second, the use of LBE results in a negative overall coolant void and temperature reactivity coefficient [9].

The integration of nuclear coolant and spallation target in the current ATW concept drastically improved the subcritical burner design by simplifying flow configuration, material compatibility and removing target structures in high proton and neutron fluxes. LBE has very high useful neutron production during spallation and extremely low neutron capture cross sections. This neutron transparency allows for a widely spaced core with much reduced pressure drop and pumping power requirement. The coolant is also self-shielding against gamma radiation.

5.1 Existing Russian LBE Nuclear Coolant Technology. Although LBE can be rather corrosive and can be contaminated by solid admixtures due to interaction with construction materials and oxygen, the Russians developed the "heavy metal technology" to mitigate these adverse 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 LBE coolant so that a self-healing protective oxide film can grow on the surface of the structural materials to prevent corrosion, while no excessive oxygen is available to form solid admixtures (mostly lead oxide). The Russians successfully deployed this technology in their nuclear submarine reactors and have over 70 reactor-years (150 MWt units) of experience [10].

The surfacing of viable Russian liquid lead/bismuth technology is opening the door to the possible commercial use in power producing nuclear plants. Russian designs for small (100MWt) and large (1000MWt) reactors are being evaluated. It is widely believed that these designs obviate many of the problems inherent to sodium-cooled reactors, such as positive void coefficients and fire hazards.

6. COST CONSIDERATIONS

In general the following key points are known about the cost of an ATW system: • the cost of the particle accelerator will not dominate the economics of ATW; • the pyrochemical waste treatment processes are acknowledged to be less expensive than traditional aqueous chemistry processes [8]; • the cost of subcritical ATW burners based upon lead/bismuth coolant technology should be comparable or lower to the cost of critical sodium-cooled reactors; • electricity produced by the ATW plant could offset operating costs and produce revenue.

Including a possible reduction in the cost of the repository introduced by ATW, it is reasonable to conclude that the economic prospects for ATW are encouraging, possibly providing an economic gain along with its other benefits.

7. CONCLUSIONS

ATW destroys virtually all the plutonium 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 that could build up large quantities of plutonium from their commercial reactor waste. ATW technology, initially proposed in the US, has received wide and rapidly increasing attention abroad, especially in Europe and the Far East, 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 the phased development of the ATW technology.

If the job of nuclear waste destruction has to be done quickly, safely, and with reasonable investment, we believe that a dedicated, once-through subcritical burner (ATW) system would provide the most effective option. ATW can provide, within a

realistic nuclear technology envelope, a way to destroy the undesired products of nuclear energy generation. This is a new instrument in the field of nuclear systems: it could accomplish the destruction of all transuranics (including plutonium) and long lived fission products, or only a residual portion, if recycle of Pu in existing critical reactors is deemed acceptable. The technologies introduced and developed for ATW (liquid lead/LBE nuclear coolant, pyrochemical processes, high power accelerators) will also have important applications to, and could well constitute the backbone of future nuclear systems (both critical and sub-critical).

ATW systems could be used in a series of different scenarios, including the expanded, sustained or declining use of nuclear power. The ability to demonstrate such a flexible means of destruction of waste will be very important in fostering the confidence that a "forever" legacy of waste is not the unavoidable consequence of having once used nuclear power, or by the same token in promoting the acceptance of nuclear power as a viable and environmentally sustainable large-scale energy source.

8. FURTHER READING

[1] Committee on Separations Technology and Transmutation Systems, "Nuclear Wastes: Technologies for Separations and Transmutations." Nat. Academy Press, Washington, DC, 1996

[2] G. Michaels, "Potential Benefits of Waste Transmutation to the U.S. High-Level Waste Repository", AIP Conference Proceedings 346: Int'l Conf. on Accelerator-Driven Transmutation Technologies and Applications, 8 (1995) and references therein.

[3] C.D. Bowman et al., "Nuclear Energy Generation and Waste Transmutation using an accelerator-driven intense thermal neutron source." Nuclear Instr. and Methods A230, 336 (1992).

[4] C. Rubbia, "A high gain Energy Amplifier operated with fast neutrons", same as [2]

[5] D.C. Christensen, L.J. Mullins, "Plutonium Metal Production and Purification at Los Alamos," in Plutonium Chemistry, Carnall and Choppin, Eds. Am. Chem. Soc., Washington D.C. , 409 (1983)

[6] J.J. Laidler, J.E. Battles, W.E. Miller, and E.C. GAY, "Development of IFR Pyroprocessing Technology," in Proceedings of Global '93, (1993).

[7] M.A. Williamson, "Chemistry Technology Base and Fuel Cycle of the Los Alamos Accelerator-Driven Transmutation System," in Proceedings of Global '97, 263 (1997).

[8] Burns and Roe Company, "ALMR - Fuel Cycle Facilities, Design Report and Cost Estimates", GEFR-00942, BRC-448 (1995).

[9] F. Venneri et al., "Accelerator-driven Transmutation of Waste (ATW) Technical Review at MIT", LANL Report, LA-UR-98-608.

[10] Communications and contract reports from the Institute of Physics and Power Engineering (Obninsk) and EDO-Gidropress (Podolsk), Russia.

This paper is based on Los Alamos Internal Report LA-UR 98-985.