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DISPOSITION OF NUCLEAR WASTE USING SUBCRITICAL ACCELERATOR-DRIVEN SYSTEMS: TECHNOLOGY CHOICES AND IMPLEMENTATION SCENARIO - ICONE-6526

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ABSTRACT

Los Alamos has led the development of Accelerator-driven Transmutation of Waste (ATW), to provide a fundamental technological solution to the nuclear waste problem. While ATW will not eliminate the need for a high-level waste repository, it offers a new technology option for altering the nature of nuclear waste and enhancing the capability of a repository. The basic concept of ATW focuses on reducing the time horizon for the radiological risk from hundreds of thousands of years to a few hundred years and on reducing the thermal loading. Furthermore, ATW will greatly reduce the amount of waste that has to be disposed of in a high-level waste repository. The goal of the ATW nuclear subsystem is to produce a three orders of magnitude reduction in the long-term radiotoxicity of the waste sent to a repository, including losses through processing. If the goal is met, the radiotoxicity of ATW-treated waste after 300 years would be less than that of untreated waste after 100.000 years.

These objectives can be achieved through the use of high neutron fluxes produced in accelerator-driven subcritical systems. While critical fission reactors can produce high neutron fluxes to destroy actinides and the selected fission products, the effectiveness of the destruction is severely limited by the criticality requirement. Furthermore, to achieve safe reactor operations, a substantial amount of excess reactivity would have to be supplied initially and compensated for by control poisons. To overcome these intrinsic limitations, we searched for solutions in subcritical systems freed from criticality requirement by taking advantage of the recent breakthroughs in accelerator technology and the release of liquid lead/bismuth nuclear coolant technology from Russia. The effort led to the selection of an accelerator-driven subcritical system that results in near complete destruction of the actinides and fission products of concern, as well as permitting easy operational control through the external control of the neutron sonroc rather than the internal control rods

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. If the level of global nuclear power generation for the near future continues as it exists today, then in the year 2015 more than 250,000 tons of spent fuel worldwide will have to be stored, containing over 2000 tons of plutonium. Well over 70,000 tons of this spent fuel will be in the US, containing more than 500 tons of plutonium (600 tons of transuranic actinides).

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 transurances 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) <u>Destroy over 99,9% of the actinides</u>. Actinide destruction eliminates concerns with their releases to the ground water and the environment, their possible diversion and use in the spent fuel for weapons construction. Their elimination will also greatly reduce long-term heat loading.

(2) <u>Destroy over 99.9% of the Tc and J.</u> By transmuting technetium and iodine, two of the major long-term radiotoxicity release hazards can be eliminated.

(3) <u>Separate Sr and Cs (short half-life isotopes)</u>. Sr-99 and Cs-137 dominate the repository short-term heat loading. These isotopes are not suited for transmutation but will be separated from the remainder of the waste for optimal storage.

(4) <u>Separate uranium</u>. Uranium is separated from the rest of the spent fuel, stored or re-enriched for further use.

(5) <u>Produce electricity</u>. The ATW resembles in many ways a nuclear reactor in that it releases energy during actinide destruction (fission) that can be converted into electricity. A small fraction (10-15%) of this electricity will be used to power the accelerator, the rest can be distributed for sale.

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. Instead of "reprocessing", the ATW approach can be fairly characterized as "once-through destruction". ATW would inhibit plutonium accumulation, proliferation and diversion. The end products of ATW are a more benign fission product waste stream, uranium similar in composition to natural uranium, and electricity. The electricity produced, and the potential cost benefits realized by enhancing the capacity of a repository (and elimination of the need for an additional repository), could offset to some extent the cost of developing and implementing the ATW technology.

Far from being limited to waste destruction, ATW technology also brings to the table new concepts that could be relevant for the next-generation power producing systems. As such, ATW has gained worldwide interest and could be an important component of strategies to deal with international nuclear materials management and promote new, proliferation-resistant, safe reactor technologies.

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 transnutation in a heterogeneous (solid fuel) core (Fig. 1). The concept is the result of many years of development at LANI. [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 (Fig. 2), 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 seni 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 (Fig. 3). 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 inake 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

Desired Outcome of Program

ATW has reached a stage where further investigation would determine its suitability and the role it may have in a future nuclear economy.

To date, studies within the US have been limited to Institution-supported investigations at rather low levels.

 $\frac{d\omega}{d\omega} = -\frac{1}{2} \sum_{i=1}^{N} \frac{1}{(\omega_i - \omega_i)^2} \sum_{i=1}$

Engineering development and demonstration activities in the 5-year program would provide useful information for bringing the technology and options to a point for informed decisions.



Presentation to AIL/ ATW REVEW

Where are we since STATS Panel?

In 1991 (report 1996) the National Research Council (STATS panel) concluded that Separations and Transmutation (including ATW) was:

- · immature
- too expensive
- » leading to proliferation
- commits to expanding nuclear energy
- » doesn't significantly impact need for repository

Technology advancements answer most of the concerns expressed seven years ago.



Concerns

Political related issues:

- Reprocessing
- **Fuel cycles**
- Licensing
- Waste management

Are we making the best investments to ensure a sustainable and favorable energy future for future generations?

Are decision makers being given the best data to make decisions that will have future impacts?



What about International Interests?

Common technical approach has evolved -- liquid Pb/Bi coolant, solid fuel, pyrochemistry, sub-critical accelerator drive.

Europe (Spain, Italy, France, Sweden, Czech Rep.) Asia (S. Korea, Japan)

ISTC funding assists Russia and other FSU countries. Collaborations -- CERN, KAERI, JAERI, CEA, Sweden.



US Participation -- Other

Laboratories

- Argonne: Have assisted us in pyrochemistry. Preliminary discussions with them on possible interests in ATW.
- Oak Ridge: Have assisted us in liquid metals and materials.
 Preliminary discussions with them on possible interests in ATW.

Universities

 Host of Ph. D. and M. Sc. thesis topics would be spun out of this program.

We would like to see them as part of the team.



US Participation -- Present

Laboratories

- » Los Alamos: Systems, Pb/Bi, Chemistry, Accelerator, Materials.
- Livermore: Systems studies, Pb/Bi, Safety.
- Sandia: Safety, Systems analysis, Materials, Diagnostics.
- » Savannah River: Engineering and Operations.

Industry

- Westinghouse: Nuclear design, Liquid Pb/Bi Technology.
- Bechtel: Engineering.
 - Northrup-Grumman: Accelerators, Systems.

Universities

- Illinois: Materials development.
- UC Berkeley: Nuclear design.



Spin-offs from 5-Year Program Important Outcome/Legacy

Liquid lead/bismuth technology for future advanced nuclear systems.

- High power spallation targets for basic science research.
- Waste disposition processes for environmental management.
- System studies that will provide advanced fuel cycle information and a basis for future decisions/directions.



Plans and Program

With industry we have developed a viable program plan for ATW and determined rough cost estimates; results indicate the need for further comprehensive study.

We have a strong team pulled together, with the intention of making it even stronger with future additions.

The five year program discussed later focuses on leveraging items and important issues-- mass flows, pyrochemistry, materials verification, technology transfer and integration, liquid Pb/Bi cooled cores with integral target, accelerator, system studies.

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ATW System

If the scenario were to transmute all of the 70,000 tons of reactor spent fuel accumulated to 2015 within a 65 year time-frame, then:

- *******:** =70,000 tons (including 600 tons TRU).
- Output: *67,000 tons uranium (LLW).

⁺<0.3 tons TRU.

*3000 tons fission products (minimal Tc and I).

*electricity.

*residual activity and radiotoxicity of waste after 300 years less than non-assisted repository after 100,000 years.



ATW Based on Three Major Functional Blocks

Subcritical burner using liquid lead/bismuth technology based on extensive Russian nuclear reactor work and on solid fuel.

Pyrochemical processes based on significant work at ANL and LANL on efficient processes that have potential for proliferation resistance and low environmental impact.

APT-class accelerator (although about 40 MW of beam power required -- re 170 MW for APT: building blocks are based on SDIO investments and APT developments in high power linac technology).



Present System is a Design Evolution

Many options for ATW have been studied at LANL, as well as comparisons of critical nuclear assemblies with accelerator-driven systems. Design has matured significantly, being based on elements that have evolved within the international community as well:

 subcritical assembly, fast n spectrum, accelerator drive, liquid Pb/Bi coolant, solid fuel and pyrochemistry.

Present design *IS NOT* based on previously considered elements such as:

1

» molten salt, thermal n spectrum, liquid fuels, centrifuge separations.



Assisting the Disposal Option

ATW can assist a repository in many ways. Whether waste disposal is based on fully buried or on monitored and retrievable, the material can be treated by an ATW system in the future. ATW ultimately impacts capacity efficiency of a repository.

ATW thereby impacts the need for a second repository:

- » increasing storage efficiency by transmutation.
- » increasing storage efficiency by types of materials.
- decreasing long-term risks.

In addition:

converting transuranics to useful energy.



ATW: What is it?

Assisting waste disposal options by transmuting waste.

Accelerator provides protons that produce copious neutrons from spallation target. These neutrons assist the nuclear system to transmute transuranics by fission, and fission products (Tc and I) by absorption. Subcriticality (accelerator drive) enables the destruction of actinides and fission products: safely, without isolation of weapons grade material, without extensive separations work : in a single-purpose device.



Past and Future Interests

Because of an interest in future energy economies for the country, LANL has made a significant investment in studies for ATW over the past eight years.

We have reached a stage where it would be prudent to take the next step in technology development -- the 5-year plan.

This path of ATW technology development could be a bridge to future nuclear options/strategies (PCAST), even if ATW was not added to the suite of systems.



Premise

Economics appear favorable for ATW-assisted waste management system.

Technical path identified; with no show stoppers.

Proposed 5-year engineering design and development program addressing key issues provides direction and provides logical exit criteria.

Positive environmental/societal impact.



Outline

Past and present. Assist waste disposal options. System description. Participation. Outcomes. Agenda.

Charge.

Details provided in following presentations.



The Problem --Nuclear Waste Issue

Issue is nuclear waste disposal -- ATW is a technology option.

ATW can assist waste disposal options -- whether fully buried, monitored and retrievable, or interim storage.

Permanent repository still necessary (but more efficient use of capacity with ATW).



Welcome and Introduction

MIT ATW Technical Review 1998, Jan. 15-16

Stan O. Schriber Deputy Division Director LANSCE Division



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genda

MIT ATW Technical Review MIT Nuclear Engineering Department Plasma Physics & Fusion Center, NW17 Second Floor Conference Room

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Chair - Professor M. Kazimi

January 15, 1998 Thursday

08:30 - 08:45	Informal Gathering							
08.45 - 09:30	Welcome and Introduction	S. Schriber, LANL						
J9:30 - 11:30	ATV Overview	F. Venneri, LANL						
11 30 - 11:45	Break							
11:45 - 13 15	ATV Demonstration Conceptual Characterization	M. Carelli, Westinghouse STC						
15:15 - 14:00	Lunch (Box lunches provided)							
14.00 - 15:00	ATW Nuclear Performance	M. Houts, LANL						
15 00 - 15:15	Break	-						
15.15 - 17:15	ATW Process Chemistry	M. Williamson, LANL A. Schake, LANL						
January 16, 1998 Friday								
28.30 - 29:30	Accelerator Design for ATW	G. Lawrence, LANL						
ng 30 - 12:15	Liquid Lead Technology and Engineering	Y. Orlov, A. Dedoul, IPPE						
11:09 - 11:15	Break							
	Continue Liquid Lead Technology	N. Li, LANL K. Woloshun, LANL						
12:15 - 13:15	Program Plan and Summary	F. Venneri, LANL						
13 15 - 14:00	Lunch (Box lunches provided)	Executive Session						
·-··· 15.00	Collaborator's Comments - Further Details as Requi	ested						
15-0-17.00	Executive Session							
17 N - 18 O	Verbal Feedback							



ATW Backlog Burndown scenario: Large Reduction in Transuranic Inventory achieved in time comparable to plant's lifetime





Accelerator Drive (Subcriticality) enables versatile and effective Nuclear Waste Destruction





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 longterm 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.

<u>3. LINAC</u>

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 because there should be no problems with the required field levels in the cavities and previous tests with these lower beta structures showed their benefits.

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 355m 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 ireatment - 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. The central development issue for process chemistry is to establish process scaling information by designing, fabricating, and testing various separation systems and then using that information to develop a more detailed material balance for the fuel treatment processes 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 [8]. The following sections provide a brief overview of process chemistry and fuel technology for the ATW system.

4.1 Process Chemistry

The flow sheet, shown in Figure 2, gives an overview of the flow of material from a spent fuel storage facility to the repository. 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 found at the front end of the flow sheet is a mechanical process that chops 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 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 teclinetium 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.

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Technetium is removed from the electrorefining anode heels and sent to the ATW system for transmutation. Tc is separated from the anode heels by oxidation of the anode metals followed by the distillation and condensation of Tc_2O_7 . Tc_2O_7 is converted to Tc by a direct oxide reduction process. 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 Te recovery process, and ultimately sent to the repository. Periodically the TRU content in the transport molten salt is decreased by using the aforementioned electrorefining salt by reductive extraction techniques and are collected, oxidized, packaged, and sent to the repository.

Although electrorefining is used in both the front-end U removal and back-end TRU recycle process, differences exist between the two systems. The front-end system is larger than the back-end system, about 400 kg of U is electrotransported in the process as compared to about 8-10 kg of TRU in back-end process. The process parameters, cell operating current and process time, are quite different. Moreover, the back-end system must be designed to process Pu-rich mixtures of TRU instead of low enriched U. Scaling of the electrowinning systems follow that of the electrorefining system.

4.2 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. Much like other development metallic fuels, ATW fuel will require both irradiation and materials compatibility testing. Specific issues include fuel swelling, burn-up limits, fission product, especially fission gas, in-growth, fuel / clad interactions, and fuel / clad bonding materials.

5. LEAD-BISMUTH EUTECTIC 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 (0.01mg/m³ in Russia, 0.03mg/m³ 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.

A schematic of the ATW nuclear subsystem is shown in Figure 3. The choice of the pool configuration is consistent with the objectives to use proven solutions (the pool configuration was chosen over the loop option for LMRs worldwide) and to maximize safety. The actinide-containing region is 2 m high by 2 m in diameter. An 0.6 m diameter central region contains LBE that is used as the spallation target. The top of the target is located 0.15 m above the mid-plane of the actinide-containing region, and a window separates the inside of the beam tube from the LBE. Passive systems are used to ensure that if LBE temperature exceeds that expected during normal operation the beam tube will be flooded with LBE, effectively removing the neutron source from the actinide-containing region and shutting down the transmutation process.

An intermediate loop could be avoided to reduce cost and in fact some Russian designs place a steam generator directly inside the primary pool. However, it was decided to adopt an intermediate heat exchanger in the ATW concept to contain polonium (produced by neutron capture in bismuth), spallation products and other radioactive isotopes. The secondary coolant is non-radioactive LBE. A minimum 1 m thick LBE reflector surrounds the waste assemblies on all sides. This reflector helps minimize required actinide loading, flattens the power density across the fissioning region, shields the vessel walls from fast neutrons, and provides thermal inertia. A core map is provided in Figure 4.

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].

In the US. Brookhaven National Laboratory first tried to use liquid lead-bismuth as a coolant, attempting to protect the container walls from corrosion by adding inhibitors, such as zirconium, possibly forming a surface layer of zirconium nitrate or carbide [11]. This solution was not satisfactory because of the difficulty in maintaining a consistent protective layer during operations. After extended study, the US project for a lead-cooled submarine reactor was abandoned.

The surfacing of viable Russian lead 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. AN IMPLEMENTATION SCENARIO

The dedicated nature of ATW and the flexibility afforded by subcriticality combined to give us many possibilities for implementing ATW for effective waste destruction. We will examine one possible implementation scenario. Many others can be developed similarly [9].

6.1 Spent Fuel Backlog Transmutation in 65 Years

By the year 2015, there will be 70,000 tons of US spent fuel containing about 600 tons of plutonium and higher actinides requiring disposal. The ATW objective is to treat the spent fuel backlog, destroy the transuranics and selected fission products, and prepare the resulting waste for permanent disposition in a geologic repository within a relevant time horizon.

Twenty one ATW systems are brought on line to accomplish the objective over a 65 year period. Each ATW system consists of an accelerator (1 GeV, variable current 20-40 mA), a subcritical 2000 MWt LBE burner, and a pyrochemical plant (50 tons/year throughput per burner). Several of these ATW systems could be located together in one facility, and we have examined the possibility of having three ATW facilities. The parameters for the ATW burners are as follows:

Fission Power: 2000 Mwt; Thermal to electric efficiency : 40%;

Power to Grid: 700 MWe; Recirculated power: 100 MWe (67 to accelerator, 33 to plant operations); Operational TRU Inventory: 3000 + 1000 kg; ConversionRratio: 0.0: Process losses: 1/1000; TRU Burn Rate: up to 650 kg/yr.; TRU Burn efficiency (Rate/inventory): 16.25%.

Of particular relevance is the fact that each 2000MWt ATW burner can destroy up to 650 kg of actinides (plutonium and transuranics) per year, and that 700 MWe would be available for distribution to the grid from each burner, after powering the driving accelerator and the plant. In the three-facility scenario, every four years a new ATW system is brought on line at each site. As the burners and accelerators reach the end of their operational life, they are decommissioned, and the fuel from the decommissioned burner is sent to feed other still operational burners. Eventually, the last 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 (Fig. 5.6).

There are two major characteristics of the ATW that distinguish the accelerator-driven subcritical systems from critical reactors that had been evaluated for actinide destruction, such as the ALMR: (1) ATW does not produce higher actinides from a fertile material such as U-238; (2) because ATW is not constrained by a criticality requirement it can burn down the end-of-life (EOL) actinide inventory to as low a level as desired and practical.

6.2 Cost considerations for the burn-down scenario

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. ACKNOWLEDGMENTS

The authors wish to thank Wolfgang Barthold and Mario Carelli for important contributions to the ATW concept. This research is funded by Los Alamos National Laboratory LDRD. The internal Los Almos Report number for this paper is LA-UR 98-985.

9. FURTHER READING

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

[11] J. R. Weeks and C. J. Klamut, Nucl. Sci. Eng. 8, 133 (1960).

Agenda for Two Days

Thursday		
08:45 -09:30	Welcome and Introduction	S. Schriber
09:30 -11:30	ATW Overview	F. Venneri
11:45 -13:15 M. Carelli, W	ATW Demonstration Conceptual estinghouse STC	Characterization
14:00 -15:00	ATW Nuclear Performance	M. Houts
15:15 -17:15 Schake	ATW Process	M. Williamson; A.
Friday		
08:30 -09:30	Accelerator Design for ATW	G. Lawrence
09:30 -12:15 Li; K. Wolos	Liquid Lead Technology hun	Y. Orlov, IPPE; N.
12:15 -13:15	Program Plan and Summary	F. Venneri
14:00 -15:00	Bechtel, N-G, Sandia, LLNL	LANSCE

Charge

Comment on ATW technology choices and status.

Provide feedback on where emphasis should be given within the program.

In what areas do you have concerns and issues? What recommendations do you have?

Comment on five year program -- elements, goals and possible accomplishments.



"Reducing the Impact of Nuclear Waste"

ATW Overview

Francesco Venneri ATW Project Leader

Los Alamos National Laboratory -- ATW Project --
ATW addresses SPENT FUEL MANAGEMENT issues:

Plutonium Accumulation Long-term Uncertainties

• Efficient use of Resources

How ATW assists Nuclear Waste Management

- Efficient destruction of transuranic waste produced in reactors
 Transmutation of long-lived fission products of impact to
- repository performance
- Full utilization of energy content in nuclear waste for power production



ATW and spent fuel: what, why

why: what:	water solubility and transport	criticality	source term	diversion	problem daughters	heat
Cladding		· · · · · · · · · · · · · · · · · · ·	1			
Uranium	hioh	law	low Dow	 Konos	lich	top.
Plutonium	low	hioh	híoh	hioh	high	hiah
Neptunium	high	lów	high	high*	hĩgh	low
Other Act.	low	low	high	low	high	high
Tc, I	high	* er =	low	-		low
Sr, Cs	high*		high*			high*
Noble metals	low		low			low
Lanthanides	low	••-	low	low	low	hìgh*
Volatiles			high	··	.	low

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ATW and spent fuel: how

Cladding	ATW plans to use the LWR cladding material (zircalloy) as matrix for its fuel elements (80-90% Zr atom fraction with remainder transuranics).			
Uranium	Uranium is separated from spent fuel, for future use in reactors or disposal.			
Plutonium	All plutonium isotopes are fissioned. Diversion concerns are eliminated. Long term heat source is eliminated.			
Neptunium	Neptunium is destroyed. ^{23°} Np is the worst contributor to aqueous dose releases.			
Minor Actinides	ATW destroys all minor actinides together with plutonium and neptunium. No buildup of heavy actinides occurs			
Technetium, I	ATW isolates technetium (we know how to do this) and iodine (we are working on it) and transmutes them into stable elements. No isotope separation is required.			
Strontium, Cs	ATW separates Strontium and Cesium (high heat producers). These are stored in optimized engineered wasteforms. • short term large heat release rate has no effect on the remainder wasteforms containing longer half-life residuals			
Noble metals Lanthanides Volatiles	ATW separates the noble metals, lanthanides and volatiles into specific wasteforms.			
	1			

Effect of Transmutation on Thermal Power of spent LWR fuel

0.1% cumulative process losses





Ξ.

Decay Time after Discharge (years)

ATW features that make a difference in dealing with nuclear waste destruction

- no new TRU production via neutron capture
- near-complete burndown of TRUs at end of life
- low-loss fuel recycle processes

ATW compares favorably to other S+T schemes:

•ATW has better transmutation performance faster, more complete, requires smaller investment

•ATW has simpler processing requirements

smaller number of steps, more proliferation resistant, less waste

details will be provided in the course of the Review

Performance Drivers in the ATW concept design :

Fast Burn Rates, Low Inventories

Capability for rapid materials inventory reduction Minimized economic impact



Engineering design clarity, defense in depth. inherent safety

Proliferation Resistance, Low Environmental Impact

Restraints (Constraints)

No Liquid Fuel

No On-line Processing

Drivers provide basis for ATW systems and technology choices

Fast Burn Rates, Low Inventories	Fast Spectrum, Subcritical Operation, Variable Current Drive, EOL burndown,		
Safety	Conversion Ratio = 0, Tc burnable poison Liquid lead-bismuth coolant/target, solid fuel		
Proliferation Resistance, Low Environmental Impact	Limited Batch Pyroprocessing		

Fast spectrum operation offers considerable neutronic advantages



- Efficient neutron production and utilization
- Avoidance of buildups of the very high actinides (Am. Cm, Bk, Cf)
- Fast spectrum eliminates need for rapid fission-product poison removal
- Leakage neutrons can be thermalized for FP transmutation (if necessary)

Accelerator Drive (Subcriticality) enables versatile and effective Nuclear Waste Destruction



why subcriticality has advantage for waste destruction:

• Power control is <u>not linked to</u> <u>reactivity feedbacks, delayed</u> <u>neutrons or to control rods</u>, but only to the accelerator drive

• ATW has <u>no need for fertile</u> <u>materials</u>. ATW uses pure transuranic cores

• Subcritical systems work independently of the fuel composition

•<u>EOL inventory is not limited by</u> <u>criticality</u>. Possible to have EOL burndown of inventory

 Neutronics and thermohydraulics are effectively <u>decoupled</u>





The insertion of 1.0\$ of reactivity in ATW systems only increases the steady state power level by ~5%



Increased coolant temperature



Because of slow and self limiting response to reactivity changes, and because of strong negative feedback of coolant temperature. ATW can use cores with little or no negative Doppler (high fraction of transuranics)

Variable accelerator drive provides constant power operation under extended burnup, without fertile materials



.... we don't have to resort to liquid fuels

Tc as Burnable "Poison"



Molybdenum Technetium alloy can be cast into rods suitable for irradiation (other possible) Ruthenium does not need to be processed because of low cross section

ATW Fuel Elements

10-15% Transuranics

Drivers: non-fertile actinide based fuel, high melting point. compatible with pyrochemical processes. recycle of waste material. existing experience.

Selection: Transuranic metal fuel in inert Zirconium matrix.

- composition 85-90% Zirconium,
- melting point ~ 1400 °C
- 316 SS clad, lead bond (He also possible)
- excellent solubility of actinides in Zr phase
- phase equilibria established
- prepared by vacuum casting
- utilizes zircalloy discard cladding from spent fuel
- considerable experience exists with metallic fuel
 - ~ 80%Zr 20%U Naval Fuel
 - ~ 75%U 15%Pu 10% Zr IFR Fuel

Issues:

Irradiation Behavior

Swelling

Bonding compatibility

Fuel bundle geometry

.... Backup: Oxide fuel

The choice of liquid lead/bismuth (LBE):

- 1. Very hard neutron spectrum
- 2. No violent reactions with air or water
- 3. Low melting, high boiling points. Low vapor pressure. Large *\t*. possible.
- 4. Excellent medium for natural convection (essential for passive safety)

5. Very low neutron absorption. Large P/D ratios are possible. Very low core pressure drops

- 6. Materials compatibility established
- 7. Very effective gamma shielding material and spallation neutron source

Early problems with LBE nuclear systems (corrosion of structural materials, oxygen balance, handling of Polonium) have been successfully solved (Russian experience)

LBE systems are a natural choice for subcritical configurations driven by accelerator beams. LBE is an excellent spallation neutron source and nuclear coolant. One-fluid systems can be readily designed.

ATW uses the same fluid for target and core cooling -- no target structure



2

Lead as spallation neutron source has been succesfully tested on large scale at Los Alamos, codes benchmarked







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Design Objectives for ATW Accelerator

A linac for an ATW facility should be designed:

- To have very high electrical efficiency (ac to beam power)
- For minimum capital and operating costs
- For minimum spatial footprint (short length)
- To vary the power on target over a wide range
- To have high availability and operational flexibility
- To use the best mix of established technology (APT) and anticipated technology advances

--- High-Gradient SuperConducting (HGSC) Linac

Nominal beam parameters:

40 MW proton beam 1000 MeV 40 mA 100% duty

THE CHOICE OF PYROCHEMISTRY : Waste Treatment Technology

The OBJECTIVES:

- Rough separation of Uranium (95-99%) from the spent fuel prior to introduction in the ATW burner
- Periodic recycle of the ATW fuel during operations
- Periodic cleanup of the processing medium
- PYROCHEMISTRY ALLOWS FAST, BULK-TYPE SEPARATIONS.
- IT IS NOT ADVERSELY AFFECTED BY HEAT OR RADIATION.
- IT PRODUCES VERY LOW AMOUNTS OF SECONDARY WASTE.

NONE OF THE PROCESSES PROPOSED PRODUCES "NAKED" PLUTONIUM.



ATW Waste Forms provide FP partitioning for good Repository performance

Transition Metal Oxides (contain reisual TRU) Rare Earth Oxides Active Metals in engineered storage Xenon, Krypton Assemblies, hardware Uranium

repository repository repository gas storage repository LLW

ATW Design Objectives

Subcriticality level: such that nature of the fuel is unimportant for operation of the system. We have 0.967 as our highest level.

Power level: economy of scale important. Maximum allowable beam size important. Proton damage to window might be driver. Windowless designs could play important role. We select 2000 MWt, with a beam drive 20-40 mA at 1 GeV.

Accelerator: 40 mA, 1 GeV. Operates at 75 % rated power (ramp 20-40 mA).

Power density: high power density means low inventory. Also short cycle length. We select 300 w/cc, 7 kw/ft, 120 day cycle, 3000 kg inventory

Passive cooling: 300 MW natural convection capability (15% of rated power)

Process rates: these are governed by the power density. 60 day wait, 60 days to process 1/3 of core. Process inventory is 1000 kg TRU.

Burndown: 5-year special operation. Objective is EOL inventory < 300 kg.

A Specific ATW Scenario:

Treatment of 70,000 tons Spent Fuel Backlog

ATW Burner Parameters for Scenario

Fission Power: 2000 MWt

Operational TRU Inventory: 3000 + 1000 kg TRU Burn Rate: 650 kg/yr TRU Burn efficiency (Rate/inventory): 16.25% Conversion ratio: 0.0 Thermal to electric efficiency : 40%. 800 MWe Recirculated power: 100 MWe (67 to accelerator, 33 to plant operations) Power to Grid: 700 MWe Process losses: 1/1000

ATW implementation scenario

Number of ATW units, coming on line, closing down

Time (years)



1 ATW burner (40 years operational life, 2000 MWt)

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Large Reduction in Transuranic Inventory achieved in time comparable to plant's lifetime



The key to the vast improvement over the use of ALMRs is the ability to handle pure transuranic cores. without fertile material and to reduce EOL inventory beyond criticality ATW implementation scenario

 Ψ = Pigford-Choi TRU reduction factor = TRU in original waste / current TRU inventory



Campaign time (years)

Rough Costs for Scenario

Baseline Assumptions;

CAPITALCOST			· · · · · · · · · · · · · · · · · · ·
Component	Unit cost (\$M)	Quantity	Total Cost (\$M)
Waste Treatment	\$1,000	1	\$1,000
Cleanup Facility	\$500	11	\$500
Accelerators	\$500	20	\$10,000
Subcritical Burner	\$750	20	\$15,000
Power Conversion & Distribution	\$750	20	\$15,000
	TOTAL CAPITA	L CHARGE ==	\$41,500
0&M	\$1,245	1	\$1,245
Electric Sales Thermal power per burner (MWI	2,000	20	40,000
coversion ratio		40%	
Electric power MWe - gross	·		16,000
Unit power per accel installtion	100	20	2000
	Net power for sa	ale ==> (MWe	14,000
Cost	of electricity (mils	/kWe-hr) ==:	30
Plant_availability ==>			- 75%
Annual rev	enue from Electric	Sale ==> (\$M) \$2.76 t
Net a	nnual revenue for A	TW option ==:	\$1,516

Rough Costs for Scenario



ATW waste destruction could provide economic benefits


	ALMR* (2000-MWt parks) each park is 4 modules	ATW (2000-MWt units)	
TRU initial	600 tons	600 tons	
TRU inventory oper unit of park. Core+ Process	$7720 + 5150 = 12.870 \ kg$	3000 + 1000 = 4000 kg	
Breeeding Ratio	ō.76	0.0	
TRU destruction rate (75% avail.)	130 kg/yr	650 kg/yr	
TRU destruction ratio (rate/inventory)	1.01 °c/yr	16.25 Gdyr **	
Decontamination Factor	1000	1000	
Campaign time	$\sim 320 \text{ yrs}(\Psi > 15)$	$\sim \underline{65 \ yr} (\Psi > 200)$	
# required (ALMR parks or ATW units)	33+24+17+12+9+ = <u>110</u> ***	<u>20</u> ***	
waste to repository	5500 tons	3000 tons	
total electricity produced (40% eff.)	~ 40.E+12 kw-hr	~ 4.0E+12 kw-hr	
residual TRU inventory	~ 39 tons	< 1 ton	
$\Psi = P_{1}g_{1}^{ford}$ -Chei factor = TRU in original wa	ste / current TRU inventory		
* from STATS panel report			
** >50% during burn-down phase			
*** 40-year plant life			

Other Scenarios

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ATW can be efficiently deployed to complement many different fuel cycles and waste management scenarios



<u>ATW Support Ratio</u> = the number of reactors whose actinide waste is destroyed by one ATW system of the same thermal power

* large fraction of ²²⁸Pu ** large fraction of minor actunides

What You Should Remember About ATW

- It is a Nuclear Waste Management enhancing technology
- Addresses issues of repository storage capacity and the longevity of radiological hazards:
 - destruction of all transuranics
 - transmutation of Tc-99 and I-129, other isotopes if required
 - partition of all other fission products for optimal disposal
 - radiotoxicity of ATW waste after 300 years is lower than direct-disposal waste after 100,000 years
- It is reactor-like in scale and function. Produces usable energy by destroying nuclear waste
- Its components are based on proven technology, can be directly transferred to nuclear power production
- Although spent fuel is processed, weapons-usable material is not made available
- It is adaptable to different fuel cycles
- In the mission of waste destruction, it is significantly faster and more complete than the other S+T schemes

ATW Development Road Map

where we are, 1998: Technology Assessment	5-year Development Plan Technology Verification and Small-scale Integration	where we are going: Large scale Integration and Implementation
 Accelerator technology status: APT-driven, ATW specific features could be developed from APT Liquid lead technology status: early stage in US, Russian technology fully developed 	 Nuclear design (including Safety) Integrated small scale waste processing plant Fuel development Russian liquid lead technology transfer High power spallation targets 	ATW Subcritical Testbed Facility (STF) at LANL 10-50 MW thermal power ATW Process Facility (APF) >100 kg scale
• Pyrochemistry status: individual parts well developed, Argonne and LANL have expertise	 Materials verification ATW-specific accelerator design Systems studies 	DEMONSTRATION of Accelerator-driven Waste Transmutation system 10-20 MW beam 500-1000 MWth
	99 2	004

ATW Consists of Three Major Functional Blocks





Pyrochemical Processes

Proliferation resistant, low environmental impact







Power to Accelerator: ~10%

ATW Design Objectives

Subcriticality level: such that nature of the fuel is unimportant for operation of the system. We have 0.967 as our highest level.

Power level: economy of scale important. Maximum allowable beam size important. Proton damage to window might be driver. Windowless designs could play important role. We select 2000 MWt, with a beam drive 20-40 mA at 1 GeV.

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Process rates: these are governed by the power density. 60 day wait, 60 days to process 1/3 of core. Process inventory is 1000 kg TRU.

Burndown: 5-year special operation. Objective is EOL inventory < 300 kg.

Many engineering, technological and safety problems of ATW are common to reactors. Russian experience will be very valuable in development of design.

Engineering	Variants	
-------------	----------	--

Coolant Type	Pb-Bi	Pb
Target design	Windowed	Windowless
Target cooling system	Separate circuit	Same as blanket cooling system
Number of circuits	2	3
Coolant circulation	Forced circulation	Natural convection
Fuel type	Oxides	Metal
Fuel rod type & fuel assembly design	Spacing by rib wrap	Spacer grid

Safety Considerations

Freezing/defreezing coolant technology	
Steam generator tube rupture	

PO decontamination Oxygen level control Loss of heat sink accident Loss of coolant circulation

Minimized-RISK APPROACH for ATW DEVELOPMENT

	Beam Power	Fission Power	Power Density	Fuel Type	Neutron Flux	Chemistry
LANL-IPPE Target (1999 - 2001) no fissionable material. Competency development. High radiation environ. Window testing, TH stressed	I MW		500 w/cc		>10**13	Oxygen control
Subcritical testbed (2001-2010) <u>Limited fissonablé inventory</u> . Could use LANSCE. TRU fuel development + test.	1-2 MW	10-50 MW 0.75-0.9 keff	100 w/cc	UO ₂ (20% enr.) metal fuel-test	>10**14	Po removal Oxygen control
ATW demonstration (2007 -) <u>Conservative approach</u> . Forgiving Operation, Could be at SRS/APT. Chemistry, FP transmutation., full TH	10 - 20 MW	500-1000 MW 0.95 keff	100 w/cc	metal fuel U - TRU core	>10**15	fully integrated waste treatment +transmutation
ATW full size burner (2015 -) Full TRU destruction performance. Accelerator load following, extend, burn.	20 - 40 MW	2000 MW 0.97-0.93 keff	350 w/cc	metal fuel full TRU core	>10**15	fully integrated waste treatment +transmutation

SUBCRITICAL REACTOR CONCEPTUAL CHARACTERIZATION

THE ATW DEMONSTRATION PLANT

Mario D. Carelli, Tom J. Fagan, Larry Green. Dmitry V. Paramonov, and Wayne L. Chase Westinghouse Science & Technology Center

M. Houts

Los Alamos National Laboratory

A presentation to MIT. Nuclear Engineering Department January 15-16, 1998



OUTLINE

- Approach
- Point Design
- Preliminary Performance Calculations
- Future Effort



HEAVY METAL (LEAD BISMUTH EUTECTIC) COOLED REACTOR

- Transfer Soviet technology (Alpha class submarines)
- No need for separate target
- Potentially safer than either water or alkali metal cooled reactors
 - Very high boiling point
 - No pressurization needed
 - No violent reaction with air or water
 - Self-plugging leaks (melting point higher than room temperature)
 - Natural circulation potential under normal operation
 - Can be designed to be passively safe

APPROACH

• Minimize development

- Russian cooperation
- Utilize water and sodium reactors experience
- Safety/design conservatism has precedence over economic performance for demo
- Outline potential improvements for future designs



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WHY A DEMO

- Demonstrates new technology
- Identifies development needs
- Easily scaleable to prototype
- Design and operation experience
- Provides realistic test bed

DESIGN CHOICES

- Pool configuration
 - LMR experience
 - Safety
- Intermediate heat exchanger (IHX)

Polonium and other radioisotopes containment
 Nuclear steam turbine

- LWR experience
- More compact IHX and steam generator
- Margin for upgrading



ATW REACTOR SCHEMATIC



DESIGN CHOICES (Cont'd.)

- Hexagonal canned assemblies
 - LMR experience
 - Orificing to offset radial power distribution
 - VVER and thermal breeder experience for triangular pitch grids
- 1000 MWt reactor design
- Core inlet/outlet temperatures 340/510°C
 - Same as Russian design
- "Diluted" core

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- Enhance natural circulation
- Moderate power density

VVER (TEMELIN) FUEL ASSEMBLY (ATW ASSEMBLY HAS 91 PINS)



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ATW REACTOR SCHEMATIC



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COMPARISON OF COOLANTS FOR NATURAL CIRCULATION

Coolant	Reactor	Tin/Tout (°C)	Driving	g Head (Δρ)	Heat Ca	pacity (pc _p)	Margin (Ti	n to Boiling _{Soit} -T _{out} i
			g/cm'	normalized	J/m ³ K 10 ⁶	normalized	°C	normalized
Pb-Bi eutectic	ATW	340/510	0.205	1	1.53	1	1160	1
Рь	BREST	-420/540	0.146	0.71	1.50	0.98	1185	1.02
Na	CRBR	388/535	0.036	0.18	1.08	Ū.71	350	0.30
H <u>-</u> O	4 loop PWR	283/325	0.073	0,36	2.93	[.9]	18	0.02

CORE GEOMETRY

Outer diameter		3 m
Inner diameter		0.8 m
Height		2 m
Hexagonal assemblies, triangular pitch		
# assemblies		416
# pins/assembly		91
Pin diameter		0.8382 cm
p/d	1.69	
Cladding thickness		0.46 mm
Pellet diameter		0.7068 cm
Gap (metal bond)*		0.2 mm
Can flat-to-flat		13.5 cm
Can thickness		3 mm
*II. and alternative is considered		

*He gap alternative is considered



Steady-State Demo-ATW Radial Power Profile

Relative Power Density vs Radial Position (cm)



Steady-State Demo-ATW Axial Power Profile

Relative Power Density Vs Axial Position (cm)



PRELIMINARY LIMITING TEMPERATURES

Coolant	None (boiling point is higher than all other limits)
Vessel	700°C (Russian data)
Cladding	800°C (Russian data)
Fuel	1200°C (LANL, Zr matrix)



PRELIMINARY UNCERTAINTY FACTORS

Enthalpy Rise	1.8
Film ΔT	2.0
Cladding ΔT	1.5
Gap ∆T	2.0
Fuel ΔT	2.0
Radial peaking factor not	1.1
compensated by orificing	

Sec. New York

Westinghouse Science & Technology Center

 $(1+1)_{i=1}^{n-1}$

PIN RADIAL TEMPERATURE PROFILE



Science & Technology Center

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HELIUM GAP TEMPERATURE DROP (NOMINAL)



Westinghouse Science & Technology Center

INTERMEDIATE HEAT EXCHANGER

- 2 halves of 60 x 70 rows 4200 tubes (2.23 cm diameter x 2.14 m length) each on a triangular pitch - P/D = 1.5
- Overall dimensions: 2.58 x 2.14 x 5.22 m
- H-27 steel (3 Cr, 0.8 Mo)
- Primary fluid (outside tubes) velocity = 0.463 m/s, ΔP= 3.6 psi
- Secondary fluid (inside tubes) velocity = 0.921 m/s, $\Delta P = 5.9$ psi



PRIMARY SYSTEM HYDRAULICS SUMMARY

	· · · · · · · · · · · · · · · · · · ·	s Alson See Age
Paramatar	Pumpe On	Pumps Off
Core Exit Temperature (°C).	510	631
Core Temperature Rise (°C)	170	295
Mass Flow Rate (kg/sec)	3.96x10 ⁴	2.32×10^4
Core Pressure Drop (psi)	5.0	1.9
Core Flow Velocity (m/s)	0.79	0.47
IHX Pressure Drop (psi)	3.6	1.3
IHX Flow Velocity (m/s)	1.0	0.6
Piping Pressure Drop (psi)*	4.6	1.7
Pump Pressure Drop (psi)	-	1.0
Total Pressure Drop (psi)	13.2	5.9
Convection Head / Total Pressure Loss	0.25	1.0
Pumping Power (MWe)	0.73	-

Conservatively estimated at ~50% of total calculated value for core and IHX

 $\sum_{i=1}^{n} \frac{1}{2} \sum_{i=1}^{n} \frac{1}{2} \sum_{i$

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PUMP FAILURE ACCIDENT -ESTABLISHMENT OF NATURAL CIRCULATION



BEAM ON, PUMPS OFF ACCIDENT - TRANSIENT THERMAL RESPONSE (NOMINAL CONDITIONS)



01/15/98

Loss of Heat Sink Accident (Beam Off, IHX Off, Pump Off)



PASSIVE SAFETY CORE "SHUTDOWN"



FUTURE HIGH PRIORITY ANALYSES

- Detailed core physics/power distribution
- Core orificing
- Reactivity insertion transients
- Window design
- Reactivity swing over lifetime
- Fuel management

01/15/98
SUMMARY CONCLUSIONS

- Established Design Philosophy and Exercised Design Choices
- Taken Maximum Advantage of Past Experience
- Established Necessary Framework for Technology
 Transfer
- Preliminary Performance Calculations are Very Encouraging
- Excellent Passive Safety Capabilities and Potential
- Outlined Plan for Next Step in Design

ATW Nuclear Performance

Los Alamos National Laboratory - ATW Project -

Presented by: Michael G. Houts Nuclear Systems Design and Analysis Group

Prepared with: David I. Poston and Holly R. Trellue

Outline

- ATW System Description
- Experience Base for the ATW
- ATW Safety
- ATW Waste Cycle
- Technetium Transmutation
- End-of-Life (EOL) Inventory Reduction
- Research Needs
- Conclusions

ATW System Description

- The ATW point design is not yet optimized, but is intended to meet ATW goals:
 - Less than 3000 kg steady-state actinide inventory.
 - Greater than 500 kg/year actinide burnup.
 - Greater than 40 kg/year technetium transmutation.
- The ATW point design has a lower inventory and a higher burnup rate than the ATW Demo.
- The ATW Demo and the Subcritical Testbed Facility provide the data and experience necessary to build the ATW.

ATW Builds on the ATW Demo

Parameter	ATW Demo	ATW
Coolant	44.5%Pb/55.5%Bi	44.5%Pb/55.5%Bi
Power (MWt)	1000	2000
Fissioning Material	(Np/Pu/Am/Cm)Zr	(Np/Pu/Am/Cm)Zr
Power Density (MW/liter)	0.076	0.34
Vessel Material / Clad Material	316 SS / 316 SS	316 SS / 316 SS
Average Linear Heat Rate (kW/ft)	6.4	7.3
Fuel Cycle, Nominal (Full Power Days)	360	100
Actinide Inventory, no Tc Burning (kg)	4900	2400
Actinide Inventory, with Tc Burning (kg)	6400	2960
Average Coolant Velocity (m/s)	1.2	< 5
Prompt Reactivity Feedback	Near-Zero	Near-Zero
Overall Reactivity Feedback	Negative	Negative

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The ATW Demo will have Demonstrated Key Requirements and Features of ATW

- Beam transport and beam window.
- Flowing LBE target.
- Assured beam shutoff (or removal of spallation source from waste region).
- Operation with small delayed neutron fraction.
- Operation with near-zero prompt reactivity feedback.
- Operation with uncertainties in the composition and physics of the waste.

In addition to utilizing Russian LBE reactor experience, ATW also stays within relevant FFTF

Parameter	FFTF	ATW
Power Density (MW/liter)	0.39	0.34
Vessel Material / Clad Material	304 SS / 316 SS	316 SS / 316 SS
Average Linear Heat Rate (kW/ft)	7.3	7.3
Fuel Cycle, Nominal (Full Power Days)	100	100
Demonstrated Capacity Factor	0.75	Not Known
Average Coolant Velocity (m/s)	6.4	< 5
Coolant delta T (C)	170	250
Prompt Reactivity Feedback	Negative	Near-Zero
Overall Reactivity Feedback	Negative	Negative
Coolant	Sodium	44.5%Pb/55.5%Bi
Power (MWt)	400	2000
Fissioning Material	25%PuO ₂ /75%UO ₂	(Np/Pu/Am/Cm)Zr

experience.

ATW has Excellent Neutronic Safety Characteristics



ATW retains subcriticality with up to 13% volumetric compaction.

ATW has Excellent Neutronic Safety Characteristics



Large reactivity insertions result in small power increases. Fission power rapidly drops to zero if source is removed.

Simple Waste-Burning Scenarios give Adequate Performance

- 3-zone scheme has adequate performance (improvements possible).
- "Fresh" waste pins placed in Zone 3, moved to Zone 2 after 1st burn cycle and Zone 1 after 2nd burn cycle.
- After burn cycle in Zone 1, waste is processed, actinides concentrated to obtain desired reactivity, and waste refabricated into pins. Then waste inserted into Zone 3.

Simple Waste-Burning Scenarios give Adequate Performance



Reflector

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ATW Steady-State Waste Cycle

Isotopics of Steady-State ATW Batch (2-GW, 4 month burn cycle)

Batch moves R3-R2-R1: then actinides concentrated and enriched with LWR discharge, and moved into R3 (the difference between the first and last bar is the SS feed rate, except for U238)



PWR vs ATW Discharge

Isotopics of PWR Discharge vs. ATW SS Discharge



Variable accelerator drive provides constant power operation under extended burnup, without fertile materials



.... we don't have to resort to liquid fuels

ATW has an Acceptable Power Profile

Relative Power Density vs Radial Position



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ATW has an Acceptable Power Profile

Relative Power Density Vs Axial Position (cm)



ATW Efficiently Transmutes Technetium

- Increases actinide loading and acceptable cycle length.
- Hardens neutron spectrum if unmoderated, increasing eta and reducing build-in of higher actinides.
- Moderated targets allow use of Tc as burnable poison, and increase Tc transmutation rate.

ATW Efficiently Transmutes Technetium

- Tc transmutation rates greater than the required rate (40 kg/yr) are easily attainable.
- Highest transmutation fraction attained by closely mixing Tc with moderator pins.
- More than adequate Tc transmutation feasible through use of moderated targets radially surrounding the fissioning region or possibly Tc impregnated end-reflectors within the fuel pins.
- Fast-spectrum transmutation of Tc in the fissioning region also has reasonable performance.

Effect of Tc Addition in Actinide

Actinide Mass Fractions with and without Fast-Spectrum Tc Transmutation in Actinide Region



Effect of Tc Addition in Actinide Burning Region

Curium Fraction in Fuel with and without Fast-Spectrum Tc Transmutation in Actinide Region



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ATW Inventory Reduced at EOL

- When a plant reaches end of operational lifetime (EOL). residual actinide inventory transferred to remaining plants.
- The final remaining site (Two 2 GWt plants) will be used for burn-down.
- As inventory drops, actinides are concentrated, waste region size reduced, and power is reduced as needed. Fast spectrum maintained as long as practical.
- Moderator introduced at EOL to attain very low (100 kg) residual inventory while maintaining reasonable multiplication.
- Reduce residual Np and Tc inventories to less than 5 kg.

Waste Mixtures with no Plutonium can be burned.



Research Needs from Subcritical Test Facility and ATW Demo

- Fuel (Waste) Performance (Demo)
 - High actinide burnup.
- Fuel (Waste) Handling (Demo)
 - Rapid shuffling, removal, and insertion.
- Materials (STF)
 - Relatively high LBE flow velocity and delta-T.
- Interface between beam and waste burner (Demo & STF).
 - High beam power, beam power density.

Summary

- A logical path exists for developing ATW systems.
- ATW has excellent neutronic safety characteristics.
- ATW burns actinides efficiently.
- ATW transmutes technetium efficiently.
- ATW has a low EOL actinide inventory.
- ATW fuel is well suited for processing.
- ATW reduces actinide and technetium waste inventories by 3 orders of magnitude.

ATW Process Chemistry

Los Alamos National Laboratory - ATW Project-

Presented by: Mark A. Williamson Nuclear Materials Technology Division

Outline

- ATW process chemistry requirements
- ATW waste treatment flow chart
- Process chemistry simulations
 - LWR spent fuel
 - ATW waste treatment
- Waste streams
- Technetium and ATW
- Conclusions

AT N Process Chemistry Is Proliferation Resistant and not Complex

- Transuranics are separated as a group for burning in ATW, no weapons usable material is produced during processing
- Technetium and iodine are separated for transmutation in ATW
- Secondary waste minimized
- Segregated waste materials produced for storage

ATW Waste Treatment



A' W Process Simulation Routines Benchmarked with Experimental Data

- SOLGAS equilibrium thermodynamic calculations based on free energy minimization, routinely used in process simulations for 25 yrs
 - Direct Oxide Reduction (DOR)
 - DOR salt regeneration
- TRAIL mass transfer modeled with diffusion layer theory, Kobayashi et al., J. Alloys and Compounds <u>197</u>, 7 (1993), code written by Y. Hu and N. Li, verified with U electrorefining data
 - Electrorefining
 - Electrowinning

LWR Waste Treatment Flow Chart



LWR Waste Treatment Simulation

- One PWR fuel assembly
- 33 MW day / kg average burn-up
- 150 days cooling time
- Composition data from Nuclear Chemical Engineering, Benedict, Pigford, and Levi

Spent Fuel Decladding Produces Feed Material for Oxide Reduction Process

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- Support hardware removed from assembly
- Pins chopped by traditional methods
- Oxide fuel separated from clad material, sent to oxide reduction process
- Clad used as source of Zr for ATW fuel
- Offgas released by decladding process is collected, sent to storage

Oxide Fuel Converted to Metal by Direct Oxide Reduction Process



Four 100 kg HM batches are required to treat one PWR fuel assembly

Electrorefining of PWR Assembly Provides Uranium Separation



Four Batches of 99.5 kg HM with U3+ re-enrichment to 6% in molten salt

Folishing Anode from Electrorefining Process Reduces TRU Discharge



Material from one PWR assembly
Electrowinning Provides Feed Material for ATW Fuel Fabrication



Material from one PWR assembly

Fuel Fabrication for ATW system



High Temperature Vacuum Casting Furnace

Reductive Extraction Process Removes Rare Earths from Molten Salt

<u>Input Materials</u> Molten salt from #5 3466 g RE

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1707 g Na as alloy 188 kg NaCl-KCl

<u>Operating Conditions</u> Time = 8 hrs T = 1000 K



Direct Oxide Reduction Salt is Recycled after Chlorination and Calcium Regeneration



Treatment of four batches of DOR salt

Treatment of 70000 t of Spent Nuclear Fuel in 60 yrs Requires 2 t / day / Plant*

System	Batch (kg HM)	Process Time (hr)	Number of Systems	Capacity per day (kg HM)	Operating Parameters
Decladding	400	24	6	2400	
DOR	120	8	6	2160	and the second s
DOR Salt Recycle	121 (Ca)	20	6	726	8000 A (8 kW)
Electrorefinning LWR fuel	400	: 17	5	2000	8000 A (8 kW)
Electrorefinning Anode Polish	3	2	5	15	500 A (500 W)
Electrowinning U/TRU/RE	18	10	5	90	500 A (500 W)
Reductive Extraction	3.5	8	2	21	
Fuel Fabrication	5.4	12	5	27	
Offgas System	Xe and Kr cryodistillation and collection		1		

Process Plant Parameters

Two chemical plants are required to treat LV and ATW waste, 300 day/yr - 24 hr/day.

ATW Waste Treatment Flow Chart



ATW Waste Treatment Simulation

- One ATW waste fuel assembly
- 182 MW day/ kg average burn-up
- 60 days cooling time
- Composition from ORIGEN calculations

Chopping ATW Assembly Produces Feed Material for the Electrorefining Process

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- Support hardware removed from assembly
- Spent ATW fuel chopped into small (~0.5 in) sections using traditional methods
- If no fuel / clad bonding material is used, cladding is sent to storage
- Offgas released by chopping is collected and stored

Electrorefining of ATW Assembly Provides U, TRU, and Fission Product Separation



One ATW fuel assembly ^ Includes Zr ATW waste

Electrowinning TRU from Process Salt Allows for Salt Recycle



Molten Salt from 10 electrorefining systems

Fuel Fabrication for ATW system



Four Batches required to prepare fuel alloy * Includes U /TRU / RE from electrowinning process #9

Reductive Extraction Process Removes Rare Earths from Process Salt



Treatment of ATW Waste Requires 360 kg / day / Plant*

Process Plant Parameters

System	Batch (kg HM)	Process Time (hr)	Number of Systems	Capacity per day (kg^)	Operating Parameters
Chopping	11.7	8	3	405	
Electrorefining	11.7	6.7	4	360	500 A (500 W)
Electrowinning	9.2	6.1	1	9.2	500 A (500 W)
Reductive Extraction	0.5	8	1	0.5	
Fuel Fabrication	4	6	5	100	
Offgas System	Xe and Kr cryodistillation and collection		1		

*Two chemical plants are required to treat LWR and ATW waste, 175 day/yr - 24hr/day.

^ Includes Zr in ATW waste

Projected TRU Waste Output to Repository

- Treatment of LWR spent fuel
 - simulations indicate 12 ppm TRU waste output with anode polishing
- Treatment of ATW spent fuel
 - simulations indicate 10 ppm TRU waste output
- Higher process loss may be present in engineering scale systems
- Total TRU sent to repository, 60 yrs operation: < 300 kg in approximately 900 t of NM

ATW Provides Segregated Waste Streams for Disposal

- Uranium low level waste, radioactivity of natural U
- Metals repository
 - spent fuel clad and fuel assembly hardware
- Transition Metal Oxides repository
- Lanthanide Oxides repository
- Active Metals
 - engineered storage containers, repository

Treatment of ATW Waste Offers Advantages over ALMR Fuel Reprocessing

- Less material to process, U is not present at significant levels
 - smaller batch size, 45 kg instead of 400 kg
 - moderate operating current, 500 A instead of 8000 A
 possible reduced cost

• No deleterious effect of fission product carry-over into fresh fuel

Process Chemistry Issues Addressed in 5 Year Program

- Develop process scalability information by designing, fabricating, and testing various size electrochemical cells, reduction systems, and reductive extraction systems
- Develop more detailed material balance for process chemistry
- Verification of separation efficiency for electrorefining and electrowinning processes at 10 kg scale
- Establish complete thermodynamic database, e.g., Cm and Np
- Verification of proposed ATW fuel preparation process at kg scale, provide fuel for irradiation experiments
- Verification of multicomponent reductive extraction process at 10 kg scale

Technetium and ATW

- ATW Project-

Presented by: Ann R. Schake Nuclear Materials Technology Division

In a Repository ...

...the high mobility in ground water and longlived radiotoxicity makes accountability of technetium extremely important.

Yucca Mountain Geology



- ~2 km thick tuff (volcanic ash) layers
- highly fractured
- water table ~500 m
 below surface
- proposed repository in water-unsaturated zone

Yucca Mountain Chemistry



- Mainly silicic rocks
- Neutral to Basic pH water (6.8 8.3)
- Water-unsaturated conditions (fairly arid)
- Oxidizing environment

Tc Mobility is a Concern for a Repository

- Tc and TcO₂ are easily oxidized to Tc₂O₇ and subsequently TcO₄⁻
 TcO₄⁻ is extremely mobile in the environment
- Reducing conditions are needed to maintain TcO₂
 - Expect oxidizing conditions at Yucca Mountain
 - In POLLUX containers a large excess of Fe is needed to retain reducing conditions (Fe:Tc = 14700:1)
 - Even under reducing conditions radiolysis forms Tc^{VII}
- Tc release rate is 1-2 orders of magnitude greater than that for Cs and Sr
- ⁹⁹Tc half-life is more than four orders of magnitude greater than ¹³⁷Cs and ⁹⁰Sr

ATW Provides a Solution to the Concerns Posed by Tc

- Tc exists as an alloyed metal in spent fuel.
 - Tc metal can be recovered and made into targets
 - 70000t inventory contains about 5.3 t of Tc
- Tc is transmuted to Ru by neutron capture in ATW
- Tc is transmuted in either fast or thermal spectrum ATW system

Technetium is Isolated in the Anode of the Electrorefining Process

Tc Recovery

- Oxidation of Tc, and anode metals, forming Tc₂O₇
 - Other metals will also be oxidized, e.g. Zr, Mo, Ru

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- Distill and condense Tc₂O₇
 - Tc forms the most volatile metal oxide of those present
- Direct oxide reduction to recover Tc
- Alloy Tc with Mo
 - Mo is a good host for Tc, ~10 atom percent solubility of Tc in Mo

Thermodynamic Properties of Tc and Related compounds

Species	Melting Point (K)	Reiling Point (K)	Vapor Pressure (torr)	
		Donnig I onit (IX)	300 K	500 K
Тс	2445	4850		
Мо	2890	4885		
Ru	2583	4173		
TcO ₂ *	(dec. 1173K)	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	3.45e-11	7.26e-5
Tc ₂ O ₇	392	583	1.26e-3	72.4
MoO ₃	1073	1428	2.72e-34	7.08e-15
Re ₂ O ₇	570	635	6.02e-10	2.54

*Thermodynamic data shown is for ReO₂.

Experiments Planned for the Five Year Program

- Separation experiments on Re / Mo and Mo / Tc oxides and alloys
 - bench-scale tests using Re as a Tc surrogate
 - distillation experiments
 - DOR experiments
 - alloy studies
- Non-aqueous oxidation potential for Mo, Re, and Tc
 - important for ER process

Summary

Recovery and transmutation of Tc in ATW reduces the uncertainty associated with Tc management.

• Chemistry of Tc in a repository environment is established

- Process chemistry is established for the recovery of Tc
 Oxidation, distillation/condensation, oxide reduction
- Tc is transmuted in either fast or thermal spectrum in ATW system

Conclusions

Pyrochemical processing provides the link between spent fuel clean-up and the TRU burning in the ATW system.

- Process chemistry builds on existing experience
- Spent fuel treatment flow chart established
- Simulations indicate processes are viable and produce minimal TRU discharge
- Minimal secondary waste produced
- Main process chemistry issues identified

George Lawrence Project Leader for APT Accelerator Design Los Alamos National Laboratory

Presentation at ATW Technical Review

Massachusetts Institute of Technology January 15-16, 1998

Outline

• Accelerator design basis

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- Proposed ATW linac design
- Relation to APT

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• Technical issues and ED&D program

Design Objectives for ATW Accelerator

A linac for an ATW facility should be designed:

- To have very high electrical efficiency (ac to beam power)
- For minimum capital and operating costs
- For minimum spatial footprint (short length)
- To vary the power on target over a wide range
- To have high availability and operational flexibility
- To use the best mix of established technology and anticipated technology advances.

Nominal beam parameters

- 40 MW proton beam
- 1000 MeV, 40 mA, 100% duty

Neutron Production on Spallation Targets as a Function of Proton Energy





High- β Superconducting Linac Architecture ($\beta = 0.71$) Singlet Lattice, 4 Cavities per Cryomodule



Energy span	240 - 1000 MeV	Section length	199.5 m
Eaccelerating	15.2 MV/m	Klystrons	38
Eaverage	4.41 MV/m	Power/coupler	200 kW
5-cell cavities	76	Power/klystron	800 kW
Cryomodules	19	SC quadrupoles	95

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Global Accelerator Parameters

Parameter	Value
Output proton current	40 mA
Duty factor	100%
Final energy of NC linac (MeV)	21.2
Final energy of SC linac (MeV)	1000
Length (m)	345
RF power (MW)	44.6
Number of klystrons	3 at 350 MHz; 53 at 700 MHz
Number of cryomodules	31
Number of NC accelerating cavities	106 x 2
Number of SC accelerating cavities	134
Number of NC quadrupoles	105 x 2
Number of SC quadrupoles	165

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NC Linac Parameters

Parameter	RFQ	CCDTL
Structure gradient (MV/m)	1.38	2.04 - 1.86
Average gradient (MV/m)	1.38	0.77 - 1.03
Length (m)	8.0	29.6
Synchronous phase (deg)	-30	-30
Quadrupole lattice period	βλ	8 - 9 βλ
Number of guadrupoles	-	105 x 2
Focusing lattice	-	singlet FODO
Aperture radius (cm)	0.23 - 0.34	1.0
Copper RF power loss (MW)	1.26 x 2	1.02 x 2
Beam power (MW)	0.26	0.58
Total RF power (MW)	2.8	2.6
Number of klystrons	3 x 2	3 x 2
Energy range (MeV)	0.075 - 6.7	6.7 · 21.2
Number of accelerating cavities	4 x 2	105 x 2
Frequency (MHz)	350	700
Superconducting Linac Parameters

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Parameter	β = 0.25	$\beta = 0.35$	β = 0.48	β = 0.71
Initial energy (MeV)	21.2	46	100	240
Final energy (MeV)	46	001	240	1000
Structure gradient (MV/m)	8.6	9.3	11.2	15.2
Active accelerating fraction	0.166	0.215	0.252	0.290
Average gradient (MV/m)	1.43	2.00	2.82	4.41
Energy gain per cavity (MeV)	2.0	3.0	5.0	10.0
Synchronous phase (deg)	-30	-30	-30	-30
RF power per cavity (kW)	80	120	200	400
RF power per section (MW)	1:0	2.2	5.6	30.4
RF coupler power (kW)	80	120	200	200
Cavities per cryomodule	6	6	4	4
Klystrons per cryomodule	1	1	1	2
Number of cavities	12	18	28	76
Number of cryomodules	2	3	7	19
Number of klystrons	2	3	7	38
Number of SC quadrupoles	14	21	35	95
Quadrupole length (cm)	28	28	28	40
Number of couplers per cavity	1	1	1	2
Focusing lattice	FODO	FODO	FODO	FODO
Cryomodule period (m)	9.7	10.4	8.2	10.5
Section length (m)	19.4	31.2	57,4	199.5
Aperture radius (cm)	3	4	5	7.5
Frequency (MHz)	700	700	700	700

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Integrated SC/NC Linac Provides Optimum Performance by Exploiting Advantages of Each Technology

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NC linac for low energies

- high-density magnetic focusing lattice
- smoothly varying accelerating and focusing parameters
- excellent emittance control of high-current beam
- minimal generation of beam halo

SC linac for high energies

- eliminating cavity RF losses provides very high power efficiency
- very large aperture dramatically reduces beam loss threat
- short cavities provide wide velocity acceptance bandwidth
- gradient adjustability and independent RF phasing allow flexibility to meet beam-power needs by energy/current tradeoff
- large apertures plus retunability provide increased tolerance to a wide range of error and fault conditions

ATW Linac Design is Based on APT Design and Technology

- Developed integrated high-power 1700-MeV, 100-mA linac design.
- Issued Conceptual Design Report and Cost Estimate (confirmed by ICE).
- Linac design and ED&D program reviewed by high-level technical panels.
- Highlights
 - Linac architecture emphasizes large apertures and smooth variation of parameters for very low beam loss.
 - High-current, high-reliability proton injector in operation.
 - High-energy 100-mA 350-MHz RFQ designed and being fabricated.
 - Designed and modeled CCDTL as solution for low-energy region.
 - Designed multicell elliptical cavities and cryomodules for SC linac.
 - Tested single-cell niobium SC cavities for medium-beta (0.48, 0.64).
 - Developed 700-MHz 1-MW CW high-efficiency klystrons.
 - Worked out control approach for coupled beam/cavity/RF-drive.
 - Designed BOP systems for support of high-power linac.

Integrated SC/NC Linac Design for APT Plant

(New Baseline, 10/97)

3-kg/yr Production Rate



High-efficiency operation over wide range of beam power and tritium production requirements.

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Diston energy	1700 14	
I TOTOL DIELAY		24
CW beam current	100 mA	
	1	
	170 MW	
Plant AC nower	470 MW	6
- Anticico ponor		
Peak coupler pow	er 210 kW	
Commodulan	E+ /777	
a yomoones	ə11.1	
Kivstrons (1-MW)	239	
	1.00	
L'ARISC IGNOIN	112U M	
Cryonlant load (2)	0 11 2 KW	
Dijopiani iodo (ci	·/ · · · · · · · · · · · · · · · · · ·	

ΔΡΊ

10/18/97

High-Level Technical Reviews Have Confirmed APT Linac Design

- JASONs have reviewed APT linac design several times as it has evolved (1988, 1992, 1995, 1997).
- 1997 JASON Review endorsed SC high-energy linac design proposed as replacement for high-energy NC linac.
- Comprehensive review of APT system design undertaken by LLNL (11/94); stimulated study of SC high-energy linac option.
- MIT Review examined APT linac design when NC high-energy linac was the baseline (10/94).
- APT External Review Committee has provided overall technical critique and guidance for design evolution.





ATW Linac Design is More Advanced than APT; Provides Efficiency and Cost Advantages

Higher accelerating gradient in SC linac

- much shorter linac; reduces length and costs
- 15 MV/m for ATW vs 5 MV/m for APT
- gradients achieved in TESLA cavity tests

• Cryomodules contain SC quadrupoles in FODO lattice

- stronger focusing; larger aperture/beam-size ratio
- higher electrical efficiency; minimizes quadrupole power

SC linac starts at lower energy

- 20 MeV instead of 210 MeV
- reduces RF losses in low-energy linac; increases efficiency
- increases aperture size at low energies
- makes use of 1/2-wave (spoke-type) SC resonators

The LEDA Injector has Demonstrated APT Beam Performance Requirements and Reliability



- H⁺ current: 110 mA
- Energy: 75 keV
- H+ fraction: 90%
- Arc power: 600 800W
- H₂ gas flow: 2 5 sccm
- Availability: 96 98%
- Emittance: 0.20 π mm-mrad (rms normalized)

6.7-MeV, 8-m-Long RFQ is Made up of Four Resonantly-Coupled Segments



Two LEDA RFQ segments are coupled for measurement of RF power distribution, confirming details of fabrication.

> One of 8 brazed RFQ sections, showing vanes, vacuum ports, & cooling lines.



As in APT Design, CCDTL Accelerating Structures will be Used in the Energy Range from 7 MeV to 21 MeV.





 $\frac{1}{2} \sum_{i=1}^{n} \frac{1}{2} \sum_{i=1}^{n} \frac{1}$

- Short DTLs, resonantly coupled.
- Combines best features of DTL and SCL.
- Strong RF coupling; field stability.
- Separation of acceleration and focusing.
- Ease of fabrication and assembly.
- Quadrupoles external to RF structure.

Spoke-Type SC Resonators Have Been Developed for Acceleration in the Low Beta Region



2-gap 1/4-wave resonator (400 MHz, $\beta = 0.15$)



BETA = 0.64 5-Cell Niobium Test Cavity



RF Power System is Major Design Driver in Terms of Costs, Operability, and Availability



APT Linac Engineering Development and Demonstration

- Low-Energy Demonstration Accelerator (LEDA) will address beam performance and integration/operation/availability issues for linac front end.
- Power test of 100-MeV CCDTL/CCL segments will confirm CW cooling and operation of NC linac sections with highest heat deposition.
- SC cavity and cryomodule prototyping. Three-stage program will confirm SC cavity and cryomodule designs and operability.
- Advanced RF tube development. Prototypes of advanced RF generators (klystrons, HOM IOTs) will enable higher electrical efficiency for plant.
- High power RF window development. Objective is to develop reliable 1-MW CW windows that have "infinite" life at 210 kW (nominal operation).
- Advanced HVDC power supply development has potential for significant cost savings.

Purpose and Functions of LEDA (Low Energy Demonstration Accelerator)



Demonstrate integrated full-power operation of APT low-energy linac. Refine equipment designs for plant. Qualify vendors Improve cost estimates

Single-Cell β = 0.48 Cavity Measurements Indicate that ATW Design Performance is Attainable



SCRF ED&D Program Will Build Prototype Cryomodules for APT Linac



 $\beta = 0.82$

 $\beta = 0.64$

ATW Linac ED&D Program Will Focus on Accelerator Design and Performance Requirements that are Different from APT

- Development of high-gradient single-cell and multicell SC elliptical cavities for medium-beta (β = 0.48, 0.71).
- Development of high-gradient single-cell and multicell SC spoke cavities for low-beta (β = 0.25, 0.35) region.
- Development of short SC quadrupoles for SC linac cryomodules.
- Development of adjustable RF power couplers to provide efficient operation over full range of beam current.
- Prototyping of complete elliptical-cavity medium-beta cryomodules (including SC quadrupoles).
- Prototyping of complete spoke-cavity low-beta cryomodules (including SC quadrupoles).

Preliminary Cost Estimate for 1000 MeV 40 mA ATW Linac (12/97)

	Labor Hours					Thousand Dollars (1997)			
	Prelim	inary &	Component Procurement,		Preliminary &	Component			
	Final (Design	Fabrication & Installation		Final Design	Procure, Fab.,	Purchased	Component	
	(P8	FD)	(PF&I)			& Install	Material	Total	
Component	Prof	Tech	Prol	Tech	Craft	(P&FD)	(PF&I)		
Injector	7,992	4,440	4,808	9,515	3,835	\$1,634	\$1,684	\$326	\$3,644
ALC OLD	7,104	2,664	28,892	65,528	5,750	\$1,350	\$10,095	\$790	\$12,235
CCDTL	8,880	5,328	11,196	38,329	3,900	\$1,847	\$5,014	\$2,542	\$9,403
Spoke Resonator Cavities	21,312	28,416	14,208	35,520	8,880	\$5,683	\$5,470	\$21,000	\$32,153
Elliptical Cavities	31,968	53,280	15,431	41,873	10,327	\$9,377	\$6,232	\$44,252	\$59,861
RF Power	15,984	22,644	31,007	153,487	120,572	\$4,369	\$22,063	\$75,576	\$102,008
Cryoplant	17,760	35,520	7,000	10,000	0	\$5,683	\$1,920	\$37,000	\$44,603
Linac Controls	26,640	46,176	62,160	79,920	26,640	\$7,956	\$17,405	\$11,873	\$37,234

		-	
Groun	drules	8	Assumptions

 Hourty Labor Rales (\$/ 	hr)		
Professional	\$160	30% Contingency ==>	\$90,342
Technician	\$80		
Craft	\$40	TOTAL LINAC SYSTEM COST ==>	\$391,483
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All estimates assume a functioning APT finac (in place talent pool)

All linac (except Spoke Resonator) scaled from APT

Spoke resonator estimates scaled from "grass roots" work done on IEMIE project

Linac controls include all components from linac through cable runs and controlling computers

· Estimates above assume successful completion of a separate technology development program

cavity development: spoke resonator & elliptical

high SC fields.

30% contingency scaled from APT/IFMIE values considering increased risk of the ATW technical requirements

Nole:

The Total ATW Linac System Cost Esilmate of \$391M is highly preliminary;

The comparable Cost Estimate for the APT (3 kg) Linac is \$980M

\$391.5M

Linac System Subtotal (\$k) ==> \$301,141

Summary

- APT accelerator design and ED&D program is firmly established, has been intensively reviewed, and has high credibility. An integrated NC/SC linac is the Project baseline.
- ATW linac design rests on APT accelerator conceptual design, and ED&D program advances.
- ATW-specific linac design requirements emphasize higher electrical efficiency and shorter length compared with APT.
- ATW design solution pushes to higher accelerating gradients, and lower starting energy for SC linac.
- A modest extension of the APT program will have high payoff for support of an ATW-specific advanced linac design.



Stages of development of Lead-Bismuth as a coolant for nuclear reactors in Russia

Prof. Yu. I. Orlov

State Scientific Center of Russian Federation Institute of Physics and Power Engineering 249020 Obninsk. Russia

IPPE. Obninsk, Russia

Alexander Ilyich Leipunskii (1903-1972)

One of the founders of the IPPE. A member of Ukraine Academy of Sciences, he worked in the IPPE in 1949-1972, was a scientific supervisor of the institute and of all the trends of liquid metal reactor design.

In the early 1950s academician Leipunskii suggested using lead-bismuth eutectic as a coolant in nuclear reactors of transport systems.



Lead-Bismuth eutectic was chosen for its unique qualities.



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The favorable properties of Pb-Bi eutectic

- The impossibility of explosion and fire at the alloy interaction with air, water and vapor because of **the low chemical lead and bismuth activity**
- The impossibility of the coolant boiling out with high energy deposition due to the high boiling point of Pb-Bi (1670° C).
- The low working pressure of the coolant in the circuit increases the facility safety, simplifies the equipment design and construction, makes significantly simpler working conditions for fuel elements

Lead-Bismuth cooled reactors

- Since 1952 the IPPE has been engaged in designing nuclear power reactors cooled by the lead-bismuth alloy. Units like these are used in high speed, highly maneuverable, nuclear submarines.
- In the course of this work a team of specialists has been formed, an experimental basis has been created, and experience in investigation, design, construction and operation of this type of installation has been gained.
- Specific features of the reactors being developed required a complex of investigations to be done on reactor physics, heat transfer, hydrodynamics, coolant technology, material corrosion, safety, control and monitoring systems, development of fuel elements and equipment capable to function in the lead-bismuth alloy medium.
- According to some publications intensive research on lead-bismuth was conducted at the same time in United States and Canada.

Lead-Bismuth cooled reactors

and the second second

- In early 1960s the first nuclear submarine, in which lead-bismuth alloy was used as a coolant, was commissioned.
- However, in the second half of 1968 there was an accident with a nuclear reactor.
- The analysis that followed showed that the cause of the accident was lack of full knowledge about the coolant.

Pb-Bi alloy properties that can negatively affect reactor performance.

- The Pb-Bi coolant is **comparatively corrosive** to the construction materials.
- Pb-Bi coolant can be contaminated by solid admixtures during operation due to interaction with construction materials and oxygen.
- At the first stage of work with Pb-Bi coolant these factors caused the following
 - closing of Pb-Bi programs in USA
 - decrease of operating life of the first Nuclear Power Plant.

IPPE, Obninsk. Russia



Pipe condition before lead-bismuth technology improvement program

Principle tasks of Pb-Bi coolant technology

- For the long and safe operation of Pb-Bi coolant facilities it was necessary to do the following.
 - Ensure the corrosion-erosion resistance of the construction materials used.
 - Ensure the cleanliness of coolant and inside surface of the equipment where the coolant circulated.
- In order to perform these tasks an extensive program of research was carried out.
 - Pb-Bi alloy properties, physical and chemical processes that take place in anisothermal circuits, corrosion resistance of construction materials, admixture sources and influence of all these factors on efficiency of the facility were studied.
 - Large experimental base, including dozens of circulation facilities, was created in Russia and is still operating.
 - Hundreds of experts were involved in solving these problems.

IPPE, Obninsk, Rissia

Corrosion Resistance of Materials

- Corrosion resistance of materials can be improved by
 - selecting suitable steel or creating a special one
 - creating protective film on the steel
 - correcting the coolant quality (O₂, particulates)
- Equipment in Pb-Bi facilities, that has to work at temperatures up to 450C, requires ordinary stainless steel of kinds ЭЯ-1T (1X18H10T), ЭП-302 (1X15H9C3E1) of the austenite class.
- Equipment that has to work at temperatures above 450C is produced from the special steel of ferritic-martensitic class.
- The protective film consists of oxide compounds of steel components. The basis of the film is Fe₃O₄.

Reaction of formation and dissociation of oxide Fe_3O_4 is described be the following formula.

$$2O_2 + 3Fe \Leftrightarrow Fe_3O_4$$

with equilibrium constant

$$K = \frac{a_{[F_{e_1}, o_{e_1}]}}{a_{[o_{e_1}]}^2 a_{F_{e_1}}^3}$$

IPPE, Obninsk, Russia



Standard free energy change for the oxide formation Fig. reactions.

g,

Corrosion Resistance of Materials

- If there are conditions for existence of phase Fe₃O₄, then $a_{[Fe_3O_4]} = 1$, and connection between $a_{[Fe]}$ and $a_{[O]}$ has fixed properties.
- Decrease in concentration of oxygen dissolved in alloy is possible up to the values that are equal or less than equilibrium with Fe3O4.
- On the other hand, existence of a considerable amount of oxygen in a circuit is undesirable as this can cause accumulation excess amount of oxide admixtures, primarily, coolant oxide.
- It is necessary to maintain amount dissolved oxygen at a fixed level.
- * Testing for dissolved oxygen concentration in alloy was done by a controller of oxygen thermodynamic activity. Controller, designed and made in IPPE, uses principle of galvanic concentration register with solid electrolyte. Methods and equipment for oxygen activity correction were developed also.
- * These device and procedures provided corrosion resistance for structural materials at all stages of Pb-Bi facility operation.

Temperature dependence of thermodynamic oxygen activity and concentration of oxygen and iron for lead-bismuth



Measuring of thermodynamic activity of oxygen.



Measuring scheme of galvanic concentration cell.





$$E^{0} = \frac{\Delta G^{0} B i_{2} O_{3} - \Delta G^{0} P b O - RT \ln a}{2F}$$

$$\lg a = -\frac{10094}{T} \left(E^{0} - 0.088 \right) - 0.18$$

$$t = \frac{E_{M}}{E^{0} M a x}$$

IPPE. Obninsk, Russia

Cross sections of steels exposed in forced-circulation lead loop for 3000 h at 550 °C

Austenitic stainless steels



Corrosion in structural materials CRISM "Prometey"





Low alloy steels



Tranzition zone

Oxide film on structural materials
Ensuring of cleanliness of circulation circuit and coolant

- During the long facility operation accumulation of coolant admixtures. which can cause some negative effects, inevitably takes place. The composition of depositions formed in the circuit depend on the construction materials, regime of operation and can vary greatly.
- The methods and devices for removal of lead oxide (the basic admixture) by reducing it to lead directly in the circuit were developed in the IPPE.
 Chemical reaction of oxides with hydrogen was applied

 $PbO + H_2 = Pb + H_2O$

- The chosen conditions for this reaction do not allow reduction of the oxides that constitute the base of the protective films on the construction materials.
- For the removal of admixtures that cannot be reduced chemically **special filters** were created.



Before cleaning



After cleaning

Conclusion

As a result of the work done on lead-bismuth cooled reactors the following problems were solved

- 1 Scientific base for Pb-Bi coolant treatment technology was determined.
- 2 New construction materials for use in contact with Pb-Bi alloy at temperatures up to 600C were chosen or created
- 3 Methods and devices for control of Pb-Bi alloy quality were created.
- 4 Methods and devices to maintain Pb-Bi alloy quality were determined.
- 5 Methods and devices for removal of admixtures from the coolant and circuit surfaces were created.

- Complex of methods and means, which are named "Heavy Coolant Technology" (HCT) was carefully tested at many experimental facilities, at a prototype of a nuclear power plant and was applied to industrial facilities.
- There were no facilities' breakdowns caused by the coolant during operations due to use of this technology.
- At present IPPE is the world leader in leadbismuth coolant technology.

IPPE, Obninsk, Russia

Influence of Spallation Products on Mass Transfer Process

- New elements generated by proton beam in spallation reactions can take part in different chemical reactions disturbing chemical equilibrium in the circuit. The most important among these reactions are oxidizing reactions with formation of nonsoluble oxides. Reactions with reduction (i.e. with taking oxygen from oxide) are especially important since they destroy protecting films on structural materials.
- New elements formed in spallation reactions may be divided into three groups
 - i elements with oxygen affinity less than lead will be dissolved in lead-bismuth without oxide formation (Au. Ag. Pt. Hg. Os. Cu. Tl. Pb. Bi) - $\Delta G \ge -160 kJ / g - atom$ at $300^{\circ} C$
 - 2. elements with oxygen affinity between the values for lead and iron will be dissolved or oxidized depending on thermodynamic activity of oxygen. (Re. As. Te, Sb. Co. Ni. Mo, Sn. Fe) $-160kJ/g - atom > \Delta G > -230kJ/g - atom$ at $300^{\circ}C$
 - elements with oxygen affinity higher than iron will be able to cause reduction in FeiO4 and destroy protective film in some conditions (lantanides, galogens, alkaline elements, Be, Y, Sc, Al, Ge, Ti, Hf)

 $\Delta G < -230 kJ fg - atom at 300^{\circ} C$

 In the whole, analysis demonstrated that for 10-20 MW circuits influence of admixtures generated by proton beam on mass transfer process can be noticeable and requires special attention

IPPE, Obninsk, Russia

Pb-Bi and Pb coolant Experimental Base in Russia

	Facilities	Investigation's field	Coolant	Temperature ⁰ C	Coolant flow rate m^3 / hr	Coolant Volume m^3
1	CBT-3M	Thermohydr aulics Technology	Pb-Bi	160-450	20	0.1
2	ИСР	Technology Corrosion	Pb, Pb-Bi	160-650	6	0.1
3	TT-1M	Technology Corrosion	Pb-Bi	240-600	6.5	0.18
4	TT-2M	Technology Mass transfer	Pb-Bi	250-550	5	0.2
5	CM-1	Corrosion Mass transfer	Pb-Bi	270-650	5	0.1
6	СМ-2	Corrosion Mass transfer	Pb	350-650	2.5	0,07
7	ЦУ-1	Corrosion Mass transfer	Pb	350-620	5	0.1
8	ЦУ-2М	Corrosion Mass transfer	Pb·Bi -	270.650	2	0.1

Pb-Bi and Pb coolant Experimental Base in Russia

-	Facilities	Investigation's field	Coolant	Temperature ⁽¹ C	Coolam flow rate $m^{\frac{3}{4}}/hr$	Coolam volume m ³
9	СПРУТ	Thermohydraulies Technology	Pb-Bi	150-500	5.0	0.12
10	H-8000	Investigation of large scale equipment performance	Pb-Bi	150-520	120.0	1.2
-	ПГ	Investigation of small and medium scale equipment performance	Pb-Bi	150-520	2.0	0.4
12	BTP	Investigation of small and medium scale equipment performance	Pb Bi	150-520	5.0	0.08
13	"Поток -1"	Corrosion	Pb·Bi	150-650	5.0	0.1
14	"Поток -2"	Corrosion	Pb	340-650	5.0	0.1
15	ΦΧΤ	Thermohydraulics Technology	Pb Bi	150-520	12.0	0.5

IPPE, Obninsk, Russia







EXPERIENCE OF THE LEAD-BISMUTH COOLANT USE.

A.Dedoul, IPPE, Obninsk, Russia V.Kutanov, EDO "Gidropress", Podolsk, Russia

Lead-bismuth coolant use issues:

• Lead-bismuth coolant technology and quality control.

- Corrosion, erosion and mass transfer.
- Radioactive safety problem including Po.
- Freezing/defreezing problem.
- Thermohydrodinamic problems.

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Issues resolved by experience of lead-bismuth coolant use in Nuclear facilities development, construction and operation:

- Two ground reactor facilities.
- Submarines with lead-bismuth cooled reactors.
- Industrial applications.
- Two test rigs for fuel rod reliability testing at reactors MIR and MR.

IPPE. EDO "Gidropress". Russia

Nuclear steam supply system module



1-st generation design

2-nd generation design



BRUS-150 Reactor plant of enhanced safety with liquid-metal coolant



Improved generation design

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LANL/IPPE/Gidropress spallation target

The target system with horizontal injection beam for LANL



Applicability of available experience to ATW development:

• Available experience is fully applicable.

• The volume of theoretical, experimental and design knowledge is enough for preliminary evaluation of ATW designs.

IPPE, EDO "Gidropress", Russia

Engineering, technological and safety problems of ATW facility design.

Engineering problem	Variants		
Coolant type	Pb-Bi	Pb	
Target design	Windowed	Windowless	
Target cooling system	Separate circuit	Joint with blanket cooling system	
Number of circuits	2	3	
Coolant circulation	Forced circulation	Natural convection	
Fuel type	Depends of fuel cycle and goals of ADS introduction into nuclear industry		
Fuel rod type & fuel assembly design	Spacing the fuel elements by the rib faces	Spacer grid use	



Complex coolant technology issues			Safety issues		
Freezind -	Coolant	Po problem	Loss of heat	Steam	Other safety
defreezing	technology		sink	generator	assurance
			accident	tube rupture	problems

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LANL/IPPE/EDO-GP COLLABORATION PROGRAMS ON LIQUID LEAD-BISMUTH TECHNOLOGY

Los Alamos National Laboratory - ATW Project -

Presented by: Ning Li

Contributions from: Y. Orlov, E. Yefimov, IPPE; EDO/GP J. Stubbins, Univ. of Illinois M. Bjornberg, X. He, W. Gregory, LANL

Outline

Part I. LANL/IPPE/EDO-GP 1-MW LBE test target IPPE: Institute of Physics and Power Engineering, Obninsk EDO-GP: Experimental Design Organization-Gidropress, Podolsk

Part II. LANL/IPPE polonium removal technology

Part I.

LANL/IPPE /EDO-GP 1-MW LBE Test Target

<u>ISTC #559</u>: Funding for IPPE/EDO-GP to design and fabricate a 1-MW liquid LBE spallation neutron target to be tested at Los Alamos by 2000

<u>Objective</u>: Perform thermal hydraulic and in-beam radiation testing

Task Plan(5-year):

- LANL assists IPPE in design and verification: target specifications. neutronic, thermal hydraulic simulations (97-98)
- IPPE/EDO-GP will fabricate the target hardware (98-99)
- LANL will prepare the testing facility and support systems (98-00)
- LANL/IPPE/EDO-GP will operate and test the target (2000-2001)
- Post-test examination and component performance assessment

ADTT Simulation Work for Target Design

- Neutronic calculation to find neutron production and heat deposition
- Thermal hydraulic simulations to ensure operation within material limits

Test Target Configuration (Sideview)





through two 1.5 mm thick Inconel windows. The 1.25 mA, 800 MeV proton beam hits the lead-bismuth target after passing





Flow Path in Target Design



Simulations Show Window Cooling is Sufficient



Acceptable Temperature Limit (600°C) Can be Maintained in Target Design





Maintain Proper Oxygen Level for "Self-healing" Protective Film

Fig. Standard free energy change for the oxide formation reactions.

Effects of Spallation Products on Oxygen Level Control Are Minor

Spallation products mass in LBE target in LANSCE beam after 1 year

Group	Elements	Mass of	Mass of
46		Elements (g)	Oxide (g)
> Ph	Au, Ag, Pt, Os, Cu, Tl, Pb, Bi	110	(none)
Pb>->Fe	Re, As, To, Sb, Co, Ni, Mo, Sn, Fe	1.	1.3
< Fe	Lanthanides, Halogens,	11	14
	Alkaline Elements, Be, Y, Sc,		
	Al, Ge, Ti, Hf		

Results from Gromov, Orlov, and Gylenvsky, IPPE, Obninsk

Only a small mass of elements which oxidize preferentially to Fe are formed Oxygen level needs to be adjusted only if it falls below permissible range

Summary of Part I

(1) We have established framework for complete neutronic, thermal hydraulic and structural analysis

(2) We are in close interaction with IPPE target design and construction team

(3) Analysis shows preliminary design very promising

(4) Final target design is expected in October, 1998

Part II. Polonium Removal Technology

LANL/IPPE Contract: Develop a polonium removal device in LBE coolant systems for ATW

<u>Objective</u>: Mitigate a radiological safety concern of polonium release hazard

Task Plan(5-year):

- select most feasible technology for polonium removal (96-98)

restore coolant quality after removal(97-99)
removal system design and safety analysis (2000-2003)

Origin of Polonium in Lead-Bismuth Target

A) Bi^{209} (n, γ) $Bi^{210} \rightarrow Po^{210}$ (main mechanism)B) Bi^{209} (p, xn) Po^{210-x} , x = 1-12(not important)C) Pb^{208} (α, xn) $Po^{210-x+2}$, x = 2-14(not important)

Activity of Po Nuclides After 1-yr. Operation(LBE*: 15MW,1 GeV) at Shutdown

Nuclide	Half-life, days	α - particle yield per disintegration, %	Activity, Ci
Po-210	1.384 E+02	100	2.310 E+04
Po-209	3.726 E+04	99.74	3.610 E+01
Po-208	1.058 E+03	100	2.570 E+03
Po-207	2.429 E-01	0.02	1.724 E+04
10-206	8.800 8400	and the second se	1.902 E +#4
Po-205	7.500 E-02	0.04	1.420 E+04
Po-204	1.471 E-01	0.66	1.376 E+04
Po-203	2.549 E-02	0.11	7.700 E+03
Po-202	3.104 E-02	2	5.465 E+03
Po-201	1.062 E-02	1.6	2.853 E+03
Po-200	7.986 E-03	15	8.510 E+02

Polonium Hazard and Regulations

• Hazard: Release from active coolant in the events of circuit breakdown (e.g. repair, accidents) and consequent gas-aerosol and surface activity formation

• Regulations: Limit permissible concentration (LPC) of Po-210 in air for professionals*

- Russian (NRB-76/87):

0.93x10⁻¹³ Ci/l

- US (Federal Register of 5/21/91 Vol 56 #98)

(*: LPC for Po-209 and -208 were not established but should be about the same)

• Russian experience: proper radiological safety measures were deployed and no noticeable personnel exposure to Po in over 70 reactor-year NPP experience even without Po extraction

Polonium Release Rates from Active Coolant



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Polonium Removal Methods Based on Russian Experience

- Vacuum sublimation
 - Po removal by evaporation when coolant is heated up to 700°C
 - Used for military applications, but not well suited for ATW
- Alkaline extraction
 - Contact coolant with alkaline melt (reversible reaction)
 - $PbPo + 4NaOH <-> Na_2Po + Na_2PbO_2 + 2H_2O$
 - IPPE experiments
 - 1) 90% Po transferred in 2 minutes of contact
 - 2) oxidizer in system caused Po back-transfer*
 - Additional benefits of removing other spallation products
- Storage
 - · Back transfer to LBE or Pb from alkaline melt by adding oxidizer
 - 50,000 Ci in 100 l of solid LBE with natural convection cooling only



Removal degree: $K = A_0/A$

- 15MW LBE Target (1 yr.): Po specific activity $a_0 \sim 30$ Ci/l
- Based on consideration of some accidents: a ~ 0.03 Ci/l -> gas aerosol activity ~ 10^{-14} Ci/l (0.1 Russian LPC; 0.03 US LPC)

K = 1000

$$λ_r = (K-1) λ ~ K λ$$

For Po-210, K=1000, $a_0 = 1.6E-3$ Ci/s, $λ = 5.8E-8$ s⁻¹
 $λ_r = 5.8E-5$ s⁻¹

Efficiency of Po Removal Systems

Alkaline extraction: $\lambda_r = g \epsilon/G$

- g flow rate of the bypass coolant for extraction, kg/s
- ϵ fraction of Po transferred to alkaline, ~0.9 in 2 min.

G · total coolant mass

Case: 15 MW LBE target, $G = 10^4$ kg $g = \lambda_r G = 0.6$ kg/s In the target loop: $g_0 = 600$ kg/s, so $g = 0.1\% g_0$ $\epsilon = 0.9$: $\tau \sim 2$ min. for contact of coolant and alkaline $G_r = g\tau \sim 70$ kg



Summary of Part II

- α-active Po presents gas-aerosol release hazards
- Po exists as PbPo in LBE and evaporates much slower than Po
- Russian NPP experience shows no noticeable Po exposure in over 70 reactor-years and strongly indicates that Po hazards are controllable
- Alkaline extraction of Po is feasible and can also extract other spallation products
- Coolant restoration is needed after alkaline extraction
- In 5-yr plan: Po removal system design and safety analysis

Los Alamos Liquid Lead Bismuth Eutectic (LBE) Test Loop

Los Alamos National Laboratory - ATW Pròject -

Presented by: Keith Woloshun

LANL: N. Li, J. King, R. Hammer, V. Tcharnotskaia, T. Langston W. Teasdale, and S. Wender IPPE: Y. Orlov

Outline

- Test loop objectives
- Test loop description
- Safety measures
- Operating experience to date
- Test plans
- Summary

LBE Test Loop Objectives

- Collaborate with Russian experts for technology transfer to establish the knowledge and experience bases in U.S. for designing and operating LBE systems for ATW
- Conduct thermohydraulic and material verification experiments critical to ATW system designs

Los Alamos Liquid-LBE Loop for Thermohydraulic and Material Experiments







System Analysis and Safety Measures

- We systematically analyzed system safety and implemented mitigating measures:
- Lead hazard well below OSHA limit(50 μ g/m³ in air/8-hr); vented
- Pressurized vessel hazard within vessel design spec; relieve valves
- Electrical, mechanical, high temperature hazards access control
- Waste hazard standard Laboratory disposition procedures

The results are documented in

- "Molten Lead-Bismuth Flowing Test Loop Description. Operating Procedures, Safety Analysis"
- Special Work Permit (SWP) for initial startup operation

Loop Successfully Started and Operated

- Test loop operated on 2 occasions for ~ 4 hours at 33% to 100% pump power, nominally 67% at 250°C
- Estimated over 3.5 m/s LBE flow velocity in 2" ID loop
- Noise and vibration very low with adequate sump pressure
- No LBE leaks through valve or seals
- Level indicators worked

Existing LBE Test Loops in IPPE and LANL

IPPE Facility	Research field	Coolani	Temperature (°C)	Coolant flow rate (m ⁼ h)	Coelant volume (m ¹)
CBT-3M	Thermo- hydraulics	Pb-Bi	160-450	20	0.1
ICP	Corrosion	Pb Pb-Bı	160-650	6	0.1
TT-1M	Corresion	Ρb-Βι	240-600	6.5	Ŭ.18
TT-2M	Mass transfer	Pb-Bi	270-650	5	0.2
CM-1	Corrosion Mass transfer	РЬ-Ві	270-650	2.5	0.1
CU-2M	Corrosion Mass transfer	Pb-Bi	27(1-650	3 	Ū. 1
LANL Test Loop*	Thermo- hydraulics	Pb-Bi	150-450	<u>ה</u> בב	0.24

*: Only one in U.S.

LBE Test Plan (5-year)

- Expand instrumentation and controls
- Test thermohydraulics of critical components necessary for ATW designs: target window design, pumps, heat exchangers, fuel elements, etc., necessary for ATW designs
- Circulation hydraulics and natural convection experiments
- Material verification experiments

Summary

- Test loop has been started
- Safety issues have been recognized and adequately addressed
- Full instrumentation and control will be implemented in the next three months

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• Collaborations with Russian experts have contributed to our early successes

ATH Program Plan and Summary

Francesco Venneri ATW Project Leader

Los Alamos National Laboratory -- ATW Project --

ATW Development Path



ATW Development Road Map

where we are. 1998: Technology Assessment

 Accelerator technology status: APT-driven, ATW specific features could be developed from APT

- Liquid lead technology status: early stage in US. Russian technology fully developed
- Pvrochemistry

status: individual parts well developed, Argonne and LANL have expertise 5-year Development Plan Technology Verification and Small-scale Integration

- Nuclear design
- Integrated small scale waste treatment plant
- Fuel development
- Russian liquid lead technology transfer
- High power spallation targets
- Materials verification
- ATW-specific accelerator design

where we are going: Large scale Integration and Implementation





Decision based on completion of

2004

Materials selection, Component Performance verification, STF Preconceptual Design, APF Design, DEMO Point Design

1999

Five-year ATW development plan provides technology support for large-scale integration and deployment





"Reducing the Impact of Nuclear Waste"

Francesco Venneri ATW Project Leader

Los Alamos National Laboratory -- ATH Project --

Performance Drivers in the ATW concept design :

Fast Burn Rates, Low Inventories

Capability for rapid materials inventory reduction Minimized economic impact



Engineering design clarity, defense in depth. inherent safety

Proliferation Resistance, Low Environmental Impact

ATW Consists of Three Major Functional Blocks



ATW Technology

what does the accelerator buy?

Subcriticality

• Power control not linked to reactivity feedbacks, delayed neutrons or to control rods, but only to the accelerator drive

- Constant power during burnup achieved by variable beam power
- Pure transuranic cores, CR=0: ATW has no need for fertile materials
- Subcritical systems work independently of the fuel composition
- EOL inventory not limited by criticality. EOL burndown of inventory
- Neutronics and thermohydraulics are effectively decoupled

ATW Technology

what does liquid lead-bismuth buy?

Design simplicity: no separate target structure
Good spallationsource: possibly the best n/p outside actinides
Enhanced safety: large natural convection capability, no reaction with water or air
Good neutronics: very hard spectrum, very low neutron capture, negative temperature and void coefficient
Good thermal hydraulics: large ∆t, low operating pressure, very high boiling temperature

ATW Technology

what does pyrochemistry buy ?

- PYROCHEMISTRY ALLOWS FAST, BULK-TYPE SEPARATIONS: what is needed in ATW, no more
- High Level of Proliferation resistance
- IT IS NOT ADVERSELY AFFECTED BY HEAT OR RADIATION
- IT PRODUCES LOW AMOUNTS OF SECONDARY WASTE
- Could be less expensive that traditional aqueous chemistry

Minimized-RISK APPROACH for ATW DEVELOPMENT

	Beam Power	Fission Power	Power Density	Fuel Type	Neutron Flux	Chemistry
LANL-IPPE Target (1999 - 2001) no fissionable material. Competency development. High radiation environ. Window testing: TH stressed	I MW	· · · · · · · · · · · · · · · · · · ·	500 w/cc		>10**13	Oxygen control
Subcritical testbed (2001-2010) Limited fissonable inventory. Could use LANSCE. TRU fuel development + test.	1-2 MW	10-50 MW. 0.75-0.9 keff	- 100 w/cc	U0 ₂ (20% enr.) metal fuel-test	>10**14	Po removal Oxygen control
ATW demonstration (2007 -) <u>Conservative approach</u> , Forgiving Operation, Could be at SRS/APT. Chemistry, FP transmutation., full TH	10 - 20 MW	500-1000 MW 0.95 keff	100 w/cc	netal fuel U - TRU core	>10**15	fully integrated waste treatment +transmutation
ATW full size burner (2015 -) <u>Full TRU destruction performance</u> . Accelerator load following, extend, burn.	20 - 40 MW	2000 MW 0.97-0.93 keff	350 w/cc	metal fuel full TRU core	>10**15	fully integrated waste treatment +transmutation

PCAST Energy R&D Recommendations for Nuclear Fission:

- Proliferation-resistant reactors or fuel cycles
- New reactor designs with higher efficiency, lower cost and improved safety
- Low-power, inexpensive reactors
- New techniques for on-site and surface storage and for permanent disposal of nuclear wastes

<u>Pyrochemistry, Subcriticality and Liquid Lead/LBE</u> technologies are not restricted to ATW applications

Most development work done / planned for ATW is directly applicable to systems (critical and subcritical) and fuel cycles that respond to all the PCAST recommendations

ATW can provide a technological bridge to a sustainable US Nuclear Future



past the burnup of the present backlog, ATW's can help sustain a viable nuclear energy strategy

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Concluding remarks

ATW can positively affect US waste management by assisting the Repository meet its objectives.

The ability to demonstrate such a means of waste destruction 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.

ATW systems could be used in a series of different scenarios, including the expanded, sustained or declining use of nuclear power. ATW technology is relevant to new and attractive nuclear designs and options.

... 5-year Program is Crucial

To the ATW Review Panel,

In the following discussion, we address some of the points raised during the feedback session shortly after completion of the Review presentations.

- Energy Multiplication 1.
- Liquid fuels 2.

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- Breakdown of ATW waste to repository 3.
- 4.
- Typical neutron spectrum Extraction and transmutation of iodine 5.

We invite and welcome further questions, as look over and evaluate the material that was presented.

Francesco Venneri and the ATW team

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1. Energy Multiplication -- How large a proton beam is necessary to drive a certain fission power in a subcritical multiplying assembly.

Starting from a basic point model

(1) m=1/(1-k)

where m is the neutron multiplication of a system, one arrives to the relation

(2) $PF=PB^{6k}/((n)^{(1-k)})$

where PF is fission power of the multiplying blanket, PB is power of the driving beam, k is keff of the blanket, and n is the average neutron production per fission. 30 neutrons per GeV proton are assumed (contained in the factor 6 at the numerator).

This would be the correct way to look at the problem of a system driven by a uniformely distributed source, where all source neutrons have the same chance at leakage as the fission neutrons.

However, this is not the case for systems driven by a centrally located neutron source, such as ATW. Here all chains start at the center and the source neutrons have a lower chance to escape or being otherwise lost, if the target/blanket is properly designed. The key to proper design for subcritical source-driven systems is to skew the loss-to-fission ratio as much as possible in favor of the source neutrons compared to the subsequent fission neutrons.

The energy multiplication is therefore strongly dependent on the specific design (geometry and fuel distribution) and a simple relation is possible only for very simplified geometries. One such geometry is the point source in a multiplying cylinder. Performing simple MCNP calculations on a typical ATW geometry (2m high, 3 m diameter, with a point source at the center, surrounded by 1 m of lead) leads to the following table:

Keff	fissions / source neutron	fission energy / source neutron	point source EM (MCNP)	unif. source EM
	F/N	MeV/N	PF/PB	PF/PB
0.83400	2.7800	556.00	16.680	10.395
0.87900	4.1300	826.00	24.780	15.030
0.90100	5.3200	1064.0	31.920	18.830
0.92700	7.3300	1466.0	43.980	26.273
0.94900	10.890	2178.0	65.340	38.499
0.96400	16.990	3398.0	101.94	55.402
0.97700	28.650	5730.0	171,90	87.886

here we have varied the plutonium fraction in the simple core geometry to calculate Keff using MCNP (k-code). MCNP then calculates the number of fissions per source neutron: F/N (assumes neutrons generated in the center of the core), and the fission energy per source neutron (here reported in MeV/N, assuming 200 MeV per fission). The ratio of fission power to beam power (Energy Multiplication, EM=PF/PB) is then

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calculated from the MCNP results assuming that 1 GeV proton produces 30 usable spallation neutrons in lead (Sunnyside experiments, and lots of LAHET MCNP calculations). Energy Multiplication is also evaluated, for the given Keff, using the formula given above (2) for the uniform source approximation.

Conclusions:

• The blanket power dependency on the target power (centrally located neutron source) depends crucially on how well the source neutrons are utilized. In a breeding design (such as Rubbia's), for instance, up to 40% of the source neutrons are captured in Thorium and do not directly contribute to the power of the blanket. Our systems (containing pure actinides) can achieve a more efficient target to blanket coupling, however appropriate design practices should be employed.

• A centrally located source will always couple to a subcritical system better than a uniformely distributed source, unless major mistakes are made in the design of the source/blanket. The two curves in the attached plot should be considered as delimiting the parameter space available to design optimization.



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2. Liquid fuels

The point was raised at the Review of the possible use of liquid fuels in ATW systems.

We have looked at liquid fuels for a very long time. The first ATW concept proposed by Los Alamos used a heavy water slurry, with actinide oxides in suspension. We have also looked in depth at molten salt (fluorides) systems similar to the Oak Ridge MSR concept.

Liquid fuels, especially molten salts are very attractive from the point of view of neutronics, because they provide the means for continuous feeding of fissile material and extraction of the fission product poisons.

After extensive evaluation, we feel that the experience base for homogeneous liquid cores is too limited, and such choice for ATW, while attractive in the longer run, would require a much more substantial investment in research and development than the present one.

Among the issues that were raised and not answered to our satisfaction during the study:

1) the low melting point salts considered by Oak Ridge for their MSR concept contained beryllium fluoride, with only limited solubility for Plutonium and the higher actinides.

- 2) Many material problems still remain unsolved from the ORNL Program.
- 3) Inventory losses due to plating, evaporation and penetration in cracks.
- 4) On-line refuelling and cleaning
- 5) Lack of clear fuel containment.
- 6) Large out of core inventories

Adopting solid fuels withoud fertile material, we face the inevitable consequence of having to compensate for burnup without the option of continuosly replenishing the fissile content of the fuel. We pointed out a few ways: burnable poisons (including Technetium), movable absorbers, reflectors and source, variable beam current.

An interesting possibility to extend fuel burnup however, which we did not discuss in any depth at the Review, is the use of encapsulated "liquid" fuels. For instance sodium fluoride with actinide fluorides dissolved in it or lead with alloyed actinides. These fuels would be cast as clad solid fuel elements at room temperature. At operating coditions, however the fuel elements would be liquid.

The advantages of the concept are: no radiation induced swelling, uniform burnup, natural separation of volatile fission products, good match with the chemical processe. The disadvantages are: corrosion problems may be severe, clad rupture could lead to the dispersion of the fuel in the coolant (sodium fluoride based fuels however will freeze upon leaking).

In our 5-year program we plan to research the possibility of these encapsulated liquid fuels as an advanced alternative to the solid metallic fuel we presented at the Review.

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3. Breakdown of ATW waste to repository

Waste Output to Repository

WR Spent Fuel -	70000 t HM
700 t	rare earth oxide
910 t	noble metal oxide
370 t	gases (Xe and Kr)
315 t	active metal oxide in engineered storage (Cs. Sr. and Ba)
cladding / as	sembly hardware - the same as for the direct disposal option
66937 t	HM
600 t	TRU to ATW
15 t	iodine to ATW

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53 t technetium to ATW

ATW Waste - 21000 t

180 t	rare earths in oxide form
230 t	noble metals in oxide form
5752 t	zirconium matrix from ATW fuel as oxide, assume no recycle
90 t	gases (Xe and Kr)
80 t	active metals in oxide form in engineered storage (Cs. Sr. Ba)
67 t	Ru metal from Tc transmutation (53 t - LWR, 16 t - ATW)
18 t	Xe from iodine transmutation (15 t - LWR, 4 t - ATW)
14400 t	cladding / hardware
80 t	spent salt from two chemical plants
18 t 14400 t 80 t	Xe from iodine transmutation (15 t - LWR, 4 t - ATW) cladding / hardware spent salt from two chemical plants

Projected TRU output to the repository is <300 kg with the majority of the TRU contained in the noble metal oxide waste form.

4. Typical neutron spectrum

We enclose a typical spectrum for the liquid lead-cooled actinide burners. We are preparing more detailed spectra for the DEMO and FULL-SIZE geometries. We will submit these as soon as they are ready.

Section 2016

5. Extraction and transmutation of iodine

Iodine Recovery

lodine is one of the two long-lived radioactive elements, the other is technetium, identified for transmutation in the ATW system. In a PWR spent nuclear fuel assembly there is approximately 86 g of iodine dissolved in 455 kg of heavy metal oxide. Iodine is released from the spent fuel matrix in two processes; spent fuel decladding and crushing (vugraph #1 of the chemistry presentation) and the direct oxide reduction and salt recycle process (vugraphs #2 and #8).

Data from Nuclear Chemical Engineering by Benedict et al. indicate that approximately one to two percent of the iodine is released during the decladding process. Iodine released in this process could be adsorbed in metal getter beds, perhaps silver impregnated zeolites or a transition metal alloy, and targets manufactured for transmutation.

The bulk of the iodine is released during the direct oxide reduction process. This process converts the metal oxide to metal (vugraph #2). Iodine is present in the calcium chloride molten salt as either calcium iodide or, because cesium and strontium are also present in the salt, cesium or strontium iodide. The concentration of iodine in the spent salt is approximately 42 ppm by weight. Spent salt from the direct oxide reduction process is not discarded but instead is treated by a two-step process and recycled (vugraph #8). First, a chlorine sparge process converts the calcium oxide to calcium chloride. Then an electrowinning process regenerates calcium metal from calcium chloride and the metal and salt are reused in the direct oxide reduction process. Iodine is released during the chlorine sparge process because of the difference in stability between the metal iodides and chlorides. Volatile iodine could be collected in a cryotrap and later alloyed, perhaps with a transition metal or rare earth alloy, and fabricated into targets for transmutation. The alloys should be stable at the operating temperature of the system and possess low vapor pressure. Candidate alloy materials include chromium and lanthanum. Chemical equilibria were verified using SOLGAS. Calculations indicate that sparging the molten salt for 2 hours with chlorine gas at a rate of 1.0 l/min would be required to remove the iodine from the salt. Complete removal of iodine from the spent molten salt is not necessary because the salt is recycled rather than discarded.

Technetium

A typographical error is present on the vugraph describing the amount of Tc present in the 70000 t HM spent fuel inventory. We have listed 5.3 t of Tc instead of the correct number of 53 t of Tc.

To the ATW Review Panel.

In the following discussion, we address some further points raised after completion of the Review presentations.

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- Backlog Burndown Scenario More accurate neutron spectrum information Delayed neutron fraction and subcriticality Pb-205 production and inventory 2.
- 3.

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Francesco Venneri and the ATW team

1. Transmutation in 65 Years. (A spent fuel back-log burndown scenario)

One of the viewgraph presented at the meeting contained an error. The viewgraph illustrating the 65-year backlog burndown scenario had 20 ATW burners operating in staggered fashion for 65 years, with a forty year plant lifetime). The right number of plants is 23. We regret the error, however, this gives us a good opportunity to explain the scenario in some detail.

Why is the burndown scenario important?

To us in the ATW project, a large part of the value of the ATW concept is that it will be very effective at achieving the "clean slate" of zero-waste backlog for nuclear power, while at the same time introducing or preserving essential nuclear technologies for the next generation of nuclear systems (the technology "BRIDGE"). We believe the "clean slate", or at least the capability to reach it in a reasonable amount of time is essential in establishing confidence in an expanded role for nuclear power production.

By the year 2015, there will be 70,000 tons of US spent fuel containing about 600 tons of plutonium and higher actinides requiring disposal. The ATW objective in the backlog burndown scenario is to treat the spent fuel backlog, destroy the transuranics and selected fission products, and prepare the resulting waste for permanent disposition in a geologic repository.

A number of ATW systems is brought on line to accomplish the objective over a 65 year period. Each ATW system consists of an accelerator (1 GeV, variable current 20-40 mA, pyrochemical plant (50 tons/year throughput) and a subcritical 2000 MWt LBE burner. Several of these ATW systems can be located together in one facility. To reduce transponation and other factors related to moving fuel assemblies, one could consider three facilities with up to 8 ATW systems at each location.

Of particular relevance is the fact that each 2000 MWt ATW burner can destroy up to 650 kg of actinides (plutonium and transuranics) per year, and that 700 MWe would be available for distribution to the grid from each burner.

The scenario works as follows: Roughly every two years two new ATW systems are brought on line for the first 20 years. At year 25, the three last plants are added. As the burners and accelerators reach the end of their operational life, they are decommissioned, and the fuel is sent to feed other still operational burners. Eventually, the last three systems in operation will receive all the remnant waste and destroy it down to less than 1 ton over a protracted (5 years) inventory burndown.

Two major characteristics distinguish the accelerator-driven subcritical systems from the critical reactors that had been evaluated for actinide destruction, such as the ALMR: (1) ATW does not produce higher actinides from fertile materials such as 238U. (2). Because ATW is not constrained by a criticality requirement it can burn down the end-of-life (EOL) actinide inventory to very low levels.

Critical reactors are considerably less efficient in the destruction of transuranic elements (about 125 kg/a, for the same 2000 MWt power rating). They tie up a very large amount of actinides in the core (to maintain criticality), and compared to the actinide inventory, only a small amount of actinides can be destroyed. Critical reactors therefore would require a proportionally larger number of installations over multiple generations (about 300 years) to achieve the same degree of transuranic inventory reduction that ATW systems would reach in 65 years.

ATW Backlog Burndown scenario

Number of ATW units, coming on line, closing down

Time (years)



-1 ATW burner (40 years operational life, 2000 MWt)-





2. Neutron spectrum.

The neutron spectrum of the current 2000 MWt Accelerator Transmutation of Waste (ATW) waste burner point design is given in Figure 1. As shown in the Figure, the average total flux seen by the waste during the three burn cycles is 1.09×1016 n/cm²s. Sixty percent of this flux has an energy greater than 0.1 MeV. The neutron flux with energy greater than 20 MeV is relatively small. With the proposed fuel shuffling scheme, the total fluence seen by the fuel and clad before it is discharged after three cycles is 1.7×1023 n/cm² (E > 0.1 MeV).



3. Delayed neutron fraction and subcriticality.

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This figure was shown at the presentation, however it was not included in the material that was handed to you at the presentation.

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The figure illustrates the delayed neutron fraction for many fissionable isotopes. Delayed neutrons come from the decay of a few fission products, and are the basis for control of critical nuclear systems. The more the better: a reactor will be easier to design and operate safely. If delayed neutrons did not exist, all nuclear systems for power production would have to be externally driven.

The picture shows how close to prompt critical reactors have to get before starting to work as self sustained systems through the action of delayed neutrons. Very literally, it says that thermal reactors (based on 235U fuel) operate at 0.995 prompt Keff, and Fast reactors (based on 239Pu fuel) operate at 0.998.

The figure also show that the "dollar" measurement of reactivity (which is the amount of reactivity equivalent to the delayed neutron fraction) is not a constant. A dollar for a thermal reactor is about three times as large as a dollar for a fast reactor. A reactor has to stay close to prompt critical within this 1-dollar margin at all times.

Thermal reactors have learned to live and operate quite safely within this margin. Reactors based on plutonium (fast reactors)have a very thin operating margin, and it gets worse going towards the higher actinides. These reactors need a significant buffer of 238U to dampen reactivity excursions with significant negative prompt feedback to the point that control rods can be effectively used.

The great claim of subcritical system is that they can burn pure transuranic cores, thereby achieving very fast destruction rates. The task is doubly difficult because:1) there is no 238 to sweeten the reactivity response of the fuel (typically with negligible or positive Doppler), and 2) the delayed neutron fraction (dollar margin) is significantly smaller for the transuranic actinides than it is for the 235U based fuels. In a pure waste destruction mode, the equivalent level of control and safety present in LWR reactors can be achieved by operating the system subcritically.

Subcriticality allows any fuel originated (internal)reactivity transient to proceed slowly enough for external feedback to become dominant and reestablish equilibrium. As shown during the review, lead (being a poor neutron absorber and excellent reflector) has a strong negative temperature feedback on reactivity (this is not the case for sodium). The slow nature of the transient allows time for the negative response of the lead coolant to stabilize the excursion.

The chosen subcriticality level should allow for transients in all expected conditions to be self limiting. The specific level is strongly dependent on the burner design.



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4. Pb-205 production rate and inventory.

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The production rate of Pb-205 (from neutron capture in Pb-204) in the 2000 MWt ATW point design is calculated to be 7 curies per full power year. Because of Pb-205 burnout (and Pb-204 burnout if total coolant volume is relatively low) the total Pb-205 inventory after 40 full power years of operation is expected to be less than 200 curies.

Pb-205 decays via electron capture to the stable isotope TI-205. There are no alpha, beta, or gamma rays emitted in the decay, only an x-ray. Because the 200 curies of Pb-205 would be mixed with approximately 20° tons of lead-bismuth eutectic, the radiological hazard from the Pb-205 should be relatively small.

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