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## Disposition of Nuclear Wastes Using Subcritical Accelerator-Driven Systems

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Since the early 1990s, Los Alamos National Laboratory (LANL) has led the development of accelerator-driven transmutation of waste (ATW) to provide an alternative technological option to the disposition of nuclear waste. Although the concept of accelerator-driven systems has been proposed before under different circumstances,<sup>1,2,3</sup> it has only become technically feasible in recent years due to the advent of high-current, high-power accelerators. We arrived at the current concept of ATW after assessing the US national need for an alternative solution to the disposition of nuclear waste, and critically evaluating the state of accelerator, nuclear and processing technologies.

Current US policy for disposition of high level nuclear waste (mostly spent fuel) is to store unprocessed spent fuel in a geologic repository. By 2015, there will be over 70 000 tonnes of spent fuel with 600 tonnes of transuranic elements, of which over 500 tonnes will be plutonium. Studies have shown<sup>4,5</sup> that the long-term radiological risk associated with the repository is from the long-lived transuranic elements (TRUs) 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. While not identified as an issue, it is clear that the repository, designed for 70 000 tonnes of commercial spent fuel and high-level waste from defence-related activities, will be fully occupied by the spent fuel produced through 2015. If nuclear energy remains a viable option for electricity generation in the future, there will be a need for a second high-level waste repository.

To make an impact on the disposition of nuclear waste, an ATW system should accomplish the following:

- **Destroy over 99.9% of the TRUs.** Fissioning the TRUs eliminates concerns related to their release to the ground water and the environment as well as their possible diversion and use for weapons. Their elimination also greatly reduces long-term heat loading in the repository.
- **Destroy over 99.9% of the technetium (Tc) and iodine (I).** Two of the long-term radiotoxic hazards can be eliminated by transmuting these elements.
- **Separate Sr and Cs (short half-life isotopes).** Sr-90 and Cs-137 dominate the short-term heat loading in the repository, and although these

isotopes are not suited for transmutation, they will be separated from the remainder of the waste and placed in optimised storage systems.

- **Separate uranium.** Uranium is separated from the rest of the spent fuel, and either stored or possibly re-enriched for use in conventional reactors.
- **Produce electricity.** The ATW resembles a nuclear reactor in that it releases energy during actinide fission that can be converted into electricity. A small fraction (10–15%) of this electricity will be used to power the accelerator and the rest can be distributed.

Advances in the following three fields contribute to establishing the feasibility of such an ATW system:

- The Accelerator Production of Tritium (APT) programme has begun the engineering design and testing of a high-power accelerator,<sup>6</sup> one more powerful than is needed for ATW.
- The R&D during and after the Integral Fast Reactor (IFR) programme at Argonne National Laboratory,<sup>7</sup> the plutonium pyroprocessing experience at Los Alamos,<sup>8</sup> and other efforts worldwide to develop pyroprocesses<sup>9</sup> have given us the capability to treat spent nuclear fuels and ATW waste in a much simplified and proliferation-resistant manner, and achieve the necessary degree of TRU removal from the waste stream destined for final disposition.
- The recently released Russian lead–bismuth (LBE) nuclear coolant technology<sup>10,11</sup> has allowed the conceptualisation of integral spallation (decay caused by high energy particle bombardment) target and blanket systems with excellent transmutation efficiency.

In the ATW concept, spent fuel would be shipped to an ATW site where the plutonium, other transuranics and selected long-lived fission products would be destroyed by fission or transmutation in the facility. This approach contrasts with the present-day reprocessing practices in Europe and Japan, in which high purity plutonium is produced and used in the fabrication of fresh mixed-oxide (MOX) fuel that is shipped off-site for use in LWRs. Instead of reprocessing, the ATW approach can be characterised 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 realised by enhancing the capacity of a repository and elimination of the need for an additional repository, or reducing the long-term performance uncertainties, could offset to some extent the cost of developing and implementing the technology.

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

#### **ATW System Description**

An ATW facility consists of three major elements:

- a high-power proton linear accelerator;

- a pyrochemical spent fuel treatment/waste cleanup system;
- a liquid lead–bismuth cooled transmuter that produces and utilises an intense source-driven neutron flux for transmutation in a heterogeneous (solid fuel) core (see Figure 1).

The concept is the result of many years of development at Los Alamos National Laboratories (LANL)<sup>12</sup> and other major international research centres.<sup>13</sup>

The high-power accelerator for ATW would be based on the APT Project accelerator (1000 MeV, 100 mA, 100 MW 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 transmuter, where transuranics and select fission products would be fissioned or transmuted.

In the spent fuel treatment system (shown schematically in Figure 2), uranium and a majority of the fission products are separated from the transuranics and the targeted long-lived fission products by pyrochemical processes. One essential requirement is the separation of enough uranium (99%) so that no significant new TRU production occurs during irradiation. Fission product partitioning comes naturally from the uranium removal and transuranic recycle process. In the clean-up processes, the fission products in the irradiated waste are partitioned into three groups: active metals, noble metals and rare earths. This remnant waste is prepared for permanent storage as: oxides in engineered containers for the active metals (including Cs and Sr); oxides for the rare earths; and metal ingots and/or oxides for the noble metals including zirconium.

These separation processes also allow for a high degree of TRU removal in the waste streams destined for final disposal. The ability to tailor the waste forms to the repository environment by natural segregation in the ATW waste treatment processes offers advantages over untreated spent fuels. An average of 50 kg of fission products per tonne of spent fuel, contaminated with less than 1000 ppm of transuranics, are discharged as waste after transmutation, including the fission products originally present in the spent fuel. Most of the radioactivity in the discharged material would decay before three hundred years, leaving only weak residual activity of negligible environmental impact.

The waste transmuter consists of a heavy metal target, liquid lead–bismuth eutectic, producing the high intensity neutron source and the surrounding subcritical blanket containing the transmutation assemblies (see Figure 3). Since significant neutron multiplication and heat production occurs from the fissioning of the actinides contained in the surrounding transmutation assemblies, adequate means for heat removal must also be present. This equipment is analogous to that used for critical reactors of a 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 transmuter. 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.<sup>10</sup>

The subcritical liquid LBE systems presently being studied 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 (end of an overall campaign to transmute waste) inventories can be achieved by burn-down strategies involving gradual thermalisation of the spectrum to exploit the large capture cross sections of resonances.

Subcriticality does not make ATW by definition safer than critical reactors. Rather, subcriticality facilitates tasks that would be exceedingly difficult or

inefficient in critical systems. Subcritical systems do not rely on delayed neutrons for control and power change; they are driven only by the externally generated neutron source (i.e. by the ion beam coming from the accelerator). Control rods and reactivity feedback have low importance; these systems are neutronic, but not thermally, decoupled from their neutron source. Therefore, subcriticality allows the ATW system to work with any composition of fuel, or waste, and relaxes the required degree of separation in the waste treatment steps. This makes possible, in principle, the destruction of any isotopes of TRUs, or fission products, or a mixture of both, with little concern for their neutronic behaviour. Fertile materials are not needed to compensate for the neutronic uncertainties or undesirable reactivity responses of the fuel, and extended burn-up is achieved by increasing the power of the accelerator drive to compensate for the reactivity decrease. In practice, the degree of subcriticality could range between  $k_{\text{eff}} = 0.98$  and  $k_{\text{eff}} = 0.93$ .

Because of its subcritical mode of operation, ATW will be ideally suited as incinerator of material that is not well characterised, that transmutes very poorly or not at all in reactors, that has potentially unstable and hazardous reactivity responses, and that should not for whatever reason be isolated and placed in reactors. This includes higher actinides such as neptunium (the worst contributor to oxidising repository long-term performance uncertainties), americium, 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.

### **High-Power Linear Accelerator**

The 1000MeV 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 optimisation. 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 radio frequency quadrupole (RFQ) based on a well-defined beam emerging from a reliable injector. A suitable injector has been working for months at LANL with currents (110 mA) 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 (Chalk River Injector Test Stand) RFQ has been accelerating a proton beam with good transmission and beam parameters exceeding the ATW requirements.

Following the RFQ is a coupled cavity drift tube linac (CCDTL) that will be demonstrated in the APT Low Energy Demonstration Accelerator (LEDA) programme. This structure has revolutionised the ability to transport high quality beams from an 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 programme. 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 355 m, with a radio frequency 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 ATW demonstration unit is projected to be a 500–1000 MWt system.

### **Fuel Cycle Technology**

Spent fuel treatment technology is derived from the pyrochemical processes developed for the IFR programme at Argonne<sup>7</sup> and plutonium production at Los Alamos.<sup>8</sup> Pyrochemical processes were chosen over the conventional aqueous processes because: they are proliferation resistant (group separations are used instead of single species separations); they allow the processing media, molten salts and liquid metals, to be recycled multiple times thus reducing secondary waste; and they allow for short turnaround times for waste treatment (radiolysis and decay heat are not significant issues).<sup>14</sup>

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) programme.<sup>15</sup> The following sections provide a brief overview of process chemistry and fuel technology for the ATW system.

#### *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 to achieve the ATW requirements. 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, Zircaloy. 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 subsequent chemical processes. If it is not possible to achieve a high degree of separation between the spent fuel and clad material then the cladding will follow the fuel through the next process and be separated from the uranium in the electrorefining step. This 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 USA.

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 1025K. The process could also be completed using a lithium metal reductant in a lithium chloride molten salt. Some fission product partitioning takes place during the oxide reduction process. Fission gases are released from the matrix of the oxide fuel and recovered by the methods described for the decladding system. Active metals, such as caesium, strontium and barium, partition to the molten salt and are periodically removed from the salt during the direct oxide reduction salt recycle process. These metals are placed in engineered storage containers and sent to the repository. Iodine also partitions to the molten salt and it is recovered from the salt during the salt recycle process. It is collected by cryodistillation methods, fabricated into targets, and transmuted in the ATW system. The heavy metal produced in the oxide reduction process is sent to the electrorefining system.

Electrorefining is used to partition the uranium, transuranics and fission products. The system uses electrochemical methods to electrotransport the uranium from the anode to a solid cathode. After the transfer process is complete, the uranium-bearing cathode is removed from the system, the uranium is oxidised, and either sent to storage or recycled. An eutectic mixture of NaCl–KCl molten salt, at approximately 1000K, is used as the transport medium. Noble metal fission products (i.e. Zr, Mo, Tc, Ru, etc.) remain at the anode in the cell. The anode is sent to the technetium recovery process before being discharged to the repository. TRUs and rare earth fission products remain in the molten salt, which is treated by the electrowinning process.

Electrowinning is an electrochemical process used to electrodeposit the TRUs from the NaCl–KCl molten salt. The TRUs, 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 TRUs. The TRUs are subsequently transferred to the vacuum casting fuel fabrication system. 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 this extraction process, the rare earths are collected, oxidised, packaged, and sent to the repository.

Technetium is removed from the electrorefining anode heels and sent to the ATW system for transmutation. It 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 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 by standard techniques. It is transferred to an electrorefining system where the TRUs are partitioned from the active metal, noble metal and rare earth fission products. These TRUs are sent to the vacuum casting system where fresh ATW fuel is fabricated. Fission product gases released during the electrorefining processes are

collected by the aforementioned methods. Noble metals are removed from the electrorefining cell, transferred to the Tc recovery process, and ultimately sent to the repository. Periodically the TRU content in the transport molten salt is decreased by using the aforementioned electrowinning process. The rare earth fission products are periodically removed from the electrorefining salt by reductive extraction techniques and they are collected, oxidised, packaged, and sent to the repository.

Although electrorefining is used in both the front-end uranium removal and back-end TRU recycle processes, differences exist between the two systems. The front-end system is larger than the back-end system, about 400 kg of uranium is electrotransported in the process as compared to about 8–10 kg of TRU in the back-end process. The process parameters, cell operating current and process time, are quite different. In addition, the back-end system must be designed to process Pu-rich mixtures of TRU instead of low enriched uranium. Similar design and scaling considerations are required for the electrowinning systems.

#### *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 (Zircaloy) as the inert fuel matrix, and the desire to make the processing as simple and waste-free as possible drives the preliminary selection of fuel form to a zirconium-based metal matrix with an initial TRU loading of about 10–15 at%. The fuel is a high melting alloy (>1900K) 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 ALMRs 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.

#### **Lead–Bismuth Eutectic**

One significant improvement of the current ATW system concept over the previous ones is the choice of using liquid lead–bismuth eutectic (LBE) as nuclear coolant and spallation neutron target, in an integral one-fluid system. This is made possible by the unique physico-chemical and nuclear properties of LBE, and the feasibility is further enhanced by the recently released Russian LBE nuclear coolant technology successfully deployed in submarine propulsion reactors. Russian designs for small (100 MWt) and large (1000 MWt) 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.

#### *LBE Properties*

The basic physical properties of LBE (44.5wt% Pb, 55.5wt% Bi) are as follows:

- low melting temperature (123.5°C);

- high boiling temperature (1670°C);
- density ( $\text{kg/m}^3$ ) = 10730 - 1.22t (t in °C);
- low vapour pressures.<sup>11</sup>

In addition, Pb and Bi are chemically quite inert towards reaction with water. For instance, LBE is less likely to be oxidised than Fe, Cr and Ni, the primary elements in many common construction materials.

The low melting temperature, high boiling temperature and very low vapour pressure of LBE allow for a wide operating temperature range, eliminating coolant boiling and enhancing circuit safety. The high density of LBE combined with a 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 if incidental leaks or contact occurs. The sealed vessels and circuits readily prevent airborne lead contamination from exceeding established industrial standards (0.03 mg/m<sup>3</sup> in the USA). LBE spills are self-contained by rapid solidification. The corrosiveness of LBE can be mitigated by the Russian coolant technology, which will be explained later.

The nuclear properties of LBE make it well suited as both spallation target and nuclear coolant. LBE target can produce over 25 neutrons per 1000 MeV incident proton. Liquid metal spallation targets do not suffer the radiation damage and coolant induced neutron loss and moderation, as do solid targets. Because of the extremely low capture cross sections, LBE is transparent to neutrons, allowing much relaxed core design. The high-Z LBE offers the hardest neutron spectrum possible in a nuclear system, and is most advantageous for efficient transmutation. It is also provides gamma-shielding for added safety. The use of LBE results in a negative overall coolant void and temperature reactivity coefficient.

#### *LBE Coolant Applications*

The potential of using liquid lead–bismuth eutectic for nuclear coolant was recognised very early on in the development of nuclear reactors, and led to intensive research programmes in the USA and Canada from the 1940s to 1960s. However, many basic components in typical construction materials, especially nickel in steels, have finite solubility in LBE, and the resulting corrosion and mass transfer problems were severe and not solved satisfactorily. All the major LBE coolant programmes in the USA were terminated by the mid 1960s.

A brief summary of the main findings of those early LBE research programs on corrosion is listed here<sup>16</sup> (without the application of the now-known corrosion prevention method):

- Carbon steels are the most corrosion resistant, low alloy steels (<2.25% Cr) are moderately corrosion resistant, and high Cr steels are subject to gross attack.
- Zr, Ti and Mg additives are used as inhibitors and yield limited but inconsistent success.
- One time, passive coating of the structural materials with nitride or oxide protective films failed to consistently prevent corrosion.

Recent information reveals that intensive R&D programmes were carried out in the former Soviet Union to solve the corrosion and coolant contamination problems associated with the application of LBE technology to nuclear submarine reactors. The programmes started in the early 1950s, and intensified greatly in the 1960s to 1970s, as early LBE nuclear installations showed the similar corrosion and contamination problems encountered by the Western researchers, which led to reduced plant lifetime and accidents. The product of those programmes were LBE cooled propulsion reactors (150 MWt) installed on eight Alfa-class submarines, with a total of over 70 reactor-years experience, in addition to many test facilities, test rigs, and various LBE or liquid lead cooled civilian reactor designs.

The key to solving the LBE corrosion and contamination problem is the recognition that LBE differs from other liquid metal coolants (i.e. Na or Li) in that it is chemically more inert than Fe, Cr and other elements in many construction materials, and warrants a different corrosion control method.

The technique selected, developed and successfully deployed at the Institute of Physics and Power Engineering (IPPE) at Obninsk, Russia, is to actively control the thermodynamic activity of oxygen in LBE coolant systems to maintain self-healing protective oxide films on structural materials (i.e. in situ surface passivation) and continuously clean the coolant of lead oxide contaminant. Very simply put, if there is too little oxygen in the system, there will not be enough protective oxide films, and the structural materials are corroded; if there is too much oxygen, the excess oxygen after forming protective oxide films will react with lead (bismuth oxide is less stable) to form lead oxide, which will eventually precipitate and contaminate the coolant. There is a range of oxygen level in LBE (approximately below 1 ppm and above 0.1 ppb by weight, depending on the system temperatures) when neither of the above will occur. The operational procedure for proper control has been mapped out and tested at IPPE.

Many studies on material compatibility, irradiation properties and radioactive safety of LBE systems have been carried out at IPPE and affiliated institutions. Many kinds of structural materials were irradiated in thermal and fast fission spectra. Special steels were developed and continuously improved. There has also been direct experience of dealing with the radioactive hazards of using LBE coolant, such as polonium formation and release.

Polonium is formed in LBE when bismuth is irradiated. However, polonium tends to bond with lead, forming lead polonide, which greatly reduces its release rate from LBE as compared to that from pure bismuth (a reduction of two to four orders of magnitude). We have evaluated several ATW transmuter coolant spill or containment breach accident scenarios, and found that the short term polonium contamination level can rise above regulatory limits, but would quickly subside to safe levels.<sup>17</sup> Russian reactor experience indicates that such release, even in rather massive spills, can be contained if proper radiological measures are deployed and the spills will not pose severe health hazards to the operators. It also appears that LBE coolant itself is a

rather safe place to store polonium. Cleanup methods are being evaluated and developed.<sup>18</sup>

Preliminary study suggests that spallation products unique to spallation targets or ATW transmuters will not lead to significant changes in the existing LBE coolant technology. However, transmutation products generated in structural materials could lead to deleterious radiation damage. Little experience is available regarding this phenomenon and more experimental study is urgently needed.

With funding support from the International Science and Technology Center and co-ordinated by scientists from the USA and the EU, a team of specialists from IPPE and Gidropress (Poldolsk, Russia) is designing and will fabricate a 1 MW LBE spallation neutron target, to be delivered and tested at LANSCE after 2000. This will be a first integral test of thermohydraulics and neutronics of LBE spallation target and nuclear coolant. Separate material irradiation tests are being planned to establish a database for radiation properties in proton and high-energy spallation neutron fluxes.

To validate the LBE coolant technology, and to adapt and extend its applications to ATW systems, we have also started a development programme at Los Alamos. In collaboration with IPPE scientists, we have built and operated our first forced convection LBE test loop and a second one will be soon constructed.<sup>19</sup> The corrosion control technique will be applied and the effects of protective oxide films on structural materials will be studied.

#### *LBE-Cooled ATW Transmuter*

The integration of nuclear coolant and spallation target in the current ATW concept drastically improved the subcritical transmuter 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 can be roughly described as follows. The actinide-containing region is 2 m high by 2 m in diameter. A 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 heat exchange 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, spallation products and other radioactive isotopes. The secondary coolant is non-radioactive LBE. A minimum 1m thick LBE reflector surrounds the waste assemblies on all sides. This reflector helps minimise required actinide loading, flattens the power density across the

fissioning region, shields the vessel walls from fast neutrons, and provides thermal inertia.

### **Interface with Geologic Repository**

The adoption of an ATW option at the close of the fuel cycle, using the existing baseline repository site and design, presents a radically different waste disposition scenario. The new waste forms are designed at the process stage to be chemically compatible with the repository environment and extremely durable and leach resistant. Thus, waste form performance inherently drives the ATW concept, instead of traditional attempts to modify an existing rock mass to accommodate a fixed waste type that was not designed expressly for disposal.

The isotopes that were problematic in the baseline repository concept can be captured in the ATW process and returned to the ATW reactor for transmutation. The fission products that make up the ATW waste stream consist of active metals, rare earth oxides containing a trace of actinides, and noble metal fission products in metallic and oxide form. The rare earths can be incorporated into mineral-like ceramics, for which we have performance data on a geologic timescale. The noble metals could be used, together with the zirconium cladding, to contain the ceramic, further enhancing its performance. The net result is a compact and extremely durable and leach resistant package that is well tailored to the geo-chemical conditions at the proposed repository.

The performance benefits of ATW include a reduced performance timescale and reduced performance uncertainty since the source term for a release is mostly eliminated by the waste form. The long-lived transportable nuclides have been transmuted, or in the case of Np-237, its parents have been eliminated from the waste stream. Because uranium is removed for recycle, its decay products are also eliminated from the waste inventory.

The different heat loading from ATW wastes may require some changes in repository layout and transportation schemes. Because the ATW wastes contain most of the initial heat sources, but not the mass of the original spent nuclear fuel, heat generation per unit mass is much higher. This may require different loading geometry during the emplacement period, unless Cs and Sr are removed. However, with the heat generation falling off with a half-life of 30 years, and a reasonably long ATW campaign, this may not be much of a problem.

The concern regarding a plutonium mine is non-existent because only small traces of actinides remain as part of the ceramic waste form and their concentration is well below any reasonable economic and proliferation limit. In addition, its value is diminished by virtue of poor isotopic distribution.

Since repository costs are driven by performance uncertainty, the reduction of the uncertainty due to the development of better waste forms reduces the overall costs associated with engineered enhancements to the repository block. Because a shorter performance timescale is possible due to the mostly short-lived inventory, there is less uncertainty in predicting far into the future. Less

uncertainty and shorter timescales could translate into cost savings during licensing and potentially less public concerns.

The key point about an ATW repository is that we can consider scenarios involving no migration out of the controlled area, as opposed to the baseline case of expected releases over a geologically short timescale followed by unrestricted releases.

### **Implementation Scenarios**

The dedicated nature of ATW and the flexibility afforded by subcriticality combine to give us many possibilities for implementing ATW for effective waste destruction, and for assisting in establishing a sustainable nuclear cycle with desirable attributes in terms of resource utilisation, waste production and proliferation resistance. We will examine two possible implementation scenarios. Many others can be developed.<sup>20</sup>

The first scenario assumes that the nuclear power option is discontinued after the current generation of LWRs. The scenario therefore deals with the destruction of the transuranic and long-lived fission product component of the accumulated spent fuel from the current generation of LWRs in the USA. The outcome of the scenario is the production of limited amounts of waste, optimally tailored for storage in the selected repository geology, containing only trace amounts of transuranics and long-lived fission products. There is also a significant amount of electricity production.

The second scenario assumes a reasonable scale-up of nuclear power in the USA, to double its current installed capacity over the course of 100 years. The outcome of this scenario is:

- the destruction of the spent fuel backlog;
- the reduction of the plutonium present in the fuel cycle to a level 100 times less than presently available, and in such form as to preclude its use for military applications;
- higher utilisation of the energy content of natural resources;
- the avoidance of production of weapons-usable materials;
- the phasing-out of enrichment technology.

### *Cost Considerations for ATW Scenarios*

In general, the following points are known about the cost of an ATW system:

- The cost of the particle accelerator, based on APT studies, will not dominate the economics of ATW.
- The pyrochemical waste treatment processes are acknowledged to be less expensive than traditional aqueous processes,<sup>15</sup> and could be made simpler under the relaxed separation requirements imposed by ATW.
- The cost of subcritical ATW transmuters based upon lead–bismuth coolant technology should be comparable or lower than the cost of critical sodium-cooled reactors, as evidenced by the Russian LBE-cooled reactor designs and experience.
- 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.

*Spent Fuel Backlog Transmutation in 65 Years*

By 2015, there will be 70 000 tonnes of US spent fuel containing about 600 tonnes of TRUs 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.

In this scenario, twenty-one ATW systems are brought on line to accomplish the objective over a 65 year period. Each ATW system consists of an accelerator (1000 MeV, variable current 20–40 mA), a subcritical 2000 MWt LBE transmuter, and a pyrochemical plant (50 t/yr throughput per transmuter). 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 transmuters are as follows:

- Fission power: 2000 MWt;
- Thermal to electric efficiency: 40%;
- Power to grid: 700 MWe;
- Recirculated power: 100 MWe (67 MWe accelerator, 33 MWe plant operations);
- Operational TRU inventory: 3000+1000 kg;
- Conversion ratio: 0.0;
- Process losses: 1/1000 original TRU;
- TRU burn rate: Up to 650 kg/yr;
- TRU burn efficiency (rate/inventory): 16.25%.

Of particular relevance is the fact that each 2000 MWt ATW transmuter can destroy up to 650 kg of TRUs per year, and that 700 MWe would be available for distribution to the grid from each transmuter, 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 transmuters and accelerators reach the end of their operational life, they are decommissioned, and the fuel from the decommissioned transmuter is sent to feed other operational transmuters. Eventually, the last system in operation at each site will receive all the remnant waste and destroy it down to less than 1 tonne over a protracted (5 years) inventory burn-down period (see Figures 4 and 5).

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: ATW does not produce higher actinides from a fertile material such as U-238; and 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 is practical.

*ATW for a Closed Thorium and Uranium Fuel Cycle*

A second scenario couples separation and transmutation activities with the back-end of an alternate fuel cycle for LWRs.<sup>21</sup> This system combines denatured thorium–uranium-oxide fuelled advanced LWRs (DTU–LWR) with an ATW to process spent fuel and to separate thorium and uranium for recycle to the LWRs. This combined system addresses two major issues facing the nuclear industry: sustainability and security. The concept involves a fuel cycle that includes abundant thorium to reduce depletion of natural uranium,<sup>22</sup> which is an exhaustible resource given a projected growth of nuclear power. Non-nuclear energy resources are also not unlimited, and a worldwide realisation of the hidden costs of other energy sources or a global consensus to reduce greatly the emission of greenhouse gases and other pollutants could result in a resumption of rapid growth of nuclear power similar to that observed in the 1970s in the USA.<sup>23</sup>

This coupled concept of DTU–LWRs and ATWs produces a view of the future with greatly reduced depletion of uranium resources and volume of high level waste, and with a reduction in proliferation risk, in terms of both quality and quantity of plutonium stored external to the highly radioactive and controlled cores of reactors. The system produces less Pu because the fuel contains less uranium, and the Pu is of lower quality for proliferation because recycling of intermediate isotopes of uranium produces more Pu-238 and other non-fissile isotopes.

DTU–LWRs supported by ATWs as a backend are deployed for a future growth in nuclear energy according to the following scenario: existing and near-future production of nuclear energy in the USA was modelled in accordance with a “high nuclear” projection from the Energy Information Administration (EIA) of the US Department of Energy.<sup>24</sup> That case is characterised by extension of licences for all existing US plants by 10 years or by early retirements for some LWRs combined with 20-year licence extensions for others.

Thus, as is shown in Figure 6, during the first 20 years (2000–2020) of the scenario of the present study, nuclear energy generation in the USA first rises from today’s 70 GWe because of increasing performance (EIA projects that capacity factors will increase from 70% in 1997 to 85% in 2020). Power from existing reactors then declines to about 68 GWe in 2020, and all existing reactors are shut down by 2045. Beginning in 2020, sufficient ATWs are deployed to transmute all TRUs from existing “conventional” LWRs (CLWRs).

To supply a growth in nuclear power to 200 GWe by 2100, the DTU–LWR/ATW deployment scenario includes new LWRs plus ATWs to burn the actinides produced by these LWRs. A DTU–LWR that is fuelled with 25% UO<sub>2</sub> and 75% ThO<sub>2</sub> (volume percent) would produce about 160 kg of TRUs (mostly Pu) per year per GWe, compared to about 310 kg for a CLWR — this production depends strongly on reactor design (moderator to fuel ratio, refuelling schedule, fuel burnup, etc.), and may be optimised in future studies. To transmute this LWR-produced feed, an ATW will fission about 1200 kg of actinides per year per GWe, thus the support ratio is 7 GWe DTU–LWRs

per 1.05 GWe ATW. Therefore, in 2100, 174 GWe from DTU-LWRs and 26 GWe from ATWs would provide the 200 GWe demand.

The DTU-LWR/ATW concept addresses proliferation concerns first by reducing the quantity of Pu from the fuel cycle, which is illustrated in Figure 7, where the cumulative inventories of Pu in both DTU-LWR and CLWR spent fuel are compared. If the power growth scenario represented in Figure 6 is supplied by CLWRs alone, without actinide burning or recycle, Pu will accumulate like the upper curve in Figure 7, with a total inventory exceeding 3500 tonnes by 2100. In contrast, with the DTU-LWR/ATW deployment scenario, the asymptotic Pu in inventories of spent fuel is equal to the total annual production from all DTU-LWRs. All other spent fuel will be removed for processing and transmutation. The total inventory in spent fuel pools would be about 70 tonnes in 2100, which is less than 2% of the Pu that would accumulate from the once-through CLWR fuel cycle. All of that 70 tonnes of Pu would be contained and controlled in highly radioactive, newly discharged fuel.

The DTU-LWR/ATW concept also reduces proliferation concerns because of the quality of plutonium in the spent fuel. Attributes of spent fuel Pu that affect its attractiveness or usefulness for proliferation include critical mass, spontaneous neutron emission, and thermal energy production. The spent fuel Pu created during the scenario examined in the present study was compared with the attributes of other grades of Pu: weapons-grade Pu, reactor-grade Pu, and mixed oxide-grade Pu. Bare critical mass (BCM), and spontaneous neutron emission and thermal generation per BCM, were computed for these three grades of Pu, for the DTU-LWR Pu, and for the Pu from seed-fuel elements of the developing Radkowsky Thorium Fuel (RTF).<sup>25</sup>

Because it contains more than 7% Pu-238 and large fractions of Pu-240 and Pu-242, the plutonium from the DTU-LWR/ATW fuel cycle produces 25 times as much heat and 50 times the spontaneous neutrons as a BCM of weapons-grade Pu, and also exceeds the thermal generation and spontaneous neutron emission from all other grades of spent fuel Pu. Because of these attributes, the Pu from the DTU-LWR/ATW scenario would be considered proliferation resistant. Any attempt to build a weapon from the plutonium extracted from spent fuel from DTU-LWRs would require complex engineering solutions to prevent degradation of weapon performance.

### **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 in their commercial reactor waste. ATW technology, initially proposed in the USA, has received wide and rapidly increasing attention abroad, especially in Europe and East Asia, with major programmes now being planned, organised and funded. Substantial convergence presently exists on the technology choices among the programmes, opening the possibility of a strong and effective international collaboration on the phased development of 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 transmuter (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, and could well constitute the backbone of future nuclear systems (both critical and subcritical).

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.

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Figure 1. An ATW system consists of three major functional blocks.

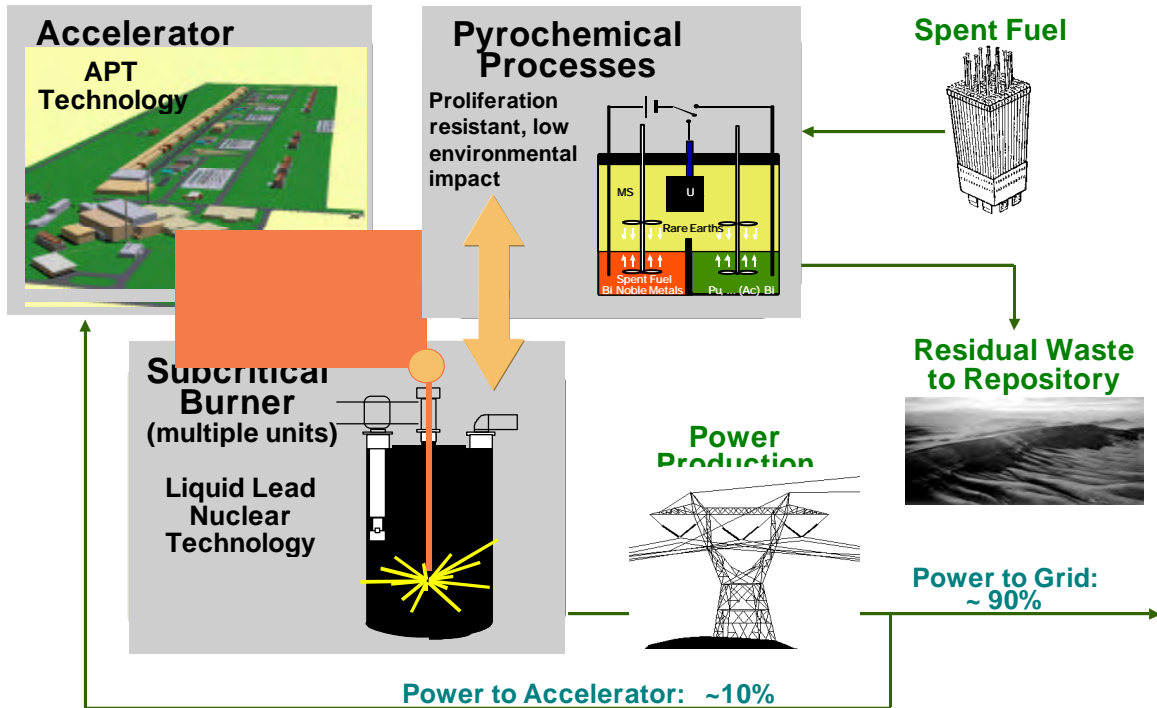


Figure 2. Schematic ATW waste treatment flowsheet based on pyrochemical processes.

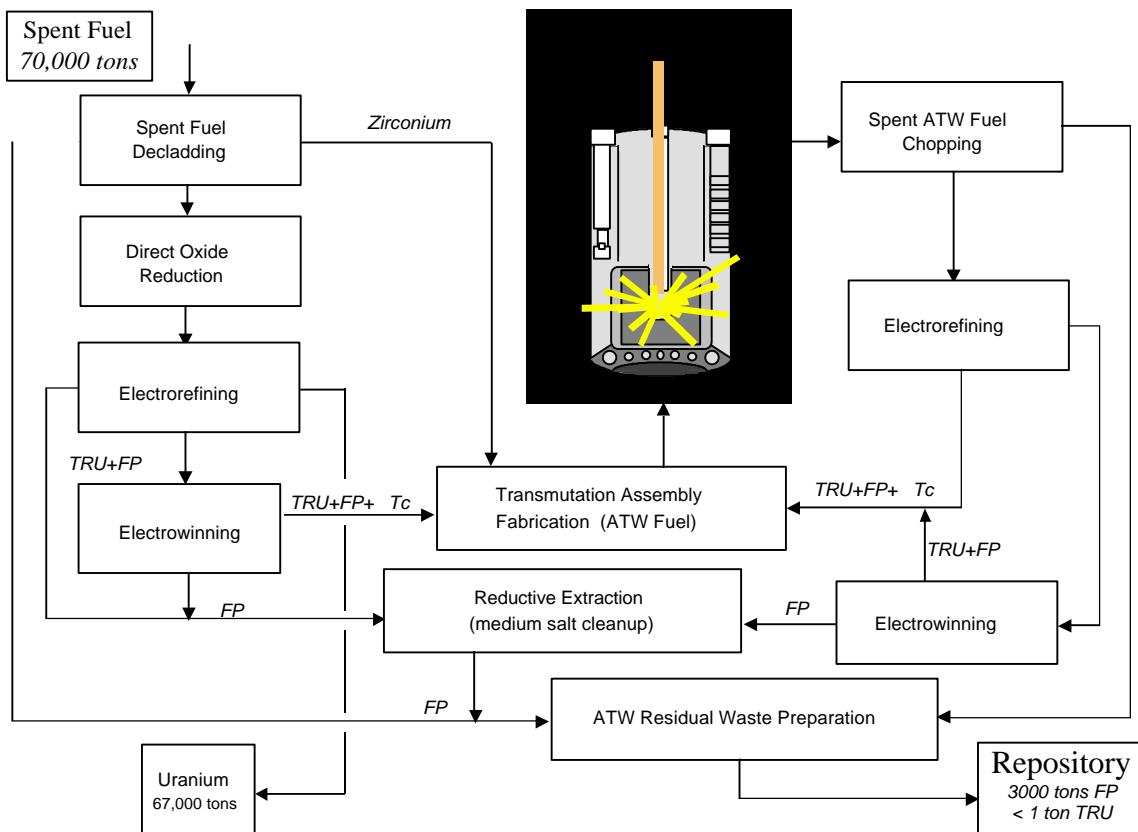


Figure 3. A schematic of an ATW transmuter based on existing reactor designs

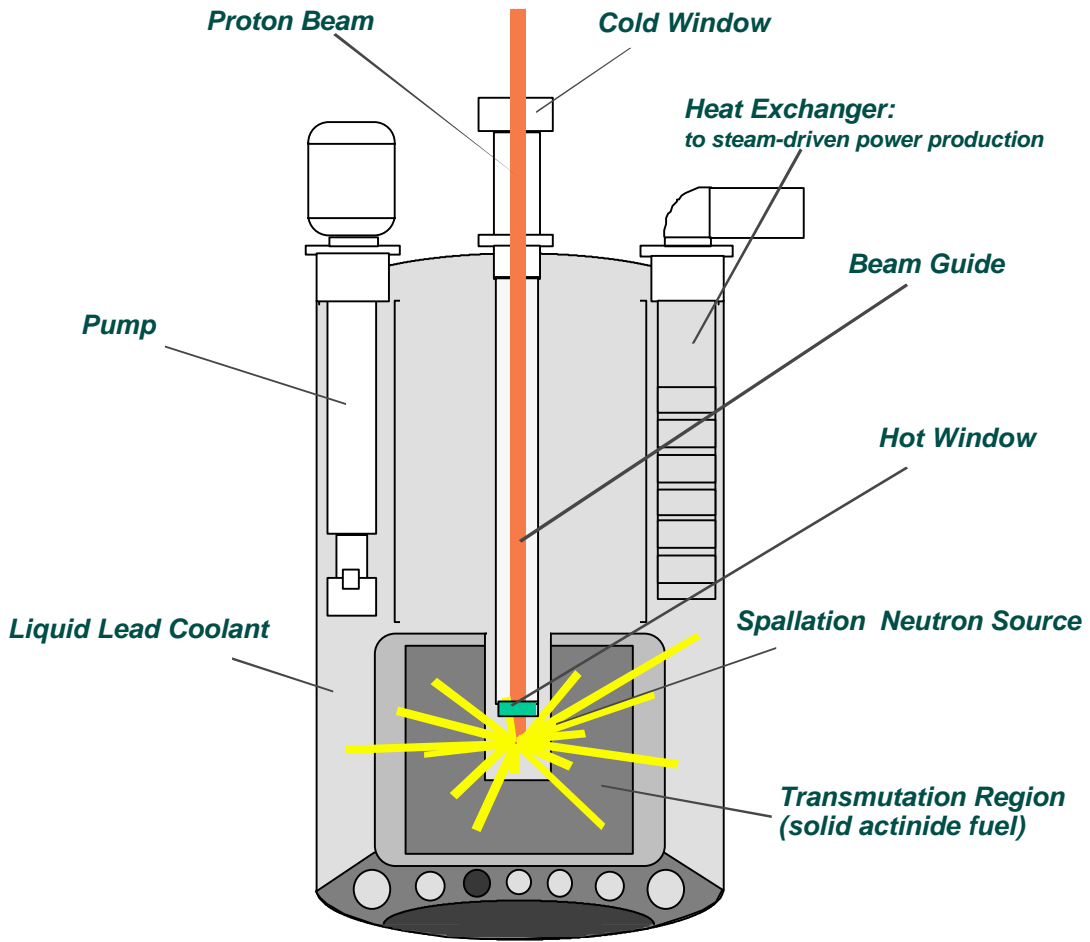
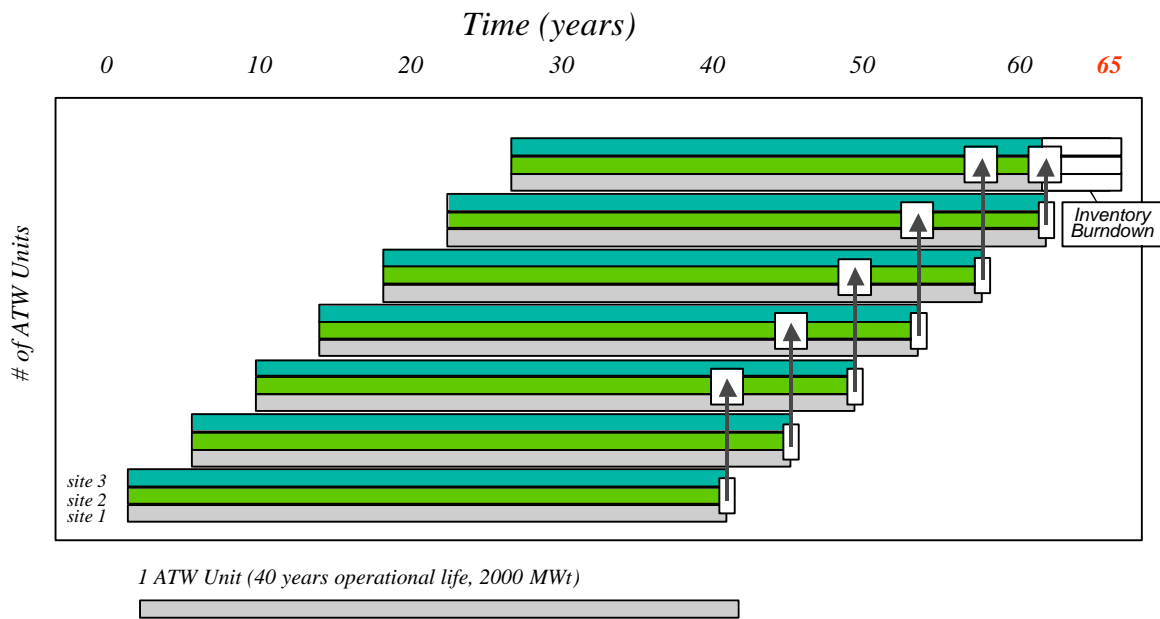


Figure 4. An implementation scenario for a 65-year burndown of spent fuel backlog.



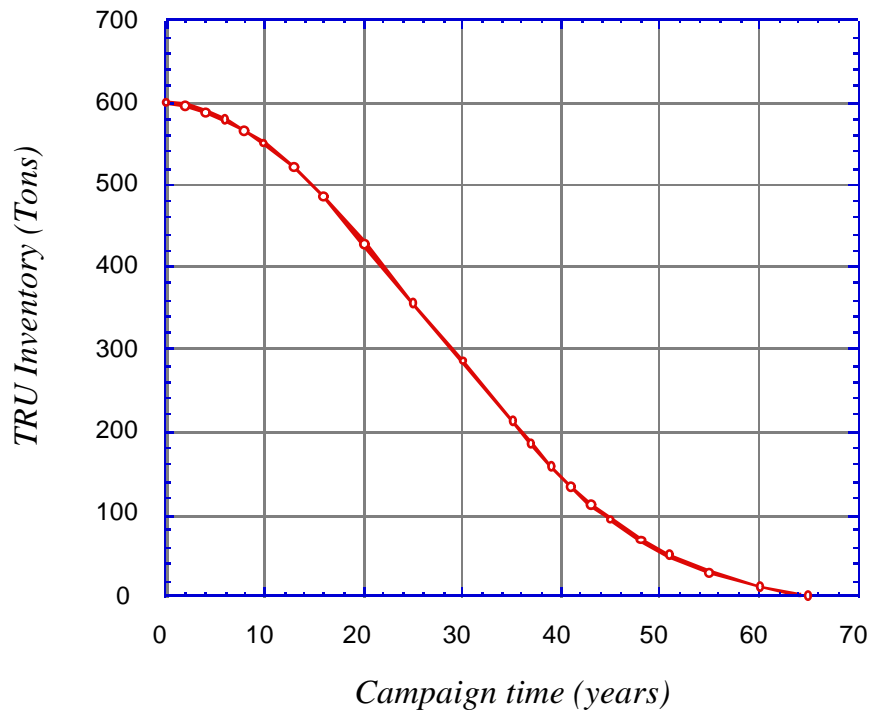


Figure 5. ATW can reduce the TRU inventory quickly.

Figure 6. A US nuclear power scenario through 2100.

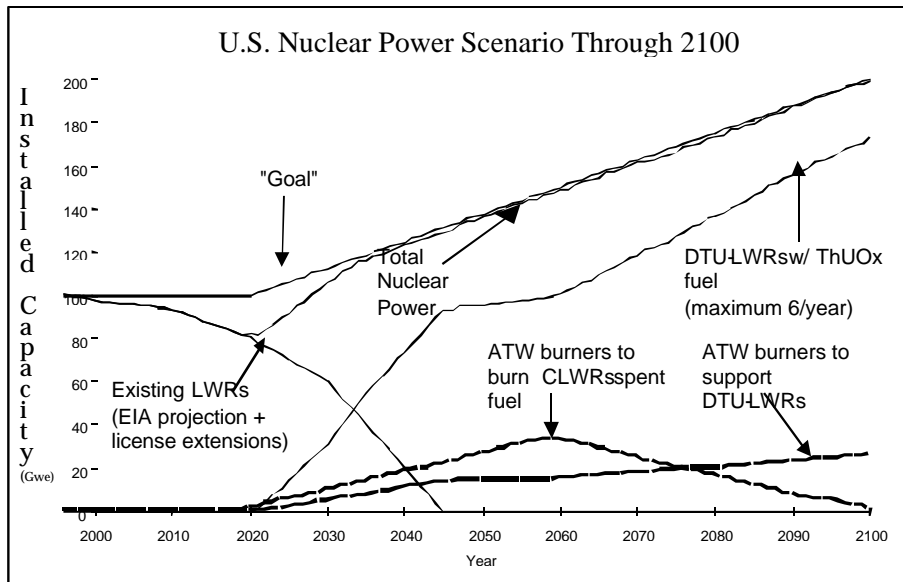


Figure 7. Pu in spent-fuel inventories resulting from a DTU-LWR/ATW deployment scenario with 200 GWe in the year 2100. The upper solid line represents Pu inventory that would accumulate if once-through UO<sub>2</sub>-fueled CLWRs provide all the growth in nuclear-generated electricity

