

ACCELERATOR-DRIVEN SUBCRITICAL FISSION TO DESTROY TRANSURANICS AND CLOSE THE NUCLEAR FUEL CYCLE*

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Abstract

A design for accelerator-driven subcritical fission in a molten salt core (ADAM) has been made for the purpose of destroying the transuranic elements in used nuclear fuel as fast as they are made in a conventional nuclear power plant. The oxide fuel is extracted from the used fuel assemblies into molten chloride salt using pyroprocessing, and the transuranic, uranium, and fission product salts are separated into three batches using electroseparation. The transuranic salt is then transferred to a subcritical core, with neutron gain 0.97. The core is driven by 800 MeV proton beams from a 12 mA CW strong-focusing cyclotron. The transuranics are destroyed and the fission heat is used to produce electric power. Simulations of many potential failure modes have been performed; the core cannot reach criticality in any failure-mode scenario considered. It operates as an energy amplifier with an energy gain ~ 5.5 .

INTRODUCTION

Today nuclear power plants generate 20% of the electric power in the United States [1]. Until recently nuclear power comprised 20% of the grid in Germany and 30% in Japan, but Germany has moved to end their nuclear power production and Japan has idled their reactor fleet. Those decisions reflect a growing public concern about the safety of nuclear power. The meltdowns at Three Mile Island [2], Chernobyl [3], and Fukushima [4] underscore that this abundant source of energy can also produce extreme hazards.

The most enduring hazard of nuclear power is the large quantity of hazardous radioisotopes in used nuclear fuel (UNF). The most dangerous among those are transuranics (TRU, elements beyond uranium in the periodic table). The transuranics contained in the $\sim 70,000$ tons of UNF in the US have a radiotoxicity $>10^{13}$ Sv and half-lives of 10^5 - 10^6 years. The present accumulation of UNF also still contains about 1/3 of the entire US reserves of uranium. Long-term storage would pose the risk unto the generations of future release of immense radiotoxicity, and would sequester a major portion of available uranium resources. ADAM has been designed to offer an alternative: to destroy the transuranics, to recover the uranium for future use, and to produce 10x more energy than was produced in the first use of the fuel.

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ADAM OVERVIEW

ADS Core Neutronics

The individual ADS core must be sized to optimize the normalized burn rate \dot{T}/T ; i.e. to minimize the TRU inventory required to sustain core operation. Figure 3 shows the energy dependence for neutron capture on ^{238}U (which breeds TRU) and for n-induced fission of the dominant TRU isotopes. The fission cross sections for most TRU isotopes are significant only for ultra-fast neutrons (>1 MeV). Optimization of fast spectrum for the ADAM core places strong constraints upon the core size and geometry and upon the fuel salt composition. The optimized core is shown in Figure 1, and its neutronics properties are summarized in Table 1. In its spectrum 20% of the neutrons have >1 MeV energy. It operates with a neutron gain $k_{\text{eff}} = 0.97$, produces $280 \text{ MW}_{\text{th}}$, and requires a 10 MW proton driver. The optimized burn rate is $\dot{T}/T = 5.6\%/\text{year}$, corresponding to a destruction time of 18 years.

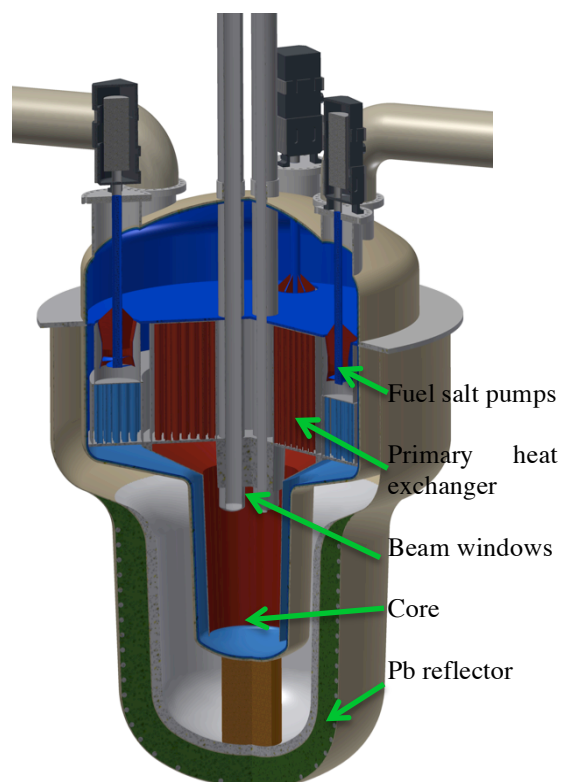


Figure 1: ADS molten salt core assembly.

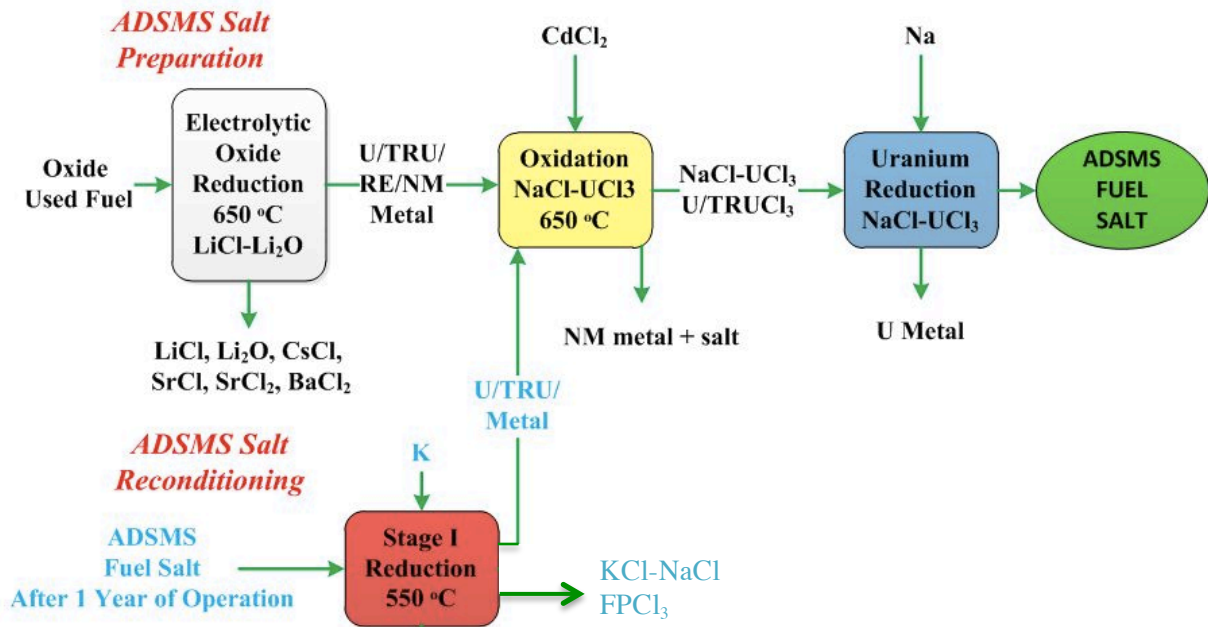


Figure 2: Pyroprocessing and electro-separation to prepare the fuel salt.

Fuel Salt Preparation and Reconditioning

The fuel salt for the ADAM core is a eutectic of TRUCl₃, UCl₃, and NaCl. It is prepared by a sequence of reduction and oxidation steps, shown in Figure 2. Fuel assemblies are chopped and crushed, and the oxide fuel is extracted from its Zircalloy cladding into molten salt (pyroprocessing [5]). Successive oxidation and reduction steps are used to plate out the uranium and to separate the remnant into separate batches of TRUCl₃ and FPCl₃ (FP = fission products). All of the steps of this electro-processing have been developed into small-scale practice at ANL, INL, and KAERI [6].

The ADAM fuel salt contains as molar constituents TRUCl₃ (15.2%), UCl₃ (13.6%), NaCl (70%), and FPCl₃ (1.2%) [7]. The fuel salt has a melt temperature of 525 C and a boiling point of ~1500 C. The primary heat exchanger is integrated directly into the Ni vessel, and operates with an inlet temperature of 675 C and outlet temperature of 575 C.

As the ADAM core burns TRU its k_{eff} decreases. We modulate the proton beam power to maintain constant thermal power in the cores (increase the drive beam power from 8 MW to 10 MW) for a period of 3 months. At the end of 3 months we restore k_{eff} to its starting value by adding 90 kg of TRUCl₃. We can continue doing this for 5 years (20 cycles), at which time the fuel salt is transferred back to the electro-processing system, the accumulated FPCl₃ is removed, and the fuel salt is returned to the core vessel to begin another 5-year operating period.

Proton Driver

Each ADAM core requires a total of 12 mA of 800 MeV continuous proton drive beam. We have developed a design for a two-stage strong-focusing cyclotron (SFC) that can provide that performance [8]. The acceleration

Table 1: Main Parameters and TRU-burning Performance of ADAM Core compared to Fast Critical Reactor Designs

System	ADAM	SFR	GFR	LFR
Net TRU Destruction	0.84	0.74	0.76	0.75 g/MW _t -day
System Power	290	840	600	840 MW_t
Outlet Temperature	665	510	850	560 C
Thermal Efficiency	44	38	45	43 %
Power Density	200	300	103	77 W/cc
TRU Inventory	1733	2250	3420	4078 kg
Fuel Volume Fraction		22	10	12 %
TRU Enrichment	53	44 - 56	57	46 - 59 % TRU/HM
Fuel Burnup	129.5	177	221	180 GWd/tHM

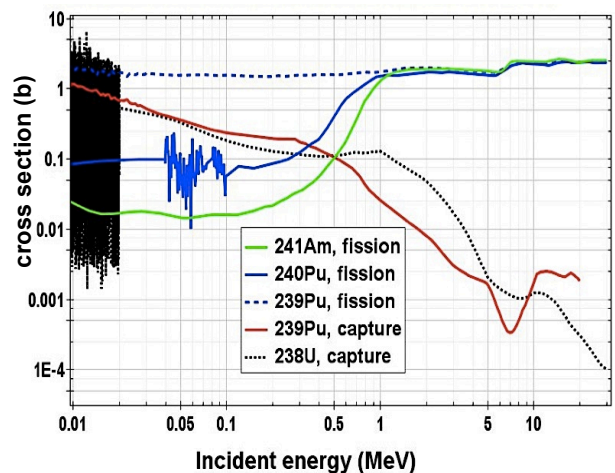


Figure 3: Energy dependence of cross sections for fission and capture by 238U and the dominant transuranics.

sequence is shown in Figure 5. It begins with acceleration of 100 mA CW to 6.5 MeV in the 350 MHz LEDA rf

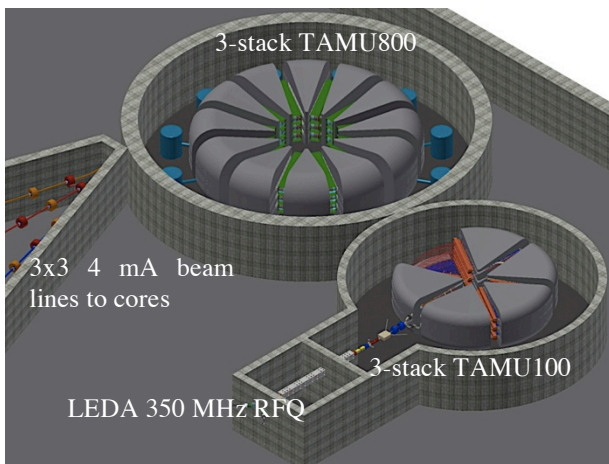


Figure 5: Acceleration chain for an ADAM site.

quadrupole (RFQ) [9]. The beam is then subharmonic-modulated, split into three 117 MHz beams, and passed through a sequence of 6-D collimators to yield three beams each with a normalized emittance $< 1\pi 10^{-6} \text{m}$ and a phase width $\pm 5^\circ$. The beams are then injected into a 3-stack of 100 MeV strong-focusing cyclotrons.

The world-record CW beam power for a proton accelerator is the PSI isochronous cyclotron [10]. It produces 2.2 mA CW at 590 MeV. Two issues pose the main limits to beam current in a cyclotron: the succeeding orbits overlap strongly so the defocusing action of space charge is exacerbated; and it has only weak focusing so that the betatron tunes migrate throughout acceleration and cross multiple resonances. We solved both of these problems in the SFC by incorporating two new elements: superconducting $\frac{1}{4}$ -wave slot-geometry cavities that provide sufficient energy gain per turn to fully separate the orbits; and beam transport channels that provide alternating-gradient strong focusing to maintain constant betatron tunes throughout acceleration.

The details of the SFC design have been presented previously [8]. Three SFCs are configured as a flux-coupled stack, in which the dipole field for each SFC is created by

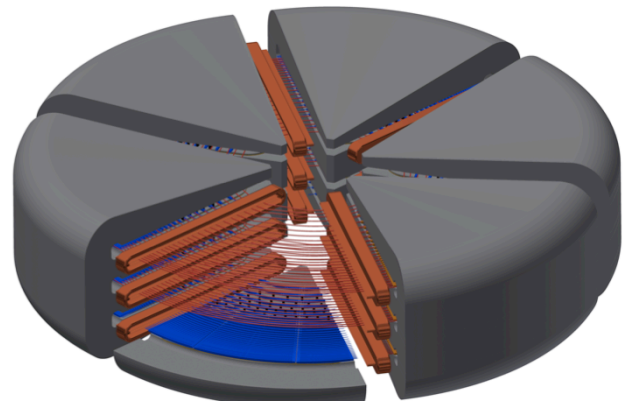


Figure 6: 3-stack of 100 MeV strong-focusing cyclotrons, with cutaway to show cavities, BTCs, and orbits.

a pair of cold-iron flux plates (Figure 4b) that are supported within a warm-iron flux return so that Lorentz forces on each flux plate cancel [11].

The superconducting cavity and beam transport channels are shown in Figure 6. The Nb superconducting cavity operates at 4.2 K and produces a ~ 2 MV acceleration. It is designed with fairly conservative surface field limits $- 21$ MV/m, 54 mT, and has provisions to suppress multipacting [12]. The rf power for each cavity is delivered to a linear array of input couplers, distributed along the upper and lower lobes of the cavity as shown in Figure 4a. Each coupler is driven by a solid-stage power source, and the linear array makes it possible to deliver input power in the same spatial distribution that is delivered to the circulating orbits of beam, so that beam loading does not drive transverse modes.

The beam transport channel (BTC) contains a single layer wire-wound Panofsky quadrupole winding and a window-frame dipole winding, both utilizing the superconductor MgB_2 which operates in the 15-20 K temperature range. An arc-shaped BTC is aligned along each equilibrium orbit in each sector as shown in Figure 4b, and is configured as an F-D doublet. The dipole winding is used to maintain precise isochronicity on all turns.

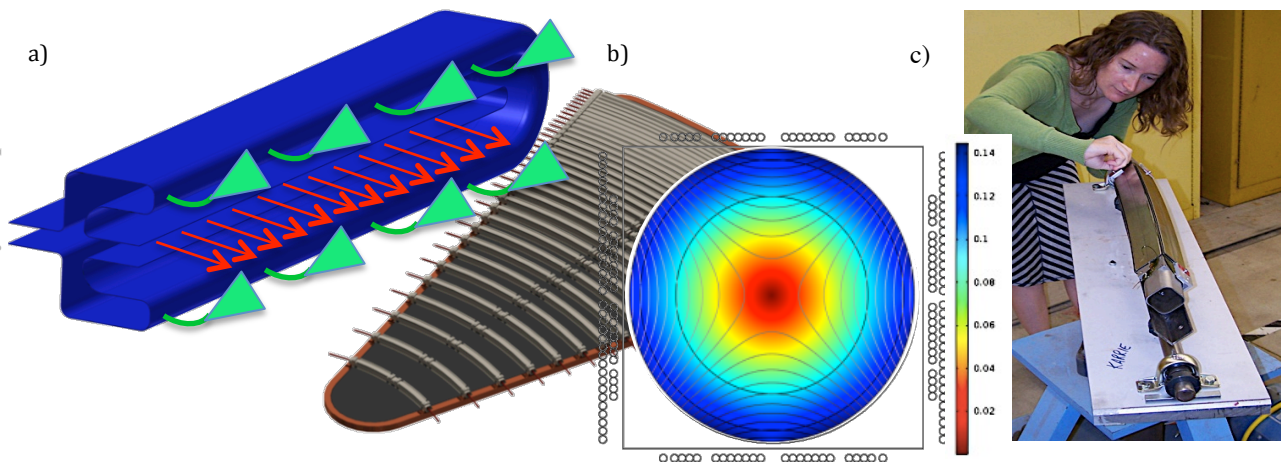


Figure 4: Innovations in the strong-focusing cyclotron: a) 117 MHz $\frac{1}{4}$ -wave superconducting cavity with linear array of input couplers (green); b) beam transport channels on a flux plate; c) detail of the MgB_2 windings on a BTC and its quadrupole field distribution (max gradient 6 T/m).

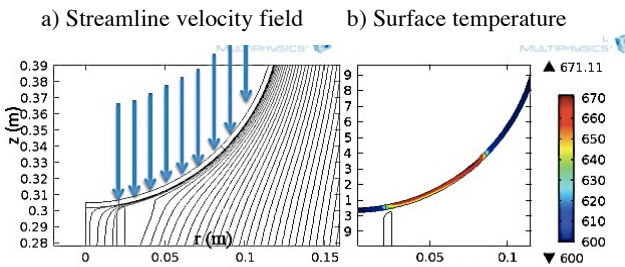


Figure 7: a) Molten salt flow on a proton beam window; b) Temperature profile on the window with 4 mA beam.

A companion paper [13] presents studies of the beam dynamics of the SFC for low-loss acceleration of high-current proton beam. We find that the elimination of overlapping orbits, the control of betatron tunes, and the suppression of transverse mode excitation by wake fields enables us to maintain stable acceleration of 12 mA CW through both SFCs to 800 MeV energy without beam breakup and with low loss for injection and extraction.

Delivery of 4 mA Proton Beam into Molten Salt

Each core requires a total of 12 mA drive beam. In order to operate within presently achieved beam window limits, we chop the proton beam after the RFQ to deliver $\sim 10 \mu\text{s}$ bunch trains for acceleration, we split the 800 MeV bunch trains from each SFC to feed 3 transport lines, and we deliver the 3 bunch trains to 3 hemispherical Nb windows (Figure 1). The closed-circuit flow of molten salt in the core is channelled to deliver a chimney flow to cool each beam window, as simulated in **Error! Reference source not found.**

Safety Considerations

The molten salt provides the spallation target and heat transfer medium for the beam windows, and it cannot be shocked by interruption of drive beam. All ADS designs that utilize a core based upon solid fuel pins have the problem that interruptions of drive beam (which happen

every day at any extant accelerator) would thermally shock the fuel cladding which can lead to cracking.

All of the fuel salt is completely contained within the Ni core vessel and 5 shells of outer structure throughout a 5-year operating period. By contrast all previous core designs using molten salt pass the molten salt frequently through external circuits for reprocessing and re-conditioning, opening the risk of leaks.

The core vessel contains a removable inner vessel (in contact with the fuel salt) made from a single-piece of CVD Ni with no weld seam, which is resistant against molten salt corrosion. The Ni is much more robust against embrittlement from neutron damage in the ultra-fast spectrum of the ADAM core than it would be for a thermal spectrum. The CVD Ni vessel is encased in a spiral-wrap Hastelloy-N structure that provides mechanical support for the Ni can. An array of K vapor heat pipes is bonded to the outer surface of the Hastelloy shell and passively heat-sinks its surface to $\sim 400 \text{ C}$ during normal operation (consuming $\sim 2 \text{ MW}$ of heat) and during power and cooling failure modes. Maintaining the temperature of the Hastelloy at 400 C preserves its high tensile strength and toughness, which would be compromised if the Hastelloy operated at core temperature.

Many failure modes have been modeled, including loss of primary and/or secondary heat exchanger, loss of drive beam, loss of controls, and cracking of the core vessel. No failure mode studied can lead to leaking fuel salt beyond the multi-layer vessel, and no failure mode can produce criticality.

IMPLEMENTATION TO DESTROY TRANSURANICS

The ADAM core is sized to optimize the destruction of transuranics \dot{T}/T . Three ADAM cores are required to destroy TRU at the same rate that it is produced in a typical GW_e nuclear power plant. Figure 8 shows the site plan for an ADAM facility containing three cores and a 3-

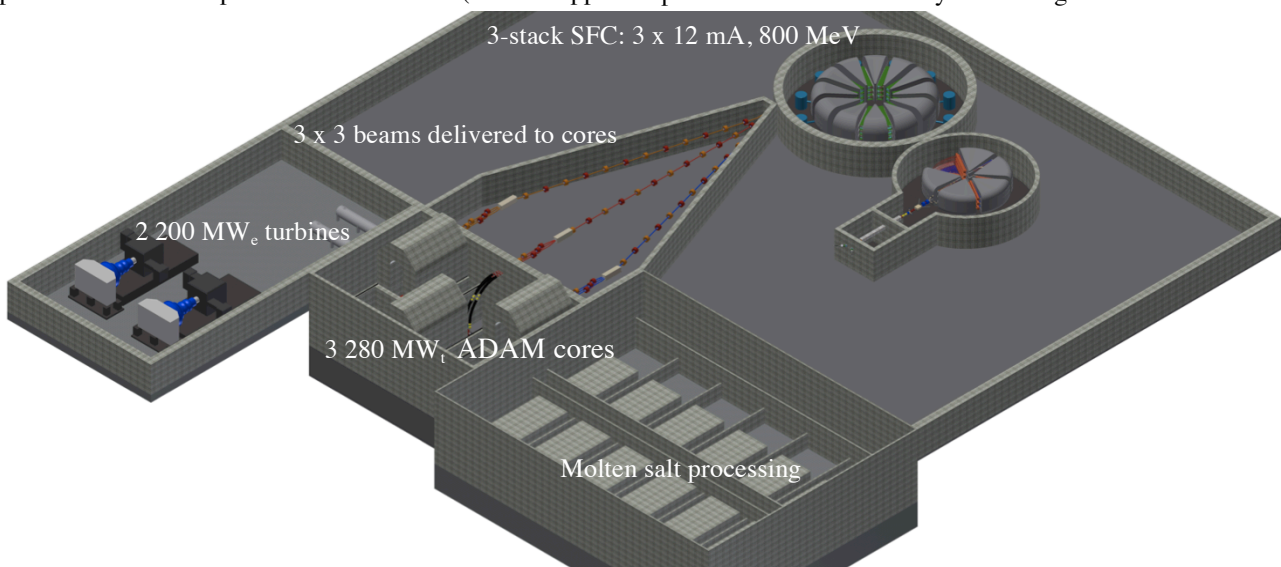


Figure 8: ADAM facility that destroys transuranics at the same rate they are produced by a GW_e nuclear power plant.

stack SFC proton driver. It is appropriate in capacity to co-locate with an existing power plant, process its spent nuclear fuel, destroy the transuranics, recover the uranium, and generate uranium. The three ADAM units produce $3 \times 290 \text{ MW}_t$ of heat, which generates $\sim 44\% \times 870 = 380 \text{ MW}_e$. The SFC systems operate with $\sim 50\%$ efficiency, so it requires $\sim 3 \times 10 \text{ MW} / 50\% = 60 \text{ MW}_e$ to operate the ADAM installation. The ADAM installation therefore is essentially an energy amplifier with a gain ~ 5.5 . For as long as the adjoining GW_e power plant operates, its ADAM companion will generate $\sim 320 \text{ MW}_e$ of co-generated power to augment the plant's output while it destroys its hazardous waste.

Table 1 summarizes the performance parameters of the ADAM core, and compares them with the performance of several fast critical reactors that have been designed to destroy transuranics [14]: SFR is a sodium-cooled fast reactor; GFR is a high-temperature He gas-cooled fast reactor; and LFR is a molten lead-cooled fast reactor. Notably the ADAM core performs as well or better than any critical core design, and conveys the benefits for safe subcritical operation discussed above. ADAM provides a feasible candidate method to destroy the transuranics in used nuclear fuel and close the nuclear fuel cycle.

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