The Future of Nuclear Energy; Chemistry is the Problem, Accelerators the Solution

Charles D. Bowman* and Ganapati Myneni#

*ADNA Corporation, Accelerator Driven Neutron Applications, Los Alamos, NM, USA #Jefferson Laboratory, Newport News, VA USA

Abstract

Throughout mankind's existence it has been impossible to mine nuclear weapons material directly from the earth. Although natural uranium can be mined, separating the ²³⁵U still requires difficult and expensive technology and time. And even if ²³⁵U is obtained by isotopic enrichment, it can be rendered useless for weapons simply by mixing it with natural uranium and returning it to the earth.

Plutonium is another matter as there is no natural plutonium with which to dilute it. If weapons plutonium (W-Pu) is returned to the earth, it can be dug up quickly and cheaply. It is commonly claimed that commercial reactor waste containing plutonium (C-Pu) containing large amounts of ²⁴⁰Pu is not weapons useful and therefore need only be made unavailable by burial. However ²³⁹Pu lives about three times as long as ²⁴⁰Pu, so the ²⁴⁰Pu decays away leaving W-Pu after one ²³⁹Pu half life. Yucca Mountain as planned therefore would eventually contain about 350 tons of W-Pu that need only be dug up and chemically separated...sufficient for about 70,000 nuclear weapons from this one site. Worldwide nuclear would eventually enable millions from many sites....an absurd legacy to permit from today's nuclear technology.

The world cannot leave underground weapons plutonium or these dangerous commercial isotopic mixtures of plutonium. These remnants must be burned in reactors optimized for this purpose. That will happen only if burning is cost competitive with present reactors, but such waste-burning critical reactors after sixty years still do not exist. The ideal reactors are GEM*STAR reactors based on accelerator-driven molten-salt thermal-spectrum systems. GEM*STAR reactors are advantageous for many reasons, but mostly because *they require <u>no chemical/isotopic</u> separation for fuel, <u>no chemistry for solid fuel preparation, and <u>no chemical reprocessing, and because they eliminate the chemical overhead</u> of fission product and higher actinide waste streams and <u>chemistry-driven</u> nuclear weapon proliferation. The implementation of accelerators displaces expensive chemistry thereby enabling nuclear energy cheaper than any other system. GEM*STAR can, without chemistry, burn safely and economically both W-Pu and C-Pu into isotopic mixtures that cannot decay to W-Pu. Nuclear energy has no long term future without eliminating the long-term legacy of mining nuclear weapons material from today's critical reactors.*</u>

Introduction

Most nuclear technologists believe that the main advantage of a subcritical reactor is the safety from its operation with less than a critical mass. They also believe that the main advantage of liquid fuel is that the fuel can incorporate automatic and fail-safe drainage of fuel if the reactor should overheat. These are nice advantages over solid fuel critical reactors, but focus on these features misses the far more important single-pass deep burning feature of subcritical systems and other advantages of the technology.

If this community is to understand our present situation in subcritical nuclear technology fully, it must examine how it began, how it evolved to our present situation, and how to break out from the present stalled status.

Twenty one years have passed since the Accelerator Transmutation of Waste (ATW) program was terminated in 1996 at Los Alamos. Our seminal paper published in Nuclear Instruments and

Methods became by 1993 the most popular paper ever to come out of Los Alamos except Robert Serbers "The Los Alamos Primer" on how to build the atomic bomb. The Los Alamos subcritical system concept swept the nuclear world with virtually every U. S. and international nuclear engineering school clamoring for a talk from Los Alamos. In addition to running its own program, the U. S. State Department asked Los Alamos ATW to undertake Project 17 of the International Science and Technology Center (ISTC) program in Moscow to help keep Russian nuclear weapons technology out of the Middle East. At its peak Los Alamos was engaged with about 450 Russian weapons scientists on ATW from six institutions, and the Russian work was of the highest quality.

The Los Alamos program search for the best means for bringing accelerators into nuclear technology eventually focused on thermal spectrum molten salt. In the meantime the international fast reactor program was viewing subcritcal systems with alarm as it threatened serious funding competition. The U. S. DOE Nuclear Energy Office, which was completely dominated by fast reactor advocates, made it clear to Los Alamos management that no element of its technology array was to be threatened by ATW. If accelerators were going to play a role, isotopic enrichment, solid fuel fabrication, fast spectrum technology, chemical reprocessing, and conventional geologic storage were not to be disturbed.

Los Alamos ATW was in deep trouble because an accelerator was going to be a significant addition to nuclear technology and the capital and operating costs were not sustainable. If the accelerator was going to enter nuclear, others of the DOE's protected technology array would have to go. In addition the ground rules from DOE-NE were that the technology would have to be fast spectrum sodium coolant technology. This technology's main advantage was in plutonium breeding in a world of imminent shrinking uranium resources, and a lesser one in fewer higher actinides to complicate reprocessing. It carried many serious disadvantages including sodium containment, more than 100 critical masses in case of a meltdown, less effective delayed neutron and Doppler broadening influence, requirement for reprocessing, a fission product and a minor actinide waste stream, and others.

In nations with competitive nuclear technology markets, fast spectrum reactors have never found a place and don't stand a chance owing to inferior economics unless the price of natural uranium skyrockets. Figure 1, showing the U. S. DOE's picture of how nuclear technology was supposed to work, is perhaps the best graphic possible for illustrating the problem with the direction of the U. S. DOE-NE's leadership. While either solar and wind could offer a slide with only one action when taking their case to Congress, nuclear required ten different interconnected and coordinated facilities or operations. Nuclear obviously needed simplification and we make the case below that the accelerator provides it.

Nevertheless these were the conditions in 1996 when internal funding from Los Alamos disappeared. It did not help that competing interests to accelerators pretended to open a small crack in the wall of opposition to accelerators by proposing that they might be used for burning Np and Am and that fast spectrum technology would be optimum for these nuclides. Some saw this as a life raft and grabbed what turned out to be a kiss of death for them as they embraced the fast spectrum and the trivial role of burning Np and Am burning that will never be separated from spent fuel.

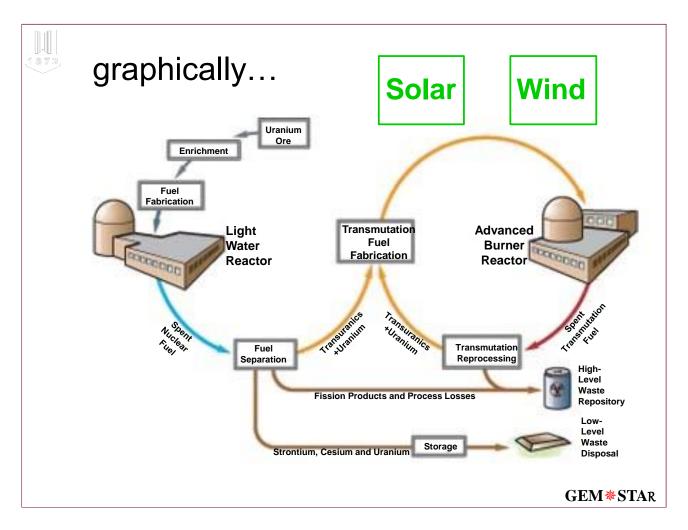


Fig. 1. U. S. DOE's fuel cycle for nuclear compared with solar and wind in green. Some wish to complicate nuclear even further by adding on an additional lines and boxes for separation of Np and Am and burning them in a subcritical fast reactor.

Rather than continuing at LANL after our ATW project was turned over to our Argonne National Laboratory competition in 1997 for a path to slow death, Bowman left LANL to form ADNA Corporation (*Accelerator Driven Neutron Applications*) for the purpose of advancing thermal-spectrum molten-salt accelerator-driven reactor technology from the private sector. ADNA has found enthusiastic support from 25 persons who are receiving stock in proportion to their effort to help advance subcritical nuclear technology. Recently we received outside investment valuing ADNA Corporation at \$12.5 million, which will enable us to advance more rapidly. ADNA's technology is called GEM*STAR. It derives from our early descriptor "Green Energy Multiplier * Subcritical Technology for Alternative Reactors." The external investment was forthcoming because of six major ADNA-led advancements that are described below.

ADNA Corporation Advances 2007 to 2016

- Continuous Flow
- Vector graphite
- Cellulosic biomass to diesel
- Viability of ocean uranium
- Much higher accelerator performance with superconductivity
- Doubling the efficiency of r.f. power generation

Fig. 2. ADNA Corporation's advances since formation in 2007

Continuous fuel flow

Organizations like the U. S. DOE Nuclear Energy Office have insisted on solid fuel for decades apparently for the simple reason that solid fuel works. Of course there are problems. One may begin with the radioactive volatiles that build up to pressures in the 1000 psi range as the fuel burns, and one must worry about this volatile release into the environment in reactor accidents. Defenders of solid fuel prominently announce that the reactor design includes "defense in depth" with three levels of protection. If (1) the fuel cladding fails, then (2) the pressure vessel confines the gases, and (3) the containment vessel is present to confine any gases that escape into the pressure vessel.

Why choose a fuel form that requires defense in depth? Molten salt liquid fuel does not allow these gases to accumulate to high pressure as the liquid form allows the gases to be collected soon after they are produced and to then be piped to storage. This storage can be as far from the reactor as necessary to avoid any concern that a reactor accident might release radioactive gases. We should mention that the solid fuel of the Fukushima reactors released almost one million times more radioactive gas than if the reactor had used liquid fuel.

As fuel burns, the solid fuel reactivity changes, so control rods are required. But in a liquid fuel reactor with properly arranged continuous flow, fuel flows in and out so that reactivity does not change. If the reactor reactivity does not change, control rods are not required. This is

illustrated in Fig. 2 showing schematically the deep burning of weapons plutonium (W-Pu) in one pass. The reactor is shown as a cup of liquid with an overflow pipe inside to a holding tank below. A pitcher is shown containing a carrier salt mixed with W-Pu with the mixture being added at the rate of 30 g of W-Pu per hour with the isotopic mixture 93% ²³⁹Pu and 7% ²⁴⁰Pu. As a pitcher of fuel is added, a pitcher overflows. Because fission power is being generated, the isotopic fuel of the overflow is different from the inflow. For a fission power of 500 MWt the outflow is 7.5 g/hr of plutonium with a fission product of 22.5 g/hr. The isotopic distribution of the burned plutonium is ^{239, 240, 241,242}Pu 52.4%, 25.4%, 10.6% and 11,7 %.

It is important to note the obvious point that the overflow isotopic composition is the same as that in the tank. This means that as soon as the pitcher of liquid is added to the cup and mixed, the fed plutonium is immediately and irreversibly transformed to non-weapons plutonium in a single pass. No initial solid fuel fabrication, no control rods, no chemical reprocessing, no fission product waste stream, and no secondary fuel fabrication.

At any point, the overflow tank can be remotely emptied by a pressurized He pipe for storage away from the reactor without personnel involvement or radiation exposure. As for Np and Am, they are deeply burned also without the need to chemically separate them. The green line at the upper right shows volatile fission products being carried away by a closed cycle He flow.

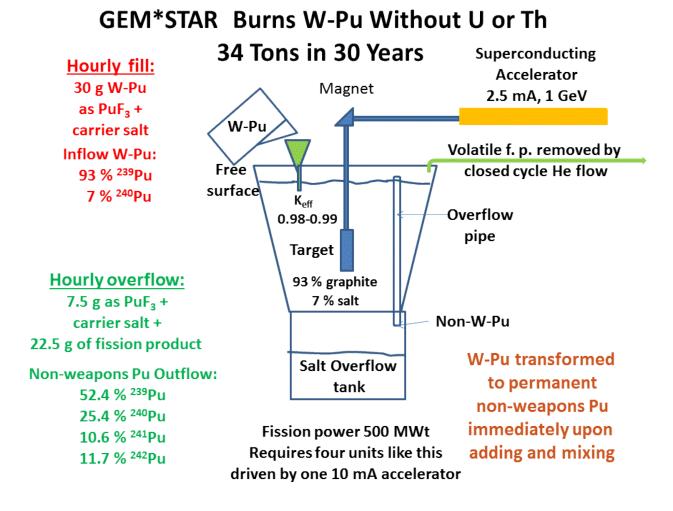


Fig. 3. The deep burning of excess weapons plutonium is illustrated using GEM*STAR's patented continuous flow system. The reactor is in the form of a cup filled 93 % with graphite containing space for a 7 % volume of molten salt and with an overflow tube so that the level in the cup stays constant. For every liter of salt poured in, a liter of salt overflows to the tank below. In one pass ³/₄ of the fed W-Pu is burned and the remainder is transformed to an isotopic mixture impossible to use for weapons because of the spontaneous fission neutrons from ²⁴⁰Pu and ²⁴²Pu. Note that the W-Pu is immediately transformed to non-weapons plutonium as soon as the added salt mixes in the cup. Four of these units driven by one accelerator buildable with today's technology could burn 34 tons of W-Pu in 30 years although logistics might make it desirable to locate the four burners on four separate sites with smaller 2.5 mA accelerators. The same system can deep-burn natural uranium fuel, LWR spent fuel, thorium or depleted uranium through at least two recycles without reprocessing.

There are no chemical separations; only the conversion of plutonium metal to PuF_3 by burning in fluorine gas. The addition of the accelerator has eliminated chemical separations as well as solid fuel fabrication, control rods, and multiple waste streams. That's why GEM*STAR will succeed economically. ADNA Corporation holds the patent to this continuous flow concept.

The burning of natural uranium fuel is much the same. Mined uranium oxide is converted to UF₄ and GEM*STAR can be fed UF₄ directly because with a subcritical system isotopic enrichment is not required. Significant subcriticality ($k_{eff} = 0.96-98$) enables UF₄ to be deeply burned in a single pass. The first pass product can be economically recycled to GEM*STAR for further burning with twice the accelerator power using today's accelerator technology. As technology improves accelerator efficiency, the second pass output can be recycled again without chemical separations as accelerator technology advances. This cycle can continue until accelerator technology ceases to advance. Much higher burn-up of mined uranium can be achieved using accelerators with GEM*STAR compared to LWRs without isotopic enrichment, and without the reprocessing and solid fuel fabrication and refabrications required by fast spectrum FBRs.

With GEM*STAR the world's enormous already mined reserve of depleted uranium is also an attractive fuel. The accelerator requirement is higher but depleted uranium is practical and is essentially free fuel.

Thorium can provide energy as cheaply as natural uranium with GEM*STAR and there is no chemistry requirement for separating out ²³³U or any proliferation concern related to ²³³U as weapons material. Because GEM*STAR fuel can be handled remotely and because with GEM*STAR there is no reprocessing requirement for deep burn-up, the hard 2.6 MeV gamma ray is not factor of concern for GEM*STAR operations.

The final advantage of molten salt is that any fuel feed can be handled, so that the difficulty of qualifying different fuels faced for solid fuel performance is not an issue. Fig. 3 shows the eliminations that are practical with molten salt and subcriticality and illustrates how new technology can drive out old technology...in this case nuclear chemistry. Nuclear must move on just like any other technology.

Vector graphite

Neutron transport programs like MCNP assume that the transport material has no structural anisotropy, but the GEM*STAR moderator material is graphite and it can be produced with anisotropic properties such as heat conductivity, electrical conductivity as well as neutron diffusion properties. The neutron thermal coefficient depends on the direction neutrons move through anisotropic graphite. In an accelerator driven system with a target at the center, it is desirable to have the neutrons move preferably away from the source. But when the neutrons near a graphite neutron reflector at the reactor boundary, it would be desirable to have them be preferably reflected back toward the neutron source.

Therefore an optimized reactor moderator will have one vector orientation near the neutron source and another for the reflector. The influence on k_{eff} is small, but when an accelerator driven system is at $k_{eff} = 0.99$, it reduces the required accelerator power by 50%, and at $k_{eff} = 0.98$ by 25%. Taking advantage of this vector property is a major factor in reducing capital and operating costs. Measurements illustrating the advantage of vector properties of graphite were studied at the Triangle University Nuclear Laboratory (TUNL) at Duke University using a graphite pile with pulsed neutrons from the TUNL tandem accelerator. MCNP code managers at Los Alamos have not yet incorporated vector properties of crystalline materials into near thermal neutron transport.

Cellulosic biomass to diesel/gasoline

ADNA Corporation recognized early the advantages of liquid fuel and molten salt in particular for an accelerator driven system. We have already discussed three of these; (1) prompt removal of volatile fission products, (2) fail safe draining of fuel from moderator if temperature requires it, (3) hands-off and remote fuel transport through tubes with helium pressure and (4) the feed of any fuel form of predominately uranium or thorium without requiring expensive and time-consuming solid fuel performance studies.

A fifth advantage is the high temperature at low vapor pressure of the fuel/coolant that enables practical use of fission heat for conversion of biomass to liquid fuels. Our sister BCLF Corporation (*Byproduct Cellulosic Liquid Fuel*) has found that charcoal from sources such as wood or straw can be combined with steam at 600 C and practical pressure to produce CO and H₂ that can then be transformed by Fischer-Tropsch to diesel, jet fuel, etc. Molten salt is an excellent heat transport medium because of its density and low vapor pressure. Molten salt enables a liquid-to-gas heat exchanger with thin walls for efficient heat transfer and without thick metal to contain high gas pressure as would be required for example by a helium coolant with a gas-to-gas heat exchanger. We consider this a major development for nuclear technology because our GEM*STAR reactor provides hot salt not only suitable for efficient electricity production but also cost-competitive production of 100 % renewable diesel/gasoline from byproduct biomass. Atmospheric CO₂ production arises almost equally from electricity and transportation fuels, and GEM*STAR can eliminate both coal/natural gas for electricity and petroleum for transportation.

Much higher accelerator performance with superconductivity

The LANSCE accelerator at Los Alamos national laboratory began operation 44 years ago with a gradient of 1 Mev/meter and requires ½ mile to reach 800 MeV. The Jefferson Lab is now

testing accelerator superconducting cavities with a gradient of 10 MeV per meter and ADNA Corporation is now considering target designs for proton energies of 350 MeV. For GEM*STAR the accelerator might be less than 50 meters long. The current loading at LANSCE is 1 mA, but the 50-meter long accelerator can accelerator 10 mA. The Nuclear Energy Office of the U. S. DOE and U. S. reactor technologists in general are completely unaware of these advances and the implications for nuclear energy production and waste burning. ADNA Corporation can't claim credit for this advancement of about 10 x 10 = 100 in accelerator technology, but we are the first to couple these advances into GEM*STAR's practical system for zero carbon electricity and 100 % renewable liquid transportation fuels from any nuclear fuel type.

Doubling the efficiency of radiofrequency power generation

Klystrons for conversion of buss-bar power to radiofrequency power were invented in 1937 and typically achieve about 45 % conversion efficiency. We are arranging a collaboration with Muons Inc. to assist them in the development of a new magnetron for radiofrequency power that promises a conversion efficiency of 80 %. The effect of this advance can be expressed several ways, but perhaps the best way is to say that it reduces the energy cost of neutrons by a factor of two. Magnetrons are much cheaper than klystrons and so there also is a major reduction in capital cost for the accelerator. The accelerator advances based on superconductivity and new magnetron technology propel subcritical reactor technology far ahead of the accelerator technology that was available to us in 1996 at Los Alamos.

Viability of ocean uranium makes breeder reactors and associated reprocessing unnecessary The amounts of uranium and thorium present in the earth's crust after the earth's formation was about the same, but uranium is soluble in water and thorium is not. Therefore 3/4^{ths} of the uranium has now dissolved and then been carried by rivers into the sea whereas all of the thorium remains in the earth's crust along with only ¹/4th of the uranium. The Japanese and Russians have both studied the extraction of uranium oxide from the Japanese current with cost estimates of three to five times that of uranium mining from the earth. If nuclear power can accommodate this higher price for uranium, the uranium supply is unlimited and not governed by mining economics.

GEM*STAR has the advantage over solid fuel systems that the UO₂ form obtained from the ocean need only be converted to UF₄ to become fuel. By contrast, critical solid fuel reactors require the conversion from UF₄ to UF₆, the UF₆ must undergo isotopic enrichment, and the UO₂ must be formed into fuel assemblies. When the costs for these extra processes for preparing uranium solid fuel are added to mining costs for the LWR or the FBR, the fuel cost is more than GEM*STAR's cost for UF₄ fuel from the ocean. Therefore *GEM*STAR will never run out of fuel from the ocean at the LWR's present fuel cost,* whereas the LWR mined fuel cost will rise as the more favorable deposits are emptied. A second point is that because there is an unlimited supply of natural uranium fuel for GEM*STAR, there is no need for breeder reactor technology and its required reprocessing with the associated proliferation problems. Lastly the ocean is accessible to virtually every nation, so one nation need never be dependent on any other for nuclear fuel.

Summary

The international consensus on reactor technology dominated for 45 years by fast breeder proponents has forced most of accelerator-driven technology into the backwater of Np and Am burning. Yet fast reactors are no more attractive for deployment now than they were in 1972 when the Clinch River Breeder Reactor (CRBR) at Oak Ridge was canceled. The Fast Flux Test Reactor (FFTF) at Hanford that was built by the U. S. DOE instead did not help the matter nor did the defunct Monju project in Japan. There is nothing magic about criticality as a necessary attribute to a successful nuclear power concept. GEM*STAR as described here with the elimination of criticality, enrichment, reprocessing, control rods, volatile containment by solid fuel, concern for minor actinides, low single cycle burnup, and multiple waste streams is the future of nuclear power.

Any doubt about that is made abundantly clear by Fig. 4 where the DOE's Fig. 1 is redisplayed with the red mark outs made possible by GEM*STAR. The green boxes show GEM*STAR requirements by comparison for both energy generation directly from natural uranium and from the bulk fluorination of the spent fuel accumulating from today's LWRs. A small arrow at the top points uranium ore to the GEM*STAR recycling subcritical reactor that enables unlimited energy without enrichment or reprocessing. A diagonal arrow from the bottom left refers to the fluorination of all LWR spent fuel without any chemical separations of fission products, actinides, or minor actinides to produce directly useful fuel for GEM*STAR. There is no need for a geologic repository for LWR spent fuel any time soon as all of it would be reburned in GEM*STAR over a period that will take probably a century. Reducing and deferring the ultimate waste stream will allow time for a permanent waste solution no large or more serious than our situation today. And it is worth noting that GEM*STAR provides a solution to LWR waste that will enable LWRs to continue during the 30 years that will be required to convert the world's LWR fleet to a subcritical GEM*STAR fleet.

In conclusion, ADNA Corporation's accelerators will drive out chemical separations that have always been a burden to nuclear power and provide a technological solution to proliferation arising from isotopic enrichment and reprocessing.

