

TITLE: ACCELERATOR-BASED SYSTEMS FOR PLUTONIUM DESTRUCTION AND NUCLEAR WASTE TRANSMUTATION

AUTHOR(S): Edward Arthur

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ACCELERATOR-BASED SYSTEMS FOR PLUTONIUM DESTRUCTION AND NUCLEAR WASTE TRANSMUTATION

E. D. Arthur

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ABSTRACT

Accelerator-based systems are described that can eliminate long-lived nuclear materials. The impact of these systems on global issues relating to plutonium minimization and nuclear waste disposal can be significant. An overview of the components that comprise these systems is given, along with discussion of technology development status and needs. A technology development plan is presented with emphasis on first steps that would demonstrate technical performance.

INTRODUCTION

The accelerator-based systems discussed in this paper are a new approach to elimination of long-lived radionuclides (actinides and long-lived fission products). These long-lived species are directly linked to nuclear material and nuclear waste related problems of worldwide interest today -- surplus weapons plutonium from retired weapons, larger inventories of plutonium in reactor spent fuel, and the general problem of the long-term disposal of high-level waste (HLW) from defense and commercial sectors.

Accelerator-based systems* under investigation at Los Alamos National Laboratory consist of an accelerator-driven, subcritical nuclear system which uses a fluid carrier for the nuclear fuel. This system and its impact on a number of problem areas are the focus of this paper. The paper also discusses the principal technology and components of the system as well as system development and demonstration.

* The accelerator-based systems discussed here are named Accelerator Based Conversion (ABC) and Accelerator Transmutation of Waste (ATW). Both employ near-identical technologies. The primary difference is that ABC refers to systems aimed at plutonium destruction while ATW refers to systems aimed at the more general application of transmutation of nuclear waste.

OVERVIEW OF ABC/ATW SYSTEM COMPONENTS

Figure 1 provides an overview of major components of ABC/ATW systems. More information concerning them and their performance is contained in a later section. In ABC/ATW systems a particle accelerator operating at an energy around 1000 MeV produces a high-intensity beam of protons. These protons are directed into a heavy metal target (e. g. lead) where up to 30 neutrons can be produced per 1000 MeV proton. Neutrons produced from this source are moderated to thermal energies and multiplied in subcritical blankets. Each blanket consists of graphite to moderate neutrons and a fluid fuel, molten salt, that contains actinides and other materials to be destroyed. As will be discussed later, an ABC/ATW system will generate large amounts of thermal energy (up to several thousand megawatts) in the process of actinide destruction via fission. Electricity is produced by conversion of this thermal power via conventional means. About fifteen percent to twenty percent is used to power the accelerator; the rest is available for the commercial grid. The use of the molten salt fuel form allows high thermal to electric conversion efficiencies (in the range of forty-four percent), so that the net efficiency for power conversion of the system (after power is provided for the accelerator) is greater than thirty-five percent. (Current reactors have similar efficiencies for conversion of thermal power into electricity.) The system also contains components used in the cleanup of the fuel and for preparation of materials arising from the feed of spent nuclear fuel into the system. Fuel cleanup is key to achieving the large destruction (factors of 100 to 1000) of long-lived radionuclides. From the fuel cleanup process high-level wastes (fission products and lanthanides) are extracted, most of which could be stored in engineered facilities for periods long enough to let the radioactivity decay (on the order of several hundred years) to low levels. Relatively small amounts of long-lived materials would be left (depending upon the overall efficiency of operation of the system and choices of materials to be transmuted). These could be stored in a repository environment or in optimized disposal forms. Finally, low-level waste would result from the transmutation of long-lived fission products into stable by-products.

IMPACTS OF MATERIAL DESTRUCTION SYSTEMS

The ABC/ATW system capability to destroy key long-lived radionuclides directly impacts:

- the area of global plutonium control and minimization, including both weapons and spent nuclear fuel sources, and
- the long-term disposal of HLW from defense and commercial nuclear reactor operation.

There has been recent attention to the issue of plutonium disposition, primarily that from weapons sources. However, as pointed out in recent studies such as the National Academy of Sciences (NAS) study¹ on Pu disposition, weapons-return Pu makes up about ten percent of the world's total inventory of Pu. Figure 2 illustrates the growth of worldwide Pu inventories as a function of fuel burnup levels in present light water reactors as well as that appropriate for future operation. Even under the conservative assumptions of the analysis² used to generate this data, large inventories of Pu would be present in the world over the next 50 years. This situation has led to some level of concern^{1,3} because of significant issues of world rearmament and proliferation that could result from the recovery and malicious use of this plutonium. Even though most is in spent fuel assemblies, it can be straightforwardly extracted. Analyses such as that of Ref. 4 illustrate there is little difference in the capability to make a nuclear explosive between weapons grade (93% ²³⁹Pu,

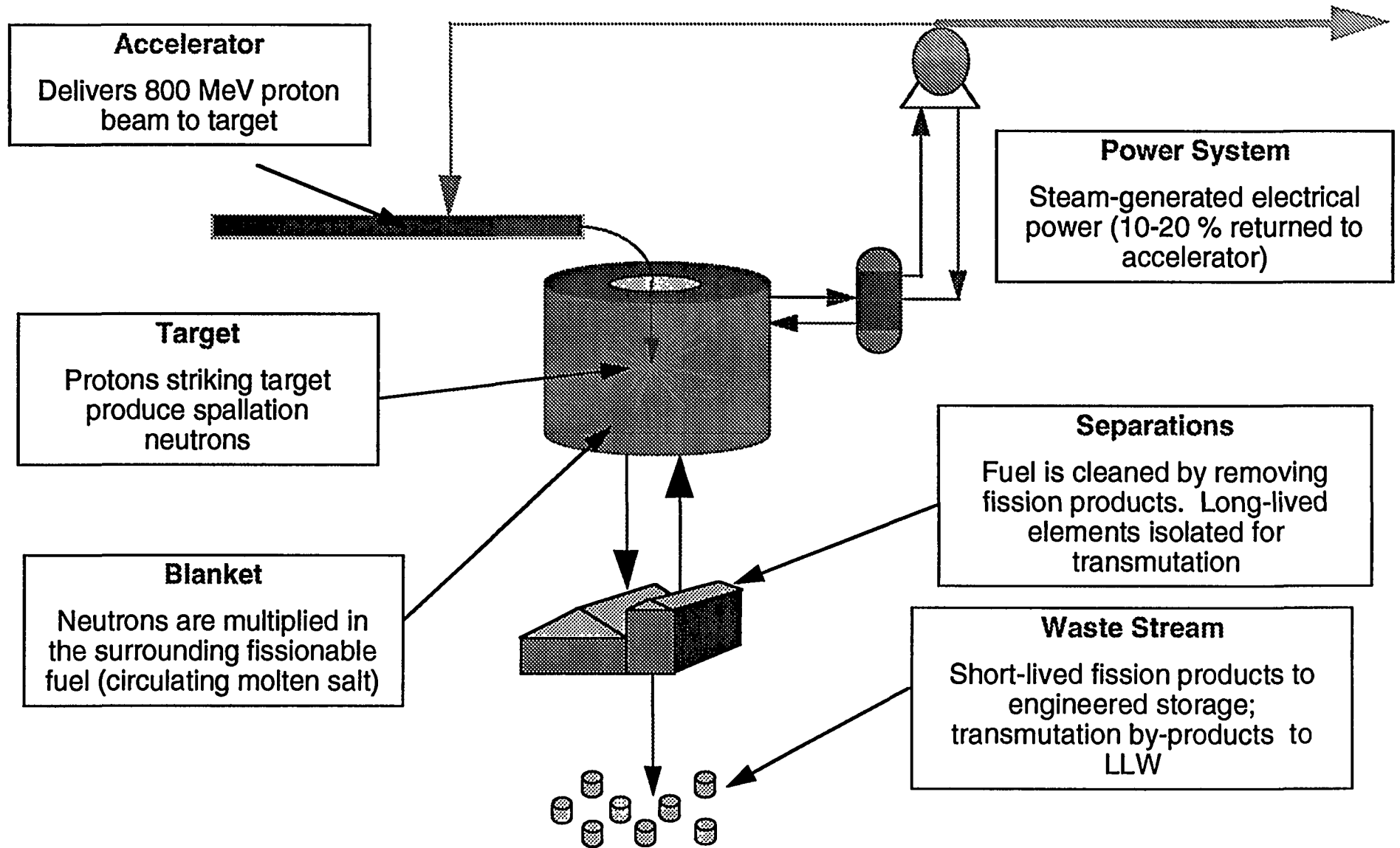


FIGURE 1

Overview of ABC/ATW system components. As described in the text, a medium energy particle accelerator produces a high-intensity beam of protons which is converted to neutrons in a number of spallation targets. A blanket configuration around each target moderates and multiplies, through fission of actinides, neutrons produced in each target. Each blanket produces significant thermal power which is converted to electricity, a portion of which is used to power the accelerator. Finally, separations are used to clean up the system's fluid fuel which is then returned to the blanket for further material burnup.

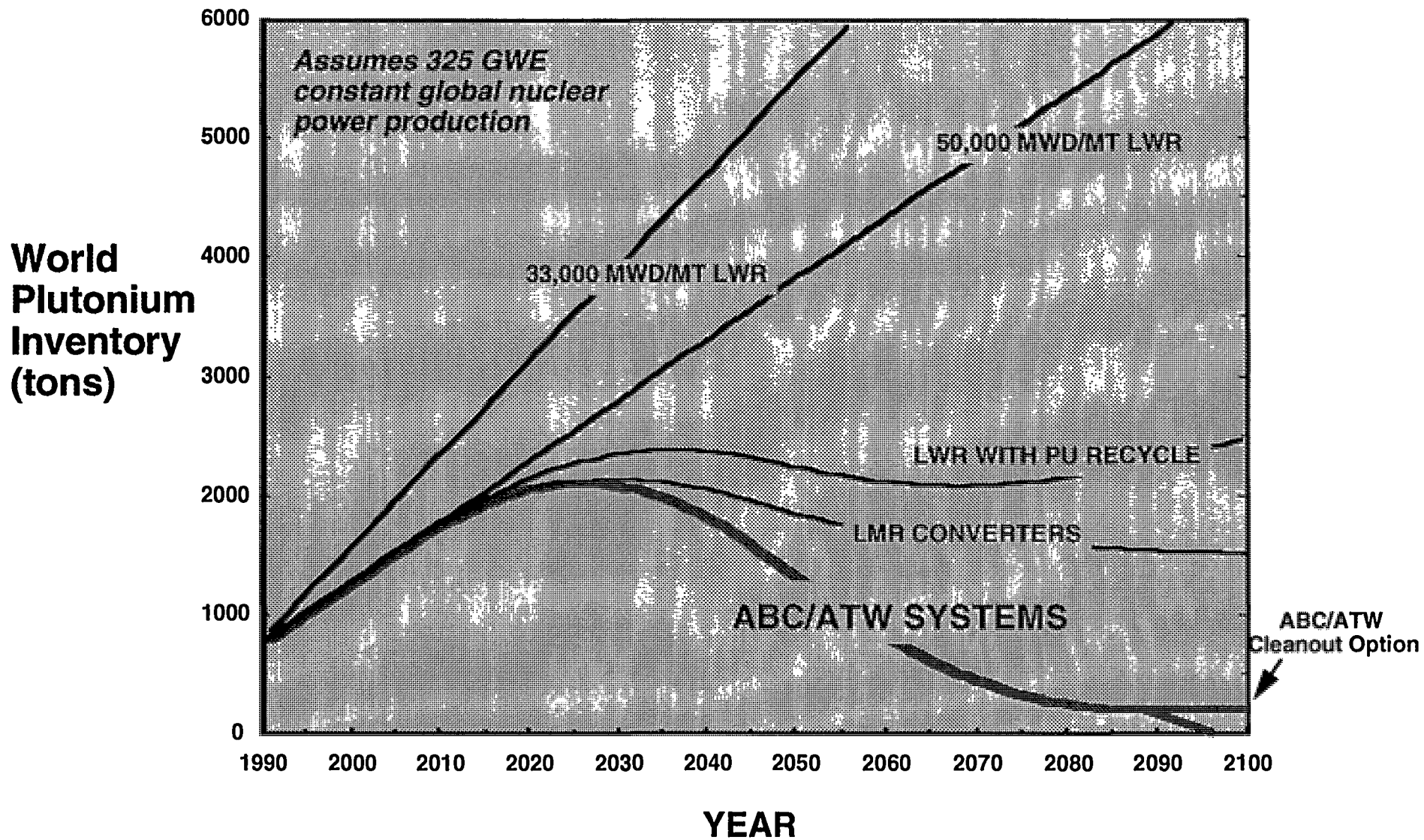


FIGURE 2

Growth in world plutonium inventories under the assumption that the world generation of electricity stays at its current level. This assumption produces minimum values in projected future plutonium inventories. The performance of three nuclear systems (light water reactors, the integral first reactor, and ABC/ATW) for reversing this growth and reducing residual plutonium inventories is also shown.

6% ^{240}Pu) plutonium and reactor grade (65% ^{239}Pu , 24% ^{240}Pu , 9% ^{241}Pu , 5% ^{242}Pu) materials.

With this rise in plutonium inventories, it is natural to ask what can be done to reduce or minimize them. Figure 2 also illustrates the performance of three nuclear systems -- light water reactors with fuel reprocessing, a fast neutron converter reactor with fuel recycle, and an accelerator-based system. For the first several decades all systems reverse the growth trend and reduce Pu inventories down to asymptotic values. For both reactor systems this limit is still 2000 tonnes -- a result of the large inventories of material tied up in the reactor and its associated fuels handling and processing. The ABC/ATW system requires a much smaller inventory of material to achieve the required level of burn. This leads to significantly smaller residual levels of plutonium than the two other nuclear systems used in this comparison. In addition, the ABC/ATW system allows a burn-down mode where the accelerator source of neutrons can be used to reduce further the remaining plutonium inventories in the system.

For the special case of weapons plutonium, an ABC system achieves burnup levels much beyond the "spent fuel standard" identified by the NAS report. Burnup levels of ninety percent for all plutonium isotopes and ninety eight percent⁵ for ^{239}Pu can be achieved without need for fuel recycle. With the accelerator-driven, fluid fuel system, a burnout mode⁶ is possible that uses high-enriched uranium to burn up all plutonium isotopes to 99% and ^{239}Pu to a level of 99.99%. The resulting mixture is primarily made up of ^{238}Pu and ^{242}Pu which have extremely unattractive properties in terms of heat loads and fission cross sections respectively⁴.

The accelerator-based system can also address a number of key issues associated with repository storage by removing long-lived actinides and fission products. A number of recent analyses^{7,8} have focused on heat load effects that would be present at a permanent disposal site such as Yucca Mountain. The presence of the actinides causes the heat load of the repository to increase with time as illustrated in Figure 3. This leads to conditions of thermal instability and cycling. This situation changes the character of the repository significantly. The usual picture of a geologically stable disposal environment would be replaced with a complex thermally-driven system characterized by thermal cycling, moisture recondensation, etc. that would occur over periods of thousands of years.

The presence of large amounts of spent fuel and other nuclear materials in a repository can lead to issues associated with proliferation and materials criticality. The 60,000 metric tonnes of spent fuel destined for repository storage at Yucca Mountain would contain over 500 tonnes of plutonium. Similar scale repositories would be required in other parts of the world. This material presence could lead to the situation of a number of "plutonium mines", dispersed throughout the world, which could be attractive to future generations and which could lead to efforts to recover plutonium from them.

The presence of significant amounts of highly fissile materials such as weapons-return plutonium and high-enriched uranium (HEU) in a repository also leads to significant criticality concerns. Most radionuclides present in a repository will decay to stable end products over time. In contrast, fissile materials, principally ^{235}U and ^{239}Pu (which decays to ^{235}U) remain concerns for time periods longer than the ^{235}U half life (700 million years). Recent studies^{9,10} have indicated that loadings of HEU and/or Pu could be on the order of less than a kilogram per waste container. For materials such as surplus plutonium, disposed of in this fashion, the requirements for disposal canisters could be on the order of 50,000 canisters. At a canister cost of \$0.5 to \$1M each, total disposal costs could be on the order of tens of billions of dollars. Disposal of HEU and weapons-return plutonium may require almost half of the proposed space at Yucca Mountain⁹. Closer packings, with more material per waste package, would be less expensive, but in the case of enriched materials, could present criticality hazards.

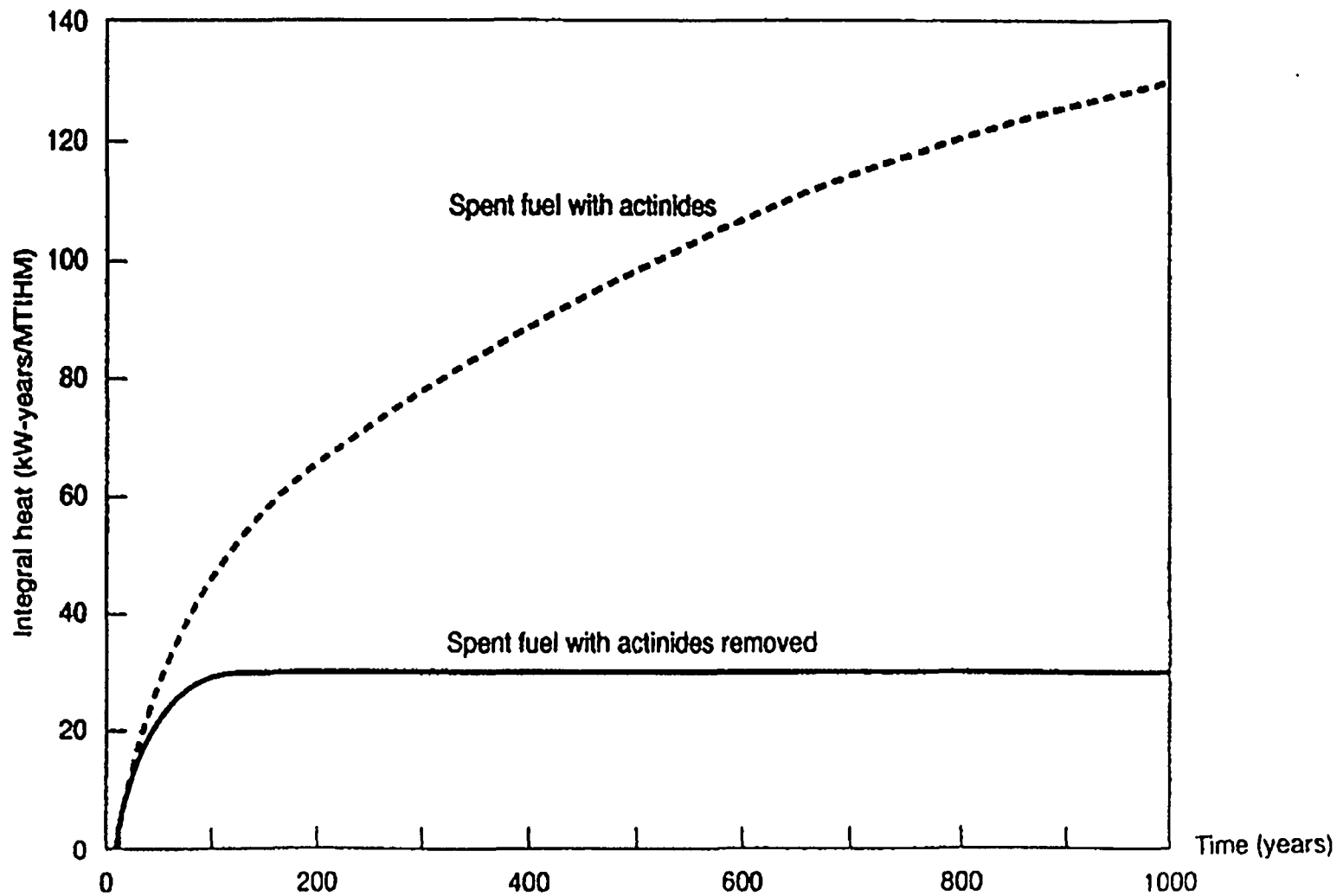


FIGURE 3

The heat load associated with storage of spent reactor fuel at the proposed Yucca Mountain site. Actinides are the major contributor to the increasing (over time) heating that would occur. Removal of the actinides produces heat loads that would be significantly lessened and which would remain constant for future storage periods.

Recent assessments of the amount of materials requiring repository storage (in addition to spent fuels and vitrified waste) indicate that the present repository space may be oversubscribed. Oak Ridge National Laboratory analyses⁹ indicate that special nuclear materials such as weapons Pu, HEU, research reactor fuels, etc. can require substantial amounts of repository space. The oversubscription may be as high as a factor of two. These analyses also confirm that actinide removal from the repository via transmutation could increase capacities as much as a factor of five.^{7,8}

In addition to actinides, other long-lived radionuclides, specifically fission products such as technetium, iodine-129, and cesium-135 can be significant contributors to the dose released at a repository boundary. In a recent assessment of the impact of actinide and fission product transmutation on waste repository performance, Pigford¹¹ found that some long-lived fission products were much more significant dose contributors to release to the biosphere than the major actinides. These results were largely based on the high solubilities of these long-lived fission products with respect to the actinides. Pigford defined the "relative dose index" parameter as a measure of the dose rate of a released radionuclide species relative to that of the dominant long-lived fission products species ⁹⁹Tc. (The dose rate was taken at the repository boundary for differing scenarios for water travel times.) This parameter accounts for both decay and sorption on rock as the species migrate from the repository and thus is a function of the water travel time to the biosphere boundary. The relative dose index values presented by Pigford are shown in Figure 4. Note that except for long water travel times, the long-lived fission products ¹²⁹I and ¹³⁵Cs are larger release dose contributors than any of the major actinides.

The actinide solubilities used by Pigford were those of actinide metals; however, recent Yucca Mountain performance assessments¹² recognize that the oxide solubilities are the relevant values. This difference in solubilities is significant as shown in Table 1. These values are from 10³ to 10⁵ larger than those used by Pigford. If Pigford's methodology is modified for these more recent actinide solubilities, the actinides become larger contributors to the repository release dose, but the long-lived fission products can still represent significant contributors. Thus if transmutation is to effect significant reductions in the repository release dose, the transmutation strategy must include the long-lived fission products, in addition to the actinides.

Table 1 - Actinide Solubilities (moles/liter) Assumed for Repository Assessments

<u>Material</u>	<u>1991 Value</u>	<u>1993 Value</u>
Uranium	10 ^{-7.7}	10 ^{-4.5}
Neptunium	10 ⁻⁹	10 ⁻⁴
Plutonium	10 ^{-12.4}	10 ⁻⁷
Americium	10 ^{-8.2}	10 ⁻⁷

CHOICE OF TRANSMUTATION SYSTEM PARAMETERS

The issues discussed associated with long-term storage above can be largely remedied through elimination of these materials through transmutation. With such an objective in mind, what are desirable features and attributes that would be required in a transmutation system. How do these requirements relate to choices made for accelerator-based systems? An overview of three important requirements is provided in Table 2 where their importance in systems aimed at several nuclear materials destruction applications is given.

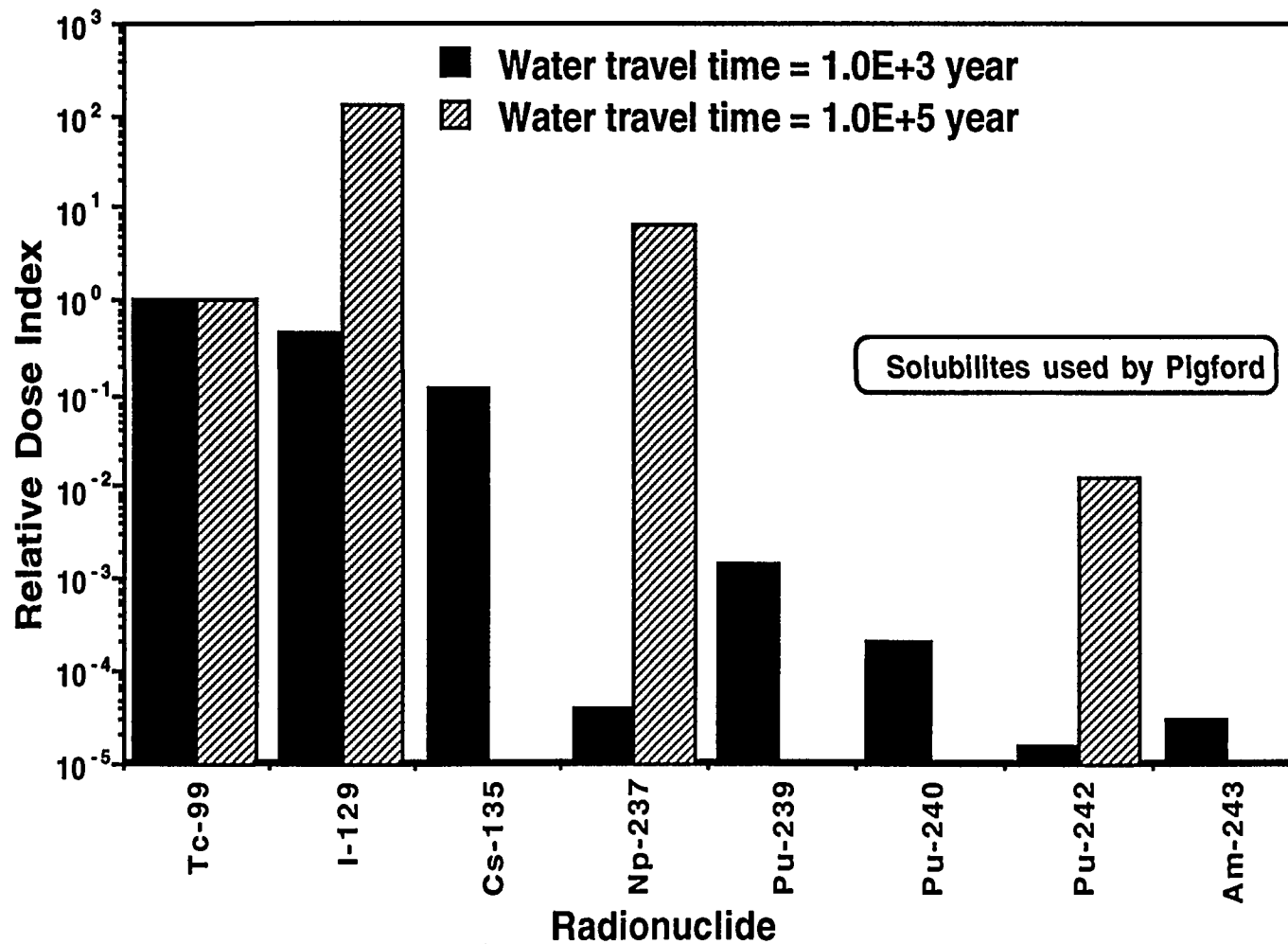


FIGURE 4

The relative dose index computed by Pigford¹¹ for fission products and actinides. The results of this analysis show the importance of long-lived fission products (technetium99, iodine129, etc.) as contributors to future dose released from a repository.

Table 2 - Operational Requirements for Accelerator-System Applications and System Components That Allow Meeting Them

	<u>Safety/Control</u>	<u>Neutron Economy</u>	<u>Fuel Cleanup</u>
Weapons Pu	accelerator		
Global Pu Management	accelerator	accelerator, fluid-fuel	fluid-fuel
Long-Lived Waste Transmutation	accelerator	accelerator, fluid fuel	fluid fuel

Safety and Control

All applications that are discussed here require maximal levels of safety and control. The use of a particle accelerator to provide subcritical operation provides an important degree of reactivity control in systems where nuclear parameters (delayed neutron fraction, temperature-dependent cross section behavior) are not as favorable as in other systems that burn, for example, primarily uranium. The degree of subcriticality can be defined at levels where reactivity effects cannot lead to uncontrolled criticality excursions.

Neutron Economy

Systems that aim at destruction of materials require enhanced neutron economy in order to deal with species that are unreactive in the nuclear sense. For applications to global plutonium management, enhanced neutron economy enables burning of threshold-fissioner plutonium isotopes (^{240}Pu , ^{242}Pu) as well higher actinides that build in during operation. For the long-lived waste transmutation system, the accelerator provides the neutron source that transmutes the long-lived fission products discussed later. For both applications, the accelerator-based neutron source and the fluid fuel (which permits rapid removal of neutron poisons) both contribute to meeting this requirement.

Fuel Cleanup

In systems aimed at global plutonium destruction and long-lived waste transmutation, multiple passes of feed material through the system's neutron environment are required to achieve large burnup values. High burnup introduces significant quantities of neutron poisons into the fuel which must be removed. The liquid fuel medium permits straightforward fuel cleanup needed to achieve large reduction factors of plutonium inventories (reduction factors of 100 to 1000). The accelerator again provides a subcritical operational environment that provides robustness required for a liquid fuel system containing variable fissile material feed and inventory isotopics.

In addition to choices aimed at satisfying these requirements, the ABC/ATW system uses low-energy neutrons for transmutation. The nuclear cross sections for thermal-neutron induced transmutation are one to two orders of magnitude greater than equivalent cross sections at higher energies associated with fast neutrons of the type used in breeder reactors. The much higher cross section means that inventories of materials occurring in the system can be an order of magnitude smaller than other fast neutron systems, providing that neutron flux levels remain at levels on the order of $3 \text{ to } 5 \times 10^{14} \text{ n/cm}^2/\text{sec}$.

COMPONENTS OF ABC/ATW SYSTEMS

Figure 1 provided a general introduction to the components of ABC/ATW systems. This section provides more details concerning them.

Figure 5 illustrates the particle accelerator that would be employed. The accelerator consists primarily of radiofrequency-driven acceleration modules. At the low-energy end of the accelerator, an ion source is used to produce the initial beam and to accelerate it up to an energy of 0.075 MeV. It then enters a radiofrequency quadrupole for further acceleration. More importantly this component is key to properly define the beam's spatial, momentum, and divergence characteristics. The radiofrequency quadrupole is a Russian invention that allows replacement of large electrostatic-based sections (a Cockcroft-Walton electrostatic accelerator) with a more compact and efficient device. After leaving the radiofrequency quadrupole, the beam is accelerated in two additional radiofrequency devices -- a drift-tube linac and a bridge-coupled linac -- up to an energy around 40 MeV. A "scraper" device is used to remove unwanted "tails" in the beam spatial distribution before introduction into the main part of the accelerator which consists of coupled-cavity structures. The removal of such tails is aimed at minimizing the activation of the high-energy portion of the accelerator so that remote maintenance can be avoided. Once in the coupled-cavity linac, the beam is accelerated up to its maximum desired energy where it is then available for transport to a number of target/blanket structures. A series of magnets are used for such transport. They also shape the beam's spatial distribution before its interaction with the target.

The beam current appropriate for an ABC/ATW is around 100 milliamperes, so that for a beam energy of 800 MeV, the total beam power is 80 megawatts. High-power linear accelerator efficiencies are predicted to be in the range of forty-five percent, so that the total electricity required to power the accelerator is approximately 180 megawatts.

The accelerator described here is based upon substantial development in high-power linac technology made under the Strategic Defense Initiative. As a result, representative portions of the accelerator that accelerate the beam up to energies of several tens of MeV have been demonstrated individually at representative performance levels. The main section of the accelerator, the coupled-cavity linac, was developed for use at the Los Alamos Meson Physics Facility (LAMPF) where such devices have successfully accelerated high-peak power beams over the past twenty years. This experience base as led a number of review committees¹³ to conclude that construction of such a high-power accelerator is feasible after creation of a lower-energy test stand that would demonstrate integrated, high-current operation.

The next major component of the system is the target/blanket shown in Figure 6. The proton beam enters from the top where it strikes a liquid metal production target comprised of either molten lead or the lead-bismuth eutectic. This material choice enables high neutron production (approximately 30 neutrons/1000 MeV proton), low cross sections for the capture of low-energy neutrons that may be scattered back into the target, plus the ability to handle high-beam power in a compact geometrical configuration. Surrounding this target is a blanket region that is made up of a moderator (graphite) that thermalizes the neutrons produced in the system so as to provide conditions where nuclear transmutation cross sections are maximized. The fuel containing the actinides (plutonium, neptunium, americium, curium) to be fissioned is a fluoride-based molten salt -- LiF-BeF₂ (denoted as FLIBE). This fuel carrier form was demonstrated in the extensive Oak Ridge National Laboratory¹⁴ program of the 1960's and 1970's. This fuel salt was chosen because of its neutronics characteristics (low neutron absorption) and attractive physical characteristics such as high-temperature, low-pressure operation. These latter properties permit high efficiency operation without the high coolant pressures characteristic of current reactors

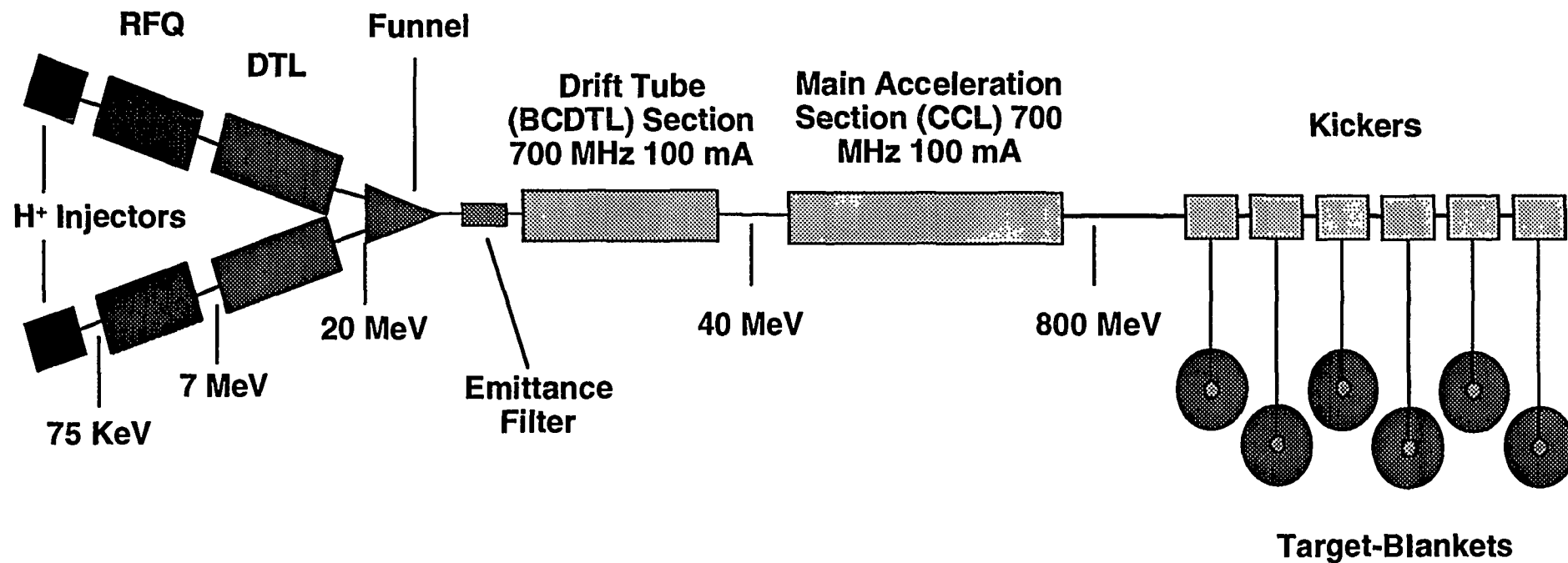


FIGURE 5

The accelerator concept proposed for ABC/ATW systems. The linear accelerator (linac) technology is an outgrowth of results from the Strategic Defense Initiative as well as operational experience from the Los Alamos Meson Physics Facility. Other technology approaches (i.e. high-current cyclotrons) could also be employed in ABC/ATW systems.

(pressurized water reactors). Blanket operation is subcritical -- that is the neutron source strength multiplication parameter, k_{eff} is less than one. (A value of 1 must be maintained in a reactor to assure that criticality, and thus conditions of self-sustaining fission reactions, occur.) In ABC/ATW systems k_{eff} values are in the range of 0.90 to 0.96. This k_{eff} range means that the neutron source strength is multiplied by factors of ten to twenty five and depends upon the types of materials to be destroyed or transmuted in the system. Reference 15 provides more discussion on the choice of k_{eff} from perspectives of safety and control, the ability to transmute long-lived fission products, and impacts on operational characteristics of the system. One target/blanket module would operate at a thermal power in the range of 500 to 700 megawatts. This range is chosen to allow passive cooling of the blanket in a fashion similar to that employed in reactors such as the Modular Helium Reactor¹⁶. This approach would also allow factory construction of the modules with the result that system costs could be lowered as a result of standardization and assembly-line construction methods. A number of such modules would be grouped together as "building blocks" so as to optimize the overall economics of the accelerator based system. A reasonable system size would be in the range of 3000 MW_t which would mean that four to six modules could be driven by one accelerator supplying less than 100 milliamperes of beam current. Because of the high thermal-to-electric conversion efficiency provided by the salt (44%) and the high accelerator efficiency (45%), the net electric output from such an ABC/ATW installation is expected to be over 1000 megawatts.

The operation of such a facility could handle the waste actinide and key long-lived fission product discharge of four 1000 MW_e light water reactors (LWR) assuming a LWR fuel burnup of 33,000 megawatt days per ton of uranium metal. If the burnup were higher (55,000 MWd/ton) then the discharge from approximately 6 LWRs could be supported by one ABC/ATW system.

In addition to the target, moderator, and fuel, the target/blanket system contains a number of other components. These include pumps required for fuel flow into the nuclear reaction region, heat exchangers to remove heat produced during fission, and a gas system to remove volatiles during operation. Also associated with each target/blanket module (or groupings of modules) would be components equivalent to those occurring in other nuclear systems. These include a multiple system of barriers to prevent radionuclide release under accident conditions and a steam system to power generators to produce electricity. In addition to components that are analogs to those occurring in current nuclear systems, the ABC/ATW target/blanket modules would have components needed for introduction of fuel, for the hold-up of volatiles (to allow their decay), and for the storage and drainage of the fuel salt. Finally, components would be needed to link the target/blanket system with the fuel salt cleanup system which is described next.

In order to achieve high material burnup in any nuclear system, neutron poisons must be removed during operation. These poisons dominate the eventual performance of any nuclear system where high material burnup is an objective. In the case of a solid-fueled system, fuel must be removed, reprocessed, and reconstituted to achieve neutron poison removal. Also in such systems, high-destruction levels (fuel burnup) are often achieved by introducing large amounts of excess reactivity into the system at the beginning of the fuel burnup cycle. This means large amounts of enriched nuclear materials must be contained in the core along with significant inventories of materials which provide required control characteristics. The choice of the liquid-fuel medium in the ABC/ATW system ensures that removal of neutron poisons is a much more straightforward process. Large amounts of excess reactivity is not needed. Instead, fuel can be added as operation progresses to aid in overcoming effects from neutron poison buildup before salt cleanup is required.

The use of the molten salt fuel provides inherent capabilities for neutron poison removal that cannot occur in standard nuclear systems. Volatile neutron poisons such as

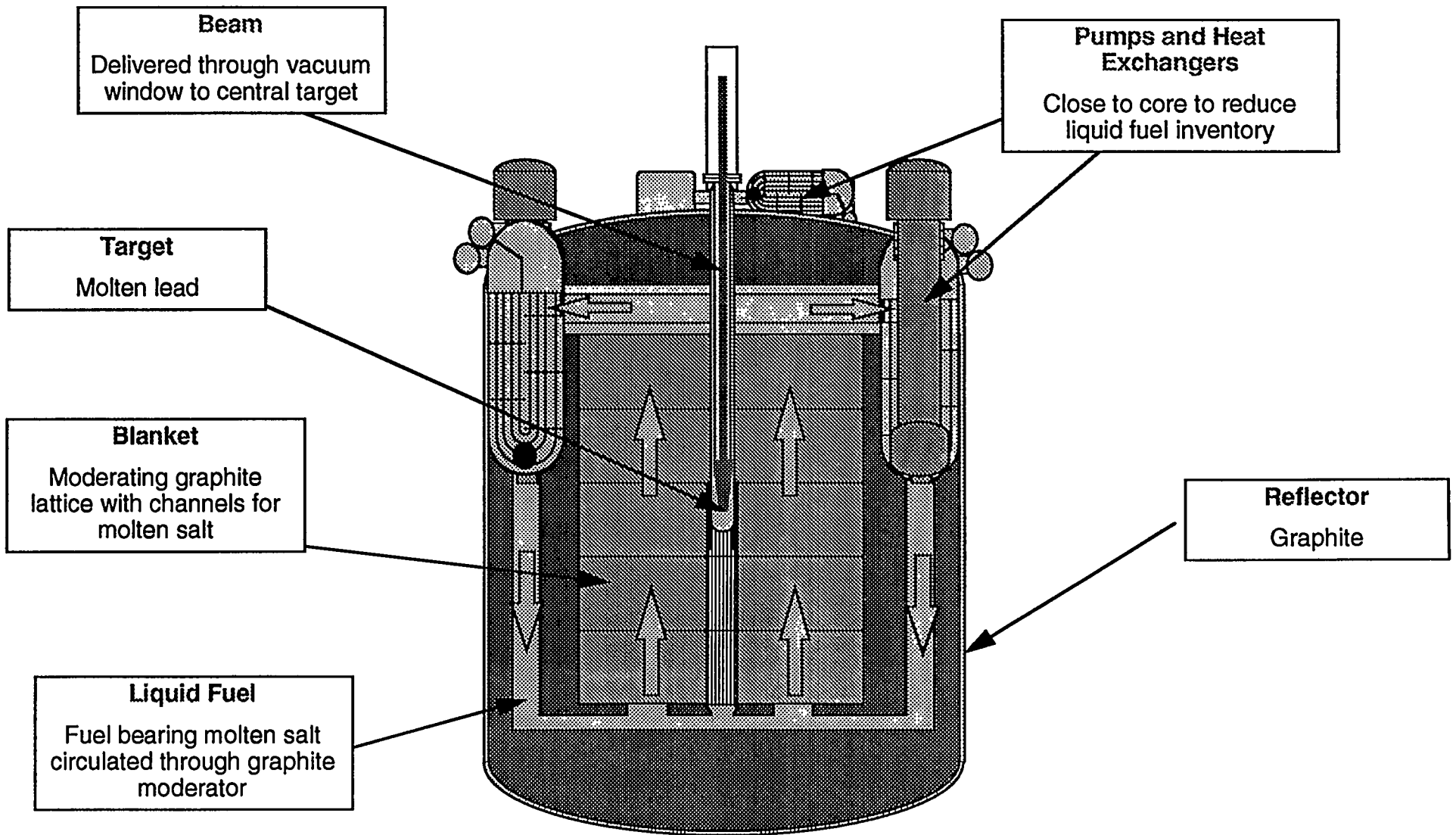


FIGURE 6

The conceptual layout for an individual ABC/ATW target/blanket component. The neutron production target would be made of lead or lead-bismuth. The multiplying blanket would use technology from the Oak Ridge molten salt reactor program and would operate at thermal powers in the range of 500 to 700 MW.

xenon and krypton migrate from the salt because they are insoluble in it. This process can be aided through use of a gas sparge (helium) so that such volatiles can be removed on timescales of a few minutes. Likewise, chemical properties of the salt can lead to conditions where noble and semi-noble metals will come out of the salt on timescales on the order of a few hours. These constituents can precipitate from the salt in the form of metals depending upon the chemical potential at which the salt is maintained during operation. During operation of the molten salt reactor experiment¹⁷ at Oak Ridge during the 1960's significant fractions of these materials collected on cold surfaces of the system in the heat exchanger. In ABC/ATW several methods can be used to specifically collect and remove such materials from the system. These methods include filtration, special cold traps, and electrolytic deposition. Again, the removal of these products contributes significantly to the neutronics performance of the system and occurs naturally because of the chemistry of the fuel salt.

Removal of neutron poisons via the above methods involve processes that are inherent and which can operate "on line". They can remove up to one-half of the neutron poison products created during fission. The remaining requirement is the removal of residual neutron poisons (mainly lanthanides) and the return of the cleaned-up salt back to the blanket system. Several general approaches are under investigation that would allow such removal to take place and which do not involve the creation of separated streams of actinides such as plutonium. Avoidance of the latter situation is important for proliferation reasons. The techniques needed for clean up of the salt are similar in many respects to that successfully developed for pyroprocessing. There, no separated actinide streams are produced, use of reagents needed for aqueous-based processing is avoided, and the operational scale can be smaller and cheaper than that required for currently-used aqueous processes such as PUREX.

Several methods are under investigation for residual salt cleanup. These include distillation, precipitation, electrochemistry, and use of centrifuge-base separations. Distillation techniques would be used to remove species that would be volatile at higher salt temperatures. The remaining fuel mixture could be converted to a chloride salt where electrochemical techniques developed and under testing at Argonne National Laboratory¹⁸ or at Rockwell corporation¹⁹ could be used for the needed actinide/lanthanide separations. Expected performance predictions¹⁹ indicate that factors of ten to thirty could be obtained per separation stage.

Alternatively, use of precipitation-based methods²⁰ could be used to sequentially extract actinides and then lanthanides. In this approach, conversion from a fluoride to a chloride salt would not be required. This technique exploits differences in free energies of formation of oxides of the materials present in the salt. Actinides would be precipitated using a catalyst such as yttria (Y_2O_3). The actinide oxide lump (containing all actinides present together) would then be fluorinated and returned to the salt. Lanthanides would be separated via use of a lithium metal reductant, extracted, oxidized, and then disposed of in a form such as glass.

A final approach to actinide/separations involves use of molecular centrifuge²¹ which would exploit mass differences between heavy actinides and the lighter lanthanides and other fission products. A dilute salt solution would be used. Early analyses indicate that enhancements of the fission product concentration (versus actinides) would be on the order of a factor of five near the outer regions of the centrifuge. A number of parallel units, consisting of a number of small-volume centrifuge stages, would be used to achieve required separation levels.

Front-end preparation of spent fuel would be required before introduction into the ABC/ATW system. The first step involves removal of the fuel clad and uranium. Pyroprocessing methods appear attractive for recovery of an actinide "lump" for introduction into the transmuted. A method based upon lithium reduction of reactor spent

fuel under development at Argonne National Lab²² could be promising for application to the ABC/ATW system. This step would produce a mixture of plutonium, americium, neptunium (also having small levels of contamination by uranium and the lanthanides) which would be fed into the transmuted. Zirconium waste produced could be recycled for use in other nuclear systems. The recovered uranium represents the largest mass (> 90 percent) constituent in spent fuel. It could be re-enriched to produce fuels used in standard light-water reactors.

Figure 7 summarizes the fuel cleanup steps associated with operation of an ABC/ATW system.

TECHNOLOGY STATUS AND DEVELOPMENT

The ABC/ATW technology has, as its basis, the substantial level of development and demonstration that resulted from the molten salt breeder reactor program at Oak Ridge National Laboratory. As a result, several assessments^{23,24} state there were no basic technology feasibility issues that need to be addressed. As an example, an assessment²³ by the Molten Salt Group (headed by Ebasco services and which included five major industrial firms and fifteen utilities) concluded in 1971 that the molten salt technology which existed at that time was sufficient to justify the construction of a molten salt reactor demonstration plant. ABC/ATW technology development also can utilize results from the Accelerator Production of Tritium (APT) program (for accelerator development) and separation results from extensive pyroprocessing development that has occurred at Argonne National Laboratory.

The major technology and feasibility issues thereby relate to the particular design of the ABC/ATW systems and the capability of the system to meet its objectives. To address these issues a multi-element technology development and demonstration plan is envisioned. It is made up of several components:

- design of an ABC/ATW prototype;
- creation of a "proof-of-principle experiment at the Los Alamos Meson Physics Facility which would demonstrate accelerator-subcritical blanket coupling and operation;
- execution of key technology risk reduction experiments in areas pertaining to the blanket, fuel chemistry, and materials; and
- a separations effort which would demonstrate approaches and components required for fuel-salt cleanup and preparation of feed materials which would go into an ABC/ATW system.

An ABC/ATW technology development program is illustrated schematically in Figure 8. The areas listed above would make up a first stage development and demonstration program which would last on the order of six years. This phase would develop information required to establish details of the expected performance of ABC/ATW system concepts and would lay the groundwork for proceeding to the second phase aimed at design and construction of a prototype system. A prototype system could include operation of one of the ABC/ATW target/blanket modules described earlier. Such a unit would operate at significant thermal power (several hundred megawatts) and would include all major auxiliary systems and components at relevant scales required for full plant operation. Destruction of plutonium and/or transmutation of other long-lived wastes could begin during this prototype development and operation phase. The third and final phase would center around the building of additional modules required for operation of a complete ABC/ATW system and would be followed by construction of follow-on systems.

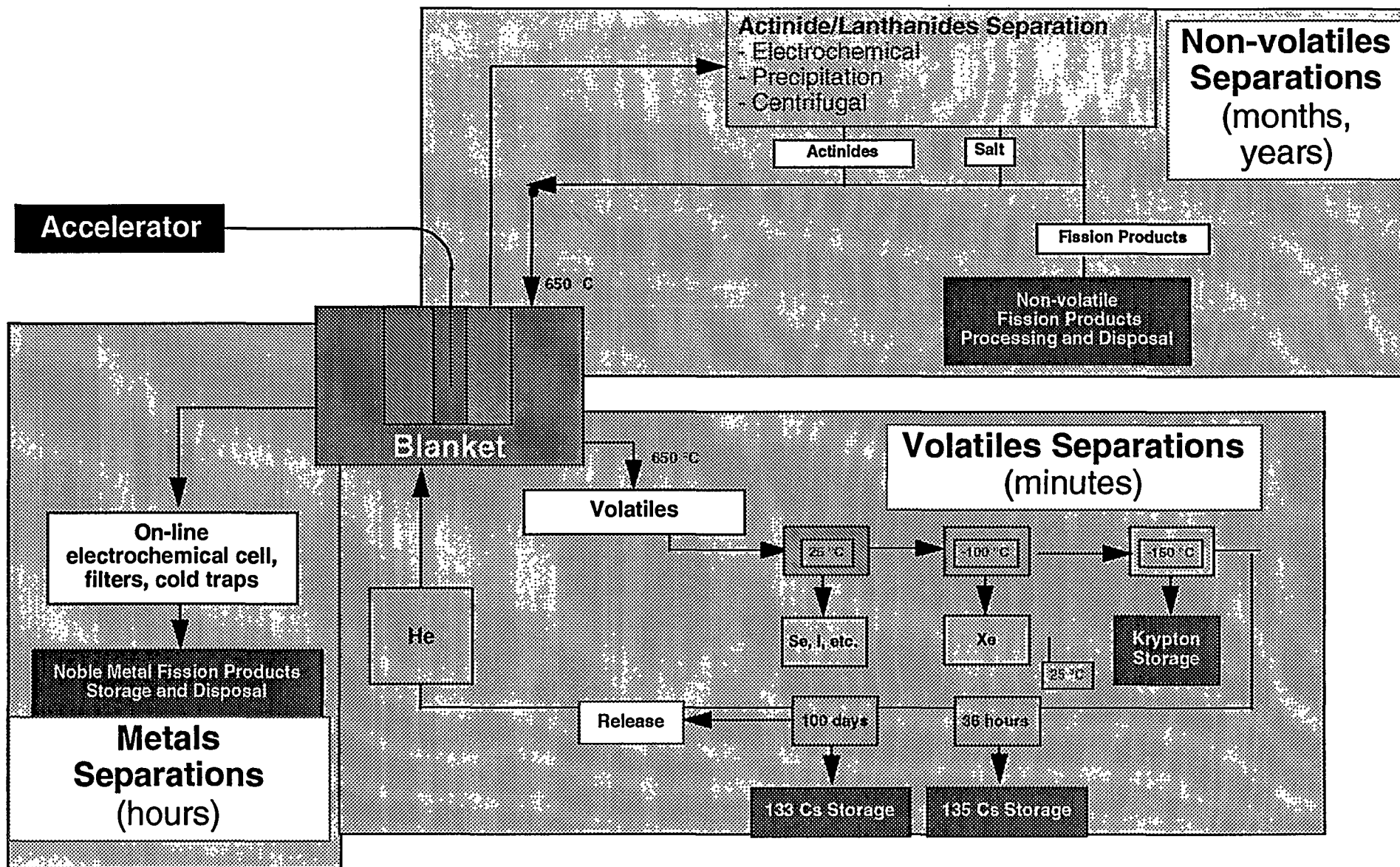


FIGURE 7

Fuel cleanup components associated with the ABC/ATW system. The use of the molten fluoride salt in the blanket permits methods for significant removal of neutron poisons through "inherent" methods. Examples of such methods include removal of volatile contributors through sparging and the removal of noble metals through chemical control of the salt. The remaining actinide/lanthanide separations can be accomplished using precipitation, centrifugation, and electrochemical means.

The technology development and demonstration first phase effort would focus on ABC/ATW areas that represent updates or departures from existing technology bases. A primary difference in the ABC/ATW system and the earlier Oak Ridge molten salt reactor program is the use of fuels (in ABC/ATW) that contain primarily plutonium and heavy actinides. In contrast, the Oak Ridge system utilized a fuel made up of large amounts of thorium and uranium because of the program's focus on creation of a thorium breeder reactor. The use of plutonium leads to issues pertaining to materials solubility in the fuel salt and chemical control of the salt. Other areas include testing of proposed secondary cooling salts, handling of tritium in the system, materials performance, and performance conditions when significant fractions of fission products are present in the system. Underlying these areas, plus others described here, is the need to reestablish the molten salt technology base developed two decades earlier. Experimental work in the areas described here (plus others) would continue beyond the first phase technology demonstration effort.

A principal focus of the first phase effort would be design, construction, and operation of a "proof-of-principle" experiment performed using the LAMPF proton beam. This beam has sufficient power (~ 1 megawatt) to drive an experimental assembly having a thermal power as high as 30 to 40 megawatts. In reality, the system would operate at a lower power, around 5 megawatts, and would be designed to demonstrate operation of a coupled accelerator-subcritical molten salt blanket system. This includes demonstration of the molten metal target operation, handling of a significant amount of fuel salt (pumping, temperature control, flows, etc.), control of the subcriticality level of the system, operation of key system components such as pumps and heat exchangers, and performance diagnostics. Figure 9 provides a conceptual layout for such a proof-of-principle experiment.

A third important area of the first phase of technology development and demonstration would focus around design of a full ABC/ATW system and the prototype described above. A significant design base exists from the earlier Oak Ridge program but this must be updated to incorporate more modern reactor design approaches, such as pool type configurations, and must address modern safety and containment requirements.

The final emphasis area for the first phase of technology development and demonstration is in the area of separations needed for fuel cleanup and feed material preparation. Small-scale experiments would be used to develop and demonstrate techniques for electrochemical, filtration, or cold-trap removal of noble metal fission products. The effort would also demonstrate techniques needed for removal of lanthanides from the fuel salt before it is returned to the system. Techniques that would be investigated would be pyroprocessing, centrifugation, distillation, and precipitation-based methods. Integral to these efforts is the overall specification of an integrated flowsheet for the system that would be used to predict material flows, size components, and define expected separation efficiencies. Finally, proposed techniques that are under development for preparation of front-end fuel preparation (for example the pyrochemical-based approach under development at Argonne) would be tested for applicability to the ABC/ATW system.

CONCLUSIONS

Material destruction systems embodied in the ABC/ATW concepts show significant promise for elimination of long-lived radionuclides that present environmental and/or proliferation threats. Transmutation of such material could have significant impacts on problem areas such as global plutonium management. Their destruction would also address many issues facing long-term repository storage of high-level nuclear waste -- ones that remain important over the entire time period for which the waste is stored.

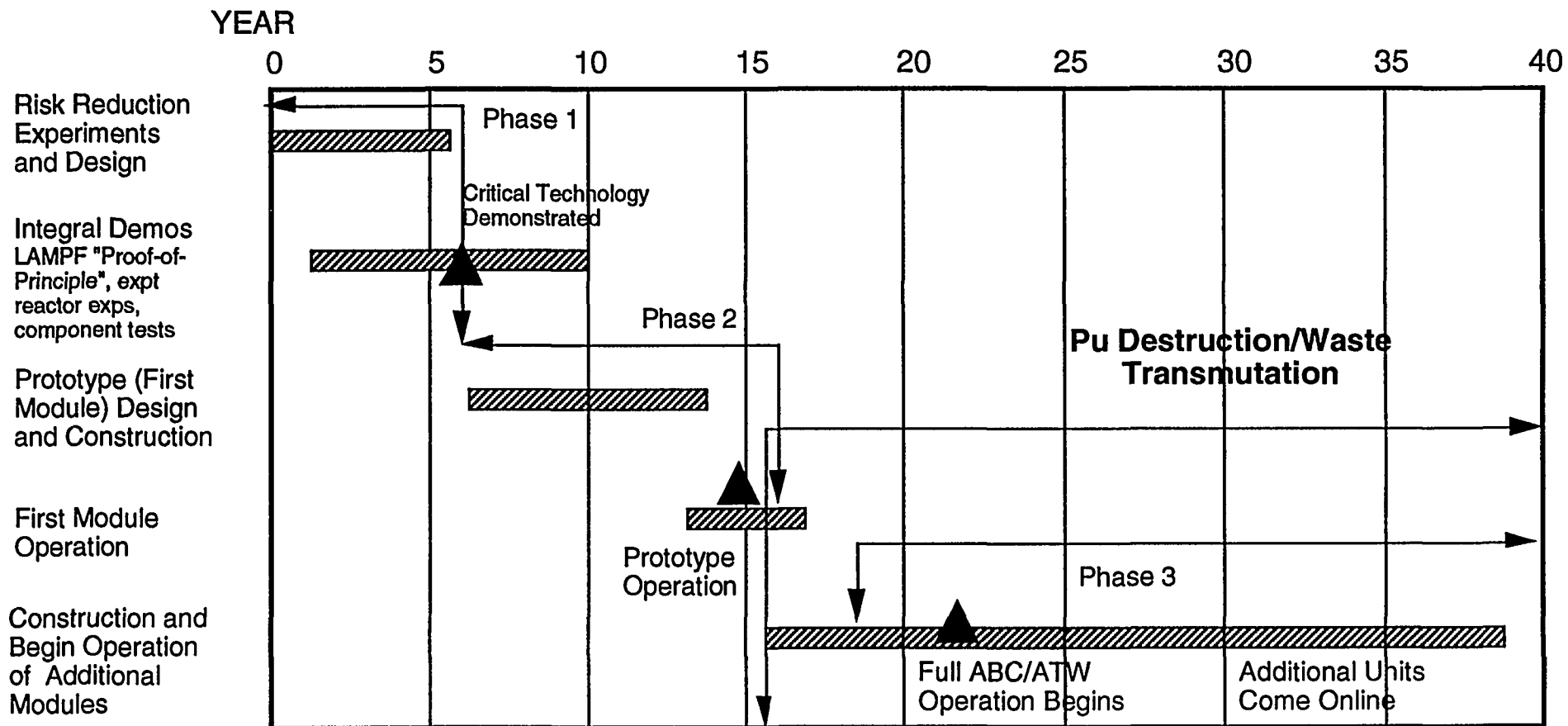


FIGURE 8

Conceptual program for ABC/ATW system development and implementation. Three phases are shown which are resolution of key issues (Phase 1), design and construction of a prototype facility (Phase 2), and prototype operation and construction of follow-on systems (Phase 3).

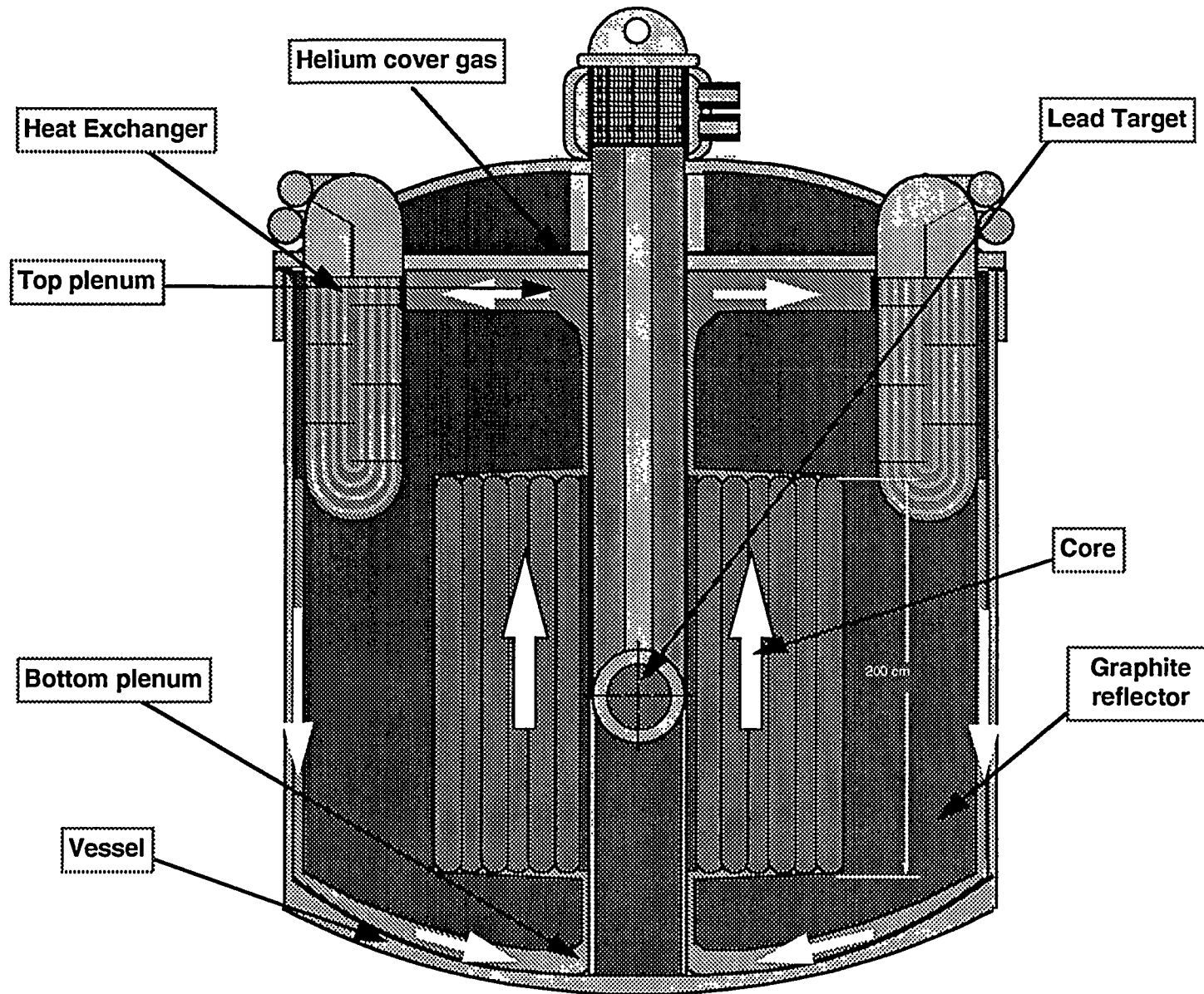


FIGURE 9

A "Proof-of-Principle" experiment proposed for the Los Alamos Meson Physics Facility (LAMPF). In contrast to the configuration shown in Figure 6, the beam enters the assembly horizontally (the beam is perpendicular to the page in this frontal perspective). This experiment would utilize the 800 MeV, one milliamperce beam of the facility to investigate coupling between an accelerator and a subcritical, molten-salt blanket system. The experiment would demonstrate operation and control using an accelerator as well as performance of key system components and diagnostics.

The ABC/ATW systems provide features for meeting these requirements that are not present in other approaches. These include completeness (ability to simultaneously transmute actinides and fission products), operation with low-inventories while achieving high material destruction rates, robust safety characteristics, and operation in modes inherently resistant to proliferation concerns.

The accelerator system coupled with a molten salt fluid fuel provides a uniquely attractive system for destruction of plutonium and other waste materials. The fluid fuel allows operation where conditions for fuel cleanup are straightforward and which can be done with minimal waste stream production and potentially cheaper costs. The accelerator adds the needed level of control required for a fluid fuel environment and provides extra neutrons that can be used to transmute fission products as well as to relax operational constraints present in a materials transmutation system.

The technology base for the ABC/ATW system is extensive, with key technology development results available from the Oak Ridge molten salt reactor program. With this technology foundation available, a technology development and demonstration effort could create an operational prototype within a period of twenty years. This timeline is comparable to times available before the process of final disposal and disposition begins for nuclear materials or spent reactor fuel. Development of the ABC/ATW system would provide the option where key components of long-lived, high-level nuclear waste are destroyed, thus eliminating uncertainties that otherwise extend for thousands of years into the future.

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