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SCIENCE OPPORTUNITIES AT HIGH POWER ACCELERATORS LIKE APT

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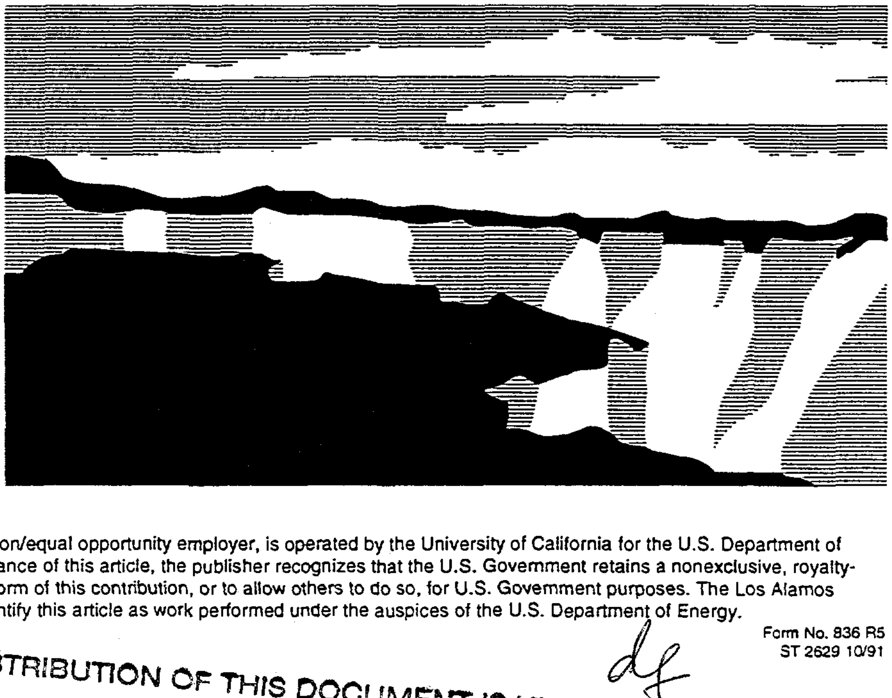

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SCIENCE OPPORTUNITIES AT HIGH POWER ACCELERATORS LIKE APT

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This paper presents applications of high power RF proton linear accelerators to several fields. Radioisotope production is an area in which linacs have already provided new isotopes for use in medical and industrial applications. A new type of spallation neutron source, called a long-pulse spallation source (LPSS), is discussed for application to neutron scattering and to the production and use of ultra-cold neutrons (UCN). The concept of an accelerator-driven, transmutation of nuclear waste system, based on high power RF linac technology, is presented along with its impact on spent nuclear fuels.

Introduction

In other papers presented at this Symposium, authors have discussed the science opportunities related to spallation neutron sources in the following areas: neutron scattering for materials science, neutron science for defense programs, neutrino physics, and materials damage research. In this paper, I will discuss opportunities in other scientific areas that would be significantly impacted by a very high power accelerator like the accelerator production of tritium (APT) accelerator.

Radioisotope Production

Radioisotopes are critical for medical diagnostics and treatment of cancers. They are also used in industrial processes to interrogate materials to detect defects. Radioisotopes are produced at high power accelerators by several processes.

First, neutron-rich isotopes can be produced by neutrons created from stopping the high energy proton beams in a heavy metal target, such as lead or tungsten. This spallation process is very efficient, producing on the order of 25 neutrons for 1 GeV protons stopping in lead. The neutrons can be thermalized in a surrounding moderator, such a water or heavy water, and subsequently captured on samples, thereby producing the radioisotope of interest. This process is essentially identical to that used at a reactor to produce radioisotopes, such as ^{99}Mo, which is the precursor to ^{99}Tc, and is widely used in diagnostics of heart function and treatment of cancer, such as bone cancer. Other important neutron-rich isotopes that can be produced include ^{131}I, used in thyroid treatment, ^{133}Xe, used in lung function diagnosis, and ^{188}Re, for bone cancer pain reduction.

There are needs for neutron-rich isotopes outside the medical field that could be impacted by a very high power accelerator. ^{238}Pu is used as a source in electric power generators, known as RTGs, for radioisotope thermal generators, which are deployed in space exploration probes requiring long-term and reliable generation. Capture of spallation neutrons on ^{237}Np could be an effective method of producing ^{238}Pu without the added problem of significant production of ^{239}Pu.

Second, neutron-deficient isotopes can be produced by protons interacting in target nuclei to produce a variety of radioisotopes of interest via (p,xn) and other similar reactions. Some of the important isotopes that can be produced via this mechanism include ^{67}Cu, which is being used in cancer research studies, ^{68}Ge, which is widely used for PET scan calibration, and ^{194}Au, which is used in arthritis research. The list of neutron-poor isotopes of research interest is quite long, and it would be difficult to include all potential isotopes and their uses in this paper. However, it is important to recognize that in the U.S. the only sources of these neutron-poor isotopes are the Brookhaven Linac Isotope Production (BLIP) facility at the 200 MeV proton linear accelerator at Brookhaven National Laboratory, and the 800 MeV proton linac at LANSCE, the Los Alamos Neutron Science Center at Los Alamos National Laboratory.

Neutron-poor isotopes are also used outside the medical field for diagnostics. Examples include ^{109}Cd, used in engine part diagnosis, and ^{32}Si, used in oceanography and environmental studies.

In the 21st Century, the opportunity for growth in the use of these neutron-poor isotopes will depend critically on their availability on a basis of year-round access, and sufficient quantities for regular application to patients. Very high power accelerator technology, like that being developed for APT, could be the enabling development to allow dedicated machines for industrial production of these isotopes.

Although it has been developed over the past 40 years, the field of radioisotope production is still in an embryonic state from the perspective of an industrial production environment. Much like the APT linac will be a production facility for tritium, so in the future other high power accelerators will be used for "production" of radioisotopes for routine use in medical and industrial settings. The present APT developments open the door for accelerator technology to demonstrate its utility in an industrial setting much as reactors did 40 years ago.

Long-Pulse Spallation Source

In other papers in this Symposium, the applications of short pulse spallation sources (SPSS) were described by several authors. In this context, short pulse refers to the pulse duration of the protons being less than the moderation time for neutrons of any given energy. Usually this means a proton pulse of a few μsec or less. In this section, I will describe a new concept called a "long-pulse spallation source (LPSS)" in which the length of the proton pulse is longer than the neutron moderation time. Figure 1 schematically shows the difference between an LPSS and an SPSS. In the discussion to follow the proton pulse will be presumed be 300 μsec or longer.

The LPSS is designed to use the protons directly from a linear accelerator rather than requiring the pulse compression in an accumulator ring or synchrotron as in a SPSS. Pulse lengths of 300 μsec or greater can be easily extracted from linacs at repetition rates in the 10s to 100s of Hz. The advantage of this LPSS method (when it can be exploited) is that the cost per neutron is

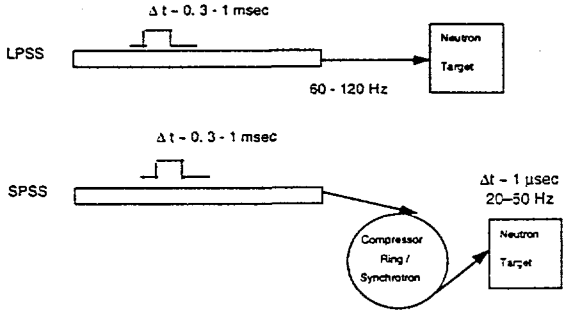


Fig. 1.

less than an SPSS of the same power because there is no cost for the time compression.

The LPSS is particularly well suited to do experiments with cold and ultra-cold neutrons. Cold neutrons refer to those neutrons with long wavelengths greater than ~3Å and are usually produced using moderators at low temperatures, such as liquid H_2 at 20K. The *average* cold neutron flux from a 1 MW LPSS was calculated by G. Russell[1] using the LAHET code system to be equivalent to the cold neutron flux from the ILL reactor multiplied by a factor of 1/4. Since the ILL reactor is a 60 MW reactor, this means that the 1 MW LPSS produces an average cold neutron flux approximately equivalent to a 15 MW reactor. This calculation assumes a coupled moderator since the resolution will depend on the proton pulse width and not the moderation time uncertainty. This comparison is shown in Fig. 2.

If the only characteristic of a 1 MW LPSS was that it was the equivalent of a 15 MW reactor, it would not be a very exciting new concept. However, it is possible to use the pulsed nature of the LPSS, similar to an SPSS, to achieve gains for many experiments as compared to a steady-state source of equivalent average neutron flux.

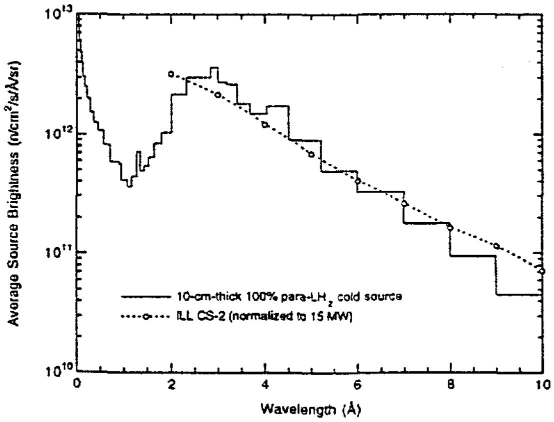


Fig. 2.

This can be demonstrated from Fig. 3, which shows a time-distance plot for a pulsed source where the time resolution, δt, is defined by the chopper. The time between pulses is denoted by Δt. The duty factor of the pulsed source is $c = \delta t / \Delta t$. The comparison between a steady-state source experiment and a time of flight experiment can be expressed in the following manner. In an experiment using a CW source one selects a narrow wavelength band, Δλ, from the neutron spectrum, $\phi(\lambda)$, using a monochromator, and this is equal to the wavelength resolution, δλ.

For a time of flight (TOF) experiment, the wavelength resolution is

$$\delta\lambda = \frac{h\,\delta t}{m\,L}$$

where h = Planck's constant, m is the neutron mass, and L is the flight path.

The wavelength band, Δλ, is given by

$$\Delta\lambda = \frac{h\,\Delta t}{m\,L}$$

Therefore, in a TOF experiment, if it is possible to use the entire bandwidth for an experiment, one can measure a gain over a CW experiment characterized by

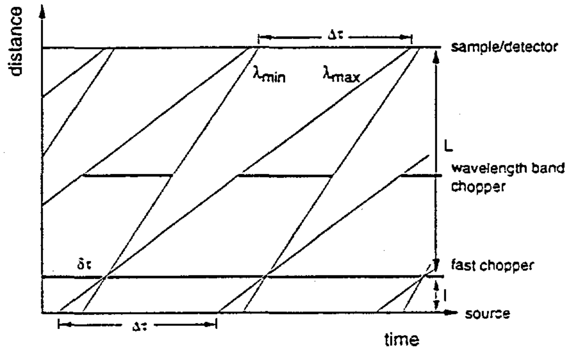


Fig. 3.

$$\Delta\lambda / \delta\lambda = \Delta t / \delta t = 1/c.$$

For a LPSS with a duty factor of 6%, this would yield a maximum gain from TOF of 17. However, this gain is dependent on the type of experiment and on whether the entire wavelength band contains useful information. Since the wavelength resolution for an LPSS is determined by the length of proton pulse or by the opening time of a chopper, then the resolution is essentially constant over the bandwidth. This is not true for an SPSS, hence the gain factor of 1/c does not apply to an SPSS.

A workshop on how neutron scattering instruments would perform at an LPSS was held at Lawrence Berkeley National Laboratory.[2] The results indicate that the gain factors vary from 1 to 17 for a variety of spectrometers examined. Refinements in the simulation of these spectrometers are being made with Monte Carlo techniques, and results will be presented at an upcoming workshop in Sept. 1996.[3]

To summarize, an LPSS provides the greatest gains for relatively low resolution experiments with long wavelength (cold) neutrons. An LPSS is complementary to an SPSS, which is most effective for high resolution experiments with epithermal and thermal neutrons. With respect to cost, an LPSS offers advantages in not having to provide the additional equipment to compress the time structure of the proton beam.

The present status of the LPSS concept is that a proposal has been made to the Department of Energy to consider an upgrade of LANSCE to provide a 60 Hz, 1.2 mA average current beam injected into a new neutron target with 16 beamlines. The LANSCE LPSS is being considered for funding in the FY1999 time frame. It would become operational in the 2002-2003 period. For more information on the performance of an LPSS compared to a steady state source, see Ref. 3.

Research with UCN

Ultra-cold neutrons, with velocities less than 5 meters/sec, can be produced by several means. The first method is simply to utilize the very long wavelength part of a Maxwellian spectrum. As can be seen from Fig. 4, which shows 2 Maxwellian spectra for temperatures of 300K and 20K, the probability of obtaining a neutron whose velocity is less than 5 meters/sec is less than 5×10^{-8} even for the very cold moderators. So UCN are very difficult to directly extract from moderators.

However, there is another approach that is schematically shown in Fig. 5. It uses a moving crystal to Doppler shift neutrons to lower velocities. If the velocity of the rotor is half of the neutron energy striking it, the result is that the reflected neutron's energy will be essentially zero (or ultra-cold). This concept was first successfully tested at Argonne National Lab at the ZING facility. Presently researchers at LANSCE are installing a rotor system for which a UCN density of 10-30 UCN/cm^3 is calculated. This should be compared to the best moderator source of 80-90 UCN/cm^3 at the ILL reactor.

A new type of UCN source has been proposed called a "superthermal" source, in which UCN are produced by the inelastic interaction of cold neutrons with a very cold medium. In this case, the neutron energies are well below molecular binding energies so that single

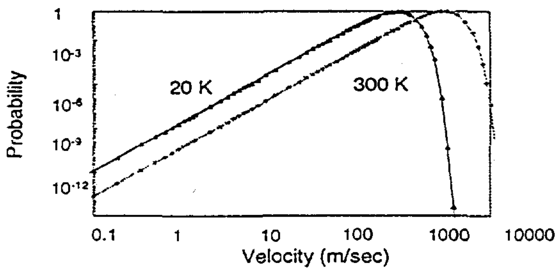


Fig. 4.

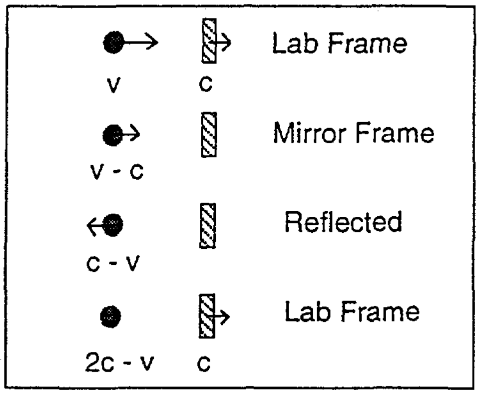
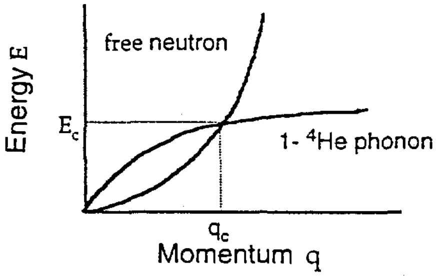


Fig. 5.

phonon scattering is the dominant inelastic process. As shown in Fig. 6, cold neutrons can create a phonon and be "down-scattered" in energy to near zero energy. Thus a UCN is created. The UCN can also absorb a phonon and be "up-scattered" to cold or thermal energies. In this latter case, the UCN is destroyed. However, the rates for upscattering are much smaller than down scattering because there are effectively no phonons of the appropriate energy to be absorbed by the UCN.

Such sources have not been used so far because at a reactor it is difficult to maintain a very low moderator temperature near the reactor core. Also inserting such a device in a reactor changes the configuration significantly leading to the requirement for major safety analysis and potential relicensing. Superthermal sources are much more likely to be successful at a spallation source than a reactor. The reasons include the following: (i) the thermal loading per neutron is reduced by a factor of 20; (ii) the spallation source is insensitive to criticality thus making it much simpler; (iii) it would be very difficult to retrofit and existing reactor.



Only a neutron at E_c can come to rest by emission of an excitation (E_c = 11K, λ_c=10 A)

Fig. 6.

In addition to superfluid ^{4}He, it has been proposed[4] to use solid H_2 as a superthermal source. This source essentially works the same way as ^{4}He but has the advantage of potentially higher UCN production. UCN densities for the superthermal sources have been projected to be as high as 10^6 UCN/cm^3 which is a factor of 10^4 higher than the best source today.

Assuming that one can produce such a high UCN density at an LPSS, how would you store the UCN? There are several different mechanisms that can be used to store UCN. Many materials have coherent scattering lengths of 5-15 fm, which means that neutrons with velocities of less than 8 meters/sec can be internally reflected from the surfaces and thus bottled and contained.

A second method of containment utilizes the fact that the gravitational potential of a 1 meter height difference is comparable to the kinetic energy of a UCN. This allows both containment and velocity selection. The third method involves containment in a magnetic field. The energy of a neutron in a 4 Tesla magnetic field is comparable to its kinetic energy of 100 neV. This is essentially 100% containment and provides for 100% polarization due to the magnetic moment of the neutron.

Now that one has stored the UCNs, what will one do with them? There are two fundamental measurements that test the Grand Unified Theories of matter. One such is the so-called standard model, which has to-date not been disproved by any experimental results although there are several recent experiments which are not predicted by the standard model. One UCN measurement is the search for the electric dipole moment (EDM) of the neutron. For the past 30 years, the limit on the neutron EDM has been one of the most important constraints on theories seeking to explain CP violation. Bottled UCNs permit the best measurements of the EDM. The history of EDM measurements is shown in Fig. 7. The next generation of measurements will put severe tests on the theories listed on the right-hand axis if the EDM is found to be lower than their predictions. The estimated sensitivity of the new UCN EDM measurements is shown to be well below these theories. This should be an exciting 10-20 years in this field.

A second measurement is that of the neutron beta-decay lifetime. The lifetime of the neutron has been measured to be 800±1.9 sec. Neutron beta decay is the archetype for all semi-leptonic weak interactions. The neutron lifetime is a critical constant for both weak interaction and nuclear theory and it has major implications in astrophysics and cosmology.

Accelerator - Driven Transmutation of Nuclear Waste (ATW)

The management of spent nuclear fuel has been a topic of debate worldwide for the past 25-30 years. Various countries have taken different approaches to the problem. In the United States, we have chosen a once-through fuel cycle for reactors that results in a large waste stream to be dealt with. This decision was based both on economics of this fuel cycle as well as a desire to reduce the potential for proliferation of nuclear weapons that was feared if countries moved to other fuel cycles involving reprocessing of spent nuclear fuel and eventually breeding of new fissile material. Other countries such as France, Britain and Japan have not been as concerned with the proliferation risks of reprocessing but were driven by their own economic analyses which showed favored gain by recycling the fissile material from spent fuel. France, in addition, has had the largest breeder reactor program in the world.

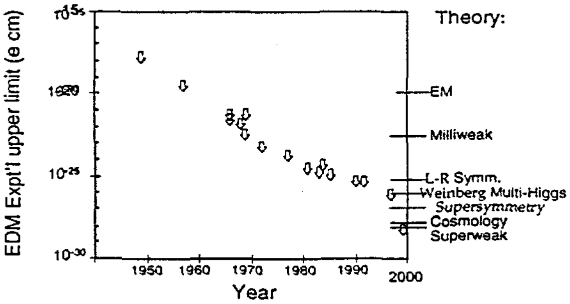


Fig. 7.

One of the major concerns of a continued global reactor economy for nuclear energy is the amount of plutonium that is contained in spent fuel. Figure 8 shows a projection of the amount of Pu that will exist in the world as a function of year depending on the light water reactor uranium burnup rate. If the world moves to a more efficient burnup than present, by the year 2015 there will be 2000 tons of Pu in the world in spent fuel. This amount dwarfs the 100 tons of excess Pu that exist as a result of the reduction in the nuclear stockpiles of the U.S. and Russia. There are methods to reduce this amount with reactor burnup schemes, but they eventually plateau at approximately 2000 T after burnup of much of the Pu. I will discuss accelerator-based systems (ATW) in this section and show how they can reduce the problem significantly.

The main goal of all reactor spent fuel programs is to minimize the risk to the environment by either encapsulating the waste in a form that will not deteriorate during the bulk of the lifetime of the waste or by

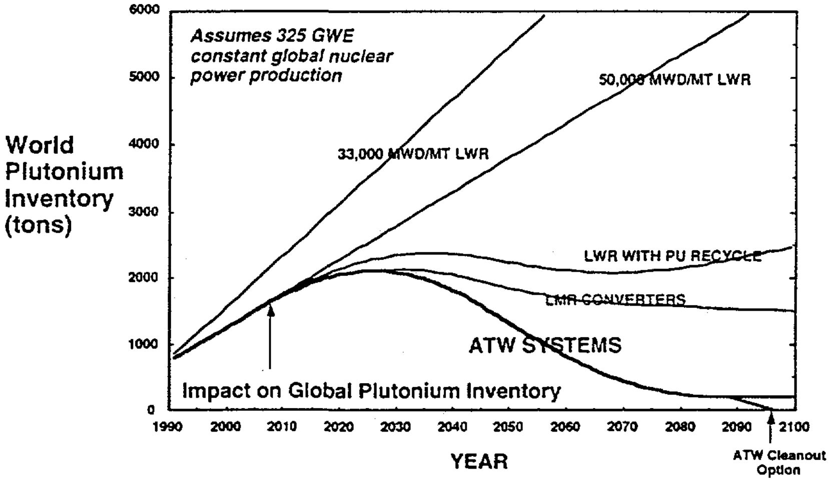


Fig. 8.

recycling the actinides and burning them in a reactor while preparing the fission products for burial in a repository. The geologic repositories have had considerable amounts of R&D done on them in past 25 years during a search for the best sites in each country. The U.S. site at Yucca Mt. is an oxidizing environment, which could cause some problems for transport of longer-lived waste after the principal containment is breached some time after it is put in the repository. This has caused problems for getting the site approved as a geologic repository for the proposed period of 10,000 years. Sites in other countries have different problems to overcome but appear to be on a path where some nuclear waste will be emplaced in the future.

If one steps back from the events of the past 30 years and attempts to ask the questions regarding what the best approach might be for nuclear waste, one is faced with environmental concerns, safety concerns, and economic concerns. All nuclear waste consists of radioactive products that live from seconds to days to years to thousands and millions of years.

One strategy that we have been pursuing at Los Alamos for the past 6 years has been to look at a different type of treatment of the spent fuel that would allow one to extract the uranium and cladding materials while leaving the plutonium and other actinides in the spent fuel. The challenge is to develop a process that is proliferation resistant so that it is difficult to extract Pu for nuclear weapons use without being detected by an international safeguards monitor. If this can be done economically, then the actinides are burned and their energy extracted.

The other challenge is to reduce the toxicity of the fission product waste. This would involve transmutation of the fission products with long-lived half-lives to products which are either stable or are much shorter lived (10s of years). This would permit a new nuclear waste strategy and policy for the U.S. since the spent fuel could be moved from reactor sites after an initial decay period followed by processing of the fuel to remove uranium and then by extraction of the actinides and fission products for burnup and transmutation.

The concept that we have put forward utilizes a high power linear accelerator to produce a copious source of spallation neutrons inside a target/blanket region in which the actinides and fission products can be burned or transmuted by the neutrons. A schematic diagram of the accelerator-based transmutation of waste (ATW) system is shown in Fig. 9.

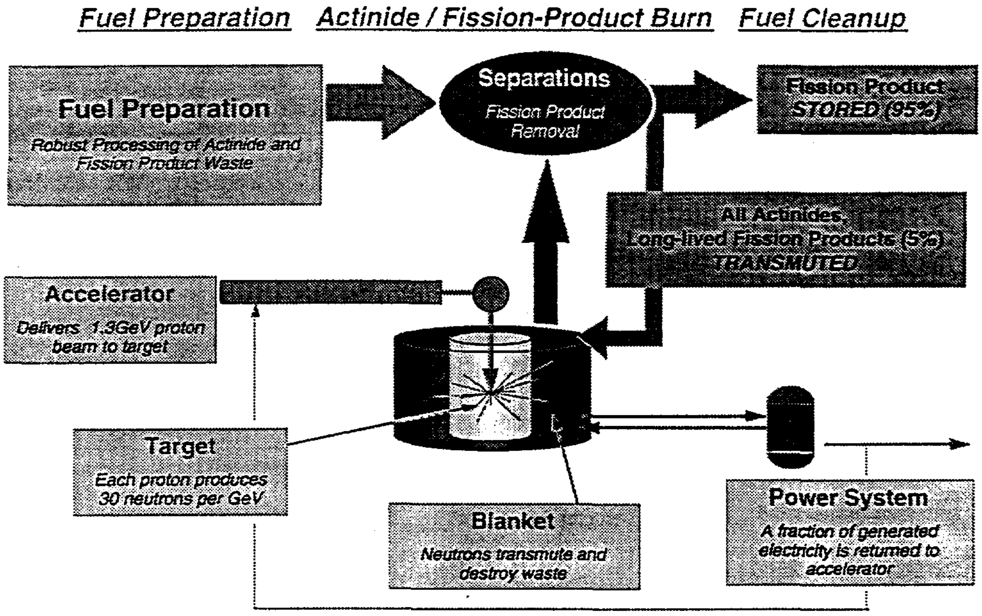


Fig. 9.

This ATW system uses a proton linac of about 1 to 1.5 GeV, which delivers the beam to a subcritical assembly of molten lead, in which approximately 30 neutrons per one GeV proton are produced.

Subcriticality is an important feature of ATW systems. As contrasted to a reactor, an ATW system precludes the occurrence of self-sustained chain reactions by operating well below criticality so that response to reactivity transients and other perturbations is small. ATW systems do not require mechanically-driven, neutron absorbing control rods or burnable poisons so that ATW neutron usage is more efficient. As a result, waste actinides that cannot be easily burned in critical reactors could be safely and effectively destroyed in the ATW burner.

A subcritical system can be defined with an "criticality coefficient," k_{eff} which is given by

$$k_{eff} = \nu \; / \; (1 + \alpha + P + T),$$

where

ν = average number of neutrons released by each fission,

α = ratio of neutron absorption to fission cross section in the active component of the fuel,

P = number of neutrons parasitically absorbed per fission, and

T = number of "working" neutrons per fission available for transmutation.

If one utilizes a fast neutron spectrum in the ATW burner, then

$$T_{fast} = (3.2/k_{eff}) - 1.8;$$

while for a thermal spectrum

$$T_{thermal} = (3.0/ k_{eff}) - 3.0.$$

For a fast spectrum, and a k_{eff} of 0.9, the number of neutrons per fission available for transmutation is 1.75

where the accelerator is supplying the neutrons to maintain the system performance.

The advantage of a thermal spectrum ATW system is that it has a low inventory of waste in the burner while the fast spectrum system has a large number of transmutation (T) neutrons available to do work.

Figure 10 shows the inventory of actinides in an ATW system at the beginning of life, after 36 years, and at end of life for fast and thermal systems. The obvious strategy is to utilize a fast spectrum until end of life followed by a thermal spectrum burndown of the remaining actinides. This can be done with the type of liquid fuel systems that are under consideration in ATW.

Why liquid fuels? Subcriticality allows much more flexible operation since fuel can be fed into the system without major perturbations. The flow of liquid fuel means more uniform burnup regardless of the flux profile in the blanket. Liquid fuels allow higher power densities, and regular fuel cleanup is possible.

Among liquid carriers for the spent fuel, lead/bismuth and fluoride salts offer a good interface for the front-end and back-end processes for fuel preparation and cleanup. Both carriers have low vapor pressure even at high temperatures, do not react chemically with water, steam or to each other, and therefore add to safety considerations.

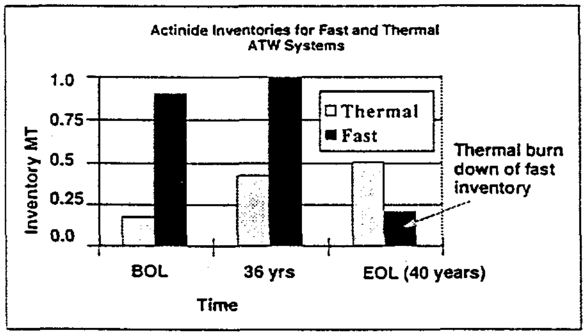


Fig. 10.

The process chemistry is an important part of the ATW system since it has a major impact both on cost and performance. Figure 11 shows a schematic diagram of the process chemistry. Fuel preparation is achieved by molten-salt hydrofluorination and electrowinning processes, which extract the uranium and zirconium fuel cladding from the spent fuel, separately extract the fission product transition metals such as Tc, for future transmutation or storage, and leave the higher actinides (Pu and above) and the lanthanides and Cs, Rb, and Sr fission products in-stream to be transmuted. Electrowinning exploits the electrochemical behavior of the materials to do this extraction without isolating the Pu. This provides no opportunity for isolation, storage, or transportation of Pu and provides a barrier to proliferation. An electrochemical potential diagram is shown in Fig. 12, which shows how this can be accomplished. More details of the front-end process are given in Ref. 5.

Similar processes are used in the back-end to clean up transmutation residues. Metals are separated by electrowinning. Volatile isotopes are separated via a helium sparge system and fractional distillation processes. Lanthanide separations are accomplished by a reductive extraction process utilizing a centrifugal contactor. Details of the back-end process are described in Ref. 5.

The effectiveness of an 3000 MWth ATW system is shown in Fig. 13. The ATW system can accept the waste from six 1000 MWe light water reactors that have utilized a burnup rate of 50,000MWd/ton of fuel. This ATW system will burn up the actinides and most egregious fission products, such as ^{99}Tc , ^{79}Se, ^{126}Sn, ^{135}Cs and ^{129}I. The accelerator required for this system is an APT class accelerator with 100 mA of protons at 1.5-2 GeV.

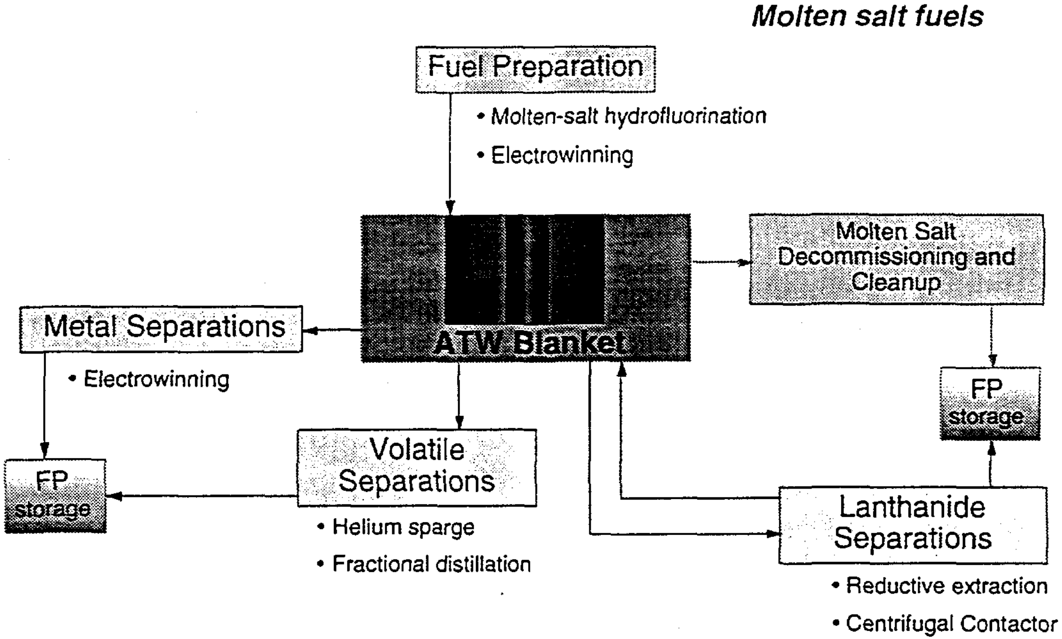


Fig. 11.

The final waste forms from an ATW system are shown in Fig. 14. If 4000 tons of spent fuel are processed in a 3000 MWth ATW plant during a 30 year lifetime, then there are 30 kg of transition metals which could be put in a Zr matrix for storage, 24 kg of lanthanides which can be placed as oxides in a glass matrix, and 6 kg of active metals such as Cs, Sr, Rb, that can be stored as oxides in engineered containers. The net result of the transmutation is a reduction in the quantity of waste that would go to a geologic repository by a factor of 1000 or more. As shown in Fig. 15, the ATW waste would drop significantly in its levels after a few hundred years of storage while achieving the factor of 1000 in the 1000-10,000 year time frame plus eliminating the main fission product concerns of Tc and I.

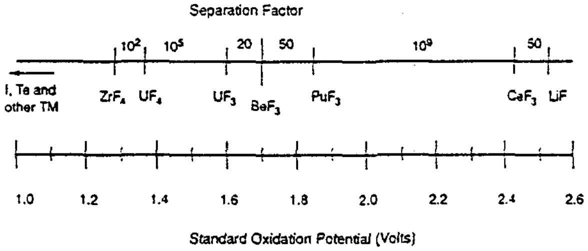


Fig. 12.

There is a lot of work to be done over the next several decades if ATW is to play a significant role in the global nuclear waste strategy. Interest is high outside the U.S. in such ATW systems with the following countries having either an active program or expressed positive interest: Belarus, Czech Republic, France, Italy, Japan, Russia, South Korea, and Sweden.

ATW offers the potential to reduce the toxicity of spent nuclear fuel considerably and to help manage the global inventory of Pu from the nuclear fuel cycle. In this regard, it fits with a strategy to reduce proliferation of nuclear material globally and to provide a more environmentally acceptable disposal of the spent fuel accumulated to date. It provides the possibility to make the job of a repository much easier since the more difficult fission products are eliminated, and the actinides are destroyed. Whether or not the world continues to

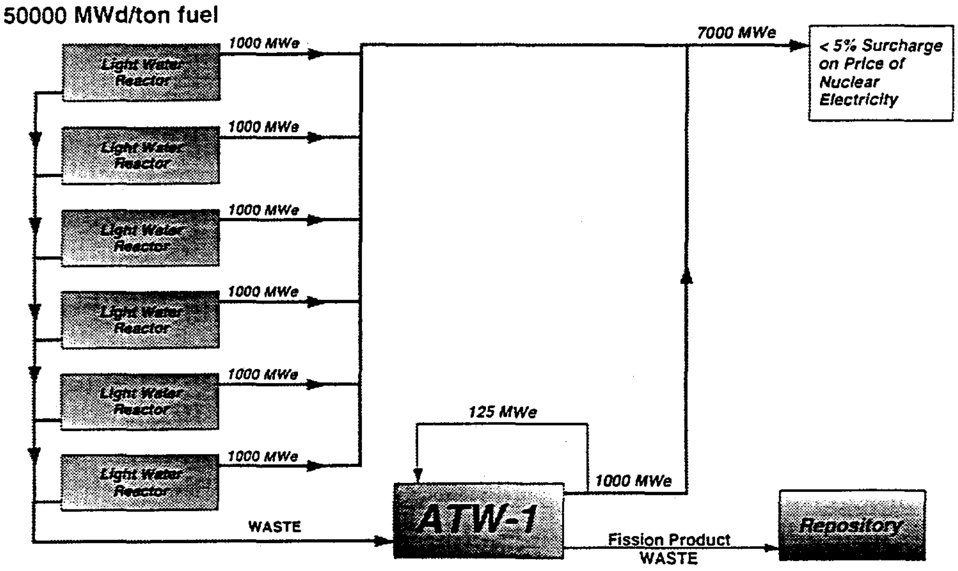


Fig. 13.

pursue nuclear power as a source of electricity, ATW will play a role for the next 100 years in cleaning up the existing legacy.

Summary

Advances in high power accelerator technology open new applications for science and technology in the 21st Century. In this Symposium, we have seen opportunities for neutron scattering in materials science and structural biology, in defense science with neutrons and protons, for fundamental studies using ultra-cold neutrons and neutrinos, in studies of materials damage, and in the production of radioisotopes. Lastly, in this paper, I have described a concept for burning nuclear waste that potentially offers an option for the world to deal with the large quantities of waste that are accumulating and doing so in a manner that reduces proliferation risks and provides a more environmentally acceptable solution than simply burying the existing waste in a geologic repository.

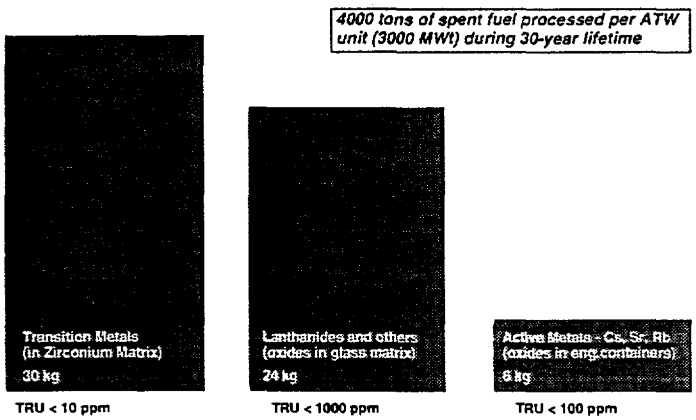


Fig. 14.

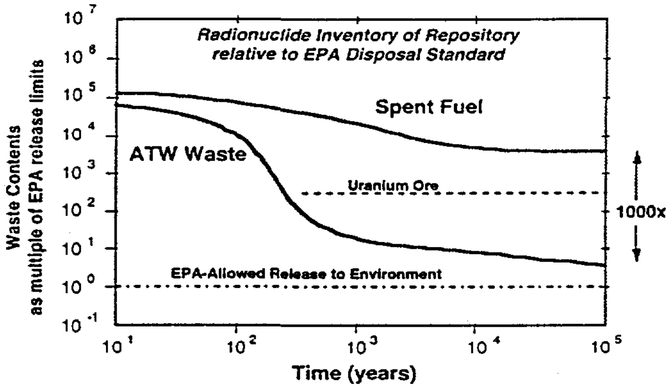


Fig. 15.

The APT accelerator that is to be built at Savannah River Site is a key enabling step to these future applications because it will demonstrate the technology and very high power operation in an industrial-plant-like environment requiring high availability on a year-round basis. Future systems will build on this experience to produce even more efficient and cost-effective designs.

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